Comments Received during the Public Review Period on the Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2012

Commenter: Giles Ragsdale
AECOM

Comment: Regarding chapter ES.2. – My opinion is that under this recent trends paragraph, note should be taken of the results of Figure ES-15. There is a positive story to tell in that despite increased population and Real GDP, emissions per capita and per $GDP have been trending downward since 1990 and by more than a negligible amount.

Comment: Regarding figure ES-3: I think the title should be revised. I might be confused, but I do not think the data on the graph reflect the title of the figure. I see that each year's annual total compared to 1990 is represented, but I do not see the "Cumulative Change" noted in the title. For cumulative change, it would seem that 1991 would be -25 as noted, but 1992 would be 52, 1993 would be 261, etc.

Commenter: William Herz
National Lime Association

Comment: In response to the last iteration of EPA's Greenhouse Gas Inventory Draft, published in March of 2013, NLA submitted comments that recommended EPA discontinue using the IPCC emission factors to account for LKD emissions, and that the agency also take into account CO2 emissions from off-spec lime, scrubber sludge, and other wastes. A copy of NLA previous comments is included in Attachment 1. This issue continues to be important to NLA members, not only to help ensure the completeness and accuracy of the data EPA publishes but also to ensuring the achievement of EPA's stated goal of agreement and alignment with the GHG mandatory reporting system.

Currently, EPA calcination emission calculations rely solely on output-based emission factors from the IPCC 2006 GHG Guidelines, which we believe are outdated. Central to the NLA's previous comments were recommendations to adopt accurate calcination emissions calculation methodology for:

- Lime Products; and
- Lime Kiln Dust (LKD); and
- Off-spec lime, scrubber sludge and other wastes.

Each of our recommendations was based on analysis of actual production data, including CaO and MgO oxide contents of lime and LKD, which had been provided to NLA by member companies. These results of this data were incorporated in the comments NLA submitted to your office last year. The comments, together with the data we provided, should be sufficient to provide EPA with the basis to generate more accurate emissions estimates for LKD and other lime products (including off-spec lime and scrubber sludge).

In sum, NLA's earlier comments concluded that while the IPCC's output-based approach for estimating calcination emissions from U.S. lime products may be accurate as to the overall data to be published, it nonetheless understates emissions from LKD and other byproducts and wastes generated in the United States. For that reason, NLA recommended that lime calcination emissions be multiplied by a factor of 1.06 (not 1.02) to account for LKD and a factor of 1.02 to account for wastes generated at lime plants; neither of these are currently accounted for which we believe is a critical error.

When the current Draft Inventory of U.S. Greenhouse Gases and Sinks 1990-2012 was published on February 21, 2014, it was disappointing that none of our recommendations concerning the use of more accurate correction factors had been adopted and EPA continued to rely on the outdated IPCC factor of 1.02 to account for LKD. Likewise, EPA took no action in relation to off-spec lime and other wastes.

As we stressed in our previous comments concerning the earlier draft, NLA conclusions and recommendations were premised on our belief in the need for EPA's published data to be accurate; especially when NLA's members are willing to supplement the agency's data with accurate data of their own. Because EPA relies solely on the questionable IPCC LKD generation rates, calcination emissions continue to be understated. Accordingly, we again urge EPA to adopt our recommendations; if there are other supporting data we can also provide that would add further weight to and/or support for our recommendations, please let us know.

In addition, we recognize that EPA has a substantive interest in having both the GHG Inventory and the Mandatory GHG Reporting system be in agreement as much as possible. This is important not only for EPA's credibility but also for the public's and stakeholders' understanding of these issues. In this regard, as we stated in our previous comments:

<u>Lime Kiln Dust</u>

"...based on data reported to NLA from our members, emissions from generating LKD account for about 6% of calcination-related emissions from lime manufacturing (in 2011, it was 5.8%). Currently the IPCC multiplies lime product-related emissions by a "correction factor" of 1.02 to account for LKD. The IPCC Guidelines acknowledge that this correction factor for LKD is borrowed from its chapter on cement, which in turn explains that the factor for cement kiln dust (CKD) is relatively low because most CKD is recycled back into the process.

By contrast, the lime industry does not recycle LKD back into the process, and thus borrowing such a factor to account for LKD-related calcination emissions is inappropriate.

EPA's reliance on the IPCC's LKD generation rate of 2% (rather than 6%) understates calcination emissions from our members alone by 535,610 tons. This is roughly 5.4% of our members' total emissions, and twenty times the understated calcination emissions described earlier for lime products."

Off-Spec Lime, Scrubber Sludge, and Other Wastes

"The IPCC Guidelines do not to take into account calcination emissions resulting from wastes commonly generated at lime plants (e.g. off-spec lime that is not recycled, scrubber sludge). Again, based on 2011 data reported to NLA from our members, calcination emissions from production of such wastes account for approximately 1.7% of total calcination emissions, or 256,000 tons. To address this omission, we recommend that EPA multiply quicklime calcination emissions by a factor of 1.02."

Conclusion:

NLA believes the deficiencies in the proposed inventory are significant and should be corrected. In the aggregate, EPA has underestimated lime emissions by approximately 814,000 CO2 tons; as the off-spec materials generate 256,000 tons (completely unaccounted for in the inventory) and 535K tons (the difference in LKD emissions when utilizing the correct emissions factor; (854K – 319K)). This represents an underestimate of approximately 5.1%, which is not insignificant.

Commenter: Marlen Eve
USDA Animal and Plant Health Inspection Service

Comment: Executive Summary:

Page 1 lines 29-30: Excellent!

Page 2 lines 9-10: Needed for effective comparison.

Page 5 Figure ES-3: Very impressive and encouraging trend!

Page 10 lines 10-14: This is excellent – it enables an accurate sectorial picture otherwise difficult to estimate.

Page 14 lines 27-31: Noteworthy point that technology improvements can be so effective in this area.

Page 15 lines 9-10: Good to see this point made in Summary – an area USDA is researching and still in need of improvement.

Page 16 lines 19-21: Suggests an area in need of more oversight and regulation in a fast growing industry.

Page 17 line 13: Noteworthy effect of improved land-use and forests as sink. Question – why have these sinks not increased since 2007 – compared to notable improving trend over 1998-2004?

Page 20 lines 7-10: Good point to note – not sure this is widely recognized – and how difficult it is to manage for lower levels. This is a clearly-needed USDA research area.

Page 20 lines 29-30: Good to mention in Executive Summary given this is a problematic area in many developing counties including China and India.

Page 27 lines 7-9: Good point to make – it identified an area that could benefit from future research.

Page 5 line 27: Is this very long table needed in Executive Summary?

Page 11 line 15: Reference that low fuel prices during period 1990-2012 in part contributed to increase in number of vehicle miles. Hopefully this can be substantiated through economic comparison – I think fuel prices increased considerably during this period relative to other consumer prices. And when I look at some internet sites such as: http://www.inflationdata.com/inflation/images/charts/Oil/Gasoline_inflation_chart.htm, they seem to reflect that the statement that gas cost has remained low and thus the conclusion that this leads to increase in number of vehicle miles could be challenged.

Page 17 lines 17-18: Is this very long table needed in Executive Summary?

Comment: Introduction:

Page 13 line 1: Very good summary of all input sources of data and expertise in one diagram!

Comment: Agriculture:

Page 1 lines 5-8: Very good way to focus on what is critical in agriculture practices!!

Page 1 lines 16-20: We liked this up-front summary and focus on what is critical!

All pages: There is a great abundance of numbers, informational statements, tables and some figures. The details can be overwhelming but we view the text and supporting data as essential, comprehensive, well-balanced, and superbly organized by easy to read, consistent sections on each source of non-CO2 GHG. The methods used should be of value to other countries as a 'role model' on what data is needed and on how to assess uncertainty and apply verifications and recalculations.

Overall—comprehensive and well-written chapter on a difficult subject.

Several locations in the Chapter: There are references to number of cattle/dairy cattle increasing/decreasing but overall CH4 emission increasing due to digestibility. I can look at some trends in the NASS that would indicate that the trends in numbers of cattle stated don't quite coincide with my quick review of NASS. But they do state a lot of adjustments that they made to the numbers that I don't have the time to work through. And I definitely don't have the background on digestibility – few, if any, in Veterinary Services would. So I can't validate or refute, and would not want our brief review to be considered a "peer review". I would hope that this section and others in the paper have been appropriately peer reviewed to avoid any improper conclusions developed which could have an undue negative influence on animal agriculture.

Comment: Land Use, Land-Use Change, and Forestry:

All pages: As commented on Agriculture Chapter we note that there is a great abundance of numbers, informational statements, tables and some figures. The details can be overwhelming but we view the text and supporting data as essential, comprehensive, well-balanced, and superbly organized by easy to read, consistent sections on each source of GHG. The methods used should be of value to other countries as a 'role model' on what data is needed and on how to assess uncertainty and apply verifications and recalculations.

Overall: Comprehensive and well-written chapter on a difficult subject. This category is especially important to developing countries where land use is in flux and where practices such as forest cutting and clearing, fire use, and extensive degradation by grazing is wide-spread.

Comment: Recalculations:

Page 1 lines 2-4: We felt this is one of the most important chapters in the Report given it provides a protocol and verification annually of the estimates. It has the salutary benefit of credibility of estimated made given they are constantly under re-evaluation as new data (past and present) and methods are developed and accessed. Some of the changes appear large in magnitude – but this may not be unusual where only imprecise data was available initially.

Possibly add a summary or a tabulation of what this report achieved in the way of new data, new methods and new findings that were not mainstream in prior analyses and thinking.

We note with interest some prior assumptions (or simple lack of information or awareness) on aspects of agriculture and land use / forestry of special interest. Some of these new perceptions

are now and in the immediate future will be important in on-going and future negotiations over land and land practice monitoring by different countries.

Commenter: Marlen Eve
USDA Agriculture Research Service

Comment: Agriculture chapter: Page1 line 6:

Seems to imply C removal is only related to land-use change. Assuming other management does not have an impact?

Comment: Agriculture chapter: Page 1 line 18:

"other cropping practices" a little vague – such as?

Comment: Agriculture chapter: Page 2 lines 16-17:

"These non-ruminants emit significantly less CH4 on a per-animal-mass basis than ruminants because the capacity of the large intestine to produce CH4 is lower."

Add … lower than in a rumen.

Comment: Agriculture chapter: Page 9 line 20:

"…increasing use of liquid manure management systems, which have higher potential CH4 emissions than dry systems." Are there any estimates on the adoption of methane capture from liquid manure?

Comment: Agriculture chapter: Page 29 line 3-4:

This sentence needs to be rewritten without all the "nots:"

"However, renewal of pasture that is **not** rotated with annual crops occasionally is **not** common in the United States, and is **not** estimated."

Comment: Agriculture chapter: Page 30:

In general DAYCENT appears to perform well, but recent work by Campbell et al., 2014 suggestion DAYCENT may underestimate N2O emissions. "Overall, DAYCENT performed

well at simulating stover yields and low N2O emission rates, reasonably well when simulating the effects of management practices on average grain yields and SOC change, and poorly when estimating high N2O emissions. These biases should be considered when DAYCENT is used as a decision support tool for recommending sustainable corn stover removal practices to advance bioenergy industry based on corn stover feedstock material." (Campbell et al., 2014). Thus, as more empirical data becomes available it could be used to improve DAYCENT.

Comment: Agriculture chapter: Page 41 Table 6-27:

Key Assumptions for Estimating Emissions from Field Burning of Agricultural Residues: Is it correct that this is refers only to residue that are burned in the field or does it include residues harvested and burned for energy – clarify.

Commenter: Other
USDA

Comment: Page 6-7, line 18: "months" should be inserted after "4-6."

Commenter: Carrie Reese
Pioneer

Comment: Gas Well Completions/Workovers with Hydraulic Fracturing:

Pioneer commends EPA's consideration of stakeholder comments to the 2013 Inventory and subsequent development of control technology-specific, net emission factors for gas well completions and workovers with hydraulic fracturing. This approach makes use of a more comprehensive data set and provides greater transparency regarding EPA's accounting of emissions reductions carried out by the industry. However, Pioneer feels that this methodology can still be improved upon.

Emissions quantified in the Greenhouse Gas Reporting Program (GHGRP) for 2011 and 2012 are based on engineering estimates and best available monitoring methods (BAMM) in addition to direct measurements. In Pioneer's initial review of 2011 and 2012 GHGRP data for "HF completions that vent", average emissions per event (Mg CH4) computed by an estimation methodology appear to be nearly tenfold that of directly-measured emissions. Until there is further understanding of the nature of these events, Pioneer suggests that EPA develop control technology-specific, net emissions factors focusing on measured data from the GHGRP and measured data contributed by other accepted sources.

Published by the University of Texas at Austin in September 2013, Measurements of Methane Emissions at Natural Gas Production Sites1 (Allen, et. al) quantifies emissions from 27 gas well completions in multiple production regions. Representative gas well completions from nine operators, which conduct about half of all new well completions, were sampled. The

measurement results, a product of peer-reviewed, scientifically-robust, and fully-disclosed methodology, present a basis to consider the reasonableness of other data provided under less controlled circumstances.

Referred to in the 2014 Inventory, the November 8, 2012 document entitled "Inventory of U.S. Greenhouse Gas Emissions and Sinks: Updating Emissions Estimate for Hydraulically Fractured Gas Well Completions and Workovers", identifies four categories of events and proposes new emission factors for each of these categories. The following table provides a comparison of the EPA emission factors in the 2014 Inventory to the measurements reported by Allen, et al. {2013). The study reports emissions data for completion flowbacks only, with no measurements for workovers with hydraulic fracturing.

Type of completion flowback or workover	*EPA Emission Factor*	*Observed Emission Factor from Allen, et al., (2013)*
Wells that vent without flaring or Reduced Emission Equipment (REC)	41	0.83 (observed mean) 0.8-124 (range)*
Wells that flare (without REC)	5	Not observed
Wells with REC that do not flare	3	4
Wells with REC that flare	6	1.5-18

*The wells that vented without flaring or REC observed by Allen, et al. (2013) had much lower potential emissions (0.83 Mg) than the average potential emissions for all of the observed wells (124 Mg). If the wells in this category observed by Allen,et al.are representative of national populations of this category of wells, then the emission factor for this category would be 0.8 Mg per event; in contrast, if the sample of all wells observed by Allen, et al.(2013) is considered representative of this category and it is assumed that the deployment of REC equipment is random among all of these wells, then the emission factor for this category would be 124 Mg per event.

Comment: Liquid Unloadings:

In previous comments to the 2013 Inventory, Pioneer expressed support for EPA's development of net emissions factors for liquid unloading events, but also noted concern that Subpart W calculation methodology may tend to overstate emissions. Pioneer requests that EPA continue to consider improvements to the calculations in this emissions category.
In the study referenced above, Allen, et al. (2013) also reported on emissions from liquid unloadings.

The sample set of nine manual unloadings proved insufficient to allow for extrapolation at a national scale, and the study team is conducting additional measurements to supplement the data collected in the first part of the study. However, Allen, et al. (2013) does report an important observation from the initial effort, demonstrating that the Subpart W methodology for liquid unloadings without plunger lifts (based on engineering calculations and not direct measurements) overestimates emissions for every measured event. Collectively, emissions are estimated five times higher than the measured emissions.

Pioneer again suggests that underlying causes for overestimation of emissions may be EPA's assumption that a full wellbore volume of gas is vented with each unloading and the assumption that a well unloads for one hour on average. In addition, Allen, et al. (2013) observed intermittent flow rates during unloading events and proposes that EPA's assumption of a continuous gas flow rate may be another contributor to overestimation .

Comment: Pioneer applauds EPA's commitment to refine emission estimates in the U.S. Greenhouse Gas Inventory to reflect the best available information. As the body of scientific and engineering knowledge continues to grow with regards to emissions from oil and gas activities, Pioneer contends that emphasis must be placed on directly-measured data and that results from these direct measurements should in turn inform corresponding estimation methodologies . Collaboration is the key to this process, and Pioneer looks forward to continued dialogue with EPA.

Commenter: Chris Busch

Energy Innovation: Policy and Technology

Comment: The EPA should take steps to address clear evidence that its inventory of GHG emissions is undercounting methane. In the short run, as part of finalizing the 2014 inventory, the agency should make the case for a significant effort to improve the inventory of emissions from the natural gas sector. In the longer run, the agency should develop a plan for integrating top-down data as well as new technologies that operate at ground level that can assist in leak detection and measurement. The federal government should be placing more emphasis and devoting more resources to this effort.

Comment: Brandt et al.'s work illustrates the value of top-down measurements to provide evidence of overall emission levels over large areas. The EPA should move to collect airborne measurements into its GHG inventories. By conducting measurement campaigns, EPA will be able to obtain atmospheric data that is more comprehensive across space and time. This will enable the agency to identify aggregate emissions levels with much greater accuracy and will help to improve confidence intervals. Current confidence intervals are much too small.

Comment: Emerging technologies can link emissions back to sources, enabling the EPA to conduct an effective ground-level measurement campaign. Infrared cameras are effective at locating leaks, and their use has been required under a recently approved Colorado regulation. Low cost stationary detectors are also under development. The newest detectors can locate leaks and estimate their magnitude from a distance, which reduces the challenge of acquiring property owner permission that bedevils direct on-site measurement.

Comment: The current oil and gas boom has been unleashed by a wave of technological innovation (directional drilling, hydraulic fracturing, and other emerging techniques, like "acidizing"). Governments need to keep pace with faster innovation on the regulatory side. New monitoring technologies are an opportunity for greater accuracy, and the EPA should move

quickly to use these technologies to transform government monitoring of emissions. Better monitoring of emissions will help the EPA solve the mystery of the missing methane and provide the best objective guidance to policymakers, regulators, and society.

Commenter: Kerry Kelly
Waste Management

Comment: WM is pleased to see that the emissions from the waste sector and landfills in particular, continue to trend downward, while methane emissions control via gas collection and combustion in renewable energy projects or flares continues to grow. We also noted with interest, the discussion of planned improvements to measuring landfill emissions by replacing the default 10 percent oxidation with a more accurate, science-based estimate. It is this aspect of the Draft Inventory that is the subject of our comments.

The Agency refers to a growing body of peer-reviewed literature describing both field and laboratory studies that all indicate that the default oxidation value of ten percent understates the oxidation rates achieved at well-managed landfills. EPA's careful analysis of peer-reviewed literature and field measurement studies resulted in recent regulatory changes to estimating methane oxidation in landfill cover in the Greenhouse Gas Mandatory Reporting Rule (GHG MRR). These changes allow greater use of site-specific conditions rather than national default assumptions and will greatly increase the accuracy of landfill facility methane emissions estimation. We urge that the Agency also update its national inventory methods to reflect these changes, so that it can improve the accuracy and reliability of the U.S. GHG Inventory.

Comment: The EPA's Decision to Revise the Methane Oxidation Factor Used in the GHG MRR is Well Supported by Peer-Reviewed Science:

Numerous studies have been conducted worldwide and referenced in the scientific literature that address and document methane oxidation in cover soils, as well as gas collection efficiency. In 2009, The Journal of Environmental Quality published a comprehensive literature review. The paper references over 60 technical documents dating from 1960 to the present, with the majority of the papers being published in the 1990s and 2000s. Overall, based on review of 42 determinations of the fraction of methane oxidized in a variety of soil types and landfill covers, the mean fraction of methane oxidized across all studies was 36 percent with a standard error of 6 percent. For a subset of 15 studies conducted over an annual cycle, the fraction of methane oxidized ranged from 11 percent to 89 percent with a mean value of 35 percent + 6 percent, nearly identical to the overall mean.

In July 2007, the Solid Waste Industry for Climate Solutions (SWICS) released its first white paper titled Current MSW Industry Position and State-of-the-Practice on LFG Collection Efficiency, Methane Oxidation, and Carbon Sequestration in Landfills (White Paper). The public and private members of SWICS shared the White Paper with EPA as it developed the

GHG MRR requirements for evaluating and reporting MSW landfill emissions. In January 2009, SWICS updated the White Paper to incorporate additional studies noted above.

Since the release of the 2009 White Paper, a number of studies have been published in peer-reviewed literature, most notably an entire special issue of the journal Waste Management (2011) on Landfill Gas Emission and Mitigation sponsored by Consortium for Landfill Emissions Abatement Research (CLEAR). CLEAR is an International Waste Working Group (IWWG) Task Group, which focuses on landfill gas emission to the atmosphere. The group has members from 12 countries, across four continents. A number of papers in the special issue focus on the use of compost biocovers, bio-windows or permeable gas dispersion layers to treat and oxidize landfill gas in situ (Huber-Humer et al., 2011; Pedersen et al., 2011; Scheutz et al., 2011; Pawlowska et al., 2011; Dever et al., 2011; and Jung et al., 2011). Additionally, several papers in the special issue, Ranchor et al., (2011); Abichou et al., (2011) and Chanton et al., (2011b), examined the response of the methane oxidizing microbial community to methane loading to the cover soil. Two key papers, Bogner et al., (2011), and Spokas et al., (2011), describe recent work in California where field measurements of emission and oxidation were coupled with extensive modeling efforts. Chanton et al., (2011a) published the results of 37 seasonal sampling events at 20 landfills with intermediate covers over a four-year period. Abichou et al. (2011b) examined the best approach towards describing central tendencies in oxidation data and reported that the results were generally distributed normally so that mean values could be used.

In November 2012 SWICS, with the participation of Dr. Jeffrey Chanton of Florida State University and Dr. Morton Barlaz of North Carolina State University, finalized an addendum (2012 Addendum) to the Methane Oxidation section of the 2009 White Paper. The 2012 Addendum includes methane oxidation results from evaluations of 90 landfills as compared to the 47 published evaluations available in 2009.

In reviewing and incorporating the results of these peer-reviewed studies of landfill methane oxidation, the 2012 Addendum updated the 2009 White Paper results as follows:

1. Clay cover: The number of studies in clay cover increased from five in 2009 to 31 in 2012. The mean fraction of methane oxidized increased from 18 percent to 30 percent, while the median fraction oxidized increased from 14 to 29 percent.

2. Sandy soils cover: The number of studies in sandy soils doubled from eight to 16, with the mean oxidation value changing very little (55 to 54 percent) while the median value increased from 43 to 50 percent methane oxidized.

3. "Other" covers: The number of studies in "other" cover soils increased by nine and both the mean and the median fraction oxidized values increased slightly.

4. The overall mean oxidation value across all of the studies increased from 35 to 38 percent while the overall median oxidation fraction increased from 31 to 33 percent.

Comment: The SWICS 2012 Addendum Definitively Supports a Significant Increase to the Current Default Value of Ten Percent:

The 2012 Addendum concluded that the EPA default oxidation value of 10 percent underestimates typical methane oxidation and is not representative of expected methane oxidation at sites utilizing organic, clay, sand or other cover types. EPA derived the default value from one field study performed at one poorly maintained landfill with no gas collection system, Czepiel et al. Analysis of the 90 studies highlighted in the 2012 Addendum indicates that if a single value is considered for methane oxidation it should be between 33 and 38 percent.

The 2012 Addendum also examined methane oxidation as a function of methane loading to the cover layer of the landfill. Recent studies show that the percent oxidation is an inverse function of the rate of emission (Stern et al., 2007; Rachor et al., 2011; Chanton et al., 2011a,b). At lower emission rates, the methanotrophs in the soil cover can consume a larger portion of the methane delivered to them, potentially oxidizing 95 to 100 percent (Humer and Lechner, 1999, 2001a, Huber-Humer 2008; Powelson et al., 2006, 2007; Kjeldsen et al., 1997). As flux rates increase, the percent oxidation decreases and the methanotrophs can become overwhelmed with methane. Thus, as methane emission increases, percent oxidation decreases (Powelson et al., 2006, 2007).

A mathematical model of cover oxidation developed by Dr. Tarek Abichou of Florida State University (Abichou et al., 2010), demonstrates that at lower methane fluxes, oxidation rates are equal to the methane loading to the soil cover. Oxidation keeps pace with flux, and the soil cover is able to oxidize all of the methane coming from below. At lower loading rates, methane oxidation is equal to 100 percent. As flux increases, the cover is not able to oxidize all of the incoming methane, and the percent oxidation falls off. Therefore, percent oxidation starts to decrease as the methane loading to the cover increases. This relationship is shown clearly in the laboratory column studies of Rachor et al., (2011). Field studies have also confirmed this relationship between methane flux and percent oxidation (Chanton et al., 2011a, b). At low rates of methane emission, the percent oxidation is near 100 percent. As emission rates increase, the percent oxidation decreases. This analysis served to support the approach that EPA finalized for determining a more accurate methane oxidation fraction by calculating the methane flux rate for the landfill.

In addition to the 2012 Addendum, the landfill sector provided data for 262 private and public landfills reporting under Subpart HH. The dataset allowed the Agency to evaluate several possible options for determining more accurate methane oxidation fractions. The data conclusively showed that the average oxidation fractions for different soil cover types are all well above the default 10 percent value required by Subpart HH, and underpin the need for a revised default value or more refined method for determining an oxidation fraction at a site.

Comment: WM recommends that the Agency carefully consider its analysis underpinning its decision to estimate facility-level methane oxidation by calculating the methane flux rate and consider how that methodology could be used at the national inventory level. The work done by

the Agency in updating methods for facility-level calculation of methane oxidation will greatly improve the accuracy and reliability of emissions estimates for landfills. We urge EPA to endeavor to make similar improvements to its national inventory of emissions and sinks at landfills.

Commenters: Anna Moritz, Kevin Bundy, Sparsh Khandeshi,
Center for Biological Diversity and Environmental Integrity Project

Comment: We are concerned that the emissions factors and global warming potential for methane that are currently employed by EPA have resulted in a significant underestimate of U.S. anthropogenic methane emissions. First, multiple studies have reported far higher leakage factors from oil and natural gas operations than EPA currently uses. It is essential that EPA arrive at accurate numbers. In addition, even reported leakage numbers necessarily underestimate emissions because they omit undiscovered and unreported leaks, such as those by smaller operators. EPA should immediately commence on-the-ground data collection and, until the results from these efforts are available, account for these factors by presenting a range of likely underreporting. Second, the climate impact of methane is underestimated because the inventory reports normalized methane emissions using solely a 100-year global warming potential ("GWP") and an outdated value for the GWP.
Methane is an important component of climate strategies to avoid Arctic disaster and other catastrophic tipping points. Unlike other traditional greenhouse gases that have atmospheric lifetimes of a century or more, methane remains in the atmosphere for only about 12 years. This means that a reduction in emissions today will not only slow the increase in radiative forcing, but also result in actual decreases in radiative forcing in a short time – just over a decade. When we are considering how to address the collapse of the Arctic cyrosphere or avoid near-term tipping points, methane and other short-lived climate pollutants present an opportunity for rapid reductions in climate forcing.

Because methane mitigation is an important climate strategy, it is essential that the current emissions levels from US sources be accurately characterized. This includes both emissions factors for various industries and quantification using the most current values for global warming potential: a 100-year GWP of 34 and a 20-year GWP of 86.

Comment: Emissions factors from oil and gas operations should be revised:

There is compelling evidence that leakage rates from oil and gas operations are far higher than EPA emission factors suggest. For instance, Miller and colleagues recently used atmospheric measurements to estimate that actual methane emissions are about 1.5 times larger than EPA estimates, with fossil fuel methane emissions more than two times higher than estimated. Observations from oil and gas operations in Colorado indicate that inventories underestimate

methane emissions by at least a factor of two. Leakage rates over a Utah gas field were recently estimated at 6.2 to 11.7%, well above the rates assumed by national inventories.

Moreover, EPA's data for oil and combined oil/gas wells omit the impact of hydraulic fracturing. A recent white paper from Environmental Defense Fund summarizes findings from a number of studies to conclude that emissions factors used in EPA's current inventory underestimate methane emissions from oil wells that employ hydraulic fracturing.

Another major source of methane emissions from the oil and gas sector is leaks from pneumatic devices. A recent study calculated emission factors for pneumatic devices to find that national emissions from this source are likely at least twice the amount predicted using the emission factors in the US GHG Inventory. This is another area where improvement of emission factors is essential.

We urge EPA to consider the range of data available and update the emission factors that are used in the GHG inventory to accurately reflect methane emissions from both venting and leakage in the oil and gas industry. These data are critical as industry leaders and decision makers consider mitigation options.

Recent reports have also substantiated an alarming rate of leaks from decaying gas pipeline systems across the country, creating the need for systematic, on-the-ground data collection to obtain an accurate quantification of emissions from this source. For example, according to a recent study, the two distributors of natural gas in New York City and Westchester County reported 9,906 leaks in their combined system for 2012 alone, and gas distributors nationwide reported an average of 12 leaks per 100 miles of the 1.2 million miles of gas main pipes across the country. More than 5,800 leaks were detected from aging gas pipelines underneath the streets of Washington, D.C. These samples indicate that EPA's data are incomplete, and we urge EPA to note this fact and undertake the efforts necessary to provide an accurate accounting next year.

Comment: The GHG Inventory should quantify methane emissions using AR5 GWPs:

EPA recently finalized technical amendments to the Greenhouse Gas Reporting Rule. These changes included updating the methane GWP from the values in the IPCC Second Assessment Report to those in the Fourth Assessment Report ("AR4") for reporting in year 2015 and beyond. While this was an important improvement, we and other organizations joined Clean Air Task Force in recommending that EPA utilize the most up-to-date science and adopt the most recent methane GWPs from the IPCC Fifth Assessment Report ("AR5") as well as require reporting of both 100-year and 20-year methane GWPs.10 EPA declined to adopt the most recent estimates of methane's GWP because current international reporting requirements under the United Nations Framework Convention on Climate Change employ only 100-year GWPs and will begin using AR4 GWPs in 2015.11

While we understand EPA's need to comply with international reporting requirements, we renew our call upon EPA to consider updating the emissions reported in the U.S. GHG Inventory to

reflect the AR5 GWPs, as well as report normalized emissions using both 20-year and 100-year GWPs for methane. The US GHG Inventory is important domestically for both government and private-sector decision-making and analysis. This is a purpose separate from international commitments and requires more precise quantification of climate impacts. We appreciate the inclusion of Appendix 6.1 in the draft GHG Inventory, which provides emissions estimates as calculated with both AR4 and AR5 100-year GWPs. We ask that EPA make this information more prominent so that users will be more likely to find and employ the updated emissions estimates. Furthermore, it is important that EPA use the most accurate GWP for methane, which includes carbon cycle feedbacks.

We further request that EPA consider reporting emissions using both the 100-year and 20-year GWP for methane as this will allow the full consideration of climate consequences. The 100-year GWP gives a better sense of how reductions can influence long-term climate stabilization, while the 20-year GWP is useful when considering tipping points and near-term climate impacts. Furthermore, the AR5 values for GWP have changed substantially since AR4. The AR5 methane GWP of 34 is significantly higher than AR4 – 36 percent higher. The AR5 20- year GWP is 86 (19% higher than the AR4 GWP). These substantial increases in GWP mean that emissions data reported using AR4 GWPs or earlier are understated. Accordingly, EPA must revise the GWPs used in the inventory and ensure that they properly reflect carbon cycle feedbacks.

Comment: EPA Must Clarify Data Sources and Emissions from Biomass Electricity Generation:

According to the Draft Inventory, CO_2 emissions from woody biomass and woody biomass consumption (measured in trillion Btus) in the electricity generation sector increased nearly tenfold between 2011 and 2012. It is not clear, however, how these emissions estimates were derived. Although emissions of biogenic CO_2 associated with electricity generation are reported primarily for informational purposes pursuant to international accounting conventions, accurate emissions data are critical to evaluating domestic renewable energy programs and accounting for the actual climate consequences of increasing biomass energy generation.

The Draft Inventory states that biogenic CO_2 emissions from the electricity generation sector data were calculated using EPA's Clean Air Market Acid Rain Program dataset, while emissions from other sectors were obtained from EIA's Monthly Energy Review.[13] An annex to the Draft Inventory explains that "there were significant differences between wood biomass consumption in the electric power sector between the EPA (2013) and EIA (2013) datasets." Accordingly, "the electricity generation sector's woody biomass consumption was adjusted downward to match the value obtained from the bottom-up analysis based on EPA's Acid Rain Program dataset."

The increase in emissions between 2011 and 2012, if accurate, represents a dramatic expansion of emissions from this industry—nearly a full order of magnitude over the course of only one year. It is impossible to discern, however, whether the Draft Inventory's emissions estimates are either comprehensive or consistent.

The EIA Monthly Energy Review data used for other biomass emissions estimates does not show a similar increase in woody biomass consumption between 2011 and 2012; in fact, these data show a slight decline in both wood and other biomass "waste" consumption. The increase thus must be reflected, if anywhere, in EPA's Clean Air Market dataset. Again, however, this is impossible to discern because the full dataset does not appear to have been included or explained further in either the Draft Inventory or the annexes.

Indeed, it appears that the AMPD dataset may be under inclusive of electrical generation facilities using woody biomass as fuel. For example, a query performed on the AMPD website for 2012 emissions data from all programs and all facilities returned 4,828 records nationwide, only 23 of which list "wood" as the primary fuel source; CO_2 emissions from these facilities in 2012, where emissions were reported at all, totaled only about 2.7 million metric tons. There are, of course, more than 23 wood-burning power plants operating in the United States; indeed, there are more than 23 such facilities in California alone, although no California plants appear in the query report generated by the AMPD dataset.[18] Of course, if there are numerous biomass power plants that are not listed in the AMPD dataset, use of this dataset for a "bottom up" emissions estimate will likely underestimate emissions from this sector.

Given these apparent inconsistencies, EPA should clarify what data set it is using to estimate biogenic CO_2 emissions from electricity generation and should ensure that these data are inclusive and comprehensive enough to produce an informative report.

Comment: Conclusion:

In sum, we commend EPA for compiling and reporting extensive data from various sources of greenhouse gases within the United States. There remain, however, some areas where improvements are needed to maximize the utility of the GHG Inventory for both international reporting and informed domestic policy-making. First, emissions factors for the oil and gas industries, including pipeline leakage, are very likely much too low to accurately reflect fugitive methane emissions. Second, we request that EPA expand its reporting of methane emissions using both 20-year and 100-year GWPs as well as report methane emissions in the main text of the Inventory using the GWPs from AR5. And finally, we request that EPA clarify the sources and accuracy of data used to estimate emissions from biomass combustion, particularly for the electricity generation sector.

Commenter: Jeff Zimmerman
Damascus Citizens for Sustainability

Comment: Over the last several years it has become apparent that stray emissions of methane from gas development projects across the United States are increasingly contributing to the greenhouse gas levels and climate change. The purpose of our submissions today to your draft inventory document is to bring to your attention a number of recent (2012-2013) studies and reports providing actual measured emissions of stray methane from unconventional gas

development using fracking. The data collected and reported in these publications documents a range of additional scientific information that needs to be factored into the GHG emissions inventory and the resultant impacts of climate change.

Three of these reports document extensive methane leakage from natural gas distribution facilities in Manhattan, New York City, NY, (Payne and Ackley, March 2013) Boston, MA, (Philips et al., 2012) and Los Angeles, CA (Peischl et al., 2013). The LA Basin report documents methane leakage at 17% of total gas production in the LA Basin. Another report provides methane leakage data in a natural gas production area in Wyalusing Township in Bradford County, PA (Payne and Ackley, November 2013) and another report documents methane leakage in fracked gas production areas of Leroy, Granville, and Franklin Townships in Bradford County, PA (Payne and Ackley, 2012). A sixth report documents 4% methane leakage in the Denver-Julesurg Basin in Colorado (Tollefson, 2012), and a seventh report documents a 9% overall methane leakage rate from fracked gas development in the Uinta Basin in Utah (Tollefson, 2013). These reports seriously call into question the much lower methane leakage rates from fracked gas development estimated by EPA. A report by Miller and many others summarizes the results of these and other similar studies and concludes that actual methane leakage rates are almost five times the earlier EPA estimates (Miller et al., 2013). Each of the reports we are providing with this comment letter should be included in the EPA inventory of climate change and GHG data. The trend in these reports demonstrates that methane leakage from unconventional gas development is far greater than previously thought. A comprehensive reexamination of leakage rates and impacts is clearly required.

Commenter: Cynthia Finley
National Association of Clean Water Agencies (NACWA)

Comment: The wastewater treatment category includes publicly owned treatment works (POTWs), septic systems, and industrial wastewater treatment systems. Although the emissions are much smaller in magnitude than for the highest ranked categories, the broadly-based wastewater category consistently ranks in the top ten emitters for nitrous oxide and methane emissions in the U.S. NACWA's review focused on emissions from POTWs, which are a fraction of the total wastewater treatment category emissions.

The emissions from POTWs in the 2012 Inventory are essentially the same as those in the 2011 Inventory, with some clarifications added to the text. NACWA's comments on the 2011 Inventory requested that all values used in the equation to calculate emissions be provided to enable the calculations to be easily reproduced. NACWA appreciates the response to this request with the addition of Table 8-15, which provides the values for the variables used in calculating the nitrous oxide emissions for 2012 and previous years.

Comment: NACWA agrees with the additions made to the Planned Improvements section and encourages EPA to investigate additional data sources as soon as possible. Since the 2008 Clean Watershed Needs Survey (CWNS) is not detailed enough to be used in the Inventory and the 2004 CWNS data is likely outdated, additional data sources are necessary to ensure the accuracy

of future Inventories. NACWA supports EPA's investigation of the data available at www.biogasdata.org and from ongoing research in the U.S. and abroad. However, NACWA also urges caution in using results from studies that were not designed to produce nationally-applicable results. Relying on studies that are not representative of utilities nationwide may actually increase the uncertainty of the estimates. NACWA agrees that EPA's plan to review inventories from other countries for additional data and methodologies may be useful, as long as any information used is directly applicable to wastewater treatment processes in the U.S.

Comment: As NACWA has explained in comments on the Inventory in previous years, the Association believes that the nitrogen loading rates for N2OEFFLUENT are sourced incorrectly and that using information from the existing National Pollution Discharge Elimination System (NPDES) database will yield more accurate and justifiable loading rates. The NPDES permitting program represents long-term, nationwide facility performance which would allow emissions estimate projections over the time series represented in the Inventory. If EPA decides not to investigate its own databases, the average nitrogen loading rate of 15.1 g N/capita-day1 represents the industry standard and is supported by a wealth of data widely confirmed in U.S. practice, as explained in our previous comments and supported by data collected by NACWA from 48 U.S. POTWs. This result represents all domestic sources of nitrogen, the use of other nitrogen-containing compounds, and both residential and commercial sources.

Comment: Outside of the Wastewater Treatment section, the Inventory's Executive Summary and Introduction should state more clearly that the Inventory's purpose is for information, not regulation. EPA should ensure that all of its offices understand the purpose of the Inventory and recognize that the Inventory's industry-wide methodologies are largely inadequate for facility level emissions, such as those required by EPA's Greenhouse Gas Reporting Rule and the Clean Air Act Title V and Prevention of Significant Deterioration (PSD) permitting programs.

Commenter: David McCabe
Clean Air Task Force (CATF)

Comment: Methane from Petroleum and Natural Gas Systems:

In our January comments on the Expert draft of the inventory, we raised a number of issues that we summarize here. Although EPA has noted most of the issues we raised in the discussion text of the public draft inventory ("Draft Inventory"), the emissions estimates in that version have not been substantially modified from the expert draft inventory. Consequently, the inaccuracies we identified remain in the inventory estimates. As such, we re-confirm our January comments, which we have attached to this document for your convenience, with some updated figures, and have made additional specific suggestions about how EPA might handle identified inaccuracies in the draft inventories here.

We raised three principle issues in the January comments: Emissions from completion of oil wells with hydraulic fracturing (HF), emissions from completion of gas wells with HF, and emissions from pneumatic controllers (PCs).

In addition we raise two additional issues in these comments: Methane from venting of associated gas from oil wells (during production), and general comments about the approach taken to tabulating emissions in the Inventory.

Comment: Emissions from completion of oil wells with hydraulic fracturing. EPA continues to use a very low emissions factor for oil well completion – 733 scf per completion – that pre-dates the use of HF on oil wells. Since most oil wells are now hydraulically fractured, and the use of HF substantially increases potential emissions per well (just as for natural gas wells), the use of this ancient emissions factor for all oil well completions substantially underestimates actual emissions.

We reference the recent comments from Environmental Defense Fund (EDF) on the Draft Inventory. EDF reviewed data from a number of sources that show that both potential and actual emissions from oil well completions after hydraulic fracturing can be hundreds of times greater than the 733 scf per well completion EPA uses in the Draft Inventory. EDF recommends that EPA use analysis of EPA's Greenhouse Gas Reporting Program (GHGRP) data for reported emissions from well completions in oil-bearing formations. This analysis finds that reported actual cumulative emissions (including wells where gas was vented, wells where gas was flared, and wells where gas was captured into pipelines, during flowback) were an average of 6.2 metric tons of methane per completion or recompletion, based on reports on 1,754 completions and recompletions. We support EDF's recommendation that EPA use this data to revise the estimates for well completion of oil wells for wells that use hydraulic fracturing, EDF's suggestion that oil well completion emissions be reported with sub-categories for wells with and without hydraulic fracturing, and EDF's suggested approach for estimating the number of oil wells that use hydraulic fracturing.

Estimating methane emissions from oil well completions in this manner would clearly be more accurate than EPA's current method. EPA must promptly address this rather manifest inaccuracy in the final 2014 inventory. If EPA is unable to provide a more accurate estimate of emissions from oil wells completions in the final 2014 inventory, a statement directly noting this issue is warranted. We suggest adding the following to page 3-55 (suggested additions in bold):

-line 11: "…increase again with the widespread use of hydraulic fracturing in tight formations."
- After the period on line 13. "Note that the inventory methodology has not been updated to reflect emissions during well completion or re-completion after hydraulic fracturing, and thus the inventory likely underestimates emissions from this source."

Comment: EPA has revised the methodology for estimating emissions of methane from completion of gas wells. As in our January comments, we generally support this revision, as the revised data appears to be based on more robust data and the result is much more transparent. However, as we noted in our January comments, EPA's methodology is flawed because it fails to account for the significant fraction of gas well completions at facilities that do not report data to

the GHGRP. Thus, the activity data reported in table A-126 of the inventory is clearly an underestimate of actual activity. As we suggested earlier, EPA should use data from state databases or commercial databases, compared to reports to GHGRP, to calculate the fraction of wells that are owned by firms that report data to the GHGRP, and then adjust the activity data from GHGRP using this fraction, to get a more accurate estimate of the number of completions occurring nationwide. The suggested approach is described in somewhat more detail in our January comments, as EPA mentions in the Public Draft Inventory (p 3-71, lines 1-3).

Although adjusting the GHGRP to a proper estimate of national activity is not trivial, the current figure is clearly an underestimate of national emissions and we believe EPA would set a poor precedent by using unadjusted data in the Inventory. If EPA chooses not to adjust the GHGRP data, as suggested or by some other approach, EPA should acknowledge in the inventory that the issue exists. Currently, in the Public Draft, this issue is not mentioned directly, but rather is indirectly referred to (in response to CATF comments) under "Well Counts and Completion and Workover Counts" under "Planned Improvements." It should be raised with a statement to the effect of, "This methodology undercounts emissions from completions and workovers with hydraulic fracturing, to the extent that it undercounts completion and workover events, because not all well facilities report emissions and activity data to GHGRP." This statement belongs in either the completions text in the QA/QC section, or the completions text in the Recalculations section. Additionally, it would be a great example to list under Uncertainty and Time Series Consistency. For example, starting on line 35 of p 3-66,

> The IPCC guidance notes that in using this method, "some uncertainties that are not addressed by statistical means may exist, including those arising from omissions or double counting, or other conceptual errors, or from incomplete understanding of the processes that may lead to inaccuracies in estimates developed from models." An example would be the probable undercount of completion and workover events with hydraulic fracturing (see below). As a result, the understanding of the uncertainty of emissions estimates for this category will evolve and will improve as the underlying methodologies and datasets improve.

Comment: Pneumatic Controller emissions:

As described in our January comments, GHGRP data shows significantly higher emissions from pneumatic controllers (PCs) than the Draft Inventory reports. Since the GHGRP uses emissions factors derived from EPA/GRI (1996), as does the Inventory, the apparent difference between the two is in device counts. Although the GHGRP clearly undercounts devices (by not capturing all wellpads, or any emissions from gathering), it must be more accurate than the current activity figures used in the Inventory. As noted in our January comments, since not all wellpads report under the GHGRP, it shows that total emissions from oil and gas wellpads were, at a minimum, 861 Gg methane in 2012. The Public Draft reports emissions from both Gas Production and Oil Production of 692 Gg methane, less than was reported in the Expert Draft (787 Gg methane), so this gap has widened significantly. While we recognize that updated activity data for 2012 may increase the figures in the Inventory, relative to the Public Draft, we anticipate that the gap

between the Inventory figure and the GHGRP figure will remain: for 2011, the Public Draft Inventory reports emissions of 752 Gg methane from oil and gas production, while the GHGRP data shows 835 Gg methane. Again, this is troubling because the GHGRP only covers a portion of the facilities that the Inventory is intended to cover yet its emissions figures are higher than the Draft Inventory.

As mentioned in our January comments, the gap would be much larger if the more accurate emissions factors from the Allen et. al (2013) study were used in place of the GHGRP emissions factors. The data available today suggests that the Allen et al. emissions factors are the best available today. As described in our January comments, correcting the GHGRP with the Allen et al. emissions factors produces a national methane emissions estimate of 1,140 Gg, even without scaling emissions up to reflect production facilities that do not report to GHGRP.

Thus it appears that both the activity data and emissions factors used in the inventory for PCs are not the most accurate data available. If EPA cannot use the more accurate, recent data we have suggested here, EPA should continue to note that data (as is done in the Public Draft) and commit to examining this data in the coming year.

Comment: Venting of Associated Gas from Oil Wells:

It appears that the Inventory underestimates venting from oil wells by a substantial amount. To our understanding, venting of associated gas from oil wells during production (i.e., casinghead gas venting) is listed in the inventory as "Stripper Wells" under Vented Emissions in Petroleum Production, and is listed as 14.2 Gg methane for 2012. Last year's inventory listed the same value for 2011.

GHGRP data shows much higher emissions of methane from "Associated Gas Venting and Flaring." For 2011, 175 Gg methane emissions were reported to GHGRP; for 2012, the figure was 90 Gg. Some of this is due to emissions of methane from flares, due to incomplete combustion in the flame. This portion of the methane emissions can be accurately estimated, by comparing CO2 emissions from associated gas venting and flaring to methane emissions from that source. As described below, CATF analyzed the GHGRP data in this way, finding that 60% to 90% of the GHGRP methane emissions from associated gas venting and flaring are due to venting, and thus the 14.2 Gg methane figure in the Draft Inventory is significantly too low.

We compared the emissions of CO2 and CH4 reported from each facility reporting "Associated Gas Venting and Flaring" emissions to the GHGRP, for both 2011 and 2012. The GHGRP uses a default factor of 2% for emissions of methane from flares, due to incomplete combustion (40 CFR Part 98.233(n)(1), Eq. W-19). Using this factor, we subtracted away the maximum methane that could be due to incomplete combustion in flares from each individual facility report. To be conservative, we also considered a case where the factor for incomplete combustion for methane in flares was 5%, in case some facilities used this higher factor to calculate their emissions.

In either case, many facilities have methane emissions well in excess of that due to incomplete combustion, and this "excess methane," which is due to venting of associated gas, is significant at the national level. In 2011, vented emissions, calculated with the 2% emissions factor, were 157.5 Gg methane; in 2012 they were 60.1 Gg CH4. Raising the incomplete combustion factor to 5% drops these figures somewhat, to 154.0 Gg CH4 and 56.3 Gg CH4, respectively. This analysis of the GHGRP data shows that venting of associated gas from oil wells is much higher than the 14 Gg reported in the Draft Inventory.

Comment: General Comments on the Organization of the Inventory for Oil and Gas:

As new data emerges on emissions from oil and gas facilities, it is very important that EPA use that data in the most appropriate way in the inventory. In some cases it may not be best for EPA to force the data into the framework used in the inventory, particularly where a) oil and natural gas production are separated and b) natural gas production is broken down, for almost every individual source, into NEMS regions.

It is appropriate that EPA has not developed emissions factors for each NEMS region for well completion and workover with HF, and it greatly improves the clarity of the inventory that these emissions are presented separately in Table A-126. We believe that EPA should have taken the same approach last year when updating the methodology for estimating liquids unloading (LU) emissions. The report submitted by API and ANGA on LU emissions did not recommend developing distinct emissions factors for each NEMS region for wells that vent during LU with and without plunger lifts. Instead, API and ANGA concluded it was more appropriate to estimate national emissions by applying their entire dataset to national activity drivers. EPA, in calculating emissions for each NEMS region, concludes that national LU emissions were substantially lower than API and ANGA concluded. Moreover, the calculated emissions factors for LU wells vary tremendously between NEMS regions that are not designed to capture differences in geology, age of wells, or anything else that might affect LU emissions. The NEMS region emissions factors are simply not credible.

EPA should use the national emissions factor approach used for completion / workover emissions for LU.

Additionally, when data from the GHGRP is superior to other available data, EPA should use that data, even if it does not readily allow separation of emissions between the oil production and natural gas production sectors. As EPA has recognized in, for example, GHGRP Subpart W and NSPS Subpart OOOO, these sectors are really one industry, and the distinction between the two is necessarily arbitrary. At present the Inventory reports that over 60% of emissions from PCs are from oil production, so it may be more appropriate to simply list PC emissions under oil production, with the "included elsewhere" designation for PCs under gas production.

Finally, we comment here on the Draft Inventory's discussion of Methane Measurement Studies (p. 3-71). First, we note that the Brandt et al. study mentioned in this section is quite specific that

emissions from oil and natural gas, specifically, were underestimated in the 2013 Inventory (which reported higher emissions than the Draft 2014 Inventory. Quoting from Brandt et al.: "We find … measurements at all scales show that official inventories consistently underestimate actual CH4 emissions, with the NG and oil sectors as important contributors…" (emphasis added). The title of Brandt et al.'s paper is "Methane Leaks from North American Natural Gas Systems." The Draft Inventory's over-generalization of Brandt et al.'s findings must be corrected.

EPA asks for input on how information from top-down ambient studies should be used to update the inventory. One non-quantitative way that EPA should use this information is to put the bottom-up inventory in context. The bottom-up inventory is essential for understanding the specifics of GHG emissions so that mitigation priorities, for example, can be examined. However, the bottom-up inventory clearly does not capture all emissions from oil and gas operations. Scientifically, it is not clear that the emissions from the sector as a whole are most accurately measured with the bottom-up measurements available to date. As such, it may be appropriate to calculate the leak rate from the entire industry (for life-cycle analysis, for example) using different methodologies. Separately, as ambient studies continue and techniques are developed, they will illuminate sources that must be reexamined in bottom-up studies. For these reasons, the top-down methodologies are strongly complementary to the bottom-up approach.

It would be appropriate for EPA to mention, in this section, the value of top-down studies in providing independent data on overall emissions from the industry, and on identifying specific potential issues in the inventory.

Comment: Emissions Data for Wood Biomass Combustion:

CATF requests that EPA provide a clearer explanation of the data on CO2 emissions from wood consumption reported in section 3.10 of the Draft Inventory, particularly the data that are reported for electricity generation units (EGUs). EPA describes the approach it used to determine the amount of CO2e emitted by EGUs that combust woody biomass in the Methodology passage at 3-79 of the Draft Inventory, but it is difficult—if not impossible—to replicate the results that EPA achieved using the database referenced by the Agency.

According to CATF's understanding of the Methodology passage at 3-79, EPA has determined that the Acid Rain Program's "bottom-up" data for woody biomass consumption by EGUs are better than the EIA Monthly Energy Review data for those same sources. EPA made the same determination in the 2013 US GHG Inventory of Emissions and Sinks, but neither the 2013 Inventory nor the 2014 Draft Inventory explains the Agency's preference for the Acid Raid data. (Id.; 2013 Inventory at 3-79). The lack of an explanation is particularly problematic because, notwithstanding its concerns about the EIA data for woody biomass consumption by EGUs, EPA considers EIA's national estimate for total woody biomass consumption to be accurate. (2014 Draft Inventory at 3-79). In any event, EGU biomass consumption data for 2012 is lower in the

Acid Rain dataset than it is in the EIA dataset. So in order to use both the Acid Rain data for woody biomass consumption by EGUs and the EIA data for total national consumption, EPA adjusts the consumption/emissions data for the other three sectors (Industrial, Residential, Commercial). (Id). It appears that that EPA calculated the difference between the EIA data and the Acid Rain data for woody biomass consumption by EGUs and then proportionally allocated that difference to those other sectors' EIA scores.

Assuming that is in fact how EPA determined the woody biomass consumption and emissions data reported in Tables 3-55 and 3-56, CATF was unable to reproduce the reported results for the Electricity Generation sector using the ampd.epa.gov database — which in turn prevented us from assessing the reported results for the other three sectors. We note, however, that the reported EGU emissions for 2012 (21.0 Tg CO2e) are an order of magnitude higher than they were in 2008 (2.8 Tg CO2e), 2009 (2.4 Tg CO2e), 2010 (2.6 Tg CO2e), and 2011 (2.4 Tg CO2e). (Draft 2014 Inventory at 3-78, Table 3-55). We also note that emissions from EGU consumption of woody biomass reported during earlier years (e.g. 2008-2011) appear to be too low when compared to emissions data that CATF received in response to queries at ampd.epa.gov. Finally, CATF notes that Acid Rain database appears to be significantly under-inclusive. For example, when CATF queried the database for national CO_2 emissions from EGUs that combust "wood" and "other solid fuel," the result was comprised of emissions from only a handful of facilities located in just five states.

