A review of methane emissions source types, characteristics, rates, and mitigation across U.S. and Canadian cities

Coleman Vollrath^{1*}, Zhenyu Xing¹, Chris H. Hugenholtz¹, Thomas E. Barchyn¹, Jennifer Winter^{2,3}

5 6

- ¹ Centre for Smart Emissions Sensing Technologies, Department of Geography, University of Calgary,
- ²Department of Economics and School of Public Policy, University of Calgary
- ³Environment and Climate Change Canada 9
- 10 *Author to whom any correspondence should be addressed.

11 12

1

2 3

4

7 8

E-mail: coleman.vollrath1@ucalgary.ca

13 14

15

16

17

18 19

20

21 22

23 24

25

26

27 28

29

30

31

32

33 34

Abstract

Cities are major aggregate sources of methane (CH₄) emissions and play a critical role in mitigating nearterm global temperature rise. However, characterizing urban CH₄ emissions remains challenging due to the diversity and spatial distribution of sources. Furthermore, limited synthesis and integration of the literature has led to a poor understanding of the characteristics and contributions of different sources, with implications for policies and mitigation. This review consolidates findings from 103 peer-reviewed articles on CH₄ emissions from cities in the U.S. and Canada, highlighting key research priorities. We find that top-down (TD) estimates of total city-level CH₄ emissions exceed bottom-up (BU) inventory estimates by a factor of 0.7 to 4.9 in 34 studies. In city-level studies that disambiguated emissions by source, natural gas distribution and use, and landfills, dominated urban CH₄ footprints. The mean natural gas loss rate in cities of $1.8\% \pm 0.9\%$ suggests a broader natural gas supply chain loss rate of $4.0\% \pm$ 0.9%. Notably, TD estimates of CH₄ emissions from six select U.S. landfills were, on average, 2.6 (± 1.8) times greater than self-reported estimates, suggesting that preferred calculation methods for reporting may systematically underestimate emissions and miss fugitive point sources. A limited number of studies examined mitigation but indicate that measurement is essential to identify mitigation opportunities and verify reductions. We raise questions and highlight challenges around existing BU inventories, urban natural gas loss rates, combustion slip, landfill emissions estimation techniques, and mitigation effectiveness. We conclude with recommendations on research priorities to address key knowledge gaps: (i) new source-level measurement datasets and modeling approaches for BU emissions estimation; (ii) more granular investigations to understand the specific sources and causes of CH₄ emissions from urban natural gas infrastructure and end use; (iii) a better coupling between measurement and modeling of landfill CH₄ emissions; (iv) mitigation-focused studies.

35 36 37

Keywords: methane emissions, cities, urban, natural gas, landfills, waste, emissions rates, mitigation

38 39

40

41 42

43

44 45

46 47

48

1. Introduction

Limiting global temperature rise to the 1.5°C Paris target is becoming increasingly unlikely (IPCC, 2023). Global mean temperatures have risen by 1.1°C relative to 1850-1900, and carbon dioxide (CO₂) and methane (CH₄) — the two most abundant greenhouse gases (GHGs) in the earth's atmosphere — are responsible for approximately 50% and 30% of this warming, respectively (IPCC, 2023). Anthropogenic CH₄ — mainly from agriculture, fossil fuel production, and waste decomposition (Saunois et al., 2020) has become an attractive target to mitigate near-term global temperature rise because of its potency and short atmospheric lifetime relative to CO₂. Ocko et al. (2021) estimate that substantial and fast reductions in CH₄ emissions this decade could slow the pace of global warming by 30% and avoid 0.25°C of additional warming by 2050.

49 50

Governments are increasingly making CH₄ a focus of broader decarbonization strategies. Over 150 countries are participating in the Global Methane Pledge, a voluntary initiative to reduce CH₄

emissions by 30% below 2020 levels by 2030. National policies to reduce CH₄ emissions from oil and gas (EPA, 2023a; European Commission, 2022; Government of Canada, 2023a) and landfills (EPA, 2023b; Government of Canada, 2023b) are in development or have been implemented in Nigeria, Canada, Colombia, the European Union, and the U.S. However, data indicate that more aggressive actions may be required: global atmospheric concentrations of CH₄ are rising quickly; the largest interannual increase measured with modern instruments was recorded in 2021 (Feng et al., 2023). Projections also show that global anthropogenic CH₄ emissions could rise by up to 13% by 2030 without executing previously announced commitments, mainly due to increasing emissions from agriculture, solid waste, and natural gas (UNEP & CCAC, 2022.) This would push Paris targets further out of reach and emphasizes the importance of accelerating and deepening measures to reduce CH₄ emissions, and translating commitments into verifiable reductions.

Cities are large aggregate sources of CH₄ emissions, with estimates based on different scaling and aggregation methods suggesting they account for between 21% and 34% of total global anthropogenic CH₄ emissions (Wunch et al., 2009; Marcotullio et al., 2013; de Foy et al., 2023). The profile of emissions sources and rates can differ considerably between cities globally depending on factors such as climate. fuel mix, industry, and the quality, age, and management of infrastructure. Key urban sources generally include solid waste disposal, natural gas distribution/end-use, and wastewater collection and treatment. In the U.S. and Canada, the abatement and mitigation potential of CH₄ emissions from these sources has not been fully realized. Natural gas and solid and liquid waste systems in cities in these countries are highly engineered and managed, which may differ from other countries (e.g., Fernandez et al., 2022; de Foy et al., 2023; Dogniaux et al., 2024). Despite oversight, the peer-reviewed literature indicates that U.S. and Canadian cities are generating substantial CH₄ emissions. For example, Plant et al. (2019) estimate that the U.S. east coast cities of Baltimore, Boston, Washington, DC, New York, Philadelphia, and Providence emit more CH₄ in aggregate than the Four Corners and Bakken oil and gas production regions in the U.S. Moreover, NASA Carbon Mapper aircraft surveys of ~250 U.S. landfills between 2018 and 2022 detected point-source plumes at 52% of landfills; for emitting landfills with data that passed quality assurance measures, the mean emissions rate was 955.3 kg/h (Cusworth et al., 2024). Notably fewer estimates of CH₄ emissions rates from Canadian cities and the sources within exist in the peer-reviewed literature; however, given similarities in infrastructure, emissions profiles and root causes in Canadian cities likely mirror U.S. cities.

Control of CH₄ emissions from key sources in U.S. and Canadian cities can be improved. In some instances, this is actioned with regulations; for example, there are new draft regulations for landfill CH₄ emissions in Canada and the U.S. (EPA, 2023b; Government of Canada, 2023b). While landfills are an important mitigation target, the problem of CH₄ emissions from U.S. and Canadian cities should be addressed broadly: there are thousands of small point- and area-sources of CH₄ emissions distributed spatially across cities offering abatement or mitigation opportunities. Policymakers and regulators build pathways to manage and reduce CH₄ emissions; for cities, this requires understanding what the major sources are, how much they emit, the causes of emissions, and what mitigation strategies are effective. To support this, we consolidated and critically reviewed research from 103 peer-reviewed journal articles published between 2003 and 2004 that examined CH₄ emissions from U.S. and Canadian cities. Our objective was to consolidate the current state of knowledge and highlight key research priorities based on the research literature covering urban CH₄ emissions source types, characteristics, measured rates, and mitigation potential.

2. Methodology

2.1. Literature search and selection

Two researchers originally searched Scopus and Web of Science databases on June 3 and June 6, 2023, respectively, using the search terms "methane emissions" AND (cities OR urban). These terms were used to prioritize the spatial context of the review (e.g., cities or urban areas) in the literature search. Databases were searched for peer-reviewed, English-language journal articles published between 2003 and 2023. Scopus returned 259 articles and Web of Science returned 365. We performed a single tier snowball

sampling strategy on the articles returned by Scopus and Web of Science to identify other relevant literature not directly returned by the searches. Two researchers with expertise on CH₄ emissions participated in this process. One was dedicated to Scopus and the other to Web of Science. If an article was deemed relevant to this review based on screening of its title and abstract with the criteria established *a priori*, we then screened the works cited and cited by lists for additional articles. Full-text screenings of articles were performed if there was potential but uncertain relevance to the review. This process was repeated for each relevant article returned in the database searches. The relevance criteria for articles to be included in the review consisted of one or more of the following:

- The article reported measurements of CH₄ emissions from, or sources within, cities/urban areas in the U.S. and Canada.
- The article focus was measuring total city-level CH₄ flux or characterizing emissions from anthropogenic sources related to the fossil fuel or waste (solid and wastewater) sectors.
- The article was a review or other article that synthesized past research on CH₄ emissions from cities/urban areas in the U.S. and Canada or presented noteworthy trends in emissions data.

Screening based on these criteria reduced the article count for review to 201 (Scopus: 96, Web of Science: 105). We assessed the articles identified by each of the two researchers to resolve potential differences. A total of 55 articles were identified by one researcher but not the other, and vice versa. The two researchers jointly performed a round of full-text review on these 55 articles to evaluate their conformance with the relevance criteria. This process was necessary to ensure that all potentially relevant articles were screened by both researchers to limit bias. Overall, initial agreement on 92 articles to review was reached.

We used the "save search" feature in Scopus and Web of Science to automatically identify new peer-reviewed articles as they were published and became available in the databases weekly after the initial searches. We also used the same feature in Google Scholar to identify new articles relevant to the review that recently emerged as pre-prints or accepted manuscripts. All new articles with potential relevance were screened and subject to the same snowball sampling strategy as all other articles. A total of 11 articles — Karion et al. (2023), Moore et al. (2023), Zeng et al. (2023), Balasus et al. (2024), Cusworth et al. (2024), Dogniaux et al. (2024), Hemati et al. (2024), Nesser et al. (2024), Stark et al. (2024), Vollrath et al. (2024a), and Xing et al. (2024) — were added to the review after the original search dates for a total of 103 articles. As such, we consider the review up to date at the timing of writing. Two of the articles (Balasus et al., 2024; Dogniaux et al., 2024) were pre-prints when reviewed. The 103 articles are listed in the Supplementary Information (S1).

We focused the review on articles that reported measurements of CH₄ emissions in cities or urban areas in the U.S. and Canada because the systems that collect and process solid waste, wastewater, and distribute natural gas in most U.S. and Canadian cities are highly engineered, managed, and share similar characteristics. Research suggests that these systems can vary significantly globally (e.g., Fernandez et al., 2022; de Foy et al., 2023; Dogniaux et al., 2024; Vogel et al., 2024). As such, cities in other countries outside of the U.S. and Canada may have significantly different profiles of emissions sources and rates. Moreover, while not exclusive to the urban domain, there is broader government, regulatory, and privatesector momentum on reducing CH₄ emissions in these countries (Government of Canada, 2023a, 2023b; EPA, 2023a, 2023b). Leveraging this momentum to quickly address urban CH₄ emissions may result in more immediate climate benefits. While most global countries have joined the Global Methane Pledge, the pledge is a commitment, and action on CH₄ in other countries may be delayed relative to the U.S. and Canada due to political or financial constraints. Last, the bulk of CH₄ emissions measurements in cities have been performed in the U.S. and to a lesser extent Canada. Equipping policymakers, regulators, and researchers in these jurisdictions with a state-of-the-art synthesis on the current state of knowledge and key research priorities from the peer-reviewed literature may accelerate momentum on understanding and reducing CH₄ emissions in cities. Together, these reasons justify prioritizing U.S. and Canadian cities for the review.

2.2. Data extraction and analysis

We extracted data from the articles pertaining to CH₄ emissions source types, characteristics, rates, and mitigation. We interpreted the effectiveness of mitigation strategies and options based on the emissions characteristics and rates reported in the articles. Other data such as the measurement methods used, spatial domain(s) studied, discrepancies between emissions estimates, and urban natural gas loss rates were also extracted from the articles. We determined that scientific knowledge gaps, areas for future research, and pathways to reduce CH₄ emissions in U.S. and Canadian cities could be identified and informed by the synthesis and interpretation of these data. Summaries of evidence in each article can be found in S1. Parameters extracted from each of the 103 articles, including emissions rates, are listed in the Supplementary Data (S2).

Hereafter, we refer to the articles as studies, as the term study better represents the research on CH_4 emissions from cities or urban areas performed in the articles.

3. Results and Discussion

3.1. City-level CH₄ emissions

3.1.1. Top-down vs. bottom-up inventory estimates of CH₄ emissions

City-level CH₄ emissions are the sum of all potential natural and anthropogenic sources within cities such as wetlands, geologic seeps, solid waste, wastewater collection and treatment, manufacturing and processing, and natural gas distribution and end-use, among others. Cities are thus large sources of CH₄ emissions in aggregate, and some sources and sectors might contribute more to total city-level emissions than others. In general, the peer-reviewed research literature suggests that cities are larger aggregate sources of CH₄ emissions than previously estimated. Total city-level CH₄ emissions can be estimated using top-down (TD) or bottom-up (BU) approaches. The studies estimated city-level emissions rates by pairing a TD measurement platform such as a stationary sensor network, aircraft, or satellite with a quantification technique like tracer scaling, atmospheric inversion, mass balance, or other emissions estimation technique. The methods reported in each study are listed in S2. In contrast, the BU approach—which underpins government and other emissions inventories—derives an emissions rate from the product of emissions factors (emissions per unit or activity) and activity factors (counts of units or activities). In the context of cities, a key difference between these two approaches is that BU estimates are constrained to known sources of CH₄ emissions, whereas TD estimates include emissions that may be unaccounted for, or underestimated in BU estimates.

We noted that satellites paired with atmospheric inversion (Hemati et al., 2024; Nesser et al., 2024) or other emissions estimation techniques (Plant et al., 2022; de Foy et al., 2023; Xing et al., 2024) were an emerging method to quantify total city-level CH₄ emissions in the studies. We attribute this to recent advancements in satellite-based sensor technology and CH₄ retrieval algorithms, which have made higher resolution observations of CH₄ emissions from different sources possible (Jacob et al., 2022), and growing interest in monitoring CH₄ emissions from urban areas to track and confirm reductions (Hopkins et al., 2016a; Cusworth et al., 2020a; Zhang et al., 2023). The Tropospheric Monitoring Instrument (TROPOMI) onboard the European Space Agency's Sentinel 5-P platform was the only satellite featured in studies that reported TD estimates of emissions at the city-level. This is unsurprising given that TROPOMI measures column-averaged dry air CH₄ mixing ratio with a 7 x 5.5 km² pixel resolution with daily global coverage, a suitable configuration for quantifying total emissions from area sources like cities (Plant et al., 2022; de Foy et al., 2023; Hemati et al., 2024; Nesser et al., 2024; Xing et al., 2024).

Figure 1 shows the distribution of ratios between estimates of city-level CH₄ emissions derived from TD measurements and BU inventories extracted from 34 (33.0%) out of the 103 studies and grouped by region. The studies benchmarked TD estimates of CH₄ emissions from individual cities against a variety of BU inventories, including the Emissions Database for Global Atmospheric Research (EDGAR; Crippa et al., 2023), and those developed by governments and academic researchers (e.g., Lamb et al., 2016; Maasakkers et al., 2016; Pitt et al., 2024), the latter of which are often highly customized. For U.S.

cities in Figure 1, we grouped the ratios into three geographic regions: U.S. West Coast, U.S. Central, and U.S. East Coast. The U.S. Central region encompasses cities located in the Rocky Mountain, Midwest, and Gulf Coast regions. These were combined into a single U.S. Central region to address fewer TD measurements in these regions compared to the high observational density on the U.S. west and east coasts. Canadian cities were grouped into a single "Canada" category given the low number of studies reporting TD city-level estimates (n = 3).

