



RESEARCH ARTICLE

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Key Points:

- Unintended (fugitive) methane emissions are ubiquitous in urban systems, and come from biogenic as well as natural gas sources
- Methane emission sources and their magnitudes vary greatly among cities
- Urban methane mitigation will require new observations with a suite of techniques, and new institutional partnerships

Supporting Information:

- Appendix S1
- Movie S1

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Mitigation of methane emissions in cities: How new measurements and partnerships can contribute to emissions reduction strategies

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Abstract Cities generate 70% of anthropogenic greenhouse gas emissions, a fraction that is growing with global urbanization. While cities play an important role in climate change mitigation, there has been little focus on reducing urban methane (CH₄) emissions. Here, we develop a conceptual framework for CH₄ mitigation in cities by describing emission processes, the role of measurements, and a need for new institutional partnerships. Urban CH₄ emissions are likely to grow with expanding use of natural gas and organic waste disposal systems in growing population centers; however, we currently lack the ability to quantify this increase. We also lack systematic knowledge of the relative contribution of these distinct source sectors on emissions. We present new observations from four North American cities to demonstrate that CH₄ emissions vary in magnitude and sector from city to city and hence require different mitigation strategies. Detections of fugitive emissions from these systems suggest that current mitigation approaches are absent or ineffective. These findings illustrate that tackling urban CH₄ emissions will require research efforts to identify mitigation targets, develop and implement new mitigation strategies, and monitor atmospheric CH₄ levels to ensure the success of mitigation efforts. This research will require a variety of techniques to achieve these objectives and should be deployed in cities globally. We suggest that metropolitan scale partnerships may effectively coordinate systematic measurements and actions focused on emission reduction goals.

1. Introduction

Cities are a major source of greenhouse gas emissions globally and are critical players in the response to climate change. Urban areas constitute emission hotspots—important targets for greenhouse gas emissions mitigation [Duren and Miller, 2012]. City governments are uniquely poised to address climate change by controlling their own emissions through management and jurisdictional control over municipal and other local emissions sources [Gurney *et al.*, 2015]. Cities face fewer political difficulties with climate change mitigation than do nation-states [Rosenzweig *et al.*, 2010] and are already taking organized action at the global scale through associations such as the C40 Cities, which comprises 63 cities and >8% of the world's population [Arup, 2014], and Local Governments for Sustainability [ICLEI USA, n.d.], which counts 1000 participating cities. Urban emissions will grow in importance along with the world's urban population, which is forecast to double over the next 40 years to encompass the vast majority of the world's population [United Nations Department of Economic and Social Affairs/Population Division, 2012].

The growing scale of urban areas globally with respect to population and land area, and the willingness of metropolitan regions to sign on to organized climate action efforts suggest a great urgency for city-scale mitigation measures. One strategy that has been proposed recently for near-term climate mitigation is control of noncarbon dioxide (CO₂), short-lived climate pollutants with large radiative forcing such as methane (CH₄) [Montzka *et al.*, 2011]. CH₄ differs from CO₂ in that mitigation is technologically and economically feasible [Shindell *et al.*, 2012]. Unlike CO₂, a large fraction of CH₄ is lost as fugitive emissions from engineered systems, such as leaks from natural gas pipelines. These unintended fugitive emissions also represent a

unique challenge for CH₄ that is different from activity-driven CO₂ emissions but also pose an opportunity to reduce loss of an economic commodity. In addition, mitigation techniques for CH₄ process emissions are well known and in wide use, such as flaring emissions from fossil fuel extraction or gas extraction from landfills. CH₄ mitigation in cities has substantial cobenefits for public health and safety by improving air quality in cities through global reductions in ozone pollution [West *et al.*, 2006; Fiore *et al.*, 2008]. CH₄ is also a safety hazard, already monitored by cities because it poses an explosion risk [e.g., Building and Safety Division, Dept. of Public Works, County of Los Angeles, 2002]. Fatal pipeline explosions in New York and San Bruno have drawn recent attention to the issue [West, 2014]. An unprecedented natural gas leak from an underground storage facility near Porter Ranch, California temporarily displaced thousands of residents and doubled CH₄ emissions from the Los Angeles Basin [Conley *et al.*, 2016; Sahagun, 2016]. Under orders from California's governor, the responsible parties are required to mitigate an equivalent amount of CH₄ elsewhere to offset the leak [Barboza, 2016].

Despite the existence of strategies to reduce CH₄ emissions that are particularly appropriate for cities [Global Methane Initiative, ; Executive Office of the President of the U.S., 2014], there are limited mitigation approaches currently in practice [Tang *et al.*, 2010]. Indeed, some strategies to reduce CO₂ emissions, such as substituting natural gas for other fossil fuels such as coal and diesel, may have the unintended consequence of increasing radiative forcing by increasing fugitive CH₄ emissions [Alvarez *et al.*, 2012]. There are substantial barriers to be overcome before urban CH₄ mitigation can be implemented successfully, starting with large uncertainties in urban CH₄ budgets [e.g., Hsu *et al.*, 2009; Wunch *et al.*, 2009]. CH₄ inventories tend to underestimate emissions compared to emissions inferred from atmospheric measurements due to the difficulty of correctly accounting for fugitive CH₄ [Brandt *et al.*, 2014]. In addition, we lack a basic understanding of the locations and temporal patterns of urban CH₄ sources at relevant scales as well as information about the cost of repairs [Forman, 2014], and cooperation needed between diverse stakeholders to carry out mitigation activities.

We suggest that these barriers can be overcome by the cooperation of key stakeholders in individual cities to develop a scientific basis for CH₄ mitigation and a process by which CH₄ reductions can be evaluated and accounted. Here, we synthesize results from several recent studies and present new observations to create a new conceptual framework for effective city-wide CH₄ mitigation policy. We begin by reviewing the state of knowledge and emerging trends in important urban CH₄ emission sectors. We then present new data from four western U.S. cities to illustrate how CH₄ emissions vary among different cities. We describe the measurement goals and multiple approaches needed to inform mitigation. Finally, we suggest a role for new city-wide partnerships and adaptive management strategies tailored specifically for each unique metropolitan area to efficiently carry out mitigation policies.