Comment: Discussion and Presentation of Global Warming Potentials (GWPs) from IPCC's Fifth Assessment Report in the introduction and Annex 6.1:

In the Draft Inventory, EPA has presented the GWPs from IPCC's Fifth Assessment Report (AR5) incompletely. We commend EPA for committing to using the GWPs from the 2007 Fourth Assessment Report (AR4) in next year's inventory, in compliance with UNFCCC guidelines. However, the more recent AR5 GWPs are now considered more accurate, and it is important that EPA let readers know about these updates. The material presented in the Introduction, and in Annex 6.1, does not accurately report what AR5 reports for GWPs, and the problem is particularly acute for methane from "fossil" sources such as coal, oil, and natural gas.

For all GHGs, AR5 reports two GWPs. For one, the climate carbon feedback ("cc-fb") effects are included when the radiative forcing from the target gas (the non-CO2 GHG) is calculated; for the other GWP, the cc-fb are not included in this calculation. However, GWPs are calculated relative to the radiative forcing caused by CO2, and the cc-fb is included for the calculation of radiative forcing from CO2 in all GWP calculations. That is, when the GWP for methane is calculated "without the cc-fb," the radiative forcing for methane without the cc-fb is compared to the radiative forcing for CO2 with the cc-fb. For this reason, IPCC states that it is likely that the GWPs with the cc-fb included are more accurate. (See page 731 of AR5). As such, the Draft Inventory, which only presents AR5 GWPs without the cc-fb, (Draft Inventory at 1-9, Box 1-2) is not presenting the most accurate information to readers.

Furthermore, specific to methane, EPA should also include the GWPs that IPCC calculates for fossil methane. The table on page 1-9 omits this. There is no reason not to include the GWP for fossil methane in the table. While the issue was not properly handled in earlier editions of IPCC Assessment reports, it is a simple matter: CO2 from the oxidation of fossil methane is additional CO2 in the climate system, whereas CO2 from oxidation of biogenic methane is not. EPA must report the best available scientific information. Consistency with earlier, less accurate IPCC reports, is not an acceptable reason to leave this information out of the Table. After all, it is just an informational table. However, many users will read this table to read that AR5 says the GWP of CH4 is 28, period. The notes at the bottom of the table are not sufficient.

In summary, a line should be added for fossil methane. It would have no entries for SAR, TAR, or AR4, so it would be clear that it is new, and that there is no analogue in the previous ARs. Secondly, an additional column with the AR5 GWPs with the cc-fb included is needed.

Likewise, Annex 6.1 is quite helpful, but it appears to not even acknowledge the GWPs in AR5 calculated with the cc-fb included, nor the separate GWPs for fossil methane. Thus, it is not accurately using the recommendations of AR5.

For example, natural gas, petroleum, coal mines & abandoned coal mines, stationary & mobile combustion, petrochemical and iron/coke production together account for 43% of US methane emissions. Thus, if using the GWPs without the cc-fb included, the correct change to methane emissions (in CO2e) for AR5, relative to AR4 (table A280), would be (5 * 0.43 + 3 * 0.57) / 25 or 15.4%, not the 12% reported in table A280. As mentioned above, AR5 says that it is likely that the values with the cc-fb included are more accurate, so the more accurate GWPs are actually 34 for biogenic methane and 36 for fossil methane. Therefore the most accurate value for the change to methane emissions (in CO2e) for AR5, relative to AR4 (table A280) would be (11 * 0.43 + 9 * 0.57) / 25 or 39%. That's a significant difference, and ignoring all of these other values for GWP does a real disservice to readers of this section.

Therefore, tables A276, A280, A281 should be updated to use the fossil methane GWP for those sources, and to discuss and show the differences if the GWPs with cc-fb included (for all non-CO_2 GHGs) are included.

Comment: Minor Suggested Corrections / Clarifications (by page number)

Page ES-14, Line 13: Strike "observed." The drop in emissions is inferred, largely from changes in activity drivers.

Page 1-6: Suggest the following addition (in bold):

> "Tropospheric ozone is produced from complex chemical reactions of volatile organic compounds and/or methane mixing with NOx in the presence of sunlight."

Page 1-9 Box 1-2: In the GWP discussion in the ES, the importance of forcing by secondary products of primary forcers is mentioned. Updates to the calculations of forcing by secondary products is an important factor in the changes in GWPs in AR5. Suggest the following addition (in bold):

> "In the AR5, the IPCC has applied an improved calculation of CO2 radiative forcing and an improved CO2 response function in presenting updated GWP values. IPCC also applied updated calculations of indirect radiative forcing for some gases. Additionally, the atmospheric lifetimes of some gases have been recalculated, and updated background concentrations were used. In addition, the values for radiative forcing and lifetimes have been recalculated for a variety of halocarbons, which were not presented in the SAR."

Page 1-10 Very end of section 1.1, Add this sentence (copied from ES):

> "The use of IPCC AR4 GWP values in future year inventories will apply across the entire time series of the inventory (i.e., from 1990 to 2013) in next year's report."

Page 3-70, line 1: the correct table reference is Annex Table A-135

Page 3-71, line 16: The Brandt et al study is not listed in the References section. Also, please make an effort to provide a URL, whenever possible, to all of the documents listed in the References section, particularly EPA documents.

Tables A125-A130 reference a number of documents not listed in the references section on pp A200-A202. EPA should attempt to get as many of the memos and other documents listed as references onto the website, and provide hyperlinks to those documents in the references section!

Table A125: the EF for Liquids Unloading w/o plunger lifts for region MC is messed up (it is written as 190,17 scfy/well, so either a digit is missing or the comma is in the wrong place.

There are no references listed for the petroleum section. (And a lot of other sections. Maybe those works are cited at the end of Annex 3?)

Commenter: Darren Smith
Devon Energy

Comment: Due to our position as an early-adopter of reducing emissions from production processes, Devon holds unique knowledge about the processes involved and the physical phenomena that shape emissions for hydraulically fractured wells. It was this expertise and knowledge – and the resulting discovery that EPA's previous estimates for methane emissions from the flowback of hydraulically fractured wells were heavily inflated – that led Devon to take an active role in encouraging EPA to refine the previously adopted emission factor for hydraulically fractured wells. It is this same expertise that leads Devon now to commend EPA

for the proposed changes in the 2012 Draft GHG Inventory. The proposed changes to this year's inventory have brought the inventory one step closer to an accurate and complete emissions profile for the natural gas production sector.

Comment: Devon supports the use of the Greenhouse Gas Reporting Program for this emission factor, replacing the previous estimate based on Natural Gas Star Data, which was not fit for the purpose of establishing emission factors. Devon has provided numerous comments criticizing the previous emission factor for methane emissions from the flowback of hydraulically fractured wells. The crux of the criticism was that Natural Gas Star recovered volumes were used as a proxy for emissions from vented well completions. Natural Gas Star data is not fit for emission factor determination. This new method drastically improves accuracy of the factor, and can form the basis for making adjustments to the inventory in the future, as industry technology continues to reduce emissions in the oil and gas sector. While there is still room for improvement due to the GHGRP's use of the choke flow calculation methodology, the change represents a significant improvement in accuracy.

More importantly, through the adoption of net emission factors, EPA provides a framework by which future greenhouse gas reporting rule results can be used to continually refine the emission factors for methane from hydraulically fractured well completions. As the greenhouse gas reporting rule further refines its calculation and reporting methodologies, and as industry improves its technology and practices to further reduce emissions, the emission factors for methane from well completions can and should be adjusted accordingly. Given that net emissions factors will closely mimic GHGRP data, updates to the emission factor can be easily automated, so that an accurate emissions profile can be captured each year. Finally, creating net emission factors that more closely match the GHGRP data will provide the public confidence in the accuracy of this particular data program.

This potential, and the ability for the public to verify greenhouse gas reporting program data, provides transparency to the method by which the factors are determined. This allows policy makers and the public to better understand the different emission profiles for different equipment configurations, and for the federal and state governments to make policy decisions based on accurate data.

Commenter: Erica Bowman
America's Natural Gas Alliance

Comment: ANGA appreciates the changes EPA has made in developing the 2014 Draft GHG Inventory, which incorporates new data sources and methodologies that more accurately reflect actual emissions. These changes include the establishment of technology-specific emissions factors for wells with hydraulically fractured completions and workovers. We encourage EPA to continue upgrading the GHG Inventory with net emission factors in place of potential emission factors as more data become available. We would also support further sub-categorization to recognize the differences between hydraulically fractured completions and hydraulically fractured workovers.

Comment: For the past several years, ANGA has submitted comments on EPA's Draft GHG Inventories. Included in those comments were concerns that EPA has overestimated emissions from natural gas production activities, particularly emissions associated with the liquids unloading, and well completions and workovers. In the 2013 GHG Inventory, EPA adjusted the methodologies for estimating the frequency of well re-fracturing and emissions from liquids unloading. These changes contributed to a reduction in estimated 2010 Field Production emissions from Natural Gas Systems of 54 percent. ANGA supported these changes, which more accurately accounted for actual field practices.

In the 2014 Draft GHG Inventory, EPA adjusts the methodology for completions and workovers with hydraulic fracturing. These adjustments establish four technology-specific emissions factors for wells with hydraulically fractured completions and workovers: (1) hydraulic fracturing completions and workovers that vent; (2) hydraulic fracturing completions and workovers that flare; (3) hydraulic fracturing completions and workovers with Reduced Emission Completions (RECs); and (4) hydraulic fracturing completions and workovers with RECs that flare. These emissions factors are based on data submitted to EPA under the 20 II and 2012 Greenhouse Gas Reporting Program (GHGRP) Subpart W. Compared to data used in the 2013 GHG Inventory, the GHGRP data shows a higher percentage of hydraulically fractured well completions and workovers using RECs, a higher percentage of hydraulically fractured well completions and workovers that flare, and fewer emissions per hydraulically fractured completion and workover that vented. We believe that the adjustment to the emissions factor for hydraulically fractured well completions and workovers that vent is closer to representing actual emissions. The GHGRP data used by EPA support ANGA's long-held contention that EPA's estimate that 9,000 thousand cubic feet (Mcf) of natural gas is released per uncontrolled well completion is flawed due to its reliance on data from the Natural Gas STAR program.

Comment: Although the new emission factors for uncontrolled well completions better represent actual industry practices, they remain higher than measured results from the recent study by researchers at the University of Texas-Austin and supported by Environmental Defense Fund (UT Austin/EDF study). At 41 metric tons (MT) methane per vented well completion, for example, the estimate in the Draft 2014 GHG Inventory is within one order of magnitude of the range found for similarly configured completions in the UT Austin/EDF Study, which found a range of 0.5-4 MT methane per completion event for those wells vented directly to atmosphere. Much of this difference can be attributed to the choke flow calculation methodology option in the GHGRP. The choke flow calculation methodology was not designed for use in multi-phase flow applications, and as such can often deliver erroneous results when compared to direct measurement. ANGA encourages EPA to remove outlier data from the emission factor calculation and use only measured data in the GHGRP for the calculation of emission factors, not data derived from the choke flow equation methodology.

Comment: As noted above, ANGA supports the use of GHGRP data to establish emission factors and strongly believes that EPA should continue using this data source to refine the emission factors for hydraulically fractured well completions and workovers. As industry

technology and practices improve to further reduce methane emissions and the GHGRP continues to update its calculation and reporting methodologies, the emission factors for hydraulically fractured wells and completions should be adjusted accordingly. In addition to improving the accuracy of the GHG Inventory which is a common goal of both EPA and the natural gas industry, creating emissions factors that more closely match the GHGRP data will provide public confidence in and increase uniformity across EPA's data programs.

While ANGA continues to believe that EPA's estimate of the number of uncontrolled well completions and workovers is too high, we understand that this number will decrease significantly in future years as the 2015 and later GHG Inventories will factor in the REC and completion combustion device requirements included in the Oil & Gas New Source Pollution Standards. This rule requires the use of RECs for almost all completions and workovers after January 1, 2015 and required flowback emissions to be routed to a completion combustion device starting in October 2012. As a result, the 2015 GHG Inventory, which reports estimated emissions from 2013, should have significantly lower emissions from these activities.

Comment: In response to EPA's request for input on the assumptions regarding the historic use of RECs, we support the recommendations made by Devon in its comments on the expert review draft and public review draft of the 2014 GHG Inventory. As EPA considers other changes to the inventory, we would support sub-categorization of pneumatic controllers to high bleed, low bleed, and intermittent categories and the use of appropriate actual emission factors for each category using GHGRP data, the UT Austin/EDF study, and other recent and upcoming studies.

Comment: Given the magnitude of the changes that the Agency has made over the past four years both increasing and decreasing estimated emissions from natural gas production, the underlying data and assumptions must be rigorous and well supported. ANGA appreciates the changes EPA has made to its methodology for estimating emissions from liquids unloading, its estimate of the frequency of work overs, and its methodology for hydraulically fractured well completions and workovers. We encourage EPA to continue updating its methodology and emissions factors with technology- and region-specific emissions factors based on valid data, assumptions and calculations. However, given the underlying uncertainties of the current data, ANGA does not support the use of the emissions estimates presented in the GHG Inventory as the basis for any analysis or regulatory action.

Commenter: Karin Ritter
American Petroleum Institute

Comment: General:

API supports the changes made to the 2012 U.S. GHG Inventory including the advances made in updating the national emission estimation methodology and increased use of site specific industry data that is becoming available through the Greenhouse Gas Reporting Program (GHGRP). When accounting for these changes the resulting non-combustion emissions from

Natural Gas Systems are estimated to be 162.3 million metric tonnes of CO2e (CO2 - 35.2; CH4 - 127.1). This represents a 1.07% of natural gas withdrawals for 2012. API encourages EPA to state this clearly early in the discussion on Natural Gas Systems to enhance understanding of the data by potential users.

Comment: General:

For this Public Review of the draft 2012 national inventory, API is providing comments regarding emission estimation for Petroleum Systems and Natural Gas Systems. Our comments reiterate some of the discussions on recalculations that were part of the U.S. GHG Inventory expert review phase and also point out areas for future collaboration where EPA is planning future improvements. API supports further review and analysis of the GHGRP data with the overarching goal of ensuring the quality and validity of data being used for deriving new national emission factors.

In addition, results from on-going GHG emission studies are expected to be published this year, and API is willing to continue its collaboration with EPA to incorporate relevant new information in the 2012 U.S. GHG Inventory and beyond.

Comment: General:

API supports the continued disaggregation of emission source information and, if applicable, emission reductions, to provide better transparency for "net" emissions for each source type. The approach historically used by EPA of lumping together reduction activities for multiple inventory sources made it difficult to attribute these reductions to specific inventory source categories. Emission reductions reported for "Other Production", "Other Processing", "Other Transmission" and "Other Distribution" in Table A-135 are larger than those shown in the Expert Review Draft and provide less transparency about the sources of these emission reductions.

Comment: General:

Where appropriate for the source category, API supports the continued use of data reported through the GHGRP and other relevant "bottoms-up" studies to develop "net" emission factors for specific source categories. API also recommends that EPA recalculate "net" emission factors for relevant source categories on an annual basis, using the GHGRP data and any relevant new "bottoms-up" studies, for each successive inventory in order to reflect changes in emissions due to expanded regulatory and voluntary reductions. This allows EPA to highlight, in the U.S. GHG Inventory, changing operating practices due to regulatory requirements being phased in by the petroleum and natural gas sector over the next few years.

Comment: General:

API advises EPA to carefully analyze and screen GHGRP reported data to identify data outliers and enable verification and/or correction or exclusion of suspect data entries and prevent the use of incorrect data in the derivation of emission factors (EFs). As discussed previously with EPA, during the Expert Review phase, the GHGRP data may potentially include incomplete or incorrect data due to ambiguity in implementation of approved EPA procedures, errors in applying the GHGRP calculations, faults in data aggregation and reporting, and partial reliance on Best Available Monitoring Methods (BAMM). Despite these discussions and detailed analysis provided to EPA to highlight the impact of erroneous data and outliers it seems that EPA did not modify their calculations published in the Public Review version of the 2012 inventory now under consideration.

Comment: Petroleum Systems Emissions:

Page 3-54 and Page 3-55. Editorial Comment:

API has noted a redundancy in the text presented in rows 28-32 of page 3.54 with rows 10–13 of page 3.55.

Comment: Petroleum Systems Emissions:

Page 3-59. Recalculation Discussion: Accounting for Voluntary Emission Reductions:

Under its recalculation discussion EPA seeks comment on its update to the Petroleum Systems section to include Natural Gas Star reduction data. EPA has added an accounting for voluntary emission reductions to the CH4 emissions from Petroleum Systems, and it indicates that this is from reassigning reductions that were previously included under the Natural Gas Systems (as referenced on page 3-70).

API supports this change but notes that the reductions attributed to the Petroleum Systems lacks the level of transparency that was previously provided for Natural Gas Systems. To address this, API recommends that Section 3.6 for Petroleum Systems in the annex should include a table that is equivalent to Tables A-135 and A-136 in the Natural Gas Systems.

Comment: Petroleum Systems Emissions:

Page 3-59. Planned Improvements Oil Well Completions and Workovers:

EPA is discussing its planned improvement to the U.S. GHG Inventory for oil production to allow for differentiation between completions with and without hydraulic fracturing. EPA is seeking comments on the topic as part of its future improvements effort since comments they received during the Expert Review phase indicate that 75-90% of all new oil wells are completed with hydraulic fracturing. Some commenters suggested that updated emission factors could be developed using data from recent studies and EPA is quoting a wide range of potential average

emission factors that are being considered without providing any explanations or justifications for these emission factors:

- 6.2 Mg CH4 (GHGRP based on gas well completions and workovers in Oil formations for wells with and without control);
- 3.1 Mg CH4 (UT Austin/EDF; wells with controls);
- 9.7 and 24.7 Mg CH4 (Wattenberg and Eagle Ford data, wells without control)

API wants to emphasize that existing data from recent field studies or from extrapolation from gas wells in oil formations do not provide a reliable representation of potential emissions from oil well completions and workovers. API is willing to work with EPA to assess data that may be used for future improvements of the emission factors used to characterize this emission source.

Comment: Petroleum Systems Emissions

As an additional item for future improvement, EPA is repeating its requests from the Expert Review draft for data on the Oil wells refracture rate, which EPA currently assumes to be 7.5% per year. As previously stated API concurs that field data for Oil well completions with and without hydraulic fracturing is currently sparse. However, EPA's assumption of a 7.5% workover (or refracture) rate for all oil wells seems higher than is expected based on industry's experience.

API is willing to work with EPA to develop a reasonable oil well refracture rate for potential use in future inventories.

Comment: Natural Gas Systems Emissions

Page 3-69. Recalculation Discussion: Gas Wells Completions and Workovers: Alternative Approach to Emission Factors Categories:

During the expert review phase of the U.S. GHG Inventory API supported EPA's derivation of new Emission Factors for gas wells completions and workovers utilizing GHGRP data. API has also noted the need for careful screening of reported data to make sure that erroneous entries and outliers are not used in these calculations.

Moreover, API has recommended that EPA collapse the proposed four categories for grouping gas well completions and workovers with hydraulic fracturing into only two categories. Therefore, in response to EPA's request for comments during the Public Review phase of the inventory, API reiterates its previous comments and maintains that the future relevance of the four distinct operating practices for which EFs were derived ought to be reconsidered. Newly proposed changes to estimating and reporting emissions for flowback events for hydraulically fractured completions and workovers1 and the phasing in of compliance with the Oil and Natural Gas (NSPS)2 will likely result in few to no events without reduced emissions completion

(RECs), and those with RECs will generally include both venting and flaring for short periods of time.

As described before, API is proposing an alternate two-category approach that may be adopted for future inventories and which would entail the derivation of emission factors that are representative of completions and workovers with hydraulic fracturing and limit significant changes in subsequent inventories in view of the expected operational changes:

- Non-REC Completions and Workovers (Vented only); and
- REC Completions and Workovers (vented and flared).

Table 1 below reintroduces a modified version of the EFs from such an alternative approach, as provided by API during the expert review phase of the U.S. GHG Inventory. The results are presented for both the 2011 and 2012 GHGRP data (with outliers removed) and these two categories are expected to provide a good characterization of emissions from these emission sources and will enable tracking industry's transition to the use of reduced emission completions and workovers. Based on discussions with EPA it became clear that EPA's count of vented completions and workovers without RECs includes completions with zero emissions. API's initial calculation approach excluded these data sets for the non-REC completions and workovers. This has been revised in API's modified analysis shown in Table1. For 2012, 466 non-REC vented completions and 95 non-REC workovers were reported with zero emissions.

Year	Category	Total CH4 Emissions, tonnes CO2e	# events	Tonnes CH4/event	Scf CH4/event	# data sets
2012 Data	Non-REC Completions and Workovers (Vented only)	1,121,164	3,037	17.58	915,596	252
	REC Completions and Workovers (vented and flared)	219,364	3,051	4.21	269,854	333
2011 Data	Non-REC Completions and Workovers (Vented only)	2,803,608	2,957	45.15	2,351,503	346
	REC Completions and Workovers (vented and flared)	430,161	4,815	4.25	221,572	319

Comment: Natural Gas Systems Emissions

Page 70. Planned Improvements: Completion and Workover Counts:

In its discussion about future Planned Improvements, EPA is addressing issues that were brought up repeatedly in API's discussions with EPA. API has provided comments before about the inconsistency in accounting for the total number of completions and workovers, due to ambiguous language in the GHG Reporting Program.

At EPA's request, API has surveyed its members and summarizes below the findings that point out the differences between EPA and API's completions and workover counts, which impact the calculated emission factors.

- EPA assumed the number of completions is equal to the sum of total completions reported and completions with purposely designed separating equipment (RECs). API assumed the RECs were a subset of the total completions reported. This was confirmed by seven (7) member companies.
- EPA assumed the number of workovers is equal to the sum of vented workovers, flared workovers, and REC workovers. API assumed the total number of workovers was equal to the sum of the vented and flared workovers, and that workovers with purposely designed separation equipment were a subset of this total. This was confirmed by five (5) member companies.
- Where data sets provided a count of workovers with REC, but no count of vented or flared workovers and zero emissions, EPA assigned these as vented workovers with REC. API treated these as invalid data sets. For 2012, this applied to 11 data sets, representing 21 workover events. The API analysis has been revised to include these data sets, as reflected in Table 1 above.

Comment: Natural Gas Systems Emissions

Page 3-71. Planned Improvement: Methane Measurement Studies:

EPA is requesting feedback on how measurements from top-down studies can be used to update its emissions estimates. As API stated before, studies such as Petron 2012 and Miller et al. 2013 focus on inverse flux modeling which employs emission concentration data from aircrafts, ground-based or towers over a regional area or on ambient hydrocarbon species ratios analysis. These studies have either been regional and do not fully represent natural gas production in the US (e.g. Petron 2012), or do not represent current operations (Miller 2013 and Petron 2012). Additionally, these studies are a "snapshot" in time and do not necessarily give any indication of emission rates over a longer time period such as annual. It is well know that bottoms-up methods like Allen et al. have much better accuracy over top-down methods. Since EPA's greenhouse gas inventory, uses a bottoms-up approach in itself, especially for quantifying CH4 emissions, it is more appropriate to use other bottoms-up approaches as data sources and for inventory verification.

There are several studies underway that attempt to combine top-down and bottoms-up methods to better understand and reconcile the differences. Until such time, EPA should only consider studies that measure emissions directly from the individual sources or activities.

Additionally, API wants to reiterate that no top-down study will be able to produce granular level information provided by the EPA inventory with respect to individual sources or activities within a sector. At best, these top-down methods can be used for gross verification of the inventory estimates.

Comment: Comments on Appendix A, Table A-135:

EPA has revised the voluntary emission reduction data in the table. The "other production" category increased from 40 Gg CH4 in the expert review version to 619.3 Gg in this version. There is no explanation of the change in reductions other than EPA reallocated some from the natural gas systems to petroleum systems.

API contends that this change is a step backward in the transparency of the emission reduction data and urges EPA to elaborate on how the change was calculated and what it includes. This does not apply only to the onshore production segment since the same increase is noted in the "other" reductions for the other industry segments listed in Table A-135.

Comment: Comments on Appendix A, Table A-141:

There seems to be an error in Table A-141. API's recalculation of the production sector emissions indicates that the value shown for condensate tanks in this table (2252 Gg) is not the net emissions. The net emissions for this source should be 164.9 Gg CH4.

Comment: Comments on Appendix A, Table A-143:

EPA revised the emission estimate for CO2 from flares. While in the Expert Review version 9,868.6 Gg CO2 were reported (Table A-141) in this version we note a value of 12,738.8 Gg (Table A-143). This appears to combine flaring from production and processing operations. API is requesting that EPA explain this new value and state specifically what industry segment it represents, or break out emissions associated with production operations separately from processing.

Comment: General Editorial:

API suggests that EPA keep the same order for the emission sources in the tables presented for each industry sector. This would certainly help when reviewing tables side by side. For example, EPA has moved the location of the emissions for gas well workovers among the different tables. In Table A-125, these emissions are presented with completions and well drilling, while in Table A 1-43, workover emissions are presented separately after tanks.

In summary, API appreciates the opportunity to provide comments during the public review phase of the 2012 U.S. GHG Inventory. EPA noted some errors and omissions that need to be addressed prior to finalizing the inventory while reiterating comments provided during the Expert Review phase and indicating areas for potential future improvements.

Commenter: David Isaacs
Semiconductor Industry Association

Comment: The current IPCC guidelines were established in 2006 with data collected in 2004 and earlier. While the data used to devise these methods represented the best available data at the time, it no longer represents the most accurate data available. The default emission factors contained in the current IPCC guidelines were based on 75 emissions characterization data sets, which may no longer be representative of the processes and equipment used throughout the industry.

Comment: In 2013 EPA issued a final rule governing the reporting of greenhouse gas emissions for the semiconductor industry in the United States, codified at 40 C.F.R. Part 98 Subpart I. As part of the development of this regulation, SIA member companies, several process equipment manufacturers, and SEMATECH, contributed to a large data collection effort resulting in a substantial amount of new data. The participants in this data collection undertook an extensive effort to characterize the processes deployed in our industry. The data collected was from equipment processing different wafer sizes and multiple semiconductor companies and equipment suppliers. It includes every fluorinated greenhouse gas currently used in semiconductor plasma etch processing and chemical vapor deposition chamber cleaning. The new data brings the total number of data sets to 1182.

SIA believes that the additional data used in the development of Subpart I will result in more accurate and more representative reporting of PFC emissions from semiconductor fabs in the United States as compared with the current IPCC guidelines used internationally to report emissions from our industry. EPA evidently concurs with this conclusion through the adoption of the regulation. Therefore, in order to improve the reporting of emissions globally and ensure consistency in reporting methods, SIA requests that EPA work to update the current IPCC guidelines to reflect this new data. Updating the IPCC guidelines will improve the consistency of the data contained in the U.S. inventory with the information available globally, and also improve the accuracy of the global data. SIA would be pleased to assist EPA in this endeavor.

Commenter: David Lyon
Environmental Defense Fund

Comment: Environmental Defense Fund (EDF) previously submitted comments on the Draft Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2012 (Draft 2014 Inventory) during the expert review comment period. As stated in our previous comments, we recommend that EPA updates the Petroleum Systems source category to account for methane emissions from co-producing well completions with hydraulic fracturing. EDF has prepared a white paper on methane emissions from co-producing well completions that summarizes our analyses of several recent data sources including the Greenhouse Gas Reporting Program Subpart W, Allen et al. 2013, and initial production data from the Eagle Ford, Bakken, and Wattenberg fields. Based on these analyses, we estimate that 2012 methane emissions from co-producing well completions are between 96 and 247 Gg CH4, comparable to the Draft 2014 Inventory estimate of 217 Gg CH4 emissions from hydraulically-fractured gas well completions and workovers.

Commenter: Nathan Matthews

Sierra Club

Comment: The Sierra Club files these comments on the February 2014 draft 1990-2012 Greenhouse Gas Inventory. We offer the following concerns:

- For gas production, although EPA proposes to revise sector wide emissions estimates downward, recent science based on atmospheric measurements indicates that a strong upward revision is appropriate.
- The "UT Austin EDF" Study provides further indication that the inventory's estimate of gas systems emissions is too low. Emissions from pneumatic controllers, in particular, are likely to be underestimated.
- The draft inventory does not include emissions from unconventional (e.g., hydraulically fractured) petroleum wells.
- EPA's outdated figure for methane's global warming potential is far lower than recent estimates.

Comment: Atmospheric Studies Indicate That Gas Systems Have Far Higher Emissions:

The February 2014 draft reduces EPA's estimate of total emissions from gas production. Yet several recent published studies based on regional atmospheric methane measurement indicate that estimates EPA proposes to lower were already too low.

We briefly summarize these atmospheric studies here. The first group of studies looked at particular regions. Two studies led by researchers with the National Ocean and Atmospheric Administration (NOAA) Earth System Research Laboratory that have directly measured methane in the atmosphere in other regions have estimated high leak rates. The first of these studies explains that by monitoring methane, propane, benzene, and other volatile organic compounds in the air around oil and gas fields, the authors can estimate oil and gas production's contributions to these pollutant levels. According to the study authors, their "analysis suggests that the emissions of the species we measure are most likely underestimated in [1990-2010] inventories," perhaps by as much as a factor of two, which would imply a leak rate of about

4.8% of production. A second announced NOAA study suggests that leak rates in the Uinta may be as high as 9%. Additionally, we note that a California study identified a 17% leak rate for oil and gas (presumably primarily oil) operations in the Los Angeles basin.

The second group of studies, released in the last four months, looks at nationwide gas production emissions and specifically criticizes the prior inventory as underestimating GHG emissions from gas production. In December of 2013, a paper published by Scot M. Miller et al. in the Proceedings of the Natural Academy of Sciences reviewed atmospheric measurements of methane and concluded that "The US EPA recently decreased its CH4 emissions factors for fossil fuel extraction and processing by 25–30% (for 1990–2011), but we find that CH4 data from across North America instead indicate the need for a larger adjustment of the opposite sign." In other words, rather than reducing the estimated leak rate from 2.4% to something approaching 1.5%, EPA should have increased its estimate to at least 3%. In February, a paper published in Science similarly concluded that the then current inventory underestimated methane emissions from gas production—indicating that the February 2014 draft is a change in the wrong direction.

Sierra Club has not identified the likely reason for the discrepancy between these "top down" assessments incorporating atmospheric measurements and EPA's "bottom up" estimate based on individual components, practices, and emission factors. Assuming the atmospheric studies to be correct, factors contributing to this discrepancy may include underestimation of the number of wells, a system wide underestimation of per component emission factors, drastic underestimation of emissions from particular sources (perhaps pneumatics or liquids unloading), or there may be some other cause. Although we are unable to recommend a particular correction to the inventory fully reconcile the inventory with these studies, we strongly encourage EPA to devote attention to this issue.

Comment: The "UT Austin-EDF" Study Further Indicates That The Inventory Underestimates Gas Systems Emissions:

The 2014 draft acknowledges a study by David Allen, of University of Texas, Austin, et al. and sponsored by the Environmental Defense Fund (EDF) as a source of additional information regarding gas and petroleum system emissions; the draft generally refers to this work as the "UT Austin EDF study." This study also generally indicates that the inventory underestimates emissions from gas systems. For the wells and completions included in this study, observed emissions were similar to average gas system emissions implied by the 2013 GHG inventory. However, the UT Austin EDF study found much higher utilization of reduced emission completions than are contemplated by the EPA inventories, resulting in drastically lower emissions from that particular slice of the lifecycle. These reduced completion emissions were offset, however, by increases from other components, such as pneumatics, in excess of those assumed by the inventory. These observed high rates of emissions from activities other than completions should be expected to apply industry wide, indicating that 2013 inventory underestimated these emissions. More generally, the UT Austin-EDF study should be assumed to

represent the top end of performers, insofar as the wells included were from large industry players who opted in to the study and who had notice that measurements would be taken. EPA should look critically at emission estimates that would indicate that the industry as a whole performs better than the subset of players and wells included in the UT Austin-EDF study.

Comment: The Inventory Likely Underestimates Emissions from Pneumatic Devices:

As noted above, the UT Austin-EDF study indicates that the inventory significantly underestimates emissions from pneumatic devices. Until individual pneumatic devices are reported pursuant to Subpart W, EPA should adopt an approach such as the one recommended by EDF in their separate comment on the February 2014 draft.

Comment: Petroleum Systems Estimates Must Account for Unconventional Production:

There can be no disputing that hydraulic fracturing has changed the face of American petroleum production and has been employed in a large percentage of petroleum wells for a number of years. FracFocus, the national hydraulic fracturing chemical registry managed by the Ground Water Protection Council and Interstate Oil and Gas Compact Commission, includes records from 12,056 oil wells that were hydraulically fractured in 2012. Yet the 2014 draft of the inventory estimates petroleum system emissions only using emission factors for conventional production. As explained in comments submitted separately by the Environmental Defense Fund, many of the tools EPA proposes to use to estimate gas systems emissions can also be applied to petroleum systems. Although these tools are imperfect, they can provide a much more accurate estimate of emissions than the draft inventory's inaccurate assumption that hydraulic fracturing is not used in petroleum wells.

Comment: EPA Uses an Outdated, and Far Too Low, Estimate of Methane's Global Warming Potential:

The inventory discusses methane's global warming potential (GWP) on the 100 year timeframe, and estimates this potential as 21. EPA explains that it uses this value pursuant to UNFCCC reporting obligations. Id. Yet as EPA recognizes, this value does not represent the best available science. As an interim measure, EPA provides an annex with many charts explaining the impact of using the 2007 Intergovernmental Panel on Climate Change (IPCC) 100 year methane GWP estimate of 25, Annex 6.1, but even that estimate has been superseded in the intervening seven years of research. Most importantly, the IPCC's Fifth Assessment Report estimates an aggregate 100-year methane GWP of 34, and an even higher estimate of 36 for methane emitted from fossil sources.

EPA must therefore take available steps to encourage this reporting obligation to be updated to reflect the best available science. These steps include including informing other federal entities participating in negotiation of these agreements of the importance of using recent science. As an

interim measure, EPA should present an annex using the methane GWP data from the IPCC AR5 report, as the draft inventory does for the IPCC AR4 data.

Supplemental Material Received

Appendix A
Environmental Defense Fund White Paper on Methane Emissions from Co-Producing Well Completions

Appendix B
Energy Innovation Missing Methane Issue Brief

Appendix C
Damascus Citizens for Sustainability Extended Report on a Preliminary Investigation of Ground-Level Ambient Methane Levels in Manhattan, New York City, New York

Appendix D
Damascus Citizens for Sustainability citing Phillips et al. 2012

Appendix E
Damascus Citizens for Sustainability citing Pieschl et al. 2013

Appendix F
Damascus Citizens for Sustainability Report on a Survey of Ground-Level Ambient Methane Levels in the Vicinity of Wyalusing, Bradford County, Pennsylvania

Appendix G
Damascus Citizens for Sustainability citing Report to the Clean Air Council of the June 8, 2012, on Field Inspection and Methane Sampling Survey

Appendix H
Damascus Citizens for Sustainability citing Tollefson 2013

Appendix I
Damascus Citizens for Sustainability citing Tollefson 2012

Appendix J
Damascus Citizens for Sustainability citing Miller et. al 2013

Appendix A

CO-PRODUCING WELLS AS A MAJOR SOURCE OF METHANE EMISSIONS: A REVIEW OF RECENT ANALYSES

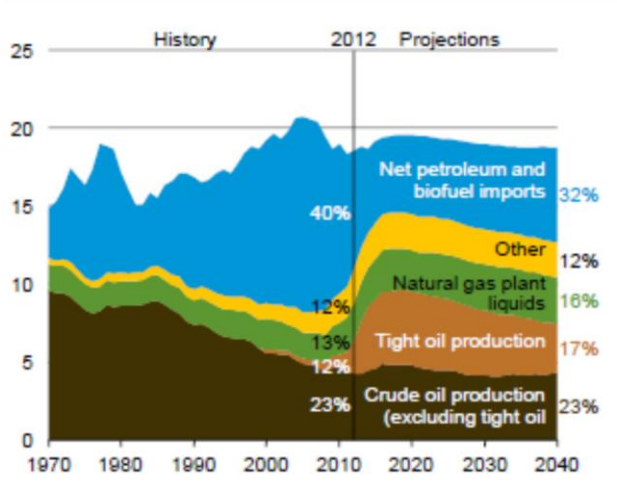
PREPARED BY ENVIRONMENTAL DEFENSE FUND

MARCH 2014

The Environmental Protection Agency's ("EPA's") New Source Performance Standards ("NSPS") for the oil and natural gas sector require that hydraulically fractured natural gas wells reduce their completion emissions using either reduced emission completions ("RECs") or flaring.[1] EPA defines a "gas well" or "natural gas well" as "an onshore well drilled principally for production of natural gas"[2] and, depending on how this definition is interpreted, a number of wells that co-produce oil (or other liquids) and natural gas ("co-producing wells") may not need to control their emissions under the REC requirements in the NSPS.

Many completions of these co-producing wells, however, produce substantial pollution that can be cost-effectively mitigated using the same clean air measures that have effectively reduced emissions from hydraulically fractured gas wells. Extending clean air protections to co-producing wells is vital given recent trends within the oil and gas industry. Over the last two years, rising oil prices and low natural gas prices have caused new drilling activity to increasingly shift to shale formations rich in oil and condensates. Reflecting this trend, the U.S. Energy Information's ("EIA's") most recent Annual Energy Outlook predicts that domestic oil production will grow significantly through 2020, driven primarily by increases in tight oil production (see Figure 1).

Figure 1. US Petroleum and Other Liquids Supply, 1970-2040 (EIA)



[1] With limited exceptions, all fractured and refractured natural gas wells will be required to use RECs as of January 1, 2015. 77 Fed. Reg. 49,490, 49,497 (Aug. 16, 2012).

[2] 40 C.F.R. § 60.5430.

This analysis synthesizes available information on per-completion emissions factors, the cost-effectiveness of mitigating those emissions using RECs or high-efficiency flaring, and, where possible, the total amount of methane that would be reduced by deploying these completion protections at co-producing wells. Table 1 synthesizes data from the following sources:

- A February, 2014 Stanford/Novim Study in the journal Science entitled "Methane Leakage from North American Natural Gas Systems;" ("Stanford/Novim Analysis")[3]

- ICF International's Report from March, 2014 entitled "Economic Analysis of Methane Emissions Reduction Opportunities in the U.S. Onshore Oil and Natural Gas Industries;" ("ICF Report")[4]

- A 2013 analysis in the Proceedings of the National Academy of Sciences led by the University of Texas entitled "Measurements of methane emissions at natural gas production sites in the United States;"[5] ("UT Study")

- EDF's analysis of the oil and natural gas portion of EPA's Greenhouse Gas Reporting Program ("EDF Subpart W Analysis");[6] and

- An analysis completed by EDF and Stratus Consulting of well completion reports in the Bakken, Eagle Ford, and Wattenberg field ("EDF/Stratus Analysis").

These sources all indicate that co-producing well completions are a substantial source of methane emissions, with total estimated emissions much larger than the figure reported in EPA's official inventory of greenhouse gas emissions. EPA's current emission factor for co-producing wells derives from a 1996 study of conventional oil wells, and very likely underestimates emissions from the hydraulic fracturing techniques that are prevalent today.

[3] A.R. Brandt et al., Methane Leaks from North American Natural Gas Systems, 343 SCIENCE 733 (Feb. 14, 2014), available at http://www.novim.org/images/pdf/ScienceMethane.02.14.14.pdf.

[4] The report is available at http://www.edf.org/sites/default/files/methane_cost_curve_report.pdf.

[5] David T. Allen et al., Measurements of methane emissions at natural gas production sites in the United States, PNAS Early Edition (2013), available at www.pnas.org/cgi/doi/10.1073/pnas.1304880110.

[6] EDF, Comments on "Draft Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2012" (included in the supplemental information for this analysis).

TABLE 1: Summary of Co-producing Emissions, Cost-Effectiveness, and Mitigation Potential

Data Sources	Potential Emission Factor (MT CH_4)	National Emissions Estimates*** (MT CH_4)	REC Cost Effectiveness ($/MT CH_4)		Flaring Cost Effectiveness ($/MT CH_4)	Methane Mitigation Potential (MT CH_4)
			without savings	with savings		
Stanford/ Novim Analysis*	40.2[7]	120,000****	778		92	114,000
ICF Report	6.6**	96,000	n/a	n/a	96.57	94,000
UT Study*	193.5		153.8	-132.7[8]	19.19	n/a
EDF Subpart W Analysis	21.8	163,000	1,435		170	140,000
EDF/Stratus Analysis	15.7	247,000	3,578	3,314	424	235,000

*Analysis includes potential emissions factor only. Cost-effectiveness and mitigation potential derived using common assumptions described below.
** This EF includes both vented emissions controlled emissions so is not a true potential emissions factor.
*** Estimates provided by the authors of each individual study.
**** This estimate only reflects emissions from three major production basins, and therefore understates total national emissions.

The remainder of this white paper provides additional information on the development of an emission factor for co-producing wells, the cost-effectiveness of mitigating these emissions, and overall methane mitigation potentials.

Potential Emission Factor

The above-described analyses determine potential emissions factors for co-producing well completions using several different methods, including direct measurement, analysis of Subpart W data, and analysis of initial oil and gas production. All of these analyses find potential emissions are significantly greater than the emissions factor for oil well completions currently in EPA's annual greenhouse gas inventory (0.0141 tons of methane per completion). Given that EPA's current emissions factor is dated and was based on emissions from completions of conventional, non-hydraulically fractured wells, the more recent studies described below suggest that the official inventory is likely underestimating the extensive methane emissions from co-producing well completions. Moreover, neither the current NSPS

[7] Weighted average of emission factors for wells in the Bakken, Eagle Ford, and Permian Basins.

[8] On average, these wells would achieve net savings of $25,630 by selling gas recovered during completions, assuming $4/Mcf.

nor the regulations of most states require control of completion emissions from co-producing wells.[9]

UT Study. The UT Study measured various large sources of methane in the production sector, including 27 well completions in various geographic areas across the country. Six of the measured completions were at co-producing wells that produced significant amounts of hydrocarbon liquids,[10] and, for each of these completions, researchers directly measured potential and actual methane emissions. Actual completion emissions from these co-producing wells ranged from 1.7 to 5.0 metric tons ("MT") CH_4, though all of the wells controlled completion flowback emissions with either flaring or a combination of RECs and flaring. The UT study estimated potential emissions as the total volume of gas vented, flared, and sent to sales from initiation of flowback until the reported completion end time. The potential emissions from these wells, which would be more indicative of uncontrolled completions, ranged from 81.9 to 414.4 MT CH_4, with an average value of 193.5 MT of CH_4/completion.[11]

Table 1. Measured and potential emissions of co-producing wells from Allen, et al. (2013)

Completion Event	Emission Controls	Measured Emissions (scf CH_4)	Potential Emissions (scf CH_4)	Measured Emissions (MT CH_4)	Potential Emissions (MT CH_4)
GC-1	Flaring	105,000	5,005,000	2.0	96.4
GC-2	Flaring	90,000	4,250,000	1.7	81.9
GC-3	REC & Flaring	260,000	21,500,000	5.0	414.1
GC-4	REC & Flaring	180,000	13,000,000	3.5	250.4
GC-6	Flaring	247,000	12,200,000	4.8	235.0
GC-7	Flaring	90,000	4,320,000	1.7	83.2
Average		162,000	10,030,000	3.1	193.5

Subpart W Analyses. EDF also evaluated completion data from 2011 and 2012 that was reported to EPA under its greenhouse gas reporting rule for oil and gas systems (known as "Subpart W").[12] Subpart W does not require reporting of oil well completion and workover

[9] Notably, Colorado does require that co-producing wells perform reduced emission completions. Co. Oil & Gas Conserv. Comm'n ("COGCC") Rule 805(b)(3)(A).

[10] David T. Allen et al., Measurements of methane emissions at natural gas production sites in the United States, PNAS Early Edition (2013), available at www.pnas.org/cgi/doi/10.1073/pnas.1304880110. See also EDF, Analysis of Co-Producing Well Completions (updated Mar. 2013) (included in the supplemental information for this analysis).

[11] EDF, Analysis of Co-Producing Well Completions (Dec. 2013). The underlying study analyzed a total of 26 well completions.

[12] EDF, Comments on "Draft Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2012" (included in the supplemental information for this analysis).

emissions. Nonetheless, in 2011 and 2012 there were 1,754 reports of completions and workovers from wells in formations classified under Subpart W as "oil formations." EDF performed a separate analysis of DI Desktop data to assess if these completions were actually oil wells.[13] In approximately 75% of the counties from which these completion reports came, over half of the wells with first production in 2011 & 2012 were oil wells. Using the same approach that EPA used to estimate emission factors for completions from the entire GHGRP dataset, EDF has derived emission factors for this subset of wells located in oil formations (Table 3). The average emission factor for all oil formation completion and workovers is 6.2 MT CH_4/event, or more than 400 times higher than the current oil well completion emission factor. EDF also developed separate emission factors for each combination of emission controls reported under Subpart W: uncontrolled ("vented") completions, completions controlled with a flare, completions controlled with a REC, and completions controlled with both flares and REC. The emission factors for the four categories range from 3.1 MT CH_4/event for completions with REC to 21.8 MT CH_4/event for vented completions.

The ICF Report also uses Subpart W data to develop an emission factor for hydraulically fractured oil wells. From this data, the Report develops an emissions factor of 344,000 scf CH_4/completion or 6.6 MT CH_4/completion, which is an average value including both controlled and uncontrolled completions.

Table 3. Oil well completion and workover emission factors developed from 2011 & 2012 GHGRP Subpart W oil formation type sub-basins using the same method as EPA for developing the natural gas completion and workover emission factors

Category	Completions (# events)	Workovers (# events)	Completions & Workovers (# events)	Completions EF (MT CH_4/event)	Workovers EF (MT CH_4/event)	Completion & Workover EF (MT CH_4/event)
Vent	320	147	467	21.8	7.6	17.3
Flare	221	66	287	3.7	2.5	3.4
REC	186	0	186	3.1	N/A	3.1
REC+Flare	17	0	17	11.7	N/A	11.7
Ambiguous	708	89	797	1.5	0.0	1.3
All events	1,452	302	1,754	6.6	4.2	6.2

Initial Production Analyses. The Stanford/Novim Analysis evaluated 2,969 well completions in the Bakken, Eagle Ford, and Permian basins for 2011 using the DrillingInfo HPDI Database.[14] The analysis estimated potential emissions from these tight oil wells by converting

[13] Data obtained from DrillingInfo, DI Desktop, http://info.drillinginfo.com/products/di-desktop/.

[14] A.R. Brandt et al., Methane Leaks from North American Natural Gas Systems, 343 SCIENCE 733 (Feb. 14, 2014), available at http://www.novim.org/images/pdf/ScienceMethane.02.14.14.pdf. The relevant data is contained in the supporting documentation for the study (http://www.sciencemag.org/content/suppl/2014/02/12/343.6172.733.DC1/Brandt.SM.datafile.xlsx).

peak gas production to a daily initial production rate. It then assumed that production during flowback increased linearly with time for 9 days prior to initial production and all such methane emissions were vented, or understood differently, that completion emissions correspond to 4.5 days of initial gas production.[15] Using this methodology, the analysis determined potential emissions factors for the Bakken (31.1 MT CH_4/completion), Eagle Ford (90.9 MT CH_4/completion), and Permian (31.2 MT CH_4/completion) Basins.

The EDF/Stratus analysis takes a similar approach, using initial production values to understand potential completion emissions at co-producing wells. Stratus Consulting initially performed an analysis of 100 well completions in the Bakken, assuming a 7 to 10 day completion event with gas production increasing from zero to the initial production value in a non-linear fashion over the course of the completion. Accordingly, Stratus assumed that total gas production over the 7-10 day completion event would equal 3 average days of gas production.[16] As with the Stanford/Novim analysis, Stratus assumed all of this gas was vented.

EDF subsequently extended this analysis to approximately 9,500 wells in the Bakken, Eagle Ford, and Wattenberg fields.[17] Only oil wells were analyzed for the Eagle Ford and Wattenberg fields; North Dakota does not distinguish between oil and gas wells so all Bakken wells were assumed to be oil wells. Across all wells, the analysis found an average potential emissions factor of 15.7 MT CH_4/completions with averages of 18.0, 24.7, and 9.5 MT CH_4/completion in the Bakken, Eagle Ford, and Wattenberg respectively.

Cost Effectiveness

Other than the ICF Report, none of the above non-EDF analyses calculated the cost-effectiveness of controlling completion emissions using RECs or high-efficiency flaring. Accordingly, we applied consistent cost assumptions to all of the analyses above, except the ICF Report. For RECs, we assumed 95% control efficiency and used EPA's cost of performing a reduced emission completion ($29,713)[18] to calculate cost-effectiveness. Across all studies, we calculated a REC cost-effectiveness without a credit for captured gas ranging from $154 - $3,578/MT CH_4 reduced. Using production data from approximately 9,500 wells in the Bakken, Eagle Ford, and Wattenberg fields, we calculated a REC cost-effectiveness with credit for gas

[15] This methodology is set forth in Francis O'Sullivan & Sergey Paltsev, Shale gas production: potential versus actual greenhouse gas emissions, ENVTL. RES. LETTERS 7(4):044030 (Nov. 26, 2012).

[16] Memorandum from Leland Deck, Stratus Consulting, to Peter Zalzal and Vickie Patton, Environmental Defense Fund, re: Methods Memo on VOC Cost-Effectiveness in Controlling Bakken Shale Combined Oil and Gas Wells During Well Completion (Mar. 30, 2012) (included in the supplemental information for this analysis).

[17] EDF, Spreadsheets analyzing Bakken, Eagle Ford and Wattenberg wells (included in the supplemental information for this analysis).

[18] EPA, Oil and Natural Gas Sector: Standards of Performance for Crude Oil and Natural Gas Production, Transmission, and Distribution, Background Technical Support Document for Proposed Standards (July 2011), available at http://www.epa.gov/airquality/oilandgas/pdfs/20110728tsd.pdf.

capture. With a credit for gas savings (based on an assumed gas price of $4.00/Mcf), we calculated a median cost-effectiveness of $3,314/MT CH_4 reduced and also calculated cost-effectiveness for the top 25% and top 10% of wells, as shown in the table below.

Table 4. EDF / Stratus REC Cost-Effectiveness for Median and Top 25 and 10 Percent of Wells

Percentile	REC Cost Effectiveness with gas capture credit ($/MT CH4)	Mitigation Potential (MT CH4)	Mitigation Potential (% of total)
10%	$544	60,643	40.9
25%	$1,266	97,430	65.7
50%	$3,314	126,508	85.3

To calculate flaring cost effectiveness, we assumed 95% destruction and removal efficiency ("DRE") and multiplied this by the emission factor to get flaring emission reductions. We then divided the EPA cost estimate of flaring completion emissions from a well ($3,523) by the flaring emission reductions for each of the analyses.[19] Across all studies (excluding the ICF Report) we calculated a flaring cost-effectiveness ranging from $19 - $424/MT CH_4 reduced.

The ICF Report includes its own cost assumptions about performing high-efficiency flaring, which are substantially higher than those in EPA's NSPS. ICF assumes flaring has a 98 percent control efficiency and a capital cost of $50,000, with an additional $6,000 in fuel costs for ignition. ICF estimates the cost-effectiveness of flaring to be $1.86/Mcf of methane ($97/MT CH_4) for completion gas. The ICF report did not examine the cost-effectiveness of RECs for co-producing wells.

Mitigation Potential

Determining inventory-wide mitigation potential requires scaling up emissions nationally and then applying percentage reductions associated with mitigation technologies. The Stanford/Novim Analysis, the ICF Report, the EDF Subpart W Analysis, and the EDF/Stratus Analysis all provide national estimates of emissions from co-producing wells, which we describe in greater detail below. The UT Study does not scale these specific emissions nationally and we have not provided a separate scale up of those emissions here.

[19] Id.

- Stanford/Novim Analysis. The Stanford/Novim analysis found that co-producing well completions accounted for approximately 120,000 MT CH_4 in 2011.[20] The analysis assumed all emissions were vented and multiplied emissions factors in the Bakken, Eagle Ford, and Permian Basins by the total number of completions in those basins. Because the 120,000 MT CH_4 figure includes only emissions from these three basins, it is not a true national figure.

- ICF Report. ICF used its emissions factor of 344,000 scf CH_4/completion (6.6 MT CH_4/completion) from Subpart W along with the most recent API Quarterly Completions Report showing 15,382 hydraulically fractured oil well completions for 2011. Using these values, ICF calculated completion emissions of 5 Bcf CH_4 or 96,000 MT CH_4.

- EDF Subpart W Analysis. EDF applied emissions factors we calculated from Subpart W to the 2012 Draft GHG Inventory activity data of 15,753 oil well completions.[21] This resulted in emission estimates between 49,000 MT CH_4 (assuming all RECs) and 343,000 MT CH_4 (assuming all emissions vented), or 182,000 MT CH_4 if the use of emission controls among the 15,753 oil well completions is assumed to be distributed in the same way as the Subpart W dataset. Because some wells are already controlled, we assumed the national proportion of uncontrolled completions was 43%, the same as the Subpart W dataset, and applied the emission factor for vented completions. We use this 147,000 MT CH_4 value for purposes of determining mitigation potential.

- EDF/Stratus Analysis. The EDF/Stratus analysis did not isolate hydraulically fractured wells, but instead derived an average emission factor applicable to all co-producing well completions. Accordingly, EDF applied emissions factors we calculated using the Stratus methodology to EPA's 2012 Draft GHG Inventory activity data of 15,753 oil well completions for an emissions estimate of approximately 247,000 MT CH_4 annually.

Translating these national emissions estimates into mitigation potential requires applying control efficiencies. The ICF Report assumes flaring achieves 98% DRE, and accordingly suggests mitigating completion emissions from co-producing wells could achieve 94,000 MT CH_4 in annual reductions.

