Company: Southern California Gas Company (U 904 G)
Application: A.25-10-008, Proposing Woody Biomass Pilot Project
Exhibit: SCG-06

Sierra Club Response to SoCalGas's Data Request 2

APPLICATION OF SOUTHERN CALIFORNIA GAS COMPANY
PROPOSING WOODY BIOMASS PILOT PROJECT

BEFORE THE PUBLIC UTILITIES COMMISSION

OF THE STATE OF CALIFORNIA

California Public Utilities Commission Docket No. A.25-10-008

Application of Southern California Gas Company (U 904 G) Proposing Woody Biomass Pilot Project

Sierra Club Response to Data Request SoCalGas-Sierra Club-02

To:	Ismael Bautista, Jr., on behalf of Southern California Gas Company ("SoCalGas")
From:	Nina Robertson, Earthjustice, on behalf of Sierra Club
Date Request Sent:	March 19, 2026
Response Due:	April 2, 2026

QUESTION 1:

Explain in detail Dr. Emily Grubert's experience with the design or operation of chemical plants or petrochemical grade equipment.

RESPONSE 1:

Dr. Grubert has worked as a consultant on projects related to process safety for petroleum refineries and related facilities. She has not worked as a design or operations engineer on chemical plants or petrochemical-grade equipment.

QUESTION 2:

Explain in detail Dr. Emily Grubert's experience with Leak Detection and Repair (LDAR) protocols and industry practices for chemical plant equipment.

RESPONSE 2:

Dr. Grubert has experience with LDAR protocols and industry practices for natural gas pipelines. She does not have direct experience with LDAR protocols and industry practices for chemical plant equipment.

QUESTION 3:

Explain in detail Dr. Emily Grubert's familiarity with U.S. Environmental Protection Agency (EPA) methodologies for calculating leakage and leakage risk from petrochemical plant equipment.

RESPONSE 3:

Dr. Grubert is familiar with U.S. EPA methodologies for calculating leakage and leakage risk from petrochemical plant equipment in an academic capacity and has published peer-reviewed research comparing U.S. EPA estimates to other estimates used in evaluating methane emissions. *See* Grubert, E., and A. R. Brandt. 2019. "Three considerations for modeling natural gas system methane emissions in life cycle assessment." Journal of Cleaner Production, 222: 760–767. https://doi.org/10.1016/j.jclepro.2019.03.096, which is attached hereto as Attachment 1.

QUESTION 4:

Explain whether any of the projects referenced in page 2 of the Testimony of Dr. Emily Grubert when responding to the question, "Are you familiar with other projects that have intentionally produced methane from biogenic sources?" involved thermochemical conversion of biomass to Bio-SNG as is proposed in the PILOT PROJECT. If so, please provide previous analysis and all assumptions as to methane leakage and supporting documents.

RESPONSE 4:

Of the several cases for which Dr. Grubert has testified in other states that have involved the intentional production of methane from biogenic sources, none have successfully implemented thermochemical conversion of biomass to Bio-SNG, and none have had 0% lifecycle methane emissions validated.

QUESTION 5:

In reference to the question "Let's first discuss the first basis for your concern. Please elaborate on why it is your expert opinion that SoCalGas's CI scores are misrepresented as validated estimates" on page 6 of the Testimony of Dr. Emily Grubert,

QUESTION 5a:

Is it possible for the GREET methodology to be applied without using the actual spreadsheet provided by Argonne National Laboratory as long as the same mathematical calculations are made?

RESPONSE 5a:

Sierra Club objects on the grounds this question is vague and ambiguous as to the term "applied."

Subject to and without waiving the foregoing objection, Sierra Club answers as follows:

No, for two reasons. First, replicating calculations externally to mimic a methodology would not appropriately be described as "using the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation ("GREET") model[,]" as claimed in the corrected testimony of James Lucas and Matthew Summers at JLMS-13, and as asserted in the November 20, 2025 memo of Eric C. D. Tan to Matthew D. Summers, included as Attachment 1 to the corrected testimony, which claims "[t]he Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model was used for the analysis" and cites "Greenhouse gases, Regulated Emissions, and Energy use in Technologies Model ® (2024 Excel) Available: https://doi.org/10.11578/GREET-Excel-2024/dc.20241203.1."

Second, while the GREET model has been validated extensively, replicative calculations have not been. This lack of validation reduces confidence in the outputs when the model itself is not being used. Thus, application of "the GREET methodology . . . without using the actual spreadsheet provided by Argonne National Laboratory" does not ensure accurate results.

QUESTION 5b:

Are there specific mathematical differences found between the calculations shown on the spreadsheet provided by SoCalGas and the calculations on the Argonne National Laboratory provided GREET spreadsheets?

RESPONSE 5b:

Sierra Club objects on the grounds this question is ambiguous and assumes facts not in evidence. It is unclear what SoCalGas means by the "Argonne National Laboratory provided GREET spreadsheets." The "Argonne National Laboratory provided GREET spreadsheets," were not made available in response to Sierra Club's request seeking the GREET documents used to generate the proposed project's carbon intensities. The corrected testimony of James Lucas and Matthew Summers references GREET on JLMS-13 and also "CA-GREET," which is provided by the California Air Resources Board and not Argonne National Labs. The spreadsheet provided by SoCalGas to Sierra Club references "R&D GREET 2024" as the source for some, but not all, data.

Subject to and without waiving the foregoing objection, Sierra Club responds as follows:

The evidence available shows that significant mathematical differences are likely, although SoCalGas has not provided enough information to clearly demonstrate specific mathematical differences.

A main source for the calculations shown on the spreadsheet provided by SoCalGas, and a main driver of the conclusion that the calculations are not GREET outputs, is Culumber et al. 2025, which was published 1 September 2025 and first appeared online on 8 April 2025. *See* https://www.sciencedirect.com/science/article/pii/S0167880925001963?via%3Dihub ("Received 18 October 2024, Revised 23 March 2025, Accepted 25 March 2025, Available online 8 April 2025, Version of Record 8 April 2025.") R&D GREET 2024 was released in 2024, and CA-GREET 4.0 has been active since 1 July 2025. *See* https://ww2.arb.ca.gov/sites/default/files/classic/fuels/lcfs/ca-greet/cagreet4_documentation_07012025_2.pdf (dated July 1, 2025). There is no evidence that Culumber et al. 2025 was incorporated into CA-GREET 4.0 within those approximate two months from CA-GREET model documentation. Even if it had been, the way that data from Culumber et al. 2025 is included in the spreadsheet provided by SoCalGas is inconsistent with the treatment of biogenic CO2 elsewhere in the GREET model family. For instance, the "Summary of Expansions and Updates in R&D GREET® 2025," which is the current R&D model documentation, explicitly states on page 9:

> The fates of biogenic carbon in forest residue are tracked across multiple pools and fluxes: sequestration in humus/microbial biomass, sequestration in inert organic matter, remaining in residues, and biogenic CO2 emissions to atmosphere. A net biogenic carbon balance is reported. Two carbon-accounting approaches biogenic CO2 emissions are implemented: (1) a carbon-neutral approach and (2) an explicit accounting approach coupled with a time-dependent discounted Global Warming Potential (GWP). https://greet.anl.gov/publication-greet-2025-summary.

Numerous other inputs to the SoCalGas-provided non-GREET spreadsheet reference sources other than GREET, as well. This further increases the likelihood that there are mathematical differences.

QUESTION 6:

In reference to the question "Let's first discuss the first basis for your concern. Please elaborate on why it is your expert opinion that SoCalGas's CI scores are misrepresented as validated estimates" on page 6 of the Testimony of Dr. Emily Grubert, Dr Grubert states, "Although CA-GREET is primarily used for the implementation of the California's Low Carbon Fuel Standard, I would generally expect a GREET-based analysis for California compliance to be consistent with, or at least very clear on its lack of consistency with, the state-approved CI calculation model for fuels like biomethane,"

QUESTION 6a:

Would the result of a global warming potential analysis using CA-GREET vs. R&D GREET emissions factors be expected to lead to vastly different results or conclusions or is the methodology generally the same for these two models?

RESPONSE 6a:

Sierra Club objects on the grounds this question is vague and ambiguous and calls for speculation as to the terms "vastly different" and "generally the same."

Subject to and without waiving the foregoing objection, Sierra Club responds as follows:

Yes, there are potentially substantial differences in results between the two models, especially given that conclusions are based on ranking carbon intensities by pathway. The weighting of particular types of emissions (essentially, what a GWP does) could change the rank. Whether this occurs in this case depends on the share of methane emissions for the overall greenhouse gas footprint, which SoCalGas did not estimate.

QUESTION 7:

In reference to page 9 of the Testimony of Dr. Emily Grubert where she states, “Culumber et al. find that 4.05 Mg C/ha of the originally applied 61.6 Mg C/ha remain in the soil after years,”

QUESTION 7a:

What amount of years does the study say the 4.05 Mg C/ha is estimated to stay in the soil?

RESPONSE 7a:

The study projects 4.05 MgC/ha remain in the soil after 20 years.

QUESTION 7b:

Does the study estimate the amount of carbon that will remain in the soil after 100 years?

RESPONSE 7b:

No, the study does not provide this estimate.

QUESTION 8:

Provide all citations and calculations supporting the following statement in the Testimony of Dr. Emily Grubert on page 10: "…I note also that accounting for the 4.05 Mg C/ha that Culumber et al. 2025 estimate are still stored in soil after 20 years post-WOR application as a sequestration would mean that WOR has a CI of -101 g CO2/kg of applied biomass, contributing to net carbon dioxide removal rather than emissions for this practice."