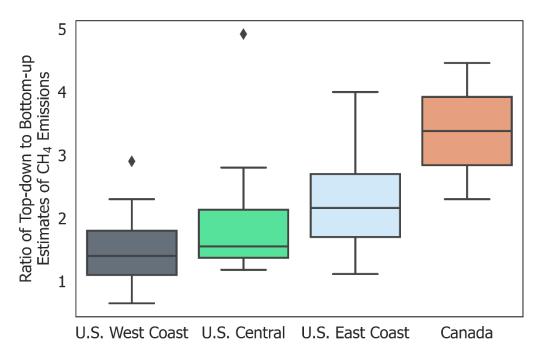


Figure 1. Distributions of ratios between TD and BU inventory estimates of city/urban CH₄ emissions reported in the studies and grouped by region: U.S. West Coast, U.S. Central, U.S. East Coast, and Canada. U.S. Central includes the Rocky Mountain, Midwest, and Gulf Coast regions. The data shown in the figure were extracted from: Anderson et al. (2021), Ars et al. (2020), Cui et al. (2015, 2019), Cusworth et al. (2020b), Fairley & Fischer (2015), Floerchinger et al. (2021), Guha et al. (2020), Hedelius et al. (2018), Heimburger et al. (2017), Hemati et al., (2024), Hsu et al. (2010), Huang et al. (2019), Jeong et al. (2013, 2016, 2017), Jones et al. (2021), Kuwayama et al. (2019), Lamb et al. (2016), Lopez-Coto et al. (2020), Nesser et al., (2024), Peischl et al. (2013), Pitt et al. (2022, 2024), Plant et al. (2019; 2022), Ren et al. (2018), Ryoo et al. (2019), Wennberg et al. (2012), Wong et al. (2015, 2016), Wunch et al. (2009), Xing et al. (2024), and Yadav et al. (2019). The total number of ratios (i.e., individual quotients between TD and BU estimates for the cities) in Figure 1 is 149 before averaging the ratios in studies that reported ratios for multiple cities in a region. After averaging, the total number of measurements is 42; by region: U.S. West Coast (n = 21), U.S. Central (n = 8), U.S. East Coast (n = 11), and Canada (n = 2).

Nesser et al. (2024) used column-averaged dry air CH₄ mixing ratio measurements from TROPOMI in an inversion framework to estimate rates for 95 different cities across the U.S. This number of city-level TD estimates substantially outweighs the number of estimates reported across the other studies in the review. Thus, to prevent weighting the results in Figure 1 substantially in favor of any one study or measurement method, we averaged the ratios from studies that measured emissions from multiple cities in a region with a single TD method before regional grouping with the ratios from the other studies. Furthermore, we did not include the ratios between TD and BU estimates of emissions for 19 U.S. cities and one Canadian city reported in de Foy et al. (2023) in our analysis, as we noted the magnitude of ratios in this study were substantially higher than the other studies, irrespective of those studies using satellites or other TD

methods to estimate emissions. de Foy et al. (2023) used column-averaged dry air CH₄ mixing ratios from TROPOMI, identical to the other satellite studies included in Figure 1, but used a 2D Gaussian technique to quantify emissions. This technique relies on using CH₄ hotspots observed with TROPOMI to inform the quantification of emissions. de Foy et al. (2023) found that urban CH₄ hotspots observed with TROPOMI had better overlap with areas of population density than gridded emissions in EDGAR v6.0, the BU benchmark used in the study. Therefore, the very large discrepancies between TD and BU estimates in de Foy et al. (2023) may be partly related to issues with the allocation of CH₄ emissions to urban areas in EDGAR v6.0 spatially and in quantity relative to the location of TROPOMI-observed urban CH₄ hotspots.

235

236

237238

239

240

241

242243

244

245

246

247

248249

250

251252

253

254

255256

257

258

259260

261262

263

264

265

266267

268

269270

271

272

273

274

275

276277

278279

280 281

282

The ratios between TD and BU estimates of city-level CH₄ emissions underpinning Figure 1 range from 0.7 to 4.9 after averaging the ratios reported in individual studies. The data allude to possible regional differences in the extent of the discrepancy between TD and BU estimates when grouped. The median ratio for the U.S. West Coast is the lowest of the four regions at 1.4, and the median ratio for U.S. Central is slightly higher than the U.S. West Coast at 1.6. Each of these regions have outliers at 2.9 for Los Angeles, CA (Plant et al., 2022) and 4.9 for Houston, TX (Hemati et al., 2024), respectively. Notably, both outliers for these regions were reported in studies that used satellites to estimate emissions. Oil and gas production in Houston, TX may explain the high ratio for this city relative to other cities. The U.S. East Coast has a higher median ratio than the other U.S. regions at 2.2 and notably larger ratio variability indicated by the lower and upper quartiles. Ratios for the U.S. East Coast range between 1.1 (Hemati et al., 2024) and 4 (Anderson et al., 2021). We typically did not observe ratios this high for non-satellite studies. The high ratio reported in the later study may be related to the timing of the stationary sensor measurements in Anderson et al. (2021), which may have corresponded with increased CH₄ emissions from natural gas sources in winter in Philadelphia, PA. Canada had the highest median ratio of all regions in Figure 1 (3.4), but a small sample size limits further comparison; the ratio is based on three TD estimates for Canadian cities reported only in two satellite-based studies (Hemati et al., 2024; Xing et al., 2024).

Ratios increasingly > 1 suggest several possible effects: (i) CH₄ emissions may be missing or underestimated with the BU approaches used, or (ii) TD methods are overestimating emissions. Several examples can be explored. Plant et al. (2019) estimate that natural gas-only CH₄ emissions in U.S. East Coast cities are ~10 times greater than estimated in inventories, which traditionally have not included CH₄ emissions from incomplete combustion at the burner tip, representing a missed emissions source in BU approaches. More recently, Pitt et al. (2024) found that TD estimates of natural gas emissions in New York were 2.3 times larger than their prior, whereas posterior biogenic emissions were 1.3 times larger. This suggests that natural gas sources — and possibly end-use emissions — are a key contributor to TD > BU in this region. Differences in the age and material type of natural gas distribution infrastructure (von Fischer et al., 2017; Weller et al., 2020) and substantial residential and commercial natural gas use in U.S. East Coast cities for space and water heating (Mittakola et al., 2024) may elevate the median TD > BU ratio for this region. More broadly, CH₄ emissions from natural gas may be driving TD > BU across the regions and cities studied. For example, diffuse natural gas emissions in Indianapolis — which we interpret as leaks and end-use emissions distributed widely across the city — are underestimated by a factor of ~1.7 to 6.9 in BU estimates (Lamb et al., 2016; Jones et al., 2021). In contrast to natural gas emissions, BU estimates of landfill emissions in Indianapolis are thought to mostly align with TD estimates (Cambaliza et al., 2015; Lamb et al., 2016; Balashov et al., 2020; Jones et al., 2021). Los Angeles-based studies indicate that emissions matching the chemical signature of dry natural gas in the distribution system and to a lesser extent oil and gas production are mainly responsible for TD > BU (Peischl et al., 2013; Kuwayama et al., 2019; Cusworth et al., 2020a). Landfills may have less influence on TD > BU in Los Angeles (Cusworth et al., 2020a) but are the main driver of TD > BU in the San Francisco Bay Area (SFBA) where landfills dominate total city-level CH₄ emissions (Jeong et al., 2016,

2017). Overall, the importance of natural gas sources to total city-level emissions and differences in the age and material type of natural gas distribution infrastructure may be driving the variability in median TD > BU ratios regionally.

TD and BU approaches can be used to track and confirm reductions, but credibility in either is undermined by disagreement between estimates. We did not observe dedicated efforts at closing gaps between TD and BU estimates of city-level CH₄ emissions in the studies. A general prerequisite to mitigating emissions is knowing individual source locations and rates, and the contribution of sources in aggregate to total city-level emissions. Carranza et al. (2018) and Pak et al. (2021) developed geospatial BU inventories of CH₄ emissions for Los Angeles and Toronto, respectively. These inventories include source locations and emissions estimates at a granular level, and as such may provide a starting point to resolve discrepancies between TD and BU estimates in cities. Field measurements of sources should be integrated into these inventories and rates updated over time. BU estimates that include empirical measurement data may have greater direct comparability to TD measurements, narrowing the discrepancy between the approaches observed at the city-level. Integrating measurement data into government BU inventories was recently shown to mostly address historic underestimation of emissions using the BU approach in the Canadian upstream oil and gas industry (Chan et al., 2024; MacKay et al., 2024). Similar approaches can be adapted to improve and better constrain estimates of urban CH₄ emissions in the U.S. and Canada.

3.1.2. Source attribution of total city-level CH₄ emissions

 Figure 2 shows results from studies in the review that reported city-level CH₄ emissions rates derived from TD measurements and disambiguated between major source types; methods used to attribute emissions are listed in S2. We extracted the studies' reported emissions rates and proportional contributions from these sources. The granularity of disambiguation differed between the studies depending on the measurement and attribution methods used; some studies only disambiguated between thermogenic and biogenic emissions, whereas others estimated emissions from landfills as a proportion of biogenic and total urban emissions. Therefore, we use a single category in Figure 2 that encompasses all biogenic emissions to avoid potential issues in understating the importance of landfills across the estimates from the studies. Notably, landfills were generally responsible for most biogenic emissions in studies that separated landfill from other biogenic emissions (e.g., Cambaliza et al., 2015; Ren et al., 2018; Kuwayama et al., 2019; Guha et al., 2020; Jones et al., 2021). Additional details on the data extraction and preparation for Figure 2 are in S1.

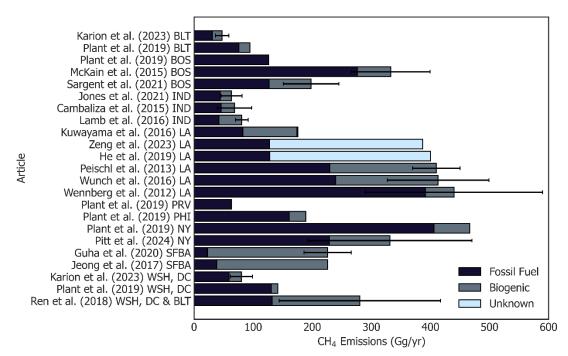


Figure 2. City/regional-level CH₄ emissions rates by major source type reported in the studies. The abbreviations for cities/urban regions following the references on the y-axis are SFBA – San Francisco Bay Area, NY – New York, LA – Los Angeles, BOS – Boston, WSH, DC – Washington, District of Columbia, BLT – Baltimore, PHI – Philadelphia, IND – Indianapolis, PRV – Providence. The fossil fuel estimates in He et al. (2019), and Zeng et al. (2023) are associated with natural gas combustion/end-use only. Error bars show measurement uncertainty as reported in the studies.

Figure 2 indicates that CH₄ emissions from fossil fuels dominate the emissions profile of most U.S. cities; however, we observe regional differences and exceptions. The mean fossil fuel percentage (FFP) derived from measurements in U.S. east coast cities is 74.6% (± 20.4%). FFPs reported by Floerchinger et al. (2021) from TD measurements in five U.S. east coast cities were included in this estimate but are not shown in Figure 2 because the study disambiguated the city-level CH₄ flux but did not quantify emissions. The FFP relative to total city-level emissions differs between individual cities and may be affected by differences in natural gas infrastructure and the time of the year when measurements are performed. Floerchinger et al. (2021) found lower FFPs in the summer than winter for some cities such as Philadelphia and Washington, DC, but less noticeable changes between seasons for others like Boston and New York. The U.S. east coast cities of Washington, DC, Baltimore, and Richmond (reported in Floerchinger et al. but not shown in Figure 2) have larger contributions from biogenic emissions, specifically landfills, and thus have smaller FFPs relative to other cities in this region (Ren et al., 2018; Floerchinger et al., 2021). The FFP does vary between studies for these cities — Plant et al. (2019) estimated that almost all measured CH₄ emissions for Washington, DC and Baltimore were of fossil fuel origin. Differences in the FFP for the same city estimated across different studies may also be due to spatial variability in the area measured between studies.

The mean FFP among studies that measured city-level CH₄ emissions on the U.S. west coast is comparatively lower ($48.4\% \pm 27.2\%$). FFPs measured by Hopkins et al. (2016b) in Los Angeles are included in this estimate but are not shown in Figure 2 because quantification was not performed. The mean FFP for Los Angeles ($62.4\% \pm 15.9\%$) is substantially higher than the SFBA ($13.5\% \pm 4.9\%$), which has a large landfill contribution (74%) to total CH₄ emissions (Guha et al., 2020). The large standard deviation for Los Angeles is partly explained by the large FFP (89.0%) reported by Wennberg et

al. (2012), which is a large percentage compared to other studies. FFP estimates in most other Los Angeles-focused studies ranged from 58% to 62% (Peischl et al., 2013; Hopkins et al., 2016b; Wunch et al., 2016). Landfills are estimated to account for most of the non-fossil CH₄ emissions in Los Angeles (Kuwayama et al., 2019).

347

348

349 350

351 352

353

354 355

356

357 358

359

360

361

362

363 364

365 366

367

368 369

370

371372

373374

375

376377

378379

380

381

382

383

384

385

386

387

388

389 390

391

392 393

394

395

396

397

FFPs relative to total emissions from cities in the U.S. Midwest such as Chicago (69.5%) and Indianapolis (57.3% \pm 18.8%) fall somewhere between estimates for U.S. west and east coast cities. One measurement performed in June 2017 with a low FFP (26.7%) lowered the mean for Indianapolis (Floerchinger et al., 2021), whereas other studies measured higher FFPs for this city (Cambaliza et al., 2015; Lamb et al., 2016; Jones et al., 2021). The South Side Landfill in Indianapolis has been characterized in several studies (Cambaliza et al., 2015; Heimburger et al., 2017; Balashov et al., 2020; Jones et al., 2021) and is estimated to account for up to ~35% of the city's total CH₄ emissions. Lamb et al. (2016) measured CH₄ from a stationary sensor network across Indianapolis and used the posterior footprint of an atmospheric inversion to estimate that biogenic emissions account for 48% of the city's total emissions. Other landfills are located on the urban fringe of Indianapolis; depending on the domain sampled, these or other biogenic sources may be contributing to the remaining biogenic CH₄ emissions that cannot be attributed to the South Side Landfill (Lamb et al., 2016). If no other landfills contribute to the 48%, then wastewater collection and treatment or other biogenic sources such as wetlands may account for up to 13% of total CH₄ emissions in Indianapolis.