---

[20] A.R. Brandt et al., Supplementary Materials for Methane Leaks from North American Natural Gas Systems 30, 343 SCIENCE 733 (Feb. 14, 2014), available at http://www.sciencemag.org/content/suppl/2014/02/12/343.6172.733.DC1/1247045.Brandt.SM.pdf.

[21] Although not all oil wells completions use hydraulic fracturing, FracFocus, the national hydraulic fracturing chemical registry managed by the Ground Water Protection Council and Interstate Oil and Gas Compact Commission, includes records from 12,056 oil wells that were hydraulically fractured in 2012. Reporting to FracFocus is voluntary in many states, which implies that the actual number of hydraulically fractured oil wells is higher than 12,056. Accordingly, we have used the draft inventory activity data as a reasonable proxy for the total number of hydraulically fractured oil well completions.

The Stanford/Novim analysis does not calculate mitigation potential, and so, consistent with the two EDF analyses, we conservatively assume flaring or gas capture achieves a 95% control efficiency. Because both the Stanford/Novim analysis and EDF/Stratus analysis assume all emissions are vented, we apply the 95% control figure directly to total emissions estimates, resulting in annual mitigation potentials of 114,000 MT CH_4 and 228,000 MT CH_4 respectively. Because EDF's Subpart W analysis assumes some wells are already controlled, we apply the 95% control effectiveness only to the subset of emissions that are vented for an annual mitigation potential of 140,000 MT CH_4.

Conclusions

Although neither EPA regulations nor the regulations of most states require control of emissions from co-producing well completions, these emissions are a potentially significant source of methane and other harmful pollutants. Recent studies and analyses – drawing from a variety of data sources including field studies of well completions, Subpart W reports, and well completion databases – suggest that emissions from an uncontrolled co-producing well completion range from 15.7 MT of CH_4 to nearly 200 MT. At a national level, these emission factors suggest total co-producing well completion emissions between approximately 96,000 to 247,000 MT, comparable to emissions from natural gas well completions (209,000 MT CH_4 in the latest EPA annual inventory). Current control technologies for natural gas well completions – including RECs where gathering infrastructure is available, and high-efficiency flaring in other situations – can be readily applied to co-producing well completions. This white paper suggests that applying those technologies to co-producing well completions would yield emission reductions on the order of 94,000 to 228,000 MT per year, or 2.63 to 6.38 million MT CO_2-e (using 100-year GWP of 28).

Appendix B

ENERGY INNOVATION
POLICY & TECHNOLOGY, LLC

The Mystery of the Missing Methane

Advances in the scientific understanding of methane emissions highlight the need for improvements to the EPA emissions inventory

By Chris Busch

25 March 2014

---

1. Summary

The U.S. Environmental Protection Agency (EPA) recently released a draft of its 1990-2012 greenhouse gas (GHG) emissions inventory. While the EPA is in many ways at the frontier of global best practice, the agency needs to take action to account for the accumulating evidence that the GHG inventory is omitting a significant fraction of methane emissions, the second most prevalent contributor to climate change. The new draft inventory estimates that emissions fell almost two percent in 2012 compared to 2011, and it revises downward previous estimates of methane emissions for the natural gas sector. For example, 2011 emissions are almost 10 percent lower in the 2014 draft inventory than they were in the 2013 inventory. These downward revisions are being made despite increasing scientific evidence that the EPA should be increasing its estimate of emissions.

Just one week before the draft inventory was released, the journal *Science* published a landmark study (Brandt et al., 2014) that concludes that the EPA inventory is undercounting emissions by a significant margin. The study brings together, for the first time, the full body of existing evidence on methane leakage. It estimates that there are 7-21 teragrams (Tg; 10^{12} grams) of methane missing from the EPA inventory and concludes that some of this methane is likely coming from the natural gas system. This quantity, 7-21 Tg, is equivalent to roughly 25–75 percent of the total methane emissions in the inventory and is two to four times the EPA's current estimate of methane emissions from the natural gas system.

The EPA needs to develop a plan to collect and analyze real-world data to narrow the uncertainty ranges and provide a better understanding of methane emissions, especially from the natural gas system. New technologies for detection and measurement of methane emissions can help the EPA achieve this goal. Additional resources should be dedicated to this objective.

2. Bottom-up vs. top-down studies of methane emissions

The EPA emission inventory relies on "bottom-up" studies of methane emissions. Bottom-up studies involve component-level sampling on the ground, at the source. The EPA uses the results from these studies to calculate emission factors for different activities that make up the natural gas system, including production, processing, transmission, and distribution. These emission factors—essentially, typical levels of emissions per unit output for different components of the system—are applied to natural gas production activities to calculate activity-specific emissions, and then are summed to estimate total system-wide emissions. As the EPA inventory for the natural gas system is constructed, uncontrolled emissions are first estimated using the process above (the "potential emissions"), then regulatory initiatives and voluntary information provided by companies are taken into account to produce estimated emissions.

Figure 1. Methane emissions are invisible to the naked eye

Methane emissions from this storage tank are visible not the naked eye but an infrared lens reveals their existence. Photo source: New York Times.

One of challenges with bottom-up studies is that they require the participation of landowners and natural gas companies. Researchers must obtain permission in order to enter a property and directly measure emissions, and have not found it easy to do this. There is some reason to believe that the producers that have voluntarily participated are the cleanest, lowest-emitting operators. This, in combination with the great heterogeneity in types of operations and geology across gas-bearing basins, means that it is difficult for bottom-up studies to collect data from a broad enough array of sources for the sampling to be representative.

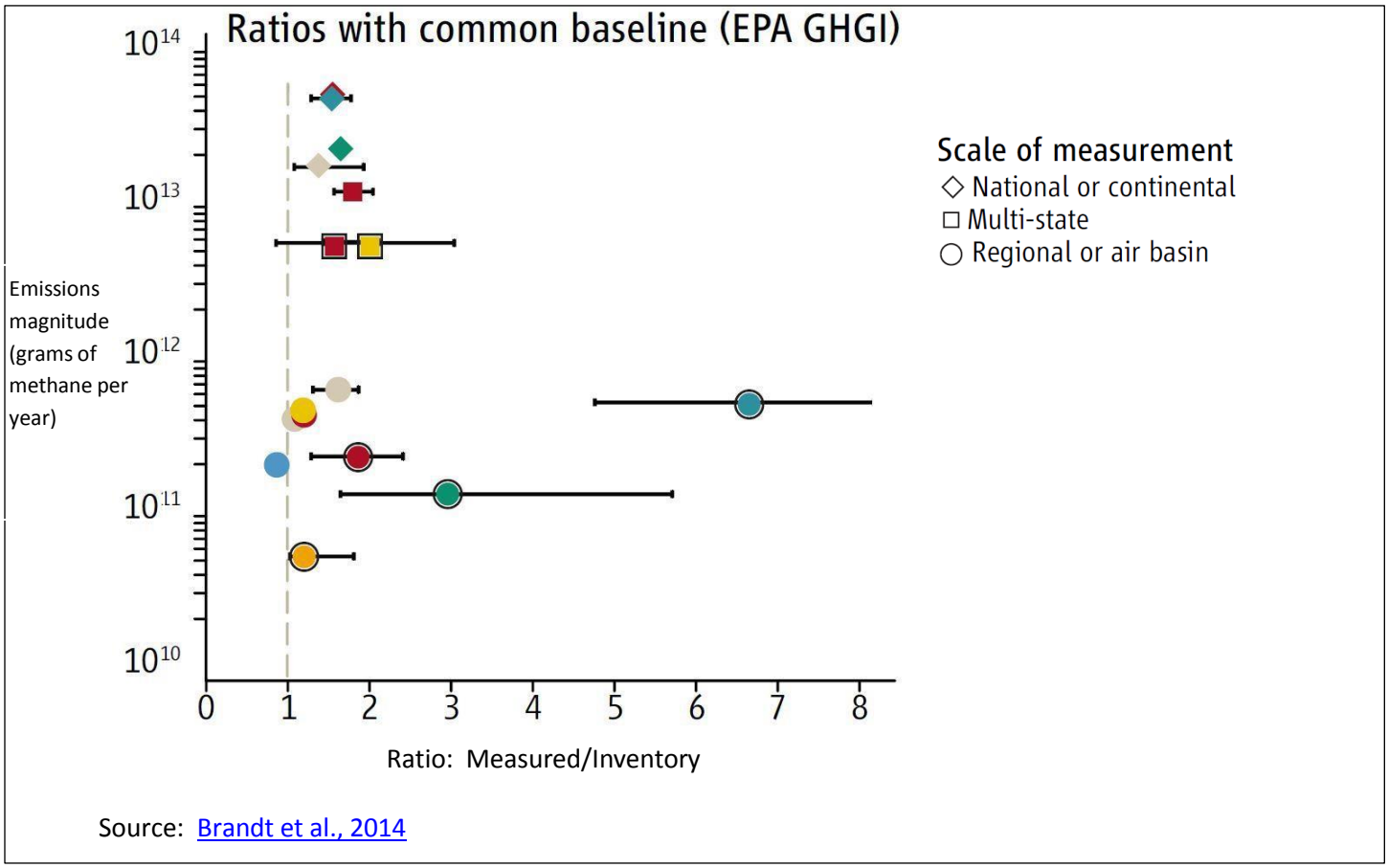
"Top-down" studies are a second, distinct approach for measuring methane emissions. These studies are based on atmospheric sampling from aircraft or tall towers. Top-down studies provide great accuracy with respect to the quantity of total emissions (though some uncertainty is introduced by wind-blown methane that might enter or exit the study area before being sampled). Traditionally, the weakness of top-down studies has been the difficulty of discerning the contribution of different sources the overall level observed level of methane. Many top-down studies have not even attempted to attribute the methane sampled in the atmosphere to particular sources on the ground. However, emerging techniques are making progress in allowing identification of likely sources for atmospherically sampled methane.

3. The missing methane

Brandt et al.'s paper is innovative in two ways. First, they provide a framework for comparison of past studies on methane emissions. In a feat of graphic creativity, Brandt et al. put all of the existing studies, bottom-up (denoted by triangles and dashes) and top-down (denoted by circles, squares, and diamonds), on a single chart. The result helps illuminate how these two threads in the literature relate to each other. Bottom-up studies measure facilities or components: the largest value found by any such study was around 10^9 g of methane emitted per year. In contrast, even the smallest of the top-down studies, which measured the Denver-Julesberg basin, reported over $4*10^{10}$ g of methane.

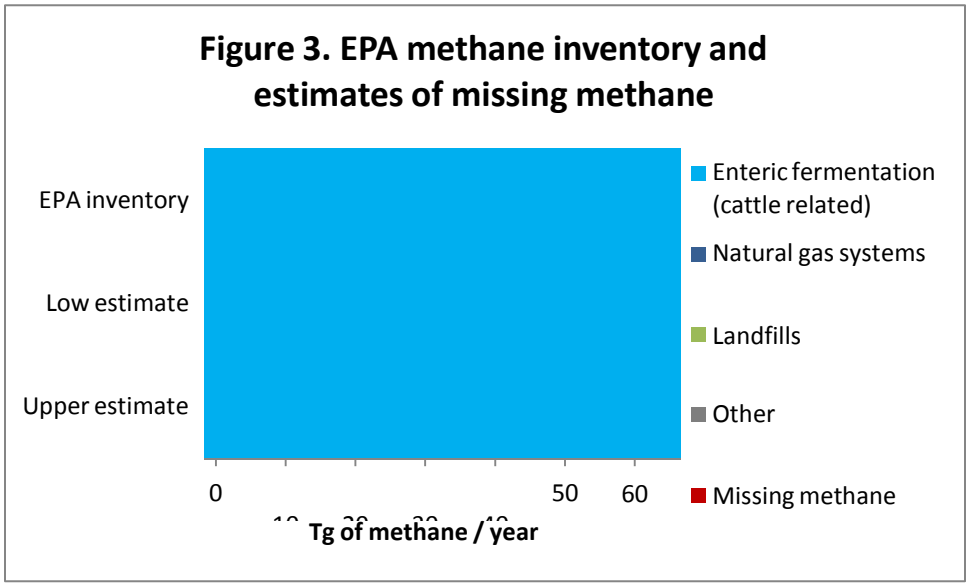
Brandt et al. also conduct a meta-analysis of national-scale, top-down studies of methane emissions. The authors develop a normalization procedure to make the multitude of studies comparable. The result indicates that the most likely range of actual methane emissions is 25–75 percent higher than the EPA inventory indicates. This range of possible emissions is illustrated in the inset panel for Brandt et al.'s principal graphic, which we reproduce as Figure 2. Note that for all of the studies that are national or continental in scale, observations all lie between 1.25 and 1.75—that is 125 percent and 175 percent of the EPA inventory.

Figure 2. Normalized comparison of top-down studies in Brandt et al.



Source: Brandt et al., 2014

Figure 3 shows in red the lower and upper estimates (7- 21 Tg) of methane emissions that the EPA did not account for in their inventory, which we are referring to as missing methane. The missing methane is shown on top of the results from the EPA's latest inventory.



Because of the limited ability of top-down studies to trace methane back to specific ground-level sources, it is not possible to determine the origin of the missing methane with great certainty. Still, there is reason to believe that at least *some* of the missing methane is coming from the natural gas system, as there are downward structural biases in the inventory. For example, it would be reasonable to expect that facility operators who believe they may have above-average emissions would be hesitant to join voluntary studies. This may have a large impact on results, as there is accumulating evidence that "super emitters" – a small number of facilities with particularly large leaks – could be a majority or a large fraction of overall emissions. Another downward structural bias is the EPA's choice to reduce the emissions estimated through the bottom-up procedure based on industry assertions that they have taken voluntary actions above and beyond those required by regulations.

The large range of uncertainty remaining about the rate of emissions in the natural gas system is an indicator of the complexity of the situation. The natural gas system is large, complex and heterogeneous, in both engineering and geologic terms. Each natural gas-bearing basin is unique, and there is great variation in how producers operate. Methane emissions come not only from wells producing natural gas, but also from those mainly producing oil. Indeed, 20 percent of the nation's gas is "associated gas" produced at oil wells. Oil wells have different emissions characteristics from wells designed to extract primarily natural gas. The intermingling of the oil and natural gas systems also introduces the question of how to attribute methane emissions. Some of the methane emissions from the petroleum system should be attributed to natural gas, but determining the appropriate fraction is challenging.

4. Computational extensions

The Brandt et al. paper concludes that some of the missing methane is likely coming from the natural gas system. It explores the specific possible sources of methane from the natural gas system beyond the EPA estimates. In the supporting materials for the article, the authors develop what they call a worst-case scenario for emissions from the natural gas system that considers the notion that all of the missing methane is from natural gas. Under such a scenario, if 7-21 Tg of extra methane was being emitted from the natural gas system, that would imply emission rates two to four times higher than the EPA inventory estimate.

While concluding that some of the missing methane almost certainly originates from the natural gas system, the Brandt et al. paper also emphasizes the continued lack of certainty regarding the extent that natural gas emissions are underestimated. To emphasize this uncertainty, the authors consciously chose to refrain from translating missing methane into emission rates. We also find it useful to illustrate the potential magnitude of the problem through some further computation, including implied emission rates for the natural gas system at different levels of missing methane.

Here, we develop four scenarios, translating the missing methane into an emission rate of methane from the natural gas system. The emission rate is calculated by adding a portion of the missing methane (varying by scenario) to the methane emissions assigned to the natural gas industry in the EPA's inventory, then dividing that value by the sum of natural gas production plus total methane emission in that scenario. We also specify the ratio of each scenario's methane emissions attributed to natural gas systems to the corresponding value from the EPA inventory. The scenarios are shown in Table 1.

Table 1. Emission scenarios

Scenario	Implied missing methane from natural gas systems	Ratio of scenario to EPA natural gas system emission	Implied natural gas system emission rate
1.	1.8 Tg	1.25	1.75%
2.	3.5 Tg	1.5	2.1%
3.	7 Tg	2	2.8%
4.	14 Tg	3	4.2%

We chose these scenarios to provide the broadest range of what seems possible in light of the work by Brandt et al. The paper explicitly says that it is not likely that the 21 Tg of methane all comes from natural gas, so that total amount is not considered. The upper bound analyzed is 14 Tg extra from natural gas systems. At the low end of the range of scenarios, we analyze 1.8 Tg of extra methane coming from the natural gas system. This would be the case if, for example, the natural gas system is responsible for 25 percent of the lowest estimate of missing methane. Additionally, we consider two intermediate scenarios, under which 3.5 and 7 Tg of missing methane due to natural gas systems.

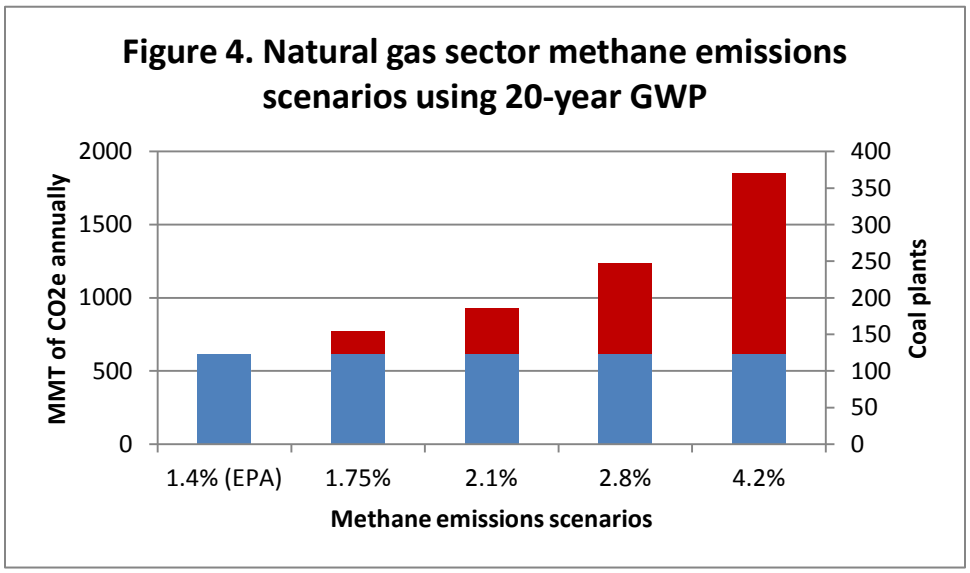
Next, we convert the methane leakage to carbon dioxide equivalent, which we use to compute an equivalency in coal plants. Coal plants comparisons are based on annual emissions using 2012 data for a generator of average efficiency, capacity factor and size for the U.S. fleet (a 543-megawatt generator operating at 85 percent capacity with a heat rate of 10,444 Btu per kilowatt-hour, from the Energy Information Administration 2013).

We use Global Warming Potential (GWP) factors to perform the conversion to CO_2 equivalent. GWP factors represent the relative contribution to global warming from GHGs other than carbon dioxide, which each have different atmospheric residence times and abilities to trap heat. All GHGs are defined in relation to carbon dioxide, the most prevalent GHG, which is assigned a GWP of one for all time periods.

Methane has an especially pronounced effect in the initial years and decades after it is released. Unlike carbon dioxide, which can continue to drive warming for hundreds or thousands of years after it is emitted, methane has an atmospheric residence time of approximately 12 years. However, while it is in the atmosphere, methane is a very potent greenhouse gas. Moreover, atmospheric chemistry transforms methane into carbon dioxide over time. The most recent Intergovernmental Panel on Climate Change (IPCC) reports GWP factors for methane of 34 over 100 years and 86 over 20 years, an increase since the prior IPCC report that reflects improved scientific understanding.

In the past, when climate change seemed like a distant problem, using 100-year GWP values was an accepted convention. The EPA inventory still refers to carbon dioxide equivalent without any reference to the timeframe with the expectation that readers will assume the numbers are on a 100-year scale. Today, with evidence of damages from climate change accumulating, there is increasing attention to near term climate disruptions. Put differently, the value of short-term climate mitigation benefits has been getting more attention from policy-makers. While carbon dioxide emissions will largely determine the extent of global warming in the long run (Harvey et al., 2013), reducing emissions of gases like methane will reduce short-run climate damages and can be used strategically to reduce peak warming (National Research Council 2011). Methane also contributes to the formation of ground-level ozone, so there are local air quality benefits to emission reductions.

This issue brief presents comparisons over both shorter and longer term time periods (20-year and 100-year GWPs). Figure 4 depicts the 20-year values in carbon dioxide equivalent (CO2e) and the comparable number of average coal plants for each of the leakage scenarios detailed in Table 1.



The first bar represents the level of methane emissions from the natural gas sector in the EPA inventory. An emissions rate of 1.4 percent implies emissions equivalent to 124 coal plants using 20-year GWP. A 1.8 percent emissions rate would imply emissions with a carbon dioxide equivalency equal to 31 additional coal plants beyond the basic inventory estimate, for a total of 155. Leakage of 4.2 percent would imply additional emissions with a carbon dioxide equivalency equal to 249 more coal plants, for a total of 373.

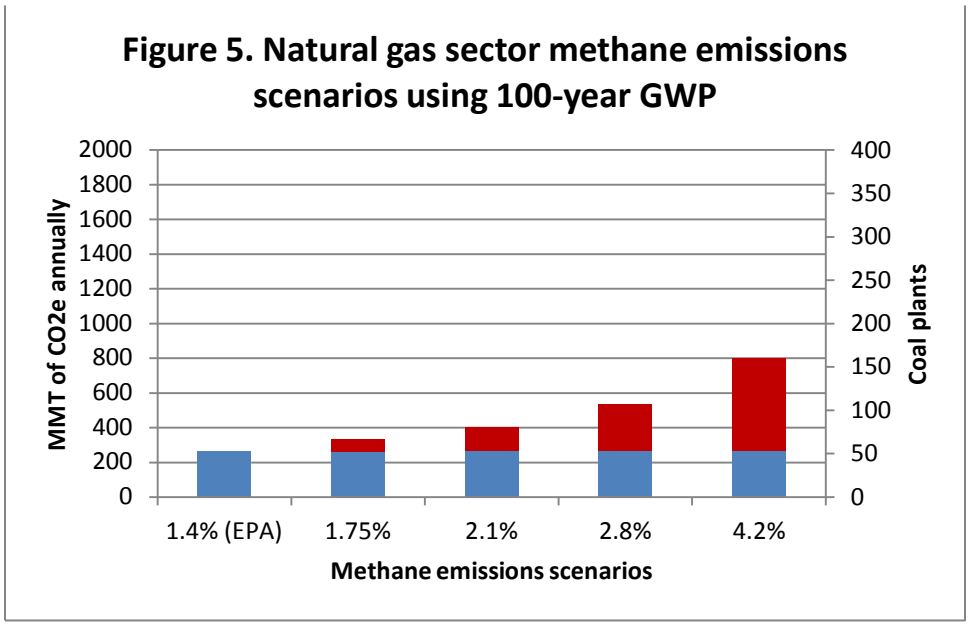


Figure 5 shows that, using 100-year GWP factors, the EPA estimate of methane leakage, 1.4 percent, has a carbon dioxide equivalency equal to 53 coal plants. A leakage rate of 1.8 percent would imply additional emissions with a carbon dioxide equivalency equal to 13 additional coal plants, for a total of

66. Doubling EPA's leakage rate to 2.8 percent results in an additional 53 coal plants, for a total of 106. A tripling of emissions to 4.2 percent would imply additional emissions with a carbon dioxide equivalency equal to 107 more coal plants, for a total of 160.

5. Implications for emissions impacts of electricity from natural gas

Proponents of natural gas have pointed to the lower carbon dioxide pollution emitted from the smokestacks of natural gas-fired electricity generators. Natural gas plants have smokestack emissions that are roughly half those of coal-fired power plants. Yet, methane emissions from the natural gas system significantly reduce this smokestack advantage. One of the reasons it is important to characterize methane emissions from the natural gas system more accurately is to provide a more accurate picture of the environmental impacts of electricity produced with natural gas. (It is worth noting that electricity generation accounted for 39 percent of natural gas consumption in 2012. Therefore, it is only appropriate to attribute that same fraction of the missing methane to electricity generated from natural gas.)

Based on the new understanding of the likely range of methane leakage provided by Brant et al., it seems very likely that substituting natural gas for coal-combustion to produce electricity actually exacerbates climate change over the short run, i.e. 20 years, and lowers greenhouse gas emissions over the long run, i.e. 100 years, (Alvarez et al. 2012). Being somewhat better than coal over a 100-year time horizon is hardly a sufficient condition to conclude that natural gas can serve as the low-carbon bridge to a clean energy future, as it is often called. In a U.S. context, it has been suggested that natural gas use will have to peak by 2030 for the Obama administration's climate goal to be achieved (Banks and Taraska 2013). From a global perspective, even those who extoll the virtues of natural gas have found that if global concentrations of carbon dioxide are to remain below 450 part per million - the level that scientists are targeting to limit the risks of dangerous climate change - then the time is very short for natural gas to serve as a useful bridge fuel (Levi 2012).

6. Conclusion

The EPA should take steps to address clear evidence that its inventory of GHG emissions is undercounting methane. In the short run, as part of finalizing the 2014 inventory, the agency should make the case for a significant effort to improve the inventory of emissions from the natural gas sector. In the longer run, the agency should develop a plan for integrating top-down data as well as new technologies that operate at ground level that can assist in leak detection and measurement. The federal government should be placing more emphasis in and devoting more resources to this effort.

Brandt et al.'s work illustrates the value of top-down measurements to provide evidence of overall emission levels over large areas. The EPA should move to collect airborne measurements into its GHG inventories. By conducting measurement campaigns, EPA will be able to obtain atmospheric data that is more comprehensive across space and time. This will enable the agency to identify aggregate emissions

levels with much greater accuracy and will help to improve confidence intervals. Current confidence intervals are much too small in light of uncertainty about the true value.

Emerging technologies can link emissions back to sources, enabling the EPA to conduct an effective ground-level measurement campaign. Infrared cameras are effective at locating leaks, and their use has been required under a recently approved Colorado regulation. Low cost stationary detectors are also under development. The newest detectors can locate leaks and estimate their magnitude from a distance, which reduces the challenge of acquiring property owner permission that bedevils direct on-site measurement.

The current oil and gas boom has been unleashed by a wave of technological innovation (directional drilling, hydraulic fracturing, and other emerging techniques, like "acidizing"). Governments need to keep pace with faster innovation on the regulatory side. New monitoring technologies are an opportunity for greater accuracy, and the EPA should move quickly to use these technologies to transform government monitoring of emissions. Better monitoring of emissions will help the EPA solve the mystery of the missing methane and provide the best objective guidance to policymakers, regulators, and society.

Acknowledgments

Thanks to Sonia Aggarwal, James Arnott, Adam Brandt, Eric Gimon, Hal Harvey, John Katzenberger, Veery Maxwell, and Jeffrey Rissman for helpful comments on this paper. Any remaining errors are the author's responsibility.

References

Alvarez, Ramon, S.W. Pacala, J.J. Winebrake, W.L. Chameides, and S.P. Hamburge. 2012. "Greater focus needed on methane leakage from natural gas infrastructure," *Proceedings of the National Academy of Sciences* 109(17): 6435-6440.

Banks, Darryl and Gwynne Taraska. 2013. *U.S. Natural-Gas Use Must Peak by 2030.* Center for American Progress: Washington, DC.

Brandt, A.R., G.A. Heath, E.A. Kort, F. O'Sullivan, G. Petron, S.M. Joraan, P. Tans, J. Wilcox, A.M. Gopstein, D. Arent, S. Wofsy, N.J. Brown, R. Bradley, G.D. Stuckey, D. Eardley, R. Harriss. 2014. "Methane Leaks from North American Natural Gas Systems," *Science* 343: 733-735.

Energy Information Administration (US Department of Energy). 2013. *Electric Power Annual*.

Harvey, Hal, Franklin Orr, and Clara Vondrich. 2013. "A Trillion Tons," Daedalus 142(1): 8-25.

Levi, Michael. 2013. "Climate consequences of natural gas as a bridge fuel," *Climatic Change* 118 (3-4): 609-623.

National Research Council. 2011. *Climate Stabilization Targets: Emissions, Concentrations, and Impacts over Decades to Millenia.* The National Academies Press: Washington, DC.

Appendix C

Gas Safety Incorporated (GSI)
16 Brook Lane
Southborough, Massachusetts 01772
774-922-4626 www.gassafetyusa.com

Report to

Damascus Citizens for Sustainability(DCS)
25 Main Street, Narrowsburg, New York 12764
mail to: P.O. Box 147, Milanville, PA 18443

Extended Report on a Preliminary Investigation of Ground-Level Ambient Methane Levels in Manhattan, New York City, New York

11 March 2013

by

Bryce F. Payne Jr.[1] and Robert Ackley[2]

[This report is subject to revision.]

EXECUTIVE SUMMARY

DCS requested that GSI extend the work effort described in our initial Report on a Preliminary Investigation of Ground-Level Ambient Methane Levels in Manhattan, New York City, New York (16 December 2012) to assess the practicality of developing an estimate of methane emissions in Manhattan. Specifically the effort was to focus on providing an estimate of methane emissions that could be used in evaluating the role of natural gas leakage in Manhattan with respect to fossil fuel dependence, climate impacts and other environmental and economic concerns.

Currently the greenhouse gas equivalence of methane is widely accepted as at least 20 times the effect of carbon dioxide over a 100-year time frame. In

---

[1] Consulting and research in environmental science since 1992. Associate Research Professor, Dept. Environmental Engineering and Earth Sciences, Wilkes University, Wilkes-Barre, PA and Senior Fellow of the Wake Forest University Center for Energy, Environment, and Sustainability, Winston-Salem, NC. bryce.payne@wilkes.edu

[2] President of Gas Safety, Inc. with 30 years experience in gas leak detection and measurement, related regulatory compliance, and training. bobackley@gassafetyusa.com

other words, leakage of 1/20th, or 5%, of the methane moving through a natural gas production-transport-distribution system will effectively double the greenhouse gas impact of the use of that natural gas. That is, leakage of only 5% of natural gas from point of production to point of use would eliminate any greenhouse gas advantage of natural gas compared to other fossil fuels. More complex efforts by others have looked into the greenhouse gas emissions advantages of using natural gas instead of other fossil fuels. It appears that those more elaborate efforts are settling in at $\leq$3.2% gas loss to leakage as the maximum leakage rate at which use of natural gas retains an advantage. Hence, the loss of even a few percent of gas during production, transport, distribution and utilization is critically important to management and planning of present and future national and international energy supply and utilization systems. Therefore, it was concluded the extended GSI work effort should be focused on the need to assess total methane emissions. The available data was from Manhattan. Among the production, transport, local distribution and utilization systems, this work addressed the collective effect of only local gas distribution and utilization systems, along with any other methane sources that might be present in Manhattan .

GSI efforts for this extended report focused on three objectives: (1) find existing estimates from industry, government or other sources, of the amount of methane being released in Manhattan, (2) develop such an estimate from the ground-level methane data collected during our preliminary investigation of methane levels in Manhattan, and (3) compare those estimates and consider their implications with regard to broader environmental and economic concerns. Since this investigation was limited to Manhattan (augmented with comparative data from the Bronx, and other areas across New York and Connecticut), ConEd is the relevant gas distribution company.

An examination of existing estimates, or methods for estimating, methane emissions led to the conclusion that such estimates have little basis in actual data. Natural gas companies are required to file yearly reports of Lost- and-Unaccounted-for (LAUF) gas. Presumably these reports would approximate the amount of gas leaked from the pipelines and other infrastructure of the reporting companies. However, the meters in those gas systems are only required to be accurate to ±2%. Each such system may contain hundreds of thousands of meters. Each meter is subject to normal wear and tear. Another problematic issue is the reported LAUF gas volume may incorporate other gas volumes by rule, contract, regulation, or for other administrative reasons. Consequently, the annual reported LAUF gas volumes should not be regarded as reliable estimates of the amounts of gas actually lost or emitted to the atmosphere. However, since the LAUF gas volume is ultimately based mostly on measurements using meters that are accurate to ±2%, it follows that long-

term average LAUF values should provide a reasonably meaningful mean with a ±2% variability. A ten-year average LAUF for ConEd was 2.2% with a range of 0.4 to 4.3%, i.e, ±2% variability. The 10-year-average-LAUF based estimate of annual methane emissions for the entire ConEd system was 2.2% or about 6.6 billion cubic feet per year.

The apparently most widely used method for estimating gas leakage and methane emissions from gas pipelines appears to be from a 1996 report by the U.S. Environmental Protection Agency and the Gas Research Institute (EPA/GRI). Estimates generated using the EPA/GRI 1996 method have such a wide confidence interval (±65%) that their general accuracy and usefulness is questionable. The report recognizes the likely importance of gas leaks that are undetectable by the standard industry leak detection practice, but the estimation method makes no attempt to account for such undetectable leaks. Finally, a related report of a more thorough study of cast iron pipelines in Brazil, suggested that the EPA/GRI method may provide estimates that are too low by almost half. Application of the EPA/GRI method to the pipeline statistics for the entire ConEd system generated an estimated methane emissions rate of 1 billion cubic feet per year, which can be meaningfully compared to the 10-year average ConEd LAUF gas estimate of 6.6 billion cubic feet per year. Since most leakage in gas delivery systems occurs from the pipes in the system, such a disparity between the EPA 1996-based estimate for ConEd pipeline leakage and the 10-year average ConEd LAUF gas volume would seem to indicate problems in one or both of those estimates.

During the research for this Report, we thoroughly reviewed the methane data collected by GSI during the previously reported Preliminary Investigation of Ground-Level Ambient Methane Levels in Manhattan. We also reviewed the meteorological literature and meteorological data available for Manhattan. Based on that information we developed a simple model (patent pending) that could process our preliminary Manhattan methane data and meteorological data from local sources to generate a preliminary estimate of total methane emissions in Manhattan. The resulting estimate was the flow of methane to the atmosphere from all sources in Manhattan. Such an estimate can be used to assess the relative importance of those emissions in terms of methane as a greenhouse gas (GHG) and the relative impact of gas service/use in Manhattan in a broader climate/GHG context. Wherever reasonable in the application of the model, input values were selected conservatively, so that any errors in the result should be to the low side.

The resulting methane emissions estimate for Manhattan alone was 8.6 billion cubic feet per year, or about 2.86% of the 300 billion cubic feet of gas handled by the entire ConEd system each year, even though Manhattan comprises only

about 5% of the land area and one-third of the customers in the ConEd service territory. There are also substantial losses that occur in the natural gas system before natural gas reaches the ConEd distribution system. It, therefore, appears inevitable that the loss of gas in the system serving NYC via ConEd is above the simple critical level of 5%, and well above the more elaborately derived critical levels of ≤3.2%. That is, the methane leakage in the system serving NYC through ConEd is likely already at a level where the methane leaked has as much or more climate impact as the remaining approximately 95% of the gas that is actually usefully burned by consumers in NYC. This necessarily raises doubts about the claimed value of natural gas as a "clean bridge fuel". Further work should be done to verify the findings we report here and to identify specific methane sources, as well as to improve natural gas leak prevention and management. Furthermore, the evidence suggests that leakage from natural gas systems has a more substantial role in climate change than was believed that has only recently begun to be appreciated.

Panoramic image looking south from the upper deck of the 'Top of the Rock' observation deck on Rockefeller Center. Image taken and assembled by Daniel Schwen on Dec 6th, 2004. {GFDL Wikipedia}

INTRODUCTION

In our initial report (dated 16 December 2012) on the preliminary investigation of ground-level ambient methane levels in Manhattan, New York City, New York we stated, “Further work is needed to determine whether an approximate estimate of the amount of methane being released to the atmosphere can be developed from the data generated by this preliminary methane survey.” To that end our efforts have focused on three objectives: (1) find existing estimates of the amount of methane being released in Manhattan from industry, government or other sources, (2) develop such an estimate from the ground-level methane data collected during our preliminary investigation, and (3) to compare those estimates and consider their implications with regard to broader environmental and economic concerns. Since this investigation was limited to Manhattan (augmented with comparative data from the Bronx, and other areas across New York and Connecticut), ConEd is the relevant gas distribution company.

Available Estimates of Methane Emissions in Manhattan

There are readily available documents that imply measurement-based estimates of methane (natural gas) releases in Manhattan have been developed.[3,4] However, review of those estimates leads to the conclusion that they are all largely based on other estimates, some periodically updated, but apparently never actual measurements of gas emissions in the field. This is presumably due in part to the historical lack of readily available, reliable approaches to actually measure methane concentrations and calculate methane emissions under field conditions.

LAUF Gas

Among the more prominent of such estimates-based-on-other-estimates would seem to be the Lost And Unaccounted For (LAUF) gas that companies are required to report to the New York State Department of Public Service (NYSDPS). Actually, the reported LAUF is a calculated number that includes volumes actually measured by meters in the gas distribution system along with various add-ins and deductions that are matters of contract, regulation, or used for operational accounting reasons. In addition to the arbitrariness of the add-ins and deductions, gas meters are only required to be accurate to ±2%. Malfunctions leading to metering errors of more than 2% can be expected to occur. It is important to realize that the estimation and reporting of LAUF gas was never intended to represent actual losses of gas from the gas distribution system, but to facilitate annual reconciliation of costs for gas purchased to revenues for gas sold while providing incentive to minimize actual loss of gas.[5] The reliability of LAUF numbers as estimates of actual gas losses is easily appreciated in the following statement (with original footnotes) found in a New

---

[3] ConEdison Gas Long Range Plan 2010-2030, December 2010 [accessed at http://www.coned.com/PublicIssues/PDF/GLRP1210c.pdf], and various ConEd annual and other reports.

[4] Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990 – 2009, USEPA, April, 2011., Annex 3 (PDF) (232 pp, 9.6 MB) – Methodological Descriptions for Additional Source or Sink Categories. [http://www.epa.gov/climatechange/ghgemissions/usinventoryreport.html].

[5] NYS DEPARTMENT OF PUBLIC SERVICE, STAFF WHITE PAPER ON LOST AND UNACCOUNTED FOR (LAUF) GAS, issued January 27, 2012. [White paper accessed at http://www.google.com/search?client=safari&rls=en&q=NYS+DEPARTMENT+OF+PUBLIC+SERVICE,+STAFF+WHITE+PAPER+ON+LOST+AND+UNACCOUNTED+FOR+(LAUF)+GAS,+Hearing+Exhibit+No.+76,+GRP-15&ie=UTF-8&oe=UTF-8]

York State Department of Public Service Staff White Paper on Lost and Unaccounted for (LAUF) Gas[3] (NYSEG is New York State Electric and Gas Corporation):

> "Negative Losses
> Staff must address negative losses because NYSEG[12] has experienced consistent negative losses for the past 3 years. Negative losses are physically impossible. However, consistent year to year calculated negative losses are possible when the offset[13] between the set of meters reading gas in and the set of meters reading gas out is negative and the natural variability is less than that offset. Additionally, natural variability in the LAUF can produce negative losses in some years for LDCs whose offset is positive.
> ---
> [12] Case 09-G-0669
> [13] Two sets of meters will never provide the same measurement. The difference between those two measurements is defined as offset."
> Note: LDCs are Local Distribution Companies

NYSEG LAUF gas values over those "past three years" (2008-2010) averaged -0.359%, while the ConEd average LAUF for the same three years was +1.249%. NYSEG is not ConEd, but gas metering and related LAUF errors inevitably affect the reported LAUF gas amounts of every company and probably in different and unforeseeable ways that change from year to year. Unaccounted for gas estimates are also reported annually to PHMSA[6]. When ten years (2002-2011) of those reported values were examined for this report, they were not the same as those stated in the NYS DPS Staff White Paper[3], presumably due to different reporting requirements. Though consistently low, the NYSEG unaccounted for gas reported to PHMSA, were never negative, ranging from 0.1% to 0.3% for the eight years 2004-2011. Though not implausible, such consistent and low numbers are interesting given that meters used in gas systems are only required to be accurate to ±2%. For the ten years 2002-2011, ConEd reported annual unaccounted for gas percentages ranging from 0.4-4.3. In contrast to the consistently low numbers of NYSEG, the ConEd numbers appear to have a variation of very close to ±2% around a mean of 2.2%. Coincidentally, 2.2% also happens to be the mean of all unaccounted for gas percentages reported to PHMSA from 2002-2011, though among those numbers individual annual reports ranged from -28% to +109%. Such examples serve to illustrate that LAUF numbers provide little if any useful insight into the actual amounts of gas lost from companies' gas distribution systems at any given time, or over a given year. Still, it is helpful to consider a bit further the implications of the average

---

[6] PHMSA – US Department of Transportation Pipeline and Hazardous Materials Safety Administration. Lost and Unaccounted for Gas reports accessed at http://www.phmsa.dot.gov/portal/site/PHMSA/menuitem.ebdc7a8a7e39f2e55cf2031050248a0c/?vgnextoid=a872dfa122a1d110VgnVCM1000009ed07898RCRD&vgnextchannel=3430fb649a2dc110VgnVCM1000009ed07898RCRD&vgnextfmt=print

unaccounted for gas percentage of 2.2%.

A Little Bit Matters

A loss of 2.2% might seem almost trivial. Each gas consumer, based on the required accuracy of the meter that measures gas consumption, can expect that they may be over or undercharged by as much as 2% anyway. Why, then, should anyone concern themselves with a loss of a few percent over the distribution system as a whole? A first answer would be a fair allocation of the monetary cost of the lost gas. In 2011 ConEd had total gas sales and transportation revenues of around 1.5 billion dollars, 2.2% of which amounts to 33 million dollars. That is a substantial amount of money and has to be accounted for and fairly allocated, a process that is regulated by the NYS Department of Public Services. Again, though, in the grand scheme of things, the consequences for each customer are relatively minor, only 0.2% more than the ±2% of metering accuracy. So, we are still left with the question, why does such a seemingly small amount matter?

There are two closely related reasons. One, it remains that, regardless of the reporting of the amounts of lost and unaccounted for gas, those reported amounts do not seem to provide a reliable indication of the actual losses of gas that are occurring. Two, when methane, which makes up over 90% of natural gas, escapes from the distribution system it can accumulate to pose direct risks of injury and property damage. A less obvious but greater global concern is the role of methane as a potent greenhouse gas. Any leakage of methane poses an effectively invisible, but potentially substantial threat to human health and the environment. These reasons provide a means of understanding why the actual amounts, and locations, of even seemingly small gas losses matter.

Even small natural gas leaks in confined spaces are dangerous, posing explosion and asphyxiation hazards. When a small underground gas leak finds a pathway to an enclosed space, such as a manhole, the gas can accumulate to explosive levels (5%-15% methane). Basements and other poorly ventilated spaces can also accumulate leaked gas to hazardous levels. Explosions related to such accumulations of leaked gas, though not common, are recurrent wherever natural gas is used. In addition, where even relatively small amounts of gas are leaked into the soil for extended periods, vegetation will be damaged, loss of urban trees being a common impact. Still, the ConEd record of gas safety with regard to direct hazards is relatively good.

ConEd, like other gas companies, has a routine program to detect, manage and repair leaks. However, the objective of such leak control programs is to detect

leaks, not measure the amount of gas lost through them. Such measurements would be impractical, especially for the potentially very large numbers of very small leaks that can be expected to develop in pipe systems that contain substantial amounts of old pipe. Over 70% of the cast iron pipe in the ConEd system is over 100 years old, and almost all was installed before 1930, i.e., is more than 80 years old.[3]

EPA Leakage Estimates for Natural Gas Pipelines

In this scenario, we are left with potentially large numbers of small leaks, and smaller numbers of larger leaks in gas pipe systems. Measurement of the gas losses that occur through such leaks is in practical terms impossible. Most of the small leaks will never be identified, let alone measured. How, then, does anyone arrive at some reasonable estimate of how much gas is being lost? In 1996 the U.S. Environmental Protection Agency (EPA) released an approach for estimating such losses.[7] This approach is of considerable importance because it has become the basis for international estimates of methane/natural gas leakage as well.[8]

The EPA approach[7] is relatively simple, based on 4 types of pipe materials, cast iron, unprotected steel, protected steel, and plastic. The estimated leak rates for the 4 types of pipe were based on data collected in a 1992 study by the EPA and the Gas Research Institute (GRI). The length of pipe of a given type in a system is multiplied by an estimated leak rate for a given length of that type pipe. For cast iron pipes, the oldest and leakiest type, the estimated leak rate is in standard cubic feet per mile of pipe per year (scf/mile-yr). That study looked at a total of 21 samples of cast iron pipe. The estimated methane leak rate for cast iron pipe was 399,867 scf/mile-yr (with a 90% confidence interval of 227,256). This was reduced by another factor intended to account for the amount of methane that would be biologically oxidized in soil before escaping into the atmosphere to produce a "Methane Emission Factor" for each type of pipe. After that reduction the estimated emission factor for cast iron pipe became 238,736 scf/mile-year (with a 90% confidence interval of 152,059).

The 90% confidence intervals and numbers of samples are mentioned in this discussion because it is important to understand how imprecise these estimates

---

[7] EPA/GRI. Methane Emissions from the Natural Gas Industry. Volume 9: Underground Pipelines. June 1996. http://www.epa.gov/gasstar/documents/emissions_report/9_underground.pdf.

[8] IPCC. 2006 IPCC Guidelines for National Greenhouse Gas Inventories. http://www.ipcc-nggip.iges.or.jp/public/2006gl/index.html>.

are. The numbers seem so imprecise that their usefulness seems questionable. The statistically strongest data set in EPA/GRI[7] was that for cast iron pipe. The data indicates that there is only 90% confidence that the true mean leak rate for cast iron pipe is somewhere in the range of 399,867±65%, that is, somewhere between 172,000 and 626,000 scf per mile of pipe per year.[9] The 90% confidence level seems low for an estimate that has implications as broad and important as this one. Accuracy is critical in estimating emissions of the second most important greenhouse gas, methane, when these estimates are being used in both national and international estimates for climate change modeling and planning of mitigation and response measures..[8] At least a 95% confidence interval would seem more traditional and appropriate to the purpose. However, back calculation from the 90% confidence levels and sample numbers in EPA/GRI[7] report indicate that the 95% confidence intervals would extend below zero for unprotected steel and plastic pipes, and would approach zero for protected steel. In fact, in the case of plastic pipe, with a high variability (range 0.008 to 61 std.cu.ft. per leak per hour) and the lowest number of samples (N=6), even at the liberal 90% confidence level, the lower limit of the confidence interval was -60,000 std.cu.ft. per leak per year, implying the impossible situation that relatively large amounts of gas could be taken in instead of emitted by leaks in plastic gas lines. One might reasonably set aside the issue of implied negative leak rates, and allow that leak rates below zero cannot occur. Even from this perspective, one is left with the predicament that the EPA/GRI[7] data for plastic pipe do not distinguish at a 90% confidence level between 260,000 scf per leak per year and no leak at all.

A Leakage Estimate from Comgas in Brazil

The EPA estimate approach is still the international norm, but more recent work reported out of Brazil provides a different picture.[10] That study by the Brazilian natural gas distribution company Comgas used a different approach to selecting samples, and a very conservative approach to disregard all suspiciously or inexplicably high leak rates. The Comgas study was apparently continuous from 2005 through at least 2009 as part of a pipe system upgrade program. Consequently, pipe sections selected for testing were each almost

---

[9] EPA/GRI[7] is not clear regarding whether a one-sided or two-sided confidence interval was used. The statement, "an overall accuracy of ±65% based on a 90% level of confidence" suggests a two-sided confidence interval was used, but repeatedly in footnotes to tables "upper bound minus the mean" may indicate a one-sided confidence interval was used. We assumed that all confidence intervals referred to in EPA/GRI[7] were two-sided.

[10] Carey Bylin et al. 2009. New measurement data has implications for quantifying natural gas losses from cast iron distribution mains. Pipeline and Gas Journal. (www.pgjonline.com).

certainly considerably larger than the minimum 20-foot sections in the EPA/GRI 1992 study[11] and were effectively more randomly selected. Random selection based on work scheduling without regard to prior detection of leaks combined with measurements of longer pipeline segments means the Comgas study would more likely measure total leakage, where the EPA/GRI approach was based on detection of leakage before testing. In the course of the Comgas work in Brazil, 912 pipe sections were tested, compared to only 21 in the EPA/GRI 1992 study. The Brazilian cast iron pipe system was reported to be otherwise comparable to the U.S. cast iron system studied by EPA/GRI in 1992. The Brazilian cast iron pipe, however, would likely be considerably younger than that in the ConEd system in which 70% of the cast iron pipe is over 100 years old. Instead of a methane leak rate of 399,867 scf/mile-yr the Brazilian study found a leak rate of 750,513 scf/mile-yr. It is interesting that though the Brazilian study may be regarded as contrasting with the EPA/GRI, in fact, it actually is statistically compatible. We back calculated the standard deviation of the EPA/GRI[7] cast iron pipe results and concluded the 750,000 scf/mile-yr appears to be within 99% confidence bound of the EPA/GRI[7] study. That is, the findings of the two studies do not seem to conflict. The Brazilian is simply a more robust, larger study that should provide a more accurate estimate and is statistically compatible with the EPA/GRI estimate.

Yet, even the higher Brazilian numbers may be too low because data from pipe sections with suspiciously or inexplicably high leak rates (>1,991,444 scf per mile per year) were excluded. The excluded data was 15.4% of the total data. The concern behind that elimination of high leak data was that such data could be caused by measurement procedural problems in the field or unmapped service lines connected to the cast iron mains. It would seem likely that leaks of this size would result in noticeable mercaptan odors and consequent leak reports. Nevertheless, it also seems reasonable that such large leaks may develop slowly and exist for some time before odor motivates reports of suspected leaks, though 15.4% of pipeline test sections seems implausibly high. The concern that such high data are due to procedural difficulties or unmapped services seems reasonable, but one avoided at the risk of entirely missing some actual large leaks. For example, if the tested sections are relatively long, there could be several moderate sized leaks that collectively cause leak rates above

---

[11] The actual lengths of cast iron pipe sections were apparently variable and not clearly specified in the 1996 EPA/GRI[7] report of the results of the 1992 EPA/GRI study of pipe leak rates: (on page 20 of that report) "The segment to be tested was either: 1) a service which was isolated … at the service-to-main connection and the customer's meter, 2) a short segment of main (at least 20 feet long) containing the detectable leak which was isolated by capping both ends, or 3) a long segment of main containing multiple leaks…isolated by capping off each end. … For cast iron pipes, a segment test approach was used since many undetected leaks are known to exist in cast iron."

the Comgas sample rejection level. Without knowing the lengths of the Comgas test sections, it is not possible to resolve this doubt. For present purposes, it is sufficient to let the Comgas test results stand as reported.

Estimates of Methane Leakage for ConEd based on EPA/GRI and Comgas Reports

Most of the oldest and leakiest pipe in many natural gas systems is cast iron. About 30% of the mains in the ConEd pipe system are cast iron, with another 30% unprotected steel, the next leakiest type. Now, using the EPA Methane Emission Factor extrapolation approach would seem reasonable enough, in fact, a practical necessity given the amount of underground pipe in natural gas distribution systems. For example, ConEd has about 1300 miles of cast iron mains, with similar amounts of unprotected steel, all of which feed eventually into hundreds of thousands of smaller service lines. Clearly the amount of gas leaking from each segment of such an extensive gas pipe system cannot be monitored continuously.[12] Given the soil conditions under the streets of Manhattan, biological oxidation of methane is probably limited. So, if one applies the (no soil methane oxidation) EPA Methane Leakage Factor of (rounded) 400,000 scf/mile-yr for cast iron mains to the 1300 miles of cast iron pipe in the ConEd system one arrives at estimated methane emissions of 520,000,000 scf/yr. If one uses the Brazilian Comgas cast iron pipe leak rate this becomes 975,000,000 scf/yr, which could also be too low.

Other Leak Sources and Other Estimates

One could similarly generate estimates for the other likely sources of gas leakage in the ConEd system in accordance with EPA estimating methods. In fact, beginning in 2010 ConEd, along with most other large emitters of greenhouse gases, has to file a report of estimated emissions of GHGs, including methane, with the EPA every year. However, during the preparation of this report only the 2010 GHG emissions report for ConEd had been filed and released by EPA. That 2010 ConEd report contained only volumes of natural

---

[12] In fact, in general any given section of pipe is checked every 1-3 years. Type 3 leaks that are detected but do not present an explosion hazard at the time of detection, and are deemed not likely to subsequently present such a hazard, are not repaired but put on a somewhat more frequent inspection schedule to assure they do not increase to a hazardous level. That is, they are left to continue leaking until they increase to an explosion hazard level or are repaired under routine leak repair efforts. Such unrepaired Type 3 leaks effectively release methane emissions without a control effort because they do not present an immediate or foreseeable explosion hazard.

gas delivered, which totaled 286,962,094,000 scf. The number of potential sources of leaked methane, besides cast iron pipe, in the ConEd system is large, perhaps explaining why the 2010 ConEd GHG emissions report to EPA is empty. For the purposes of this report, a simpler approach may serve the immediate purpose of showing that presently reported numbers are not reliable and approaches to actual measurement are needed.

Consider in this regard that through the EPA Natural Gas STAR program ConEd has been credited with reducing methane emissions by 4,393,613,000 scf cumulatively since 1993. That 18-year (or so) cumulative reduction barely makes up for somewhere between 4 and 8 years of the estimated ongoing leakage from cast iron pipes alone, depending on the leak rate factor used. ConEd reported to the EPA GasSTAR program that in its best single year, 2008, it reduced methane emissions by 158,795,000 scf. That is, in its best year, ConEd eliminated the equivalent of barely 30% of just one year of losses from the cast iron pipe alone. So, given there are still 1300 miles of cast iron pipe in the ConEd system, and there are many other potential leaks in the ConEd system, ConEd may well be losing ground with respect to overall net methane emissions. Further, if one considers that the total gas handled annually by ConEd amounts to about 300,000,000,000 scf[1], then the estimated cast iron pipe leakage alone amounts to in the range of 0.17-0.33%, and this estimate could still be low.