2. Anthropogenic Urban Methane Sources

According to global inventories of anthropogenic CH₄ sources, the most important sectors for urban CH₄ emissions are energy, waste, agriculture, and transportation [Table 1; Marcotullio *et al.*, 2013]. Energy and transportation primarily emit fossil CH₄ derived from natural gas, whereas waste treatment and agriculture produce biogenic CH₄ from the process of anaerobic decomposition (Figure 1). Fossil sources produce CH₄ as a result of combustion or as fugitive emissions of natural gas from natural gas distribution networks or combustion units. Biogenic CH₄ is primarily produced from anaerobic decomposition but can also be unintentionally released as fugitive emissions from engineered systems designed to handle biogenic CH₄. Biogenic CH₄ is also produced (and consumed) by soils and from agricultural sources but will not be discussed further in this study. Observations suggest that the magnitude of CH₄ flux from urban soils is reduced relative to soils under native vegetation and is likely to be several orders of magnitude smaller than citywide areal fluxes [i.e., <0.9 nmol m⁻² s⁻¹; Kaye *et al.*, 2004; Groffman and Pouyat, 2009]. Given these low rates, biogenic soil fluxes will not be further discussed.

A large fraction of natural gas consumption and waste treatment, along with their respective CH₄ emissions, are concentrated in cities. CH₄ emissions from both sectors are likely to increase—natural gas is currently promoted as a clean burning fuel for electricity generation and vehicles, and waste CH₄ emissions are growing along with the urban population. In addition, biogenic CH₄ is increasingly considered a source of renewable energy, resulting in increased production and utilization of biogas [Global Methane

Table 1. Inventory Estimates of Global and Urban Methane (CH₄) Emissions by Sector for Year 2000, From Marcotullio *et al.* [2013]

| Sector | Global Emissions ^a | Urban Emissions ^a | Urban CH ₄ as Percent of Sectoral CH ₄ | Urban CH ₄ as Percent of Total CH ₄ |
|----------------|-------------------------------|------------------------------|--|---|
| Agriculture | 168 | 9 | 5 | 3 |
| Energy | 74 | 31 | 42 | 10 |
| Waste | 69 | 27 | 40 | 9 |
| Transportation | 1 | <1 | 43 | <1 |
| Total | 312 | 67 | | 21 |

^aEmissions are given as Tg CH₄-y⁻¹.

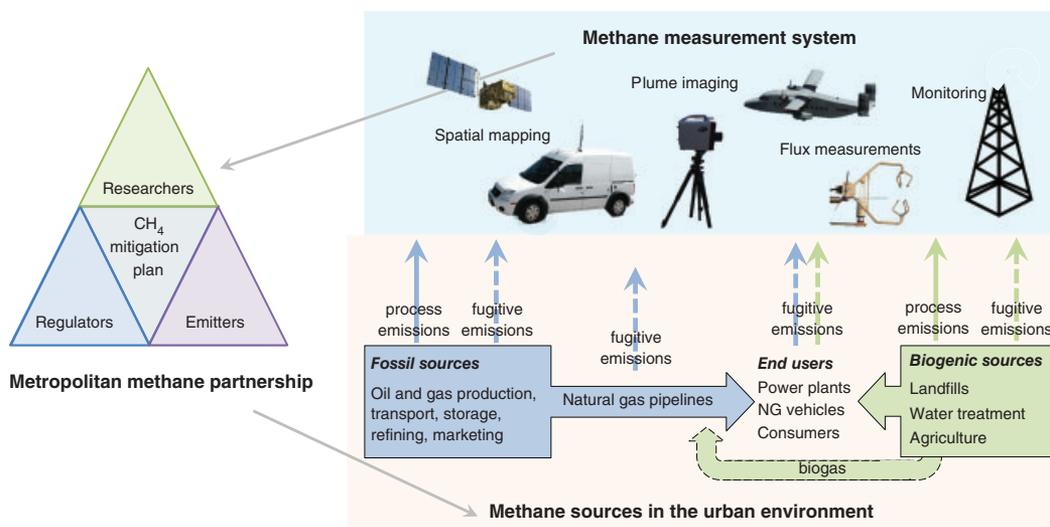


Figure 1. Conceptual framework for urban methane (CH₄) mitigation. CH₄ emissions in the urban environment originate directly from either fossil or biogenic sources or escape unintentionally from engineered systems and end users as fugitive emissions. A variety of measurement approaches, spanning scales of meters to hundreds of kilometers, gather data that can be used to understand complex patterns of urban CH₄ emissions. These observations can inform a shared CH₄ mitigation plan developed by a metropolitan CH₄ partnership, consisting of emitters, researchers and regulators, with the shared goal of adaptive management of urban CH₄ for safety and climate mitigation.

Initiative, 2011). These diverse CH₄ sources pose an attribution challenge for atmospheric scientists; however, their urban confluence provides an opportunity for CH₄ mitigation and the development of new renewable energy sources.

Here, we present a detailed review of major CH₄-producing sectors in the urban environment, including the state of knowledge, emerging trends, challenges, and opportunities for mitigation. Across sectors, there is a need for improved quantification of emissions, cooperation between stakeholders for measurements and mitigation, development and deployment of new mitigation technologies, and verification of emissions reduction efforts.

2.1. New Approaches Are Needed for Reducing Natural Gas Leaks in Cities

Natural gas systems are the second largest anthropogenic source of CH₄ globally [U.S. Environmental Protection Agency, 2012]. CH₄, the primary component of natural gas, escapes to the atmosphere in nearly every step of the natural gas supply chain, namely production, gathering and processing, transmission, storage, distribution, and use. Fugitive emissions from the natural gas fuel cycle are primarily a function of use but also depend on the extraction and processing techniques, the distance gas travels to the end user, and the leakage rate of the pipeline [U.S. Environmental Protection Agency, 2013]. Natural gas consumption is growing rapidly because economic factors—recent advances in gas drilling, such as hydraulic fracturing with horizontal drilling—have lowered the price of the commodity. Natural gas use is also being promoted by governments as a means to improve air quality and reduce greenhouse gas emissions as it is a more

efficient and clean burning energy source relative to other fossil fuels (i.e., combustion produces less CO₂, SO₂, NO₂, and particulate matter per unit of energy). However, recent studies have called attention to the potential for fugitive emissions in the natural gas fuel cycle to undermine greenhouse gas reduction goals of fuel switching [Alvarez et al., 2012]. Recent estimates of natural gas leakage indicate that CH₄ emission rates are currently underestimated in greenhouse gas inventories [Brandt et al., 2014], and thus, it is unclear if switching to CH₄-based fuels provides a net benefit for climate mitigation.