The FFPs of total city-level emissions reported by He et al. (2019) and Zeng et al. (2023) for Los Angeles as shown in Figure 2 were excluded from the analyses in the preceding paragraphs. The attribution methods used in these studies only account for emissions that correlate with natural gas enduse. Operating pressures of natural gas distribution pipelines and other infrastructure and thus leak rates are thought to be to be mostly consistent year-round (Sargent et al., 2021). If correct, this means that emissions that correlate with end-use arise from 'slip' or the incomplete combustion of natural gas. As such, other urban fossil fuel emissions are excluded, and the estimates are not directly comparable to other studies. Despite this, these studies reported high FFPs. He et al. (2019) quantified CH₄ emissions for the Los Angeles basin at 27 Gg/month for the months of June-July and 45 Gg/month for the months of December-January. They regressed monthly CH₄ emissions on natural gas consumption in the region, yielding a y-intercept of 22.9 (± 1.1) Gg/month. The intercept may provide an approximation of the CH₄ emissions rate from all other sources in the region (i.e., those not associated with metered natural gas consumption). In this context, 15.2% (summer) and 49.1% (winter) of total monthly CH₄ emissions are estimated to be explained by sources related to the metered consumption of natural gas — mainly residential and commercial. Zeng et al. (2023) performed a similar analysis for Los Angeles as He et al. (2019) but included three additional years (2018-2020) in their estimates, finding a comparable proportion of total CH₄ emissions attributed to metered consumption (33% versus 32% from He et al., 2019).

Other studies reported estimates of CH₄ emissions that correlate with end-use in addition to disambiguating between major source types, or data that we used to estimate the contribution of potential end-use emissions to total city-level emissions. Karion et al. (2023) found a slightly lower contribution from metered consumption to total CH₄ emissions in Washington, DC and Baltimore (21% to 22%). For Boston, Sargent et al. (2021) reported mean annual natural gas CH₄ emissions of 127 Gg/yr over an eightyear study period (2012 to 2020). We attribute 73.4% of these emissions to metered consumption using the y-intercept of natural gas CH₄ emissions regressed on residential and commercial gas consumption reported in the study. This is equivalent to 47.1% of total CH₄ emissions in Boston, although this number may be higher given the higher FFPs reported in other studies for Boston (McKain et al., 2015; Plant et al., 2019; Floerchinger et al., 2021). For Indianapolis, we estimate that end-use emissions contribute at least 17.2% to total city-level emissions. This is based on fossil fuels accounting for an estimated 52% of total emissions in Indianapolis and end-use combustion estimated to contribute 33% to total fossil fuel emissions (Lamb et al., 2016). If the FFP in Indianapolis is higher (~67%), as suggested in other studies (Cambaliza et al., 2015; Jones et al., 2021), and end-use combustion is 33% of total fossil fuel emissions, then end-use emissions may contribute up to 22.1% of total CH₄ emissions in this city, similar to Washington, DC and Baltimore.

If approximately 32.5% (Los Angeles), 17.2% to 22.1% (Indianapolis), 21.5% (Washington, DC and Baltimore), and 47.1% (Boston) of total CH₄ emissions are tied to slip, this constrains the quantity of fossil emissions that can be attributed to leaks and vents from distribution pipelines and other natural gas infrastructure in the cities. Using these values and the mean FFPs for each individual city, we estimate that other fossil emissions (those not associated with natural gas end-use) contribute 29.9% (Los Angeles), 35.2% to 40.1% (Indianapolis), 25.5% (Washington, DC and Baltimore), and 36.2% (Boston) to total CH₄ emissions.

The observable differences in FFPs of total emissions in U.S. cities are likely driven by the presence and distribution of different sources in the areas studied. Other factors such as climate and weather (i.e., increased natural gas use at different times of the year), fuel mix for electricity and heating, and the age and material composition of distribution pipelines and other natural gas infrastructure may also contribute to the observed differences (von Fischer et al., 2017; Weller et al., 2020; Sargent et al., 2021). Moreover, FFPs may be somewhat modulated by the spatial domains sampled, and if landfills potentially located in urban fringe areas are included in estimates. In general, it is challenging to draw further conclusions on the potential reasons contributing to higher FFPs in some U.S. cities relative to others given the heterogeneity of sources within the spatial domains measured and without a structured causal inference method.

Evidence from the studies may indicate that CH₄ emissions from combustion slip could be substantial, varying between 17.2% to 47.1% of total city-level CH₄ emissions depending on the city. Slip emissions likely have a seasonal trend, which increase as temperatures decrease and more natural gas is used for heating (He et al., 2019; Sargent et al., 2021; Karion et al., 2023; Zeng et al., 2023). While this assumes that leaks from pipelines and other natural gas infrastructure do not vary seasonally, as operating pressures are held constant (Sargent et al., 2021), we note that this may only be true for leak rates. In contrast to leak rates, operating pressure likely does not affect the number of leaks, which may be affected by seasonality and temperature changes. Metal shrinks in cold temperatures and thus may result in the emergence of new leaks from pipelines and other components involved in the distribution of natural gas during winter. These may be transient and self-resolve upon temperatures warming. More leaks from natural gas distribution infrastructure during colder periods of the year could challenge our findings, as these emissions would also correlate with metered gas consumption (which increases in winter), reducing the amount of emissions attributable to slip. However, it is unclear from the studies if seasonal changes in leak frequency in the natural gas distribution segment are noteworthy enough to substantially increase non-slip emissions during winter. Moreover, this phenomenon would be highly dependent on a city's latitude and annual temperature range and may not be a widespread issue geographically. If fossil fuel emissions do have a larger component of slip (instead of leaks), this misattribution could explain why annual emissions for Boston remain unchanged over eight years despite Massachusetts requiring that utilities improve detection, reporting, and repair of distribution pipeline leaks (McKain et al., 2015; Sargent et al., 2021). Policies and actions to mitigate slip emissions may include servicing, repair, and efficiency improvements to appliances and other gas-fired equipment or replacements with electrified or other alternatives.

3.1.3. Natural gas loss rates

Estimates of urban fossil fuel CH₄ emissions in the studies indicate that noteworthy proportions of delivered natural gas are being lost to the atmosphere (Wennberg et al., 2012; Peischl et al., 2013; McKain et al., 2015; Wunch et al., 2016; Jeong et al., 2017; Ren et al., 2018; He et al., 2019; Huang et al., 2019; Sargent et al., 2021; Karion et al., 2023; Zeng et al., 2023). Figure 3 shows urban natural gas loss rates reported in the studies for select U.S. cities. We combined these urban loss rates, which account for CH₄ emissions from natural gas distribution and end-use, with loss rate estimates for the upstream and midstream natural gas supply chain segments (Alvarez et al., 2018) to estimate a total supply chain loss rate. The loss rates for the individual segments in Figure 3 can be interpreted as the amount (expressed as a percentage) of CH₄ emitted relative to total produced (e.g., Alvarez et al., 2018), throughput, or delivered/consumed natural gas. We included the Oil and Gas Climate Initiative's (OGCI's) target loss rate

of 0.2% for the upstream segment in Figure 3 to demonstrate the effect reducing upstream CH₄ losses has on the total supply chain loss rate.

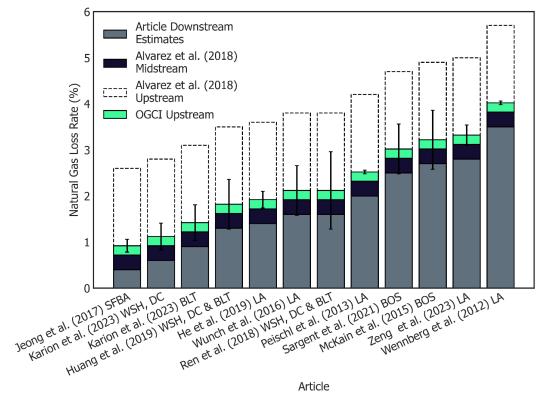


Figure 3. Natural gas loss rates for different supply chain segments. Estimates for downstream loss rates were extracted from the studies and thus account for CH₄ emissions from distribution and end-use (i.e., total urban natural gas CH₄ emissions). Loss rates for the upstream and midstream segments were extracted from Alvarez et al. (2018). The Oil and Gas Climate Initiative's (OGCI's) target loss rate of 0.2% for the upstream segment is included as an alternative to Alvarez et al. (2018) to demonstrate the effect upstream CH₄ emissions reductions have on the total supply chain loss rate. To visualize this effect, readers should eliminate the dashed bar sections in the figure. Uncertainties are the sum of downstream (if reported) and midstream uncertainties. The abbreviations for cities/urban regions on the x-axis are SFBA – San Francisco Bay Area, WSH, DC – Washington, District of Columbia, BLT – Baltimore, LA – Los Angeles, and BOS – Boston.

In Figure 3, the mean downstream loss rate reported in the studies was 1.8% (\pm 0.9%). The lowest and highest downstream loss rates were reported for SFBA ($0.4\% \pm 0.1\%$; Jeong et al., 2017) and Los Angeles (3.5%; Wennberg et al., 2012). On average, the downstream losses in Figure 3 are responsible for 41.6% (\pm 14.2%) of the total natural gas supply chain loss rate. Notably, the downstream loss rates estimated for Boston (McKain et al., 2015; Sargent et al., 2021) and two out of the five estimates for Los Angeles (Wennberg et al., 2012; Zeng et al., 2023) are > 50% of the total supply chain loss rate. Downstream loss rates reported for Washington, DC and Baltimore (Ren et al., 2018; Huang et al., 2019) and Los Angeles (Peischl et al., 2013; Wunch et al., 2016; He et al., 2019) are generally comparable. Zeng et al. (2023) estimated a downstream loss rate for Los Angeles two times greater than He et al. (2019) but for a slightly longer period. The TD estimates of CH₄ emissions used to calculate the loss rate in Zeng et al. (2023) are larger than those reported in He et al. (2019), possibly due to a slight difference in the methods used to estimate emissions between the studies. Both studies used the tracer scaling quantification method, but

Zeng et al. (2023) accounted for seasonal variability in the Los Angeles CO_2 flux whereas He et al. (2019) did not. We note that the loss rate for Ren et al. (2018) for the Washington, DC/Baltimore region is the average of two estimates over winter 2015 $(1.1\% \pm 0.6\%)$ and 2016 $(2.1\% \pm 1.0\%)$. Seasonally higher natural gas emissions in winter and timing of the measurements could explain why this loss rate is higher relative to others reported for the same region (Huang et al., 2019; Karion et al., 2023). In SFBA (Jeong et al., 2017) and Washington, DC and Baltimore (Karion et al., 2023), the upstream sector dominates total supply chain losses using the Alvarez et al. (2018) loss rate for this segment.

U.S. and Canadian O&G industries have focused the bulk of their efforts on reducing total supply chain CH₄ emissions within the upstream segment, and compliance with regulations and conformance to voluntary initiatives continue to drive reductions. We illustrate the potential effect reducing CH₄ losses upstream has on the total supply chain loss rate by substituting the Alvarez et al. (2018) upstream loss rate (1.9%) with the OGCI upstream target loss rate of 0.2%. In Figure 3, the OGCI loss rate (green fill) is superimposed on the Alvarez et al. (2018) upstream loss rate (dashed black line). As such, if upstream losses were reduced from 1.9% to 0.2%, the Alvarez et al. (2018) estimates would be eliminated from Figure 3. This would leave only the other legend categories remaining. This replacement results in a 1.7% reduction in the upstream loss rate, lowering the mean total supply chain loss rate across these studies from 4.0% ($\pm 0.9\%$) to 2.3% ($\pm 0.9\%$). As upstream emissions shrink, downstream emissions dominate supply chain emissions, averaging 72.8% ($\pm 13.3\%$) of the total across the studies.

Our estimated total natural gas supply chain loss rates do not match the target loss rates of new voluntary 'differentiated gas' schemes marketing lower-emitting natural gas. Even using an upstream loss rate of 0.2%, our estimates are ~5.8 times higher, on average, than the mean total supply chain loss rate of 0.4% reported by One Future — a low emissions natural gas industry group in the U.S. — for their partners in 2022 (One Future, 2023). It is likely that One Future's estimated loss rate excludes combustion slip emissions at the point of the customer. These are Scope 3 CH₄ emissions and therefore are mostly outside of the O&G industry's control, but their exclusion from supply chain loss rate estimates creates an incomplete picture of the climate impact of natural gas use. It is possible the O&G industry may be producing and delivering increasingly lower emissions natural gas, but additional downstream CH₄ leakage at the burner-tip may be eroding potential climate benefits. There may be cases where widespread combustion efficiency ratings are over-estimated or idealized, and most appliances and end-uses are emitting more than intended, leading to large CH₄ emissions from slip in cities in aggregate. End use CH₄ emissions are comparatively understudied and require further examination for prioritizing mitigation.

3.2. Source-level CH₄ emissions

3.2.1. Waste

3.2.1.1. Landfills

The number of studies focused on CH₄ emissions from landfills surged towards the end of the review period (2003-2024). Notably, all recently published landfill-focused studies in the review used aircraft or satellite platforms to measure point-source plumes emitted from the landfill surface or associated infrastructure or to quantify total landfill emissions (Balasus et al., 2024; Cusworth et al., 2024; Dogniaux et al., 2024; Nesser et al., 2024). Several landfills were observed with different measurement methods at different points in time across the studies. We noted that six U.S. landfills were most frequently observed between studies. Thus, we extracted emissions data from eight studies that measured CH₄ emissions from these landfills with TD methods to compare rates (Figure 4) and identify patterns for future research. Additional details on the data extraction and preparation for Figure 4 are in S1.

Figure 4 compares TD estimates of emissions for six different U.S. landfills reported across eight studies (Cambaliza et al., 2015; Lamb et al., 2016; Heimburger et al., 2017; Guha et al., 2020; Balasus et al., 2024; Cusworth et al., 2024; Dogniaux et al., 2024; Nesser et al., 2024) and U.S. Greenhouse Gas Reporting Program (GHGRP) self-reported emissions. In general, measured CH₄ emissions from landfills were much larger than estimates of emissions by landfill operators submitted to the U.S. GHGRP; TD estimates of emissions for the six landfills in Figure 4 were on average 2.6 times (± 1.8) higher than self-

reported rates. While this mean was derived from the emissions data for the six landfills in Figure 4 only, it aligns with the mean discrepancy (2.7 times) between TD aircraft estimates of point-source emissions and GHGRP self-reported rates for 96 U.S. landfills quantified in Cusworth et al. (2024). The largest discrepancy in Figure 4 was observed for the Charlotte Motor Speedway Landfill (1003099); TD estimates from four studies (Balasus et al. 2024; Cusworth et al., 2024; Dogniaux et al. 2024; Nesser et al., 2024) were on average $3.4 (\pm 0.5)$ times higher than the GHGRP self-reported rate for this landfill. The lowest mean discrepancy was observed for the Keller Canyon Landfill (1004011); TD estimates were $1.2 (\pm 0.6)$ times higher than the GHGRP self-reported rate.