When Is a Leak a Leak?

When It Is Detectable.

Another matter worth considering is the functional definition of a leak. In the ConEd Long Range Gas Plan (2010)[1] there is the following statement (including associated original footnotes).

> "Con Edison also performs extensive leak repairs annually and has managed to reduce the backlog of leaks … . In 1988, the gas leak backlog was just over 15,000 leaks and year-end 2009 leaks were under 1,400. Most of the leaks in the leak backlog are Type 3[23] leaks which are not hazardous. We enter each winter with less than 100 hazardous leaks. Gas leak repairs are a major commitment of our O&M expenses. Con Edison has the highest amount of leak reports issued annually of all NYS utilities. Con Edison has committed to the NYS Public Service Commission that ConEd will maintain a leak backlog of less than 1,600[24] leaks at the end of the year.
>
> ---
>
> [23] A Type 3 leak is not immediately hazardous at the time of detection and can be reasonably expected to remain that way. However, Type 3 leaks shall be reevaluated during the next required leakage survey or annually whichever is less.
>
> [24] NYS PSC mandates a leak backlog less than 1600 leaks at the end of the year."

The contention of ConEd regarding the total number of leaks may be reasonable given industry leak detection practices, but not at all accurate in terms of actual total pipe leakage. A similar statement has to be made with respect to the previously discussed 1996 EPA/GRI report[7] providing the now widely used methane emission factors for gas pipelines.

Cast iron gas distribution (pipe) mains have been in the ground longest among all the predominant pipe types in the commercial natural gas system. EPA/GRI[7] reported that cast iron pipelines were found to be much leakier than the pipelines of the other pipe materials. The high leakage from cast iron pipes is due to large number of small leaks, "For cast iron pipes, a segment test approach was used since many undetected leaks are known to exist in cast iron." EPA/GRI[7] also reported experiments indicated 40.3% of the methane leaked from cast iron pipes was oxidized during its rise to the soil surface, but only 1.8-3.0% for the other pipe types. Soil methane oxidation rates measured around cast iron pipes were much higher than for other types because the methane leakage is spread more widely around and along cast iron pipes. For the other pipe types, detected leaks tended to be larger but fewer in number resulting in more concentrated methane and less oxidation in the soil.

So, when, then, is a leak a leak? When gas escapes from a pipeline is it like the proverbial tree falling in the forest? When gas escapes from a pipeline is it a leak, or is it not a leak until the gas company detects it? The following quote from the EPA/GRI report[7] explains the typical industry approach to detecting gas leaks.

> "Gas distribution operators use leak detection procedures to locate and classify leaks for repair. To identify a leak in a section of pipe, a portable hydrocarbon analyzer or flame ionization detector (FID) was used to screen immediately above the ground level while walking the pipeline. Any excursions above the background level (typically 2-3 ppm) may indicate a nearby leak."

However, the EPA/GRI[7] report also states that "many undetected leaks are known to exist" in cast iron gas mains. That is, there are undetectable leaks, and potentially a lot of them. Again quoting the EPA/GRI[7] report (page 20),

> "This technique was based on testing leaks which are detected using leak survey procedures (i.e., detected leaks), and may exclude smaller or more diffuse leaks that are not detected at the soil surface."

Now, having established there are undetectable leaks, and since undetectable

leaks are undetectable, they are not included in the leak counts of ConEd, or any other gas company using a similar leak detection method. Similarly, since this method was used in the EPA/GRI[7] pipeline leakage study to select pipe sections for leak testing, whether or not it accounts for any undetected leaks is unclear. That report states,

> "The leak flow rate measurement used should have accounted for all leaks in a pipe segment. … The segment of pipe tested was also surveyed to determine the number of detected leaks and the corresponding concentration of methane detected for each leak in the segment."

However, it is not clear whether or how this survey "to determine the number of detected leaks" might have included "undetectable leaks".

So, we are left with data in industry records and the widely used EPA/GRI[7] study results that by default do not seem to address "undetectable" leaks even though those records and that report clearly indicate substantial amounts of such leaks do occur. At least we do know that a leak is a leak no matter how small.

A Consideration of Undetectable Leaks

In Cast Iron Pipe

At this point one may wonder what then might an undetectable leak be like and what difference, if any, might such leaks make? The question would seem to resolve to how many undetectable leaks might there be that would escape detection by the typical industry leak detection method. Leaks are usually detected by surveying at the ground surface above a pipe with an FID instrument set to alarm if methane (actually combustible gas) levels rise above background levels. EPA/GRI[7] accepted and included in their emission factors an estimate by Southern Cross Corporation that 15% of detectable leaks are simply missed using the standard leak survey. It would seem to make sense that those 15% might be predominantly smaller, hence, harder to detect leaks.

Actual individual leak data were not provided in the EPA/GRI[7] report except for the 6 data points for plastic pipe. The lowest leak measured, hence, presumably detected, was 0.008 scf per leak per hour. It is not clear, however, that this was a leak that actually allowed detection as the next nearest leak rate, 0.700 scf per leak hour, was approaching 100 times larger. EPA/GRI[7] reported that this 0.008 scf per hour leak value was a potential statistical outlier. Coincidentally, it also happens to be the smallest of 6 data points, and,

therefore, comprises roughly the bottom 15% of the leaks, i.e., the percentage estimated to be routinely missed in leak surveys. So, if the 0.008 scf/leak-hour value is disregarded, among the remaining five data points, the next highest 3 fall in the range of 0.7-1.62 (average 1.15) scf/leak-hour. Since these are the only data immediately available, we will assume for this discussion that the smallest leak that can be reliably detected using the industry leak detection method will have a leak rate of 1 scf/leak-hour.[13] As discussed below, it matters little whether the actual undetectable leak is 1 scf per hour or considerably lower.

It would seem to follow that if two 1-scf-per-hour leaks were next to each other, then at the soil surface they would present the same methane concentration as one 2-scf-per-hour leak. That is, they would be detectable. So, then, at what distance of separation would they cease to be detectable? Gas Safety, Inc. experience with gas leak detection indicates that under a paved surface small leaks are detectable over a surrounding, roughly circular area in the range of 20-25 feet in diameter, and about half that if the soil surface is not paved over. Recall the test sections in the EPA/GRI[7] study were around 20 feet which would, therefore, imply that small ($\leq$1 scf -per-hour) leaks separated by more than 20 feet would not likely have been detected or measured in that study. To provide some notion of what such leaks might mean, one could assume there ought to be a range of such small undetectable leaks that should vary from just more than zero to just less than 1 scf per hour, which would generate an average undetectable leak size of 0.5 scf per hour.

Because undetectable leaks are undetectable, there is at present no data that provide direct indications how many there might be per length of pipe, regardless of the material the pipe is made of.[14] Nevertheless, a rough indication can be extracted from the data in the EPA/GRI[7] report. For ten reporting gas distribution companies, there was an average of 1.38 leak repairs per mile of cast iron pipe. It follows that if a repair were undertaken, then it was because a detectable leak had been found. This is actually a conservative approach because a repair implies a detected leak, but not all detected leaks are repaired (within a year of detection). EPA/GRI[7] estimated the average

---

[13] Based on decades of experience in gas pipeline leak detection, Gas Safety, Inc, considers such small leaks unlikely to be detectable by conventional gas leak surveys in open field, unpaved soil surface conditions. In urban settings, i.e., where gas lines are under paved surfaces that can cause methane to accumulate in the soil or in underground channels or spaces, a larger proportion of such leaks might be detected. The urban/rural setting of the EPA/GRI[7] sampling sites was not specified.

[14] Except for the Comgas study[7] in Brazil regarding leaks from cast iron pipes, implications of which are discussed later in this report.

number of active detectable leaks per repair was 2.14. Converting 1.38 repairs per mile to the distance between detected leaks (repairs) yields one detected leak for every 0.725 miles of pipe. Applying the EPA/GRI[7] estimate of 2.14 actual detectable leaks per repair reduces the distance between detectable leaks to 0.725/2.14=0.339 miles. Since the (presumably) total leak rate for cast iron pipe was 399,867 scf per mile per year, the total leak rate for the average length of pipe between two adjacent detectable leaks, i.e., 0.339 mile, would be (0.339 X 399,867) = 135,469 scf per year.

We are trying to develop some understanding of the potential importance of undetectable leaks. The EPA/GRI[7] cast iron leakage rate supposedly includes both detectable and undetectable leaks. So, if we deduct the rate for detectable leaks in cast iron pipe from the total leakage, we should have the rate for undetectable leaks. Unfortunately, there was no reported leak rate per leak in cast iron pipe because, as previously discussed, cast iron pipe typically has a large number of small leaks. As an alternative, we used the leak rate of 52,748 scf per leak per year for the most similar pipe, unprotected steel. Each detectable leak is on average 0.339 miles from the next, and each 0.339 miles of pipe has a total leakage of 135,469 scf per year. The undetectable leakage should be the difference between the total leakage (135,469 scf/yr) and leakage from the detectable leak (52,748 scf/yr), which is 82,721 scf per year. This then is an estimated average leakage from undetectable leaks for the pipe between each two detectable leaks, which occur on average every 0.339 miles. Converting this undetectable leakage rate to leakage per mile per year yields 244,000 scf per mile per year.

This volume of leakage would be accounted for by one undetectable 0.5-scf-per-hour leak every 95 feet along the cast iron pipeline. Perhaps, though, the actual undetectable leak size is smaller. Even if the average undetectable leak were smaller, say, 0.2 scf per hour, then the interval between undetectable leaks that would account for 82,271 scf/0.339 mile per year would be 39 feet, still farther apart than the likely 20-foot interval that might make 1-scf-per-hour leaks detectable and well beyond the ends of the 20-foot test segments used in the EPA/GRI[7] study. So, it matters little whether the threshold for leak detection is 1, 0.5 or 0.2 scf/hour, the implications of undetectable leaks remain large, at least for cast iron pipe. With regard to the plausibility of this estimate of leakage from undetectable leaks in cast iron pipe, one may consider that adding this 244,000 scf per mile per year to the EPA/GRI[7] estimated 400,000 scf per mile per year (presumably based on pipe sections with detectable leaks) generates a total estimated leakage of 644,000 scf per mile per year, still well below the 750,000 scf per mile per year total leakage actually measured in the Comgas study in Brazil.

Undetectable Leaks In Pipelines Made of Other Materials

This potential importance of undetectable leaks cannot be simply ruled inapplicable to pipes made of other materials. There seems no reason to rule out occasional minor manufacturing defects, damage during installation and due to natural underground processes and animal and human activities after installation. Indeed, unprotected steel is subject to corrosion problems, as is protected steel, though to a lesser degree. The question becomes, then, how to generate an estimate of the potential importance of undetectable leaks in steel and plastic gas lines. One approach would seem to be to again exploit the logical association of repairs to detected leaks. It was estimated above that leaks as large as 1 scf/hour and as close together as every 20-25 feet would likely be undetectable using the typical industry leak detection method. Once again referring to EPA/GRI[7], the reported repair interval for unprotected steel pipeline was 1.09 repairs per mile per year, and 0.08 for both protected steel and plastic. These can be converted, as above, to miles between adjacent repairs, which are 0.917 miles for unprotected steel and 12.5 miles for both protected steel and plastic. Now, it would seem reasonable to conclude if pipe injury/defects/etc. were causing detectable leaks in cast iron, then undetectable leaks in other pipe materials will ultimately be due to the same causes. So, if leaks have the same causes in all pipe materials, then the ratio of detectable leaks to undetectable leaks should be reasonably similar for all pipe materials.

Applying this same-ultimate-causes-for-leaks reasoning and extrapolating the estimated undetectable leakage rate for cast iron pipelines to unprotected steel pipelines yields an effective distance between detectable leaks of 0.429 miles, and an estimated leakage from undetectable leaks of 47,543 scf per year for each 0.429 miles of pipe, or 111,000 scf per year per mile of unprotected steel pipeline. Extrapolating the above approach indicates flows from undetectable leaks are likely to be <10% of those for detectable leaks in plastic and protected steel pipes. It should be borne in mind, however, that these pipe materials have not yet progressed far into their expected service lives, whereas cast iron pipes still in service are old, 70% over 100 years for ConEd. It would seem that monitoring for leaks previously regarded as undetectable would be advisable to assure environmentally safe management of natural gas leaks in a future where so much more gas and presumably so many more gas lines are expected to be in use, regardless of the pipe material.

Why Are More Accurate Measures of Natural Gas Leakage Needed?

Whether one considers the ConEd LAUF as reported to NYSDPS, or to PHMSA or

to EPA based on factors given in EPA/GRI[7], the reality is we have little reason to believe any of these estimates provide a reliable indication of how much natural gas is leaking from natural gas distribution systems, or of how much methane that leakage is releasing to the atmosphere. Hopefully it is at this point obvious to the reader that actual identification and measurement of every gas leak, or even leakage of gas from every segment of gas pipeline in service, is an impossible, and perhaps meaningless task. In the end there remain three objectives:

1. Fair and reasonable allocation of unaccounted for costs in the natural gas public service system.
2. Prevention of hazardous situations related to accumulation of leaked gas to levels that are explosive or asphyxiating (to humans, animals or plants).
3. Mitigation of the expected climate affecting impacts of methane emissions to the atmosphere.

At present there are, as already discussed, procedures in place that achieve the first two of these objectives to a reasonably satisfactory level. The third, however, is not effectively addressed at all by those approaches, and apparently inadequately by currently used estimation methods based on EPA/GRI[7].

RESULTS

An Estimate Based on Ground-Level Ambient Methane Levels

We developed a method (patent pending) to generate a preliminary estimate of total methane emissions in Manhattan from the data collected by GSI during the previously reported Preliminary Investigation of Ground-Level Ambient Methane Levels in Manhattan. The method appears to be broadly applicable to other trace gases, sites and situations. In the present case of Manhattan, such emissions estimates can be used to assess the relative importance of those emissions in terms of methane as a greenhouse gas (GHG) and the relative impact of gas service/use in Manhattan in a broader climate/GHG context. More precisely, the estimate that can be generated from the GSI Manhattan preliminary ground-level methane data is the rate of flow of methane from Manhattan to the atmosphere beyond.

The approach used is relatively simple. Only four pieces of information are needed to calculate a flow rate, in this case for methane from Manhattan into the atmosphere. What are the boundaries of the source area for the flow; in this case what are the effective boundaries for air flow to/from Manhattan? What is the concentration of methane in the air when the air enters the source

area, i.e., Manhattan? What is the methane concentration when the air exits Manhattan? How fast is the air entering/exiting Manhattan?

The GSI preliminary Manhattan methane data provide a large set of (over 700,000) measurements of the concentration of methane at various points around the island, and other areas in the vicinity and region, at various times over a period of five days. The challenge is to sort that data into subsets such that the methane concentration data can be associated with air moving into Manhattan, picking up methane in Manhattan, and then departing, and how to estimate how much air was moving during the relevant sampling times. Fortuitously, during certain parts of the GSI Manhattan preliminary methane survey winds and survey pathways occurred in such patterns that evaluation of the methane concentration in air entering and leaving Manhattan is practical. In order to enable use of that methane data, it was necessary to gather information and data from meteorological literature and monitoring and reporting programs. The times and conditions of one relevant data subset from the GSI Manhattan methane survey were as follows.

The 29 November 2012 Methane Survey Data

From roughly 4 PM to 5 PM on the afternoon of 29 November 2012 a survey run was made along the west, south, and eastern sides of Lower Manhattan near the shorelines. At that time the wind was consistent, from roughly the southwest (compass bearing 240 degrees) at 8 miles per hour. These wind conditions and that survey path provided data for distinct upwind and downwind areas along the near-shoreline areas around Lower Manhattan. The upwind data provided methane concentration of air arriving on the island, while downwind data provided methane concentration of air departing the island on the same wind direction path. The City College of New York has a robust weather monitoring program. By accessing the NYCMetNet website an estimated height for the mixing layer of the atmosphere over Manhattan for the same time period was obtained.[15] The length of the travel paths in the upwind

[15] The mixing layer is the lowermost layer of air in the atmosphere where air flows over and is influenced by the land or water surface below (see image on page 22). Above the mixing layer, winds tend to have a smoother, laminar flow, but within the mixing layer winds tend to have turbulent flows that cause most gases or aerosols released near the land or water surface to disperse rapidly laterally and vertically throughout the air to the upward limit of turbulent flow. The height of the mixing layer changes over time, but is consistent for time periods longer than necessary for the purposes of the current data interpretation effort. Height of the mixing layer and other meteorological data are accessible through the NYCMetNet, provided by the Optical Remote Sensing Laboratory of The City College of New York (ORSL), http://nycmetnet.ccny.cuny.edu.

and downwind portions of that survey run were estimated using Google Earth. These data were as follows:

Methane Concentrations in Ground-Level Air
Upwind 1.92 ppm ±0.003ppm (99.9999% Confidence Interval)
Downwind 2.165 ppm ±0.021ppm (99.9999% Confidence Interval)

Wind speed (speed of air entering/exiting Manhattan) 8 mph (11.7 feet per second)
Wind direction (from) WSW (compass bearing 240 degrees)
Manhattan wind cross-sectional length: 7 miles (36960 feet)
Mixing layer height: 2600 ft.[14]

These data can be applied in the following sequence of calculations:

To get the volume of air entering/leaving Manhattan per second:
Wind speed X wind cross-sectional length of Manhattan X mixing layer height =
11.7 ft/sec X 36960 ft X 2600 feet = 1.1 billion cubic feet per second

To get the amount of methane added while the air passed over Manhattan, take the difference between the upwind and downwind methane concentrations and apply it to the amount of air leaving Manhattan per second:
(Downwind methane concentration – Upwind concentration) X Volume of air leaving Manhattan per second =
(2.16 ppm – 1.92 ppm) X 1,100,000,000 cu.ft./sec. = 270 cubic feet per second

To get cubic feet per second of methane added by Manhattan to cubic feet of methane added per year:
Cubic feet per second added by Manhattan X 60 seconds per minute X 60 minutes per hour X 24 hours per day X 365 days per year =
270 cu.ft./sec X 60 sec/min X 60 min/hr X 24 hr/day X 365 days/ yr =
8,600,000,000 or 8.6 billion cubic feet per year.

This estimated annual methane flow rate from Manhattan is approximate. Each of the measured data values used could be a source of error. The methane data for a given time frame is highly reliable, 99.9999% confidence intervals ± <1% (0.021 ppm). However, methane concentrations in the air vary with location, time, wind, temperature, barometric pressure, humidity/precipitation, and the complex collective interactions of all these and possibly other factors. To examine the likely accuracy of the 29 November methane data used in the above Manhattan flux estimate other data subsets from the full data set were

examined. Each of these data subsets was collected at different times, covered different locations on and off Manhattan island, and occurred under different weather conditions. Nevertheless each data set is still relatively large, the smallest containing over 2000 methane data points. The following subsets were identified and examined:

Manhattan mean methane levels relative to reference area for given date					
Date (2012)	11-27	11-29	11-30	12-09	
Wind (from)	NE	WSW	NE	NNE	
Mean Methane Concentration (ppm)					
					Means over all 4 dates
Manhattan	2.079	2.165	2.345	2.261	2.213
Reference Area	1.866	1.92	2.008	2.002	1.949
Increase while over Manhattan Island	0.213	0.245	0.337	0.259	0.264
99.9999% Confidence interval for all Manhattan and Reference Area Mean Methane Concentrations was $\leq$ 1 % relative (0.002 to .022 ppm)					

On 27 November data were collected on Manhattan island that generated a mean methane level of 2.079 ppm, while the average methane level traveling to NYC was 1.866 ppm. The wind that day was out of the NE (compass bearing 50 degrees) at an average speed of 5.8 mph. On this day the wind was blowing from the area travelled to arrive in Manhattan. Hence, deducting the average methane level before arrival in Manhattan, 1.866 ppm, from that measured in Manhattan, 2.079 ppm, indicates the increase due to methane sources on Manhattan island, 0.213 ppm. This compares reasonably well with the 0.245 ppm increase due to methane sources on Manhattan island on 29 November.

Similar data subsets were available in the 30 November and 09 December data sets, each day with different wind conditions and, consequently, different upwind areas used as sources of reference methane levels. On 30 November the indicated methane concentration increase due to methane sources on Manhattan island was 0.337 ppm. On 09 December the increase was 0.259 ppm. The table above summarizes the indicated increases in methane

concentration due to sources on Manhattan island.

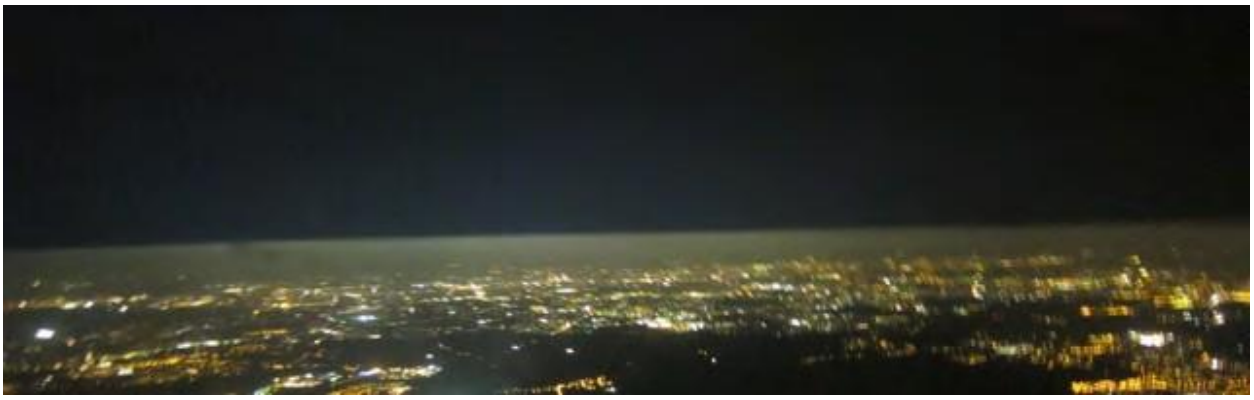
Given that these data subsets were for different survey paths on Manhattan, different reference zones off the island, and on different days, times of day and weather conditions, all effectively random, unplanned occurrences within the body of general methane survey data, the consistency of the indicated increase in methane concentration over Manhattan is actually impressive. In fact, the 99% confidence interval for the mean of the four days mean methane levels was ±0.068 ppm, or ±25%. Recall that the EPA/GRI[7] 90% confidence interval for cast iron pipelines was ±65%. For the purposes of evaluating the likely accuracy of the estimate of methane emissions on Manhattan Island, we will use ±25% as the likely accuracy of the data for increases in methane concentration in the air while passing over Manhattan Island. For data quality and field observational reasons, and to maintain a conservative approach, the 29 November data was regarded as most reliable and was used in the above calculation of annual methane flux to the atmosphere from Manhattan.

Weather data were obtained from online sources based on National Weather Service data or CCNY observations.[15] Wind speed is likely accurate to within 0.1 mph or 0.15 feet per second. Winds were moderate averaging 5.5 to 6.8 mph on the 4 survey days in the table above. The actual winds during the survey times in the table above tended to be above the average wind speed for the day. Since the data for 29 November was to be used in the calculation of the Manhattan methane flux rate, the wind speed for 4PM to 5PM on that day was estimated to be 8 mph and was the wind speed used. Potential error should not have been greater than 10% for the wind speed used in the calculation.

Wind direction was used for two purposes. One was identification of appropriate upwind methane reference areas and selection of an appropriate reference data subset within the full set of methane data. The other was to determine the length of the extent of Manhattan Island perpendicular to the direction of the wind. This length was used because the actual volume of air flowing over Manhattan should be related to the direction of the wind with respect to the greater N-S length and shorter E-W width of the island. If wind were blowing along the N-S length of the island, then, near the land surface, the band of air blowing onto and off the island would be about 2.5 miles wide. If the wind were blowing across the N-S length of the island, then the band of air would be closer 10 miles wide. So, at the land surface less air would be flowing onto and off the island for roughly N-S winds than for roughly E-W. It might seem this would cause some difficulty in that days with N or S winds would seem to have less air flowing over the island than days with E or W winds of the same speed. However, the height of the mixing layer increases with time over land compared to over water. So, this effect is probably in part

compensated by related changes in the mixing layer height. In fact, on only one (09 December) of the four days did the wind run directly along the length of the island, and on that day the mixing layer height did increase substantially to a height of approximately 7200 feet.[15]

The width of the band of air blowing over the island was the length of the projection of the profile of Manhattan onto a line perpendicular to the wind direction, which we call the cross-wind length. On three of the four days the winds were nearly opposite in direction, from either the southwest or the northeast, so the cross-wind lengths of Manhattan were very similar except on 9 December when there was a compensating increase in mixing layer height. The cross-wind length of the island for any given wind direction can be relatively easily estimated to within a few percent using Google Earth.



A nighttime image showing the mixing layer over Berlin, Germany. Aerosol particles dispersed in the mixing layer cause light from below to be diffracted/reflected revealing the mixing layer as brighter and distinct from the clear (dark), uncontaminated air in the overlying layers of the atmosphere. Ralf Steikert http://userpage.fu-erlin.de/~kyba/images/night_boundary_layer.html

Another potential error source that might affect the calculation was the thickness or height of the mixing layer (see image above). Equipment capable of measuring the height of the top of the mixing layer is not common, but such equipment is in place in Manhattan.[15] Initially, the data was obtained in a graphic format and a 5% error was assumed due to graph reading inaccuracies. The graphs were read conservatively to assure the height of the mixing layer was not overestimated. The mixing layer occasionally has a somewhat diffuse upper boundary. This occurred at 4PM-5PM on 29 November. Only the mixing height that appeared to have the same or stronger composition (backscatter) as near the land surface was used. This predisposes the height of the mixing layer to underestimation as well as the resulting estimate of the actual methane flux,

but, again, a conservative approach was preferred.[16]

Another potential source of error is the thoroughness of upward mixing of methane in the mixing layer at the time measurements were taken in the downwind sampling area, i.e., where air was leaving the island. Less than thorough mixing vertically throughout the mixing layer would seem likely if certain conditions were present. The land surface was relatively smooth, with few tall obstructions. The gas of concern was relatively dense and diffused slowly in the air. Winds were weak or inconsistent. The conditions during the relevant periods of the preliminary Manhattan methane survey were the opposite of these. Methane is lighter than air and diffuses rapidly through it, with a tendency to move upward. Winds were appreciable and consistent. With over 90 buildings more than 600 feet tall among many others of considerable height (see the image of the view from the "Top of the Rock" at the beginning of this report) the land surface of Manhattan is nearly the opposite of smooth. Further, the graphic representations of the ceilometer data for the relevant time periods indicated diffuse layers of air between the mixing layer and the overlying free atmosphere. Those diffuse layers were not included in the height of the mixing layer used in our calculations. At the time of this report, there did not appear to be reason to assume less than thorough vertical mixing of methane in the mixing layer. We anticipate opportunities to collect data that more directly address this possible source of error soon, and to revise our Manhattan methane emissions estimate in the near future.

Counter to a potential overestimate of methane emissions due to incomplete vertical mixing of methane in the mixing layer over Manhattan, there is also an unaccounted for potential loss of methane through the upper boundary of the mixing layer. Methane is only about half as dense as air, and is, therefore, strongly disposed to migrate upward in the atmosphere regardless of other conditions. It is, therefore, likely that at any given time a portion of the methane in the mixing layer is moving through the top of the mixing layer and on up into the atmosphere. Such "excessive vertical mixing" would not be accounted for in our calculations and would cause our emissions estimate to be low. We had no data on the thoroughness of vertical mixing of methane before the air in the mixing layer departs the island on the downwind side. We also have no data on what proportion of methane escapes out through the top of the mixing layer, but it seems unreasonable to expect that vertical methane loss

---

[16] In the final stages of preparation of this report, the results of the application of two different mixing layer algorithms to the raw ceilometer data were provided courtesy of Mark Arend and Yonghau Wu of the City College of New York Optical Remote Sensing Lab and made available through the NOAA CREST NYCMetNet (http://nycmetnet.ccny.cuny.edu/). The average of the twelve results (6 time intervals X 2 algorithms) for 4PM-5PM 29 November time period was 0.815 kilometers, just 0.015 kilometers over our graphic estimate of 0.8 kilometers.

would be zero. It also seems likely that either incomplete or excessive mixing may be dominant in different areas within the downwind sampling area. Ultimately we assumed both processes were in effect, the effects of both countering each other in the overall data set. That is, we assumed that on average the vertical mixing was neither incomplete nor excessive. Again, we anticipate opportunities to collect data that will help us address this possible source of error, and hope to release those findings, and update our emissions estimate at the earliest practical date.

The potential error due to inadequate or excessive vertical excessive mixing in the mixing layer could not be estimated. At the time of preparation of this report, we had found only two publications on comparable measurement-based methane emissions from another large metropolitan area.[17,18] Both were for Krakow, Poland. The first of these, Kuc et al. (2003), estimated methane emissions were around 760 million cubic feet per year (2.15×10^{-7} m^3 yr^{-1}) over the period 1996-1997. The later, Zimnoch et al. (2010), reported around 220 million cubic feet per year (6.2×10^{-6} m^3 yr^{-1}) over the period 2005-2009, an apparent 3.5-fold decrease from the 1996-1997 estimate. In the intervening years the gas service operator in Krakow had undertaken a substantial gas infrastructure improvement program, presumably substantially reducing gas leakage. The population of Krakow is about 800,000[19], while Manhattan is very close to twice that, at 1.6 million[20]. The per capita gas consumption in Poland is around 16,000 cubic feet per year[21] and for New York is around 200,000 cubic feet per year[22]. Adjusting the 1996-1997 Krakow emissions for the higher population of Manhattan and New York per capita gas consumption rate, one obtains an emissions level of 19 billion cubic feet per year. The 2005-2009 Krakow emissions adjusted to Manhattan population and NY consumption rates becomes 5.5 billion cubic feet per year. We concluded

---

[17] T. Kuc et al. 2003. Anthropogenic emissions of CO_2 and CH_4 in an urban environment. Appl. Energ. 75(3-4), 193-203.

[18] M. Zimnoch et al. 2010. Assessing surface fluxes of CO_2 and CH_4 in urban environment: a reconnaissance study in Krakow, Southern Poland. Tellus (2010), 62B, 573–580.

[19] http://www.krakow-info.com/people.htm

[20] http://www.nyc.gov/html/dcp/html/census/popcur.shtml

[21] http://www.indexmundi.com/map/?t=0&v=137000&r=eu&l=en (in cubic meters per year per capita, converted to cubic feet per year per capita)

[22] http://www.usnews.com/news/slideshows/the-10-states-that-use-the- least-energy-per-capita/11 (in BTU per capita in 2008, converted to cubic feet per capita per year)

our estimate of 8.6 billion cubic feet per year for Manhattan is reasonable in light of the estimates of Kuc (2003) and Zimnoch (2010) for Krakow.

In summary, among the measured data that were potential sources of error the 99% confidence interval of 25% relative for the methane concentration increase over Manhattan was the largest likely error. Each of the other potential sources of error were considered subject to errors of <10% relative. Further, when interpretation of data was required, those interpretations were conservative. It would seem reasonable at this point to hold that the estimated annual methane flux for Manhattan may contain an error of as much as ±25%.

Comparisons of the Estimated Emissions from Manhattan

An EPA/GRI[7]-Factors-Based Estimate

Applying the EPA/GRI[7] factors for pipe lengths and materials in the entire ConEd system[1], we arrived at an estimate of 915 million cubic feet as total gas leakage from the entire ConEd system of gas mains and service connection lines (services). Allowing an additional arbitrary 85,000,000 cubic feet for potential leakage from other ConEd gas infrastructure, we arrived at an estimated total methane leakage of around 1 billion cubic feet per year. Also, because soil conditions under Manhattan probably do not support optimal conditions for methane oxidation, we used the EPA/GRI[7] methane leakage factors instead of the methane emission factors. Use of the methane emission factors would have generated an even lower estimate of natural gas losses/ methane emissions.

An Average Long-Term LAUF Estimate

The ConEd ten-year average of LAUF gas (reported to PHMSA) was 2.2%. Even though the LAUF does not represent actual measured gas losses from the ConEd system, its preparation does involve metered gas flows albeit through many meters. Consequently, the LAUF might provide some indication of gas losses if inherent variability can be overcome, which can be accomplished by taking a long-term average. It should be kept in mind that 2.2% was the average ConEd LAUF over 10 years. As the average of 10 years this value is more reliable than the annual LAUF estimates used to calculate the average, but this greater reliability comes with costs. The average provides a more reliable estimate for leakage over times greater than one year, but may not be reliable for an individual year, say, a year impacted by a major storm. Also, leak detection and repair efforts are continuous. Use of a ten-year reporting period in order to have a reliable leakage rate would be useless with respect to annual

or more frequent efforts to identify and control leakage. For present purposes of estimating total leakage, however, the 10-year average is the best value we can extract from the reported ConEd LAUF estimates. At 2.2% the ConEd LAUF for the entire ConEd gas system that handles about 300 billion scf/yr[1] would be 6.6 billion cubic feet of lost gas, or around 6.1 billion cubic feet of methane.

The GSI Estimate Based on Preliminary Ground-Level Methane Survey Data
The actual measured levels of methane in Manhattan and adjacent areas were used to develop an estimate of the likely rate of methane emissions from the natural gas system in Manhattan. The estimate did not include any ConEd gas distribution or service beyond the shorelines of Manhattan Island. The estimate used conservative criteria in selection of which data from outside (meteorological) sources would be used to generate the estimate. The resulting estimate of total emissions of methane (functionally losses of natural gas) was 8.6 billion cubic feet per year (≈9.2 billion cubic feet of natural gas).

This estimate is 1/3 larger than the 10-year average LAUF losses and nearly 10 times greater than the methane leakage estimates using the EPA/GRI[7] factors applied to the entire ConEd system of mains and services. Given that the primary function of reported values for LAUF gas is accounting reconciliation and equitable cost allocation, the error of 33% over the long term might be acceptable. However, given that the 33% higher estimate was based on methane-in-air measurements only in Manhattan, which accounts for only about one-third of the customers and 5% of the land area in the ConEd gas service territory, the question of how much more gas may be leaking in the remainder of the ConEd gas system service area stands unaddressed. Similarly, we leave for others to discuss the implications of the difference between our estimated methane emission rate for Manhattan and the reported LAUF gas from the entire ConEd system.

The difference between the annual Manhattan methane emission rate developed from GSI methane survey data and that generated by application of the EPA/GRI[7] pipelines leakage factors is more striking. If one were to assume that the EPA/GRI[7] data did account for distribution and service gas lines leakage within the accuracy given in that report (90% confidence interval was ±65% relative), then one would would expect that the entire ConEd system might have an emission rate up to 65% greater than the above mentioned estimate of 1 billion cubic feet per year based on the EPA/GRI[7] factors. That is, at the extreme upper limit proposed by EPA/GRI[7], the methane emissions for the entire ConEd system should be something around 1.65 billion cubic feet per year. Even if one uses this upper limit of an EPA/GRI[7]-based estimate, our estimate based on actual methane measurements in Manhattan alone is still almost 6 times greater.

Again, a Little Bit Matters

Returning to the issue of how much methane leakage is of practical concern, we need to put some perspective on the 8.6 billion cubic feet per year of methane emissions that we derived from our preliminary methane data for Manhattan. To do that we will need to make some assumptions. Our first assumption is pipeline natural gas is 93% methane (EPA/GRI[7]). Our second is that natural gas pipelines are the only sources of methane emissions on Manhattan. Our third assumption is there are no natural gas leaks from the ConEd system outside of Manhattan. This third assumption is obviously not true, but allows us to put 8.6 billion cubic feet into some perspective, while assuring that our conclusion is certainly conservative. Again, for clarification, Manhattan comprises about only 5% of the land area and accounts for only about 1/3 of the customers in the ConEd service territory.

Our measurements do not distinguish between methane sources. There could be methane sources in Manhattan other than the ConEd natural gas system. Given no data on this question at present, and based on GSI experience with methane surveys over fairly broad areas of the Northeast, our opinion is that it is unlikely methane from other sources would approach 10% of the emissions level indicated by our methane survey data in Manhattan. So, for purposes of this discussion the effects of the first two assumptions counter each other, plus ≈10% due to 93% methane content of pipeline natural gas, and minus ≈10% due to other potential methane sources in Manhattan.

Putting a number on the perspective for the estimated 8.6 billion cubic feet per year methane emissions from Manhattan now requires only comparison of that volume of gas to that handled by the ConEd system as a whole, i.e., ≈300 billion cubic feet per year. So our estimated annual methane emissions for Manhattan amount to only (100 X 8.6 billion / 300 billion =) 2.86%. Once again, why does this matter?

As mentioned back in the discussion of LAUF gas, this gas loss is actually 0.66% greater than the long-term average ConEd LAUF of 2.2%. With respect to hazards of explosive concentrations of methane in susceptible locations, this amount is probably not particularly important or informative. Though it seems reasonable to conclude such risks could increase proportionately with gas leakage (methane emissions), that would seem to matter little as the ConEd leak detection and management program has been running relatively effectively for decades with no real knowledge of what actual methane emissions have been. With respect to cost reconciliation and fair allocation, using the annual ConEd gas sales and services revenue of 1.5 billion dollars, 0.66% is 9.9 million dollars, consideration of which we will leave for ConEd, its customers, and

NYSDPS. With respect to the impacts of methane as a greenhouse gas, however, there is more to be said.

Methane is a potent greenhouse gas. A widely accepted minimum relative greenhouse gas strength of methane is 21 times greater than that of carbon dioxide over a 100-year time frame.[23] There have been complex and ongoing discussions about what the greenhouse equivalence of methane actually is, which the reader may want to consult.[24] Those discussions generally are resulting in incremental increases in the accepted value for methane greenhouse gas equivalence, but for this presentation we will use the simpler approach of using the lowest widely used greenhouse equivalence for methane. For convenience, we will further lower this by rounding it to 20 times greater than that of carbon dioxide. So, if methane is approximately 20 times stronger than carbon dioxide as a greenhouse gas, and if the natural gas upon reaching its destination is entirely burned to carbon dioxide (and water), then how important are gas (methane) leaks from the natural gas production and delivery system that delivered it?

We can restate that methane as a greenhouse gas is 20 times stronger than carbon dioxide by stating that it only takes 1/20 or 5% as much methane to cause as much atmospheric warming as a given quantity of carbon dioxide. If the natural gas arrives at its intended destination and is burned, it will form carbon dioxide (and water), so its original form (as methane) does not matter since it is now carbon dioxide. However, if only 5% of natural gas escapes as it moves from within the earth through the production, transport and delivery systems, that 5% will have as much GHG impact as the other 95% burned as fuel.

FINDINGS

The findings suggest the role of leakage from natural gas systems has a more substantial role in climate change than has been appreciated.[24] Apparently present provisions in state utility regulations allow gas companies to charge their customers for up to 2% (varies by state) of their handled gas volume as lost and unaccounted for gas (discussed earlier in this report). Depending on

---

[23] http://epa.gov/climatechange/ghgemissions/gases/ch4.html, or, http://www.ipcc.ch/publications_and_data/ar4/wg1/en/ch2s2-10-2.html, among others.

[24] Alvarez, R. A., Pacala, S. W. Winebrake, J. J., Chameides, W. L. & Hamburg, S. P. Greater focus needed on methane leakage from natural gas infrastructure. Proc. Natl Acad. Sci. USA 109, 6435–6440 (2012).

the state, presumably such allowances apply to each sector of the gas system separately, i.e., production (gas wells), transportation (long distance pipelines), and distribution (gas utilities). In the end the methane emissions that affect the greenhouse gas impact of natural gas as fuel are the total methane emissions along the whole path the gas travels through the entire production-transport-distribution network. The infrastructure in each sector in that network can and does leak natural gas.

A 2.86% leakage of all the natural gas handled by ConEd in Manhattan alone leaves only 2.14% for the rest of the ConEd system, and the production and transport system feeding it, to leak collectively before total losses exceed the 5% level at which the greenhouse gas cost of using natural gas is effectively at least doubled. So far GSI efforts to gather data on volumes of gas lost by leakage or other processes in the natural gas system have indicated all such data are based on methods that are not founded in well-documented data on actual leaks, let alone actual measurements of leaks or field emissions. Some actual field data have recently been reported for production and early stage transport of shale gas. In the Denver-Julesberg Fossil Fuel Formation, largely in Weld County in northeast Colorado, emissions of methane were estimated at 2.3% to 7.7% of production.[25] Preliminary results from the Uinta Basin in Utah discussed at recent meetings of the American Geophysical Union indicated methane leakage in the field reached 9% of total production.[26] Even if the Marcellus shale gas fields planned to serve New York City release methane emissions at the lowest rate indicated by field data from northeast Colorado, and if that were added to just the GSI estimated methane emission for Manhattan alone, that would already put the total methane emission leak rate for Marcellus Shale gas delivered through the ConEd system at 5.16%. This leakage rate, which does not account for leakage from gas transmission lines to ConEd or from the rest of the ConEd system outside Manhattan, is already in excess of our simple calculation for the total leakage rate (5%) at which the leaked gas has as much potential climate impact as the burned gas. In fact, this leakage is well in excess of the total leakage rate of 3.2% at which other authors using more elaborate approaches have concluded that natural gas ceases to have a “clean fuel” advantage over coal for power production.[18]

---

[25] Gabriel Petron et al. Hydrocarbon Emissions Characterization in the Colorado Front Range – A Pilot Study. National Oceanic and Atmospheric Administration, Earth System Research Laboratory, Boulder, Colorado, USA. (Nature 482, 139–140; 2012)

[26] http://www.nature.com/news/methane-leaks-erode-green-credentials-of-natural-gas-1.12123#/ref-link-4

Caveats and Cautions Regarding the GSI Preliminary Estimate of the Manhattan Methane Emissions Rate

The GSI method (patent pending) used to estimate the Manhattan methane emissions rate from preliminary mobile methane survey data does not provide an estimate that is relative to natural background levels for natural areas in the region. It is difficult to imagine that there might even be an area anywhere in the vicinity of New York City where natural background methane emissions rates might be evaluated. The GSI approach was instead based on an alternate approach that could be evaluated because Manhattan is an island making physical boundaries of the Manhattan land surface emissions area relatively easy to define. Further, because of observations during the methane survey and analyses of the survey data, it became apparent that air arriving on the upwind and departing the downwind sides of the island at any given time necessarily provide a functional methane baseline and impacted air concentration level for the island. Hence, it is not necessary to know the natural methane baseline for the area or region, or even the surrounding waters, in order to calculate an emission rate for the island. Also, this approach eliminates any need to understand or attempt to correct off-island incoming air methane concentrations for methane sources within the geographical methane reference area since the only needed data is methane concentration in the incoming air.

The height of the mixing layer is important to the accuracy of the GSI approach to estimating area methane emissions based on ground level methane concentrations. Fortunately mixing height data is measured in Manhattan. However, the measurement used was collected at a single location not in the area where the departing air methane concentration data were collected. Nevertheless due to the mixing layer measurement location being relatively upwind from the air departure area it is more likely the mixing layer height used was too low rather than too high. Also, the measurement used was chosen to exclude diffuse zones at the upper edge of the mixing layer. Actual above ground and airborne measurements would be useful to assess variations of concentration of methane throughout the mixing layer.

There are potential and actual sources of methane in Manhattan other than the ConEd natural gas system. The GSI approach to estimating methane emissions cannot distinguish the contributions of various potential sources of methane to the overall methane emissions rate. One clearly distinguishable localized release of possible “sewer gas” was observed in the GSI Manhattan methane survey data collected at the outlet of a storm drain on the east side of the island. The elevated methane level was apparent, but not particularly high. How many other methane elevations might have been due to sewer gas or other

potential, non-ConEd, methane sources, e.g., old fill areas, is not known. However, based on GSI experience in other urban and rural areas, the effects of using conservative allowances and assumptions wherever reasonable likely exceed the influence of landfill, sewer or other biologically generated methane in the GSI Manhattan preliminary methane emissions estimate. The relative importance of biogenic methane sources in Manhattan probably could be assessed using methane isotopic composition analysis. It is also worthwhile to note that just because gas is being released from a sewer or storm drain does not necessarily confirm that the gas is actually generated in the sewage or storm water and residues. Sewers and storm drains can also receive and transport gas leaked from gas pipes.

There is also potential for losses due to pirated or illegal gas taps, and post-metering losses at the consumer level. Again, such losses cannot be distinguished within the GSI Manhattan methane emissions estimate, but seem likely to be small in comparison to leakage from ConEd gas infrastructure and operations.

RECOMMENDATIONS

The estimated Manhattan methane emission rate presented in this report indicates the need for actual measurements of methane flux for urban, petroleum and gas field areas, etc. instead of estimates based on extrapolations of typically very limited and generally indirect data.

In Manhattan, additional ground level methane survey work seems needed to support more effective and rapid detection and identification of gas leaks, to determine areas where gas pipe is in need of general replacement or lining rather than stop-gap repairs. Additional ground level work is needed that is specifically designed to develop and refine the approach developed and presented in this report for rapid actual-measurement-based estimation of methane emissions. Additional supplementary work is needed to explore and refine the level of knowledge regarding the height of the mixing layer and methane distribution within it for Manhattan and other urban and non-urban settings.

The findings from this data analysis effort indicate there is need to re-evaluate:

- Methane emissions estimates and assumptions being used as the basis for global climate modeling and projections regarding the path and speed of climate change
- Plans and projections regarding short-term high-impact opportunities to reduce greenhouse gas emissions by focusing initially on methane emissions

associated with fossil fuel development, production, and utilization
- Regulation of the fossil fuel industry
- The actual economic and environmental costs of fossil fuel compared to alternative energy technologies over all time frames.

Our findings, based on actual measurements, necessarily raise doubts about the claimed value of natural gas as a “clean, bridge fuel” and call for further work to verify the reported findings and to begin to identify specific methane sources and improve natural gas leak prevention and management.

ACKNOWLEDGEMENTS

We want to express our appreciation to the scientists and other colleagues who reviewed drafts and provided comments and suggestions during preparation of this report.

Appendix D

Environmental Pollution xxx (2012) 1e4

Contents lists available at SciVerse ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Rapid communication

Mapping urban pipeline leaks: Methane leaks across Boston

Q1 Nathan G. Phillips [a,*], Robert Ackley [b], Eric R. Crosson [c], Adrian Down [d], Lucy R. Hutyra [a], Max Brondfield [a], Jonathan D. Karr [d], Kaiguang Zhao [d], Robert B. Jackson [d]

[a] Boston University, Department of Earth and Environment, 675 Commonwealth Avenue, Boston, MA 02215, USA
[b] Gas Safety, Inc., Southborough, MA 01772, USA
[c] Picarro, Inc., Santa Clara, CA 95054, USA
[d] Duke University, Nicholas School of the Environment and Center on Global Change, Durham, NC 27708, USA

article info

Article history:
Received 25 July 2012
Received in revised form
31 October 2012
Accepted 3 November 2012

Keywords:
Carbon isotopes
Infrastructure
Methane
Natural gas
Urban

abstract

Natural gas is the largest source of anthropogenic emissions of methane (CH_4) in the United States. To assess pipeline emissions across a major city, we mapped CH_4 leaks across all 785 road miles in the city of Boston using a cavity-ring-down mobile CH_4 analyzer. We identified 3356 CH_4 leaks with concentrations exceeding up to 15 times the global background level. Separately, we measured $d^{13}CH_4$ isotopic signatures from a subset of these leaks. The $d^{13}CH_4$ signatures (mean ¼ -42.8& ± 1.3& s.e.; n ¼ 32) strongly indicate a fossil fuel source rather than a biogenic source for most of the leaks; natural gas sampled across the city had average $d^{13}CH_4$ values of -36.8& (±0.7& s.e., n ¼ 10), whereas CH_4 collected from landfill sites, wetlands, and sewer systems had $d^{13}CH_4$ signatures w20& lighter (m ¼ -57.8&, ±1.6& s.e., n ¼ 8). Repairing leaky natural gas distribution systems will reduce greenhouse gas emissions, increase consumer health and safety, and save money.

1. Introduction

Methane (CH_4) is a greenhouse gas more potent molecule for molecule than carbon dioxide (Shindell et al., 2012). In the United States, leaks of CH_4 from natural gas extraction and pipeline transmission are the largest human-derived source of emissions (EPA, 2012). However, CH_4 is not just a potent greenhouse gas; it also influences air quality and consumer health. CH_4 reacts with NO_x to catalyze ozone formation in urban areas (West et al., 2006). Incidents involving transmission and distribution pipelines for natural gas in the U. S. cause an average of 17 fatalities, 68 injuries, and $133 M in property damage each year (PHMSA, 2012). A natural gas pipeline explosion in San Bruno, CA, for instance, killed eight people and destroyed 38 homes in 2010. Detecting and reducing pipeline leaks of CH_4 and other hydrocarbons in natural gas are critical for reducing greenhouse gas emissions, improving air quality and consumer safety, and saving consumers money (West et al., 2006; Han and Weng, 2011; Shindell et al., 2012; Alvarez et al., 2012).

To assess CH_4 emissions in a major urban metropolis, we mapped CH_4 emissions over the entire 785 centerline miles of Boston's streets. To evaluate the likely source of the street-level CH_4 emissions, we also measured the $d^{13}CeCH_4$ carbon isotope composition, which can differentiate between biogenic (e.g., landfill, wetland, sewer) and thermogenic (e.g., natural gas) sources (Schoell, 1980).

2. Materials and methods

We conducted 31 mobile surveys during the period 18 August, 2011e1 October, 2011, covering all 785 road miles within Boston's city limits. We measured CH_4 concentration ($[CH_4]$, ppm) using a mobile Picarro G2301 Cavity Ring-Down Spectrometer equipped with an A0491 Mobile Plume Mapping Kit (Picarro, Inc, Santa Clara, CA). This instrument was factory-calibrated on 15 August 2011, immediately prior to use in this study, and follow-up tests of the analyzer were made during 11e21 August, 2012, comparing analyzer output to a National Oceanic and Atmospheric Administration (NOAA) primary standard tank. In both pre- and post-checks, the analyzer output was found to be within 2.7 parts per billion of known $[CH_4]$ in standard tanks, three orders of magnitude below typical atmospheric concentrations. Spectrometer and mobile GPS data were recorded every 1.1 s. To correct for a short time lag between instantaneous GPS location and a delay in $[CH_4]$ measurement due to inlet tube length (w3 m), we used an auxiliary pump to increase tubing flow throughput to within 5 cm of the analyzer inlet; we also adjusted the time stamp on the $[CH_4]$ readings based on a 1-s delay observed between analyzer response to a standard CH_4 source that we injected into the instrument while driving, and the apparent GPS location. We also checked the GPS-based locations of leaks with dozens of street-level sampling to confirm specific leak locations and the estimated sampling delay. Air was sampled through a 3.0 um Zefluor filter and Teflon tubing placed w30 cm above road surfaces.

For our mobile survey data, we defined a "leak" as a unique, spatially contiguous group of $[CH_4]$ observations, all values of which exceed a concentration threshold of 2.50 ppm. This was used as a threshold because it corresponded to the 90th

* Corresponding author.
E-mail address: nathan@bu.edu (N.G. Phillips).

0269-7491/$ e see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.envpol.2012.11.003

percentile of the distribution of data from all road miles driven, and, relative to global background, is w37% above 2011 mean mixing ratios observed at Mauna Loa (NOAA, 2012).

Independently of mobile street sampling of CH_4, we measured $d^{13}CH_4$ from a subset of the leaks with a Picarro G2112i Cavity Ring-Down Spectrometer (Crosson, 2008). This instrument is calibrated monthly using isotopic standards from Isometric Instruments (Victoria, BC, Canada). The instrument was checked at least once daily to ensure analyzer output was within 1& of a tank of CH_4 with $d^{13}CH_4$ measured by a private lab (Isotech Labs, IL). Samples were collected in 1-L Tedlar sampling bags with valve and septa fittings, manufactured by Environmental Supply Company (Durham, NC). A Gas Sentry CGO-321 handheld gas detector (Bascom-Turner, MA) was used to identify the area of highest ambient [CH_4] at each site sampled for $d^{13}CH_4$. Sampling bags were pre-evacuated and filled at the area of highest ambient concentration at the sampling site using a hand pump. $d^{13}CH_4$ was analyzed using a Picarro G2112i with a sample hold time typically of a few days and always less than two weeks.

At a subset of sampling sites (n ¼ 12), we collected duplicate samples in glass vials to assess potential leaking or fractionation by the Tedlar sampling bags. We also sent duplicate samples from a different subset of sampling sites (n ¼ 5) to a private lab (Isotech Labs, IL) for independent $d^{13}CH_4$ analysis. These analyses suggest no significant fractionation or bias either from the sampling bags or the Picarro G2112i analyzer. Most samples were analyzed at less than the maximum hold time of two weeks, at which bag diffusion could account for a 1.2& drift in our measurements of $d^{13}CH_4$.

We compared $d^{13}CH_4$ of these locations with samples taken from area landfills, wetlands, and the Deer Island Water Treatment Facility. Sampling equipment and procedures, as well as laboratory analyses, for landfill and wetland sites were similar to those for $d^{13}CH_4$ sampling locations described above. Samples were collected from three capped, inactive landfills (there are currently no active landfills in the Boston area). At one former landfill site, samples were collected at approximately three-month intervals between September, 2011 and April, 2012. The $d^{13}CH_4$ signature of the landfill was consistent over this period (±3.4& s.e.). At all wetland sampling sites, a plastic chamber (10 cm x 25 cm x 5 cm) connected to a sampling tube was placed over the surface of exposed moist sediment or shallow (>5 cm) water. Sediment below the chamber was disturbed gently before drawing air samples from the headspace within the chamber. The sample from the Deer Island Treatment Facility was drawn from the headspace of a sample bottle of anaerobic sludge, collected onsite by Deer Island staff for daily monitoring of the facility's anaerobic sludge digesters.