Atmospheric and facility-level measurements suggest that leaks in the natural gas network are also significant in cities, where storage, distribution, and use are concentrated [Wunch et al., 2009; Gioli et al., 2012; Lamb et al., 2015; McKain et al., 2015; Subramanian et al., 2015]. CH₄ leaks from urban gas distribution systems have long been recognized from atmospheric observations in Europe [Shorter et al., 1996]. More recently, road surveys of CH₄ in Washington, DC and Boston have uncovered many pipeline leaks, including several with CH₄ levels high enough to pose an explosive hazard [Phillips et al., 2013; Jackson et al., 2014]. Several recent explosions caused by pipeline gas leaks have resulted in mortality and extensive damage in incidents in San Bruno, California, in September 2010, and in New York, in March 2014 [Lagos et al., 2010; Santora, 2014], drawing attention to the aging natural gas infrastructure of most North American cities [Forman, 2014; McGeehan et al., 2014; West, 2014]. More leaks have been found in areas served by aged, cast-iron mains [Phillips et al., 2013; Jackson et al., 2014]; however, problems also have been identified with steel and plastic pipes [Van Derbeken, 2011]. A large leak at a natural gas storage facility near Porter Ranch, California in 2015–2016 recently drew attention to the vulnerability of underground gas storage to large CH₄ leaks. While this event is likely the largest gas release from a storage operation, dozens of leaks in similar facilities have been documented globally [Evans, 2008].

A lack of systematic understanding of pipeline leaks makes it difficult to prioritize repairs. Replacement of all pipelines in the next decade is impractical, with main pipeline replacement costing up to \$8 million dollars per mile [Forman, 2014]. Hence, utilities require information with which to identify those pipelines with the highest risk. Systematic evaluation of leaks is needed to understand the extent to which fugitive emissions originate from fittings, couplings with buildings, and other aboveground infrastructure downstream of consumer meters, including individual combustion units that may be less costly to repair. Utility companies use leak detection equipment designed for detecting explosive CH₄ concentrations, with a threshold for detection that is about ~10,000 times higher than the criteria for pipeline leak detection in recent urban studies by academic researchers [e.g., Phillips et al., 2013; Jackson et al., 2014]. To identify climate-relevant leaks and quantify gas losses, partnerships between utilities that understand and manage infrastructure, researchers with state-of-the-art equipment, and regulators with the ability to incentivize leak repair are critical [Executive Office of the President of the United States, 2014].

2.2. Minimize Leaks From Natural Gas Fueled Vehicles and Fueling Infrastructure

Currently, CH₄ emissions from transportation comprise a small portion (roughly 0.3%) of inventoried CH₄ emissions, globally and within the United States [Marcotullio et al., 2013; U.S. Environmental Protection Agency, 2014]. Inventory estimates, however, represent a lower bound for transportation CH₄, as fugitive emission sources are often excluded [Hopkins et al., 2016]. At present, transportation CH₄ emissions in the United States primarily come from conventional gasoline-fueled vehicles, which emit small amounts of CH₄ from the tailpipe as a result of incomplete combustion [Kirchstetter et al., 1996; Lipman and Delucchi, 2002]. These emissions have declined over the past several decades due to improvements in emissions control technologies [Lipman and Delucchi, 2002]. However, the future of transportation CH₄ emissions will be transformed by increasing use of natural gas as a transportation fuel. Natural gas-powered vehicles are currently promoted as a means to reduce air pollution in cities [e.g., Delhi; Goyal and Sidhartha, 2003; Chelani and Devotta, 2007] as they produce fewer criteria pollutants than diesel and gasoline-powered vehicles [Wang, 1996]. Natural gas is a particularly popular choice for municipal fleets [Yang et al., 1997; Johnson, 2010]. However, a switch to natural gas as a vehicle fuel is not likely to reduce radiative forcing in the near term because fugitive CH₄ emissions outweigh reduced CO₂ production from tailpipes [Venkatesh et al., 2011; Alvarez et al., 2012; Burnham et al., 2012].

Fugitive CH₄ arises from vehicles that directly combust natural gas, in the form of compressed natural gas (CNG) or liquefied natural gas (LNG), and from vehicles that use natural gas as an energy feedstock, such as a fuel cell or hydrogen-powered cars. In addition to fugitive emissions from gas production and



Figure 2. Methane measurements at a compressed natural gas vehicle fueling station in Irvine, California. Fueling station outlined in blue.

transport, directly fueled CNG and LNG vehicles emit CH_4 during operation. CH_4 emissions from combustion are about 20 times higher from CNG vehicles than gasoline-powered vehicles [Lipman and Delucchi, 2002]. However, CH_4 from combustion only constitutes 4–10% of total CH_4 emitted from CNG vehicles, with the remainder from fugitive emissions [Alvarez et al., 2012]. Emissions may also result from the process of converting pipeline gas to vehicle fuel and during the fueling process itself. For CNG vehicles, pipeline gas must be compressed before delivery to the fuel tank. For LNG vehicles, natural gas is liquefied and kept at low temperatures and high pressures to maintain a liquid state. CNG and LNG both require storage, compressors, fueling lines, valves, pump systems, and nozzles downstream of the natural gas pipeline [Marathon Technical Services, 2004]; however, there is little information about leak rates of these components [but see Transportation Research Board, 1998].

Recent surveys provide evidence for fugitive emissions in fueling stations—elevated CH_4 levels (>200 ppb above background) were observed at 12 of 13 different CNG filling stations surveyed by a mobile laboratory in Orange County, California (Figure 2; Appendix SI, Supporting Information). Particularly, high levels were found near storage tanks and connecting pipes; however, CH_4 enhancement was highly variable across stations, suggesting that fugitive leaks are responsible. The largest CH_4 enhancement observed in a mobile laboratory campaign in the Los Angeles Basin was attributed to a natural gas fueling station for heavy-duty vehicles in the Port of Long Beach [Hopkins et al., 2016]. These observations suggest that CH_4 emissions are likely underestimated for CNG vehicles, particularly as commonly used lifecycle assessment models that calculate well-to-wheels emissions from natural gas-powered vehicles such as GREET (Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation model [Wang, 1996]) neglect fugitive CH_4 emissions (A. Burnham, personal communication 2014).

Natural gas vehicle use has grown rapidly over the past decade and will continue to grow globally, particularly in developing countries in South Asia and Latin America [Nijboer, 2010]. In the United States, use

of natural gas as a transportation fuel is growing most rapidly for heavy-duty and mass transit vehicles [Nijboer, 2010]. Many cities are switching fleets to natural gas with the assistance of the U.S. Department of Energy Clean Cities program in an effort to reduce petroleum use [Johnson, 2013]. In Los Angeles, nearly half of the refuse trucks run on natural gas fuel, and a large number of buses, taxis, and drayage trucks in the port have been switched to natural gas [Nijboer, 2010; Udy, 2011; Weikel, 2011; San Pedro Bay Ports, 2014]. Municipalities may also have access to biogas sources to fuel these vehicles from local landfills or wastewater treatment.