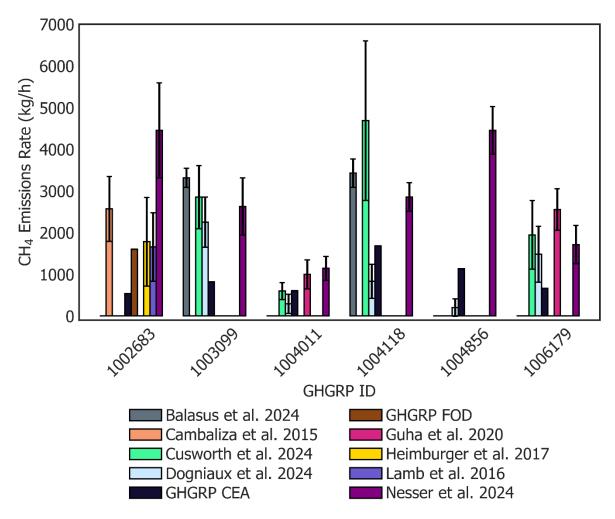


Figure 4. Comparison of TD estimates of CH₄ emissions and Greenhouse Gas Reporting Program (GHGRP) self-reported rates for six U.S. landfills grouped by GHGRP ID. From left to right on the x-axis, landfill names corresponding to GHGRP IDs are: South Side Landfill (1002683 – Indianapolis, IN), Charlotte Motor Speedway Landfill (1003099 – Charlotte, NC), Keller Canyon Landfill (1004011 – Pittsburg, CA), Sampson County Landfill (1004118 – Roseboro, NC), Rumpke Sanitary Landfill (1004856 – Cincinnati, OH), and Newby Island Landfill (1006179 – Milpitas, CA). The emissions data for these landfills were extracted from eight studies: Cambaliza et al. (2015), Lamb et al. (2016), Heimburger et al. (2017), Guha et al. (2020), Balasus et al. (2024), Cusworth et al. (2024), Dogniaux et al. (2024), Nesser et al. (2024) and the U.S. GHGRP. Error bars show reported or estimated measurement uncertainty. Additional details on data extraction and preparation are in S1. FOD (first order decay — gold bar) and CEA (collection efficiency assumption — black bars) are the two calculation methods used in U.S. GHGRP reporting for landfills (see Stark et al., 2024).

The studies estimated both point- and area-source emissions for five out of the six landfills in Figure 4. For the Charlotte Motor Speedway Landfill (1003099) and the Newby Island Landfill (1006179), TD estimates mainly agree within uncertainties. The fact that point- and area-source measurement methods quantified comparable rates suggests that most emissions from these landfills may be related to large fugitive sources or intentional releases from discrete locations; point-source imagers cannot observe

 unoxidized landfill gas permeating through landfill cover soils as diffuse CH₄ emissions. Point-source imagers attributed individual plumes to active/working faces, gaps in intermediate cover, construction of new waste cells or gas capture infrastructure, leaking gas capture wells, and unlit gas capture flares (Duren et al., 2019; Cusworth et al. 2020b, 2024; Dogniaux et al., 2024). Some of these sources can be targeted for abatement or mitigation; however, if surface activity is a proxy for the location of large emissions (Dogniaux et al., 2024), then waste deposition and collection and modification of the landfill surface may rank highest in importance to total emissions for landfills.

567

568

569 570

571

572

573

574575

576577

578

579

580

581

582 583

584

585

586 587

588 589

590 591

592

593

594 595

596

597

598

599 600

601

602

603 604

605 606

607

608 609

610

611

612 613

Detection of a plume from the active face of a landfill may depend partly on the detection limits of the measurement methods used and the time at which observations are made. First, the aircraft and satellite platforms used in recent studies have 5-10 kg CH₄/h (Cusworth et al., 2024, see also Thompson et al., 2016; Frankenberg et al., 2016) and ~100 kg CH₄/h (Dogniaux et al., 2024) detection limits, respectively. Thus, the active face may dominate total landfill emissions generally (e.g., Cambaliza et al., 2017; Goldsmith Jr. et al., 2012), but plumes from smaller active faces or those accepting less waste may be below the detection limits of these methods, or atmospheric conditions may result in non-detects. Topography of the active face and operator traffic may hamstring efforts to corroborate this hypothesis with on- or near-surface methods (Goldsmith Jr et al., 2012; Cambaliza et al., 2017; Cusworth et al., 2020b). Second, remote sensing methods that rely on measuring CH₄ abundance in a column of air using solar backscatter are limited to daytime observations. Emissions from a landfill's active face may only be detectable or elevated during daytime working hours when waste is being added to the active face or managed. Landfill operators may add a thin layer of cover soil (Cusworth et al., 2020b; Guha et al., 2020) or another barrier over the active face at the end of each working day to control odors, birds, and prevent the wind from mobilizing dust/aerosols off the surface before removing it at the start of the next working day. It is unknown whether this is standard industry practice, but covering the active face temporarily at night may temporarily modify its emissions rate. Thus, higher daytime emissions rates and observational density may contribute to TD estimates of point-sources > GHGRP self-reported rates for the Charlotte Motor Speedway Landfill (1003099) and Newby Island Landfill (1006179). Elevated daytime emissions rates from urban sources of CH₄ are not unique to landfills; incomplete combustion from manufacturing (Mitchell et al., 2018) or intentional releases from other sources during daytime operations may also be higher.

In Figure 4, the Keller Canyon Landfill (1004011) and Rumpke Sanitary Landfill (1004856) differ in that TD estimates of point-source emissions align with or are lower than GHGRP self-reported rates and are generally much lower than estimates of total emissions for these landfills. The emissions data for Rumpke Sanitary (1004856) are noteworthy in that Cusworth et al. (2024) detected CH₄ from this landfill during a single overpass but did not quantify a rate, possibly due to failure of the data to meet quality assurance criteria. Dogniaux et al. (2024) estimated a rate of 200 kg CH₄/h with GHGSat, whereas Nesser et al. (2024) estimated a rate of 4452.1 kg CH₄/h based on atmospheric inversion with TROPOMI. These are 0.2 and 3.9 times the GHGRP self-reported rate of 1131 kg CH₄/h, respectively. For these landfills, point-sources may contribute less to total emissions, GHGRP self-reported rates are underestimates, estimates of total landfill emissions are overstated, or some combination thereof. The emissions data for the Sampson County Landfill (1004118) show widely different estimates of point source emissions between methods (Dogniaux et al., 2024; Cusworth et al., 2024). This emphasizes that temporally variable emissions tied to temperature, moisture, and barometric pressure changes (Czepiel et al., 2003; Bogner et al., 2011; Goldsmith Jr. et al., 2012; Xu et al., 2014; Ren et al., 2018) may underpin these estimates. Alternatively, different point-sources emitting higher and lower rates may have been detected in Cusworth et al. (2024) and Dogniaux et al. (2024), respectively. The estimate from Cusworth et al. (2024) of 4686.2 kg CH₄/h does agree with estimates of total emissions for this landfill, albeit over a large uncertainty range.

A change to U.S. GHGRP requirements for landfills in 2016 may mostly explain the substantial disagreement between TD estimates of emissions and GHGRP self-reported rates in Figure 4 generally (EPA, 2009; Balasus et al., 2024; Nesser et al., 2024; Stark et al., 2024). Two methods are used to estimate CH₄ emissions from U.S. landfills with gas capture systems, which account for the majority (~75%) of landfills and waste emissions reporting to the U.S. GHGRP: first-order decay (FOD) and collection efficiency assumption (CEA, Stark et al., 2024). Operators of landfills with gas collection systems may choose which estimate they report to the GHRGP annually. Most are opting to report estimates derived with the CEA method, which consistently estimates lower emissions than the FOD method (Stark et al., 2024). Latitude in the method used for landfill CH₄ emissions reporting may be unique to the U.S. (Stark et al., 2024); to our knowledge, the FOD method is followed in Canada to estimate and report landfill emissions rates which are submitted annually to the Intergovernmental Panel on Climate Change (IPCC, ECCC, 2023, 2024). Reporting favoring the CEA method has produced a misleading trend that CH₄ emissions from landfills in the U.S. are decreasing; actual emissions are likely higher and may be trending upwards in response to increasing waste deposition. We refer readers to Stark et al. (2024) for a detailed overview of these estimation methods for GHGRP reporting and trends in reported U.S. landfill CH₄ emissions. Results from the studies (Nesser et al., 2024; Stark et al., 2024) suggest that key issues with the CEA method are overestimates of gas collection system efficiencies by landfill operators and higher assumed soil oxidation rates relative to the FOD method. More CH₄ emissions are likely being emitted diffusely through the landfill surface, cover gaps, or other associated infrastructure than anticipated.

614

615

616 617

618

619

620

621 622

623

624

625

626

627

628

629

630

631

632

633 634

635 636

637 638

639

640

641 642

643

644

645

646 647

648

649

650

651

652 653

654

655 656

657

658

659

660

In Figure 4, we use the South Side Landfill (1002683) as an example to illustrate the effect of the GHGRP requirement change on self-reported rates. Several studies estimated and reported total emissions from this landfill using a variety of platforms and methods (Cambaliza et al., 2015; Lamb et al., 2016; Heimburger et al., 2017; Nesser et al., 2024). Three of the studies (Cambaliza et al., 2015; Lamb et al., 2016; Heimburger et al., 2017) reported estimates for this landfill prior to the change in reporting requirements: Figure 4 shows general agreement between these TD estimates and the GHGRP selfreported rate using the FOD method. The most recent TD estimate of emissions from this landfill was reported in Nesser et al. (2024), along with a GHGRP self-reported rate based on the CEA method that is 3 times lower than the FOD estimate from a decade prior. Nesser et al. (2024) note that their TD estimate for the South Side Landfill is much larger than the other studies possibly due to the construction of a new gas capture facility in 2019 that raised emissions temporarily. Nonetheless, a > 1000 kg CH₄/h decrease in the GHGRP self-reported rate for this landfill over the last decade is inconsistent with the evolution of an active landfill's emissions profile as the volume of accepted waste increases. FOD models have known issues such as using a generalized soil oxidation coefficient across the landfill surface and failing to account for the effects of climate and weather on oxidation rates (Bogner et al., 2011; Cambaliza et al., 2017; Stark et al., 2024); however, our example demonstrates a case where FOD models may perform well in accurately estimating total landfill CH₄ emissions relative to TD measurements and the CEA method. Recent analysis of emissions data for Sampson County and Charlotte Motor Speedway in Balasus et al. (2024) corroborates this, as TD estimates aligned with GHGRP self-reported rates based on the FOD method for these landfills. This may not be the case for every landfill — the FOD method may underestimate total emissions, or TD methods may overestimate emissions (e.g., Balasus et al., 2024).

Deploying aircraft to acquire snapshot observations of CH₄ emissions from landfills once or repeatedly can be costly and logistically complex. Satellites address some concern related to survey frequency and possibly costs but are affected by spatial resolution, higher detection limits, cloud cover, and the accuracy of emissions priors if the data are used in an inversion framework to estimate an annual emissions rate (e.g., Nesser et al., 2024). As such, there is value in calculating landfill CH₄ emissions using models, similar to the calculations used by operators in reporting to national GHGRPs. Currently,

models used to estimate CH₄ emissions from landfills have known issues, such as overestimating gas capture system efficiencies and soil oxidation rates (Nesser et al., 2024; Stark et al., 2024), and lack of accounting for fugitive emissions from construction or other infrastructure (Cusworth et al., 2024). The FOD method may perform better than the CEA method for estimating emissions, but landfill CH₄ generation can diverge from FOD principles — generation and thus emissions rates can be higher and sustained over longer periods (Spokas et al., 2015; Nesser et al., 2024). This underscores that measurement and modeling are complimentary efforts in accurately constraining estimates of total landfill CH₄ emissions. Empirical data should inform gas collection system efficiency coefficients in models, and measurements of fugitives should be integrated into models. More nuanced models for landfills do exist and should be used broadly, such as the California Landfill Methane Inventory Model (CALMIM), which was developed to better account for variability in soil cover oxidation rates depending on the cover type, weather, and climate (Bogner et al., 2011). Addressing these issues may narrow the mismatch between estimates reported across the studies, enhance the credibility of estimates of CH₄ emissions from landfills, and better inform targeted abatement and mitigation efforts.

3.2.1.2. *Wastewater*

We identified only three studies dedicated to characterizing CH₄ emissions from wastewater collection and treatment sources (Fries et al., 2018; Williams et al., 2022; Moore et al., 2023). It was more common for wastewater sources to be measured in campaigns targeting street-level leaks or a variety of sources (Chamberlain et al., 2016; Williams et al., 2018; Guha et al., 2020). The studies identified several biogenic CH₄ sources related to wastewater: manholes, sewers, pavement cracks, storm drains, a sewage vent pipe on a building, and wastewater treatment plants (WWTPs). There is a challenge in disambiguating some of these smaller point sources; stable carbon isotope 13 for CH₄ (δ^{13} C) — a common method used for attribution — can show mixed biogenic and natural gas source signatures (Chamberlain et al., 2016; Fries et al., 2018; Williams et al., 2022), likely due to the co-location of emitting natural gas and sewage pipelines under city streets. CH₄ enhancements attributed to flooded storm drains and sewers in Williams et al. (2018) were associated with rising urban water levels, suggesting that these emissions were intermittent. Guha et al. (2020) detected a CH₄ plume from an anaerobic digester at one WWTP in SFBA. It is unclear if this plume was fugitive or a vent.

The studies in the review generally indicate that wastewater collection and treatment do not dominate U.S. and Canadian cities' emissions profiles. For example, Moore et al. (2023) quantified CH₄ emissions from 63 WWTPs across the U.S. mid-Atlantic and California using a vehicle-based system and integrated Gaussian plume modeling combined with Bayesian inference. The mean TD rate derived from the estimates in the study was 28.4 kg CH₄/h, which is about 34 times lower than the mean CH₄ point-source emissions rate from 96 U.S. landfills quantified in Cusworth et al. (2024). Guha et al. (2020) estimated that WWTPs are responsible for 6% of total CH₄ emissions in aggregate in SFBA, and Fries et al. (2018) estimated that the Rumpke Landfill in Cincinnati emitted 37 million times more CH₄ in 2015 than all 84,000 manholes and sewer grates in the city. These examples illustrate that CH₄ emissions from wastewater collection and treatment in U.S. and Canadian cities may be small compared to other sources.

Two reports challenge these findings: First, Williams et al. (2022) measured three "super-emitting" manholes in Montreal. When scaled up to the city-level, they estimated that manholes (including sewers and storm drains) are the third largest CH₄ source in Montreal, following active and historic landfills. The super-emitting manholes measured in Williams et al. (2022) were close to or within historical landfills in Montreal and it is possible that landfill gas is migrating into these manholes and escaping to the atmosphere, or that the manhole within the landfill is associated with a permanent leachate or run-off management system. The overlapping source signatures of δ^{13} C for manholes and historic landfills reported in the study corroborate this hypothesis (Williams et al., 2022). This could have high-biased the city-level estimate and potentially misattributed the CH₄ to wastewater when the source origin was the landfill. Second, de Foy et al. (2023) estimated total CH₄ emissions from 61 cities and urban areas globally using TROPOMI and a 2D Gaussian fit method, including 19 U.S. cities and one Canadian

city (Toronto). The pairing of this measurement platform and method does not permit identification of individual sources of CH₄ emissions; however, de Foy et al. (2023) found that city-level emissions rates correlated with rates of untreated wastewater, inclusive of cities in high-income countries such as the U.S. and Canada. While this suggests that the collection and discharge of untreated wastewater may be an important source of emissions in cities in these countries, most studies in the review indicate that total city-level CH₄ emissions are dominantly related to natural gas and landfills (§3.1.2.).