3. Results and discussion

We identified 3356 CH_4 leaks (Figs. 1 and 2) exceeding 2.50 parts per million. Surface concentrations corresponding to these leaks ranged up to 28.6 ppm, 14-times above a surface background concentration of 2.07 ppm (the statistical mode of the entire concentration distribution). Across the city, 435 and 97 independent leaks exceeded 5 and 10 ppm, respectively.

Based on their $d^{13}CH_4$ signatures, the CH_4 leaks strongly resembled thermogenic rather than biogenic sources (Fig. 3). Samples of natural gas from the gateway pipelines to Boston and from other consumer outlets in the city were statistically indistinguishable, with an average $d^{13}CH_4$ signature of -36.8& (±0.7& s.e., n ¼ 10; & vs. Vienna Pee Dee Belemnite). In contrast, CH_4 collected from landfill sites, wetlands, and sewer systems reflected a greater fractionation from microbial activity and $d^{13}CH_4$ signatures w20& lighter. Biogenic values ranged from -53.1& to -64.5& (m ¼ -57.8&, ±1.6& s.e., n ¼ 8) for samples collected in four wetlands, three capped landfills, and the primary sewage facility for the city, Deer Island Sewage Treatment Plant, which had the heaviest sample observed for non-natural-gas sources (-53.1&). Our results for biogenic CH_4 carbon isotope signatures are consistent with other studies of the $d^{13}CH_4$ signature of CH_4 from landfills (Bergamaschi et al., 1998; Borjesson et al., 2001) and wetlands (Hornibrook et al., 2000).

Peaks of [CH_4] detected in the road surveys strongly reflected the signature of natural gas rather than biogenic sources (Table 1). The average $d^{13}CH_4$ value for peaks was -42.8& ± 1.3& (n ¼ 32), reflecting a dominant signal from natural gas, likely altered in some cases by minor fractionation of natural gas traveling through soils and by mixing with background air ($d^{13}CH_4$ ¼ -47&; Dlugokencky et al., 2011). A minority of samples had $d^{13}CH_4$ more negative than that of background air, reflecting apparent influence of biogenic CH_4. All 32 samples emitted a distinct odor of the mercaptan additive associated with natural gas, including those with a larger apparent biogenic influence on $d^{13}CH_4$.

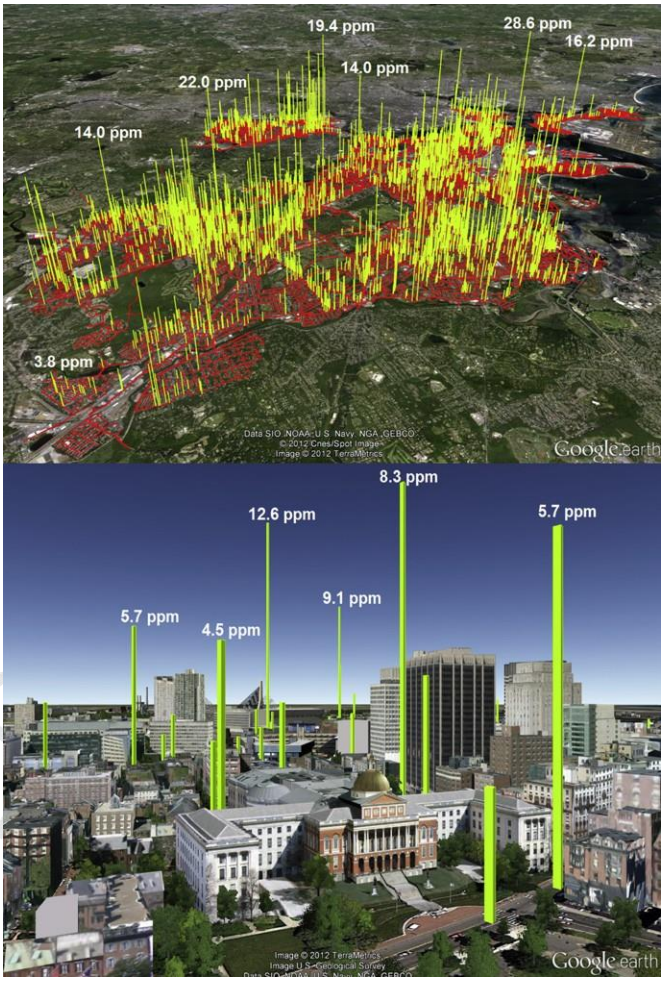


Fig. 1. Upper Panel: Methane leaks (3356 yellow spikes > 2.5 ppm) mapped on Boston's 785 road miles (red) surveyed in this study. Lower Panel: Leaks around Beacon Hill and the Massachusetts State House. Sample values of methane concentrations (ppm) are shown for each panel. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

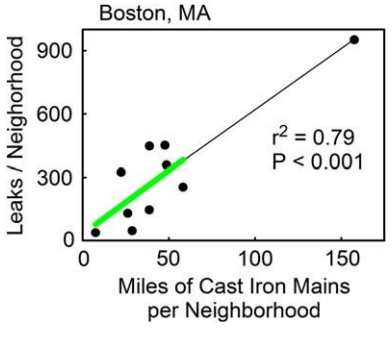


Fig. 2. Leak prevalence is associated with old cast iron pipes across ten Boston neighborhoods. (The combined line is the regression across all ten neighborhoods (P < 0.001); the green regression line [r^2 ¼ 0.34; P ¼ 0.08], which eliminates the influence of the leverage point [Dorchester neighborhood], has a slope and intercept indistinguishable (P > 0.10) from the combined regression.). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Please cite this article in press as: Phillips, N.G., et al., Mapping urban pipeline leaks: Methane leaks across Boston, Environmental Pollution (2012), http://dx.doi.org/10.1016/j.envpol.2012.11.003

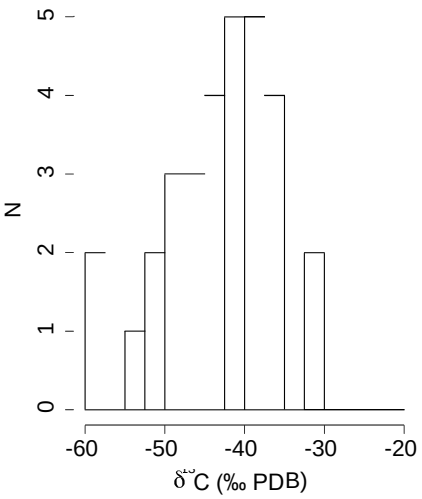


Fig. 3. $d^{13}CH_4$ of [CH_4] peaks detected in road surveys (n ¼ 32). Red lines represent means of thermogenic (-36.8&, ±0.7& s.e., n ¼ 10) and biogenic (-57.8&, ±1.6& s.e., n ¼ 8) sources, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Leaks across Boston (Fig. 1), were associated primarily with cast iron mains that were sometimes over a century old (Fig. 2). Across ten Boston neighborhoods, leak frequency was linearly related to number of miles of cast iron mains (r^2 ¼ 0.79, P < 0.001; Fig. 2), but only marginally to miles of non-cast-iron piping (r^2 ¼ 0.27; P ¼ 0.12, data not shown). Leak counts did not differ statistically by neighborhood or by socio-economic indicators for the neighborhoods obtained from the 2010 US Census (P > 0.1 for number of housing units and ethnicity) or the 2000 US Census (P > 0.1 for median income and poverty rate).

Table 1
Locations and isotopic values from discrete street leak samples.

Latitude	Longitude	$d^{13}CH_4$ (&PDB)
42.3654	-71.0612	-53.959
42.3439	-71.2628	-47.898
42.3493	-71.2265	-57.590
42.3583	-71.1749	-40.818
42.3411	-71.2440	-37.323
42.3543	-71.2441	-38.241
42.3559	-71.1898	-39.412
42.3513	-71.2092	-41.978
42.3515	-71.2081	-39.531
42.3614	-71.2314	-41.796
42.3426	-71.1012	-44.100
42.3443	-71.0949	-41.848
42.3328	-71.0761	-37.516
42.3360	-71.0738	-46.414
42.3441	-71.0673	-45.490
42.3303	-71.0569	-37.476
42.3409	-71.0542	-40.029
42.3524	-71.0445	-43.127
42.3799	-71.0272	-48.182
42.3722	-71.0361	-57.693
42.3785	-71.0681	-48.429
42.3730	-71.0632	-37.471
42.3593	-71.0629	-42.689
42.3584	-71.0644	-52.033
42.3546	-71.1271	-47.241
42.2943	-71.1891	-52.028
42.2793	-71.1514	-37.648
42.2887	-71.1428	-32.467
42.3285	-71.0792	-28.251
42.3215	-71.0692	-36.214
42.3269	-71.0796	-30.662
42.3553	-71.0573	-43.836
	Mean	-42.793
	Standard error	1.259

Reducing CH_4 leaks will promote safety and help save money. Although our study was not intended to assess explosion risks, we observed six locations where gas concentrations in manholes exceeded an explosion threshold of 4% [CH_4] at 20 °C (concentrations measured using a Gas Sentry CGO-321 handheld gas detector; Bascom-Turner, MA). Moreover, because CH_4, ethane (C_2H_6), and propane (C_3H_8) interact with NO_x to catalyze ozone formation, reducing these hydrocarbon concentrations should help reduce urban ozone concentrations and respiratory and cardiopulmonary disease (West et al., 2006; Shindell et al., 2012). CH_4 is also a potent greenhouse gas, with an estimated 20-year global warming potential 72 times greater than CO_2 (Alvarez et al., 2012; Townsend-Small et al., 2012). Replacing failing natural gas mains will reduce greenhouse gas emissions, thereby providing an additional benefit to the fewer mercury, SO_2 and particulate emissions that natural-gas burning emits compared to coal (Shindell et al., 2012). Finally, leaks contribute to $3.1 B of lost and unaccounted natural gas annually in the United States (EIA, 2012; 2005e2010 average).

Our ongoing and future research evaluates how surface [CH_4] values correspond to individual, and city-wide, urban leak rates and greenhouse-gas emissions. Two approaches to this question are useful: "bottom-up" chamber measurements taken on representative samples of individual leaks, and "top-down" atmospheric mass-balance estimates of the collective urban leak rate that exploit the known isotopic signature of natural gas versus that of biogenic sources and other fossil fuel sources. The instrumentation used in this study is well-suited for both approaches.

We propose that a coordinated campaign to map urban pipeline leaks around the world would benefit diverse stakeholders, including companies, municipalities, and consumers. Repairing the leaks will bring economic, environmental, and health benefits to all.

Acknowledgments

The Barr Foundation, Conservation Law Foundation, Picarro, Inc., Duke University's Center on Global Change and Nicholas School of the Environment, and Boston University's Sustainable Neighborhood Laboratory supported this research. Dr. Michael Delaney of the Massachusetts Water Resources Agency facilitated sewage influent sampling. Additional support was provided by the US National Science Foundation ULTRA-ex program (DEB 0948857). Shanna Cleveland, Adrien Finzi and Steven Wofsy provided helpful comments on the manuscript.

References

Alvarez, R.A., Pacala, S.W., Winebrake, J.J., Chameides, W.L., Hamburg, S.P., 2012. Greater focus needed on methane leakage from natural gas infrastructure. Proceedings of the National Academy of Sciences U.S.A. 109, 6435e6440.

Bergamaschi, P., Lubina, C., Knigstedt, R., Fischer, H., Veltkamp, A.C., Zwaagstra, O., 1998. Stable isotopic signatures (d13C, dD) of methane from European landfill sites. Journal of Geophysical Research 103, 8251e8265.

Borjesson, G., Chanton, J., Svensson, B.H., 2001. Methane oxidation in two Swedish landfill covers measured with carbon-13 to carbon-12 isotope ratios. Journal of Environmental Quality 30, 369e376.

Crosson, E.R., 2008. A cavity ring-down analyzer for measuring atmospheric levels of methane, carbon dioxide, and water vapor. Applied Physics B: Lasers and Optics 3, 403e408.

Dlugokencky, E.J., Nisbet, E.G., Fisher, R., Lowry, D., 2011. Global atmospheric methane: budget, changes and dangers. Philosophical Transactions of the Royal Society A 369, 2058e2072.

Energy Information Administration (EIA), 2012. http://205.254.135.7/dnav/ng/ng_sum_lsum_dcu_nus_a.htm, http://205.254.135.7/naturalgas/annual/pdf/table_a01.pdf, http://205.254.135.7/naturalgas/annual/archive/2009/pdf/table_a01.pdf.

Environmental Protection Agency (EPA), 2012. http://epa.gov/methane/sources.html.

Han, Z.Y., Weng, W.G., 2011. Comparison study on qualitative and quantitative risk assessment methods for urban natural gas pipeline network. Journal of Hazardous Materials 189, 509e518.

Hornibrook, E.R.C., Longstaffe, F.J., Fyfe, W.F., 2000. Evolution of stable carbon isotope compositions for methane and carbon dioxide in freshwater wetlands and other anaerobic environments. Geochimica Cosmochimica Acta 64, 1013e1027.

National Oceanic and Atmospheric Administration, Annual Greenhouse Gas Index (AGGI), 2012. Earth System Research Laboratory, Global Monitoring Division. http://www.esrl.noaa.gov/gmd/aggi/ (accessed 10.09.12.).

Pipeline and Hazardous Materials Safety Administration (PHMSA), 2012. www.phmsa.dot.gov/pipeline/library/data-stats.

Schoell, M., 1980. The hydrogen and carbon isotopic composition of methane from natural gases of various origins. Geochimica Cosmochimica Acta 44, 649e661.

Shindell, D., Kuylenstierna, J.C.I., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z., Anenberg, S.C., Muller, N., Janssens-Maenhout, G., Raes, F., Schwartz, J., Faluvegi, G., Pozzoli, L., Kupiainen, K., Höglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V., Hicks, K., Oanh, N.T.K., Milly, G., Williams, M., Demkine, V., Fowler, D., 2012. Simultaneously mitigating near-term climate change and improving human health and food security. Science 335, 183e189.

Townsend-Small, A., Tyler, S.C., Pataki, D.E., Xu, X., Christensen, L.E., 2012. Isotopic measurements of atmospheric methane in Los Angeles, California, USA: influence of "fugitive" fossil fuel emissions. Journal of Geophysical Research 117, D07308.

West, J.J., Fiore, A.M., Horowitz, L.W., Mauzerall, D.L., 2006. Global health benefits of mitigating ozone pollution with methane emission controls. Proceedings of the National Academy of Sciences U.S.A. 103, 3988e3993.

Please cite this article in press as: Phillips, N.G., et al., Mapping urban pipeline leaks: Methane leaks across Boston, Environmental Pollution (2012), http://dx.doi.org/10.1016/j.envpol.2012.11.003

Appendix E

Quantifying sources of methane using light alkanes in the Los Angeles basin, California

J. Peischl,[1,2] T. B. Ryerson,[2] J. Brioude,[1,2] K. C. Aikin,[1,2] A. E. Andrews,[3] E. Atlas,[4] D. Blake,[5] B. C. Daube,[6] J. A. de Gouw,[1,2] E. Dlugokencky,[3] G. J. Frost,[1,2] D. R. Gentner,[7] J. B. Gilman,[1,2] A. H. Goldstein,[7,8] R. A. Harley,[7] J. S. Holloway,[1,2] J. Kofler,[1,3] W. C. Kuster,[1,2] P. M. Lang,[3] P. C. Novelli,[3] G. W. Santoni,[6] M. Trainer,[2] S. C. Wofsy,[6] and D. D. Parrish[2]

Received 26 November 2012; revised 10 April 2013; accepted 12 April 2013; published 28 May 2013.

[1] Methane (CH_4), carbon dioxide (CO_2), carbon monoxide (CO), and C_2–C_5 alkanes were measured throughout the Los Angeles (L.A.) basin in May and June 2010. We use these data to show that the emission ratios of CH_4/CO and CH_4/CO_2 in the L.A. basin are larger than expected from population-apportioned bottom-up state inventories, consistent with previously published work. We use experimentally determined CH_4/CO and CH_4/CO_2 emission ratios in combination with annual State of California CO and CO_2 inventories to derive a yearly emission rate of CH_4 to the L.A. basin. We further use the airborne measurements to directly derive CH_4 emission rates from dairy operations in Chino, and from the two largest landfills in the L.A. basin, and show these sources are accurately represented in the California Air Resources Board greenhouse gas inventory for CH_4. We then use measurements of C_2–C_5 alkanes to quantify the relative contribution of other CH_4 sources in the L.A. basin, with results differing from those of previous studies. The atmospheric data are consistent with the majority of CH_4 emissions in the region coming from fugitive losses from natural gas in pipelines and urban distribution systems and/or geologic seeps, as well as landfills and dairies. The local oil and gas industry also provides a significant source of CH_4 in the area. The addition of CH_4 emissions from natural gas pipelines and urban distribution systems and/or geologic seeps and from the local oil and gas industry is sufficient to account for the differences between the top-down and bottom-up CH_4 inventories identified in previously published work.

Citation: Peischl, J., et al. (2013), Quantifying sources of methane using light alkanes in the Los Angeles basin, California, J. Geophys. Res. Atmos., 118, 4974–4990, doi:10.1002/jgrd.50413.

1. Introduction

[2] In California, methane (CH_4) emissions are regulated by Assembly Bill 32, enacted into law as the California Global Warming Solutions Act of 2006, requiring the state's greenhouse gas (GHG) emissions in the year 2020 not to exceed 1990 emission levels. To this end, the California Air Resources Board (CARB) was tasked with compiling and verifying an inventory of GHG emissions for the state. Two published works [Wunch et al., 2009; Hsu et al., 2010] have concluded that atmospheric emissions of CH_4 in the Los Angeles (L.A.) area were greater than expected from a per capita apportionment of the statewide 2006 CARB GHG inventory and from a bottom-up accounting of CH_4 sources, respectively.

[3] Several recent works have estimated CH_4 emissions to the South Coast Air Basin (SoCAB; Figure 1a), which are summarized in Table 1. Wunch et al. [2009] used a Fourier transform infrared spectrometer at the Jet Propulsion Laboratory (JPL) in Pasadena, California to measure vertically integrated total column enhancement ratios of

[1]Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, Colorado, USA.
[2]Chemical Sciences Division, National Oceanic and Atmospheric Administration Earth System Research Laboratory, Boulder, Colorado, USA.
[3]Global Monitoring Division, National Oceanic and Atmospheric Administration Earth System Research Laboratory, Boulder, Colorado, USA.
[4]Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, Florida, USA.
[5]Department of Chemistry, University of California, Irvine, California, USA.
[6]School of Engineering and Applied Science and Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts, USA.
[7]Department of Civil and Environmental Engineering, University of California, Berkeley, California, USA.
[8]Department of Environmental Science, Policy, and Management, University of California, Berkeley, California, USA.

Corresponding author: J. Peischl, Chemical Sciences Division, National Oceanic and Atmospheric Administration Earth System Research Laboratory, Boulder, CO, USA. (jeff.peischl@noaa.gov)

2169-897X/13/10.1002/jgrd.50413

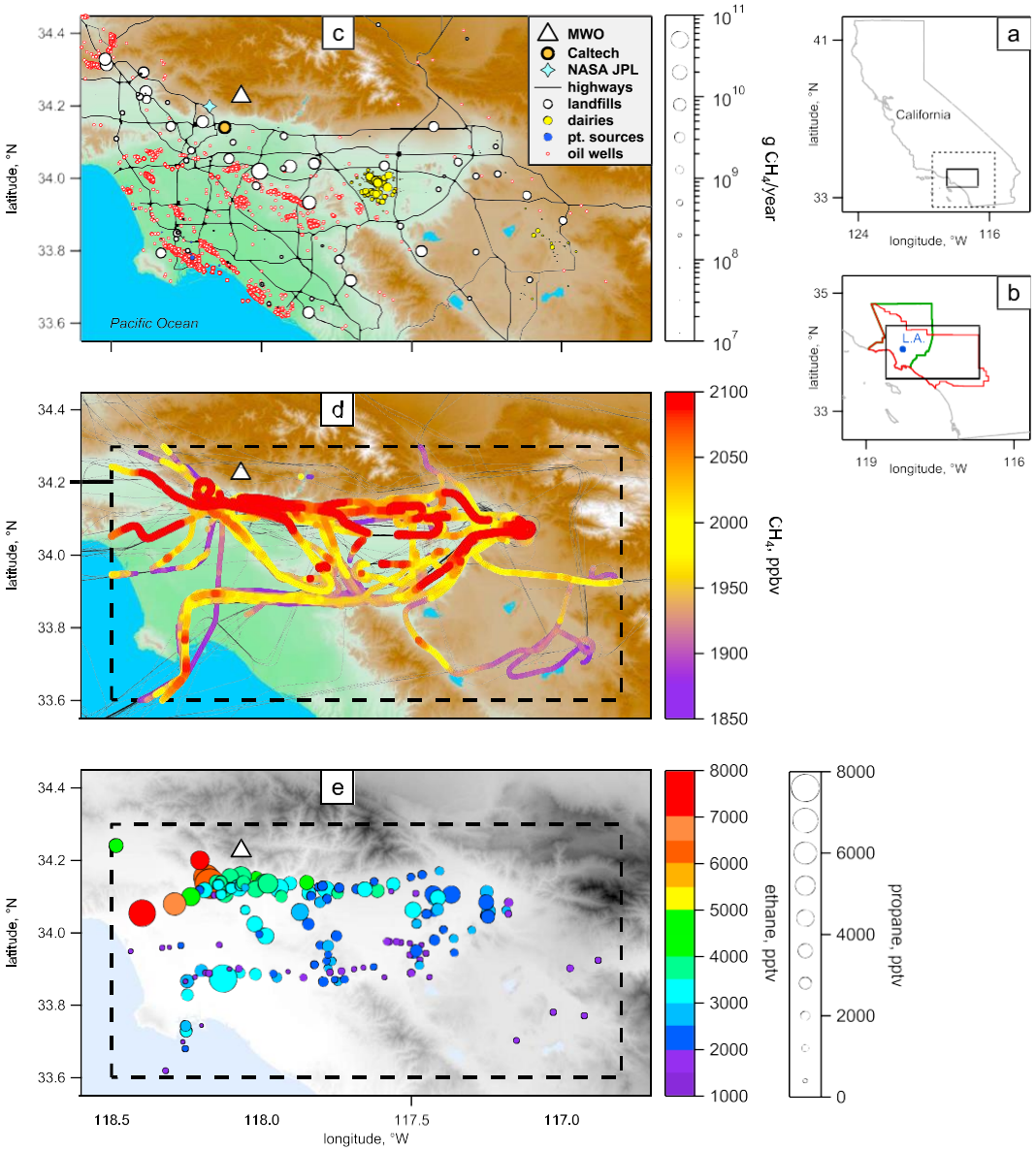


Figure 1. (a) Map of California. The dashed box shows the inset for Figure1b; the solid box shows the extent of the map boundaries for Figures 1c–1e. (b) Map of southern California showing the location of downtown L.A. (blue dot), the Los Angeles County boundary (green), the South Coast Air Basin boundary (red), and the extent of the map boundaries for Figures 1c–1e (black box). (c) Map of the L.A. region showing known sources of CH_4 in the L.A. basin. The white triangle shows the location of the Mount Wilson Observatory (MWO), where ground-based measurements were made by Hsu et al. [2010] and in this study. The light blue star shows the location of the Jet Propulsion Laboratory, where Wunch et al. [2009] made their measurements. The California Research at the Nexus of Air Quality and Climate Change (CalNex) Pasadena ground site was located on the California Institute of Technology (Caltech) campus, located at the orange-filled circle. Landfills (white circles) and CH_4 point sources (filled blue circles; negligibly small) are sized by emissions in the 2008 CARB greenhouse gas inventory. Dairies (filled yellow circles) are sized by the estimated emissions from the number of cows from Salas et al. [2008] multiplied by the 2009 CARB GHG inventory annual CH_4 emission per cow from enteric fermentation. (d) Same map of the Los Angeles region as in Figure 1c, with flight tracks from 16 daytime flights of the NOAA P-3 (thin black lines). CH_4 measurements from the daytime boundary layer are color-coded atop these tracks according to the legend to the right. (e) Locations of whole air samples in the L.A. basin are colored by ethane mixing ratio and sized by propane mixing ratio as indicated in the legends to the right. JPL, Jet Propulsion Laboratory.

Table 1. Summary of Past Studies Investigating CH_4 Emissions in the L.A. Basin

Study	Time of Study	Geographic Area	Percentage of California Population in Geographic Area	CH_4 Emission (Gg/yr)	Inventory Referenced	Bottom-up CH_4 Emission Inventory (Gg/yr)
Wunch et al. [2009]	August 2007 to June 2008	SoCAB	43%	400 ± 100 600 ± 100	CARB CO 2007 (CARB CO_2 2006 + EDGAR CO_2 2005)/2	260[b]
Hsu et al. [2010]	April 2007 to May 2008	L.A. County ∩ SoCAB	27%	200 ± 10	CARB CO 2007	140
Wennberg et al. [2012]	April 2007 to May 2008	SoCAB	43%	$380^a \pm 100$	CARB CO 2007	-
	June 2008	SoCAB	43%	470 ± 100	CARB CO 2008	-
	May 2010 to June 2010	SoCAB	43%	440 ± 100	CARB CO 2010	-

[a]Wennberg et al. [2012] recalculated the data reported by Hsu et al. [2010] to estimate a CH_4 emission from the entire SoCAB.
[b]Wunch et al. [2009] apportioned the statewide CARB GHG inventory for CH_4, less agriculture, and forestry emissions, by population.

CH_4 relative to CO and to CO_2. The observed column enhancement ratios, multiplied by CARB inventory values of CO for 2008 and an average of 2006 CARB GHG inventory and 2005 Emission Database for Global Atmospheric Research (EDGAR) for CO_2, were used to derive a lower limit to CH_4 emissions of 400 ± 100 Gg CH_4/yr (based on CO) or 600 ± 100 Gg CH_4/yr (based on CO_2) for the SoCAB. One reason for the discrepancy in their top-down analysis was that their observed CO/CO_2 enhancement ratio of 11 ± 2 ppb CO/ppm CO_2 was greater than the 8.6 ppb CO/ppm CO_2 calculated from the inventories. Wunch et al. [2009] contrasted these top-down assessments to a bottom-up estimate of 260 Gg CH_4/yr using the statewide 2006 CARB GHG inventory apportioned by population after removal of agricultural and forestry emissions, and concluded that 140–340 Gg CH_4/yr were not accounted for in the CARB CH_4 inventory for the SoCAB.

[4] Hsu et al. [2010] took a similar top-down approach and used observed atmospheric enhancement ratios of CH_4 to CO from in situ whole air samples taken at Mount Wilson (34.22°N, 118.06°W, 1770 m above sea level), scaled by the projected CARB CO inventory for 2008, to derive CH_4 emissions of 200 ± 10 Gg CH_4/yr for just the Los Angeles (L.A.) County (Figure 1b) portion of the SoCAB (L.A. County ∩ SoCAB). They used methods prescribed by the Intergovernmental Panel on Climate Change (IPCC) to create the CARB GHG inventory and reached a bottom-up estimate of 140 Gg CH_4/yr, or 60 Gg less than their top-down calculation for the L.A. County portion of the SoCAB. Hsu et al. [2010] used higher spatial resolution emissions data from CARB to construct their bottom-up inventory and therefore did not have to rely on population apportionment methods used by Wunch et al. [2009].

[5] The difference between the top-down CH_4 emissions reported by Wunch et al. [2009] and by Hsu et al. [2010] (400 Gg and 200 Gg, respectively, both based on the CARB CO inventory) are in part due to the different geographic areas for which they calculate CH_4 emissions, and in part due to differences in observed CH_4/CO enhancements between these two studies: 0.66 ± 0.12 mol/mol for Wunch et al. [2009] [Wennberg et al., 2012] and 0.52 ± 0.02 mol/mol for Hsu et al. [2010]. Both works suggested that fugitive losses of natural gas (NG) could be the source of the CH_4 missing from the bottom-up inventories.

[6] More recently, Townsend-Small et al. [2012] analyzed stable CH_4 isotope ratios in atmospheric samples taken at Mount Wilson and elsewhere in the western L.A. basin and showed they were consistent with isotope ratios in natural gas sources. Wennberg et al. [2012] used the different atmospheric ethane/CH_4 enhancement ratios observed from research aircraft during the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) field project in 2008 and the California Research at the Nexus of Air Quality and Climate Change (CalNex) field project [Ryerson, 2013] in 2010 to estimate an upper limit of 400 Gg CH_4/yr from natural gas leakage in the SoCAB. Further, their top-down analysis resulted in a calculated total emission of 440 Gg CH_4/yr in the SoCAB. Wennberg et al. [2012] also recalculated the data used by Hsu et al. [2010] to derive CH_4 emissions for the entire SoCAB and calculated a SoCAB CH_4 emission from 2008 using data from ARCTAS. The results are summarized in Table 1.

[7] Here we use ambient measurements in the SoCAB taken in May and June 2010 aboard the National Oceanic and Atmospheric Administration (NOAA) P-3 research aircraft during the CalNex field study to derive CH_4 emissions from the SoCAB using methods different from Wennberg et al. [2012]. We further examine CH_4 emissions from landfills and dairy farms in the SoCAB identified in the bottom-up CH_4 inventories reported by Hsu et al. [2010] and Wennberg et al. [2012]. We then expand on these previous studies by examining light alkane emissions from Los Angeles area data sets. In addition to CH_4 and ethane, we examine propane, n- and i-butane, and n- and i-pentane measurements to derive emissions of each of these light alkanes in the SoCAB, and use them in a system of linear equations to further quantify the source apportionment of CH_4 in the L.A. basin.

2. Measurements

[8] We use trace gas measurements from a subset of platforms and sites from the CalNex field study. The NOAA P-3 research aircraft flew all or parts of 16 daytime flights in and around the L.A. basin. Two independent measurements of CH_4 and CO_2 were made aboard the aircraft by wavelength-scanned cavity ring-down spectroscopy (WS-CRDS; Picarro 1301 m) [Peischl et al., 2012], and by quantum cascade laser direct absorption spectroscopy (QCLS) [Kort et al., 2011].

Imprecision of the 1 Hz Picarro CH_4 measurement is ±1.4 ppbv (all uncertainties herein are 1-s), and inaccuracy is estimated at ±1.2 ppbv. Imprecision of the 1 Hz QCLS CH_4 measurement is ±1 ppbv, and inaccuracy is estimated at ±15 ppbv. Imprecision of the 1 Hz Picarro CO_2 measurement is ±0.14 ppmv, and inaccuracy is estimated at ±0.12 ppmv. Imprecision of the 1 Hz QCLS CO_2 measurement is ±0.05 ppmv, and inaccuracy is estimated at ±0.10 ppmv. All CH_4 and CO_2 measurements are reported as dry air mole fractions. For this work, CH_4 and CO_2 data from the Picarro instrument are used, and QCLS CH_4 data from May 8 are used when the Picarro instrument was not operating. The 1 Hz CO data used in this analysis were measured by vacuum ultraviolet fluorescence spectroscopy [Holloway et al., 2000]. Imprecision of the 1 Hz CO data is ±1 ppbv; inaccuracy is estimated at ±5%. C_2 to C_5 alkanes, and their structural isomers, were measured in whole air samples [Colman et al., 2001], periodically filled during flight. Imprecision of these alkane measurements is ±5%; inaccuracies are estimated at ±10%. Wind measurements were derived from various sensors aboard the NOAA P-3; the uncertainty of the 1 Hz wind speed is estimated to be ±1 m/s. Sensors aboard the NOAA P-3 also measured relative humidity, ambient temperature, and potential temperature with an estimated 1 Hz uncertainty of ±0.5 °C, ±0.5 °C, and ±0.5 K, respectively.

[9] At the CalNex Pasadena ground site, located on the California Institute of Technology (Caltech) campus, measurements of C_2–C_5 alkanes were made by a gas chromatograph-mass spectrometer on 5 min integrated samples taken every half hour [Gilman et al., 2010]. Imprecision of these measurements are ±8% for ethane and ±6% for propane; inaccuracy is estimated at ±15% for each. Data from the ground site were taken between 15 May and 15 June 2010. CH_4 was not measured at the Pasadena ground site.

[10] Additionally, whole-air flask samples were taken twice daily at the Mount Wilson Observatory (MWO) for most days during May and June 2010 and analyzed for a variety of trace gas species, including CH_4, CO_2, CO, and hydrocarbons [Dlugokencky et al., 2011; Conway et al., 2011; Novelli and Masarie, 2010]. Imprecision of the CH_4 measurement is ±1 ppb; imprecision of the CO_2 measurement is ±0.1 ppm; imprecision of the CO measurement is ±1 ppbv, and inaccuracy of the CO measurement is estimated to be ±5%.

[11] We also analyze alkane data from whole air samples taken in the L.A. basin prior to 2010. Ethane and propane were measured in whole air samples taken on four flights in L.A. aboard an instrumented National Aeronautics and Space Administration (NASA) DC-8 research aircraft during ARCTAS in June 2008 [Simpson et al., 2010]. Ethane and propane were also measured on one flight in L.A. aboard the NOAA P-3 during the Intercontinental Transport and Chemical Transformation (ITCT) study in May 2002 [Schauffler et al., 1999].

3. Methods

[12] To ensure sampling from the L.A. basin, we consider aircraft data collected between 33.6 and 34.3°N latitude and 118.5 and 116.8°W longitude (Figure 1d, dashed box) in the following analysis. Aircraft data were further limited to samples taken between 1000 and 1700 PST, between 200 and 800 m above ground, and below 1400 m above sea level, to ensure daytime sampling was within the well-mixed boundary layer, which averaged 1000 ± 300 m above ground level for the daytime L.A. flights [Neuman et al., 2012]. Ground-based measurements at Pasadena were retained between 1000 and 1700 PST to ensure sampling of a well-mixed daytime boundary layer. For MWO measurements, afternoon samples, which typically occurred between 1400 and 1500 PST, were retained to capture upslope transportation from the L.A. basin [Hsu et al., 2010]. Linear fits to the data presented below are orthogonal distance regressions [Boggs et al., 1989] weighted by instrument imprecision (weighted orthogonal distance regression (ODR)). The total uncertainty in the fitted slope is calculated by quadrature addition of the fit uncertainty and the measurement uncertainties.

[13] For flux determinations, crosswind transects were flown downwind of known point sources. Enhancements of CH_4 above background levels were integrated along the flight track, and a flux was calculated using the following equation:

$$\text{flux} = n \cos(a) \int_{z_0}^{z_1} dz \int_{-y}^{y} X_m(y)\,dy \qquad (1)$$

where v cos(a) is the component of the average wind velocity normal to the flight track, n is the number density of the atmosphere, z_0 is the ground level, z_1 is the estimated boundary layer height, and X_m is the measured mixing ratio enhancement above the local background along the flight track [White et al., 1976; Trainer et al., 1995; Ryerson et al., 1998; Nowak et al., 2012]. Boundary layer heights are estimated from vertical profiles of relative humidity, ambient temperature, and potential temperature made prior to and after the crosswind transects. We assume the plume is vertically homogeneous within the mixed layer at the point of measurement, and the wind velocity is constant between emission and measurement. We estimate the uncertainty in these assumptions, combined with the uncertainties of the wind speed, wind direction, temperature, and integrated atmospheric enhancements to be ±50% for the plumes studied here [Nowak et al., 2012]. Weighted averages of the fluxes are calculated following Taylor [1997]. When calculating the CH_4 flux from dairies, CH_4 variability immediately upwind of the dairies is sufficiently large to complicate interpolation from the downwind local background. To account for this, we take the weighted ODR slope of CH_4/CO immediately upwind, multiply this ratio by the measured CO downwind of the dairies, and integrate the plume CH_4 enhancement calculated from CO ($CO \times [CH_4/CO]_{upwind}$), similar to the integrations performed by Nowak et al. [2012]. This assumes the dairies emit a negligible amount of CO.

[14] As with previously published works [Wunch et al., 2009; Hsu et al., 2010; Wennberg et al., 2012], we estimate total CH_4 emissions in the SoCAB by multiplying enhancement ratios of CH_4 to CO and CO_2 by inventory estimates of CO and CO_2 for that region:

$$E_{CH_4} = \left(\frac{CH_4}{X}\right)_{ODR\ slope} \times \left(\frac{MW_{CH_4}}{MW_X}\right) \times E_X \qquad (2)$$

where E_{CH4} is the emission of CH_4, X is either CO or CO_2, MW is the molecular weight, and E_X is the inventory

Table 2. Inventories Used in Current Analysis

Emission	Inventory	Year	Geographic Area
180 Tg CO_2/yr	CARB GHG[a]	2009	SoCAB[c]
979 Gg CO/yr	CARB[b]	2010	SoCAB
301 Gg CH_4/yr	CARB GHG[a]	2009	SoCAB[c]

[a]2009 CARB CO_2 and CH_4 emissions (http://www.arb.ca.gov/cc/inventory/data/data.htm).
[b]projected 2010 CARB CO emissions (http://www.arb.ca.gov/app/emsinv/fcemssumcat2009.php).
[c]statewide inventory apportioned by SoCAB population.

emission value of either CO or CO_2. Although not necessarily emitted from the same sources, we assume emissions of CH_4, CO, and CO_2 are well-mixed by the time they are sampled from the NOAA P-3.

[15] We use the following latest available inventories for our analysis below: the 2010 CARB emissions inventory for CO projected from the base-year 2008 inventory (http://www.arb.ca.gov/app/emsinv/fcemssumcat2009.php) and the 2009 CARB GHG inventory (http://www.arb.ca.gov/cc/inventory/data/data.htm). Both inventories were accessed in November 2012.

[16] CARB projects the total 2010 annually averaged CO emissions in the SoCAB at 979 Gg CO/yr (Table 2). We use the annually averaged CARB inventory that excludes biomass burning CO emissions because no known biomass burning events were observed in the L.A. basin during CalNex. This estimate is 4% less than the summertime CO inventory without biomass burning emissions, and approximately 6% less than the annually averaged CO inventory including biomass burning emissions used by Wennberg et al. [2012]. To estimate 2010 CH_4 emissions in the SoCAB using the 2009 CARB GHG inventory, we follow the method used by Wunch et al. [2009] and take the total statewide emission of 1525 Gg CH_4/yr, less agricultural and forestry CH_4 emissions of 898 Gg CH_4/yr, then apportion the remainder by population. In 2010, the SoCAB comprised 43% of California's population (http://www.arb.ca.gov/app/emsinv/trends/ems_trends.php). However, unlike Wunch et al. [2009], we include SoCAB dairy emissions of 31.6 Gg CH_4/yr, which are calculated in section 4.3 below. Therefore, we attribute a total of 301 Gg CH_4/yr to the SoCAB based on the 2009 CARB GHG inventory (Table 2).

[17] According to CARB's mobile source emission inventory for the Los Angeles County portion of the SoCAB (http://www.arb.ca.gov/jpub/webapp//EMFAC2011WebApp/emsSelectionPage_1.jsp), mobile source CO_2 emissions remained essentially unchanged between 2009 and 2010 (39.94 versus 39.95 Tg CO_2/yr). Additionally, the statewide CARB GHG inventory for CO_2, with out-of-state electricity generation emissions removed, decreased by less than 2% between 2008 and 2009. Therefore, we assume errors due to sampling year are negligible in examining the CO_2 emission inventories in the SoCAB from 2009 to 2010. To estimate 2010 CO_2 emissions in the SoCAB using the 2009 CARB GHG inventory, we take the total statewide emission of 465.7 Tg CO_2/yr, subtract out-of-state electricity generation of 47.9 Tg CO_2/yr, and then apportion the remainder by population. We therefore attribute 180 Tg CO_2/yr to the SoCAB using the 2009 CARB GHG inventory (Table 2). We do not compare to the Vulcan CO_2 inventory [Gurney et al., 2009] because at present, it is only available for the 2002 reporting year.

4. Results and Discussion

4.1. Total Derived Emission of CH_4 in L.A. and Comparison to Inventories

[18] In this section, we use P-3 measurements of CH_4, CO, and CO_2 to calculate enhancement ratios representative of the integrated emissions from the L.A. basin. We then use tabulated CO and CO_2 emissions taken from the CARB inventories to derive total CH_4 emissions based on enhancement ratios observed in CalNex and compare to earlier estimates of total CH_4 emissions in L.A.

[19] Figure 1c shows known stationary sources of CH_4 in the L.A. area, which include landfills, dairies, wastewater treatment facilities, and oil fields, as well as the location of measurement sites used in this study. Dairy sources are sized by estimated CH_4 emissions from enteric fermentation, as explained in section 4.3. Landfills are sized by CH_4 emissions from the 2008 CARB GHG inventory (L. Hunsaker, personal communication, 2011). Point sources are sized by 2009 CARB individual facility CH_4 emissions (https://ghgreport.arb.ca.gov/eats/carb/index.cfm) but do not stand out in the map due to their low CH_4 emissions relative to the landfills and dairies. Figure 1d shows the locations of daytime boundary-layer CH_4 data from the P-3, colored by observed mixing ratio, that were retained for the analysis as described previously. The largest concentrations of CH_4 were typically encountered along the mountains at the north edge of the L.A. basin, likely driven by transport of air within the basin, as typical daytime winds in the L.A. basin were from the west and southwest during May and June 2010 [Washenfelder et al., 2011]. CalNex CH_4 data are plotted against observed CO in Figure 2a. Weighted ODR fits to these data resulted in derived enhancement ratios of 0.74 ± 0.04 and 0.68 ± 0.03 ppbv CH_4/ppbv CO from the NOAA P-3 and MWO, respectively. We note that the same CH_4/CO enhancement ratio of 0.74 ± 0.03 was reported by Wennberg et al. [2012] using the CalNex P-3 data with different selection criteria. We include box and whisker plots in Figure 2a to show that the weighted ODR fit to the data is insensitive to the relatively few data points of higher CH_4. The ratio calculated from the CARB inventory (Table 2) is 0.54 ppb CH_4/ppb CO and is displayed for comparison.

[20] CalNex CH_4 data are plotted against observed CO_2 in Figure 2b. The slope from a weighted ODR of P-3 data is 6.70 ± 0.01 ppb CH_4/ppm CO_2 and of MWO data is 6.60 ± 0.04 ppb CH_4/ppm CO_2. The ratio of the CARB inventories from Table 2 is 4.64 ppb CH_4/ppm CO_2 and is displayed for comparison. In this case, because CH_4 and CO_2 are measured with high precision and accuracy, the largest uncertainties in interpreting the slope as an emissions ratio are likely determined by the extent of mixing of emissions from different sources within the Los Angeles air shed. Similarly, Figure 2c shows a correlation plot of CO against CO_2. The slope from a weighted ODR of P-3 data is 9.4 ± 0.5 ppb CO/ppm CO_2 and of MWO data is 10.4 ± 0.5 ppb CO/ppm CO_2. The ratio of the CARB inventories from Table 2 is 8.5 ppb CO/ppm CO_2 and is plotted

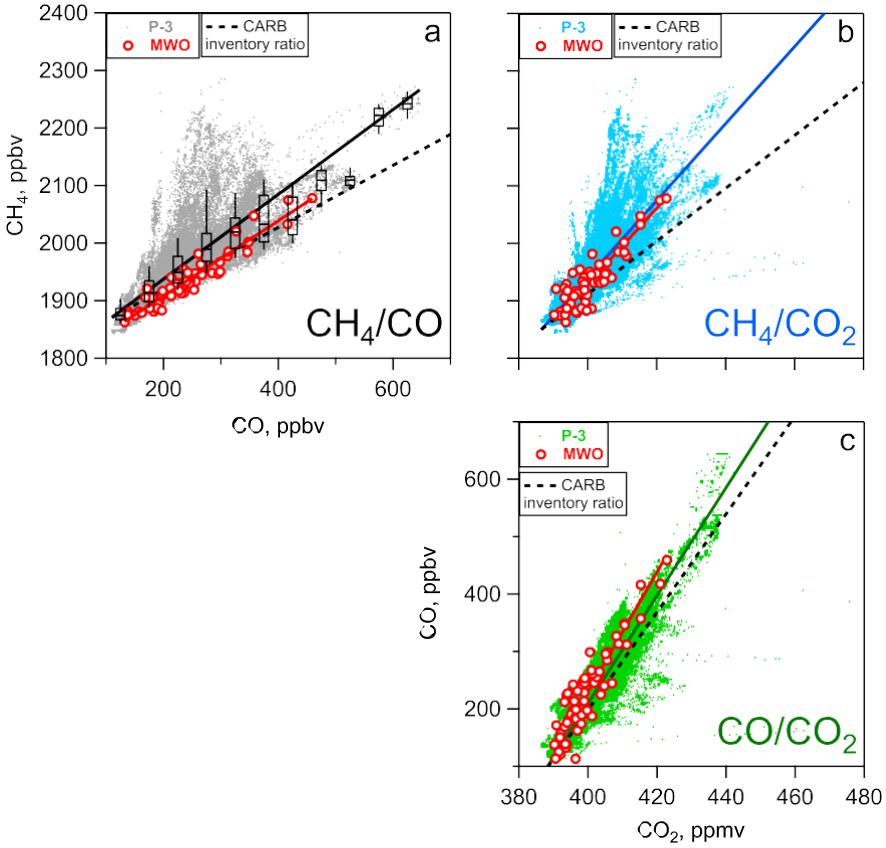


Figure 2. Scatter plots of CH_4, CO_2, and CO from all 1 s data points along flight track highlighted in Figure 1. Dots are from the NOAA P-3, while red circles are from NOAA GMD flask samples taken at the Mount Wilson Observatory during CalNex. Weighted ODRs (solid lines) result in slopes of (a) 0.74 ± 0.04 and 0.68 ± 0.04 ppb CH_4/ppb CO; (b) 6.70 ± 0.01 and 6.60 ± 0.04 ppb CH_4/ppm CO_2; and (c) 9.4 ± 0.5 and 10.4 ± 0.5 ppb CO/ppm CO_2 from the NOAA P-3 and Mount Wilson Observatory, respectively. The black dotted lines represent molar ratios of the CARB inventories listed in Table 2: CH_4:CO = 0.54, CH_4:CO_2 = 4.64×10^{-3}, and CO:CO_2 = 8.5×10^{-3}, where the background values used are the same as those determined from the fitted slopes. Also, plotted in Figure 2a are boxes (25th–75th percentiles), whiskers (10th–90th percentiles), and the median (horizontal line) for distributions of CH_4 data calculated for 50 ppbv wide bins from the NOAA P-3 CO data.

for comparison. We estimate a ±7.5% uncertainty in each of the CARB CO and CO_2 inventories, which is sufficient to explain the difference between the CO/CO_2 enhancement ratio measured from the NOAA P-3 and the ratio calculated from the CARB inventories. Quantitative agreement between emission ratios derived from P-3 and MWO data (Figures 2a–2c) is likely due to the fact that the transport within the basin was driven by the land-sea breeze, meaning typical daytime winds in the Pasadena area near Mount Wilson were from the southwest [Washenfelder et al., 2011]. This transport, and the highest values of CH_4 and CO_2 in the P-3 data that are not seen at MWO (Figures 2a and 2b), also suggests that MWO preferentially samples the western part of the L.A. basin [Hsu et al., 2010]. We therefore use enhancement ratios determined from the NOAA P-3 data to derive CH_4 emissions from the entire basin.

[21] We note that the ratio of the latest CARB CO and CO_2 inventories (Table 2) are in better agreement with ambient enhancement ratios in the CalNex data than was the case for Wunch et al. [2009]. This is likely due to either improved CARB inventories, the present use of a basin-wide data set to determine basin-wide emission ratios, or both.

[22] With the slopes and inventory values quantified, we next derive a CH_4 emission using equation (2). Using the CH_4/CO slope derived from the weighted ODR fit to the 2010 NOAA P-3 data and the projected 2010 CARB annually averaged CO emission inventory in equation (2) yields an estimated SoCAB emission of 410 ± 40 Gg CH_4/yr. The stated uncertainty is the quadrature propagation of the measurement uncertainty, errors on the slope of the ODR fit to P-3 data, and an estimated uncertainty in the CARB CO inventory. We note our derived emission of 410 ± 40 Gg CH_4/yr is similar to that derived from the P-3 data by Wennberg et al. [2012], which was 440 ± 100 Gg CH_4/yr using different selection criteria. It is further consistent with the emission derived by Wunch et al. [2009] of 400 ± 100 Gg CH_4/yr, which assumed a CARB CO inventory uncertainty of 15%. We also determine CH_4 emissions using estimates of CO_2 emissions in the SoCAB. P-3 measurements of the CH_4/CO_2 enhancement ratio observed during CalNex and SoCAB CO_2 emissions inferred from

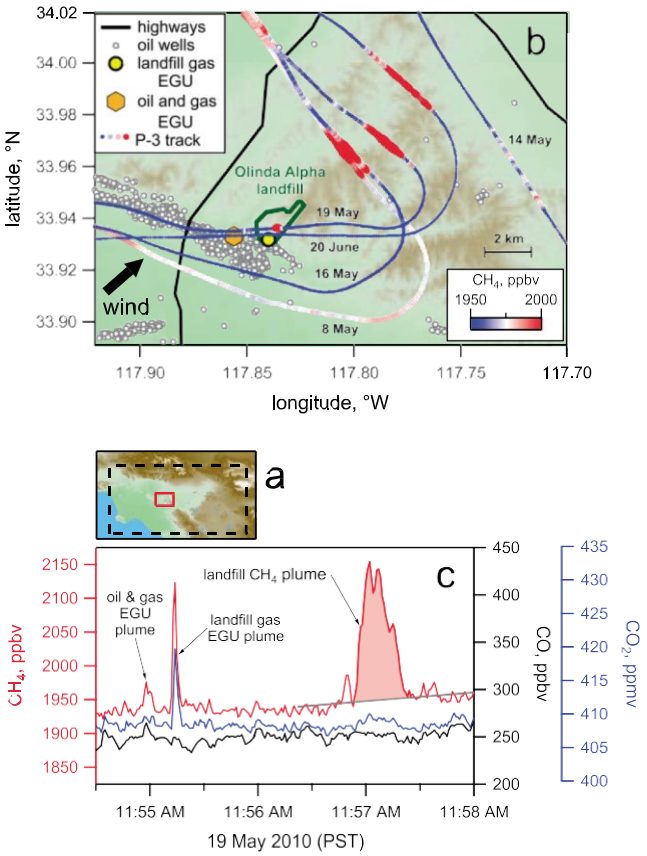


Figure 3. (a) The map from Figures 1c to 1e shows the inset for Figure 3b in red. (b) Five downwind transects, sized, and colored by CH_4 mixing ratio, showing enhancements in CH_4 downwind of the Olinda Alpha landfill (green outline). Winds were from the southwest, except on 14 May, when they were from the west and southwest. (c) Example of integration of the CH_4 plume from the 19 May flight. The filled pink area is integrated above the surrounding background (gray line). The upwind transect on this day passed downwind of two power plant (Electric Generating Unit (EGU)) plumes.

the 2009 CARB GHG inventory result in a derived CH_4 emission rate of 440 ± 30 Gg CH_4/yr, with the stated uncertainties determined by quadrature propagation of the measurement uncertainty, errors on the slope of the ODR fit to P-3 data, and an estimated uncertainty in the CARB CO_2 inventory. This value, based on the CO_2 inventory, is consistent with that derived using P-3 measurements and the CO inventory, further supporting both our assessment of uncertainties in the CARB CO and CO_2 inventories, and our assumption of sampling well-mixed emissions in the SoCAB, since any outlying CH_4 data do not affect the overall emission estimates significantly.

[23] The derived 2010 top-down SoCAB CH_4 emission of 410 and 440 Gg CH_4/yr reported here using the CARB CO or CO_2 inventories, respectively, are in quantitative agreement, in contrast to that reported for 2008 [Wunch et al., 2009]. The 2010 estimates are a factor of 1.35 to 1.45 greater than the modified population-apportioned 2009 CARB GHG inventory value of 301 Gg CH_4/yr (Table 2). A concurrent inverse modeling study by Brioude et al. [2012] has found no statistical difference between the total SoCAB CO emissions reported by CARB for 2010 and a top-down approach that estimated CO emissions in the SoCAB region using the same CO measurements used in this paper. For this reason, and for consistency with published works [Wunch et al., 2009; Hsu et al., 2010; Wennberg et al., 2012], we use 410 ± 40 Gg CH_4/yr from the top-down CH_4 assessment based on 2010 P-3 measured CH_4/CO enhancement ratios and the CARB CO inventory for the remainder of our analysis.

4.2. Methane Emissions From L.A. Basin Landfills

[24] Landfills are the largest nonfossil fuel CH_4 emission source in the bottom-up inventories compiled by Hsu et al. [2010] and by Wennberg et al. [2012], but these two studies disagree on the magnitude of this source. Hsu et al. [2010] estimated annual emissions from landfills totaled 90 Gg CH_4/yr from the Los Angeles County portion of the South Coast Air Basin. Wennberg et al. [2012] reported landfill emissions of just 86 Gg CH_4/yr for the entire South Coast Air Basin. However, that number is too low due to an error in their gridded landfill emissions inventory (P. Wennberg, personal communication, 2012) and is discarded in the following analysis.

[25] In the CARB GHG inventory, CH_4 emissions are calculated for individual landfills using methods prescribed by the IPCC and summed over all landfills to estimate a statewide total. Annual CH_4 emission values for individual landfills were obtained directly from CARB (L. Hunsaker, personal communication, 2011) to facilitate direct comparison to the P-3 data from CalNex. We use the P-3 data to calculate emissions from two of the largest CH_4-emitting landfills in the statewide GHG inventory, both of which are located in the SoCAB.