RESPONSE 8:

See Response 2 to SoCalGas's first data request.

QUESTION 9:

In response to the question "Please summarize your concerns with SoCalGas's methodology for calculating the CI score of the Project" on page 11 of the Testimony of Dr. Emily Grubert, Dr. Emily Grubert states, "CI (e.g., through CA-GREET or a modified version of that model that has been vetted and approved by the CPUC)."

QUESTION 9a:

If CPUC has not vetted and approved a modified version of the GREET model, what reasons make R&D-GREET emissions factors invalid for the preliminary analysis of the Global Warming Potential impacts of this proposed facility?

RESPONSE 9a:

Sierra Club objects on the grounds that this question mischaracterizes the testimony of Dr. Grubert.

Subject to and without waiving the foregoing objection, Sierra Club responds as follows:

The testimony of Dr. Grubert does not assert that the California Public Utilities Commission ("CPUC") has not vetted and approved a modified version of the GREET model. The testimony of Dr. Grubert also does not state that using R&D-GREET emissions factors is invalid. The testimony of Dr. Grubert does assert that SoCalGas did not provide evidence that it used a validated GREET model at all, as is clear from the remainder of the sentence excerpted above, which reads in full:

> In my opinion, SoCalGas has not provided a CI score estimate that is consistent with California's validated approaches for calculating CI (e.g., through CA-GREET or a modified version of that model that has been vetted and approved by the CPUC), and as explained above, it appears to have misrepresented that its CI calculations—which have grave inconsistencies embedded—were done through a widely used, validated model (e.g. GREET).

QUESTION 10:

In reference to the study cited in footnote 21 of the Testimony of Dr. Emily Grubert on page 11,[1]

a. In this study, were any of the facilities studied using thermochemical conversion of biomass to Bio-SNG in a closed-loop system using refinery grade equipment?
b. In this study, were any of the facilities staffed 24/7 with technicians tasked with preventing leaks?
c. In this study, did any of the facilities use continuous leak detection or LDAR protocols?

RESPONSE 10:

Sierra Club objects to this request on the grounds that it calls for speculation.

Subject to and without waiving the foregoing objection, Sierra Club responds as follows:

a. Dr. Grubert did not perform the study and does not have complete knowledge of what facility practices were in place, including whether any used thermochemical conversion of biomass to Bio-SNG in a closed-loop system using refinery grade equipment. Dr. Grubert's testimony cites the study as evidence that methane emissions can be high at biogas facilities such that an assumption of 0 emissions is likely inaccurate.
b. Dr. Grubert did not perform the study and does not have complete knowledge of what facility practices were in place, including whether any of the facilities were staffed 24/7 with technicians tasked with preventing leaks. Relevant to the question, the study (attached hereto as Attachment 2) states on page 45 that "[w]ithin the last few years, this organisation has initiated a voluntary measurement programme with the aim of keeping CH4 loss at a minimum via a target of 1% loss for the sector."
c. Dr. Grubert did not perform the study and does not have complete knowledge of what facility practices were in place, including whether they use continuous leak detection or LDAR protocols. Relevant to the question, the study states on page 45 that "[w]ithin the last few years, this organisation has initiated a voluntary measurement programme with the aim of keeping CH4 loss at a minimum via a target of 1% loss for the sector." The study also states on page 38, "[t]he on-site approach measures emissions from various single sources at the plant, and it is the method most commonly used[,]" and explains on page 39:

> Recent measurement comparison studies have found that methods measuring the plant's total CH_4 emission often result in a higher emission rate in comparison to on-site measurements, where the total emission is obtained by summing up those measured from

[1] *Citing* Scheutz, Charlotte, and Anders M. Fredenslund. 2019. "Total Methane Emission Rates and Losses from 23 Biogas Plants." Waste Management 97 (September): 38–46.

single sources. The reason for the discrepancy between on-site and ground-based remote sensing approaches is most likely that single sources are overlooked and/or not identified, or they are technically not quantifiable when using a particular measurement technique (e.g. open tanks).

QUESTION 11:

In reference to footnote 22 of the Testimony of Dr. Emily Grubert on page 11,[2]

a. In this study or any studies referenced, were any of the facilities studied using thermochemical conversion of biomass to Bio-SNG in a closed-loop system using refinery grade equipment?
b. In this study or any studies referenced, were any of the facilities staffed 24/7 with technicians tasked with preventing leaks?
c. In this study or any studies referenced, did any of the facilities use continuous leak detection or LDAR protocols?

RESPONSE 11:

Sierra Club objects to this request on the grounds that it calls for speculation.

Subject to and without waiving the foregoing objection, Sierra Club responds as follows:

a. Dr. Grubert does not have complete knowledge of whether any of the facilities studied across every reference in the cited study used thermochemical conversion of biomass to Bio-SNG in a closed-loop system using refinery-grade equipment. Dr. Grubert's testimony cites the study as evidence that methane emissions can be high at biogas facilities such that an assumption of 0 emissions is likely inaccurate.
b. Dr. Grubert does not have complete knowledge of whether any of the facilities studied across every reference in the cited study were staffed 24/7 with technicians tasked with preventing leaks. Relevant to the question, and as explained in Response 10(b) above, one cited study (i.e., Scheutz, Charlotte, and Anders M. Fredenslund. 2019. "Total Methane Emission Rates and Losses from 23 Biogas Plants." Waste Management 97 (September): 38–46) states on page 45 that "[w]ithin the last few years, this organisation has initiated a voluntary measurement programme with the aim of keeping CH4 loss at a minimum via a target of 1% loss for the sector."
c. Dr. Grubert does not have complete knowledge of whether any of the facilities studied across every reference in the cited study use continuous leak detection or LDAR protocols. Relevant to the question, and as explained in Response 10(c) above, one cited study (i.e., Scheutz, Charlotte, and Anders M. Fredenslund. 2019. "Total Methane Emission Rates and Losses from 23 Biogas Plants." Waste Management 97 (September): 38–46) states on page 45 that "[w]ithin the last few years, this organisation has initiated a voluntary measurement programme with the aim of keeping CH4 loss at a minimum via

[2] Citing Grubert, Emily. 2020. "At Scale, Renewable Natural Gas Systems Could Be Climate Intensive: The Influence of Methane Feedstock and Leakage Rates." Environmental Research Letters 15 (8): 084041.

a target of 1% loss for the sector." The same study also states on page 38, "[t]he on-site approach measures emissions from various single sources at the plant, and it is the method most commonly used[,]" and explains on page 39:

> Recent measurement comparison studies have found that methods measuring the plant's total CH_4 emission often result in a higher emission rate in comparison to on-site measurements, where the total emission is obtained by summing up those measured from single sources. The reason for the discrepancy between on-site and ground-based remote sensing approaches is most likely that single sources are overlooked and/or not identified, or they are technically not quantifiable when using a particular measurement technique (e.g. open tanks).

QUESTION 12:

Provide all DOCUMENTS, including calculations, relied upon to support the response of Dr. Emily Grubert on page 12 of her testimony to the question, "Please describe how you think methane emissions of this Proposed Project should be evaluated."

RESPONSE 12:

The document is the open-access model provided in footnote 22 of Dr. Grubert's testimony. It is attached hereto as Attachment 3.

Attachment 1

Journal of Cleaner Production 222 (2019) 760–767

Contents lists available at ScienceDirect

Journal of Cleaner Production

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

Three considerations for modeling natural gas system methane emissions in life cycle assessment

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ARTICLE INFO

Article history:
Received 27 August 2018
Received in revised form
7 March 2019
Accepted 8 March 2019
Available online 13 March 2019

Keywords:
Methane
Inventory
Climate change
Carbon footprint
Leakage
Natural gas

ABSTRACT

Natural gas is a fossil fuel accounting for about 30% of US primary energy consumption. Climate change is one of the primary environmental issues associated with natural gas use: natural gas combustion releases carbon dioxide. A less emphasized issue is that natural gas is mostly methane, a potent greenhouse gas (GHG). The climate impact of natural gas use is thus sensitive to the amount of methane that escapes from the natural gas system unburned. We call attention to three considerations for modeling natural gas-related methane emissions in life cycle assessment (LCA). First, natural gas system methane leakage is inconsistently characterized and likely systematically underestimated by commonly used life cycle inventory (LCI) databases. Second, studies are often imprecise in assumptions about process boundaries. This matters because not all natural gas uses rely on the same infrastructure and induce the same methane leakage. Third, there is not yet a stable estimate for the global warming potential (GWP) of methane. Newer estimates tend to be larger, which further exacerbates the underestimation of GHG impacts from natural gas systems. Data uncertainty is common in LCA, but natural gas-related methane emissions deserve special attention due to their influence on a decision-relevant parameter (GHG intensity) in product systems across the economy.

1. Introduction

Natural gas accounts for about 30% of US primary energy consumption (EIA, 2018) and is widely used for electricity generation, heating, and industrial purposes. Given its prevalence, natural gas is part of the life cycle of a large number of product systems. Natural gas is primarily comprised of methane (CH_4), the second most significant GHG for anthropogenic climate change, after carbon dioxide (CO_2) (Weyant et al., 2006). Methane has a relatively high global warming potential (GWP): even a small amount of emitted methane can result in substantial carbon dioxide-equivalent (CO_2e) GHG emissions, one of the most commonly studied environmental indicators (Grubert, 2017). Questions about the amount of methane that escapes to the atmosphere unburned from natural gas systems, which we will call methane leakage, are thus highly relevant to environmental impact evaluations like life cycle assessment (LCA).