New mitigation strategies are needed to prevent CH₄ emissions from increasing with natural gas usage in the transport sector. Better quantification is needed for CH₄ emissions from downstream portions of the natural gas fuel cycle, including fueling infrastructure and combustion emissions of CH₄, which can vary widely across vehicles [Lipman and Delucchi, 2002]. Policy decisions promoting natural gas vehicles should consider the impact of fugitive CH₄ leaks in lifecycle analyses, even in the case of vehicles powered indirectly by natural gas (e.g., fuel cells). Also, decisions should consider whether natural gas vehicles actually contribute to air quality goals given that volatile organic compounds, such as ethane, are also emitted from fugitive natural gas leaks [Moore et al., 2014]. Natural gas is a promising transportation fuel because of potential renewable sources (e.g., landfill gas); however, fugitive CH₄ may undermine climate mitigation goals without careful monitoring and leak repair.

2.3. Current Landfill Methane Mitigation Efforts Are Insufficient

The majority of waste CH₄ emissions globally come from urban landfills (Table 1) and will increase globally with urbanization and population growth. Landfills are the predominant means of waste disposal in urban settings [Themelis and Ulloa, 2007] and among the most CH₄-intensive form of waste management. Cities require landfills as a means to avoid the sanitation and air pollution problems of open dumping and trash burning common in rural areas, but produce more CH₄ due to anaerobic conditions created by waste compaction and burial [Bogner et al., 2008; Vergara and Tchobanoglous, 2012]. Landfill CH₄ emissions from developed countries have declined in recent decades owing to increased landfill CH₄ recovery and waste diversion practices [Bogner et al., 2008]. However, emissions from developing countries are likely to increase with urbanization, population growth, and higher standards of living [Bogner and Matthews, 2003; Vergara and Tchobanoglous, 2012].

CH₄ emissions from landfills are determined by several factors, including the amount and type of waste disposed, the physical environment, and CH₄ capture systems in place. Waste management practices control landfill CH₄ emissions by two general strategies: (1) reducing CH₄ production and (2) capturing landfill CH₄. CH₄ production can be prevented by reducing the amount of waste that ends up in landfills—cities have implemented this approach with pay-as-you-throw pricing and diversion of organic waste to alternative treatments such as composting [Bogner et al., 2008; Vergara et al., 2011]. Emissions from landfilled waste can be further reduced by altering the decomposition process that converts organic waste to CH₄ and CO₂, which is largely a function of anaerobic conditions in the landfill [Bogner et al., 2008]. Landfill management strategies such as the use of caps and liners alter temperature, moisture, oxygen availability in a landfill, and hence the amount and rate of CH₄ production [Bogner and Matthews, 2003; Scheutz et al., 2009].

Engineered systems to physically remove CH₄ produced in landfills are currently thought to be the most effective landfill mitigation technique [Bogner and Matthews, 2003]. Landfill gas collection systems use extensive networks of wells and pipes to extract gases produced inside the landfill. Captured landfill gas is vented to the atmosphere, flared, or used as a renewable fuel for electricity generation or vehicle fueling [Cosulich et al., 1992]. Another strategy is to use microbial oxidation of CH₄ in landfill cover materials to destroy CH₄ before it reaches the atmosphere. Biological CH₄ oxidation can be promoted by additions of soil, compost, and sludge over landfills [Bogner and Matthews, 2003; Scheutz et al., 2009].

Landfill gas collection systems alone are insufficient. Landfill gas recovery systems were designed primarily to prevent explosive hazard and for odor control, not to reduce CH₄ emissions [Cosulich et al., 1992]. Some landfill gas recovery systems may paradoxically increase emissions by venting recovered CH₄ directly to the atmosphere, thereby preventing any oxidization by methanotrophic soil microorganisms that would otherwise occur. For example, CH₄ emissions from a closed landfill in Orange County, California are vented directly to the atmosphere from a landfill gas collection system (Web Object 1; Appendix SI). Extensive plumbing systems used for landfill gas recovery create ample opportunities for fugitive emissions, as recently

observed by airborne infrared imaging at a large Los Angeles landfill [Tratt *et al.*, 2014]. The airborne spectrometer detected large plumes emanating from CNG fueling and gas flaring infrastructure. At sites where landfill gas is recovered for use as biogas, landfills may be managed to optimize CH₄ collection rather than to reduce CH₄ emissions [Spokas *et al.*, 2006; Sierra Club, 2010]. This suggests that biogas production may undermine greenhouse gas reduction goals of a landfill gas recovery project if there are significant fugitive emissions in the biogas lifecycle.

To maximize the potential of CH₄ mitigation, CH₄ emissions reduction should become an explicit goal of landfill management, and attainment should be verified with regular surveys. More research is needed to understand the effectiveness of currently practiced and proposed landfill mitigation activities. In particular, a better understanding of fugitive emissions from these highly engineered systems, e.g., from leaks in gas collection pipes or gaps between liners [Spokas *et al.*, 2006], could be useful to both mitigation efforts and improved quantification of landfill emissions in inventories [Bogner and Matthews, 2003]. Use of CH₄ imaging technology could enable better surveys of landfill areas and rapid determination of the location of leaks [ARCADIS U.S. Inc., 2012]. Improving landfill cover technology that enhances biological CH₄ oxidation (e.g., Adams *et al.*, 2011; Scheutz *et al.*, 2011; Lamb *et al.*, 2014) is a promising route for reducing CH₄ emissions from landfills and other waste systems. This strategy has been demonstrated in combination with existing landfill gas recovery systems [Spokas *et al.*, 2006], can be used for former landfills that continue to emit CH₄ decades after closure [Hopkins *et al.*, 2016], and is likely the most cost-effective mitigation solution [Bogner *et al.*, 2010]. Going forward, cities need to develop and implement alternatives to landfilling organic waste to prevent the production of waste CH₄ and account for lifecycle greenhouse gas emissions, such as with composting programs [Jaffe, 2013] and mechanical biological treatment [Bogner *et al.*, 2008].