[26] The first landfill results we examine are from the Olinda Alpha landfill (33.934°N, 117.841°W) in Brea, Orange County, California. The NOAA P-3 flew five daytime boundary-layer transects on five different days downwind of this landfill (Figure 3), and a CH_4 emission flux was determined for each transect using equation (1). The results are summarized in Table 3. For the three transects when both the WS-CRDS and QCLS CH_4 instruments were sampling ambient air, flux determinations using these independent CH_4 measurements agreed within

Table 3. Landfill Emission Fluxes Determined Aboard the NOAA P-3 in 2010 From Downwind Plume Transects

Landfill	Transect Date	Flux (10^{25} molecules/s)	Flux (Gg/yr)	2008 CARB GHG Inventory[a] (Gg/yr)
Olinda Alpha	8 May	1.13	9.5	11.0
	14 May	1.45	12.2	
	16 May	1.74	14.6	
	19 May	1.61	13.5	
	20 June	2.90	24.3	
	Average[b]	1.49 ± 0.35	12.5 ± 2.9	
Puente Hills	8 May	4.29	36.0	38.8
	19 May	3.62	30.4	
	20 June	4.48	37.6	
	Average[b]	4.06 ± 1.18	34.0 ± 9.9	

[a]data from CARB (L. Hunsaker, personal communication, June 2011).
[b]weighted average, assuming a 50% uncertainty in the individual flux determinations [Taylor, 1997].

3%. In these cases, the flux was averaged and reported in Table 3. Three nearby CH_4 point sources are identified in the 2009 CARB GHG inventory: an oil and gas field power plant, which burns natural gas for fuel; the landfill power plant at Olinda Alpha, which burns landfill gas for fuel; and general stationary combustion from the landfill operations. Inventory data suggest that these three sources together emit between 0.0004 and 0.0015 Gg CH_4/yr, negligible amounts relative to CH_4 emitted directly from the landfill. On 19 May, the NOAA P-3 sampled plumes from the nearby oil and gas power plant and the landfill's power plant, both of which burn natural gas as fuel (Figure 3c). A large spike in CO_2, some CH_4, and perhaps a small amount of CO were encountered in the landfill power plant plume. However, downwind of the landfill in the large plume of CH_4, the CO_2 enhancement does not stand out significantly above the background variability. Therefore, our analysis of P-3 data supports the conclusion from the inventory that landfill CH_4 emissions dominate the observed plume enhancements downwind of Olinda Alpha landfill. Using NOAA P-3 CH_4 data from all five transects, we directly calculate a weighted average CH_4 emission flux via equation (1) of $(1.49 \pm 0.35) \times 10^{25}$ molecules/s, equal to 12.5 ± 2.9 Gg CH_4/yr assuming a constant emission, where the weights are the 50% uncertainty of each determination. For comparison, the CARB GHG inventory emission estimate from the Olinda Alpha landfill is 11.0 Gg/yr for 2008, showing agreement within the errors of the direct estimate using P-3 airborne data.

[27] The second landfill results we examine in depth are from the Puente Hills landfill (34.020°N, 118.006°W) in City of Industry, Los Angeles County, California. Of all California landfills, Puente Hills is the largest emitter of CH_4 in the 2008 CARB GHG inventory. Nearby sources of CH_4 in the 2008 CARB GHG inventory include the Puente Hills power plant (0.00045 Gg CH_4/yr) and the Savage Hills Canyon landfill (1.1 Gg CH_4/yr), both of which are small relative to the CARB GHG inventory of 39 Gg CH_4/yr emission rate for Puente Hills. The NOAA P-3 conducted three daytime boundary layer plume transects from which we determine an average emission flux of $(4.06 \pm 1.18) \times 10^{25}$ molecules/s, which extrapolates to 34.0 ± 9.9 Gg CH_4/yr assuming a constant emission (Table 3). Similar to the findings for Olinda Alpha, the CARB GHG inventory of 39 Gg CH_4/yr for the Puente Hills landfill is in agreement within the errors of the direct estimate using P-3 airborne data.

[28] Quantitative agreement between CH_4 flux estimates from the NOAA P-3 and the 2008 CARB GHG inventory for these two examples supports the use of that inventory to quantify total CH_4 emissions from landfills in the South Coast Air Basin. According to the 2008 CARB GHG inventory, CH_4 emissions from landfills totaled 117 Gg CH_4/yr in the L.A. County portion of the SoCAB, 30% higher than the 90 Gg CH_4/yr for the same geographic area using the CARB GHG inventory in 2008 reported by Hsu et al. [2010], which we attribute to different versions of the CARB GHG inventory.

[29] The 2008 CARB GHG inventory further predicts an emission from landfills of 164 Gg CH_4/yr for the entire SoCAB. On the basis of the agreement with the CARB inventory described above for the emission rates from the two landfills quantified directly by the CalNex P-3 data (50 Gg CH_4/yr, or 30% of the inventory total for the SoCAB), we assume the remaining CARB landfill CH_4 emission estimates are accurate.

4.3. Methane Emissions From L.A. Basin Dairies

[30] Salas et al. [2008] published dairy locations in California for the year 2005, with an estimate of dairy cow population for each. The locations are plotted as filled yellow circles in Figure 1c, and sized by the expected CH_4 emission from enteric fermentation according to the 2009 CARB GHG inventory (144 kg CH_4 per cow per year). According to Salas et al. [2008], all dairies in San Bernardino and Riverside counties were also located in the SoCAB, and 87% of the dairy cows in the SoCAB in 2005 were located in the Chino area (the large grouping of dairies in Figure 1c). The Chino-area dairy operations, which at one time were distributed across the Riverside-San Bernardino county line in satellite images, now appear to be located mainly in San Bernardino County as the Riverside dairies have been converted to residential neighborhoods (e.g., see Google Earth historical imagery since 2000). This declining number of dairies is confirmed by the United States Department of Agriculture (USDA) (http://www.nass.usda.gov/Statistics_by_State/California/Publications/County_Estimates/201005lvscef.pdf), which reports a decrease in dairy cows in San Bernardino and Riverside Counties from 200,000 head in 2005 to 137,500 head in 2010. In addition to dairy cows, dairies also stock immature heifers. Further, there are beef operations in the SoCAB, but these are negligible compared to the San Bernardino and Riverside dairy populations. According to the USDA, there were a total of 431,000 cattle in San Bernardino and Riverside counties in 2005, and 295,000 cattle in 2010. For both years, dairy cows represented approximately 46.5% of the cattle population in the SoCAB. From these dairy and cattle populations, we construct a bottom-up emissions inventory for the SoCAB using the same emission factors as the CARB GHG inventory.

[31] We begin with CH_4 emissions from enteric fermentation. We assign to each of the 137,500 dairy cows in the SoCAB an emission factor of 144 kg CH_4/yr. We assume the remaining 157,500 head are dairy replacements, and assign each an emission factor of 57.7 kg CH_4/yr, or the average emission factor for 0–1 and 1–2 year old dairy replacements in the CARB GHG inventory. We calculate a total of 28.9 Gg CH_4/yr emitted solely from enteric fermentation in the SoCAB.

[32] In addition to enteric fermentation, manure management practices have a substantial effect on CH_4 emissions from livestock operations. In the L.A. basin, dairies typically practice solid storage (http://www.aqmd.gov/rules/doc/r1127/pr1127_task1rpt_20020101.pdf and http://www.arb.ca.gov/planning/sip/sjv_report/addtl_resources.pdf), which emits relatively low levels of CH_4 (17 kg/yr per cow) according to the 2009 CARB GHG inventory. The tradeoff for this practice is that it emits larger amounts of NH_3 than other types of manure management (http://www.epa.gov/ttn/chief/ap42/ch09/draft/draftanimalfeed.pdf). Therefore, if we attribute dry manure management emissions to the SoCAB dairy cow population, and the dry lot emission rate of 2.1 kg CH_4/yr for the remaining heifers, we get an additional 2.7 Gg CH_4/yr from dairy operation manure

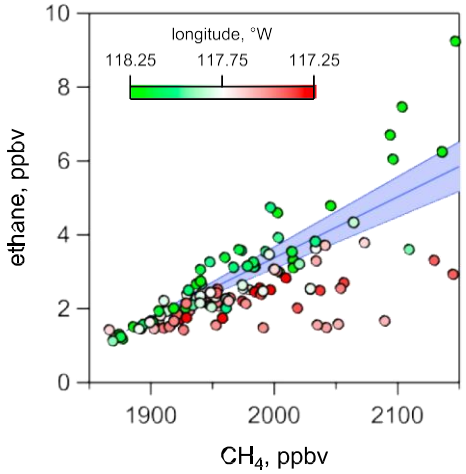


Figure 4. Scatter plot of ethane versus CH_4 from the NOAA P-3 data in the L.A. basin. Data points are colored by longitude to show the different distributions of ethane to CH_4 in the eastern (red) and western (green) parts of the basin. The blue line represents the slope of 1.65 ± 0.25 % used by Wennberg et al. [2012] to represent the estimated ethane/CH_4 ratio of pipeline-quality dry natural gas from the Southern California Gas Company's pipelines.

management in the SoCAB. This results in a total of 31.6 Gg CH_4/yr from enteric fermentation and manure management for the SoCAB dairy operations. This is the emission from agriculture and forestry that we add back into the population-apportioned CARB CH_4 inventory above (Table 2).

[33] Our estimate of 31.6 Gg CH_4/yr, based on inventory data, is less than half of the 76 Gg CH_4/yr estimated by Wennberg et al. [2012]. We attribute this difference in bottom-up inventories to the different assumptions of manure management practices. Wennberg et al. [2012] scaled total California CH_4 emissions by livestock population, which also assumes the manure management practices from the San Joaquin Valley apply to the L.A. basin. For example, the anaerobic lagoons more commonly used in the San Joaquin Valley emit 325 kg CH_4 per cow per year according to the 2009 CARB GHG inventory, significantly higher than 17 kg CH_4 per cow per year from dry manure management practices typical of the L.A. basin.

[34] Nowak et al. [2012] used P-3 data from CalNex to derive emissions of ammonia (NH_3) from dairy farms in the Chino area. From NOAA P-3 measurements, we determine a CH_4 flux from the Chino-area dairies for the same three downwind transects analyzed by Nowak et al. [2012]. Using the Chino to SoCAB population apportionment by Salas et al. [2008], we expect these same Chino-area dairies to emit approximately 28 Gg CH_4/yr. CH_4 fluxes determined from equation (1) range from 24 ± 12 to 88 ± 44 Gg CH_4/yr, and the average of the three transects is 49 ± 25 Gg CH_4/yr. This value derived from airborne flux determination lies between the 28 Gg CH_4/yr calculated from the inventory assuming dry manure management practices described above, and the estimate by Wennberg et al. [2012] of 76 Gg CH_4/yr (less livestock emissions from the SoCAB that are not in the Chino area) assuming mainly wet management practices. We attribute the differences to actual practices in the region, which are likely a mixture of the two manure management approaches. Satellite images of the area show what appear to be several anaerobic lagoons near Chino, California. Our flux determination is therefore consistent with our bottom-up CH_4 emission inventory, with room for a mixture of manure management practices, including some anaerobic lagoons, in the L.A. basin.

4.4. Spatial Distribution of Methane Sources

[35] Townsend-Small et al. [2012] concluded that the CH_4 emissions in the L.A. region had a stable isotope ratio similar to that of fossil-fuel CH_4. This conclusion was based on measurements made at the Mount Wilson Observatory. A back-trajectory [White et al., 2006; http://www.esrl.noaa.gov/psd/programs/2010/calnex/traj/] from MWO for 5 August 2009, the specific day that Townsend-Small et al. [2012] used to determine the excess CH_4 stable isotopic ratio, shows the prevailing winds to MWO were from the southwest, or from downtown L.A. and the coast west of downtown L.A. The trajectory tool also shows winds from the eastern basin on the previous day, which was excluded by Townsend-Small et al. [2012] due to lower correlation between the excess CH_4 and $\delta^{13}C$. We conclude that the MWO data interpreted by Townsend-Small et al. [2012] were dominated by emissions from the western basin only and were not influenced by emissions from either the largest landfills (Puente Hills and Olinda Alpha), or from the dairies in the eastern part of the L.A. basin. This spatially biased sampling is consistent with their conclusion that landfills do not contribute significantly to the total atmospheric CH_4 burden in L.A.

[36] Evidence for the heterogeneous spatial distribution of CH_4 sources in the SoCAB can be seen in the NOAA P-3 data. Figure 4 shows that the correlation of ethane with CH_4 is dependent on the sample location in the L.A. basin. Also, shown in Figure 4 is the slope used by Wennberg et al. [2012] to represent the ethane/CH_4 ratio (16.5 ± 2.5 ppt ethane/ppb CH_4) in pipeline-quality dry natural gas from the Southern California Gas Company (SoCalGas), the major provider of natural gas to the SoCAB, for 2010. The chemical data in Figure 4 reflect the known source types shown on the map in Figure 1c: the large CH_4 sources in the eastern L.A. basin, primarily landfills and dairies, are not significant sources of ethane relative to CH_4.

[37] We can reconcile the conclusions of Townsend-Small et al. [2012] and Wennberg et al. [2012] with the CARB GHG inventory by noting that fossil fuel CH_4 emissions predominate in the western basin and that landfill and livestock CH_4 emissions predominate in the eastern basin. However, in contrast to the findings of Wennberg et al. [2012], we find that natural gas leaks from the SoCalGas and in-home pipelines are not the only possible source of fossil fuel CH_4 to the western basin, as described below.

4.5. Light Alkane Emissions From Local Natural Gas Production

[38] Los Angeles was one of only three out of 28 cities characterized by propane and ethane levels within 10% of one another in the atmosphere [Baker et al., 2008], consistent with an enhanced propane source term in L.A. Figure 5 shows correlations of propane versus ethane in whole-air

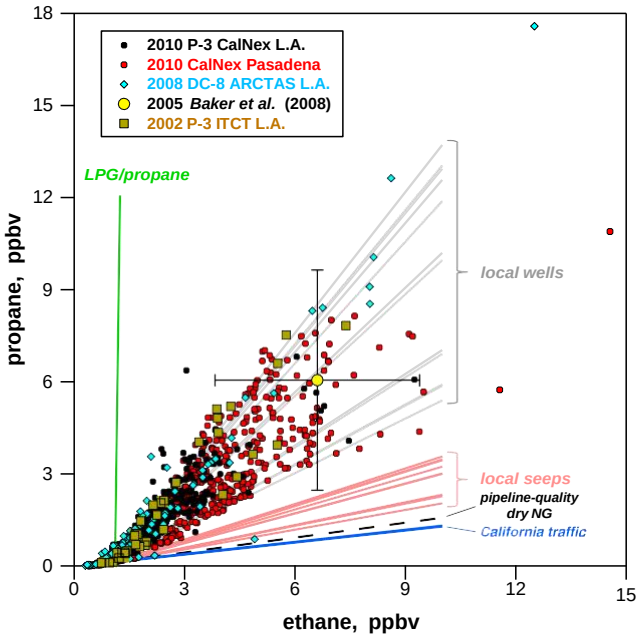


Figure 5. Correlation plot of propane versus ethane from four Los Angeles data sets. Also, plotted are composition ratios of local wells (gray lines) and local seeps (salmon lines) reported by Jeffrey et al. [1991], the composition ratio of pipeline-quality dry natural gas (black dashed line), the propane/ethane emission ratio from a San Francisco Bay-area tunnel study reported by Kirchstetter et al. [1996], and the average composition ratio of liquefied petroleum gas (LPG) or propane (green line).

samples from various aircraft projects in the Los Angeles region (ITCT 2002, ARCTAS 2008, and CalNex 2010), as well as measurements from the CalNex Pasadena ground site in 2010. Also, plotted are lines representing the composition ratios of other possible sources of ethane and propane in Los Angeles.

[39] The L.A. basin is home to oil and gas operations (Figure 1c); the composition ratios depicting possible emissions from local natural gas (gray lines) and local geologic seeps (salmon lines) in Figure 5 are those reported by Jeffrey et al. [1991]. The lower propane content relative to ethane seen in the seeps (e.g., the La Brea tar pits) compared to the local natural gas is attributed to near-surface microorganisms forming shorter chain alkanes from longer chain alkanes during the time the natural gas migrates toward the surface [Jeffrey et al., 1991]. The average propane/ethane ratio for processed gas in SoCalGas pipelines [Wennberg et al., 2012] is plotted as a dashed black line. Pipeline-quality dry natural gas has a low propane/ethane ratio because the natural gas has been processed (i.e., the higher alkanes have been removed from the natural gas) before distribution. The SoCalGas ratio is representative of natural gas piped in from out of state (e.g., from Texas, Wyoming, and Canada); approximately 90% of natural gas used in California is imported (http://www.socalgas.com/regulatory/documents/cgr/2010_CGR.pdf). The on-road emissions are taken from a San Francisco Bay-area tunnel study by Kirchstetter et al. [1996], who reported a vehicular emission ratio of 0.13 mol propane/mol ethane roughly similar to those by Fraser et al. [1998] (0.27 mol propane/mol ethane) and by Lough et al. [2005] (0.06–0.18 mol propane/mol ethane). Vehicle engine exhaust typically contains small, decreasing amounts of CH_4, ethane, and propane due to incomplete combustion, as gasoline and diesel fuel do not contain significant amounts of these light alkanes. The on-road emissions, local geologic seeps, and the pipeline-quality dry natural gas from SoCalGas contain three to five times more ethane than propane and therefore cannot alone explain the ambient ratios measured in the L.A. basin. The propane and ethane composition of unprocessed natural gas from local wells, on the other hand, closely matches the SoCAB ambient measurements from three aircraft campaigns, the CalNex ground site measurements, and the Baker et al. study [2008]. Propane and ethane were also typically enhanced at the same time, with the exception of one sample with elevated propane near the Long Beach area (Figure 1e).

[40] The data in Figure 5 suggest that local oil and gas wells contribute significantly to the atmospheric propane burden in the SoCAB. However, Wennberg et al. [2012] invoked a large source of propane from fugitive losses from the liquefied petroleum gas (LPG) industry (i.e., propane tanks), in addition to leaks from the pipeline-quality dry natural gas distribution system in the L.A. basin. This would be consistent with past works that have found significant fugitive losses of propane in other cities, such as Mexico City [Blake and Rowland, 1995]. We therefore extend our analysis to incorporate ethane, propane, and C_4 (n- and i-butane) and C_5 (n- and i-pentane) isomers to better attribute and quantify the sources of light alkanes and CH_4 to the SoCAB atmosphere. Light alkanes are plotted in Figure 6, with lines depicting the composition of natural gas in SoCalGas pipelines [Wennberg et al., 2012] and of on-road emissions [Kirchstetter et al., 1996]. We neglect chemical processing of these long-lived alkanes ($t \geq 3$ days at OH = 1×10^6 molecules/cm^3) as we find no detectable difference between daytime and nighttime enhancement ratios relative to CO, similar to the findings of Borbon et al. [2013] for n-butane and CO at the CalNex Pasadena ground site. Atmospheric enhancement ratios of propane, n-butane, and i-butane (Figures 6b–6d) relative to ethane are consistent with emissions having the composition of local natural gas [Jeffrey et al., 1991]. On-road emissions do not appear to contribute significantly to the CH_4, ethane, and propane in the L.A. atmosphere, and pipeline-quality dry natural gas and/or local geologic seeps do not appear to contribute significantly to the propane and n-butane relative to ethane in the L.A. atmosphere. Based on these observations, we conclude that the local natural gas industry contributes a significant fraction to the total atmospheric C_2–C_4 alkane abundances, including propane, in the L.A. basin. We infer CH_4 emissions from the local natural gas industry are non-negligible as well, as discussed below.

4.6. Source Attribution

[41] Here we quantify total emissions of C_2–C_5 alkanes in the L.A. basin by multiplying their observed enhancement ratios to CO by the CARB SoCAB emission inventory for CO. Figure 7 shows C_2–C_5 alkanes plotted versus CO with their respective ODR fits. The slopes from these fits are used in equation (2) along with the projected 2010 CARB CO

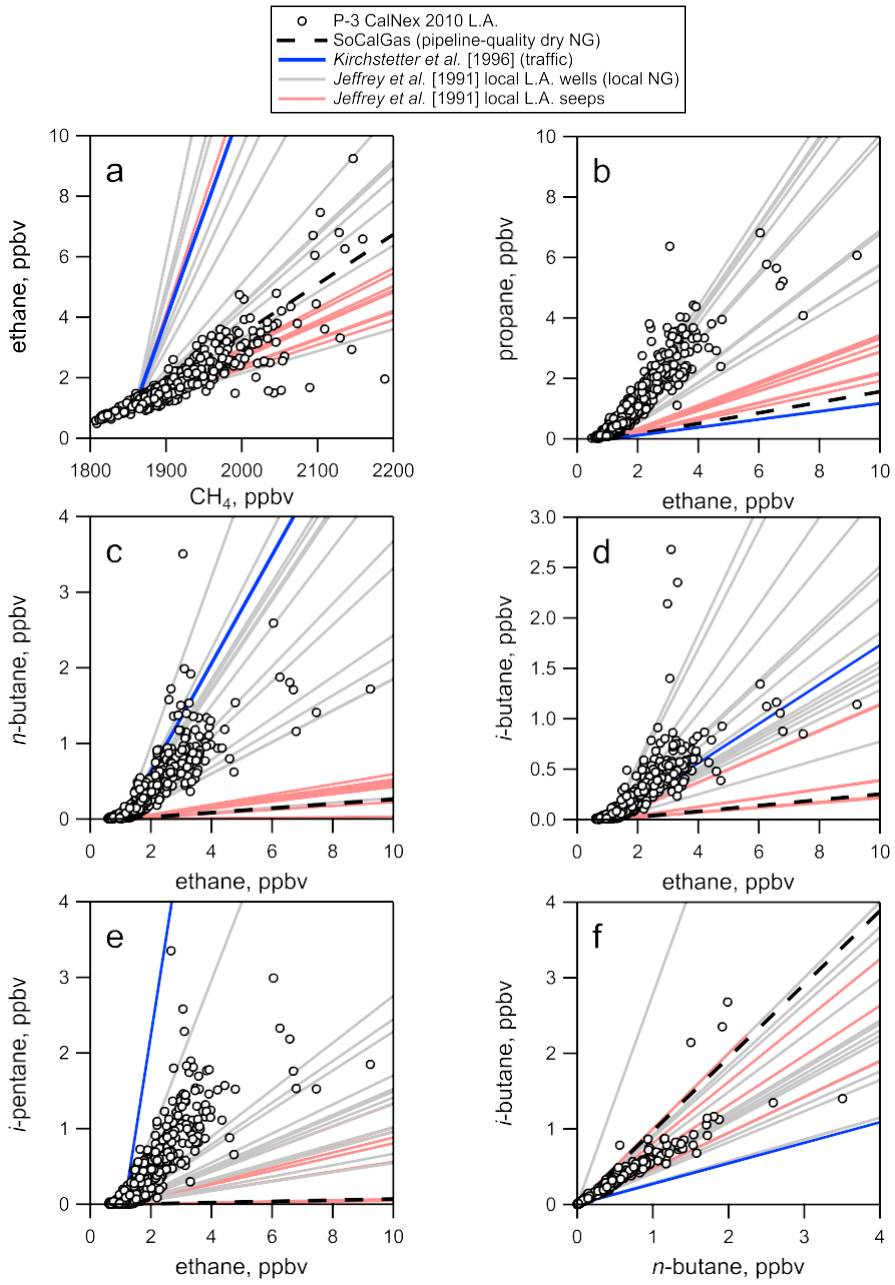


Figure 6. Plots of CH_4 and C_2–C_5 alkanes from the NOAA P-3 CalNex data set, selected for the SoCAB (black circles). Nighttime and high-altitude data are included. Also, included for reference are the emission ratios of mobile sources from Kirchstetter et al. [1996] (blue line), composition ratios measured by Jeffrey et al. [1991] for local natural gas (gray lines) and local geologic seeps (salmon lines), and composition ratios from pipeline-quality dry natural gas (NG) delivered by SoCalGas (dashed black line). These ratios were plotted from daytime background levels.

inventory to calculate annual alkane emissions in the SoCAB. We assume the slopes represent a direct emission with no chemical aging. These emissions are listed in the rightmost column of Table 4. Also, listed in Table 4 are the estimated contributions from mobile sources in the SoCAB, using C_1–C_5 to CO emission ratios from Kirchstetter et al. [1996] (modified as discussed below) and CO emissions from the mobile sources category in the projected 2010 CARB CO inventory, equal to 925 Gg CO/yr, in equation (2).

[42] Wennberg et al. [2012] attributed the inventory CH_4 shortfall [Wunch et al., 2009; Hsu et al., 2010] by ascribing much of the CH_4 and ethane enhancements to fugitive losses of processed pipeline-quality dry natural gas. They further suggest the majority of atmospheric propane is due to LPG industry/propane tank fugitive losses. Here, we consider other possible explanations of the sources of CH_4 and light alkanes in the L.A. basin for the following two reasons. First, the source attribution by Wennberg et al. [2012] leaves little room for CH_4 emissions from landfills, wastewater

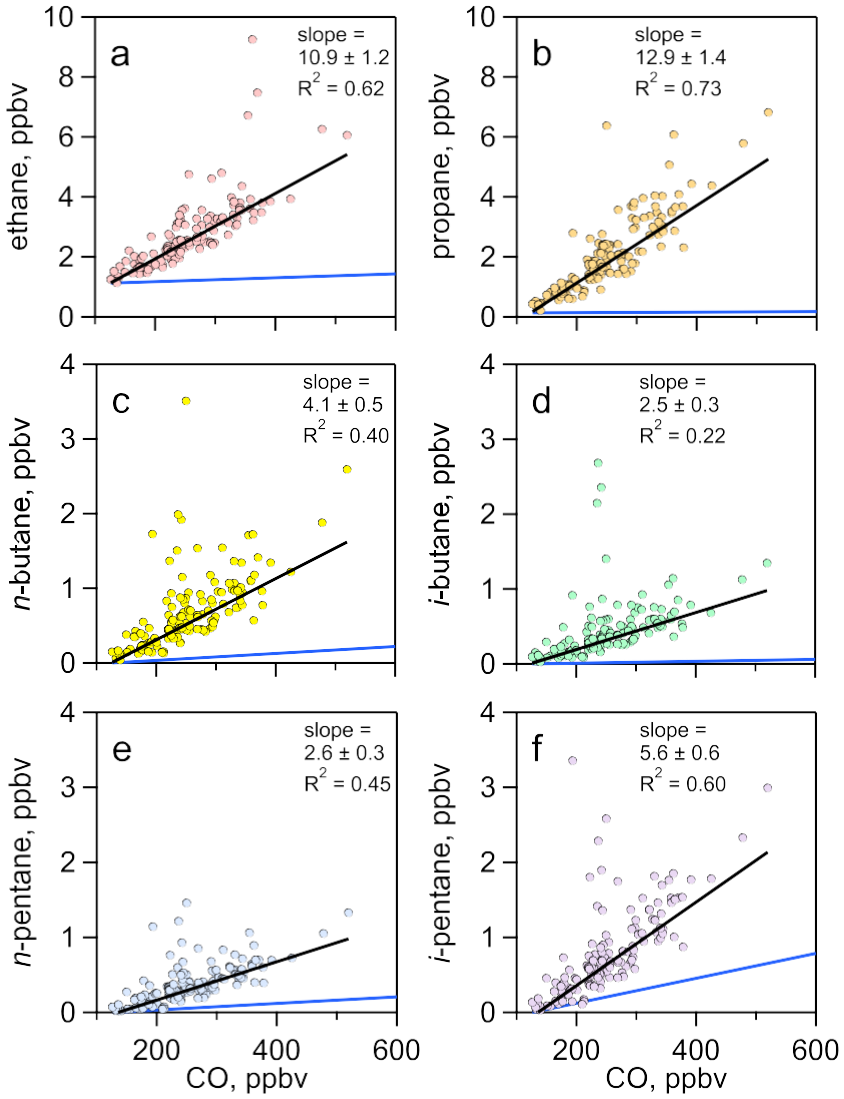


Figure 7. (a–f) Daytime measurements of alkanes versus CO from the NOAA P-3 in the L.A. basin during CalNex are plotted as filled circles. For comparison, the alkane/CO emission ratios from a San Francisco Bay-area tunnel study [Kirchstetter et al., 1996] are plotted as a solid blue line, which extends to the right axis. The slope from a weighted ODR (given as ppt alkane/ppb CO), total slope uncertainty, and R^2 are given in each panel.

treatment plants, and dairies in the L.A. basin. This solution seems unlikely based on direct emissions flux estimates using the P-3 data downwind of landfills and dairies in the SoCAB, as described above. Second, the attribution by Wennberg et al. [2012] would leave a shortfall in both n- and i-butane emissions that cannot be explained by gasoline evaporation or emissions from mobile sources. We use a multivariate approach based on a linear combination of the CH_4 and light alkane compositions from known sources in order to attribute and quantify total CH_4 and C_2–C_5 alkane emissions in the South Coast Air Basin.

[43] We include seven different source types (sectors) with distinct and known CH_4 and C_2–C_5 alkane compositions (Figure 8) in the following analysis: (1) Leaks of processed dry natural gas from pipelines, and/or emissions from local geologic seeps (this approach cannot distinguish between pipeline-quality dry natural gas and local seeps); (2) CH_4-dominated emissions, such as from landfills, wastewater treatment plants, and dairies; (3) Leaks of unprocessed, local natural gas; (4) Leaks of liquefied petroleum gas from propane tanks; (5) On-road combustion emissions from mobile sources; (6) Emissions of CH_4 and C_2–C_5 alkanes in the SoCAB from other source sectors; and (7) Evaporative emissions from gasoline. These are described briefly below.

[44] 1. The South Coast Air Basin contains 14.8 million people, and SoCalGas delivers approximately 11 Tg/yr of natural gas to the Los Angeles area. Additionally, the Earth's natural degassing is a known source of CH_4, ethane, and propane to the atmosphere [Etiope et al., 2008; Etiope and Ciccioli, 2009], and the L.A. basin contains abundant geologic hydrocarbon reserves [Jeffrey et al., 1991]. We group fugitive losses from processed pipeline-quality dry natural gas with the emissions from local geologic seeps because the C_1–C_4 emissions from these sources are not sufficiently different to be treated separately in our linear

Table 4. Derived Emissions in the South Coast Air Basin (In Gg/yr) for 2010 From Each Source Sector Used in Linear Analysis

	Pipeline-Quality Dry NG/Local Seeps	CH_4-Dominant (Landfills, Dairies, Etc.)	Local NG	LPG/Propane	Evaporated Gasoline	Mobile Sources	CARB Other	Summed Source Totals	Estimated SoCAB Total[a]
CH_4	192 ± 54	182 ± 54	32 ± 7	-	-	4.9 ± 1.3	1.2 ± 0.3	411 ± 77	411[b] ± 37
Ethane	5.9 ± 1.7	-	4.5 ± 1.0	0.05 ± 0.02	0.0 ± 0.0	0.6 ± 0.1	0.3 ± 0.1	11.4 ± 1.9	11.4[b] ± 1.6
Propane	1.5 ± 0.4	-	9.9 ± 2.0	6.6 ± 2.9	0.006 ± 0.001	0.1 ± 0.0	1.6 ± 0.4	19.8 ± 3.6	19.8 ± 2.7
n-Butane	0.3 ± 0.1	-	5.9 ± 1.2	0.02 ± 0.01	0.5 ± 0.1	0.3 ± 0.1	1.4 ± 0.4	8.5 ± 1.3	8.3 ± 1.2
i-Butane	0.3 ± 0.1	-	2.2 ± 0.5	0.13 ± 0.06	0.08 ± 0.02	0.04 ± 0.01	1.8 ± 0.5	4.6 ± 0.6	5.1 ± 0.7
n-Pentane	0.07 ± 0.02	-	2.2 ± 0.5	-	2.6 ± 0.4	1.0 ± 0.1	0.3 ± 0.1	6.6 ± 0.6	6.5 ± 0.9
i-Pentane	0.11 ± 0.03	-	2.4 ± 0.5	0.003 ± 0.001	7.6 ± 1.0	3.9 ± 0.5	0.03 ± 0.01	14.1 ± 1.2	14.1 ± 1.8

[a]includes measurement, ODR fit, and inventory uncertainty.
[b]Wennberg et al. [2012] estimate emissions to the SoCAB of 440 ± 100 Gg CH_4/yr and 12.9 ± 0.9 Gg ethane/yr.

combination analysis (illustrated by the similarity in slopes of the dashed black and salmon-colored lines in Figure 6). Both pipeline-quality dry natural gas and local seep emissions contain similar amounts of CH_4 and ethane relative to one another and have less C_3–C_5 alkanes relative to ethane than local, unprocessed natural gas. For pipeline-quality dry natural gas, most C_{3+} alkanes are removed during the processing stage, which is typically done close to the source, which for ~90% of the natural gas used in California is in Canada, Wyoming, and/or Texas. For local seeps, most C_{3+} alkanes are either preferentially adsorbed in shallow sediments compared to CH_4 or biodegraded by microbes in the Earth's crust during the seepage of local natural gas to the surface [Jeffrey et al., 1991]. We use SoCalGas samples of pipeline-quality natural gas from 2010 [Wennberg et al., 2012] to represent this source and estimate the uncertainty of the composition at 15%.

[45] 2. CH_4-dominant emission sources, which for this analysis include landfills, wastewater treatment plants, and livestock, emit CH_4 but no significant amounts of C_2–C_5 alkanes. This is represented in our analysis as a unit vector containing only CH_4.

[46] 3. From 2007 to 2009, the oil and gas industry in the L.A. basin produced roughly 12–13 billion cubic feet of natural gas per year, mostly associated gas from oil wells (http://www.conservation.ca.gov/dog/pubs_stats/annual_reports/Pages/annual_reports.aspx). We use an average of the samples reported by Jeffrey et al. [1991] weighted by 2009 gross natural gas production per field and estimate the uncertainty of this composition at 25%.

[47] 4. Two types of LPG are sold in the Los Angeles area: One is almost completely composed of propane; the other has traces of n- and i-butane (http://www.arb.ca.gov/research/apr/past/98-338_1.pdf). We use the ratios reported by Blake and Rowland [1995] from direct analysis of LPG in Los Angeles, which is consistent with an average of the two types of LPG sold in L.A., and estimate the uncertainty of the composition at 10%.

[48] 5. On-road combustion emissions are modified from the work of Kirchstetter et al. [1996] by multiplying emission ratios of alkanes to CO by the 925 Gg CO/yr from on-road sources in the projected 2010 CARB CO inventory. The C_4–C_5 emissions represent unburned fuel and are typically proportional to the fuel composition; the C_1–C_3 emissions typically represent incomplete combustion products. To account for differing fuel compositions since the time of the Kirchstetter et al. [1996] study, the i- and n-butane emissions calculated for mobile sources in the SoCAB (Table 4) have been scaled to the i-pentane emissions based on their relative abundance in gasoline [Gentner et al., 2012].

[49] 6. There are additional sources of light alkanes in the SoCAB. We use the 2010 CARB speciated inventory for total organic gases (http://arb.ca.gov/ei/speciate/interopt10.htm) and projected 2010 total organic gas emissions (http://www.arb.ca.gov/app/emsinv/fcemssumcat2009.php) for the SoCAB to estimate emissions of light alkanes not specified in other source sectors. These include emissions from aerosol spray cans and other consumer products, coatings and solvents, adhesives and sealants, and fiberglass and plastics manufacturing. For example, propane, n-, and i-butane are commonly used as propellants in aerosol spray cans, having replaced CFCs in the United States in the 1970s (e.g., CARB estimates 0.6 Gg of aerosol antiperspirant vapors were emitted to the SoCAB in 2010, of which 0.14 Gg, 0.03 Gg, and 0.15 Gg were propane, n-, and i-butane, respectively). These emissions are summed and listed in the "CARB Other" column in Table 4. Emissions from natural gas leaks, petroleum refining, petroleum marketing (gas stations), landfills and composting, and mobile sources are not included in these totals, because they are accounted for elsewhere in other source sectors. We estimate a 25% uncertainty in the "CARB Other" inventory.

[50] 7. Emissions ratios from evaporated gasoline were calculated from 10 gasoline samples from five Pasadena gas stations in the summer of 2010, weighted by estimated sales of 80% regular and 20% premium [Gentner et al., 2012]. Uncertainties are those reported by Gentner et al. [2012].

[51] First, we start with estimated annual C_1–C_5 emissions in the SoCAB (rightmost column of Table 4), then subtract modified on-road emissions [Kirchstetter et al., 1996] and projected emissions of C_1–C_5 alkanes from other sources (source sector 6, above). Next, we place the remaining source sector characteristics into a matrix and solve for the fraction each source contributes to the remaining alkane observations for the L.A. basin based on each source's relative abundances of various light alkanes. The matrix has five columns representing the five remaining source sectors, and seven rows containing C_1–C_5 alkanes. We solve the following equation [e.g., see section 4.2 of Kim et al., 2011]

$$A_{i;j}x_j \text{ ¼ } b_i \quad (3)$$

where $A_{i,j}$ is a matrix of the C_1–C_5 alkane composition, i, for the source sectors, j, defined above; x_j is the fraction each source contributes to the total observed emissions; and b_i

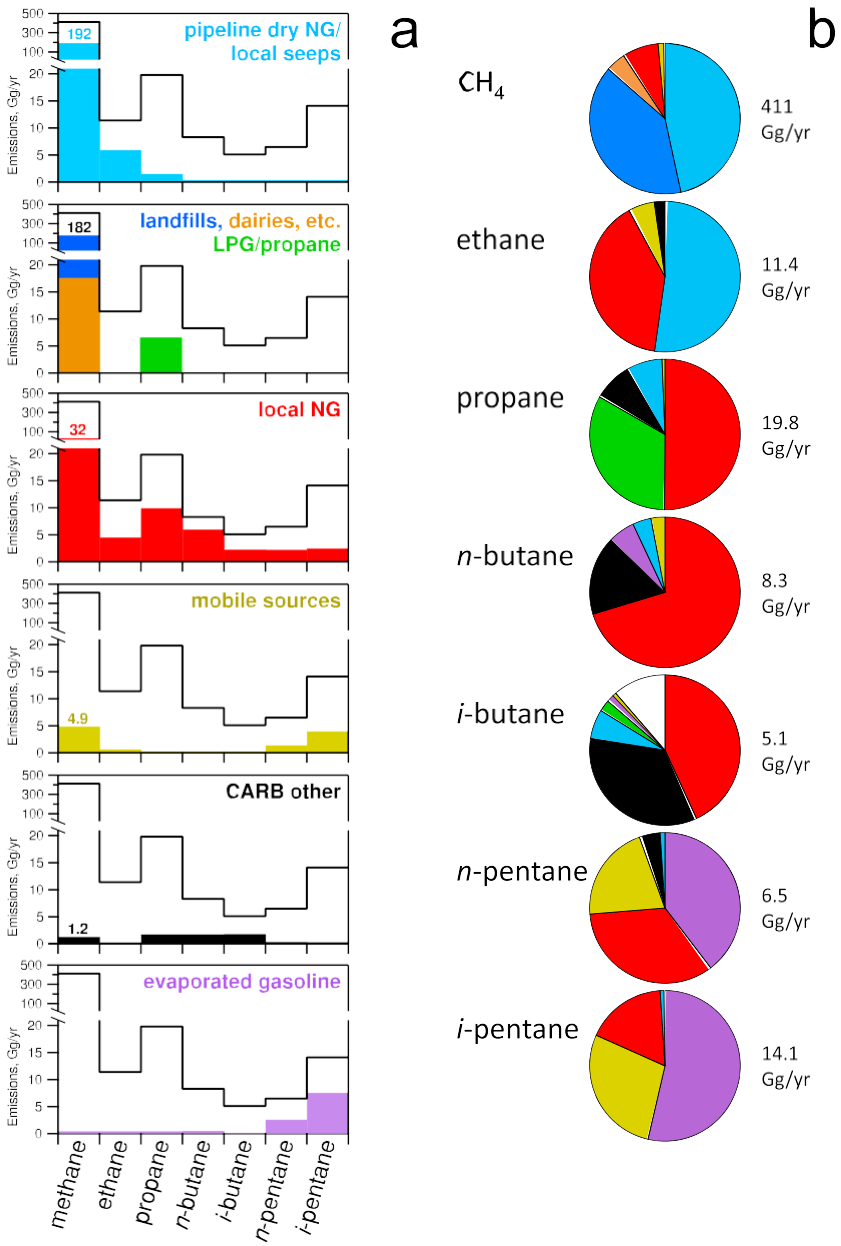


Figure 8. (a) Results from a linear least squares solution to a combination of six sources and seven trace gas species in the SoCAB. The thick black line represents the estimated total annual emission to the SoCAB for seven hydrocarbons (CH_4 and C_2–C_5). The colored bars represent the fraction of the total contributed by each of the six source sectors used in the linear analysis. CH_4 emissions are written above the bar. (b) Pie charts for the same data in Figure 8a showing the relative contributions from each source for each of seven alkanes, colored as in Figure 8a. The white region in the i-butane pie chart represents the 11% shortfall between our source attribution and our estimated emission to the SoCAB, though it is within the uncertainties of these two values. The total emission of the alkane to the SoCAB is given to the right of each pie chart.

is the total observed emission of alkane i minus the contributions from the mobile and "other" source sectors (Table 4). The columns of the matrix A are proportional to the first five columns of Table 4. We solve for the linear least squares solution that minimizes $(Ax - b)$. Uncertainties in the derived x_j are estimated by a sensitivity study, where we run the solution 1,000,000 times by randomly varying $A_{i,j}$ and b_i according to their estimated uncertainties, then use the standard deviation of the 1,000,000 x_j determinations to estimate the uncertainty in the source attribution fraction. The source attribution fractions and their uncertainties are multiplied by the total estimated SoCAB emission for each alkane and then are summed with the uncertainties added in quadrature. CH_4 and C_2–C_5 alkane emissions totals, their uncertainties, and the contributions from each source type are given in Table 4. The source attribution solution solves

the observed SoCAB alkane emission to within each alkane's emission uncertainty.

[52] Our modeled source attribution differs from the alkane source distribution in the L.A. basin as set forth by Wennberg et al. [2012]. From a total calculated source of 410 ± 40 Gg CH_4/yr in the SoCAB, we determine that 47% comes from leaks of processed pipeline-quality dry natural gas and/or from local geologic seeps; 44% of the CH_4 comes from the sum of landfill, wastewater treatment, and dairy emissions; 8% from the leaks of unprocessed natural gas from production in the western L.A. basin; and 1% from mobile sources. The attribution is presented graphically in Figure 8. Figure 8a displays the total SoCAB emissions as a black horizontal line in each panel, with contributions from the different source sectors given below the line by the filled bars. Figure 8b shows the proportion that each source sector contributes to the derived total emissions of each alkane.

[53] Our analysis attributes CH_4 emissions of 192 ± 54 Gg CH_4/yr to leaks of pipeline-quality dry processed natural gas and/or leaks from local geologic seeps but does not distinguish further between these two different sources. This value is nearly a factor of 5 greater than the population-apportioned 2009 CARB GHG emissions inventory estimate of 40 Gg CH_4/yr lost from natural gas pipelines in the SoCAB. Our estimate of 192 Gg CH_4/yr is less than the maximum emission of 400 ± 150 Gg CH_4/yr estimated by Wennberg et al. [2012]. Our estimate would represent approximately 2% of the natural gas delivered to customers in the SoCAB and, including storage and deliveries to customers outside the SoCAB, 1% of the gas flowing into the basin [Wennberg et al., 2012]. These percentages would decrease linearly with any CH_4 emissions attributed to local geologic seeps. Farrell et al. [2013] estimate up to 55 Gg CH_4/yr are emitted from the La Brea Tar Pits in western L.A. County alone; if accurate, this would imply pipeline leaks of only 0.7% of the gas flowing into the basin, or a factor of at least two lower than the 2% proposed by Wennberg et al. [2012].

[54] Our analysis attributes 182 ± 54 Gg CH_4/yr in the SoCAB to emissions from landfills, wastewater treatment, and dairies. SoCAB landfills account for 164 Gg CH_4/yr in the 2008 CARB GHG inventory; a value supported by our analysis in section 4.2. In section 4.3, we estimated in a bottom-up inventory that SoCAB dairies emitted 31.6 Gg CH_4/yr. Wennberg et al. [2012] estimated an emission of 20 Gg CH_4/yr from wastewater treatment. These independent estimates sum to 216 Gg CH_4/yr and are consistent with our source apportionment using NOAA P-3 data.

[55] CH_4 emissions of 31.9 ± 6.5 Gg CH_4/yr are ascribed to leaks of local, unprocessed natural gas and would represent 17% of the local production in 2009, the latest year for which data are available (http://www.conservation.ca.gov/dog/pubs_stats/annual_reports/Pages/annual_reports.aspx). This number assumes a CH_4 composition of 72.5% by volume for natural gas produced in the South Coast Air Basin, which is calculated as an average from the samples reported by Jeffrey et al. [1991] weighted by 2009 production. Our derived value of 17%, although a surprisingly high amount of local production, is consistent with a nascent bottom-up estimate under way at CARB. A new bottom-up inventory survey, conducted by CARB for the calendar year 2007 but not yet incorporated into the official GHG inventory, indicates that 109 Gg CH_4/yr, since revised to 95.5 Gg CH_4/yr (S. Detwiler, personal communication, October 2012), were emitted throughout California by the oil and gas industry via combustion, venting, and fugitive losses (Table 3-1, http://www.arb.ca.gov/cc/oil-gas/finalreport.pdf). This updated value is a factor of 2.5 larger than the current CARB GHG inventory tabulation of 38 Gg CH_4/yr from oil and gas extraction for 2007 in California. CH_4-specific emissions for the South Coast Air Quality Management District in the new CARB survey report show 24.6 Gg CH_4/yr were emitted in the SoCAB (S. Detwiler, personal communication, October 2012). According to the survey, emissions in the SoCAB accounted for 26% of the revised statewide total oil and gas operations CH_4 emission in 2007, despite accounting for only 4.4% of statewide natural gas production in the basin that year (http://www.conservation.ca.gov/dog/pubs_stats/annual_reports/Pages/annual_reports.aspx). Thus, the survey responses suggest a CH_4 leak rate of 12% of local production in the L.A. basin. Thus, our estimate of CH_4 emissions from local natural gas for 2010 based on P-3 data from CalNex is within a factor of 1.5 of the CARB bottom-up inventory currently in development based on the 2007 survey. According to the survey, other oil and gas-producing regions in California show smaller CH_4 loss rates than that from the SoCAB. For instance, statewide losses of CH_4 represent approximately 2.1% of statewide production, and CH_4 losses from the San Joaquin Air Quality District represent approximately 1.4% of production (from Oil and Gas Districts four and five). This indicates that losses from natural gas production are proportionally larger in the L.A. basin than elsewhere in the State of California.

[56] A propane emission of 6.6 ± 2.9 Gg/yr from LPG/propane tanks would represent approximately 1% of sales (http://www.aqmd.gov/ceqa/documents/2012/aqmd/finalEA/PAR1177/1177_FEA.pdf), which is less than the ~4% calculated by Wennberg et al. [2012], and closer to the 0.6% estimated from the document cited.

[57] Finally, our analysis suggests a resolution to the discrepancies noted above between previous top-down assessments and the bottom-up inventory calculations for CH_4 in the SoCAB [e.g., Wunch et al., 2009; Hsu et al., 2010; Townsend-Small et al., 2012; Wennberg et al., 2012]. We conclude the most probable source for the excess atmospheric CH_4 is likely due to a combination of primarily leaks, not accurately represented in the current CARB GHG inventory, from natural gas pipelines and urban distribution systems and/or from local geologic seeps, and secondarily leaks of unprocessed natural gas from local oil and gas production centered in the western L.A. basin. This finding is based on the characteristic enhancement ratios of CH_4 and the various C_2–C_5 alkanes consistently observed in the L.A. atmosphere, and is further supported by the spatial information provided by P-3 samples during CalNex. Finally, the updated values for local oil and gas industry emissions in the recent GHG survey commissioned by CARB, when incorporated fully into the official CARB GHG record, will likely help to reduce this long-standing discrepancy between top-down assessments and bottom-up inventories.

5. Conclusions

[58] We use aircraft measurements of CH_4, CO, and CO_2 during the CalNex field campaign to show that emissions of CH_4 to the L.A. basin are greater than can be explained by official state bottom-up inventories apportioned by population, consistent with published work. The ratio of the CARB CO and CO_2 inventories is in better agreement with our measurements of CO/CO_2 in the Los Angeles atmosphere than was the case for the analysis by Wunch et al. [2009], which we attribute either to improved CARB inventories, the present use of a basin-wide data set to determine basin-wide emission ratios, or both.

[59] From crosswind plume transects downwind of the two largest landfills in the basin, we determine CH_4 fluxes that are consistent with the 2008 CARB GHG inventory values, which total 164 Gg CH_4/yr emitted from all landfills in the South Coast Air Basin. CH_4 emission fluxes were also determined for Chino-area dairies in the eastern L.A. basin. Flux estimates from these dairies ranged from 24 ± 12 to 87 ± 44 Gg CH_4/yr, and the average flux is consistent with a revised bottom-up inventory originally compiled by Salas et al. [2008] and with previous inventory estimates [Wennberg et al., 2012].

[60] Finally, we present a top-down assessment of C_2–C_5 alkane sources in the L.A. basin, and then apportion CH_4 and the C_2–C_5 alkanes to specific source sectors in the region. Using this source apportionment approach, we estimate that 32 ± 7 Gg of CH_4/yr, or 8% of the total CH_4 enhancement observed in the SoCAB during CalNex, came from the local oil and gas industry. This number represents approximately 17% of the natural gas produced in the region, within a factor of 1.5 of that calculated from a recent survey that will be used to update the CARB bottom-up inventory. We estimate 182 ± 54 Gg CH_4/yr are emitted by landfills, dairies, and wastewater treatment, which is consistent with bottom-up inventories, and 192 ± 54 Gg CH_4/yr are emitted of processed pipeline-quality dry natural gas and/or from geologic seeps in the region. We further conclude that leaks of processed pipeline-quality dry natural gas and/or local geologic seeps, and unprocessed natural gas from local oil and gas production are the most likely major contributors to the previously noted discrepancy between CH_4 observations and State of California inventory values for the South Coast Air Basin. Our findings suggest that basin-wide mobile studies targeting CH_4 and C_2–C_5 alkane emissions from natural gas pipelines and urban distribution systems, geologic seeps, and local oil and gas industry production sites would be useful to further distinguish the sources of CH_4 in the L.A. basin.

[61] Acknowledgments. This work was supported in part by the NOAA Health of the Atmosphere Program and by the NOAA Atmospheric Chemistry, Carbon Cycle, and Climate Program. We thank Larry Hunsaker of CARB for landfill CH_4 emission data and Stephanie Detwiler of CARB for the updated bottom-up oil and gas inventory data.

References

Baker, A. K., A. J. Beyersdorf, L. A. Doezema, A. Katzenstein, S. Meinardi, I. J. Simpson, D. R. Blake, and F. S. Rowland (2008), Measurements of nonmethane hydrocarbons in 28 United States cities, Atmos. Environ., 42, 170–182, doi:10.1016/j.atmosenv.2007.09.007.

Blake, D. R., and F. S. Rowland (1995), Urban leakage of liquefied petroleum gas and its impact on Mexico City air quality, Science, 269, 953–956.

Boggs, P. T., et al. (1989), Algorithm 676 – ODRPACK: Software for weighted orthogonal distance regression, ACM Trans. Math. Software, 15, 348–364.

Borbon, A., et al. (2013), Emission ratios of anthropogenic VOC in northern mid-latitude megacities: Observations vs. emission inventories in Los Angeles and Paris, J. Geophys. Res. Atmos., 118, 2041–2057, doi:10.1002/jgrd.50059.

Brioude, J., et al. (2012), Top-down estimate of surface flux in the Los Angeles Basin using a mesoscale inverse modeling technique: Assessing anthropogenic emissions of CO, NO_x, and CO_2 and their impacts, Atmos. Chem. Phys. Discuss., 12, 31439–31481, doi:10.5194/acpd-12-31439-2012.

Colman, J. J., A. L. Swanson, S. Meinardi, B. C. Sive, D. R. Blake, and F. S. Rowland (2001), Description of the analysis of a wide range of volatile organic compounds in whole air samples collected during PEM-tropics A and B, Anal. Chem., 73(15), 3723–3731, doi:10.1021/ac010027g.

Conway, T. J., et al. (2011), Atmospheric carbon dioxide dry air mole fractions from the NOAA ESRL carbon cycle cooperative global air sampling network, 1968–2010, Version: 2011-10-14, Path: ftp://ftp.cmdl.noaa.gov/ccg/co2/flask/event/.

Dlugokencky, E. J., et al. (2011), Atmospheric methane dry air mole fractions from the NOAA ESRL carbon cycle cooperative global air sampling network, 1983–2010, Version: 2011-10-14, Path: ftp://ftp.cmdl.noaa.gov/ccg/ch4/flask/event/.

Etiope, G., and P. Ciccioli (2009), Earth's degassing: A missing ethane and propane source, Science, 323, doi:10.1126/science.1165904.

Etiope, G., K. R. Lassey, R. W. Klusman, and E. Boschi (2008), Reappraisal of the fossil methane budget and related emission from geologic sources, Geophys. Res. Lett., 35, L09307, doi:10.1029/2008GL033623.

Farrell, P., D. Culling, and I. Leifer (2013), Transcontinental methane measurements: Part 1. A mobile surface platform for source investigations, Atmos. Environ., doi:10.1016/j.atmosenv.2013.02.014.

Fraser, M. P., G. R. Cass, and B. R. T. Simoneit (1998), Gas-phase and particle-phase organic compounds emitted from motor vehicle traffic in a Los Angeles roadway tunnel, Environ. Sci. Technol., 32, 2051–2060.