There are opportunities to expand on emissions measurements of wastewater infrastructure. WWTPs with anerobic digesters — which are used to treat sludge and capture CH₄ for sale, on-site use, or destruction — merit further attention. The studies identified anerobic digestors as a potential notable source of fugitive CH₄ (Guha et al., 2020; Moore et al., 2023). Thus, using measurement as a tool to determine if this is a widespread issue and root cause may improve the reliability of rate estimates and inform emissions reductions from WWTPs. There are also contrasting results in characterizing CH₄ emissions from sewers; targeted study to reduce uncertainty in sewer emissions estimates and ranking of their importance to total city-level emissions is warranted. Last, other wastewater infrastructure with potential to emit CH₄ such as biosolids lagoons and lift and pumping stations are not well characterized; future research could address these knowledge gaps.

3.2.2. Natural gas

3.2.2.1. Street-level natural gas leaks

Of all natural gas sources studied in the articles (S1), natural gas distribution pipelines were a prominent research target, and most studies used vehicle systems to measure emissions from pipeline leaks in city streets (Phillips et al., 2013; Jackson et al., 2014; Gallagher et al., 2015; Lamb et al., 2015; Chamberlain et al., 2016; von Fischer et al., 2017; Sanchez et al., 2018; Weller et al., 2018; 2020; Ars et al., 2020; Keyes et al., 2020; Luetschwager et al., 2021; Xia et al., 2022; MacMullin & Rongère, 2023). U.S. east coast cities were emphasized geographically in the studies due to the higher prevalence of old, leak-prone cast-iron pipe in these cities. The number of leaks per kilometer of natural gas distribution pipeline in cities is closely related to material type; cities with more cast-iron pipe have leakier natural gas distribution pipeline networks than cities with low or no prevalence of cast-iron pipe (Jackson et al., 2014; Lamb et al., 2015; Chamberlain et al., 2016; Sanchez et al., 2018; Weller et al., 2020).

Vehicle-based surveys are one of the most accessible techniques to measure spatially distributed sources in cities, which may account for the relatively high number of studies reporting this approach. Other methods such as flux chambers, surface enclosures, tracer releases, and mapping with handheld CH₄ sensors were also used to measure distribution pipeline emissions (Lamb et al., 2015; Hendrick et al., 2016; Fries et al., 2018; Weller et al., 2018; Edwards et al., 2021). Measurements of ethane (C_2H_6) and $\delta^{13}C$ were common methods in the studies to attribute the emissions to natural gas, although the number of samples taken for attribution relative to the total number of detected plumes is often very limited (e.g., Phillips et al., 2013; Jackson et al., 2014; Gallagher et al., 2015; Chamberlain et al., 2016). Moreover, natural gas and wastewater sources can mix, which results in ambiguous chemical signatures (§3.2.1.2.). In general, there is significant uncertainty in attributing city-level estimates of leaks to different sources based on current methods.

Methods to attribute street-level natural gas leaks with vehicle-system surveys have relied mainly on indirect approaches. Several studies used plume persistence (detectable on 2 or more passes or at least one half of the passes) and width (< 160m) as methods to attribute emissions detected at the surface to underground natural gas distribution pipeline leaks (von Fischer et al., 2017; Weller et al., 2018; 2020; Ars et al., 2020; Xia et al., 2022). The premise of these criteria is that natural gas plumes are narrow and persistent because distribution pipeline pressures remain constant. However, there are several issues with these criteria. First, the plume length threshold (< 160m) was based on a controlled release testing (CRT) experiment in an open field (von Fischer et al., 2017). Turbulence and plume dispersion in an open field are very different than on a busy city street where wind is modified by buildings and moving vehicles. Second, using persistence as a metric assumes that there are no other natural gas sources that could be emitting continuously. For example, leaks from natural gas service lines running up the sides of buildings

or customer meters (e.g., Vollrath et al., 2024a) could be persistent sources. Third, the general focus of research on natural gas distribution pipeline leaks has narrowly framed the problem along city streets. There are many other potential natural gas sources adjacent to streets — such as customer meters, valve vaults, farm tap units, vents from heaters and boilers, and industrial sources — that may be emitting but have been poorly characterized or not characterized at all.

In most cases, street-level CH₄ emissions detected with vehicle-based systems are remotely diagnosed and interpreted as natural gas distribution pipeline leaks from inside the vehicle or during post-processing; there is limited evidence of direct follow-up to confirm the source in the studies. There are potentially hundreds to thousands of plumes from different and unknown sources in cities that are only detected on a single vehicle pass but remain unattributed by measurement. Persistence is problematic because detection, particularly for sources adjacent to roadways, is strongly modulated by wind conditions; if the wind direction shifts between passes or surveys, the source may be undetectable even though the emission persists. A tiered research approach may be more effective, where a vehicle is used to screen for emissions and portable methods (i.e., sniffer) are used with follow-up to identify the source (e.g., Defratyka et al., 2021). Simultaneous measurements of CH₄ and C₂H₆ could support disambiguation for sources like cracked pavement or sewers when the origin (natural gas or a sewage pipeline) is not immediately obvious (e.g., Chamberlain et al., 2016; Hendrick et al., 2016; Fries et al., 2018). While the tiered approach to measurement likely consumes more resources, it should yield a more detailed understanding of emissions sources required to guide abatement.

3.2.2.2. Natural gas distribution facilities

Three studies examined CH₄ emissions rates from natural gas distribution facilities such as city-gate and meter and regulator (M&R) stations (Lamb et al., 2015; Hugenholtz et al., 2021; Williams et al., 2022). Lamb et al. (2015) used a hi-flow sampler and downwind tracer releases at 229 facilities operated by 13 different gas utility companies across the U.S. They found significant reductions in CH₄ emissions relative to Harrison et al. (1996), which acquired measurements from several of the same facilities in a 1992 campaign. Lamb et al. (2015) attributed these reductions to equipment replacements, facility rebuilds, and greater identification and repair of leaks. Overall, nation-wide estimates of emissions from M&R facilities in Lamb et al. (2015) were 7 to 13 times smaller than EPA estimates. Hugenholtz et al. (2021) mapped CH₄ concentrations at the fenceline of 33 distribution facilities in Calgary, adapting a version of the EPA's Other Test Method (OTM) 33A to quantify emissions downwind at six of the facilities. Based on concentration maps, wind direction, and mercaptan odor, 22 out of the 33 facilities had a high probability of emitting. In Montreal, Williams et al. (2022) used flux chambers to measure CH₄ emissions from sources at 27 individual facilities. These measurements were performed across six unique facility subtypes including branch, gate, predetention, district regulating, block valve, and telemetry stations. Many of these facility subtypes can be categorized as M&R stations (e.g., Lamb et al., 2015), but the differences between these facilities, their processes, and equipment are not well documented in the literature.

The duration of the measurement campaigns reported in these studies spanned May to November 2013 for Lamb et al. (2015), May to June 2020 for Hugenholtz et al. (2021), and August 2019 to March 2021 for Williams et al. (2022). It is unclear from Williams et al. (2022) if any measurements were performed at distribution facilities in winter because the duration of the field campaign was reported coarsely and spanned 3 years; actual measurement dates were not reported. Larger natural gas distribution facilities like city-gate stations and M&R facilities are known to have line heaters that vent burner emissions, which may include slip and other CH₄ emissions. These are presumably more active during colder periods. This may affect the emissions profile of these facilities, where emissions may be higher in winter. Emissions factors (EFs) derived from the measurements in these studies are used in national BU estimates (e.g., Lamb et al., 2015). EFs may underestimate total emissions if seasonality is not captured. Winter sampling at distribution facilities could improve knowledge of emissions sources and rates at different periods of the year and enable more accurate assessment of their seasonal contribution to total city-level CH₄ emissions.

 3.2.2.3. Natural gas combustion

Despite our estimates based on the data in several studies suggesting that natural gas combustion slip is a potentially large source of CH₄ emissions at the city-level (§3.1.2., Lamb et al., 2016; He et al., 2019; Sargent et al., 2021; Karion et al., 2023; Zeng et al., 2023), we found that specific sources of slip emissions are not yet clear based on source-level studies in the review. Slip emissions may occur during the pressure regulation and distribution of natural gas or downstream of customer gas meters (Vollrath et al., 2024a) and may vary between cities according to differences in the age of infrastructure and end uses.

Only a handful of studies examined CH₄ emissions from residential end-use, finding that pilot lights and operating gas-fired appliances such as furnaces, water heaters, and stoves emit nontrivial amounts of CH₄ (Fischer et al., 2018; Merrin and Francisco, 2019; Lebel et al., 2020). Saint-Vincent and Pekney (2020) integrated data from Fischer et al. (2018) and Merrin and Francisco (2019) to estimate that whole house and major appliance emissions could amount to 144.4 Gg CH₄/yr emitted across the U.S. residential sector, accounting for $\sim 2\%$ of total U.S. natural gas emissions. While this suggests residential slip emissions are small in the context of other, mostly non-urban natural gas sources, our calculations in §3.1.2 indicate that slip could be a much more important source. For example, if slip contributes approximately 32.5% and 47.1% to total CH₄ emissions in Los Angeles (He et al., 2019; Zeng et al., 2023) and Boston (Sargent et al., 2021), respectively, then on the basis of mass slip emissions would total 127.9 Gg CH₄/yr and 93.2 Gg CH₄/yr in these cities. Combined, this estimate of slip emissions from two U.S. cities only is 1.5 times larger than Saint-Vincent and Pekney's (2020) estimate of total U.S. residential slip emissions. Similarly, Fischer et al. (2018) scaled up measurements of CH₄ emissions from pilot lights and operating appliances in 75 California homes to estimate that 35.7 Gg CH₄/yr is emitted from residential natural gas use across the state. Our calculation for potential slip emissions in Los Angeles alone is 2.6 times larger. We note that these are not direct comparisons, as the estimates for CH₄ emissions that correlate with metered natural gas use in Los Angeles and Boston include emissions from the commercial and industrial (He et al., 2019; Zeng et al., 2023) and commercial (Sargent et al., 2021) sectors in addition to residential. However, unless commercial and industrial slip dominate total slip emissions in cities, emissions from residential slip may be largely underestimated across the U.S.

Higher natural gas use in the residential and commercial sectors in winter that overlaps with elevated city-level CH₄ emissions rates may signal that the majority of slip emissions in cities is tied to using natural gas for space or water heating, cooking, or other purposes in these sectors (e.g., see He et al., 2019 SI). Commercial and industrial end-use sources were not targeted in the studies, although passive CH₄ detections from a light rail train suggest these types of sources are emitting (Mitchell et al., 2018). Further investigation of end use sources is warranted. Studies in other cities globally can inform future research targets such as compressed natural gas vehicles (Hu et al., 2018; Pan et al., 2020) and heating, cooking, and other CH₄ emissions from festivals and events (Chen et al., 2020), among others.

3.3. Mitigation

- Mitigation was a very small component of the 103 studies reviewed but an important extension of measurements to guide and verify CH₄ emissions reductions. Generally, studies identified targets for mitigation (Duren et al., 2019; Guha et al., 2020; Cusworth et al., 2020b, 2024), assessed mitigation effectiveness by comparing emissions measurements between regions with and without mitigation programs (von Fischer et al., 2017), or directly compared measurements at different levels before and after mitigation was applied (Lamb et al., 2015; Fries et al., 2018; Kuwayama et al., 2019; Cusworth et al., 2020a, 2020b; Edwards et al., 2021; Yadav et al., 2023).
- *3.3.1. Landfills*
- Landfill management practices and organic diversion were highlighted in a handful of studies. Cusworth et al. (2020b) detected very large CH₄ emissions from a landfill in Los Angeles, which were attributed to a specific management practice. The landfill operator was placing several inches of daily cover overtop of

fresh waste to minimize odor complaints from residents downwind, but this practice — which did not include peel-back before new waste was added — created pockets of CH₄ that the landfill would intermittently "burp" into the atmosphere. Cusworth et al. (2020b) notified the landfill operator of the emissions and reductions were verified with follow up aerial measurements after the operator changed the practice and improved gas capture from this area of the landfill. The reductions achieved at this landfill were also verified in regional-level measurements (Cusworth et al., 2020a). These examples emphasize that measurement is a powerful tool to identify mitigation opportunities and verify emissions reductions.

In California, policies to separate and divert organic waste from landfills may also be diverting the associated CH₄ emissions to alternative locations. Cusworth et al. (2020b) and Guha et al. (2020) reported large CH₄ plumes from dry digestion and open-air composting facilities, attributing the plumes to an exhaust system, organic waste bags, and curing compost. The studies noted that composting facilities are currently excluded from state or federal emissions self-reporting and that these sources are missing from BU inventories (Cusworth et al., 2020b; Guha et al., 2020). Whether organic waste diversion is reducing or shifting CH₄ emissions is an outstanding research question.

3.3.2. Wastewater

Mitigation of wastewater CH₄ emissions was largely unaddressed in the studies compared to landfills or natural gas. This could be due to the comparatively low emissions relative to city-level totals (e.g., Fries et al., 2018; Guha et al., 2020) or the poor scalability of current solutions to reduce CH₄ emissions from urban wastewater networks (Williams et al., 2022). Anaerobic digesters at WWTPs were identified as notable emissions sources in aerial and vehicle-based system surveys across California and other areas of the U.S. (Duren et al., 2019; Guha et al., 2020; Moore et al., 2023). These emissions reduce the mitigation effectiveness of the digesters but to what extent is unclear.

3.3.3. Natural gas

3.3.3.1. Natural gas distribution pipelines

Several studies that used vehicle-based systems to measure street-level leaks in cities suggested that replacing leak-prone, cast-iron segments of the natural gas distribution pipeline network is effective in reducing emissions. This is a focus of mitigation efforts in U.S. east coast cities given their age and high prevalence of cast iron pipelines. von Fischer et al. (2017) found that cities in the U.S. with advanced pipeline replacement programs (APRPs) had leak rates that were 95% lower (0.08 L/min/km) compared to cities without these programs (2.0 L/min/km). In Toronto, ON, Ars et al. (2020) noted very few leaks per km of road surveyed, attributing this to a program (completed in 2012) that replaced 1800 km of cast-iron and steel pipeline. While these examples suggest pipeline replacement programs reduce emissions, other studies in the review indicate different results. For example, Edwards et al. (2021) measured 61 distribution pipeline leaks in Boston before and within a year after repair, finding that 15 repairs eliminated the emissions, 22 partially eliminated the emissions, and 24 were ineffective. Edwards et al. (2021) attributed the low repair success rate to temporary fixes, "super-failures" over time at the same location, or utilities only resolving one of multiple potential co-located leaks. The varying effectiveness of pipeline replacements reported across the studies indicates that follow up measurements to verify CH₄ emissions reductions may be needed.

3.3.3.2. Other natural gas sources

Other examples include post-mitigation measurements of CH₄ emissions following equipment replacement, upgrades, and facility rebuilds at city-gate and M&R stations across the U.S. (Lamb et al., 2015). In three cases, aerial measurements led directly to mitigation of CH₄ emissions from natural gas distribution pipelines and a liquified natural gas storage tank by notifying operators of the emissions (Duren et al., 2019). Last, Yadav et al. (2023) partly attributed a declining regional-level trend in CH₄ emissions in the Los Angeles Basin from 2015 to 2020 to leak detection and repair (LDAR) at O&G sites in the region; however, the exact sources mitigated are unclear based on the scale of measurement. Notably, mitigating CH₄ emissions from combustion slip was not featured in the studies, but potential

approaches could include appliance efficiency improvements, retrofits or replacements, and end-use fuel switching.