2.4. More Systematic Approaches Are Needed for Water Treatment Systems

CH₄ from wastewater is the fastest growing emission source outside of fossil fuels, expected to increase by 19% over the next two decades as population grows, particularly in developing economies [U.S. Environmental Protection Agency, 2013]. Centralized wastewater treatment in cities tends to reduce CH₄ emissions relative to more primitive forms of treatments such as lagoons, latrines, and septic systems. For cities in developing countries that lack centralized wastewater systems, development of urban sewer infrastructure could reduce CH₄ emissions while also improving sanitation and public health [Rosso and Stenstrom, 2008; U.S. Environmental Protection Agency, 2013]. In developed countries, urban wastewater treatment systems minimize CH₄ emissions by the energy-intensive process of aerobic sludge digestion [Global Methane Initiative, 2013]. More affordable anaerobic digester systems are common in the developing world and are thought to reduce climate impacts of wastewater treatment through reduced electricity use [Greenfield and Batstone, 2005], but do not account for fugitive CH₄ emissions. Anaerobic digestion produces large amounts of CH₄ that is usually recovered and combusted to produce energy [Cakir and Stenstrom, 2005]. Other anaerobic systems such as wastewater lagoons can also be retrofitted with biogas capture systems [Global Methane Initiative, 2013]. Nevertheless, anaerobic digestion tends to produce higher CH₄ emissions than aerobic treatment, even with CH₄ capture technology [Daelman *et al.*, 2013]. A recent survey of anaerobic wastewater treatment plants in Pennsylvania uncovered CH₄ leaks in five of the six tested facilities [Erndwein, 2012]. The most consistent leaks came from condensation drip traps, suggesting that preventative maintenance on these components can improve safety and reduce CH₄ emissions. The high frequency of leaks illustrates the need for regular leak monitoring with anaerobic wastewater treatment.

Apart from the digestion phase, other parts of wastewater treatment require new measurements to quantify and minimize CH₄ leaks. Sewer mains may emit nearly as much CH₄ as wastewater treatment plants, yet, these emissions have been neglected by greenhouse gas emissions inventories [Guisasola *et al.*, 2008]. In situ mapping of sewer gas concentration using new sensor technology shows promise for improving estimates of CH₄ emissions, with the dual goal of informing sewer maintenance and repair in a cost-effective manner [Lim *et al.*, 2013]. Anaerobically treated effluent contains large quantities of dissolved CH₄ that can escape to the atmosphere without further treatment. Methods to capture dissolved CH₄ after it leaves the reactor, such as in a closed column with high turbulence or a subsequent aerobic treatment to allow biological oxidation should be developed and widely implemented [Cakir and Stenstrom, 2005; Global Methane Initiative, 2013].

2.5. Different Cities: Different Methane Sources, Different Mitigation Opportunities

Greenhouse gas emissions vary among cities, depending on population size and density, geographic factors such as climate and access to resources, and economic factors, including industries, affluence, and technology [Kennedy *et al.*, 2009]. Specifically, differences in latitude and climate alter the rates of biogenic CH₄ production from anthropogenic sources such as landfills, and from natural sources, such as local soils. Cultural and economic factors also play an important role in determining the presence and integrity of CH₄ emitting infrastructure, including fuel distribution networks and waste treatment facilities. Hence, cities require local information about emissions for greenhouse gas mitigation planning [Fong *et al.*, 2014]. To this end, many cities have initiated the development of their own greenhouse gas emission inventories; however, most efforts to date are limited to CO₂ emissions [Hoornweg *et al.*, 2011]. In the case of CH₄, local observations are required because of the importance of fugitive emissions.

Here, we present CH₄ data collected by on-road sampling for four urban regions in the western United States during the summer of 2013 (Figure 3). We used fast-response in situ measurements of CH₄ made from a laboratory vehicle while driving in Fairbanks, Alaska; Los Angeles and San Diego, California; and Salt Lake City, Utah [Appendix S1; see also Bush *et al.*, 2015; Hopkins *et al.*, 2016]. These regions represent a range of sizes and urban forms, from roughly 100,000 residents in Fairbanks to 12.8 million inhabitants in the Los Angeles metropolitan area [U.S. Census Bureau, 2011]. The highest CH₄ levels, reported as excess above local background levels, were observed in Los Angeles, followed by Salt Lake City, San Diego, and Fairbanks (Table 2). The number of CH₄ hotspots, defined as locations with CH₄ levels exceeding 200 ppb above background, was highly variable among cities (see Appendix S1). Numerous CH₄ hotspots were observed in the Los Angeles Basin, which serves as a gateway for trade and fossil fuel inputs to the region, and contains one of the largest active oil fields in the United States. More hotspots were detected in Salt Lake City than in San Diego, possibly reflecting greater industrial activity despite its smaller size or a more aged infrastructure, and no hotspots were detected in Fairbanks.

The frequency of hotspots observed in Los Angeles, Salt Lake, and San Diego were more than an order of magnitude lower than previously reported for Boston [Phillips *et al.*, 2013] and Washington, DC [Jackson *et al.*, 2014] (11–19 vs. 390–420 hotspots per 100 road miles; Table 2). Higher emissions in cities of the northeast United States are likely caused by leaks in older natural gas pipeline infrastructure, compared to more recently developed cities in the western United States. Indeed, Fairbanks does not have pipeline distribution of natural gas. Differences such as these are likely to be especially large between these developed urban regions and those of the developing world, which typically do not have extensive pipeline distribution networks. Research in these regions is needed as cities in the developing world are one of the fastest growing sources of emissions [Duren and Miller, 2012].

3. Methane mitigation strategy for cities

New research on urban CH₄ emissions can provide a scientific basis for mitigation activities. We suggest three hypotheses to drive future research: (H1) *Urban methane emissions are poised to grow because of ineffective current mitigation practices and increasing use of natural gas and biogas fuels.* More studies of the distribution, frequency and rate of fugitive CH₄ across different engineered urban systems are needed, as well as a better understanding of how different management practices influence emissions rates. (H2) *Mitigating urban methane emissions will require new measurements over a range of scales using multiple techniques.* Facility-level measurements, attribution studies, and new inventory approaches are needed to resolve discrepancies between purely top-down estimates inferred from atmospheric measurements and traditional activity-based inventories typically used by governments to calculate emissions. (H3) *Cities differ greatly in their methane emissions and require unique mitigation approaches.* Interdisciplinary research is needed to understand how differences in economy, population, climate, geography, policy, and development affect CH₄ emissions, and how differences in governance structures, culture, economy, and emitting sectors affect CH₄ mitigation activities and their effectiveness. This work will help determine mitigation priorities and best practices among regions.