Despite recent studies on methane leakage from natural gas systems, most of which suggest that leakage is underestimated (Alvarez et al., 2018; Balcombe et al., 2018; Brandt et al., 2014; Farquharson et al., 2017; Heath et al., 2014; Littlefield et al., 2016; Sanchez and Mays, 2015; Zhang et al., 2014), little attention has been paid to the impact of methane leakage inventories on product carbon footprints. Natural gas is an indirect input to many products via electricity, heating, and chemical feedstock supply. As these inputs are typically background processes for product-specific LCAs, database defaults are often used. This ubiquity means that inaccurate methane leakage inventories are widely relevant to LCA and, especially given the small number of processes involved, should be priorities for LCA database improvement.

1.1. Methane leakage from natural gas systems

We will use the terms methane “leakage” or methane “emissions” interchangeably to refer to all methane emitted unburned from natural gas systems. Not all leakage is unintentional: for example, some safety systems are designed to release gas when a system becomes overpressured due to unusual operating conditions or equipment problems. Leakage is generally a combination of non-purposeful emissions (sometimes called fugitives) and

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https://doi.org/10.1016/j.jclepro.2019.03.096
0959-6526/

designed or purposeful emissions (sometimes called vents).

Researchers have been concerned for decades about methane leakage from natural gas systems and its influence on climate change (Lelieveld et al., 2005, 1993; Meier et al., 2005), with substantial disagreement about the value and dynamics of leakage rates (Kirchgessner et al., 1997). Data from outside North America are sparse (though see Yacovitch et al., 2018, a recent study from the Netherlands), neglecting possibly significant regional variability (Bouman et al., 2015; Gibon et al., 2017). For example, a study comparing GHG emissions from coal versus shale gas-fired power generation in China uses United States (US) emissions factors due to a lack of Chinese data (Chang et al., 2015). Recent literature has suggested that methane emissions might be systematically underestimated by official inventories, in part because a substantial portion of emissions is due to large, infrequent, and unintentional releases that can be difficult to detect (Brandt et al., 2016, 2014). The exact volume of leakage is not precisely understood (Alvarez et al., 2012; Brandt et al., 2014; Burnham et al., 2012; Hausfather, 2015, 2014; Heath et al., 2014; Jeong et al., 2014; Lelieveld et al., 2005, 1993) due to regional variability (Allen et al., 2013; Jiang et al., 2011; Karion et al., 2015, 2013; Pétron et al., 2014), operational practices, regulatory context, and other factors.

The energy analysis community has primarily focused on leakage while examining whether substituting natural gas for coal in the electricity sector will reduce overall climate change impacts (Farquharson et al., 2017; Gilbert and Sovacool, 2017; Hausfather, 2015; Howarth et al., 2011; Roy et al., 2015). Less attention has been paid to the relative GHG impacts of natural gas-versus electricity-based residential and commercial applications like heating and cooking. Although leakage from the natural gas distribution system has been studied (Costello, 2014; Hendrick et al., 2016; Jackson et al., 2014; McKain et al., 2015; Phillips et al., 2013), end use leakage from equipment, homes and businesses is rarely assessed (for an exception see Lavoie et al., 2017). This lack of study is particularly apparent for residential applications where methane leakage from appliances might be environmentally relevant in a comparison across heating options (Oliphant, 1994).

1.2. Three claims about methane leakage in LCA databases

In this paper we make three claims with respect to the treatment of methane in LCA databases.

First, commonly used inventory databases are inconsistent and likely systematically underestimate methane leakage from the natural gas system. Major ambiguities within and across inventory databases are common. We examine how existing databases model methane leakage from the natural gas system and quantify the relevance of natural gas system leakage rates to the GHG footprint of basic materials to demonstrate the scope of this problem. Our results show that methane leakage from natural gas systems should be a priority target for inventory improvements.

Second, databases (and individual studies) have unclear or incorrect assumptions about which natural gas system processes are applicable for a given supply chain. This is important, as it affects both how users might respond to leakage and what methane emissions are assigned to a given use of natural gas.

Third, there is not yet a stable estimate for the GWP of methane, affecting comparability of LCA studies over time. Newer estimates of methane's GWP tend to be larger. Analysts studying comparative energy systems should perform sensitivity analysis on the GWP and should report actual methane masses rather than CO_2e only, to allow forward compatibility and comparability of results.

The analysis below examines and supports these claims in order. We will focus on US methane leakage in LCA databases, though many of the same issues apply to carbon footprinting or environmental impact assessment studies.

2. Methods

2.1. Methane leakage in life cycle inventory databases

Given the vast amount of data needed for LCA and a desire for consistency across studies, LCA practitioners often use one of a few common life cycle inventory (LCI) databases. For major databases, we ask: 1) What do the databases assume about US natural gas system leakage rates? And 2), how significant is embodied natural gas-related methane leakage for GHG footprints of non-natural gas product systems?

2.1.1. Database assumptions

We examine leakage assumptions in several common LCI databases. Our primary goal is to illustrate current status and discrepancies rather than to provide a set of recommended values. We do, however, incorporate synthetic insights from the many recent studies that have estimated leakage outside the context of life cycle inventories (Abrahams et al., 2015; Allen et al., 2014, 2013; Alvarez et al., 2018; Balcombe et al., 2018; Brandt et al., 2014; Ge et al., 2016; Heath et al., 2014; Hendrick et al., 2016; Jeong et al., 2014; Jiang et al., 2011; Karion et al., 2015, 2013; Lavoie et al., 2017; Littlefield et al., 2016; Mitchell et al., 2015; Pétron et al., 2014; Phillips et al., 2013; Zimmerle et al., 2015).

We examine three databases, each of which is the most current version available:

1) Economic Input-Output Life Cycle Assessment (EIO-LCA) Model (Carnegie Mellon University Green Design Institute, 2008), 2002 Producer Price model;
2) The National Renewable Energy Laboratory (NREL) United States Life Cycle Inventory database (USLCI) (NREL, 2012); and
3) The ecoinvent database, version 3.5 (ecoinvent, 2018).

USLCI and EIO-LCA data are older and less frequently updated than ecoinvent data, but relevantly, these databases are free while ecoinvent is not. Thus, it is reasonable to expect that they remain in use. Some other common LCA tools base their US natural gas-related methane emissions on databases we assess, so these are implicitly assessed as well. For example, GREET uses EPA inventories and GaBi/Thinkstep uses the USLCI as source data.

In each case, we: 1) identify unit process data for US-based natural gas systems; 2) match available inventory processes with processes in the US natural gas system to evaluate coverage; 3) convert data from each database's inventory format to a mass of methane basis, using process-specific data on energy densities, pressure, and standard gas conditions; and 4) calculate methane emissions as a mass percentage of gross methane withdrawals for each included process.

To investigate the accuracy of LCI data, we compare them with three non-LCI leakage estimates: 1) a recent inventory for 2015 natural gas supply chain methane emissions based on facility-scale ground measurements and aircraft observations (Alvarez et al., 2018); 2) Environmental Protection Agency (EPA) greenhouse gas inventory (GHGI) estimates for 2013 (EPA, 2015); and 3) EPA GHGI estimates for 2015 (EPA, 2017). EPA GHGI mass emissions are converted to leakage rates using the method of Brandt et al. (2014). Year 2015 EPA GHGI results are the basis for many leakage parameters in Argonne National Laboratory's GREET model (Burnham, 2017), and these data reflect a recent change in calculation methodology for natural gas systems. EPA 2013 GHG data are also included because they are of a similar vintage to the USLCI (released in 2012) dataset.

2.1.2. Significance beyond natural gas systems

We are interested in LCI data on methane leakage from the natural gas system largely because of the hypothesis that these background data have a large influence on GHG footprint results beyond the natural gas system itself. To test this hypothesis, we investigate the contribution of natural gas system methane leakage to the GHG footprints of six illustrative materials: plastic, fertilizer, aluminum, steel, electricity, and cement. These materials were chosen because they are themselves common inputs for products throughout the economy. Demonstrating that natural gas system methane leakage matters for the GHG footprints of these materials thus indicates its relevance for GHG footprinting more generally.

2.2. The location of methane leakage

Not all processes that are part of the natural gas supply chain apply to all uses of natural gas. For example, leakage from the natural gas distribution system (a low-pressure network delivering natural gas to typically nonindustrial end users, like homes) does not in most cases affect the carbon intensity of natural gas-fired electricity from power plants. Most natural gas-fired power plants are supplied from the high-pressure transmission system either directly or via high pressure laterals (personal communication, Dynegy Investor Relations, March 2018). We illustrate the relevance of this system structure using natural gas consumption data from the Energy Information Administration (EIA) to provide guidelines on how natural gas end uses map to natural gas system subprocesses (EIA, 2017).

2.3. The influence of global warming potential on methane leakage impact

Use of different GWPs in the literature means that methane emissions reported on a CO_2e basis, even evaluated over the same time horizon, are not necessarily comparable. To illustrate the impact of this instability, we examine methane's GWP in the five Intergovernmental Panel on Climate Change (IPCC) Assessment Reports (the First, Second, Third, Fourth, and Fifth are referred to as FAR, SAR, TAR, AR4, and AR5) (Intergovernmental Panel on Climate Change, 2018). We do not address the separate question of which time horizon to use when comparing climate impacts of methane to carbon dioxide (Farquharson et al., 2017; Gilbert and Sovacool, 2017; Hausfather, 2015; Howarth et al., 2011; Roy et al., 2015).