4. Conclusions and Recommendations

912

913

914 915

916

917

918

919 920

921

922 923

924

925

926 927

928 929

930 931

932

933 934

935

936

937

938 939

940

941

942

943 944

945

946 947

948

949

950

951

952

953

954

955 956

957 958

959 960

961

This work reviewed 103 studies of CH₄ emissions from cities in the U.S. and Canada to consolidate the current state of knowledge and highlight key research priorities. The review found that estimates of citylevel CH₄ emissions derived from TD measurements were dominantly larger than estimates from BU inventories. The extent of the discrepancy varied regionally, which is likely related to regional-based differences between cities in the age and material type of natural gas distribution infrastructure and overall importance of emissions from natural gas sources to city-level CH₄ emissions. The largest discrepancies were generally reported in satellite-based TD studies, suggesting that the extent of the discrepancy may be sensitive to the measurement method. As noted by Vollrath et al. (2024b) for upstream O&G methane emissions, TD > BU is not an issue unique to the urban space, but likely indicates that existing BU approaches are not capturing all the sources in cities and that the emissions and activity factors are not representative. Therefore, a refocusing of research to improve BU approaches to emissions estimation is warranted. Targeted measurements to expand source coverage and reduce assumptions, new and accessible datasets for researchers, and development of new urban-specific modeling approaches are required. Priority should be given to natural gas sources given that the studies showed the largest discrepancies between TD and BU estimates for this source category and their importance to total CH₄ emissions in most U.S. and Canadian cities.

While the studies indicated that natural gas is the dominant source of CH₄ emissions from U.S. and Canadian cities, on average, they also revealed that the specific sources are not well understood. This makes it challenging to develop policies, regulations, and mitigation strategies with a precision approach. Three key findings from the review point to a research need to develop a better resolved understanding of specific sources and root causes of emissions from natural gas in cities. First, the average measurementderived CH₄ loss rate for the urban areas studied $(1.8\% \pm 0.9\%)$ contradicts the O&G industry's claims of low-carbon differentiated gas, raising questions about industry estimation methods. The O&G industry cannot control CH₄ leakage downstream of customer meters, but nontrivial slip emissions at the burner tip are consequential for estimates of the total supply chain CH₄ intensity of natural gas and the climate. Second, analyses in the studies and our estimates based on the studies' emissions data point to end-use slip as a potential major contributor to urban CH₄ emissions, but attributing the emissions to specific end uses (e.g., furnaces, boilers, appliances, gas turbines, and industrial processes) is impeded by a very limited number of emissions rate measurements. Third, despite numerous studies, the characterization of natural gas leaks from pipelines is largely speculative, because the attribution methods are imprecise. Research targeting these and related knowledge gaps has potential to inform mitigation and improve the climate performance of natural gas.

Landfill studies included in the review indicate that TD estimates of CH₄ emissions are generally much larger than those self-reported to government GHGRPs. In the U.S., low GHGRP self-reported rates for landfills relative to TD estimates can mainly be tied to the CEA calculation method preferred by operators for reporting, which is systematically under-estimating emissions. The studies highlighted that the main issues with this method are overestimated gas capture system efficiencies and soil oxidation rates. Methods to calculate CH₄ emissions from landfills have value owing to the cost of snapshot observations of emissions with aircraft, satellites, or other measurement methods, but calculation methods are also missing emissions from fugitive point-sources within landfills at present. Thus, closing the gap between TD estimates and GHGRP self-reported CH₄ rates for landfills broadly will require advances in CH₄ emissions modeling approaches to address current shortcomings and the need to integrate empirical measurement data. We note that models to estimate CH₄ emissions from landfills have improved considerably (e.g., CALMIM) and should be used more widely for reporting. These models should be adapted to incorporate new parameters and measurement data based on research findings and insights, like the fugitive emissions highlighted in several studies or measurement-based estimates of gas capture

system efficiencies. Overall, a more integrative approach to landfill CH₄ measurements and emissions modeling is recommended.

Mitigation has not been a major focus of research on urban CH₄ emissions to date, although several key studies in the review indicate a strong connection between emissions measurement and mitigation through identification of CH₄ mitigation opportunities and verification of mitigation effectiveness. Regulations may provide impetus for expanding the scope of measurement and mitigation research through increased requirements around measurement-based monitoring, mitigation actions, reporting, and verification. Key research priorities include emissions from organic waste diversion, fugitive emissions from landfill cover systems and gas capture, and targeted studies of programs and technologies for reducing CH₄ emissions from the natural gas distribution system and end-use slip.

It must be recognized that stakeholder collaboration will be required to address many of the research priorities outlined here, particularly those involving more granular investigations of emissions sources, causes, and mitigation. This includes public and private organizations that may be hesitant to exchange data and information, grant site access, and who may oppose public-facing dissemination of research findings. In some cases, negotiating agreements with stakeholders may consume valuable research time and stymie progress. Thus, successful strategies to engage and manage stakeholders are crucial to enable the research and maintain momentum. These efforts should parallel the development of independent urban research projects that can be completed quicker and with less potential for bias.

Data Availability Statement

Supplementary Information

Additional details on the data extraction and preparation for Figures 2 and 4, list of studies in the review that measured CH₄ emissions from natural gas sources in cities by major natural gas source type, full citation list of all 103 articles reviewed, and summaries of evidence in each of the 103 studies reviewed.

Supplementary Data

962

963

964

965

966

967

968

969 970

971

972 973

974

975

976 977

978

979

980 981

982 983

984

985

986 987

988

989

990

991

992

993

994 995

996

997

998 999

1000 1001

1002 1003 1004 Parameters extracted from each article: author, article title, publication year, journal, first author institution, city or urban area studied, measurement technology/platform, CH₄ quantification method, attribution method, CH₄ emissions sources, emissions rates, urban natural gas loss rates, discrepancies between CH₄ emissions estimates, reported source signatures, and area of spatial domain. Individual sheets with more detailed data on emissions rates or source signatures are also included for 29 studies in addition to the main sheet with data from every study.

Acknowledgements

This project was undertaken with the financial support of the Government of Canada. Ce project a été réalisé avec l'appui financier du gouvernement du Canada. Additional financial support was provided by a doctoral scholarship from the Natural Sciences and Engineering Research Council (NSERC) of Canada. These organizations were not involved in this research in any capacity.

Competing Interests

The authors declare that they have no competing interests.

References

Alvarez, R. A., Zavala-Araiza, D., Lyon, D. R., Allen, D. T., Barkley, Z. R., Brandt, A. R., ... Hamburg, S. 1005 P. (2018). Assessment of methane emissions from the U.S. oil and gas supply chain. *Science*, 361(6398), 1006 1007 186–188. https://doi.org/10.1126/science.aar7204

1008 Anderson, D. C., Lindsay, A., DeCarlo, P. F., & Wood, E. C. (2021). Urban emissions of nitrogen oxides, carbon monoxide, and methane determined from ground-based measurements in 1009 1010

Philadelphia. Environmental Science & Technology, 55(8), 4532-4541.

- 1011 Ars, S., Vogel, F., Arrowsmith, C., Heerah, S., Knuckey, E., Lavoie, J., ... & Wunch, D. (2020).
- 1012 Investigation of the spatial distribution of methane sources in the greater Toronto area using mobile gas 1013 monitoring systems. Environmental science & technology, 54(24), 15671-15679.
- 1014 Balashov, N. V., Davis, K. J., Miles, N. L., Lauvaux, T., Richardson, S. J., Barkley, Z. R., & Bonin, T. A.
- 1015 (2020). Background heterogeneity and other uncertainties in estimating urban methane flux: results
- from the Indianapolis Flux Experiment (INFLUX). Atmospheric Chemistry and Physics, 20(7), 4545-1016 4559. https://doi.org/10.5194/acp-20-4545-2020 1017
- Balasus, N., Jacob, D. J., Maxemin, G., Jenks, C., Nesser, H., Maasakkers, J. D., ... & Wang, X. (2024). 1018
- 1019 Satellite monitoring of annual US landfill methane emissions and trends. arXiv preprint 1020 arXiv:2408.10957.
- Bogner, J., Spokas, K. A., & Chanton, J. P. (2011). Seasonal greenhouse gas emissions (methane, carbon 1021
- 1022 dioxide, nitrous oxide) from engineered landfills: Daily, intermediate, and final California cover soils. 1023 Journal of Environmental Quality, 40(3), 1010–1020. https://doi.org/10.2134/jeq2010.0407
- Cambaliza, M. O. L., Bogner, J. E., Green, R. B., Shepson, P. B., Harvey, T. A., Spokas, K. A., ... 1024
- Corcoran, M. (2017). Field measurements and modeling to resolve m2 to km2 CH4 emissions for a 1025 complex urban source: An Indiana landfill study. Elementa (Washington, D.C.), 5. 1026
- https://doi.org/10.1525/elementa.145 1027
- 1028 Cambaliza, M. O., Shepson, P. B., Bogner, J. E. A. N., Caulton, D. R., Stirm, B., Sweeney, C., ... &
- Richardson, S. (2015). Quantification and source apportionment of the methane emission flux from the 1029 1030 city of Indianapolis. Elementa, 3, 000037.
- Cambaliza, M. O. L., Shepson, P. B., Caulton, D. R., Stirm, B., Samarov, D., Gurney, K. R., ... 1031
- Richardson, S. J. (2014). Assessment of uncertainties of an aircraft-based mass balance approach for 1032
- 1033 quantifying urban greenhouse gas emissions. Atmospheric Chemistry and Physics, 14(17), 9029–9050.
- https://doi.org/10.5194/acp-14-9029-2014 1034
- Carranza, V., Rafiq, T., Frausto-Vicencio, I., Hopkins, F. M., Verhulst, K. R., Rao, P., ... Miller, C. E. 1035
- (2018). Vista-LA: Mapping methane-emitting infrastructure in the Los Angeles megacity. Earth System 1036 Science Data, 10(1), 653-676. https://doi.org/10.5194/essd-10-653-2018 1037
- 1038 Chamberlain, S. D., Ingraffea, A. R., & Sparks, J. P. (2016). Sourcing methane and carbon dioxide
- emissions from a small city: Influence of natural gas leakage and combustion. Environmental 1039 1040 pollution, 218, 102-110.
- 1041 Chan, E., Vogel, F., Smyth, S., Barrigar, O., Ishizawa, M., Kim, J., ... Worthy, D. E. J. (2024). Hybrid
- bottom-up and top-down framework resolves discrepancies in Canada's oil and gas methane 1042
- 1043 inventories. Communications Earth & Environment, 5(1), 566-10. https://doi.org/10.1038/s43247-024-1044 01728-6
- Chen, J., Dietrich, F., Maazallahi, H., Forstmaier, A., Winkler, D., Hofmann, M. E., ... & Röckmann, T. 1045
- 1046 (2020). Methane emissions from the Munich Oktoberfest. Atmospheric Chemistry and Physics, 20(6), 1047 3683-3696.
- 1048 Covidence (2022). Covidence systematic review software. Veritas Health Innovation.
- https://www.covidence.org/ 1049
- Crippa, M., Guizzardi, D., Pagani, F., Banja, M., Muntean, M., Schaaf E., Becker, W., Monforti-Ferrario, 1050
- F., Quadrelli, R., Risquez Martin, A., Taghavi-Moharamli, P., Köykkä, J., Grassi, G., Rossi, S., Brandao 1051
- 1052 De Melo, J., Oom, D., Branco, A., San-Miguel, J., Vignati, E., GHG emissions of all world countries,
- 1053 Publications Office of the European Union, Luxembourg, 2023, doi:10.2760/953322, JRC134504.
- Cui, Y. Y., Vijayan, A., Falk, M., Hsu, Y.-K., Yin, D., Chen, X. M., ... Croes, B. (2019). A Multiplatform 1054
- Inversion Estimation of Statewide and Regional Methane Emissions in California during 2014–2016. 1055
- Environmental Science & Technology, 53(16), 9636–9645. https://doi.org/10.1021/acs.est.9b01769 1056
- 1057 Cui, Y. Y., Brioude, J., McKeen, S. A., Angevine, W. M., Kim, S.-W., Frost, G. J., ... Trainer, M. (2015).
- Top-down estimate of methane emissions in California using a mesoscale inverse modeling technique: 1058
- The South Coast Air Basin. Journal of Geophysical Research. Atmospheres, 120(13), 6698-6711. 1059
- https://doi.org/10.1002/2014JD023002 1060

- Cusworth, D. H., Duren, R. M., Ayasse, A. K., Jiorle, R., Howell, K., Aubrey, A., ... & Thorneloe, S. (2024). Quantifying methane emissions from United States landfills. *Science*, 383(6690), 1499-1504.
- Cusworth, D. H., Duren, R. M., Yadav, V., Thorpe, A. K., Verhulst, K., Sander, S., ... Miller, C. E.
- 1064 (2020a). Synthesis of Methane Observations Across Scales: Strategies for Deploying a Multitiered Observing Network. *Geophysical Research Letters*, 47(7). https://doi.org/10.1029/2020GL087869
- Cusworth, D. H., Duren, R. M., Thorpe, A. K., Tseng, E., Thompson, D., Guha, A., ... Miller, C. E.
- 1067 (2020b). Using remote sensing to detect, validate, and quantify methane emissions from California solid waste operations. *Environmental Research Letters*, *15*(5), 54012-. https://doi.org/10.1088/1748-1069 9326/ab7b99
- 1070 Czepiel, P. M., Shorter, J. H., Mosher, B., Allwine, E., McManus, J. B., Harriss, R. C., ... Lamb, B. K. (2003). The influence of atmospheric pressure on landfill methane emissions. *Waste Management* (Elmsford), 23(7), 593–598. https://doi.org/10.1016/S0956-053X(03)00103-X
- de Foy, B., Schauer, J. J., Lorente, A., & Borsdorff, T. (2023). Investigating high methane emissions from
 urban areas detected by TROPOMI and their association with untreated wastewater. *Environmental Research Letters*, 18(4), 044004.
- Defratyka, S. M., Paris, J. D., Yver-Kwok, C., Fernandez, J. M., Korben, P., & Bousquet, P. (2021).
- Mapping urban methane sources in Paris, France. *Environmental science & technology*, *55*(13), 8583-1078 8591.
- Dogniaux, M., Maasakkers, J. D., Girard, M., Jervis, D., McKeever, J., Schuit, B. J., ... & Aben, I. (2024).
 Satellite survey sheds new light on global solid waste methane emissions. *Earth ArXiv pre-print*.
 https://doi.org/10.31223/X5TB09
- Duren, R. M., Thorpe, A. K., Foster, K. T., Rafiq, T., Hopkins, F. M., Yadav, V., ... & Miller, C. E. (2019).
 California's methane super-emitters. *Nature*, 575(7781), 180-184
- Edwards, M. R., Giang, A., Macey, G. P., Magavi, Z., Nicholas, D., Ackley, R., & Schulman, A. (2021).
 Repair Failures Call for New Policies to Tackle Leaky Natural Gas Distribution Systems.
- 1086 Environmental Science & Technology, 55(10), 6561–6570. https://doi.org/10.1021/acs.est.0c07531
- 1087 Environment and Climate Change Canada (ECCC). (2024, May 2). Additional NIR Annexes:
- Methodology for Waste Sector. https://data-donnees.az.ec.gc.ca/data/substances/monitor/canada-s-official-greenhouse-gas-inventory/G-Additional-NIR-Annexes?lang=en
- Environment and Climate Change Canada (ECCC). (2023). Facility Greenhouse Gas Reporting:
 Technical Guidance on Reporting Greenhouse Gas Emissions.
- publications.gc.ca/collections/collection 2023/eccc/En81-29-2023-eng.pdf
- European Commission. (2022). Global Methane Pledge: From Moment to Momentum.
- $1094 \qquad https://energy.ec.europa.eu/system/files/2022-11/GMP\%20COP27\%20Joint\%20Factsheet.pdf$
- Fairley, D., & Fischer, M. L. (2015). Top-down methane emissions estimates for the San Francisco Bay Area from 1990 to 2012. *Atmospheric Environment*, 107, 9-15.
- Feng, L., Palmer, P. I., Parker, R. J., Lunt, M. F., & Bösch, H. (2023). Methane emissions are predominantly responsible for record-breaking atmospheric methane growth rates in 2020 and 2021. *Atmospheric Chemistry and Physics*, *23*(8), 4863-4880.
- Fernandez, J. M., Maazallahi, H., France, J. L., Menoud, M., Corbu, M., Ardelean, M., ... & Röckmann,
 T. (2022). Street-level methane emissions of Bucharest, Romania and the dominance of urban
 wastewater. *Atmospheric Environment: X*, 13, 100153.
- Fischer, M. L., Chan, W. R., Delp, W., Jeong, S., Rapp, V., & Zhu, Z. (2018). An Estimate of Natural Gas
 Methane Emissions from California Homes. *Environmental Science & Technology*, 52(17), 10205–
- 1105 10213. https://doi.org/10.1021/acs.est.8b03217
- Floerchinger, C., Shepson, P. B., Hajny, K., Daube, B. C., Stirm, B. H., Sweeney, C., & Wofsy, S. C.
- 1107 (2021). Relative flux measurements of biogenic and natural gas-derived methane for seven US cities. *Elem Sci Anth*, *9*(1), 000119.
- Frankenberg, C., Thorpe, A. K., Thompson, D. R., Hulley, G., Kort, E. A., Vance, N., ... Green, R. O.
- 1110 (2016). Airborne methane remote measurements reveal heavy-tail flux distribution in Four Corners