3.1. Urban Methane Monitoring

If mitigation efforts are successful, emissions reductions over time may leave a trend in surface concentrations that is detectable with a citywide monitoring network [National Research Council, 2010]. Specifically,

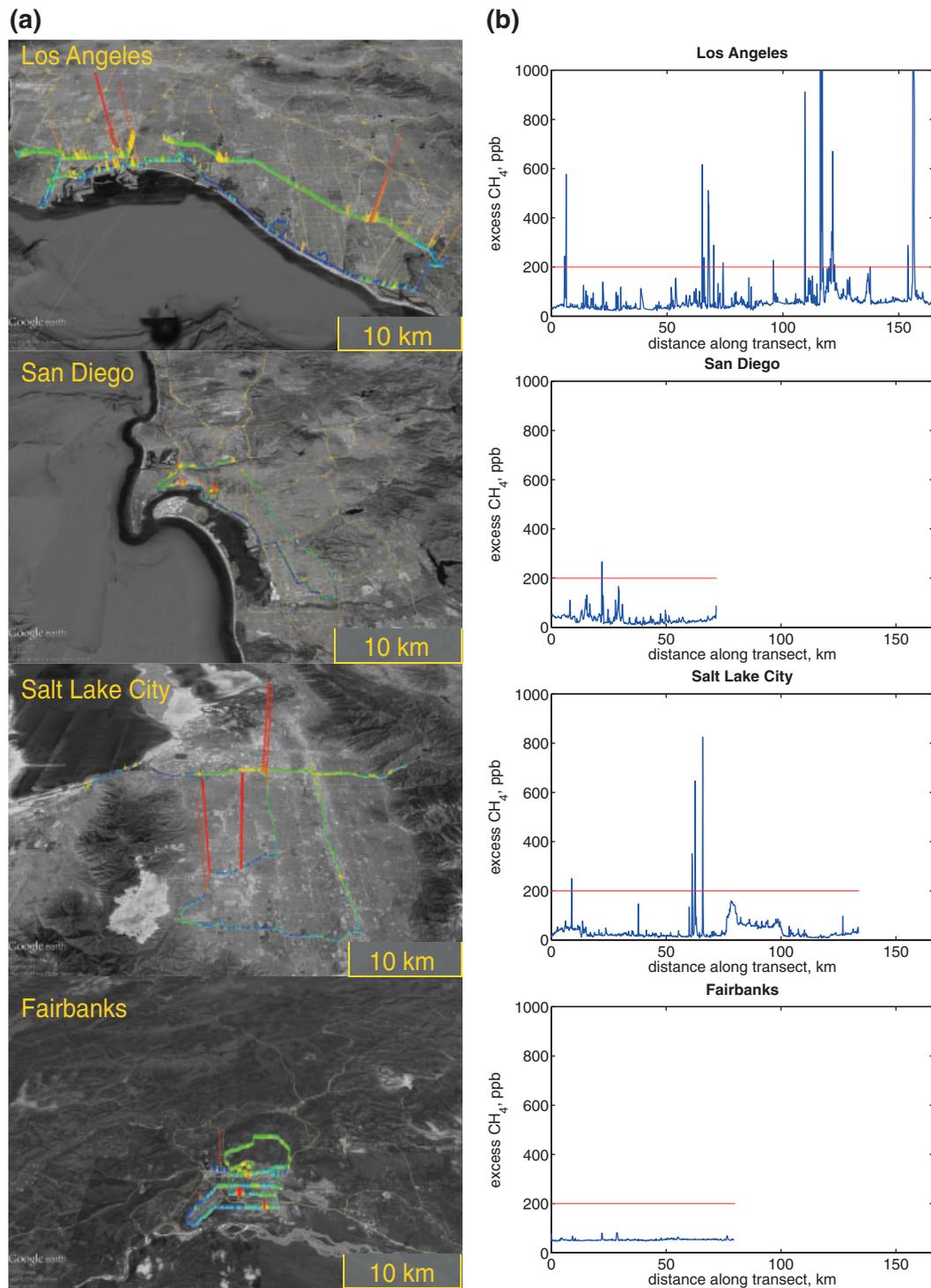


Figure 3. Atmospheric methane (CH_4) patterns from mobile laboratory studies in four western U.S. cities. Spatial patterns of CH_4 from transects covering major emission sources in each city conducted during summer 2013. Colored bars on map panels (a) show measured CH_4 mole fractions. Bar heights are proportional to absolute CH_4 excess over background levels, and bar colors were determined by the range of values observed for an individual city. Plot panels (b) show CH_4 excess values for the same transects plotted in blue by distance traveled. CH_4 excess values >200 ppb (red line) are considered to be CH_4 hotspots.

Table 2. Comparison of Four Metropolitan Regions in the Western United States Based on Mobile Methane (CH₄) Sampling During Summer 2013
Summer 2013 Transects

| Metropolitan Region | Population | | Density (pop./km ²) [United States Census Bureau, 2011] | Number of Transects | Distance Traveled (km) | Background CH ₄ (ppm) ± 1σ | Mean Excess CH ₄ (ppm) | 95th Percentile Excess CH ₄ (ppm) | Percent of | | Hotspot Frequency (Hotspots/km) |
|---------------------|---|---|---|---------------------|------------------------|---------------------------------------|-----------------------------------|--|-------------------------------|----------|---------------------------------|
| | Metro area (Millions) [United States Census Bureau, 2011] | Density (pop./km ²) [United States Census Bureau, 2011] | | | | | | | Transect >3σ Above Background | Hotspots | |
| Los Angeles | 12.8 | 3100 | 11 | 1585 | 1.821 (±0.007) | 0.185 | 0.353 | 95 | 190 | 0.12 | |
| San Diego | 3.1 | 1400 | 5 | 354 | 1.847 (±0.002) | 0.057 | 0.205 | 99 | 23 | 0.07 | |
| Salt Lake Valley | 1.1 | 700 | 4 | 718 | 1.824 (±0.003) | 0.082 | 0.157 | 82 | 62 | 0.09 | |
| Fairbanks | 0.1 | 400 | 7 | 574 | 1.823 (±0.005) | 0.047 | 0.077 | 90 | 0 | 0 | |

surface concentrations are elevated over urban areas compared with more remote locations because of the intensity of local emissions and finite rates of removal by atmospheric transport. The magnitude of this gradient, sometimes described as an “urban dome” is proportional to emission levels over time, assuming that atmospheric mixing remains unchanged [Pataki *et al.*, 2010].

Recent advances in cavity-enhanced laser absorption spectroscopy and tunable lasers have enabled new instruments capable of making high-frequency, high-precision measurements of CH₄ and key tracer species [Hendriks *et al.*, 2007; Crosson, 2008; Mønster *et al.*, 2014; Yacovitch *et al.*, 2014]. This capability facilitates new data collection approaches, such as mobile street-level sampling of multiple trace gases [Bush *et al.*, 2015] and long-term stationary monitoring [Andrews *et al.*, 2014]. In this context, the science community is poised to make a valuable contribution to mitigation planning by developing locally appropriate, comprehensive sampling strategies that place fugitive emissions from natural gas in the context of other sources.