Gentner, D. R., et al. (2012), Elucidating secondary organic aerosol from diesel and gasoline vehicles through detailed characterization of organic carbon emissions, Proc. Natl. Acad. Sci. U. S. A., 109(45), 18318–18323, doi:10.1073/pnas.1212272109.

Gilman, J. B., et al. (2010), Surface ozone variability and halogen oxidation throughout the Arctic and sub-Arctic springtime, Atmos. Chem. Phys., 10, 10,223–10,236, doi:10.5194/acp-10-10223-2010.

Gurney, K. R., D. L. Mendoza, Y. Zhou, M. L. Fischer, C. C. Miller, S. Geethakumar, and S. de la Rue du Can (2009), High resolution fossil fuel combustion CO_2 emissions fluxes for the United States, Environ. Sci. Technol., 43, 5535–5541, doi:10.1021/es900806c.

Holloway, J. S., R. O. Jakoubek, D. D. Parrish, C. Gerbig, A. Volz-Thomas, S. Schmitgen, A. Fried, B. Wert, B. Henry, and J. R. Drummond (2000), Airborne intercomparison of vacuum ultraviolet fluorescence and tunable diode laser absorption measurements of tropospheric carbon monoxide, J. Geophys. Res., 105(D19), 24,251–24,261, doi:10.1029/2000JD900237.

Hsu, Y.-K., T. VanCuren, S. Park, C. Jakober, J. Herner, M. FitzGibbon, D. R. Blake, and D. D. Parrish (2010), Methane emissions inventory verification in southern California, Atmos. Environ., 44, 1–7, doi:10.1016/j.atmosenv.2009.10.002.

Jeffrey, A. W. A., et al. (1991), Geochemistry of Los Angeles Basin Oil and Gas Systems, in Active Margin Basins, Memoir 52, edited by K. T. Biddle, pp. 197–219, Amer. Assoc. Petr. Geologists, Tulsa, Okla.

Kim, S.-W., et al. (2011), Evaluations of NO_X and highly reactive VOC emission inventories in Texas and their implications for ozone plume simulations during the Texas Air Quality Study 2006, Atmos. Chem. Phys., 11, 11361–11386, doi:10.5194/acp-11-11361-2011.

Kirchstetter, T. W., B. C. Singer, R. A. Harley, G. R. Kendall, and W. Chan (1996), Impact of oxygenated gasoline use on California light-duty vehicle emissions, Environ. Sci. Technol., 30, 661–670.

Kort, E. A., P. K. Patra, K. Ishijima, B. C. Daube, R. Jiménez, J. Elkins, D. Hurst, F. L. Moore, C. Sweeney, and S. C. Wofsy (2011), Tropospheric distribution and variability of N_2O: Evidence for strong tropical emissions, Geophys. Res. Lett., 38, L15806, doi:10.1029/2011GL047612.

Lough, G. C., J. J. Schauer, W. A. Lonneman, and M. K. Allen, (2005), Summer and winter nonmethane hydrocarbon emissions from on-road motor vehicles in the midwestern United States, J. Air Waste Manage. Assoc., 55, 629–646.

Neuman, J. A., et al. (2012), Observations of ozone transport from the free troposphere to the Los Angeles basin, J. Geophys. Res., 117, D00V09, doi:10.1029/2011JD016919.

Novelli, P. C., and K. A. Masarie (2010), Atmospheric carbon monoxide dry air mole fractions from the NOAA ESRL carbon cycle cooperative

global air sampling network, 1988–2009, Version: 2011-10-14, Path: ftp://ftp.cmdl.noaa.gov/ccg/co/flask/event/.

Nowak, J. B., J. A. Neuman, R. Bahreini, A. M. Middlebrook, J. S. Holloway, S. A. McKeen, D. D. Parrish, T. B. Ryerson, and M. Trainer (2012), Ammonia sources in the California South Coast Air Basin and their impact on ammonium nitrate formation, Geophys. Res. Lett., 39, L07804, doi:10.1029/2012GL051197.

Peischl, J., et al. (2012), Airborne observations of methane emissions from rice cultivation in the Sacramento Valley of California, J. Geophys. Res., 117, D00V25, doi:10.1029/2012JD017994.

Ryerson, T. B., et al. (1998), Emissions lifetimes and ozone formation in power plant plumes, J. Geophys. Res., 103(D17), 22,569–22,583.

Ryerson, T. B., et al. (2013), The 2010 California Research at the Nexus of Air Quality and Climate Change (CalNex) field study, J. Geophys. Res. Atmos., doi:10.1002/jgrd.50331.

Salas, W. A., et al. (2008), Developing and applying process-based models for estimating greenhouse gas and air emission from California dairies, California Energy Commission, PIER Energy-Related Environmental Research, CEC-500-2008-093, http://www.energy.ca.gov/2008publications/CEC-500-2008-093/CEC-500-2008-093.PDF.

Schauffler, S. M., E. L. Atlas, D. R. Blake, F. Flocke, R. A. Lueb, J. M. Lee-Taylor, V. Stroud, and W. Travnicek (1999), Distributions of brominated organic compounds in the troposphere and lower stratosphere, J. Geophys. Res., 104(D17), 21,513–21,535, doi:10.1029/1999JD900197.

Simpson, I. J., et al. (2010), Characterization of trace gases measured over Alberta oil sands mining operations: 75 speciated C_2–C_{10} volatile organic compounds (VOCs), CO_2, CO, CH_4, NO, NO_y, O_3 and SO_2, Atmos. Chem. Phys., 10, 11,931–11,954, doi:10.5194/acp-10-11931-2010.

Taylor, J. R. (1997), An Introduction to Error Analysis, The Study of Uncertainties in Physical Measurements, 2nd Edition, p. 174, University Science Books, Sausalito, Calif.

Townsend-Small, A., S. C. Tyler, D. E. Pataki, X. Xu, and L. E. Christensen (2012), Isotopic measurements of atmospheric methane in Los Angeles, California, USA: Influence of "fugitive" fossil fuel emissions, J. Geophys. Res., 117, D07308, doi:10.1029/2011JD016826.

Trainer, M., B. A. Ridley, M. P. Buhr, G. Kok, J. Walega, G. Hübler, D. D. Parrish, and F. C. Fehsenfeld (1995), Regional ozone and urban plumes in the southeastern United States: Birmingham, a case study, J. Geophys. Res., 100(D9), 18,823–18,834.

Washenfelder, R. A., et al. (2011), The glyoxal budget and its contribution to organic aerosol for Los Angeles, California, during CalNex 2010, J. Geophys. Res., 116, D00V02, doi:10.1029/2011JD016314.

Wennberg, P. O., et al. (2012), On the sources of methane to the Los Angeles atmosphere, Environ. Sci. Technol., 46(17), 9282–9289, doi:10.1021/es301138y.

White, W. H., J. A. Anderson, D. L. Blumenthal, R. B. Husar, N. V. Gillani, J. D. Husar, and W. E. Wilson Jr. (1976), Formation and transport of secondary air pollutants: Ozone and aerosols in the St. Louis urban plume, Science, 194, 187–189, doi:10.1126/science.959846.

White, A. B., C. J. Senff, A. N. Keane, L. S. Darby, I. V. Djalalova, D. C. Ruffieux, D. E. White, B. J. Williams, and A. H. Goldstein (2006), A wind profiler trajectory tool for air quality transport applications, J. Geophys. Res., 111, D23S23, doi:10.1029/2006JD007475.

Wunch, D., P. O. Wennberg, G. C. Toon, G. Keppel-Aleks, and Y. G. Yavin (2009), Emissions of greenhouse gases from a North American megacity, Geophys. Res. Lett., 36, L15810, doi:10.1029/2009GL039825.

Appendix F

Gas Safety Incorporated
16 Brook Lane
Southborough, Massachusetts 01772
774-922-4626 www.gassafetyusa.com

Report to

Damascus Citizens for Sustainability
25 Main Street
Narrowsburg, New York 12764

Report on a Survey of Ground-Level Ambient Methane Levels in the Vicinity of Wyalusing, Bradford County, Pennsylvania

November 2013

by

Bryce F. Payne Jr.[1] and Robert Ackley[2]

[This report is subject to revision.]

NOTE: Figures follow text.

There have been numerous reports of methane emissions related to shale gas development in the vicinity of Wyalusing, Bradford County, Pennsylvania. In the interest of furthering the understanding of those fugitive methane events Damascus Citizens for Sustainability engaged Gas Safety, Inc. to survey ambient air methane levels in the vicinity of Wyalusing, PA. The survey covered parts of 9 townships on both sides of the Susquehanna River (Figure 1 –

---

[1] Consulting and research in environmental science since 1992. Associate Research Professor, Dept. Environmental Engineering and Earth Sciences, Wilkes University, Wilkes-Barre, PA and Senior Fellow of the Wake Forest University Center for Energy, Environment, and Sustainability, Winston-Salem, NC. bryce.payne@wilkes.edu

[2] President of Gas Safety, Inc. with 30 years experience in gas leak detection and measurement, related regulatory compliance, and training. bobackley@gassafetyusa.com

following text) from Towanda on the northwest to Wyalusing on the central eastern side. Survey coverage was restricted to readily identifiable public roadways. Consequently, the survey was most intense from the Susquehanna River west to Pennsylvania Route 187.

Though the survey results do not prove a relationship between ambient air methane contamination and groundwater contamination, it is clearly suggestive. Further, it also suggests shale gas well operations in that area still did not have control of the gas that has been developed there. In fact, as will be discussed, survey data indicates there may be gas control problems in about 10% of the survey area resulting in elevated methane levels in most of the area.

In addition, detection of any level of methane above normal background for an area indicates only two possible conditions: diffuse, non-point emissions are occurring over some portion of the area, or, one or more point sources are active within the area.

Conditions during the Survey

The survey effort involved two separate survey field work efforts, one on 31 January and the other 3-4 June 2013. Weather conditions at the time of the January survey were not ideal. Winds were from the west at speeds consistently near 20 miles per hour (29 feet per second). Under these conditions methane emissions from any source disperse rapidly. Consequently, elevated methane levels due to such emissions are more difficult to detect than under more favorable wind conditions. Functionally this means that, during a road survey, detection of elevated methane levels requires the sources be larger or more intense and in closer proximity to the survey vehicle path than under more favorable wind conditions. However, such wind conditions do cause methane emissions to be swept along the ground surface farther and faster. Consequently, methane emissions appear as a general elevation of methane levels over a wider area, instead of localized markedly elevated peaks.

During the 3-4 June field work weather conditions were more favorable. The wind was from the north-northwest at an average speed of 5 miles per hour (around 8 feet per second). Under these conditions methane emissions would be expected to be detectable as low concentration plumes extending for an appreciable distance to the south-southeast of the source. Mixing layer structure and height was not estimated during the survey, but conditions should have favored typical lower atmospheric mixing patterns in which most methane emissions diffuse rapidly upward.

Results of the January Survey

As anticipated due to the wind conditions the methane levels were moderately elevated widely over the survey area. Typical methane level observed during the survey was low. The average methane level was 1.86 ppm, with a minimum of 1.79 ppm, 90% were below 1.91 ppm, and 99% below 2.08 ppm.[3] Under such high wind conditions, the layer of the atmosphere that normally forms next to the land surface[4] is swept away by air that would normally move at altitudes of a few hundred to a few thousand feet above. Under gentler wind conditions gases released into the air tend to accumulate in plumes as they dissipate into the turbulent but lower-wind-speed layer of air next to the land surface. Under sustained high wind conditions the air from the higher layer sweeps down and across the land surface rapidly sweeping any released gases across the land surface and up into the atmosphere.

Figure 2 shows an oblique westward view of the survey area in which the data was processed to remove values lower than 2.2 ppm and vertically exaggerate those over 2.2 ppm by a factor of 1000. In effect, this approach visually defines methane levels above 2.2 ppm as elevated methane levels (EMLs). This graphical rendering shows around 18 locations with elevations above 2.2 ppm. There also appear to be many locations with EMLs near 2.2 ppm. This, however, is an artifact of the low resolution of this image and the high resolution of the survey data set. When this image is examined at higher resolution most of the apparent near-2.2-ppm EMLs disappear.

To allow examination of smaller EMLs another image of data was prepared with the methane data processed to remove values below 1.9 ppm and vertically exaggerate values >1.9 ppm by a factor of 100. The lower 1.9-ppm cutoff and vertical exaggeration preserved EMLs that were not apparent upon high resolution examination of Figure 2, as illustrated by Figures 3 and 4. The >1.9-ppm image is not shown as it is visually nearly flat at the resolution that can be rendered on a single page of this report. In the >1.9-ppm image 57 EMLs were indentified as sufficiently clear to merit further examination (see Appendix B for a listing of those EMLs by location). Of those 57 EMLs, 43 were in proximity to and nearly-downwind of gas pipelines, gas well pads, farms, industrial facilities with apparent waste water treatment ponds or lagoons.

---

[3] During survey runs the vehicle has to make stops. The CRDS methane instrument collects data continuously. Consequently, geographically disproportionate amounts of data accumulate whenever the vehicle stops. Geographically disproportionate data accumulations are removed from the data set before statistical analysis. Images are generated using the full raw data sets.

[4] Planetary boundary layer or mixing layer. See Manhattan extended report for more detailed discussion.NEED LINK HERE

Further identification of the methane sources causing the other 14 EMLs was beyond the scope of the survey work.

Despite the strong wind conditions a relatively large methane plume was detected. The plume was detected over an area running from Wysox 2.5 miles southward along the river and up to 3.6 miles to the east. The plume was not present on a later pass through the same area. The extent and consistency of this plume over such a large area under such windy conditions, and its relatively sudden disappearance suggest a sizeable release of methane upwind of the plume area that ended sometime during the survey. Identification of a likely source was beyond the scope of the survey work. It is noteworthy that this plume was again present during the June survey. The plume may have been related to a number of gas wells generally north of Wysox.

Conclusions from 31 January Survey

The strong wind conditions during the methane survey caused rapid mixing and lateral dispersal of methane from any sources in or near the survey area. Under such conditions detection of elevated methane levels is limited to those resulting from larger emissions or those from sources in close proximity to the roadway. The rapid mixing and lateral dispersal causes methane levels in the area to appear more uniformly elevated than would be the case under less windy conditions. This was indicated by the slightly elevated mean (1.86 ppm) and narrow range of methane levels (1.79-1.91 ppm) that accounted for the 90% of the data (further discussed in comparison to the June data follows below). All the other 10% of the data indicating methane levels above 1.91 ppm occurred at less than 60 locations. Among those locations, 43 were in the vicinity of candidate potential methane sources, in most cases gas pipelines or gas well pads. At 14 locations with elevated methane levels candidate potential methane sources were not readily apparent.

Results of the 3-4 June Survey

As expected under the more favorable wind conditions on 3-4 June, methane plumes were detectable over much larger areas than during the extreme wind conditions of the 31 January survey. Elevated methane levels occurred over much of the survey area. Additionally the methane instrument (cavity ring down spectrometer[5]) was run during travel from the survey area and during a brief observational trip to the Leroy Township area. Those two legs of the

[5] http://www.picarro.com/technology/cavity_ring_down_spectroscopy

survey trip provided methane measurements in geographically and geologically adjacent areas that can be reasonably regarded as comparable areas with limited or no shale gas well activity. That area is referred to as the Reference Area in the remainder of this report. It includes data from valleys, along a river, and two town/city areas. Hence, the Reference Area can be reasonably considered to have all likely natural and human-caused methane sources typical for the geographical/geological area, but with minimal large-scale agricultural, industrial or shale gas sources. Also, of some interest is recognition that the methane survey work included parts of two areas under Pennsylvania Department of Environmental Protection Consent Orders. An image displaying the results of the June survey is provided in Figure 5.

It should be borne in mind that the survey work was limited to publicly accessible roads. The survey, therefore, measures the impacts of methane emissions sources at considerable distances from those sources. Consequently, seemingly minor changes, in the tenths or hundredths of a part per million, in ambient air methane levels are of considerable importance in locating methane emissions sources and assessing their broader area impacts.

The June survey average methane level was 1.83 ppm, with a minimum of 1.75 ppm, 90% were below 1.88 ppm, and 99% below 2.05 ppm.[3] Given the difference in wind conditions, these levels were quite similar to those seen in the January survey. For comparison, in the Reference Area the average methane level was 1.78 ppm, with a minimum of 1.76 ppm, 90% were below 1.79 ppm, and 99% below 1.81 ppm.[3] Since much of the survey area is affected by the same type and frequency of methane sources that occur in the Reference Area, one would expect that much of the survey area data would be similar. This was, in fact, found to be the case. It can be seen in Figure 6 that in the Reference Area 97% of the methane levels were below 1.8 ppm, while in the survey area in June, 37% were, but in the survey area in January less than 1% were below 1.8 ppm. These results suggest that methane emissions in about 37% of the survey area are effectively similar to the Reference Area. The strong winds during the January compared to the June survey were probably the cause of the apparent reduction in total area with readings below 1.8 ppm (37% of the area in June compared to <1% in January), Emissions that on 3-4 June were rising into the air more normally, whereas on 31 January emissions were being rapidly mixed and swept over the land surface by the strong winds.

Looking at another methane value of interest, the maximum methane level measured in the Reference Area was 1.88 ppm. In the survey area on 3-4 June 10% of the measurements exceeded the Reference Area maximum, and on 31 January 16%. Consequently, it is reasonable to conclude that at least 10% of the survey area is impacted by methane sources that do not occur in the Reference Area. As previously mentioned, these are agricultural and industrial sources. Field observations and examination of satellite imagery allowed determination

that some of the methane sources causing the elevated methane were agricultural or industrial, other than shale gas development. The plumes of the ag/industrial sources appeared less extensive than the plumes of the sources associated with shale gas development. Most of the shale gas methane emissions sources appeared likely to be well pads and pipelines.

With regard to the relationship between ambient air methane surveys and locations of methane sources potentially impacting an area, it is interesting to consider the survey covered parts of the areas under two PaDEP Consent Orders. Those two Orders were between the PaDEP and Chesapeake Appalachia, LLC, dated 16 May 2011[6]. The two Orders were designated for impact areas referred to by PaDEP as Paradise Road and Sugar Run. It should be borne in mind that at the time of the survey, the Consent Order impact areas were not specifically known to GSI and were not specifically targeted. The general outline of the survey area was selected by DCS based on reports in the media and from residents. The specific area was determined by the operational conditions GSI encountered in the field. Consequently, the survey covered the Consent Orders impact areas only coincidentally. Still the survey did include about 2/3 of the Paradise Road and ½ of the Sugar Run Consent Order impact areas. It can be readily observed in Figure 5 that elevated methane levels were concentrated within the Paradise Road impact area compared to the remainder of the survey. There were elevated methane levels in other parts of the survey area but the concentration in the central part of the Paradise Road impact area is distinct. Though this does not prove a relationship between ambient air methane contamination and groundwater contamination, it is clearly suggestive. Further, it also suggests shale gas well operations in that area still did not have control of the gas that has been developed there. In fact, as already mentioned, the survey data indicates there may be gas control problems in about 10% of the survey area resulting in elevated methane levels over 60-90% of the area.

In addition, detection of any level of methane above normal background for an area indicates only two possible conditions: diffuse, non-point emissions are occurring over some portion of the area, or, one or more point sources are active within the area. Non-point sources are difficult to assess, precisely because they are diffuse. As mentioned previously, at the end of the survey work reported here a cursory evaluation run was made to the area of a previously documented shale gas well impact in Leroy Township. NEED LINK HERE That site is of interest in this discussion because on the land surface methane emissions occur as a non-point source, with gas emerging from many points over a area of uncertain extent. During the earlier evaluation of that site

---

[6] This PA DEP Consent Order available HERE: https://www.dropbox.com/s/3r34e3ggb88qxbo/161%20Consent%20Agreem%20Susquehana%20River.pdf

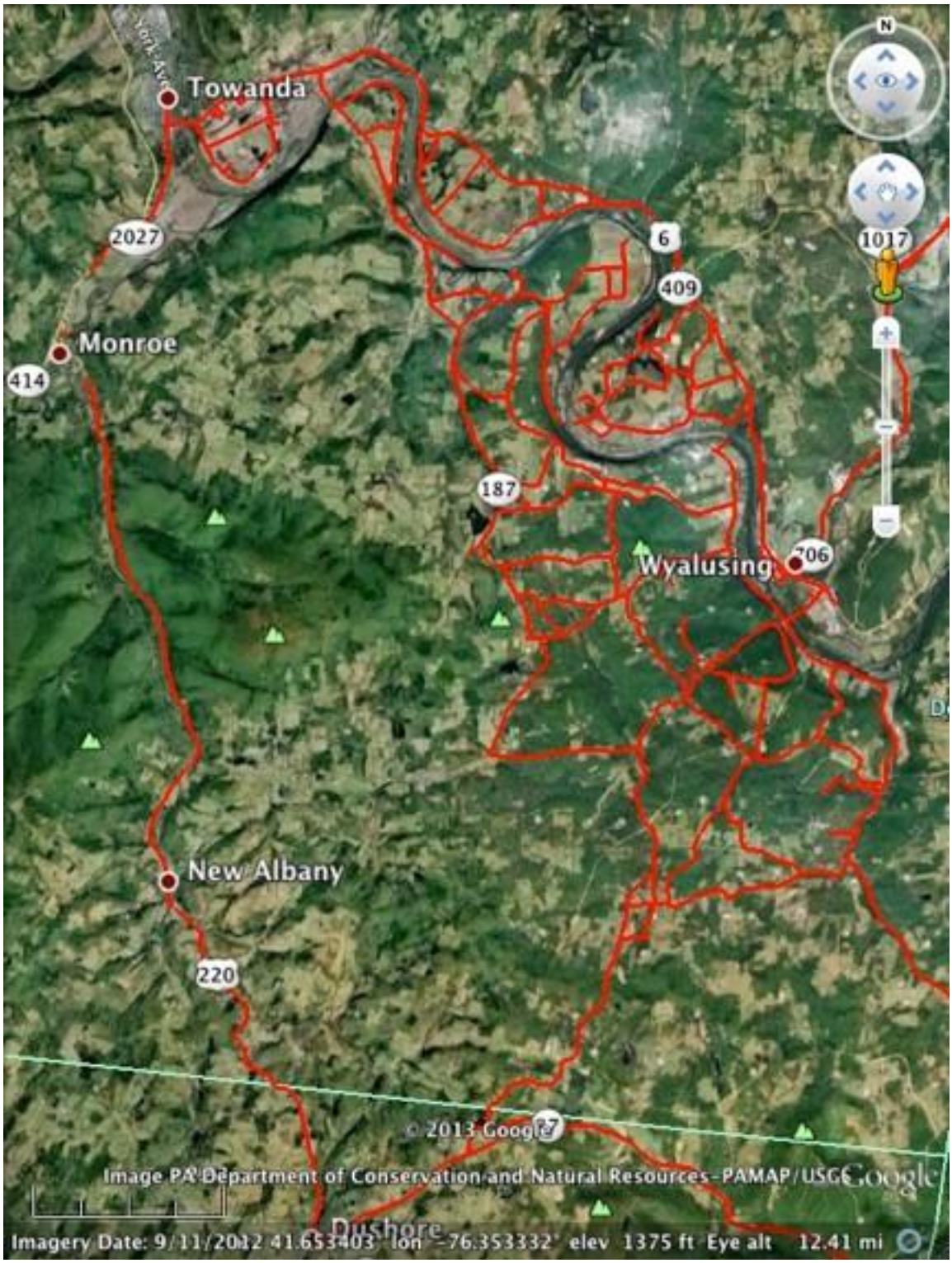
nearly pure natural gas was encountered within inches of the soil surface, but on the nearest road, about 100 yards away, and downwind at the time, only a few ppm of methane were detected. Despite gas well remediation measures, the 4 June run along the same roads confirmed methane levels remain in the range of a few ppm, suggesting the methane migration problem still exists. A cursory water sample test also indicated water in the area still has very high methane levels. Methane contamination was prevalent in the area during the prior evaluation. The Leroy Township situation is troubling with regard to health and safety, and discouraging with regard to the capability of industry to effectively correct gas well problems when they occur.

Point sources of methane present a slightly different set of concerns. A substantial amount of methane is necessary to raise methane levels even slightly over an extensive area, as measured from our survey over public roads. If that amount of methane is being emitted at one or a few point sources, then the concentration of methane in the vicinity of those sources will likely be hazardous with respect to explosion or asphyxiation. Consequently, the methane levels measured during the survey indicate there likely are point sources associated with some shale gas wells in the area that do give rise to hazardous conditions. Those point sources need not necessarily be at the gas well itself, as the gas may find underground pathways to emerge in water wells, homes or other structures, as occurred in Leroy Township, and the Paradise Road and Sugar Run impact areas.

Conclusions

Methane from any source rapidly diffuses and rises in the air. Consequently, detection of possible methane sources from any distance away requires extremely sensitive measurement capabilities. The GSI survey approach takes advantage of extremely sensitive measurement instrumentation to detect small increases in ambient air methane levels as an indication of probable methane emissions sources in a given area. Based on the data collected using that equipment, we conclude that the Towanda-Wyalusing area is probably substantially impacted by methane emissions from shale gas wells both within and beyond the survey area, depending on wind conditions. The coincidence of two DEP methane migration impact areas, Paradise Road and Sugar Road, and the most marked ambient air methane levels suggests there are still gas control problems associated with the shale gas wells there, as well as in another documented impact area in Leroy Township also cursorily measured following the main survey. A rapid water test in the Leroy area confirmed the water in that area is still contaminated with methane. These survey results suggest methane contamination continues and measures taken by gas well operators with regard to methane migration problems that have occurred in these three areas have likely been only partially effective.

Figure 1. Overhead image of roads traveled during the survey of ambient air methane levels in the vicinity of Wyalusing, PA on 31 January 2013 (Google Earth).



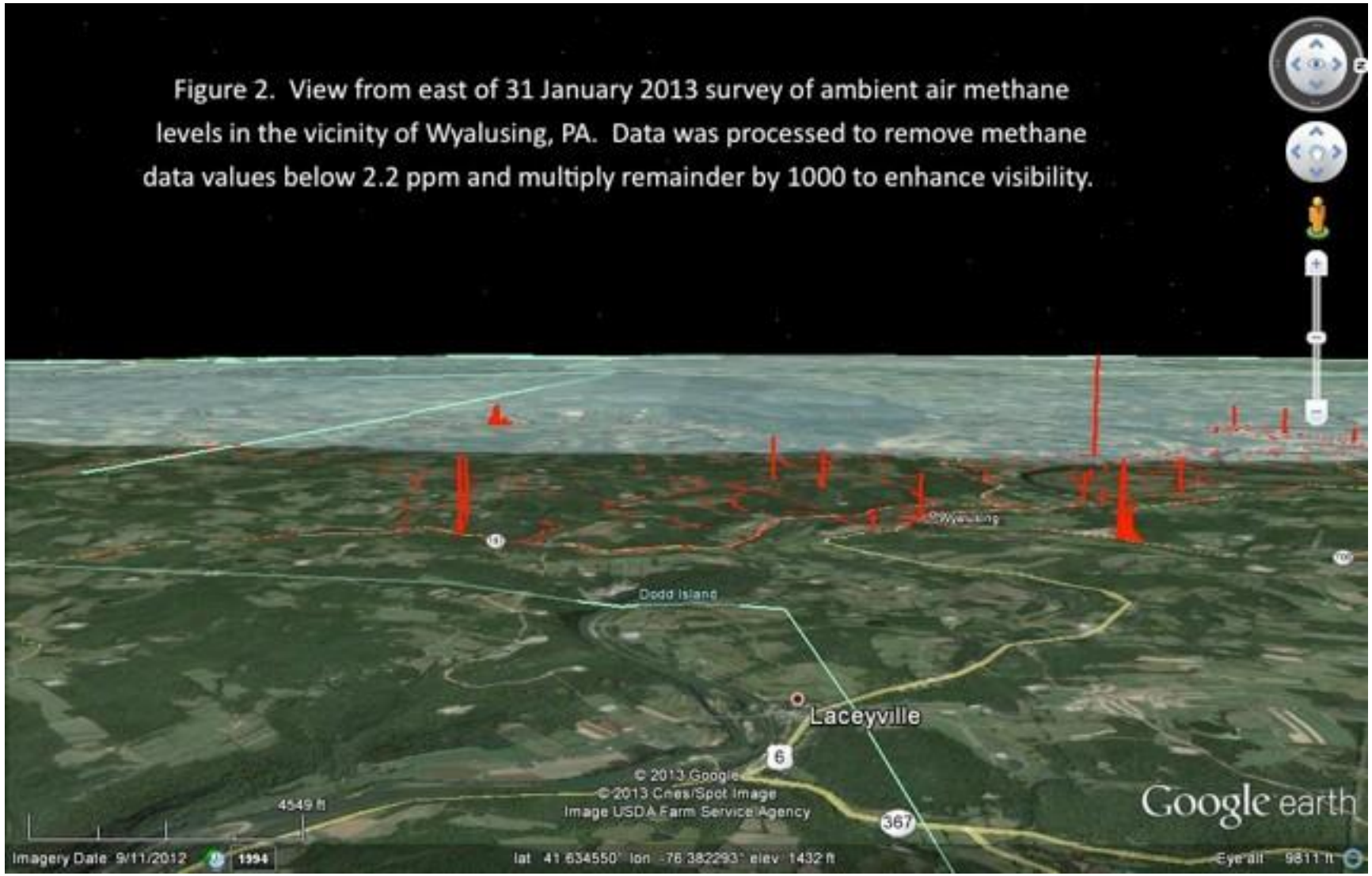


Figure 2. View from east of 31 January 2013 survey of ambient air methane levels in the vicinity of Wyalusing, PA. Data was processed to remove methane data values below 2.2 ppm and multiply remainder by 1000 to enhance visibility.

Figure 3. An elevated methane level as rendered by processing of the Wyalusing 31 January 2013 methane survey data to remove values <2.2ppm and multiply remainder by 1000. Compare to same elevated methane location in Figure 4.

Figure 4. An elevated methane level as rendered by processing of the Wyalusing 31 January 2013 methane survey data to remove values <1.9ppm and multiply remainder by 100. Compare to same elevated methane location in Figure 3.

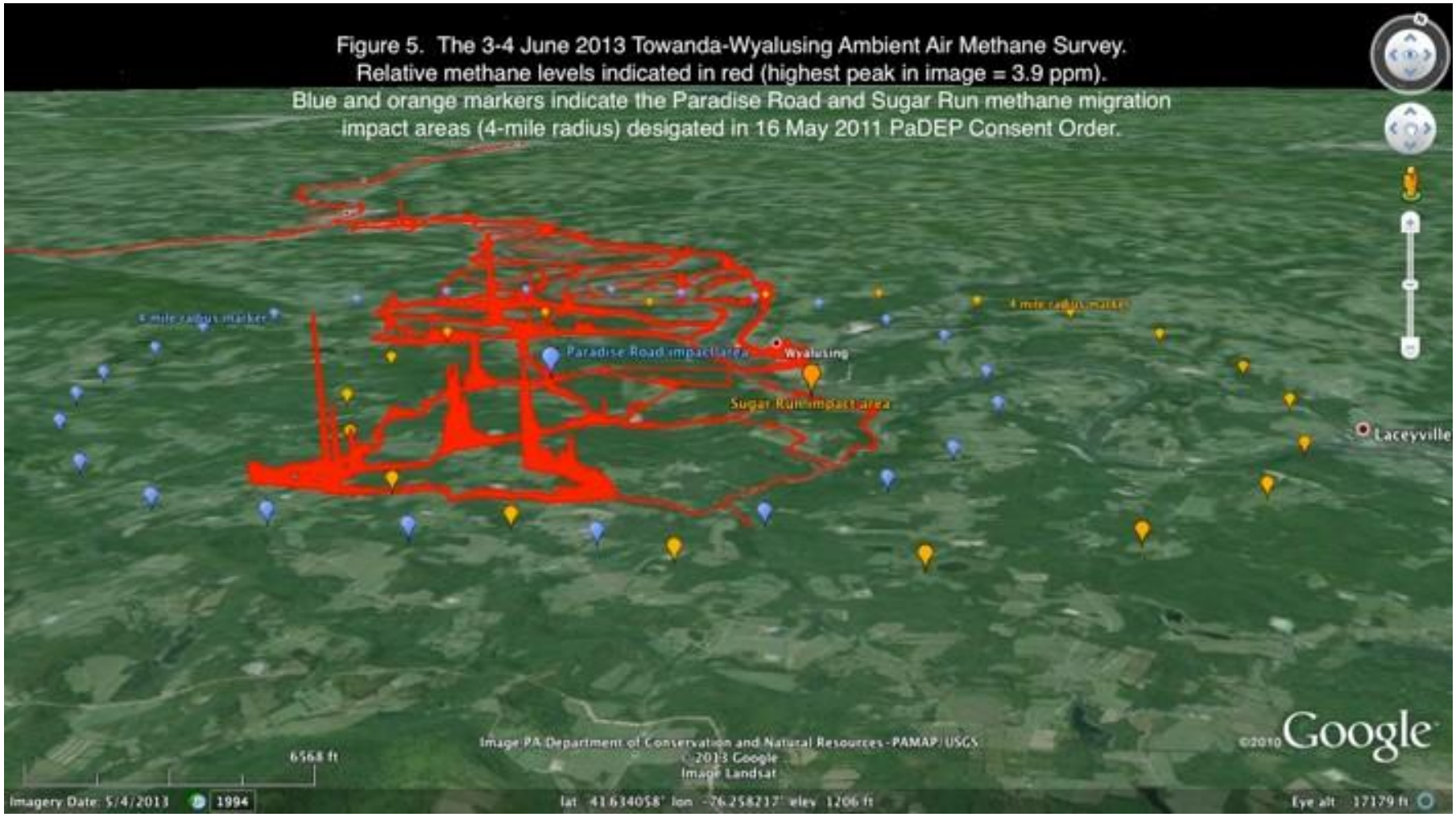
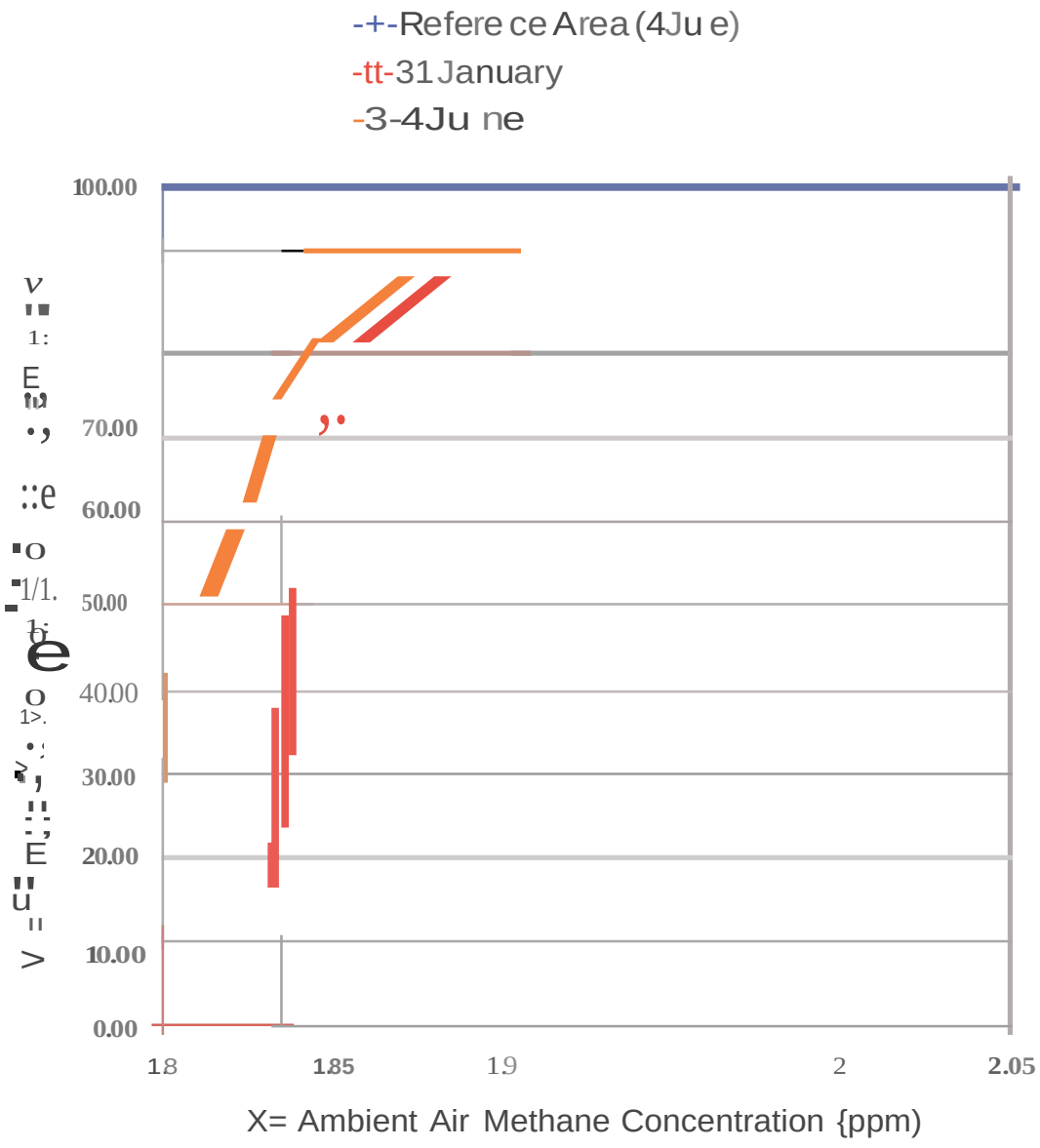


Figure 5. The 3-4 June 2013 Towanda-Wyalusing Ambient Air Methane Survey.
Relative methane levels indicated in red (highest peak in image = 3.9 ppm).
Blue and orange markers indicate the Paradise Road and Sugar Run methane migration
impact areas (4-mile radius) desigated in 16 May 2011 PaDEP Consent Order.
4 mile radius marker
Paradise Road impact area
Wyalusing
Sugar Run impact area
4 mile radius marker
Laceyville
Image PA Department of Conservation and Natural Resources - PAMAP / USGS
©2013 Google
Image Landsat
©2010 Google
6568 ft
Imagery Date: 5/4/2013
1994
lat 41.634058° lon -76.258217° elev 1206 ft
Eye alt 17179 ft

Figure 6. Ambient Air Methane Surveys Towanda-Wyalusing Area, PA January and June 2013



Appendix G

Gas Safety Incorporated
16 Brook Lane
Southborough, Massachusetts 01772
774-922-4626
www.gassafetyusa.com

Report to the Clean Air Council
on
8 June 2012 Field Inspection and Methane Sampling Survey
of
Parts of Leroy, Granville and Franklin Townships
Bradford County, Pennsylvania

NOTE: Specific location identification information is avoided in this report pending approval of involved or potentially involved parties.

SUMMARY
A portable laser-based methane measurement system was used to survey methane levels in northeastern Leroy Township, Bradford County, Pennsylvania and adjacent parts of Granville and Franklin Townships on 8 June 2012. The methane system reports methane levels in air to the nearest part per billion (ppb) every 3-4 seconds. During the survey over 7,600 methane measurements were made. The survey data indicated one or more substantial methane emissions were occurring in an area near and to the west of H Rockwell Road and Route 414 giving rise to a ground level plume that expanded to cover at least 4.2 square kilometers over a period of 3.5 hours. The size and rate of expansion of the plume suggested large amounts of methane were being emitted to the atmosphere. Heavily methane contaminated residential water wells occurred in and around the same area, and documentation indicated heavy contamination has existed for at least several months. Bubbling gas in Towanda Creek suggest fugitive gas from shale gas wells may be travelling through faults and fractures, which also carry local ground water, hence, impact local water wells. Collectively the data and observations suggest natural gas has pervaded an extensive subsurface area beyond the area where elevated ground-level methane was found during this survey effort. If that is correct, then more surface emissions are likely and should be expected. The issues and concerns presented in this report require more thorough investigation for confirmation and quantification.

BACKGROUND

A number of dramatic fugitive methane emissions were reported to have begun in Leroy Township on 19 May 2012. Reports suggested a substantive loss of control of natural gas flows from one or more of the shale gas wells in the Township may have occurred. In the interest of verifying and developing independent documentation of the reportedly large increases in natural gas emissions, the Clean Air Council ("CAC", Philadelphia, PA) contracted Gas Safety, Inc. ("GSI", Southboro, MA) to do a one-day sampling and area visit to ascertain the locations of observed or suspected natural gas emissions. The intention was for GSI to use a customized, portable Cavity Ring-Down Spectrometry (CRDS) methane measurement instrument to investigate and document the occurrence (or not) of the reported emissions.

A major concern was to perform the assessment as soon as possible to better ascertain the possible initial intensity and extent of the event; that is, this would preferably be a short notice, rapid response effort. Other concerns were GSI instrument availability and efficient use of field time with the instrument. In order to assure a one-day effort would be as productive as practical, GSI and CAC contacted various parties in pursuit of information regarding specific locations of reported point-source gas emissions. Like the survey trip itself, such requests for information had to be short notice, rapid response efforts. In the interest of openness and sharing of information GSI proposed to provide through CAC its findings to cooperating parties. GSI contacted the Emergency Management Agency of Bradford County, which preliminarily offered to share its own records regarding the initial reports of the sudden onset gas emissions event. CAC and GSI also contacted various private parties with similar data sharing offers and rapid response requests for information and property access. Within 24 hours of such requests, and despite initially positive responses, only 3 private parties agreed to provide information or access to suspected emissions or impacted areas on private property. Ultimately no specific identification of or authorization for access to the actual point locations of ongoing natural gas emissions was obtained in time for the survey. Hence, work was limited to surveying methane levels on public roadways and verifying methane in well water in three residences and collecting anecdotal reports on three others.

Weather conditions were mild and favorable. Barometric pressure was steady. Winds were from the west-northwest increasing steadily throughout the day from nearly calm to a few miles per hour by the end of the survey work.

The group involved in the methane sampling survey (listed just below) met at the junction of Routes 414 and 514 in the northwest corner of Franklin Township at approximately 09:15 AM on 8 June 2012.

Carolyn Knapp, Bradford County resident
Dan Natt, Bradford County resident
Matt Walker, Clean Air Council
Ron Kanter, videographer, Clean Air Council
Dr. Brian Redmond, PG, Wilkes University,
Dept. of Environmental Engineering and Earth Sciences
Bob Ackley, Gas Safety, Inc.
Dr. Bryce F. Payne Jr., Gas Safety, Inc.

METHANE IN RESIDENTIAL WATER WELLS

A total of four residences (referred to as house1, house2,…) were visited, all served by on-site wells with an interview at house 5 regarding houses 5 and 6. House1 was vacant. It was reported that the residents had vacated due to the inconvenience of and health concerns related to elevated levels of methane and contamination in well water. The house could not be entered, and due to lack of power, no well water could be sampled. Concentrations of methane in the air on the property were normal (normal background ambient air methane 1.75-1.95 ppm).

House2 was located on a farm near a gas well pad (Morse 3H and 5H wells). Most of the area had elevated ambient air methane levels (max. 2.5 ppm) that appeared possibly associated with animal manure accumulations on the farm. However, in areas where there was substantial manure, the presumed likely source of the methane, ambient methane levels were rarely above background and never exceeded 2.2 ppm. Upslope from the farm building area ambient air methane levels were normal. At 200 meters east of the gas well pad methane levels were normal. Down slope, toward Towanda Creek methane levels were elevated, with three locations showing >100 ppm in the ambient air just above the surface of the creek bank. In these instances the methane could have been biogenic or fugitive thermogenic, but sampling conditions did not permit collection of samples for isotope analysis.

The water at the kitchen tap in house3 and house4 was supersaturated with methane. Upon flowing from the faucet the water appeared "milky" due to the large amount of fine methane bubbles present. The fine bubbles coalesced over a period of several seconds causing a pronounced effervescence. High levels of methane in the gas evolved from the tap water were confirmed with the CRDS instrument. No attempt was made to verify initial methane concentration in the tap water, but reports of analyses of samples previously collected by PaDEP or contractors indicated that the well water in these homes had been confirmed to contain methane levels from 50 to 100 milligrams per liter, much greater than saturation under atmospheric pressure (about 28 milligrams per liter). Ongoing supersaturation of well water can only occur if there is substantial water "head" pressure in the well and the methane is under sufficient pressure to reach aquifers under such pressure. It should also be noted that such methane levels are sufficient to pose asphyxiation hazards if used for showering or other high water uses in close quarters, symptoms of which the residents of these properties reported.

An additional inquiry was made at another residence (designated house 5). This was a no-notice contact initially to request information on ownership of the adjacent property. The occupant at house5 reported the well water at that house was similarly heavily contaminated, as well as the well of a close relative who lived in another nearby house (house6).

The wellheads at houses 3-6 had been equipped with passive or wind turbine vents, reportedly by either PaDEP or gas company contractors. Such vents are not designed to prevent or treat contamination of water in wells with conditions and methane exposures of the type that can

cause such super-saturation with methane. Presumably the passive vents were installed to prevent pressure driven flow of methane into the homes through possible underground pathways. Though clearly better than the risk of not venting, the application of only passive vents leave the residents under continuing risk of exposures to asphyxiating concentrations of methane, ignore the at least substantial nuisance of having to use methane-super-saturated water, and the potential for serious eruptive releases of methane up through the water well. In addition, such levels of methane contamination necessarily imply the possibility of indirect effects on water quality due to induced biological and chemical changes in the ground water and the mineral medium through which it flows. Such effects might take months or years to become fully apparent, and present a serious concern with regard to long-term degradation of aquifers in areas where even less intensive methane contamination occurs.

In summary, of 6 houses visited or about which information was obtained, 5 had well water that was supersaturated with methane. All 5 of those in which methane contamination was observed or reported lie north of Towanda Creek. Four of the five contaminated residences were occupied at the time of this inquiry, and at all 4, passive vents had been installed with the foreseeable lack of effect on methane contamination of the water. The intensity of the methane contamination seems to require more definitive treatment measures as well as efforts to identify the source or sources of the contamination and actions to prevent long-term degradation of aquifers.

SURVEY OF THE AREA FOR METHANE IN THE AIR

Cavity Ring-Down Spectrometry and Baseline Ground-Level Methane Data

The CRDS instrument is extremely sensitive, runs continuously, and is robust. Consequently the unit quickly generates large volumes of highly reliable methane measurements on a continuous basis. During the one-day area survey reported here, the instrument generated 7,697 methane measurements. In combination with similar quantities of data from prior surveys in the eastern Marcellus Shale region, GSI has determined that a reliable (99.99% confidence level) upper bound for background methane levels in ground level air is 1.95 parts per million (ppm). GSI also has identified thousands of gas leaks in commercial pipelines in a variety of settings and based on that experience has concluded that CRDS measured levels of methane in excess of 2.05 ppm reliably indicate a natural gas leak in the surrounding area. Based on these findings, GSI interprets methane levels above 1.95 ppm as presumptive, and above 2.05 ppm as highly probable methane contamination. There is potential for some biogenic sources to generate enough methane to cause such readings, but such potential biogenic sources are usually readily identifiable, and limited in both extent and intensity in comparison to fugitive natural gas from wells or infrastructure. When more definitive evidence is needed, gas samples are collected and analyzed for isotopic composition for comparison to similar data for suspected sources of contaminating gas.

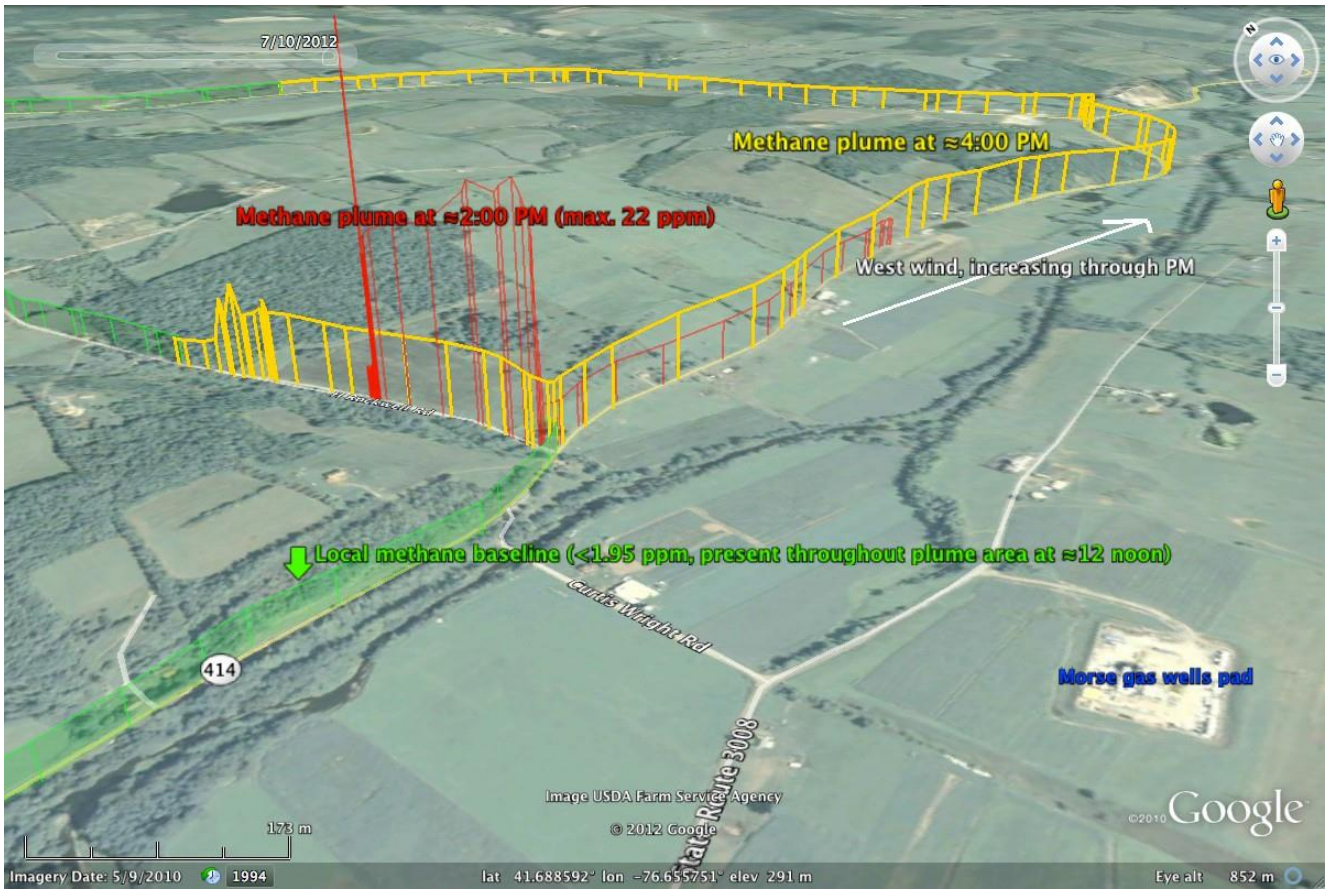
The areas in Leroy, Granville, and Franklin Townships surveyed and reported here had background levels and variations typical for the region, the lowest methane reading being 1.674 ppm (nominal accuracy of the CRDS is 0.001 ppm). Some areas of elevated methane in

the air occurred near areas on farms with long-term animal manure loads. No elevated methane levels were found for carcass handling, and other agricultural areas that might be conventionally considered suspect for biogenic methane production. Interestingly no elevated methane levels were measured at the nearest access (200 meters) to the natural gas well pad (Morse 3H and 5H wells) within the area covered by this survey. This would seem a reasonable finding given the well is new, with limited and new infrastructure, and there was no wind during sampling in that area. With no wind and the low density of methane (half that of air), any gas leaks comprised primarily of methane would likely rise directly upward and go undetected without adequately close access to the vicinity of the leak.

Elevated methane levels, however, were detected as soon as the instrument was activated at the junction of Routes 414 and 514. All of the initial 157 readings were above 1.95 ppm, 152 were above 2.00 ppm. Such sustained levels above 1.95 indicate a fugitive methane source upwind. An initial drive and walk survey along and near Route 514 covering approximately 2 kilometers to the north and back indicated no methane above reasonable background levels. The initially observed elevated readings at the junction of Routes 414 and 514 had diminished when the instrument was returned to the location just over one hour later.

A driving survey west on Rt 414 (0.6 kilometers), south on Cross Road (0.5 kilometers), and west on South Side Road (2.2 kilometers) again revealed no elevated methane levels, as did a walking survey upslope from South Side Road, downwind from the Morse gas well pad.

Upon descending to the banks of Towanda Creek, methane levels rose above baseline in the vicinity of the creek banks. Random sampling at three locations showed maximum methane levels immediately above the soil surface of 133, 391, and 713 ppm. At the time of the observations there was no basis for inferring whether the methane was more likely biogenic or fugitive thermogenic gas. Methane levels were slightly elevated over most of the surveyed area along the creek.



The next leg of the survey involved a return east on South Side Road, then north across the bridge, and west along Rt 414 (2.2 kilometers) and north on H Rockwell Road (1 kilometer) (below red methane spike in image above). Methane levels were normal (indicated by green methane level markers in image above) until reaching H Rockwell Road, where slightly elevated levels were again encountered (average of 32 readings = 2.068, range = 1.967 to 2.184ppm) northbound along the first approximately 500 meters of that road. About an hour later, on the return trip south on H Rockwell Road and east on Rt 414 the methane levels (red methane level markers in image above) had risen substantially and the affected area expanded south and east. Methane levels began to rise relatively suddenly about 500meters north of Rt 414 from 2.01 ppm to a maximum of 21.979 ppm, then settled into a range of 10 to 14 ppm. The area of elevated methane levels had expanded to the south and east as indicated by measurements along Rt 414 showing levels descending from 4.620 ppm at H Rockwell Road to 2.049 ppm approximately 1 kilometer to the east. Another survey pass was made through the area approximately 1 hour 50 minutes later driving eastbound on Rt 414 (yellow methane level markers in image above). The elevated methane levels were then found to have expanded to cover an area from Rockwell Road east along Rt 414 for 2.8 kilometers then north along Rt 514 (2.8 kilometers) at an overall average concentration of 3.8 ppm. The data clearly indicated that one or more methane emissions were present and releasing substantial amounts of methane into the atmosphere probably within 500 meters to the north of Rt 414, near and to the west of Rockwell Road along with other possible emissions occurring or developing within the area enclosed by Rockwell Road and Rts 414 and 514. The measured

plume covered an area of approximately 4.2 square kilometers, however, methane data and wind direction indicate the plume probably extended considerably farther to the south and east. Time was insufficient to measure the full extent of the plume to the south and east.