- region. Proceedings of the National Academy of Sciences PNAS, 113(35), 9734–9739.
- 1112 https://doi.org/10.1073/pnas.1605617113
- 1113 Fries, A. E., Schifman, L. A., Shuster, W. D., & Townsend-Small, A. (2018). Street-level emissions of
- methane and nitrous oxide from the wastewater collection system in Cincinnati, Ohio. *Environmental*
- 1115 *Pollution (1987)*, 236, 247–256. https://doi.org/10.1016/j.envpol.2018.01.076
- 1116 Gallagher, M. E., Down, A., Ackley, R. C., Zhao, K., Phillips, N., & Jackson, R. B. (2015). Natural gas
- pipeline replacement programs reduce methane leaks and improve consumer safety. *Environmental Science & Technology Letters*, *2*(10), 286-291.
- Global Methane Pledge (n.d.) https://www.globalmethanepledge.org/.
- Goldsmith, Jr. C. D., Chanton, J., Abichou, T., Swan, N., Green, R., & Hater, G. (2012). Methane
- emissions from 20 landfills across the United States using vertical radial plume mapping. *Journal of the Air & Waste Management Association*, 62(2), 183–197. https://doi.org/10.1080/10473289.2011.639480
- Government of Canada. (2023a, December 16). Regulations Amending the Regulations Respecting
- Reduction in the Release of Methane and Certain Volatile Organic Compounds (Upstream Oil and Gas
- Sector). https://www.gazette.gc.ca/rp-pr/p1/2023/2023-12-16/html/reg3-eng.html
- Government of Canada. (2023b, June 14). Reducing Canada's landfill methane emissions: Proposed
- 1127 regulatory framework. https://www.canada.ca/en/environment-climate-change/services/canadian-
- environmental-protection-act-registry/publications/reducing-landfill-methane-emissions.html
- 1129 Guha, A., Newman, S., Fairley, D., Dinh, T. M., Duca, L., Conley, S. C., ... Martien, P. T. (2020).
- 1130 Assessment of Regional Methane Emission Inventories through Airborne Quantification in the San
- 1131 Francisco Bay Area. Environmental Science & Technology, 54(15), 9254–9264.
- https://doi.org/10.1021/acs.est.0c01212
- 1133 Harrison, M., T. Shires, J. Wessels, AND M. Cowgill. METHANE EMISSIONS FROM THE NATURAL
- GAS INDUSTRY VOLUME 1: EXECUTIVE SUMMARY. U.S. Environmental Protection Agency,
- 1135 Washington, D.C., EPA/600/R-96/080a (NTIS PB97-142921), 1996.
- He, L., Zeng, Z., Pongetti, T. J., Wong, C., Liang, J., Gurney, K. R., ... Sander, S. P. (2019). Atmospheric
- Methane Emissions Correlate With Natural Gas Consumption From Residential and Commercial
- 1138 Sectors in Los Angeles. *Geophysical Research Letters*, 46(14), 8563–8571.
- 1139 https://doi.org/10.1029/2019GL083400
- 1140 Hedelius, J. K., Liu, J., Oda, T., Maksyutov, S., Roehl, C. M., Iraci, L. T., ... Wennberg, P. O. (2018).
- 1141 Southern California Megacity CO2, CH4, and CO Flux Estimates Using Ground- and Space-Based
- Remote Sensing and a Lagrangian Model. *Atmospheric Chemistry and Physics*, 18(22), 16271–16291.
- 1143 https://doi.org/10.5194/acp-18-16271-2018
- Heimburger, A. M. F., Harvey, R. M., Shepson, P. B., Stirm, B. H., Gore, C., Turnbull, J., ... Gurney, K.
- 1145 R. (2017). Assessing the optimized precision of the aircraft mass balance method for measurement of
- urban greenhouse gas emission rates through averaging. Elementa (Washington, D.C.), 5.
- 1147 https://doi.org/10.1525/elementa.134
- Hemati, M., Mahdianpari, M., Nassar, R., Shiri, H., & Mohammadimanesh, F. (2024). Urban methane
- emission monitoring across North America using TROPOMI data: an analytical inversion
- approach. Scientific Reports, 14(1), 9041.
- Hendrick, M. F., Ackley, R., Sanaie-Movahed, B., Tang, X., & Phillips, N. G. (2016). Fugitive methane
- emissions from leak-prone natural gas distribution infrastructure in urban environments. *Environmental Pollution*, *213*, 710-716.
- Hopkins, F. M., Ehleringer, J. R., Bush, S. E., Duren, R. M., Miller, C. E., Lai, C. T., ... & Randerson, J. T.
- 1155 (2016a). Mitigation of methane emissions in cities: How new measurements and partnerships can
- contribute to emissions reduction strategies. *Earth's Future*, 4(9), 408-425.
- Hopkins, F. M., Kort, E. A., Bush, S. E., Ehleringer, J. R., Lai, C., Blake, D. R., & Randerson, J. T.
- 1158 (2016b). Spatial patterns and source attribution of urban methane in the Los Angeles Basin. *Journal of*
- Geophysical Research. Atmospheres, 121(5), 2490–2507. https://doi.org/10.1002/2015JD024429
- Hsu, Y. K., VanCuren, T., Park, S., Jakober, C., Herner, J., FitzGibbon, M., ... & Parrish, D. D. (2010).
- Methane emissions inventory verification in southern California. *Atmospheric Environment*, 44(1), 1-7.

- Hu, N., Liu, S., Gao, Y., Xu, J., Zhang, X., Zhang, Z., & Lee, X. (2018). Large methane emissions from natural gas vehicles in Chinese cities. *Atmospheric Environment*, *187*, 374-380.
- Huang, Y., Kort, E. A., Gourdji, S., Karion, A., Mueller, K., & Ware, J. (2019). Seasonally Resolved
- Excess Urban Methane Emissions from the Baltimore/Washington, DC Metropolitan Region.
- Environmental Science & Technology, 53(19), 11285–11293. https://doi.org/10.1021/acs.est.9b02782
- Hugenholtz, C. H., Vollrath, C., Gough, T., Wearmouth, C., Fox, T., Barchyn, T., & Billinghurst, C.
- 1168 (2021). Methane emissions from above-ground natural gas distribution facilities in the urban
- environment: A fence line methodology and case study in Calgary, Alberta, Canada. *Journal of the Air*4 & Waste Management Association (1995), 71(11), 1319–1332.
- https://doi.org/10.1080/10962247.2021.1942316
- 1172 IPCC, 2023: Summary for Policymakers. In: Climate Change 2023: Synthesis Report. Contribution of
- Working Groups I, II and III to the Sixth Assessment Report of the Intergovernmental Panel on Climate
- 1174 Change [Core Writing Team, H. Lee and J. Romero (eds.)]. IPCC, Geneva, Switzerland, pp. 1-34, doi: 10.59327/IPCC/AR6-9789291691647.001
- 1176 Jackson, R. B., Down, A., Phillips, N. G., Ackley, R. C., Cook, C. W., Plata, D. L., & Zhao, K. (2014).
- Natural gas pipeline leaks across Washington, DC. *Environmental science & technology*, 48(3), 2051-1178 2058.
- Jacob, D. J., Varon, D. J., Cusworth, D. H., Dennison, P. E., Frankenberg, C., Gautam, R., ... & Duren, R.
 M. (2022). Quantifying methane emissions from the global scale down to point sources using satellite
- observations of atmospheric methane. *Atmospheric Chemistry and Physics*, 22(14), 9617-9646.
- 1182 Jeong, S., Cui, X., Blake, D. R., Miller, B., Montzka, S. A., Andrews, A., ... Fischer, M. L. (2017).
- Estimating methane emissions from biological and fossil-fuel sources in the San Francisco Bay Area.
- 1184 Geophysical Research Letters, 44(1), 486–495. https://doi.org/10.1002/2016GL071794
- Jeong, S., Newman, S., Zhang, J., Andrews, A. E., Bianco, L., Bagley, J., ... Fischer, M. L. (2016).
- Estimating methane emissions in California's urban and rural regions using multitower observations.
- Journal of Geophysical Research. Atmospheres, 121(21), 13,031-13,049.
- 1188 https://doi.org/10.1002/2016JD025404
- Jeong, S., Hsu, Y.-K., Andrews, A. E., Bianco, L., Vaca, P., Wilczak, J. M., & Fischer, M. L. (2013). A
- multitower measurement network estimate of California's methane emissions. *Journal of Geophysical Research*. *Atmospheres*, *118*(19), 11,339-11,351. https://doi.org/10.1002/jgrd.50854
- 1192 Jones, T. S., Franklin, J. E., Chen, J., Dietrich, F., Hajny, K. D., Paetzold, J. C., ... Wofsy, S. C. (2021).
- Assessing urban methane emissions using column-observing portable Fourier transform infrared (FTIR)
- spectrometers and a novel Bayesian inversion framework. *Atmospheric Chemistry and Physics*, 21(17), 13131–13147. https://doi.org/10.5194/acp-21-13131-2021
- 1196 Karion, A., Ghosh, S., Lopez-Coto, I., Mueller, K., Gourdji, S., Pitt, J., & Whetstone, J. (2023). Methane
- Emissions Show Recent Decline but Strong Seasonality in Two US Northeastern Cities. *Environmental Science & Technology*.
- 1199 Keyes, T., Ridge, G., Klein, M., Phillips, N., Ackley, R., & Yang, Y. (2020). An enhanced procedure for urban mobile methane leak detection. *Heliyon*, *6*(10).
- 1201 Kuwayama, T., Charrier-Klobas, J. G., Chen, Y., Vizenor, N. M., Blake, D. R., Pongetti, T., ... Herner, J.
- D. (2019). Source Apportionment of Ambient Methane Enhancements in Los Angeles, California, To
- Evaluate Emission Inventory Estimates. *Environmental Science & Technology*, 53(6), 2961–2970.
- 1204 https://doi.org/10.1021/acs.est.8b02307
- Lamb, B. K., Cambaliza, M. O. L., Davis, K. J., Edburg, S. L., Ferrara, T. W., Floerchinger, C., ...
- Whetstone, J. (2016). Direct and Indirect Measurements and Modeling of Methane Emissions in
- 1207 Indianapolis, Indiana. Environmental Science & Technology, 50(16), 8910–8917.
- 1208 https://doi.org/10.1021/acs.est.6b01198
- Lamb, B. K., Edburg, S. L., Ferrara, T. W., Howard, T., Harrison, M. R., Kolb, C. E., ... & Whetstone, J.
- R. (2015). Direct measurements show decreasing methane emissions from natural gas local distribution
- systems in the United States. *Environmental Science & Technology*, 49(8), 5161-5169.

- Lebel, E. D., Lu, H. S., Speizer, S. A., Finnegan, C. J., & Jackson, R. B. (2020). Quantifying Methane
- Emissions from Natural Gas Water Heaters. *Environmental Science & Technology*, 54(9), 5737–5745.
- 1214 https://doi.org/10.1021/acs.est.9b07189
- Lopez-Coto, I., Ren, X., Salmon, O. E., Karion, A., Shepson, P. B., Dickerson, R. R., ... & Whetstone, J.
- R. (2020). Wintertime CO2, CH4, and CO emissions estimation for the Washington, DC-Baltimore
- metropolitan area using an inverse modeling technique. *Environmental science & technology*, *54*(5), 2606-2614.
- 1219 Luetschwager, E., von Fischer J. C., & Weller, Z. D. (2021). Characterizing detection probabilities of
- advanced mobile leak surveys: Implications for sampling effort and leak size estimation in natural gas
- distribution systems. *Elementa*, 9(1). https://doi.org/10.1525/elementa.2020.00143
- Maasakkers, J. D., Jacob, D. J., Sulprizio, M. P., Turner, A. J., Weitz, M., Wirth, T., ... & Fischer, M. L.
- 1223 (2016). Gridded national inventory of US methane emissions. *Environmental science & technology*, *50*(23), 13123-13133.
- 1225 MacKay, K., Seymour, S. P., Li, H. Z., Zavala-Araiza, D., & Xie, D. (2024). A Comprehensive Integration
- and Synthesis of Methane Emissions from Canada's Oil and Gas Value Chain. *Environmental Science & Technology*, *58*(32), 14203–14213. https://doi.org/10.1021/acs.est.4c03651
- MacMullin, S., & Rongère, F. X. (2023). Measurement-based emissions assessment and reduction
- through accelerated detection and repair of large leaks in a gas distribution network. *Atmospheric Environment: X*, 17, 100201.
- Marcotullio, P. J., Sarzynski, A., Albrecht, J., Schulz, N., & Garcia, J. (2013). The geography of global
- urban greenhouse gas emissions: An exploratory analysis. *Climatic Change*, 121, 621-634.
- 1233 McKain, K., Down, A., Raciti, S. M., Budney, J., Hutyra, L. R., Floerchinger, C., ... & Wofsy, S. C.
- 1234 (2015). Methane emissions from natural gas infrastructure and use in the urban region of Boston,
- Massachusetts. *Proceedings of the National Academy of Sciences*, 112(7), 1941-1946.
- 1236 Merrin, Z., & Francisco, P. W. (2019). Unburned Methane Emissions from Residential Natural Gas
- 1237 Appliances. Environmental Science & Technology, 53(9), 5473–5482.
- 1238 https://doi.org/10.1021/acs.est.8b05323
- 1239 Mitchell, L. E., Crosman, E. T., Jacques, A. A., Fasoli, B., Leclair-Marzolf, L., Horel, J., ... Lin, J. C.
- 1240 (2018). Monitoring of greenhouse gases and pollutants across an urban area using a light-rail public
- transit platform. Atmospheric Environment (1994), 187(C), 9–23.
- 1242 https://doi.org/10.1016/j.atmosenv.2018.05.044
- 1243 Mittakola, R. T., Ciais, P., Schubert, J. E., Makowski, D., Zhou, C., Bazzi, H., ... Davis, S. J. (2024).
- Drivers of natural gas use in U.S. residential buildings. *Science Advances*, 10(14), eadh5543–eadh5543. https://doi.org/10.1126/sciadv.adh5543
- Moore, D. P., Li, N. P., Wendt, L. P., Castañeda, S. R., Falinski, M. M., Zhu, J.-J., ... Zondlo, M. A.
- 1247 (2023). Underestimation of Sector-Wide Methane Emissions from United States Wastewater Treatment.
- 1248 Environmental Science & Technology, 57(10), 4082–4090. https://doi.org/10.1021/acs.est.2c05373
- 1249 Nesser, H., Jacob, D. J., Maasakkers, J. D., Lorente, A., Chen, Z., Lu, X., ... & Randles, C. A. (2024).
- High-resolution US methane emissions inferred from an inversion of 2019 TROPOMI satellite data:
- contributions from individual states, urban areas, and landfills. *Atmospheric Chemistry and Physics*,
- 24(8), 5069-5091. https://doi.org/10.5194/acp-24-5069-2024
- Ocko, I. B., Sun, T., Shindell, D., Oppenheimer, M., Hristov, A. N., Pacala, S. W., ... & Hamburg, S. P.
- 1254 (2021). Acting rapidly to deploy readily available methane mitigation measures by sector can
- immediately slow global warming. *Environmental Research Letters*, 16(5), 054042.
- Oil and Gas Climate Initiative (OGCI). *Methane emissions*. https://www.ogci.com/methane-emissions
- One Future. (2023). 2023 Annual Report on Calendar Year 2022 Methane Intensities.
- https://onefuture.us/2023-annual-report/
- 1259 Pak, N. M., Heerah, S., Zhang, J., Chan, E., Worthy, D., Vogel, F., & Wunch, D. (2021). The Facility
- 1260 Level and Area Methane Emissions inventory for the Greater Toronto Area (FLAME-
- 1261 GTA). *Atmospheric Environment*, 252, 118319.