One major challenge to mitigation and monitoring that has been identified consistently across cities is that emission estimates derived from atmospheric measurements are larger than can be explained by CH₄ inventories (e.g., London [Helfter *et al.*, 2016] and Los Angeles [Wong *et al.*, 2015]). Resolving this discrepancy will require a new combination of investigative approaches. Citywide CH₄ emission studies using multiple measurement techniques and a consortia of cooperating researchers have been tried in several cities, including Indianapolis and Boston [McKain *et al.*, 2015; Lamb *et al.*, 2016], and are currently underway in many other North American and European cities. A metropolitan CH₄ monitoring system should (1) estimate regional CH₄ emissions and detect changes over time, (2) apportion CH₄ among different source sectors, and (3) systematically characterize important CH₄ point sources. Achieving these objectives requires a variety of measurement platforms and approaches can be deployed in urban settings (Table 3).

Top-down budgets of city-wide emissions can be determined with representative measurements of CH₄ and another trace gas with a known urban emission rate [Blake *et al.*, 1984; Morizumi *et al.*, 1996]. This approach works with regional-scale measurements that observe well-mixed air with an integrated urban signal that varies in time or space, such as remote sensing, tall towers, and aircraft. For example, several recent studies have used CO and CO₂ inventories to quantify CH₄ emissions in Los Angeles, revealing emissions up to 50% larger than inventory estimates [Hsu *et al.*, 2009; Wunch *et al.*, 2009]. Top-down budgets can also be made with smaller scale, in situ observations like urban tower networks, but require inverse modeling to delineate the region of influence on the measurement and a prior emission map that reasonably represents the spatial distribution of emissions [McKain *et al.*, 2015; Lamb *et al.*, 2016].

To understand relative contributions of different CH₄ sources to urban emissions, apportionment can be performed at a variety of scales. Measurements of co-emitted gases such as ethane and propane or CH₄ isotopes can be used to distinguish fossil and biogenic sources from the regional down to the facility level [e.g., Hopkins *et al.*, 2016]. In Los Angeles, tracer measurements show that fossil sources constitute a majority of CH₄ emissions and are the likely source of discrepancy between measurement and inventory [Townsend-Small *et al.*, 2012; Wennberg *et al.*, 2012; Peischl *et al.*, 2013]. Spatial surveys can also help distinguish among CH₄ sources and explain differences between inventory and observed emissions. For example, airborne infrared imaging surveys can locate uninventoried fugitive CH₄ sources over large areas and multiple facilities [Hulley *et al.*, 2016]. Fugitive gas leaks observed by mobile sampling in Boston are consistent with findings of high contributions of fossil CH₄ to city budgets [Phillips *et al.*, 2013; McKain *et al.*, 2015].

Finally, systematic measurements of individual emission sources are needed to refine bottom-up estimates that rely on accurate facility counts and emission factors. Specifically, uninventoried fugitive emissions should be included, as well as super-emitters that disproportionately contribute to sectoral emissions [Brandt *et al.*, 2014]. Quantification of emissions at the facility level can improve inventories and help determine priorities for mitigation. New, fine-scale inventories of CH₄ emissions that include all potential sources of fugitive CH₄ could be useful for designing targeted, statistically robust sampling and for interpreting urban-scale CH₄ surveys.

3.2. Design and Attributes of a Methane Mitigation Partnership

Urban CH₄ mitigation will require the involvement of diverse stakeholders in addition to new observational data [Kennedy *et al.*, 2009; Hoornweg *et al.*, 2011]. Cooperation among partners is needed, for example, to

Table 3. Urban CH₄ Measurement Approaches

| Application | Technique | Advantage | Disadvantage | References |
|--------------------------|--|---|--|---|
| Top-down regional budget | Remote sensing (e.g., satellite, scanning Fourier transform spectrometer) | <ul style="list-style-type: none"> Regional to global coverage Repeated surveys | <ul style="list-style-type: none"> Measurement confined to specific overpass times during cloud-free, daylight hours Coarse spatial resolution | Wong et al. [2014] |
| | Total column measurements of CH ₄ (ground-based Fourier transform spectrometer) | <ul style="list-style-type: none"> Avoids influence of planetary boundary layer height on measurement Effective for trend detection | <ul style="list-style-type: none"> Requires separation of city and free troposphere contributions to column Limited to cloud-free periods | Wunch et al. [2009] |
| | Stationary tall tower site or tower network | <ul style="list-style-type: none"> Continuous, long term in situ measurements can detect trends in time Source attribution possible with additional tracer measurements | <ul style="list-style-type: none"> Requires measurements of planetary boundary layer height, meteorology, and inverse models to calculate a flux Height and location determines footprint; access to appropriate sites is challenging | Hsu et al. [2010]; McKain et al. [2015]; Jeong et al. [2013] Source attribution: Wennberg et al. [2012]; Lowry et al. [2001] |
| | Airborne in situ sampling (e.g., aircraft, unmanned aerial vehicles) | <ul style="list-style-type: none"> Extensive spatial coverage: city to regional scale Measurements of individual facilities/area sources Source attribution possible with additional tracer measurements | <ul style="list-style-type: none"> Use of airspace requires air traffic control permission/approval Infrequency and expense of campaigns limits time resolution; just a “snapshot” Spatial resolution limited by minimum allowable height | Miller et al. [2013]; Berman et al. [2012]; Khan et al. [2012] Source attribution: Wennberg et al. [2012]; Peischl et al. [2013] |
| Source apportionment | Inventoried trace gas with other urban sources (e.g., CO) | <ul style="list-style-type: none"> Necessary to estimate a top-down budget using the scaling ratio method | <ul style="list-style-type: none"> Relies on accuracy of inventory | Wunch et al. [2009]; Hsu et al. [2010]; Blake et al. [1984]; Morizumi et al. [1996] |
| | Co-emitted species (e.g., alkanes such as ethane) | <ul style="list-style-type: none"> Apportionment of biogenic and fossil sources May be able to distinguish pipeline gas from unprocessed fuel emissions | <ul style="list-style-type: none"> Other sources of these co-emitted species may not be well constrained | Peischl et al. [2013]; Katzenstein et al. [2003] |
| | Isotopes of CH ₄ | <ul style="list-style-type: none"> Apportionment of biogenic and fossil sources | <ul style="list-style-type: none"> Cannot clearly distinguish natural gas pipeline leaks from other fossil sources | Townsend-Small et al. [2012] |