3. Results and discussion

3.1. Methane leakage in life cycle inventories

3.1.1. Database assumptions

Table 1 summarizes the leakage assumptions in studied LCI databases. The supplementary material (SM) contains calculation details.

For each life cycle stage, leakage is reported as a cumulative mass percentage of gross withdrawals of the methane that reaches each stage. That is, a value of 1.4% under "distribution" means that for natural gas in the distribution system, life cycle methane emissions to that point are 1.4% of the mass of the gross withdrawals of methane that reached the distribution system, not that 1.4% of methane is lost at the distribution stage or that 1.4% of gross methane withdrawals have been lost due to distribution and upstream processes. Percent emissions are computed as follows (Equation (1)):

$$\text{percent emissions through stage } n = \sum_{i=1}^{n} \frac{\text{mass methane emitted at stage } i}{\text{mass methane reaching stage } i \times \frac{\text{mass methane withdrawn (incl. losses)}}{\text{unit mass methane reaching stage } i}} \quad (1)$$

Leakage rate estimates for a stage can be computed by subtracting the cumulative losses listed for upstream processes. Note also that gross withdrawals of methane are not equal to gross withdrawals of natural gas because natural gas is not pure methane. Relatedly, the embodied noncombustion GHG intensity of natural gas is also affected by the presence of other GHGs in withdrawn natural gas, including CO_2 and ethane. These nonmethane contributions are outside the scope of this study but could be relevant for some systems, particularly those using natural gas sources containing large mole fractions of CO_2.

Table 1 indicates several issues with the LCI databases. First, all investigated databases likely underestimate methane emissions from natural gas systems: total leakage reported by each database is similar to EPA estimates, which evidence suggests are too low (e.g., Alvarez et al., 2018). Also, they disagree. For example, data from ecoinvent (dated 2018) are much different from USLCI data, even though documentation for ecoinvent suggests its values are based on the USLCI. USLCI and EIO-LCA data are both based on US federal data from the late 1990s and early 2000s, but leakage rates by process are meaningfully different.

Second, databases have variable coverage of the natural gas system. This matters in part because LCA is sometimes used for hotspot identification, allowing analysts to focus on life cycle stages with the most potential for improvement. For example, even though the EIO-LCA production leakage of 0.57% is not that different from the USLCI production + processing leakage of 0.54%, users would draw very different conclusions about where to focus leakage reduction efforts. Note that none of the investigated databases account for methane leakage associated with nonindustrial end users (for example, residential cooking and heating), though its existence has long been understood (Oliphant, 1994) and can affect the climate implications of appliance choices.

Third, databases include numerous documentation discrepancies and nonintuitive definitions. Minor changes in interpretation or approach result in very different values for Table 1. For example, although ecoinvent 3.5 shows a leakage rate slightly higher than EPA estimates, this value is highly sensitive to a nonobvious parameter related to natural gas production from gas versus oil wells: through ecoinvent 3.3, use of a different value for that parameter, but essentially identical data otherwise, led to implied leakage of 1.2% rather than 1.6%. Even in version 3.5, the parameter does not reflect actual US conditions (see SM, S1.4). In another example, in EIO-LCA, calculating emissions based on the "Pipeline transportation" process suggests transmission-stage leakage of 0.42% (when emissions are allocated to coproducts based on monetary value) to 1.6% (when emissions are allocated to coproducts based on the 2002 EPA Greenhouse Gas Inventory), not the 0.34% implied by decomposing the "Natural gas distribution" sector (see SM, S1.2).

Table 1
Methane leakage as cumulative mass percentage of gross withdrawals of methane reaching a given stage, assuming natural gas progresses through each stage in sequence.

Data source	Production	Processing	Transmission & Storage	Distribution	End Use
Reference estimates					
Alvarez et al. (2018)	2.0%	2.1%	2.4%	2.5%[a]	n/a[b]
EPA (2015)	0.82%	0.93%	1.1%	1.3%	n/a
EPA 2013	0.38%	0.63%	1.0%	1.4%	n/a
LCA databases					
EIO-LCA[c]	0.57%	n/a	0.91%	1.4%	n/a
USLCI	0.34%[d]	0.54%	0.92%	n/a	n/a
ecoinvent v3.5	1.3%[e]	n/a	1.6%	n/a	n/a

[a] Note Alvarez et al., 2018 estimates for distribution leakage are based on the 2017 EPA GHGI, which they consider a lower bound for the true value (see Alvarez et al., 2018, S1.5).
[b] "n/a" denotes a process that is not explicitly included and where no data are available.
[c] Methane leakage from the natural gas system is based on the furthest downstream sector "Natural gas distribution:" see SM, S1.2 for details.
[d] The 0.34% value cited is based on the process "natural gas, extracted," which documentation suggests includes all natural gas production. The process "natural gas, at extraction site" is associated with 1.4% leakage. The distinction between the two is unclear: documentation indicates that the database distinguishes between associated and nonassociated natural gas, but both "natural gas, at extraction site" and "natural gas, extracted" indicate in their "technology description" that the process includes both natural gas from natural gas-only wells (nonassociated) and from oil wells (associated). See SM, S1.3 for details.
[e] Based on life cycle inventory data. Note that the production value is based on the mass-weighted percentage of contributions from "natural gas production" (methane leakage: 1.8%; ecoinvent contribution: 70%) and the natural gas allocation of "petroleum and gas production, on-shore" (methane leakage: 0.1%; ecoinvent contribution, 30%). See SM, S1.4 for details.

In USLCI and ecoinvent, ambiguous process names and definitions can easily lead to unintended choices with large implications for results. In both cases, there are multiple processes for natural gas extraction with unclear or no guidance on which to use. In USLCI, natural gas produced from gas-only wells versus from any well appear to be differentiated by the terms "extracted" (0.34% leakage) and "at extraction site" (1.4% leakage), respectively. Process descriptions are nearly identical, and both imply that all natural gas (both associated and nonassociated) is included. Further, "natural gas, extracted" uses a mass-denominated output while "natural gas, at extraction site" uses a volume-denominated output, so users might choose one or the other based on the units they are using without realizing the data are different. This introduces unnecessary possible confusion.

A similar issue exists for ecoinvent users, who might notice that the cumulative value for leakage post-transmission is smaller than leakage associated with "natural gas, production," even though the reference flow for the two is the same: 1 m^3 of "natural gas, high pressure." This discrepancy exists because the post-transmission process is also drawing on a second production process, "petroleum and gas production, on-shore" (see SM, S1.4). The use of the term "natural gas, high pressure" to refer to different systems at different places in the inventory is confusing. As with USLCI, users likely will not anticipate that data associated with "natural gas, production" are very different from those associated with the natural gas output of "petroleum and gas production, on-shore:" life cycle methane leakage for these processes is 1.8% or 0.10%, respectively. Less ambiguous names would help (e.g., calling one flow "natural gas, high pressure, at oil and gas co-production facility" and the other "natural gas, high pressure, at gas production facility").

Another serious source of confusion is use of the term "natural gas" itself. Without data on the composition of natural gas, converting among mass, energy, and volume units can introduce serious errors. Further, users might reasonably assume that the type of "natural gas" being referenced is relevant to the process at

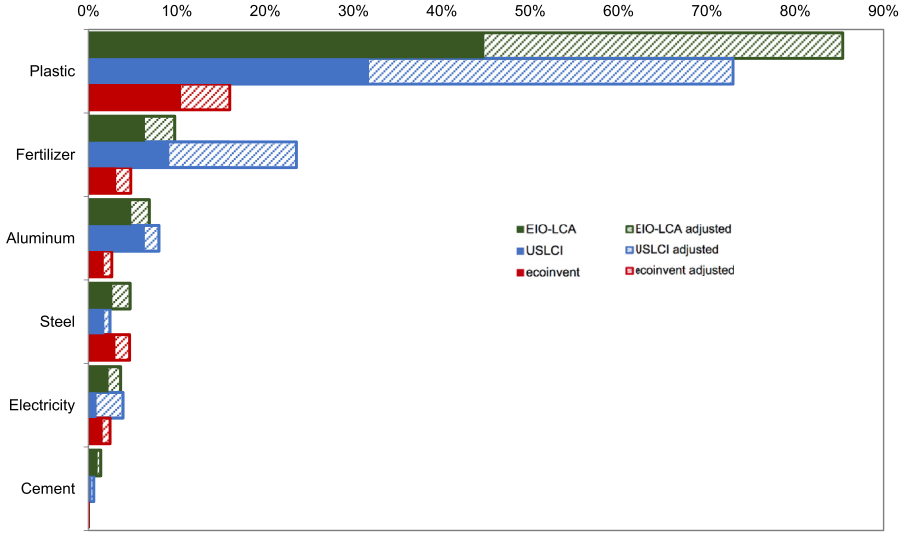

Fig. 1. Contribution of natural gas system methane leakage to greenhouse gas intensity for six basic materials in three life cycle inventory databases.
Notes: Specific processes used to proxy the six materials are found in the SM Data File. "Plastic" is "Plastics material and resin manufacturing" for EIO-LCA and polyethylene terephthalate (PET) for USLCI and ecoinvent. "Fertilizer" is "Fertilizer manufacturing" for EIO-LCA, "nitrogen fertilizer" for USLCI, and "urea ammonium nitrate" for ecoinvent. "Electricity" is the broadest category identified for US electricity in each case. "Adjusted" values show recalculated values assuming that leakage rates in each database matched values from Alvarez et al., 2018.

hand, which is not always the case. For example, in ecoinvent 3.5, the stated mass density of "natural gas, high pressure" of 0.84 kg/m^3 implies that the reference flow is raw gas at standard temperature and pressure, not pipeline-quality gas at high pressure (see SM, S1.4). USLCI uses energy rather than mass densities to describe flows but has similar problems (see SM, S1.3). Additionally, databases rarely document whether energy densities are reported on a higher heating value (HHV, sometimes called "gross") or lower heating value (LHV, sometimes called "net") basis. The difference between these two measures is about 10%, and the impact might not be noticed outside the energy community (e.g., one should not expect that a polymer chemist performing an LCA of a novel plastics process would know to check for this definitional detail).