Gas was reported to have been bubbling up in Towanda Creek beneath the Cross Road bridge. The bridge was visited to view the gas bubbling, if present. Upon arrival the bubbling proved to be relatively easily observed. Batches of bubbles were rising to the surface at consistent time intervals and locations, fairly regularly spaced along a line running roughly east-northeast for the entire distance visible from the bridge, about 100 meters west to a somewhat shorter distance east. The directional orientation of the line of bubbles and regular spacing between bubbling points suggested association with a local fault or related subsurface structure. The total volume of bubbles per batch was very roughly estimated to be at least 300 cubic centimeters. Over the visible length of the bubble line the bubbling was nearly always occurring at one or more of the locations. Hence, the observed bubbling area was estimated to have been releasing at least 300 cubic centimeters per second, or 18 liters per minute, or 38 cubic feet per hour.

The volume and spatial distribution of the bubbling locations make other potential explanations, e.g., a biogenic methane source in the creek bottom, seem implausible. When the direction of the bubbling line under the bridge was extended to the west-southwest, it intersected the area where methane had been measured in the creek bank soils earlier in the day, suggesting the possibility that methane emissions may have been occurring along a fault line, but due to lack of access and time there was no opportunity to evaluate this possibility.

It is important and useful to note that the gas released in the creek under the bridge could not be confirmed to be methane with the CRDS instrument due to wind conditions and no access to the bubbling points in the creek due to the high elevation of the deck of the bridge. Further, there is the possibility that the gas in the bubbles is comprised of other gases besides methane. This could presumably be due the air normally present in local faults and fractures being displaced by methane intruding under pressure. If this were the case, then the methane content of the gas in the bubbles would initially contain little or no thermogenic methane, with relatively sudden increase in methane concentration once intruding methane effectively purges the fracture

The data available from 3 survey drive-by passes over this area spanned a period of 3.5 hours. Assuming the measured concentration is consistent from the ground surface to 2 meters above, the volume of ground level air in the plume area is 4.2 square kilometers X 2m = 4,200,000 square meters x 2m = 8,400,000 cubic meters. A methane concentration increase of 1.8 ppm would require 15.2 cubic meters of methane. Given the 3.5 hours over which this accumulation occurred, the implied emission rate is 4.3 cubic meters, or 150 cubic feet per hour. This, however, is a major underestimation of the likely volume of gas being released in the identified plume. Methane is a low-density gas, about half the density of air. Consequently, methane will tend to rise in the air relatively rapidly and the lowest methane concentrations in the vicinity of a surface methane emission will be expected to occur at ground level. It follows, therefore, that an estimate of the likely methane emission rate in the identified plume area that includes the vertical extent of the plume would be orders of

magnitude greater than the above estimate (150 cubic feet per hour) based on ground level methane only. Application of air contaminant diffusion models appropriate to estimating the full-height methane emission rate was beyond the scope of this effort. The most definitive and reliable approach would be direct investigation of methane emissions through water and soil surfaces using the CRDS instrument and appropriate related equipment. However, this approach requires direct access to the properties on which the methane emissions are occurring, which could not be obtained for this effort. Further, emissions through soil surfaces typically are invisible and may occur for prolonged periods with no recognition until vegetation is damaged or killed by asphyxiation of the roots. Hence, many property owners may be heavily impacted but be unaware, and, therefore, reluctant to participant in methane emission survey efforts.

In summary, the methane survey data collected on 8 June 2012 in parts of Leroy, Granville, and Franklin Townships, Bradford County, Pennsylvania indicated one or more substantial methane emissions were occurring in an area centered roughly on the intersection of H Rockwell Road and Route 414. A ground level plume was detected that increased in area substantially over a period of 3.5 hours, which, when expanded to account for above ground level methane, suggests large amounts of methane were being emitted to the atmosphere. Heavily methane contaminated residential water wells occurred in and around the same area, and documentation indicated heavy contamination had existed for at least several months. Bubbling gas in Towanda Creek suggested fugitive gas from shale gas wells might be travelling through faults and fractures, which also carry local ground water, hence, impact local water wells. Collectively the data and observations suggest natural gas has pervaded an extensive subsurface area beyond the area where elevated ground-level methane was found during this survey effort. If that is correct, then more surface emissions should be expected. The issues and concerns presented in this report require more thorough investigation for confirmation and quantification.

Appendix H

ENERGY

Methane leaks erode green credentials of natural gas

Losses of up to 9% show need for broader data on US gas industry's environmental impact.

BY JEFF TOLLEFSON

Scientists are once again reporting alarmingly high methane emissions from an oil and gas field, underscoring questions about the environmental benefits of the boom in natural-gas production that is transforming the US energy system.

The researchers, who hold joint appointments with the National Oceanic and Atmospheric Administration (NOAA) and the University of Colorado in Boulder, first sparked concern in February 2012 with a study[1] suggesting that up to 4% of the methane produced at a field near Denver was escaping into the atmosphere. If methane — a potent greenhouse gas — is leaking from fields across the country at similar rates, it could be offsetting much of the climate benefit of the ongoing shift from coal- to gas-fired plants for electricity generation.

Industry officials and some scientists contested the claim, but at an American Geophysical Union (AGU) meeting in San Francisco, California, last month, the research team reported new Colorado data that support the earlier work, as well as preliminary results from a field study in the Uinta Basin of Utah suggesting even higher rates of methane leakage — an eye-popping 9% of the total production. That figure is nearly double the cumulative loss rates estimated from industry data — which are already higher in Utah than in Colorado.

"We were expecting to see high methane levels, but I don't think anybody really comprehended the true magnitude of what we would see," says Colm Sweeney, who led the aerial component of the study as head of the aircraft programme at NOAA's Earth System Research Laboratory in Boulder.

Whether the high leakage rates claimed in Colorado and Utah are typical across the US natural-gas industry remains unclear. The NOAA data represent a "small snapshot" of a much larger picture that the broader scientific community is now assembling, says Steven Hamburg, chief scientist at the Environmental Defense Fund (EDF) in Boston, Massachusetts.

The NOAA researchers collected their data in February as part of a broader analysis of air pollution in the Uinta Basin, using ground-based equipment and an aircraft to make detailed measurements of various pollutants, including methane concentrations. The researchers used atmospheric modelling to calculate the level of methane emissions required to reach those concentrations, and then compared that with industry data on gas production to obtain the percentage escaping into the atmosphere through venting and leaks.

Natural-gas wells such as this one in Colorado are increasingly important to the US energy supply.

L DAVID ZALUBOWSKI/AP

The results build on those of the earlier Colorado study[1] in the Denver–Julesburg Basin, led by NOAA scientist Gabrielle Pétron (see *Nature* **482,** 139–140; 2012). That study relied on pollution measurements taken in 2008 on the ground and from a nearby tower, and estimated a leakage rate that was about twice as high as official figures suggested. But the team's methodology for calculating leakage — based on chemical analysis of the pollutants — remains in dispute. Michael Levi, an energy analyst at the Council on Foreign Relations in New York, published a peer-reviewed comment[2] questioning the findings and presenting an alternative interpretation of the data that would align overall leakage rates with previous estimates.

Pétron and her colleagues have a defence of the Colorado study in press[3], and at the AGU meeting she discussed a new study of the Denver–Julesburg Basin conducted with scientists at Picarro, a gas-analyser manufacturer based in Santa Clara, California. That study relies on carbon isotopes to differentiate between industrial emissions and methane from cows and feedlots, and the preliminary results line up with their earlier findings.

A great deal rides on getting the number right. A study[4] published in April by scientists at the EDF and Princeton University in New Jersey suggests that shifting to natural gas from coal-fired generators has immediate climatic benefits as long as the cumulative leakage rate from natural-gas production is below 3.2%; the benefits accumulate over time and are even larger if the gas plants replace older coal plants. By comparison, the authors note that the latest estimates from the US Environmental Protection Agency (EPA) suggest that 2.4% of total natural-gas production was lost to leakage in 2009.

To see if that number holds up, the NOAA scientists are also taking part in a comprehensive assessment of US natural-gas emissions, conducted by the University of Texas at Austin and the EDF, with various industry partners. The initiative will analyse emissions from the production, gathering, processing, long-distance transmission and local distribution of natural gas, and will gather data on the use of natural gas in the transportation sector. In addition to scouring through industry data, the scientists are collecting field measurements at facilities across the country. The researchers expect to submit the first of these studies for publication by February, and say that the others will be complete within a year.

In April, the EPA issued standards intended to reduce air pollution from hydraulic-fracturing operations — now standard within the oil and gas industry — and advocates say that more can be done, at the state and national levels, to reduce methane emissions. "There are clearly opportunities to reduce leakage," says Hamburg. ■

1. Pétron, G. *et al. J. Geophys. Res.* **117,** D04304 (2012).
2. Levi, M. A. *J. Geophys. Res.* **117,** D21203 (2012).
3. Pétron, G. *et al. J. Geophys. Res.* (in the press).
4. Alvarez, R. A., Pacala, S. W. Winebrake, J. J., Chameides, W. L. & Hamburg, S. P. *Proc. Natl Acad. Sci. USA* **109,** 6435–6440 (2012).

Appendix I

NEWS IN FOCUS

J. SARTORE/NATIONAL GEOGRAPHIC STOCK

Natural-gas operations in areas such as Wyoming's Jonah Field could release far more methane into the atmosphere than previously thought.

CLIMATECHANGE

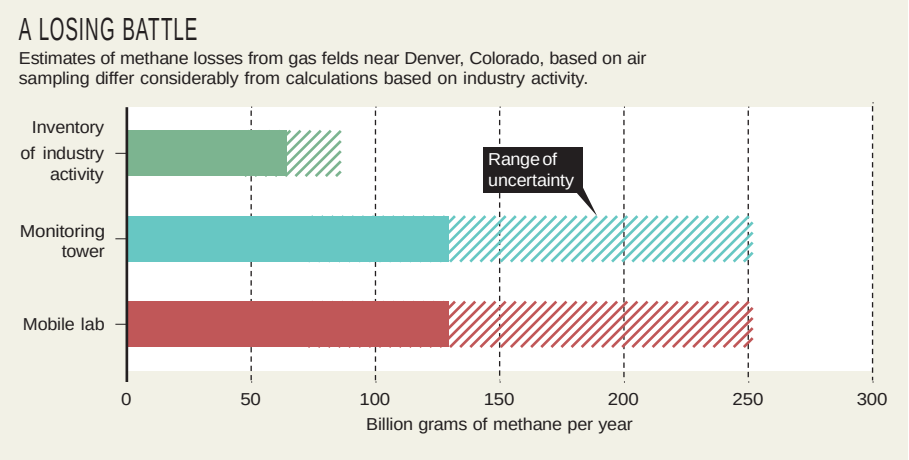
Air sampling reveals high emissions from gas field

Methane leaks during production may offset climate benefits of natural gas.

BY JEFF TOLLEFSON

When US government scientists began sampling the air from a tower north of Denver, Colorado, they expected urban smog — but not strong whiffs of what looked like natural gas. They eventually linked the mysterious pollution to a nearby natural-gas field, and their investigation has now produced the first hard evidence that the cleanest-burning fossil fuel might not be much better than coal when it comes to climate change.

Led by researchers at the National Oceanic and Atmospheric Administration (NOAA) and the University of Colorado, Boulder, the study estimates that natural-gas producers in an area known as the Denver-Julesburg Basin are losing about 4% of their gas to the atmosphere — not including additional losses in the pipeline and distribution system. This is more than double the official inventory, but roughly in line with estimates made in 2011 that have been challenged by industry. And because methane is some 25 times more efficient than carbon dioxide at trapping heat in the atmosphere, releases of that magnitude ▶



G. PÉTRON *ET AL. J. GEOPHYS. RES.*

▶ could effectively offset the environmental edge that natural gas is said to enjoy over other fossil fuels.

"If we want natural gas to be the cleanest fossil fuel source, methane emissions have to be reduced," says Gabrielle Pétron, an atmospheric scientist at NOAA and at the University of Colorado in Boulder, and first author on the study, currently in press at the *Journal of Geophysical Research*. Emissions will vary depending on the site, but Pétron sees no reason to think that this particular basin is unique. "I think we seriously need to look at natural-gas operations on the national scale."

The results come as a natural-gas boom hits the United States, driven by a technology known as hydraulic fracturing, or 'fracking', that can crack open hard shale formations and release the natural gas trapped inside. Environmentalists are worried about effects such as water pollution, but the US government is enthusiastic about fracking. In his State of the Union address last week, US President Barack Obama touted natural gas as the key to boosting domestic energy production.

LACK OF DATA

Natural gas emits about half as much carbon dioxide as coal per unit of energy when burned, but separate teams at Cornell University in Ithaca, New York, and at the US Environmental Protection Agency (EPA) concluded last year that methane emissions from shale gas are much larger than previously thought. The industry and some academics branded those findings as exaggerated, but the debate has been marked by a scarcity of hard data.

"It's great to get some actual numbers from the field," says Robert Howarth, a Cornell researcher whose team raised concerns about methane emissions from shale-gas drilling in a pair of papers, one published in April last year and another last month (R. W. Howarth *et al. Clim. Change Lett.* **106,** 679–690; 2011; R. W. Howarth *et al. Clim. Change* in the press). "I'm not looking for vindication here, but [the NOAA] numbers are coming in very close to ours, maybe a little higher," he says.

Natural gas might still have an advantage over coal when burned to create electricity, because gas-fired power plants tend to be newer and far more efficient than older facilities that provide the bulk of the country's coal-fired generation. But only 30% of US gas is used to produce electricity, Howarth says, with much of the rest being used for heating, for which there is no such advantage.

ON THE SCENT

The first clues appeared in 2007, when NOAA researchers noticed occasional plumes of pollutants including methane, butane and propane in air samples taken from a 300-metre-high atmospheric monitoring tower north of Denver. The NOAA researchers worked out the general direction that the pollution was coming from by monitoring winds, and in 2008, the team took advantage of new equipment and drove around the region, sampling the air in real time. Their readings led them to the Denver-Julesburg Basin, where more than 20,000 oil and gas wells have been drilled during the past four decades.

"A big part of it is just raw gas that is leaking from the infrastructure."

Most of the wells in the basin are drilled into 'tight sand' formations that require the same fracking technology being used in shale formations. This process involves injecting a slurry of water, chemicals and sand into wells at high pressure to fracture the rock and create veins that can carry trapped gas to the well. Afterwards, companies need to pump out the fracking fluids, releasing bubbles of dissolved gas as well as burps of early gas production. Companies typically vent these early gases into the atmosphere for up to a month or more until the well hits its full stride, at which point it is hooked up to a pipeline.

NATURE.COM
Should fracking stop?
go.nature.com/adox2r

The team analysed the ratios of various pollutants in the air samples and then tied that chemical fingerprint back to emissions from gas-storage tanks built to hold liquid petroleum gases before shipment. In doing so, they were able to work out the local emissions that would be necessary to explain the concentrations that they were seeing in the atmosphere (see 'A losing battle'). Some of the emissions come from the storage tanks, says Pétron, "but a big part of it is just raw gas that is leaking from the infrastructure". Their range of 2.3–7.7% loss, with a best guess of 4%, is slightly higher than Cornell's estimate of 2.2–3.8% for shale-gas drilling and production. It is also higher than calculations by the EPA, which revised its methodology last year and roughly doubled the official US inventory of emissions from the natural-gas industry over the past decade. Howarth says the EPA methodology translates to a 2.8% loss.

The Cornell group had estimated that 1.9% of the gas produced over the lifetime of a typical shale-gas well escapes through fracking and well completion alone. NOAA's study doesn't differentiate between gas from fracking and leaks from any other point in the production process, but Pétron says that fracking clearly contributes to some of the gas her team measured.

Capturing and storing gases that are being vented during the fracking process is feasible, but industry says that these measures are too costly to adopt. An EPA rule that is due out as early as April would promote such changes by regulating emissions from the gas fields.

Officials with America's Natural Gas Alliance, based in Washington DC, say that the study is difficult to evaluate based on a preliminary review, but in a statement to *Nature* they add that "the findings raise questions and warrant a closer examination by the scientific community". Environmental groups are pushing the EPA to strengthen pollution controls in the pending rule, but industry is pushing to relax many of the requirements. Many companies are already improving their practices and reducing emissions throughout the country, either voluntarily or by regulation, the alliance says.

Not all studies support the higher methane numbers. Sergey Paltsev, assistant director for economic research at the Massachusetts Institute of Technology Energy Initiative in Cambridge, and his colleagues are gathering information about industry practices for a study on shale-gas emissions. He says that their figures are likely to come in well below even the lower EPA estimate. He calls the NOAA results "surprising" and questions how representative the site is.

Pétron says that more studies are needed using industry inventories and measurements of atmospheric concentrations. "We will never get the same numbers," she says, "but if we can get close enough that our ranges overlap in a meaningful way, then we can say we understand the process." ■

Appendix J

Anthropogenic emissions of methane in the United States

Scot M. Miller[a,1], Steven C. Wofsy[a], Anna M. Michalak[b], Eric A. Kort[c], Arlyn E. Andrews[d], Sebastien C. Biraud[e], Edward J. Dlugokencky[d], Janusz Eluszkiewicz[f], Marc L. Fischer[g], Greet Janssens-Maenhout[h], Ben R. Miller[i], John B. Miller[i], Stephen A. Montzka[d], Thomas Nehrkorn[f], and Colm Sweeney[i]

[a]Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA 02138; [b]Department of Global Ecology, Carnegie Institution for Science, Stanford, CA 94305; [c]Department of Atmospheric, Ocean, and Space Sciences, University of Michigan, Ann Arbor, MI 48109; [d]Global Monitoring Division, Earth System Research Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO 80305; [e]Earth Sciences Division, and [g]Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720; [f]Atmospheric and Environmental Research, Lexington, MA 02421; [h]Institute for Environment and Sustainability, European Commission Joint Research Centre, 21027 Ispra, Italy; and [i]Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, CO 80309

Edited by Mark H. Thiemens, University of California, San Diego, La Jolla, CA, and approved October 18, 2013 (received for review August 5, 2013)

This study quantitatively estimates the spatial distribution of anthropogenic methane sources in the United States by combining comprehensive atmospheric methane observations, extensive spatial datasets, and a high-resolution atmospheric transport model. Results show that current inventories from the US Environmental Protection Agency (EPA) and the Emissions Database for Global Atmospheric Research underestimate methane emissions nationally by a factor of ~1.5 and ~1.7, respectively. Our study indicates that emissions due to ruminants and manure are up to twice the magnitude of existing inventories. In addition, the discrepancy in methane source estimates is particularly pronounced in the south-central United States, where we find total emissions are ~2.7 times greater than in most inventories and account for 24 ± 3% of national emissions. The spatial patterns of our emission fluxes and observed methane–propane correlations indicate that fossil fuel extraction and refining are major contributors (45 ± 13%) in the south-central United States. This result suggests that regional methane emissions due to fossil fuel extraction and processing could be 4.9 ± 2.6 times larger than in EDGAR, the most comprehensive global methane inventory. These results cast doubt on the US EPA's recent decision to downscale its estimate of national natural gas emissions by 25–30%. Overall, we conclude that methane emissions associated with both the animal husbandry and fossil fuel industries have larger greenhouse gas impacts than indicated by existing inventories.

climate change policy | geostatistical inverse modeling

Methane (CH_4) is the second most important anthropogenic greenhouse gas, with approximately one third the total radiative forcing of carbon dioxide (1). CH_4 also enhances the formation of surface ozone in populated areas, and thus higher global concentrations of CH_4 may significantly increase ground-level ozone in the Northern Hemisphere (2). Furthermore, methane affects the ability of the atmosphere to oxidize other pollutants and plays a role in water formation within the stratosphere (3).

Atmospheric concentrations of CH_4 [~1,800 parts per billion (ppb)] are currently much higher than preindustrial levels (~680–715 ppb) (1, 4). The global atmospheric burden started to rise rapidly in the 18th century and paused in the 1990s. Methane levels began to increase again more recently, potentially from a combination of increased anthropogenic and/or tropical wetland emissions (5–7). Debate continues, however, over the causes behind these recent trends (7, 8).

Anthropogenic emissions account for 50–65% of the global CH_4 budget of ~395–427 teragrams of carbon per year ($TgC \cdot y^{-1}$) (526–569 Tg CH_4) (7, 9). The US Environmental Protection Agency (EPA) estimates the principal anthropogenic sources in the United States to be (in order of importance) (i) livestock (enteric fermentation and manure management), (ii) natural gas production and distribution, (iii) landfills, and (iv) coal mining (10). EPA assesses human-associated emissions in the United States in 2008 at 22.1 TgC, roughly 5% of global emissions (10).

The amount of anthropogenic CH_4 emissions in the US and attributions by sector and region are controversial (Fig. 1). Bottom-up inventories from US EPA and the Emissions Database for Global Atmospheric Research (EDGAR) give totals ranging from 19.6 to 30 $TgC \cdot y^{-1}$ (10, 11). The most recent EPA and EDGAR inventories report lower US anthropogenic emissions compared with previous versions (decreased by 10% and 35%, respectively) (10, 12); this change primarily reflects lower, revised emissions estimates from natural gas and coal production Fig. S1. However, recent analysis of CH_4 data from aircraft estimates a higher budget of 32.4 ± 4.5 $TgC \cdot y^{-1}$ for 2004 (13). Furthermore, atmospheric observations indicate higher emissions in natural gas production areas (14–16); a steady 20-y increase in the number of US wells and newly-adopted horizontal drilling techniques may have further increased emissions in these regions (17, 18).

These disparities among bottom-up and top-down studies suggest much greater uncertainty in emissions than typically reported. For example, EPA cites an uncertainty of only ±13% for the for United States (10). Independent assessments of bottom-up inventories give error ranges of 50–100% (19, 20), and

Significance

Successful regulation of greenhouse gas emissions requires knowledge of current methane emission sources. Existing state regulations in California and Massachusetts require ~15% greenhouse gas emissions reductions from current levels by 2020. However, government estimates for total US methane emissions may be biased by 50%, and estimates of individual source sectors are even more uncertain. This study uses atmospheric methane observations to reduce this level of uncertainty. We find greenhouse gas emissions from agriculture and fossil fuel extraction and processing (i.e., oil and/or natural gas) are likely a factor of two or greater than cited in existing studies. Effective national and state greenhouse gas reduction strategies may be difficult to develop without appropriate estimates of methane emissions from these source sectors.

Author contributions: S.M.M., S.C.W., and A.M.M. designed research; S.M.M., A.E.A., S.C.B., E.J.D., J.E., M.L.F., G.J.-M., B.R.M., J.B.M., S.A.M., T.N., and C.S. performed research; S.M.M. analyzed data; S.M.M., S.C.W., A.M.M., and E.A.K. wrote the paper; A.E.A., S.C.B., E.J.D., M.L.F., B.R.M., J.B.M., S.A.M., and C.S. collected atmospheric methane data; and J.E. and T.N. developed meteorological simulations using the Weather Research and Forecasting model.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

[1]To whom correspondence should be addressed. E-mail: scot.m.miller@gmail.com.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1314392110/-/DCSupplemental.

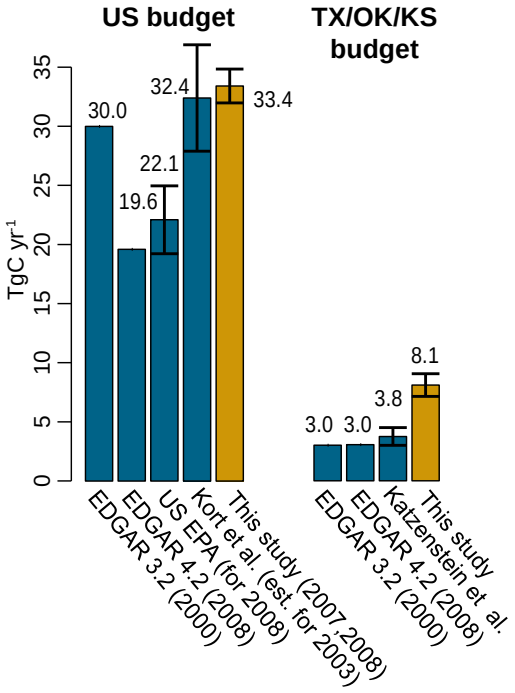


Fig. 1. US anthropogenic methane budgets from this study, from previous top-down estimates, and from existing emissions inventories. The south-central United States includes Texas, Oklahoma, and Kansas. US EPA estimates only national, not regional, emissions budgets. Furthermore, national budget estimates from EDGAR, EPA, and Kort et al. (13) include Alaska and Hawaii whereas this study does not.

values from Kort et al. are 47 ± 20% higher than EPA (13). Assessments of CH_4 sources to inform policy (e.g., regulating emissions or managing energy resources) require more accurate, verified estimates for the United States.

This study estimates anthropogenic CH_4 emissions over the United States for 2007 and 2008 using comprehensive CH_4 observations at the surface, on telecommunications towers, and from aircraft, combined with an atmospheric transport model and a geostatistical inverse modeling (GIM) framework. We use auxiliary spatial data (e.g., on population density and economic activity) and leverage concurrent measurements of alkanes to help attribute emissions to specific economic sectors. The work provides spatially resolved CH_4 emissions estimates and associated uncertainties, as well as information by source sector, both previously unavailable.

Model and Observation Framework

We use the Stochastic Time-Inverted Lagrangian Transport model (STILT) to calculate the transport of CH_4 from emission points at the ground to measurement locations in the atmosphere (21). STILT follows an ensemble of particles backward in time, starting from each observation site, using wind fields and turbulence modeled by the Weather Research and Forecasting (WRF) model (22). STILT derives an influence function ("footprint," units: ppb CH_4 per unit emission flux) linking upwind emissions to each measurement. Inputs of CH_4 from surface sources along the ensemble of back-trajectories are averaged to compute the CH_4 concentration for comparison with each observation.

We use observations for 2007 and 2008 from diverse locations and measurement platforms. The principal observations derive from daily flask samples on tall towers (4,984 total observations) and vertical profiles from aircraft (7,710 observations). Tower-based observations are collected as part of the National Oceanic and Atmospheric (NOAA)/Department of Energy (DOE) cooperative air sampling network, and aircraft-based data are obtained from regular NOAA flights (23), regular DOE flights (24), and from the Stratosphere-Troposphere Analyses of Regional Transport 2008 (START08) aircraft campaign (25); all data are publicly available from NOAA and DOE. These observations are displayed in Fig. 2 and discussed further in the SI Text (e.g., Fig. S2). We use a GIM framework (26, 27) to analyze the footprints for each of the 12,694 observations, and these footprints vary by site and with wind conditions. In aggregate, the footprints provide spatially resolved coverage of most of the continental United States, except the southeast coastal region (Fig. S3).

The GIM framework, using footprints and concentration measurements, optimizes CH_4 sources separately for each month of 2007 and 2008 on a 1° × 1° latitude–longitude grid for the United States. The contributions of fluxes from natural wetlands are modeled first and subtracted from the observed CH_4 (2.0 $TgC \cdot y^{-1}$ for the continental United States); these fluxes are much smaller than anthropogenic sources in the United States and thus would be difficult to independently constrain from atmospheric data (SI Text).

The GIM framework represents the flux distribution for each month using a deterministic spatial model plus a stochastic spatially correlated residual, both estimated from the atmospheric observations. The deterministic component is given by a weighted linear combination of spatial activity data from the EDGAR 4.2 inventory; these datasets include any economic or demographic data that may predict the distribution of CH_4 emissions (e.g., gas production, human and ruminant population densities, etc.). Both the selection of the activity datasets to be retained in the model and the associated weights (emission factors) are optimized to best match observed CH_4 concentrations. Initially, seven activity datasets are included from EDGAR 4.2, (i) population, (ii) electricity production from power plants, (iii) ruminant population count, (iv) oil and conventional gas production, (v) oil refinery production, (vi) rice production, and (vii) coal production.

We select the minimum number of datasets with the greatest predictive ability using the Bayesian Information Criterion (BIC) (SI Text) (28). BIC numerically scores all combinations of available datasets based on how well they improve goodness of fit and applies a penalty that increases with the number of datasets retained.

The stochastic component represents sources that do not fit the spatial patterns of the activity data (Fig. S4). GIM uses

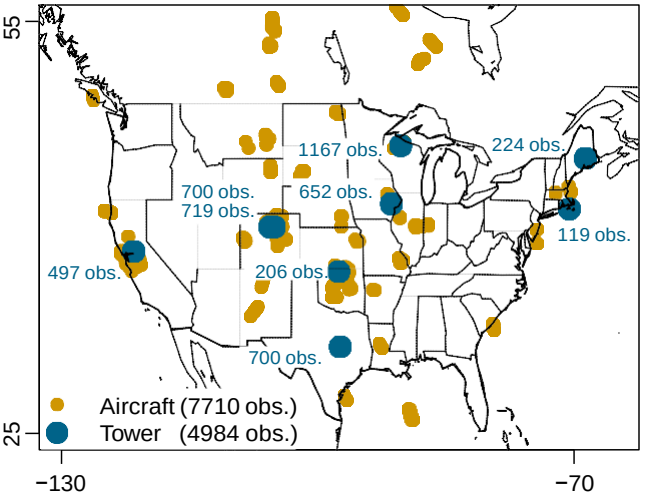


Fig. 2. CH_4 concentration measurements from 2007 and 2008 and the number of observations associated with each measurement type. Blue text lists the number of observations associated with each stationary tower measurement site.

a covariance function to describe the spatial and temporal correlation of the stochastic component and optimizes its spatial and temporal distribution simultaneously with the optimization of the activity datasets in the deterministic component (SI Text, Fig. S5) (26–28). Because of the stochastic component, the final emissions estimate can have a different spatial and temporal distribution from any combination of the activity data.

If the observation network is sensitive to a broad array of different source sectors and/or if the spatial activity maps are effective at explaining those sources, many activity datasets will be included in the deterministic model. If the deterministic model explains the observations well, the magnitude of CH_4 emissions in the stochastic component will be small, the assignment to specific sectors will be unambiguous, and uncertainties in the emissions estimates will be small. This result is not the case here, as discussed below (see Results).

A number of previous studies used top-down methods to constrain anthropogenic CH_4 sources from global (29–33) to regional (13–15, 34–38) scales over North America. Most regional studies adopted one of three approaches: use a simple box model to estimate an overall CH_4 budget (14), estimate a budget using the relative ratios of different gases (15, 37–39), or estimate scaling factors for inventories by region or source type (13, 34–36). The first two methods do not usually give explicit information about geographic distribution. The last approach provides information about the geographic distribution of sources, but results hinge on the spatial accuracy of the underlying regional or sectoral emissions inventories (40).

Here, we are able to provide more insight into the spatial distribution of emissions; like the scaling factor method above, we leverage spatial information about source sectors from an existing inventory, but in addition we estimate the distribution of emissions where the inventory is deficient. We further bolster attribution of regional emissions from the energy industry using the observed correlation of CH_4 and propane, a gas not produced by biogenic processes like livestock and landfills.

Results

Spatial Distribution of CH_4 Emissions. Fig. 3 displays the result of the 2-y mean of the monthly CH_4 inversions and differences from the EDGAR 4.2 inventory. We find emissions for the United States that are a factor of 1.7 larger than the EDGAR inventory. The optimized emissions estimated by this study bring the model closer in line with the observations (Fig. 4, Figs. S6 and S7). Posterior emissions fit the CH_4 observations [R^2 = 0:64, root mean square error (RMSE) = 31 ppb] much better than EDGAR v4.2 (R^2 = 0:23, RMSE = 49 ppb). Evidently, the spatial distribution of EDGAR sources is inconsistent with emissions patterns implied by the CH_4 measurements and associated footprints.

Several diagnostic measures preclude the possibility of major systematic errors in WRF–STILT. First, excellent agreement between the model and measured vertical profiles from aircraft implies little bias in modeled vertical air mixing (e.g., boundary-layer heights) (Fig. 4). Second, the monthly posterior emissions estimated by the inversion lack statistically significant seasonality (Fig. S8). This result implies that seasonally varying weather patterns do not produce detectable biases in WRF–STILT. SI Text discusses possible model errors and biases in greater detail.

CH_4 observations are sparse over parts of the southern and central East Coast and in the Pacific Northwest. Emissions estimates for these regions therefore rely more strongly on the deterministic component of the flux model, with weights constrained primarily by observations elsewhere. Therefore, emissions in these areas, including from coal mining, are poorly constrained (SI Text).

Contribution of Different Source Sectors. Only two spatial activity datasets from EDGAR 4.2 are selected through the BIC as meaningful predictors of CH_4 observations over the United States: population densities of humans and of ruminants (Table S1). Some sectors are eliminated by the BIC because emissions are situated far from observation sites (e.g., coal mining in West Virginia or Pennsylvania), making available CH_4 data insensitive to these predictors. Other sectors may strongly affect observed concentrations but are not selected, indicating that the spatial datasets from EDGAR are poor predictors for the distribution of observed concentrations (e.g., oil and natural gas extraction and oil refining). Sources from these sectors appear in the stochastic component of the GIM (SI Text).

The results imply that existing inventories underestimate emissions from two key sectors: ruminants and fossil fuel extraction and/or processing, discussed in the remainder of this section.

We use the optimized ruminant activity dataset to estimate the magnitude of emissions with spatial patterns similar to animal husbandry and manure. Our corresponding US budget of 12.7 ± 5.0 $TgC \cdot y^{-1}$ is nearly twice that of EDGAR and EPA (6.7 and 7.0, respectively). The total posterior emissions estimate over the northern plains, a region with high ruminant density but little fossil fuel extraction, further supports the ruminant estimate (Nebraska, Iowa, Wisconsin, Minnesota, and South Dakota). Our total budget for this region of 3.4 ± 0.7 compares with 1.5 $TgC \cdot y^{-1}$ in EDGAR. Ruminants and agriculture may also be

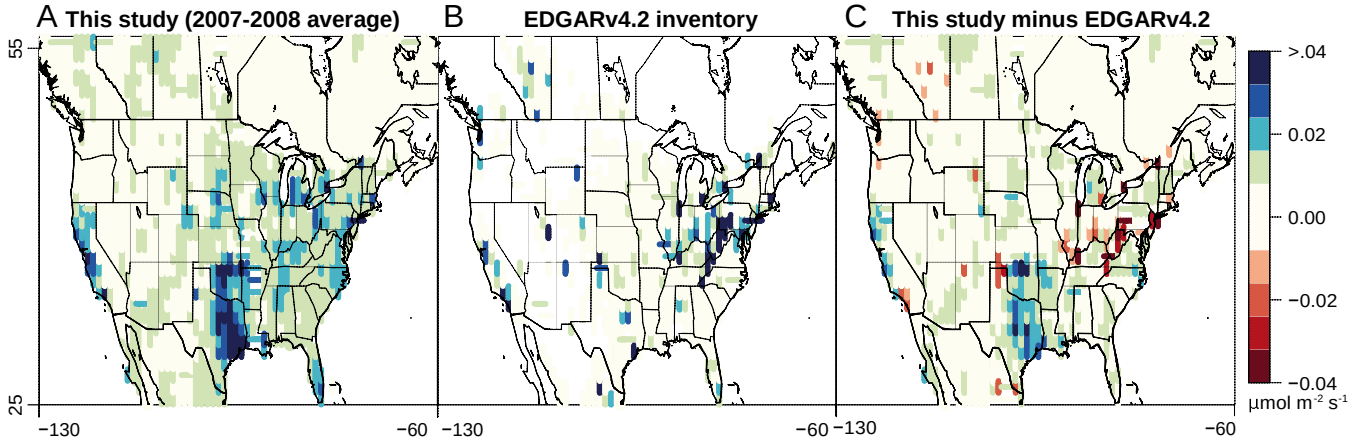


Fig. 3. The 2-y averaged CH_4 emissions estimated in this study (A) compared against the commonly used EDGAR 4.2 inventory (B and C). Emissions estimated in this study are greater than in EDGAR 4.2, especially near Texas and California.

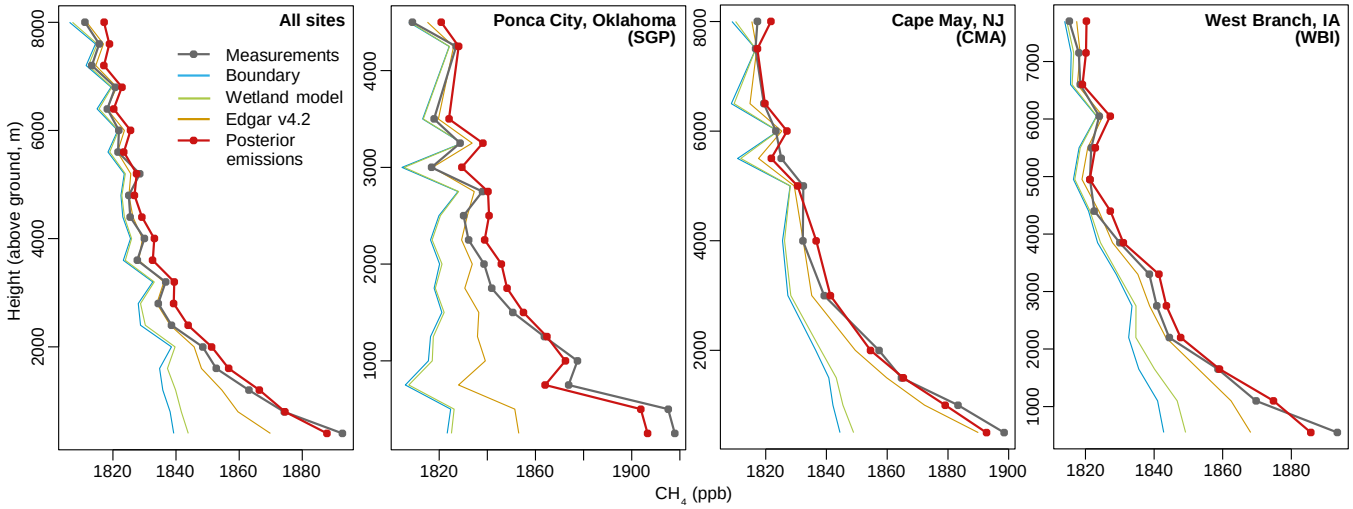


Fig. 4. A model–measurement comparison at several regular NOAA/DOE aircraft monitoring sites (averaged over 2007–2008). Plots include the measurements; the modeled boundary condition; the summed boundary condition and wetland contribution (from the Kaplan model); and the summed boundary, wetland, and anthropogenic contributions (from EDGAR v4.2 and the posterior emissions estimate).

partially responsible for high emissions over California (41). EDGAR activity datasets are poor over California (42), but several recent studies (34, 36–38, 41) have provided detailed top-down emissions estimates for the state using datasets from state agencies.

Existing inventories also greatly underestimate CH_4 sources from the south-central United States (Fig. 3). We find the total CH_4 source from Texas, Oklahoma, and Kansas to be 8.1 ± 0.96 $TgC \cdot y^{-1}$, a factor of 2.7 higher than the EDGAR inventory. These three states alone constitute ~24 ± 3% of the total US anthropogenic CH_4 budget or 3.7% of net US greenhouse gas emissions [in CO_2 equivalents (10)].

Texas and Oklahoma were among the top five natural gas producing states in the country in 2007 (18), and aircraft observations of alkanes indicate that the natural gas and/or oil industries play a significant role in regional CH_4 emissions. Concentrations of propane (C_3H_8), a tracer of fossil hydrocarbons (43), are strongly correlated with CH_4 at NOAA/DOE aircraft monitoring locations over Texas and Oklahoma (R^2 = 0:72) (Fig. 5). Correlations are much weaker at other locations in North America (R^2 = 0:11 to 0.64).

We can obtain an approximate CH_4 budget for fossil-fuel extraction in the region by subtracting the optimized contributions associated with ruminants and population from the total emissions. The residual (Fig. S4C) represents sources that have spatial patterns not correlated with either human or ruminant density in EDGAR. Our budget sums to 3.7 ± 2.0 $TgC \cdot y^{-1}$, a factor of 4.9 ± 2.6 larger than oil and gas emissions in EDGAR v4.2 (0.75 $TgC \cdot y^{-1}$) and a factor of 6.7 ± 3.6 greater than EDGAR sources from solid waste facilities (0.55 $TgC \cdot y^{-1}$), the two major sources that may not be accounted for in the deterministic component. The population component likely captures a portion of the solid waste sources so this residual methane budget more likely represents natural gas and oil emissions than landfills. SI Text discusses in detail the uncertainties in this sector-based emissions estimate. We currently do not have the detailed, accurate, and spatially resolved activity data (fossil fuel extraction and processing, ruminants, solid waste) that would provide more accurate sectorial attribution.

Katzenstein et al. (2003) (14) were the first to report large regional emissions of CH_4 from Texas, Oklahoma, and Kansas; they cover an earlier time period (1999–2002) than this study. They used a box model and 261 near-ground CH_4 measurements taken over 6 d to estimate a total Texas–Oklahoma–Kansas CH_4 budget (from all sectors) of 3.8 ± 0.75 $TgC \cdot y^{-1}$. We revise their

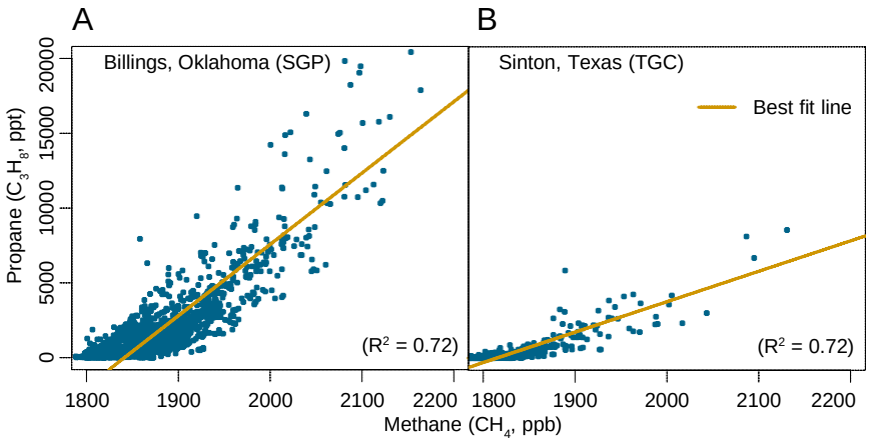


Fig. 5. Correlations between propane and CH_4 at NOAA/DOE aircraft observation sites in Oklahoma (A) and Texas (B) over 2007–2012. Correlations are higher in these locations than at any other North American sites, indicating large contributions of fossil fuel extraction and processing to CH_4 emitted in this region.

estimate upward by a factor of two based on the inverse model and many more measurements from different platforms over two full years of data. SI Text further compares the CH_4 estimate in Katzenstein et al. and in this study.

Discussion and Summary

This study combines comprehensive atmospheric data, diverse datasets from the EDGAR inventory, and an inverse modeling framework to derive spatially resolved CH_4 emissions and information on key source sectors. We estimate a mean annual US anthropogenic CH_4 budget for 2007 and 2008 of 33.4 ± 1.4 $TgC \cdot y^{-1}$ or ~7–8% of the total global CH_4 source. This estimate is a factor of 1.5 and 1.7 larger than EPA and EDGAR v4.2, respectively. CH_4 emissions from Texas, Oklahoma, and Kansas alone account for 24% of US methane emissions, or 3.7% of the total US greenhouse gas budget.

The results indicate that drilling, processing, and refining activities over the south-central United States have emissions as much as 4.9 ± 2.6 times larger than EDGAR, and livestock operations across the US have emissions approximately twice that of recent inventories. The US EPA recently decreased its CH_4 emission factors for fossil fuel extraction and processing by 25–30% (for 1990–2011) (10), but we find that CH_4 data from across North America instead indicate the need for a larger adjustment of the opposite sign.

ACKNOWLEDGMENTS. For advice and support, we thank Roisin Commane, Elaine Gottlieb, and Matthew Hayek (Harvard University); Robert Harriss (Environmental Defense Fund); Hanqin Tian and Bowen Zhang (Auburn University); Jed Kaplan (Ecole Polytechnique Fédérale de Lausanne); Kimberly Mueller and Christopher Weber (Institute for Defense Analyses Science and Technology Policy Institute); Nadia Oussayef; and Gregory Berger. In addition, we thank the National Aeronautics and Space Administration (NASA) Advanced Supercomputing Division for computing help; P. Lang, K. Sours, and C. Siso for analysis of National Oceanic and Atmospheric Administration (NOAA) flasks; and B. Hall for calibration standards work. This work was supported by the American Meteorological Society Graduate Student Fellowship/Department of Energy (DOE) Atmospheric Radiation Measurement Program, a DOE Computational Science Graduate Fellowship, and the National Science Foundation Graduate Research Fellowship Program. NOAA measurements were funded in part by the Atmospheric Composition and Climate Program and the Carbon Cycle Program of NOAA's Climate Program Office. Support for this research was provided by NASA Grants NNX08AR47G and NNX11AG47G, NOAA Grants NA09OAR4310122 and NA11OAR4310158, National Science Foundaton (NSF) Grant ATM-0628575, and Environmental Defense Fund Grant 0146-10100 (to Harvard University). Measurements at Walnut Grove were supported in part by a California Energy Commission Public Interest Environmental Research Program grant to Lawrence Berkeley National Laboratory through the US Department of Energy under Contract DE-AC02-05CH11231. DOE flights were supported by the Office of Biological and Environmental Research of the US Department of Energy under Contract DE-AC02-05CH11231 as part of the Atmospheric Radiation Measurement Program (ARM), ARM Aerial Facility, and Terrestrial Ecosystem Science Program. Weather Research and Forecasting–Stochastic Time-Inverted Lagrangian Transport model development at Atmospheric and Environmental Research has been funded by NSF Grant ATM-0836153, NASA, NOAA, and the US intelligence community.

1. Butler J (2012) The NOAA annual greenhouse gas index (AGGI). Available at http://www.esrl.noaa.gov/gmd/aggi/. Accessed November 4, 2013.
2. Fiore AM, et al. (2002) Linking ozone pollution and climate change: The case for controlling methane. Geophys Res Lett 29:1919.
3. Jacob D (1999) Introduction to Atmospheric Chemistry (Princeton Univ Press, Princeton).
4. Mitchell LE, Brook EJ, Sowers T, McConnell JR, Taylor K (2011) Multidecadal variability of atmospheric methane, 1000-1800 CE. J Geophys Res Biogeosci 116:G02007.
5. Dlugokencky EJ, et al. (2009) Observational constraints on recent increases in the atmospheric CH_4 burden. Geophys Res Lett 36:L18803.
6. Sussmann R, Forster F, Rettinger M, Bousquet P (2012) Renewed methane increase for five years (2007-2011) observed by solar FTIR spectrometry. Atmos Chem Phys 12: 4885–4891.
7. Kirschke S, et al. (2013) Three decades of global methane sources and sinks. Nat Geosci 6:813–823.
8. Wang JS, et al. (2004) A 3-D model analysis of the slowdown and interannual variability in the methane growth rate from 1988 to 1997. Global Biogeochem Cycles 18: GB3011.
9. Ciais P, et al. (2013) Carbon and Other Biogeochemical Cycles: Final Draft Underlying Scientific Technical Assessment (IPCC Secretariat, Geneva).
10. US Environmental Protection Agency (2013) Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2011, Technical Report EPA 430-R-13-001 (Environmental Protection Agency, Washington).
11. Olivier JGJ, Peters J (2005) CO_2 from non-energy use of fuels: A global, regional and national perspective based on the IPCC Tier 1 approach. Resour Conserv Recycling 45:210–225.
12. European Commission Joint Research Centre, Netherlands Environmental Assessment Agency (2010) Emission Database for Global Atmospheric Research (EDGAR), Release Version 4.2. Available at http://edgar.jrc.ec.europa.eu. Accessed November 4, 2013.
13. Kort EA, et al. (2008) Emissions of CH_4 and N_2O over the United States and Canada based on a receptor-oriented modeling framework and COBRA-NA atmospheric observations. Geophys Res Lett 35:L18808.
14. Katzenstein AS, Doezema LA, Simpson IJ, Blake DR, Rowland FS (2003) Extensive regional atmospheric hydrocarbon pollution in the southwestern United States. Proc Natl Acad Sci USA 100(21):11975–11979.
15. Pétron G, et al. (2012) Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study. J Geophys Res Atmos 117:D04304.
16. Karion A, et al. (2013) Methane emissions estimate from airborne measurements over a western United States natural gas field. Geophys Res Lett 40:4393–4397.
17. Howarth RW, Santoro R, Ingraffea A (2011) Methane and the greenhouse-gas footprint of natural gas from shale formations. Clim Change 106:679–690.
18. US Energy Information Administration (2013) Natural Gas Annual 2011, Technical report (US Department of Energy, Washington).
19. National Research Council (2010) Verifying Greenhouse Gas Emissions: Methods to Support International Climate Agreements (National Academies Press, Washington).
20. Dlugokencky EJ, Nisbet EG, Fisher R, Lowry D (2011) Global atmospheric methane: Budget, changes and dangers. Philos Trans A Math Phys Eng Sci 369(1943): 2058–2072.
21. Lin JC, et al. (2003) A near-field tool for simulating the upstream influence of atmospheric observations: The Stochastic Time-Inverted Lagrangian Transport (STILT) model. J Geophys Res Atmos 108(D16):4493.
22. Nehrkorn T, et al. (2010) Coupled Weather Research and Forecasting-Stochastic Time-Inverted Lagrangian Transport (WRF-STILT) model. Meteorol Atmos Phys 107:51–64.
23. NOAA ESRL (2013) Carbon Cycle Greenhouse Gas Group Aircraft Program. Available at http://www.esrl.noaa.gov/gmd/ccgg/aircraft/index.html. Accessed November 4, 2013.
24. Biraud SC, et al. (2013) A multi-year record of airborne CO_2 observations in the US southern great plains. Atmos Meas Tech 6:751–763.
25. Pan LL, et al. (2010) The Stratosphere-Troposphere Analyses of Regional Transport 2008 Experiment. Bull Am Meteorol Soc 91:327–342.
26. Kitanidis PK, Vomvoris EG (1983) A geostatistical approach to the inverse problem in groundwater modeling (steady state) and one-dimensional simulations. Water Resour Res 19:677–690.
27. Michalak A, Bruhwiler L, Tans P (2004) A geostatistical approach to surface flux estimation of atmospheric trace gases. J Geophys Res Atmos 109(D14):D14109.
28. Gourdji SM, et al. (2012) North American CO_2 exchange: Inter-comparison of modeled estimates with results from a fine-scale atmospheric inversion. Biogeosciences 9: 457–475.
29. Chen YH, Prinn RG (2006) Estimation of atmospheric methane emissions between 1996 and 2001 using a three-dimensional global chemical transport model. J Geophys Res Atmos 111(D10):D10307.
30. Meirink JF, et al. (2008) Four-dimensional variational data assimilation for inverse modeling of atmospheric methane emissions: Analysis of SCIAMACHY observations. J Geophys Res Atmos 113(D17):D17301.
31. Bergamaschi P, et al. (2009) Inverse modeling of global and regional CH4 emissions using SCIAMACHY satellite retrievals. J Geophys Res Atmos 114(D22):D22301.
32. Bousquet P, et al. (2011) Source attribution of the changes in atmospheric methane for 2006-2008. Atmos Chem Phys 11:3689–3700.
33. Monteil G, et al. (2011) Interpreting methane variations in the past two decades using measurements of CH_4 mixing ratio and isotopic composition. Atmos Chem Phys 11: 9141–9153.
34. Zhao C, et al. (2009) Atmospheric inverse estimates of methane emissions from central California. J Geophys Res Atmos 114(D16):D16302.
35. Kort EA, et al. (2010) Atmospheric constraints on 2004 emissions of methane and nitrous oxide in North America from atmospheric measurements and receptor-oriented modeling framework. J Integr Environ Sci 7:125–133.
36. Jeong S, et al. (2012) Seasonal variation of CH_4 emissions from central California. J Geophys Res 117:D11306.
37. Peischl J, et al. (2012) Airborne observations of methane emissions from rice cultivation in the Sacramento Valley of California. J Geophys Res Atmos 117(D24):D00V25.
38. Wennberg PO, et al. (2012) On the sources of methane to the Los Angeles atmosphere. Environ Sci Technol 46(17):9282–9289.
39. Miller JB, et al. (2012) Linking emissions of fossil fuel CO2 and other anthropogenic trace gases using atmospheric 14CO2. J Geophys Res Atmos 117(D8):D08302.
40. Law RM, Rayner PJ, Steele LP, Enting IG (2002) Using high temporal frequency data for CO_2 inversions. Global Biogeochem Cycles 16(4):1053.
41. Jeong S, et al. (2013) A multitower measurement network estimate of California's methane emissions. J Geophys Res Atmos, 10.1002/jgrd.50854.
42. Xiang B, et al. (2013) Nitrous oxide (N2O) emissions from California based on 2010 CalNex airborne measurements. J Geophys Res Atmos 118(7):2809–2820.
43. Koppmann R (2008) Volatile Organic Compounds in the Atmosphere (Wiley, Singapore).