- Pan, D., Tao, L., Sun, K., Golston, L. M., Miller, D. J., Zhu, T., ... & Zondlo, M. A. (2020). Methane emissions from natural gas vehicles in China. *Nature communications*, 11(1), 4588.
- 1264 Peischl, J., Ryerson, T. B., Brioude, J., Aikin, K. C., Andrews, A. E., Atlas, E., ... & Parrish, D. D. (2013).
- Quantifying sources of methane using light alkanes in the Los Angeles basin, California. *Journal of Geophysical Research: Atmospheres*, 118(10), 4974-4990.
- Phillips, N. G., Ackley, R., Crosson, E. R., Down, A., Hutyra, L. R., Brondfield, M., ... & Jackson, R. B.
- 1268 (2013). Mapping urban pipeline leaks: Methane leaks across Boston. *Environmental pollution*, 173, 1-4.
- 1269 Pitt, J. R., Lopez-Coto, I., Karion, A., Hajny, K. D., Tomlin, J., Kaeser, R., ... Shepson, P. B. (2024).
- Underestimation of Thermogenic Methane Emissions in New York City. *Environmental Science & Technology*, 58(21), 9147–9157. https://doi.org/10.1021/acs.est.3c10307
- 1272 Pitt, J. R., Lopez-Coto, I., Hajny, K. D., Tomlin, J., Kaeser, R., Jayarathne, T., ... Shepson, P. B. (2022).
- New York City greenhouse gas emissions estimated with inverse modeling of aircraft measurements.
- 1274 Elementa (Washington, D.C.), 10(1). https://doi.org/10.1525/elementa.2021.00082
- 1275 Plant, G., Kort, E. A., Murray, L. T., Maasakkers, J. D., & Aben, I. (2022). Evaluating urban methane
- emissions from space using TROPOMI methane and carbon monoxide observations. *Remote Sensing of Environment*, 268, 112756.
- 1278 Plant, G., Kort, E. A., Floerchinger, C., Gvakharia, A., Vimont, I., & Sweeney, C. (2019). Large fugitive
- methane emissions from urban centers along the US East Coast. *Geophysical research letters*, 46(14), 8500-8507.
- 1281 Ren, X., Salmon, O. E., Hansford, J. R., Ahn, D., Hall, D., Benish, S. E., ... & Dickerson, R. R. (2018).
- Methane emissions from the Baltimore-Washington area based on airborne observations: Comparison to emissions inventories. *Journal of Geophysical Research: Atmospheres*, 123(16), 8869-8882.
- Rohatgi, A. (2022). WebPlotDigitizer. v4.6. https://automeris.io/WebPlotDigitizer/
- 1285 Ryoo, J.-M., Iraci, L. T., Tanaka, T., Marrero, J. E., Yates, E. L., Fung, I., ... Chang, C. S. (2019).
- Quantification of CO2 and CH4 Emissions over Sacramento, California, Based On Divergence
- Theorem Using Aircraft Measurements. *Atmospheric Measurement Techniques*, *12*(5), 2949–2966. https://doi.org/10.5194/amt-12-2949-2019
- Saint-Vincent, P. M. B., & Pekney, N. J. (2020). Beyond-the-Meter: Unaccounted Sources of Methane
- Emissions in the Natural Gas Distribution Sector. *Environmental Science & Technology*, *54*(1), 39–49. https://doi.org/10.1021/acs.est.9b04657
- 1292 Sanchez, N. P., Zheng, C., Ye, W., Czader, B., Cohan, D. S., Tittel, F. K., & Griffin, R. J. (2018).
- Exploratory study of atmospheric methane enhancements derived from natural gas use in the Houston urban area. *Atmospheric Environment*, *176*, 261-273.
- Sargent, M. R., Floerchinger, C., McKain, K., Budney, J., Gottlieb, E. W., Hutyra, L. R., ... & Wofsy, S. C.
- 1296 (2021). Majority of US urban natural gas emissions unaccounted for in inventories. *Proceedings of the National Academy of Sciences*, *118*(44), e2105804118.
- 1298 Saunois, M., Stavert, A. R., Poulter, B., Bousquet, P., Canadell, J. G., Jackson, R. B., ... & Zhuang, Q.
- 1299 (2020). The global methane budget 2000–2017. *Earth system science data*, 12(3), 1561-1623.
- Spokas, K., Bogner, J., Corcoran, M., & Walker, S. (2015). From California dreaming to California data:
- 1301 Challenging historic models for landfill CH4 emissions. *Elementa (Washington, D.C.)*, 3.
- 1302 https://doi.org/10.12952/journal.elementa.000051
- Stark, B. M., Tian, K., & Krause, M. J. (2024). Investigation of US landfill GHG reporting program methane emission models. *Waste Management*, *186*, 86-93.
- Thompson, D. R., Thorpe, A. K., Frankenberg, C., Green, R. O., Duren, R., Guanter, L., ... Ungar, S.
- 1306 (2016). Space-Based Remote Imaging Spectroscopy of the Aliso Canyon CH4 Superemitter.
- 1307 Geophysical Research Letters, 43(12), 6571–6578. https://doi.org/10.1002/2016GL069079
- 1308 United Nations Environment Programme (UNEP) and Climate and Clean Air Coalition (CCAC). (2022).
- Global Methane Assessment: 2030 Baseline Report Summary for Policymakers.
- https://www.ccacoalition.org/sites/default/files/resources//2022 Methane-baseline-summary 0.pdf
- U.S. Environmental Protection Agency (EPA). (2023a, December 18). EPA's Final Rule for Oil and
- Natural Gas Operations Will Sharply Reduce Methane and Other Harmful Pollution.

- https://www.epa.gov/controlling-air-pollution-oil-and-natural-gas-operations/epas-final-rule-oil-and-natural-gas
- U.S. Environmental Protection Agency (EPA). (2023b, March 29). Municipal Solid Waste Landfills: New
- Source Performance Standards (NSPS), Emission Guidelines (EG) and Compliance Times.
- https://www.epa.gov/stationary-sources-air-pollution/municipal-solid-waste-landfills-new-sourceperformance-standards
- U.S. Environmental Protection Agency (EPA). (2009, October 30). 40 CFR Part 98 Subpart HH:
 Municipal Solid Waste Landfills. https://www.ecfr.gov/current/title-40/part-98/subpart-HH
- Vogel, F., Ars, S., Wunch, D., Lavoie, J., Gillespie, L., Maazallahi, H., ... & Calcan, A. (2024). Ground-
- Based Mobile Measurements to Track Urban Methane Emissions from Natural Gas in 12 Cities across Eight Countries. *Environmental Science & Technology*, *58*(5), 2271-2281.
- Vollrath, C., Hugenholtz, C. H., Barchyn, T. E., & Wearmouth, C. (2024a). Methane emissions from residential natural gas meter set assemblies. *Science of The Total Environment*, 172857.
- 1326 https://doi.org/10.1016/j.scitotenv.2024.172857.
- Vollrath, C., Hugenholtz, C. H., & Barchyn, T. E. (2024b). Onshore methane emissions measurements from the oil and gas industry: A scoping review. *Environmental Research Communications*.
- 1329 DOI 10.1088/2515-7620/ad3129.
- von Fischer, J. C., Cooley, D., Chamberlain, S., Gaylord, A., Griebenow, C. J., Hamburg, S. P., ... & Ham,
- J. (2017). Rapid, vehicle-based identification of location and magnitude of urban natural gas pipeline leaks. *Environmental Science & Technology*, *51*(7), 4091-4099.
- Weller, Z. D., Hamburg, S. P., & von Fischer, J. C. (2020). A National Estimate of Methane Leakage from
- Pipeline Mains in Natural Gas Local Distribution Systems. *Environmental Science & Technology*,
- 1335 *54*(14), 8958–8967. https://doi.org/10.1021/acs.est.0c00437
- Weller, Z. D., Roscioli, J. R., Daube, W. C., Lamb, B. K., Ferrara, T. W., Brewer, P. E., & von Fischer, J.
- 1337 C. (2018). Vehicle-Based Methane Surveys for Finding Natural Gas Leaks and Estimating Their Size:
- Validation and Uncertainty. *Environmental Science & Technology*, 52(20), 11922–11930.
- 1339 https://doi.org/10.1021/acs.est.8b03135
- 1340 Wennberg, P. O., Mui, W., Wunch, D., Kort, E. A., Blake, D. R., Atlas, E. L., ... & Fischer, M. L. (2012).
- On the sources of methane to the Los Angeles atmosphere. *Environmental science &*
- 1342 *technology*, 46(17), 9282-9289.
- Williams, J. P., Ars, S., Vogel, F., Regehr, A., & Kang, M. (2022). Differentiating and Mitigating Methane
- Emissions from Fugitive Leaks from Natural Gas Distribution, Historic Landfills, and Manholes in
- 1345 Montréal, Canada. Environmental Science & Technology, 56(23), 16686–16694.
- 1346 https://doi.org/10.1021/acs.est.2c06254
- Williams, P. J., Reeder, M., Pekney, N. J., Risk, D., Osborne, J., & McCawley, M. (2018). Atmospheric
- impacts of a natural gas development within the urban context of Morgantown, West Virginia. *The*
- 1349 *Science of the Total Environment*, 639, 406–416. https://doi.org/10.1016/j.scitotenv.2018.04.422
- 1350 Wong, C. K., Pongetti, T. J., Oda, T., Rao, P., Gurney, K. R., Newman, S., ... Sander, S. P. (2016).
- Monthly trends of methane emissions in Los Angeles from 2011 to 2015 inferred by CLARS-FTS
- observations. *Atmospheric Chemistry and Physics*, *16*(20), 13121–13130. https://doi.org/10.5194/acp-1353 16-13121-2016
- 1354 Wong, K. W., Fu, D., Pongetti, T. J., Newman, S., Kort, E. A., Duren, R., ... Sander, S. P. (2015).
- Mapping CH4:CO2 ratios in Los Angeles with CLARS-FTS from Mount Wilson, California.
- 1356 Atmospheric Chemistry and Physics, 15(1), 241–252. https://doi.org/10.5194/acp-15-241-2015
- Wunch, D., Toon, G. C., Hedelius, J. K., Vizenor, N., Roehl, C. M., Saad, K. M., ... & Wennberg, P. O.
- 1358 (2016). Quantifying the loss of processed natural gas within California's South Coast Air Basin using
- long-term measurements of ethane and methane. *Atmospheric Chemistry and Physics*, *16*(22), 14091-1360 14105.
- Wunch, D., Wennberg, P. O., Toon, G. C., Keppel-Aleks, G., & Yavin, Y. G. (2009). Emissions of
- greenhouse gases from a North American megacity. *Geophysical research letters*, 36(15).

- Xia, T., Raneses, J., & Batterman, S. (2022). Improving the performance of pipeline leak detection algorithms for the mobile monitoring of methane leaks. *Atmosphere*, *13*(7), 1043.
- 1365 Xing, Z., Barchyn, T. E., Vollrath, C., Gao, M., & Hugenholtz, C. (2024). Satellite-Derived Estimate of
- 1366 City-Level Methane Emissions from Calgary, Alberta, Canada. *Remote Sensing*, *16*(7), 1149.

 1367 Xu, L., Lin, X., Amen, J., Welding, K., & McDermitt, D. (2014). Impact of changes in barometric
- Xu, L., Lin, X., Amen, J., Welding, K., & McDermitt, D. (2014). Impact of changes in barometric
 pressure on landfill methane emission. *Global Biogeochemical Cycles*, 28(7), 679–695.
 https://doi.org/10.1002/2013GB004571
- Yadav, V., Verhulst, K., Duren, R., Thorpe, A., Kim, J., Keeling, R., ... Whetstone, J. (2023). A declining trend of methane emissions in the Los Angeles basin from 2015 to 2020. *Environmental Research Letters*, 18(3), 34004-. https://doi.org/10.1088/1748-9326/acb6a9
- Yadav, V., Duren, R., Mueller, K., Verhulst, K. R., Nehrkorn, T., Kim, J., ... Miller, C. (2019). Spatio-temporally Resolved Methane Fluxes From the Los Angeles Megacity. *Journal of Geophysical Research*. *Atmospheres*, 124(9), 5131–5148. https://doi.org/10.1029/2018JD030062
- Zeng, Z. C., Pongetti, T., Newman, S., Oda, T., Gurney, K., Palmer, P. I., ... & Sander, S. P. (2023).
 Decadal decrease in Los Angeles methane emissions is much smaller than bottom-up estimates. *Nature Communications*, 14(1), 5353.
- Zhang, S., Ma, J., Zhang, X., & Guo, C. (2023). Atmospheric remote sensing for anthropogenic methane
 emissions: Applications and research opportunities. *The Science of the Total Environment*, 893,
 164701–164701. https://doi.org/10.1016/j.scitotenv.2023.164701.