3.1.2. Significance beyond natural gas systems

Problems with the characterization of methane leakage from natural gas systems are important for GHG footprinting beyond studies focused on natural gas itself. To illustrate this point, we use the EIO-LCA, USLCI, and ecoinvent databases to estimate natural gas system methane leakage GHG emissions as a percentage of direct process GHG emissions for six major industrial materials: plastic, fertilizer, aluminum, steel, electricity, and cement (Fig. 1). Calculation details, including the exact processes referenced, can be found in the SM Data File. Fig. 1 uses a GWP of 34 for methane (AR5, 100 year, with climate carbon feedback) and also presents "adjusted" values that show what results would be using leakage rates from a recent empirical study of US natural gas system leakage (Alvarez et al., 2018).

For some materials, embodied methane emissions from natural gas systems are not large compared to direct emissions. For example, cement manufacturing releases substantial process CO_2 emissions and uses little natural gas, so the influence of leaking methane is negligible. For others, however, most notably plastic, embodied methane leakage is substantial.

Fig. 1 suggests that correctly representing natural gas-system methane leakage should be a priority for LCI databases. Only a small number of processes need be updated, a minor task relative to the potential benefits to accuracy. Ensuring these emissions are visible in LCA studies can also help make results actionable. Reducing methane leakage from natural gas systems is likely an easier mitigation opportunity (Gallagher et al., 2015; Gladd, 2016; Hopkins et al., 2016; Lamb et al., 2015; Ravikumar and Brandt, 2017; von Fischer et al., 2017) than reducing byproduct emissions inherent to the production of a product (e.g., from calcining or

			Life Cycle Stages Relevant to End Use					
End Use	**Description[1]**	**2016 Consump-tion, Bcf**	**Produc-tion**	**Process-ing**	**Trans-mission**	**Storage**	**Distri-bution**	**User leakage[2]**
Lease fuel	natural gas used for production	1,169	yes					
Plant fuel	natural gas used for processing	421	yes	yes				
Pipeline and distribution use[3]	natural gas used for transmission and distribution pipelines	697	yes	yes	some	some	some	
Transmission	*not reported separately*	*n/a*	*yes*	*yes*	*some*	*some*		
Distribution	*not reported separately*	*n/a*	*yes*	*yes*	*yes*	*yes*	*some*	
Residential	natural gas used in residences	4,345	yes	yes	yes	yes	yes	yes
Commercial	natural gas used in commercial settings	3,105	yes	yes	yes	yes	yes	yes
Industrial	natural gas used in non-power generation industrial settings	7,722	yes	yes	yes	yes	*rare*	yes
Vehicle Fuel	natural gas used in vehicles	41	yes	yes	yes	yes	yes	yes
Electric Power	natural gas used for electricity generation	9,983	yes	yes	yes	yes	*rare*	yes
Approximate end use volume affected, %[4]		27,486	~100%	~96%	<90%	<90%	<23%	~87%

Fig. 2. Relationship between end uses of natural gas in the United States and upstream processes for which leakage and other impacts are embedded. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Notes: Bcf = billion cubic feet. Green/yes = "upstream process for this end use," yellow/some = "upstream process for some of this end use," where "some" means that neither condition is unusual, red = "not an upstream process for this end use," where "rare" means that analysts should assume the upstream process does not apply unless they specifically know about an unusual situation. Source: https://www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_a.htm.

[1]Formal definitions at https://www.eia.gov/dnav/ng/TblDefs/ng_cons_sum_tbldef2.asp.

[2]All users have the potential to emit fugitive (leaked) methane. These emissions vary widely by customer type and by specific customer: averages should be used very carefully, with effort to match the leakage rate to the end use type. Natural gas system fuel use (i.e., lease, plant, and pipeline and distribution consumption) is assumed not to have additional end-use leakage not accounted for as a system leakage.

[3]Pipeline use is not differentiated between the transmission and distribution system. Given common uses of natural gas for compressor stations along the pipeline route, natural gas used for these purposes will be subject to some leakage from the systems they support (i.e., transmission or distribution) as well as upstream emissions.

[4]Approximate; exact relationships between users and the natural gas system are not clearly known. For example, some industrial users might be located on distribution lines.

combustion) or unintentional emissions in more dispersed settings (e.g., methane emissions from abandoned coal mines or nitrous oxide emissions from fertilized soils).

3.2. The location of methane leakage

Natural gas has unusually diverse consuming sectors: in the US, 40% is used for electricity generation and 30% each for industrial and residential/commercial uses (EIA, 2017). For LCA, this matters not only because it means that natural gas system methane leakage has pervasive GHG footprint implications (see Section 3.1.2), but also because not all parts of the natural gas supply system are used to serve all end users. Correct representations of a product system include methane leakage only from those upstream processes that are invoked by the product system. Studies of methane leakage often do not explicitly clarify which processes are appropriate for which types of end use, so this section provides rules of thumb for modeling the natural gas system.

Some processes are common to essentially all uses of natural gas. These include extraction, processing, and transmission through high pressure pipelines. Emissions from these processes can vary (e.g., by basin of origin, processing plant, or pipeline), but all natural gas users connected to the same infrastructure will induce the same upstream impacts.

Other processes are not common to all uses. The most significant divergence is that some end uses are served by low-pressure distribution systems downstream of high-pressure transmission systems, while others draw on the high-pressure system directly. Although both the transmission and distribution systems are sometimes called “pipelines” or “transportation,” they are separate systems. Thus, distribution leakage should not be allocated to users supplied directly by the transmission system. A rule of thumb is that natural gas used for power plants, industrial heat, and chemical plant feedstocks generally does not pass through the distribution system, while natural gas used for commercial and residential purposes generally does (Fig. 2, and see SM, S2). The most decision-relevant application of this observation is for natural gas-fired power plants, which are rarely fed by distribution lines, and which have frequently been compared to coal-fired power on the basis of relative carbon emissions. Given methane's high GWP, incorrectly including distribution leakage for natural gas-fired power can lead to nonneglible overestimates of GHG intensity (Alvarez et al., 2012).

We recommend that authors explicitly state which natural gas system processes are included in analysis, rather than citing an overall leakage rate. For high resolution studies, note that actual (rather than system-average) leakage can be highly site specific given different use of technologies, levels of training, safety and operational practices, year and/or season, legal context, company practice, or geology and land characteristics. Customer-side leakage is not well characterized by the empirical literature.

3.3. The influence of global warming potential on methane leakage impact

The scientific community's estimate of methane's GWP has not yet stabilized. Notably, published estimates have tended to increase over time, both for 20- and 100-year estimates (Fig. 3). For methane, IPCC-estimated GWP values that include climate-carbon feedback are slightly higher than GWP values that do not include this feedback, which is intended to account for inconsistent estimates of GWP for the reference gas (CO_2) versus other species (Gasser et al., 2017). The current IPCC estimate for methane's 100-year GWP (with climate-carbon feedback) is about 60% higher than the 1996 estimate and about 35% higher than the 2007 estimate; for the 20-year GWP, the current estimate is about 50% higher than the 1996 estimate and 20% higher than the 2007 estimate. The anticipated release of the Sixth Assessment Report in 2022—nearly ten years after the release of the current Fifth Assessment Report (AR5)—might update GWP estimates again.

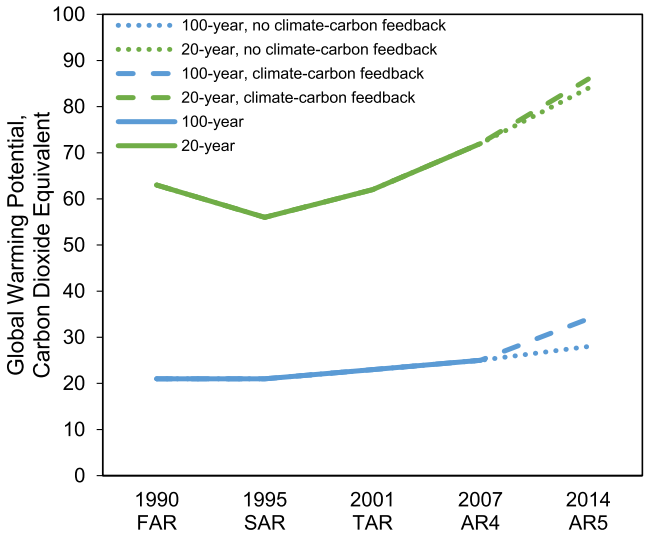

Fig. 3. Estimated global warming potential of methane by IPCC assessment report. **Data source:** Intergovernmental Panel on Climate Change (2018).