Table 3. Continued

| Application | Technique | Advantage | Disadvantage | References |
|--|-----------------------------------|---|---|--|
| Point source/facility-level characterization | On-road mobile sampling | <ul style="list-style-type: none"> • Fine-scale pattern of CH₄ in a city • Point source measurements over city to continental scales • CH₄ hotspot location • Source attribution with additional tracer measurements • Global coverage | <ul style="list-style-type: none"> • Difficult to integrate to obtain a regional emissions flux • Point source access limited by road network and facility access | <p>Phillips et al. [2013]; Jackson et al. [2014]; Bush et al. [2015]; Pétron et al. [2012]; Farrell et al. [2013]; Leifer et al. [2013]</p> <p>Source attribution: Hopkins et al. [2016]</p> |
| | Satellite CH ₄ imaging | <ul style="list-style-type: none"> • Broad spatial surveys possible (~100 s of km²) | <ul style="list-style-type: none"> • Difficult to resolve city-scale | <p>Frankenberg et al. [2011]; Kort et al. [2014]</p> |
| | Airborne CH ₄ imaging | <ul style="list-style-type: none"> • Additional tracer gases can be retrieved (e.g., NH₃) | <ul style="list-style-type: none"> • Air traffic control height restrictions in urban areas | <p>Tratt et al. [2014]; Harig et al. [2003]; Thorpe et al. [2014]; Hulley et al. [2016]</p> |
| | Eddy covariance towers | <ul style="list-style-type: none"> • Flux measurements at neighborhood scale (2–3 km²) [Gioli et al., 2012] • Direct measurement of emissions rate | <ul style="list-style-type: none"> • Infrequency and expense | <p>Gioli et al. [2012]; Helfter et al. [2016].</p> |
| | Ground infrared imaging | <ul style="list-style-type: none"> • Visualize plume, pinpoint emission sources | <ul style="list-style-type: none"> • Does not give quantitative results except with custom made sensor [see Gálffalk et al., 2015] | <p>Gálffalk et al. [2015]</p> |
| | Chamber-based approaches | <ul style="list-style-type: none"> • Temporal evolution of emission source | <ul style="list-style-type: none"> • Spatial scaling for area sources | <p>Bogner et al. [2011]; Kaye et al. [2004]</p> |
| | Tracer release (e.g., acetylene) | <ul style="list-style-type: none"> • Direct flux measurements • Quantify flux from a point or area source | <ul style="list-style-type: none"> • Chamber shape limits applications • Need to know location of source • Does not work for collocated sources | <p>Mønster et al. [2014]</p> |

determine the cost of appropriate repairs. Cost estimates require knowing the location of CH₄ leaks, only possible with precise CH₄ detecting equipment that is often too costly for individual operators. Meanwhile, researchers who have this specialized equipment cannot make these measurements without site access. In order to overcome these barriers, we suggest the formation of regional CH₄ partnerships composed of researchers, regulators, and representatives of CH₄ emitting industries, including public utilities and for-profit corporations, that share the goal of managing regional CH₄ emissions. The Environmental Defense Fund, a nongovernmental organization, recently coordinated a partnership to study CH₄ emissions from the U.S. natural gas supply chain that included research and industry partners [Environmental Defense Fund, 2016]. This partnership led to new bottom-up measurements of leaks in natural gas distribution, transmission, and storage [Lamb *et al.*, 2015; Subramanian *et al.*, 2015] and demonstrated the value of multiple measurement techniques [Harris *et al.*, 2015]. As yet, however, no new natural gas regulations of these sectors have resulted.

Metropolitan regions could be an effective scale for these partnerships, particularly at the level of regulatory action (e.g., air pollution control district) and because of stakeholders' shared interest in local economic development, public health, and consumer perception. For industries where CH₄ is an economic commodity, technical and perhaps financial assistance to repair fugitive CH₄ leaks could be an incentive for participation. Even in cases where CH₄ capture is not economical, reduced liability from CH₄ safety hazard in populated areas may be sufficient motivation. Municipal contracts with landfill operators or natural gas utilities could also require participation. In addition to reaching climate and air quality goals, municipalities could benefit from collaborations with researchers to verify emission reductions for international mitigation initiatives [Hsu *et al.*, 2015]. Government and academic researchers from a variety of disciplines could use the partnership as a springboard for research on the urban CH₄ budget, to develop and test new mitigation technologies, and a host of other topics related to climate mitigation. One promising area of collaboration is in the design a city-wide CH₄ inventory, drawing on knowledge of infrastructure held by industry, experience, and tools needed to compile inventories from government partners, and new data collected by researchers. In addition to better accounting for emissions, the inventory can be used as a prior for inverse studies and for policy analysis.

The partnership should have a specific mitigation target and allotted time to achieve the goal, e.g., 10 years to reduce CH₄ emissions by 20%. To achieve this goal, the partnership needs to build a scientific basis for mitigating CH₄ emissions in the metropolitan region. Strategic, coordinated measurements can eliminate discrepancies between bottom-up inventories and top-down atmospheric measurements that impede communication between regulators and researchers. Of particular importance is resolving the contribution of super-emitters that are thought to contribute disproportionately to urban emissions, which may be effective mitigation targets. The partnership also needs to create a process by which leaks can be systematically detected and evaluated as mitigation targets and then repaired as appropriate in the context of a cost-efficient, city-wide mitigation goal.

There are major organizational challenges that must be overcome for these partnerships to be realized. One potential conflict is the requirement of privacy and liability issues for emitters, and possibly regulators, versus academic freedom needs of researchers. To motivate participation and free sharing of information among partners, deliberations by the team could be protected by confidentiality agreements, similar to the operation of the National Research Council committees overseen by the U.S. National Academies. Other products from such a partnership, including information on outcomes of mitigation efforts, would be freely accessible by the public. Another challenge is the variety of different industries involved; however, several multisectoral CH₄ projects already exist that can provide guidance to metro-scale efforts. Examples include the Global Methane Initiative, a voluntary partnership among governments, private sector members, development banks, universities, and nongovernmental organizations and the United Nations Environment Program's Climate and Clean Air Coalition to Reduce Short-Lived Climate Pollutants [United Nations Environment Program,]. Organizations such as U.S. DOE's Clean Cities Program, U.S. EPA's Natural Gas Star Program, and C40 Cities may already have established some of the collaborations needed among potential participants. Finally, metropolitan scale coalitions might not be able to influence major emitters, for example, as natural gas utilities are often regulated at the state level. However, lobbying at the metropolitan scale may be able to influence future regulations, as demonstrated by state and national level responses to natural gas leak

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events in cities [Simon, 2011; Wire, 2016]. These organizations may also be able to extend their reach by sharing best practices with other cities globally.

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