How much does GWP matter? Using a GWP of 25 (100-year, AR4), the 2015 GHGI estimates that natural gas systems accounted for 2.8% of total US GHG emissions (EPA, 2017). Simply updating the GWP to 34 (100-year, AR5 with climate-carbon feedback) raises this estimate to 3.6% of total US GHG emissions. Updating both GWP and natural gas system leakage rates to reflect state of the science estimates (Alvarez et al., 2018) suggests that methane leakage from natural gas systems alone accounts for 7.1% of total US GHG emissions—2.5 times the recorded estimate (EPA, 2017). Researchers using CO_2e-based literature estimates for methane impacts should take care to identify and harmonize GWP assumptions from previously published work. Especially for studies that directly address natural gas systems, conducting GWP sensitivity analysis is advisable.

4. Conclusion

The impact of methane leakage from natural gas systems is systematically underestimated and imprecisely characterized, which affects GHG footprints across product systems. Underestimated natural gas system methane leakage and low GWPs are both significant. LCA databases often underestimate leakage through key life cycle stages even relative to recorded estimates, and the connection between database estimates and relevant data is frequently unclear. The small number of processes involved, and the large impact to product LCA uncertainty, makes natural gas system methane leakage a priority target for data quality assurance in inventory databases. Meaningful improvements can be made with attention to the issues of leakage rates, system boundaries, and GWP described in this work.

Acknowledgements

Neither author has a conflict of interest to declare. This research did not receive any specific grant from funding agencies in the

public, commercial, or not-for-profit sectors.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2019.03.096.

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Attachment 2

Waste Management 97 (2019) 38–46

Contents lists available at ScienceDirect

Waste Management

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

Total methane emission rates and losses from 23 biogas plants

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ARTICLE INFO

Article history:
Received 25 March 2019
Revised 16 July 2019
Accepted 20 July 2019
Available online 1 August 2019

Keywords:
Anaerobic digestion
Fugitive emissions
Greenhouse gas emissions
Tracer gas dispersion
Carbon footprint assessment

ABSTRACT

Methane losses from biogas plants are problematic, since they contribute to global warming and thus reduce the environmental benefits of biogas production. Total losses of methane from 23 biogas plants were measured by applying a tracer gas dispersion method to assess the magnitude of these emissions. The investigated biogas plants varied in terms of size, substrates used and biogas utilisation. Methane emission rates varied between 2.3 and 33.5 kg CH_4 h^{-1}, and losses expressed in percentages of production varied between 0.4 and 14.9%. The average emission rate was 10.4 kg CH_4 h^{-1}, and the average loss was 4.6%. Methane losses from the larger biogas plants were generally lower compared to those from the smaller facilities. In general, methane losses were higher from wastewater treatment biogas plants (7.5% in average) in comparison to agricultural biogas plants (2.4% in average). In essence, methane loss may constitute the largest negative environmental impact on the carbon footprint of biogas production.

1. Introduction

Biogas from anaerobic digestion, using various substrates such as manure, food waste, organic industrial waste and sludge from wastewater treatment, may result in several greenhouse gas (GHG) mitigation effects, including fossil fuel substitution, the possible balancing of energy sources in a supply system with a high proportion of wind and solar power and a reduction in methane (CH_4) emissions from manure management (Clemens et al., 2006; IPCC, 2011; Sommer et al., 2004). Fugitive CH_4 emissions from biogas plants, however, will reduce the environmental benefits of biogas production, mainly because of the relatively high global warming potential of CH_4, in that releasing just 1 kg of CH_4 into the atmosphere has the same effect with regards to global warming as the release of 28 kg of carbon dioxide (CO_2) integrated over a 100-year period (not including climate feedback) (Myhre et al., 2013). Data on the magnitude of these emissions are sparse, which in turn causes uncertainty with regards to the environmental assessment of biogas production concerning global warming (Meyer-Aurich et al., 2012; Møller et al., 2009). Recent studies suggest that the extent of CH_4 emissions expressed as a fraction of production lost to the atmosphere (also referred to as "CH_4 loss") may vary between facilities. Liebetrau et al. (2013), for instance, monitored CH_4 emissions from ten German biogas plants, using an on-site approach where individual leaks were identified and emission rates were subsequently measured. It was found that CH_4 emissions relative to the energy output of the biogas plants varied by approximately one order of magnitude between plants, and that open digestate storage tanks in many cases were the most significant emission source. Other sources of CH_4 emission from biogas plants may include unburnt CH_4 from gas engine exhausts, pressure relief valves, biogas upgrading units, ventilation from buildings, leaks in pipes, tanks, etc. (Kvist and Aryal, 2019; Angelidaki et al., 2018; Fredenslund et al., 2018; Liebetrau et al., 2013; Reinelt et al., 2017, 2016; Samuelsson et al., 2018).

An important step in understanding and subsequently reducing CH_4 emissions in the biogas sector is the reliable identification and quantification of single emission sources and the quantification of overall plant emissions. In general, two main approaches can be used for gas emission quantification: on-site and ground-based remote sensing approaches. The on-site approach measures emissions from various single sources at the plant, and it is the method most commonly used (Reinelt et al., 2016; Daniel-Gromke et al., 2015; Westerkamp et al., 2014; Liebetrau et al., 2013). Often a two-step procedure is followed where the first step includes a leakage search performed by using infrared cameras or handheld methane analysers. The second step includes quantification of each identified leakage or emission source often using the stationary or dynamic flux chamber technique. The ground-based remote sensing approach includes different methodologies and measures emissions from a good distance (for example one kilometre) away from the plant, thus providing plant-integrated emission numbers (Fredenslund et al., 2018; Delre et al., 2017; Groth et al., 2015; Yoshida et al., 2014; Hrad et al., 2014; Westerkamp et al., 2014; Flesch et al., 2011). Ground-based remote sensing techniques

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https://doi.org/10.1016/j.wasman.2019.07.029
0956-053X/

encounter inverse dispersion techniques using for instance open-path lasers and tracer gas dispersion methods. Recent measurement comparison studies have found that methods measuring the plant's total CH_4 emission often result in a higher emission rate in comparison to on-site measurements, where the total emission is obtained by summing up those measured from single sources (Fredenslund et al., 2018; Reinelt et al., 2017). The reason for the discrepancy between on-site and ground-based remote sensing approaches is most likely that single sources are overlooked and/or not identified, or they are technically not quantifiable when using a particular measurement technique (e.g. open tanks). For GHG emission reporting or environmental assessment, the plant's total emissions are important; however, if the purpose of measuring CH_4 emissions at a biogas plant is to identify mitigation options, and thereby provide options to improve the environmental benefits of biogas production, on-site methods are needed.

The objective of this study was to quantify CH_4 emission rates and losses from full-scale biogas plants. The study focused primarily on large, centralised, manure-based biogas plants, which produce the bulk of biogas in Denmark. Production capacity at this type of facility was in the expansion phase nationally at the time of this study. In addition, CH_4 emission rates and losses were measured at biogas plants located at wastewater treatment plants (WWTPs). Landfill gas extraction and utilisation sites were not included. The paper compiles the results taken from several biogas plants, in order to provide an estimate of CH_4 losses from biogas production and to assess the importance of minimising this issue. CH_4 emissions were measured using the tracer gas dispersion method, which measures plant-integrated emission rates. The environmental importance of fugitive CH_4 emissions from biogas plants was evaluated by performing CO_2 footprint calculations for a generic, manure-based agricultural biogas plant.

2. Methodology

2.1. Site descriptions

The biogas plants included in this study all utilise continuously stirred anaerobic digesters to produce biogas, and they all are commercially operated facilities. They varied in terms of feedstocks, size, rate of gas production, type of gas utilisation and other factors. Table 1 provides an overview of the main characteristics of the plants.

Thirteen of the biogas plants (plants 1–13) are categorised herein as "agricultural", which means that the feedstocks consist mainly of manure, energy crops and agricultural waste, though they can also receive other feedstocks such as slaughterhouse waste or food waste. Out of the 13 agricultural plants, nine receive manure as the main feedstock (>75% of dry matter input is manure), whereby organic waste (organic industrial waste and/or food waste) is used as a supplement to increase gas production. Two biogas plants (plants 8 and 12) rely on energy crops (grass, maize silage and forage rye), one (plant 5) receives mainly organic waste (~80% of dry matter input) but also receives manure and one plant (plant 13) mainly uses food waste. Plant 13 (and possibly also plant 5) could depending on definition be termed a waste treatment biogas plant as it mainly treated slaughterhouse waste, food industry waste and household food waste. However, as this plant was the only one of this type and also as the generated digestate is spread on farmland, the plant was included in the agricultural biogas plant category.

Five of the agricultural biogas plants (plants 1, 2, 5, 6 and 8) were recently constructed (constructed in 2013 or later), whereas the remaining agricultural plants generally were constructed in the 1980s or 1990s. For a number of reasons, the 2000s saw very low levels of investment in Danish biogas production (Raven and Gregersen, 2007), whereas increases and diversification in subsidies in recent years have led to a "second wave," with most new production capacity emanating from large facilities that upgrade and inject the biogas into the Danish natural gas distribution grid. At the smaller and older plants, it is more common that the biogas is utilised on-site in a combined heat and power (CHP) unit.

Biogas plants 14–23 are categorised herein as "wastewater treatment biogas plants". These plants utilise sludge from wastewater treatment to produce biogas, and they are all located on the grounds of the WWTP from where the sludge originates. They can thus be considered part of a larger plant, the primary function of which is to remove pollutants from wastewater before discharge to a recipient, with energy production as secondary function. The biogas plants categorised as "agricultural biogas plants" all rely on gas production for revenue. Although the wastewater treatment biogas plants receive revenue from their gas production, their primary function is to stabilise and reduce the volume of sludge, and thereby the costs of further sludge treatment. The WWTPs may thus arguably have less incentive to minimize loss of methane compared to the agricultural biogas plants.

The size of the plants varied in size in terms of treated feedstocks, from 30,000 to 600,000 tonnes (wet weight) per year for agricultural biogas plants, while the WWTPs treated between 60,000 and 805,000 PE, which corresponded to a load to the on-site biogas plants of between 3,000 and 112,000 tonnes (wet weight) per year.

The biogas plants differ with regards to gas utilisation (Table 1). At 12 plants, all or some of the produced biogas is utilised on site in a CHP unit. At plants 3, 4 and 7 some of the gas (~20–30%) is used on site in a CHP unit providing process heat for the biogas reactors. The generated electricity is sold to the grid. The remaining part of the gas is routed off site to a nearby power plant (where it is used in a CHP unit). At eight plants, all or some of the biogas is upgraded to biomethane, using technologies such as water scrubbers or chemical scrubbers. At these facilities, the gas is either compressed and transported off site or is injected into a natural gas distribution network. At four plants (5, 11, 14 and 21), all gas utilisation occurs off site. An example of this type is plant 5, where the biogas is led to a nearby power plant (and used in a CHP unit) to generate electricity to the grid and heat to a district heating network.

Open digestate storage units may be significant emitters of CH_4 from biogas plants (Samuelsson et al., 2018; Reinelt et al., 2017; Baldé et al., 2016; Liebetrau et al., 2013). Table 1 lists those facilities, which store digestate in open tanks on site. All biogas plants were equipped with gas storage units with capacities typically corresponding to ~1 to 2 days gas production.

In all, the 23 biogas plants included in this study represent a variety of continuously stirred reactor biogas plant types with regards to amounts of feedstock utilised, feedstock types, gas production rates and gas utilisation.

2.2. Tracer gas dispersion method

CH_4 emission rates from each biogas plant were quantified using a tracer gas dispersion method, whereby a gaseous tracer (here acetylene gas – C_2H_2) is continuously released at the biogas plant, and concentrations of CH_4 and C_2H_2 are then measured while traversing the CH_4/C_2H_2 plume at distances up to ~2 km away, using a vehicle-mounted, high-precision gas analyser. The method has been applied to quantify fugitive emissions from various facilities such as landfills, composting facilities, WWTPs and biogas plants (Andersen et al., 2010; Fredenslund et al., 2018; Mønster et al., 2014; Scheutz et al., 2011; Yoshida et al., 2014). An advantage of this method compared to on-site methods, where emission sources are quantified individually, is the measurement

Table 1
Overview of the main characteristics of the investigated biogas plants.

Agricultural biogas plants	Type of feedstock and annual total amount treated at the plant (in tonnes wet weight per year)	On site gas utilisation (CHP[1]/biogas upgrade)	Digestate storage (open/closed)
1*	Manure, maize silage, organic waste (600,000)	Biogas upgrade: chemical scrubber, gas grid injection	Closed
2*	Manure, slaughterhouse waste (240,000)	Biogas upgrade: water scrubber, gas grid injection	Closed
3	Manure, organic waste (300,000)	CHP (partly off site)	Closed
4	Manure, slaughterhouse waste, other organic waste (235,000)	CHP (partly off site)	Closed
5*	Industrial waste, manure (200,000)	None – routed for off-site use in a CHP	Closed
6*	Manure, maize silage (118,000)	Biogas upgrade: chemical scrubber, gas grid injection	Closed
7	Manure, slaughterhouse waste, other organic wastes (225,000)	CHP (partly off site)	Closed
8*	Maize silage, forage rye	CHP and biogas upgrade: chemical scrubber, gas grid injection	Closed
9	Manure, organic waste, maize silage (170,000)	CHP	Closed
10	Manure, organic waste (37,000)	CHP	Closed
11	Manure, maize and grass silage, glycerol (30,000)	None – routed for off-site use in a CHP	Closed
12	Grass and maize silage, manure	CHP	Open
13	Organic waste (slaughterhouse waste, industrial food waste and household food waste) (104,000)	Biogas upgrade: chemical and water scrubber, gas to vehicle fuel	Open
Wastewater treatment biogas plants	**Feedstock (amount given in person equivalent)**	**On site gas utilisation (CHP[1]/biogas upgrade)**	**Digestate storage (open/closed)**
14	Sludge from wastewater treatment (750,000 PE[2])	None – routed for off-site biogas upgrading	Closed
15	Sludge from wastewater treatment (265,000 PE)	CHP	Open
16	Sludge from wastewater treatment (150,000 PE)	CHP	Closed
17	Sludge from wastewater treatment (420,000 PE)	Biogas upgrade: chemical scrubber, gas grid injection	Closed
18	Sludge from wastewater treatment (95,000 PE)	CHP	Closed
19	Sludge from wastewater treatment (60,000 PE)	CHP	Open
20	Sludge from wastewater treatment (125,000 PE)	CHP	Closed
21	Sludge from wastewater treatment (805,000 PE), industrial food waste and sewage sludge from small WWTPs	None – routed for off-site biogas upgrading, gas to vehicle fuel	Open
22	Sludge from wastewater treatment (95,000 PE), food waste	Biogas upgrade: chemical scrubber, gas to vehicle fuel	Open
23	Sludge from wastewater treatment (120,000 PE)	Biogas upgrade: chemical scrubber, gas to vehicle fuel	Open

[1] CHP: Combined heat and power.
[2] PE: Person equivalent.
* Constructed in 2013 or later.

of the biogas plant's total CH_4 emission, with little risk of underestimating them due to undetected emission sources (Fredenslund et al., 2018).

The method and instrumentation are described in detail in Mønster et al. (2014) and Yoshida et al. (2014). The overall error of the method has been the subject of a recent validation study, and it was found very likely to be less than 20% (Fredenslund et al., 2019). The potential error of the tracer gas dispersion measurement technique was determined to 15% by establishment of an error budget including the analytical error, error in the tracer gas release rate, data processing, and error in tracer gas placement and source simulation. The error of a measurement is the combined error of the method and the variability of the quantification, which was found to be about 20% in a controlled release test and comparable to the error obtained by comparison of the measured emission rate and the known controlled release rate (Fredenslund et al., 2019).

Emission rates are calculated using Eq. (1):

$$E_{target} = Q_{tracer} \times \frac{\int_{plume\ start}^{plume\ end} C_{target} - C_{target,backgruound})dx}{\int_{plume\ start}^{plume\ end} C_{tracer} - C_{tracer,backgruound})dx} \times \frac{MW_{target}}{MW_{tracer}} \quad (1)$$

where E_{target} is the emission rate of CH_4 in kg h^{-1}; Q_{tracer} is the release rate of the acetylene tracer gas in kg h^{-1}; C_{target} and C_{tracer} are the measured downwind concentrations in parts per billion (ppb); $C_{target,\ background}$ and $C_{tracer,\ background}$ are the measured background concentrations in parts per billion (ppb) and MW_{target} and MW_{tracer} are the molar weights of the two gases.

The measurements were taken by driving through the downwind plumes several times (typically 10 to 20 traverses per measurement campaign). Each plume traverse resulted in one CH_4 emission measurement, calculated using Eq. (1). The CH_4 emission rate (in kg h^{-1}) was calculated as the average value of the individual plume traverses, and any uncertainty was estimated as the standard error of the mean of the measurements (Fredenslund et al., 2019).

CH_4 loss (%) was determined as the ratio of the measured CH_4 emission to the CH_4 production of the biogas plants, logged the day the measurement was performed.

2.3. Measurement campaigns

The measurements were performed July 2013 through June 2018. At six biogas plants, this happened on a single day, whereas for the remaining 17 plants, measurements were repeated up to a maximum of six days (Table 2).

All measurements were performed using the same analytical equipment and the method described in Section 2.2. The measurements were performed during normal operation of the biogas plants. No malfunctions were reported by the plants for the periods of measurement. As implementation of the method required certain adjustments in each case, some variability with regards to tracer gas release rates and number of release points exists.

Table 2
Overview of measurements performed.

Plant number	Days of measurement campaigns	Number of plume traverses	Tracer gas release rate (kg C_2H_2 h^{-1})
Agricultural biogas plants			
1	1	20	2.24
2	2	66	1.07
3	1	14	2.29
4	3	54	1.90
5	2	32	1.44
6	2	42	0.97
7	3	54	0.83
8	5	166	1.32
9	2	39	1.24
10	1	17	0.91
11	2	29	1.50
12	4	138	1.51
13	2	21	0.44
Wastewater treatment biogas plants			
14	6	82	0.57
15	1	21	0.92
16	4	63	0.51
17	2	37	1.68
18	2	40	0.93
19	4	89	0.48
20	1	16	0.90
21	1	16	–
22	3	81	0.91
23	3	82	0.78

The average tracer gas release rate varied between 0.11 and 2.29 kg C_2H_2 h^{-1}, and the number of tracer gas release points varied between one and three. The measurement distance varied from a few hundred metres up to more than 1 km, according to the availability of drivable roads downwind and the detectability of elevated concentrations of CH_4 and C_2H_2 in the plume – low emission rates and high wind speeds increase dilution, and so it may be necessary to traverse the plume closer to the source of emission.

2.4. Impact of methane emissions on the overall CO_2 footprint of biogas plants

The impact of CH_4 loss on the overall CO_2 footprint of biogas plants was evaluated by using a calculation model provided by the Danish Ministry of Environment for environmental impact assessments of biogas projects (Danish Nature Agency, 2014). The model considers the following factors in determining the overall CO_2 footprint of biogas plants:

Substitution of fossil fuels
Substitution of chemical fertiliser
Transportation of feedstock and digestate
Change in manure management compared to conventional storage and use of manure in agriculture (fewer GHG emissions from manure storage at farms when manure is digested before storage)
Energy use of the biogas plant
Direct GHG emissions from biogas production and utilisation

Emissions and savings were determined by considering five levels of direct CH_4 loss from an agricultural biogas plant: 1%, 2%, 5%, 10% and 20%. Losses of produced biogas contribute directly (CH_4 emitted into the atmosphere) and indirectly (less substitution of fossil fuel as a result of lost biogas production), so both losses were included in the model.

In this assessment, we considered a generic agricultural biogas plant receiving 50,000 tonnes yr^{-1} of cattle manure, 60,000 tonnes yr^{-1} pig manure and 5000 tonnes yr^{-1} organic waste, which in combination produced 2.2 million m^3 CH_4 yr^{-1}. This calculation example is similar to one described by The Danish Nature Agency (2014). Two biogas utilisation options were considered: CHP and biogas upgrade and injection into the natural gas grid.

Table 3 provides an overview of emission factors as well as energy use and CHP energy conversion efficiencies. Two emission factors regarding the use and production of electricity were considered in terms of CHP gas utilisation, namely average and marginal. The average emission factor corresponds to the average emissions associated with the provision of electricity in Denmark, whereas the marginal factor is derived from an estimate of which electricity sources are reduced when production from (for example) biogas plants is increased – also in Denmark. The provision of electricity, on average, consisted of 17% coal, 6% natural gas, 55% wind, hydro and solar, 18% waste incineration, biomass and biogas, 1% oil and 3% nuclear – as reported by the Danish national authority on electricity production (Energinet.dk, 2018), whereas the provision of marginal electricity consisted of 80% coal, 15% natural gas and 5% renewables from a recent study on CO_2 emissions caused by

Table 3
Overview of parameters used in carbon footprint calculations.

Parameter	Value	Reference
Emission factors		
Provision of electricity (average)[a]	0.053 kg CO_2-eq. MJ^{-1}	Energinet.dk (2018)
Provision of electricity (marginal)[b]	0.24 kg CO_2-eq. MJ^{-1}	Ea Energianalyse (2016)
Provision and consumption of natural gas	0.057 kg CO_2-eq. MJ^{-1}	Danish Energy Agency (2018)
Provision of heat (district heating, Danish average value)[c]	0.056 kg CO_2-eq. MJ^{-1}	Danish Nature Agency (2014)
Production of N fertiliser	7.0 kg CO_2-eq. kg N^{-1}	Danish Nature Agency (2014) and Wood and Cowie (2004)
Production of P fertiliser	0.5 kg CO_2-eq. kg P^{-1}	Danish Nature Agency (2014) and Wood and Cowie (2004)
Transportation of digestate, manure, etc.	0.09 kg CO_2-eq. $tonne^{-1}km^{-1}$	(Danish Nature Agency, 2014)
Emission of CH_4	28 kg CO_2-eq. kg CH_4^{-1}	Myhre et al. (2013)
Manure management, cattle	−15 kg CO_2-eq. tonne $manure^{-1}$	Danish Nature Agency (2014)
Manure management, pigs	−23 kg CO_2-eq. tonne $manure^{-1}$	Danish Nature Agency (2014)
Other factors		
Process heat	8.4% of energy output	Danish Energy Agency (2017a)
Electricity use, biogas plant	3.7% of energy output	Danish Energy Agency (2017a)
Electricity use, biogas upgrade and compression	5.3% of energy output	Danish Energy Agency (2017a)
Electrical efficiency, CHP unit	44%	Danish Energy Agency and Energinet.dk (2014)
Total efficiency, CHP unit	92%	Danish Energy Agency and Energinet.dk (2014)

[a] Provision of electricity, average: 17% coal, 6% natural gas, 55% wind, hydro and solar, 18% waste incineration, biomass and biogas, 1% oil and 3% nuclear.
[b] Provision of electricity, marginal: 80% coal, 15% natural gas and 5% renewables.
[c] Average value of Danish district heating networks utilising various energy sources (waste incineration, solar, surplus heat from coal and biomass electricity production and more).

increasing electricity demand in Denmark (Ea Energianalyse, 2016). The differences in energy mix in the provision of average electricity, and provision of marginal electricity cause a relatively large difference in emission factors at 0.053 kg CO_2-eq MJ^{-1} (average) and 0.24 kg CO_2-eq MJ^{-1} (marginal). In the scenario considering the biogas upgrade, for simplicity we only considered the average emission factor for electricity use. In all scenarios, the consumption of heat by the biogas plant was presumed to be in the form of natural gas. In both CHP scenarios, the same emission factor for heat substitution was used, namely an average value of district heating networks in Denmark.

Both feedstock and digestate were assumed to be transported 5 km to and from the biogas plant. The anaerobic digestion of organic waste and the land application of digestates, and thereby recycling of the contained nutrients, was assumed to result in the reduced use of chemical fertiliser at 10 tonnes N yr^{-1} and 5 tonnes P yr^{-1}. The nutrient content of the manure was not considered to contribute to the reduced use of chemical fertiliser, since these nutrients would be applied to agricultural land anyway as raw manure without digestion.

3. Results and discussion

3.1. Measured CH_4 emission rates

Table 4 lists the measured CH_4 emission rates and losses for the 23 biogas plants in this study. The table also lists the biogas production rate of each plant, which was reported by individual plant operator in each case in the form of average daily production at the time of measurement. In those cases where CH_4 emission rates were measured over several campaigns, the listed CH_4 emission rates, gas production rates and CH_4 losses are average values.

Overall, the average CH_4 emission rates varied between 2.3 and 33.5 kg CH_4 h^{-1}. CH_4 losses (CH_4 emission relative to CH_4 production) varied between 0.4 and 14.9%, with the average being 4.6%. These results are comparable to Liebetrau et al. (2013), who found CH_4 losses from single, dominant sources (CHP units and open digestate storage) equating to between 0.22 and 11.2% of the utilised gas at 10 biogas plants. They are also comparable to the results of a study of a Canadian biodigester, where losses under normal operating conditions corresponded to 3.1% of CH_4 production (Flesch et al., 2011).

In general, CH_4 losses were higher from wastewater treatment biogas plants (average 7.5%) than from agricultural plants (2.4%) (Table 4). At seven of the 23 biogas plants, the average measured CH_4 loss was higher than the overall average (4.6%) (Table 4). Of these seven plants, six were WWTPs. The agricultural biogas plant that emitted more than 4.6% (plant 11, Table 4) actually had the lowest level of biogas production (Table 4). Of the agricultural plants, the highest CH_4 loss was 8.4% (biogas plant 11). The reported loss was based on two measurement campaigns, which both showed high CH_4 emissions. There was no on-site gas utilisation and no open mixing tanks, digestate storage tanks or similar. A specific reason as to why the biogas plant had a higher loss than

Table 4
Overview of measured average CH_4 emission rates and losses.

Plant number	On-site sources included in the measured emission (CHP or biogas upgrade unit)	Average biogas production kg CH_4 h^{-1}	Average CH_4 emission rate kg CH_4 h^{-1}	Average CH_4 loss %	Estimated revenue loss[e] k€ y^{-1}	Off-site sources not included in the measured emission (CHP or biogas upgrade unit
1	Biogas upgrade unit	1469	6.5 ± 0.6	0.4 ± 0.04	27.4 ± 2.3	–
2[a]	Biogas upgrade unit	1083	19.1 ± 2.5	1.8 ± 0.23	80.9 ± 10.6	–
3	CHP[f]	888	23.2 ± 1.7	2.6 ± 0.19	98.5 ± 7.2	CHP
4	CHP[f]	858	6.4 ± 0.5	0.7 ± 0.06	27.0 ± 2.0	CHP
5	–	498	3.0 ± 0.3	0.6 ± 0.06	12.9 ± 1.4	CHP
6[a]	Biogas upgrade unit	411	10.7 ± 0.5	2.6 ± 0.12	45.2 ± 2.1	–
7[a]	CHP[f]	404	6.4 ± 0.2	1.6 ± 0.06	27.1 ± 1.0	CHP
8	CHP and biogas upgrade unit	400	2.3 ± 0.4	0.6 ± 0.10	9.9 ± 1.6	–
9	CHP	333	14.9 ± 0.9	4.5 ± 0.26	63.1 ± 3.6	–
10	CHP	234	6.1 ± 0.8	2.6 ± 0.35	26.1 ± 3.5	–
11	–	74	6.4 ± 0.4	8.6 ± 0.50	27.2 ± 1.6	CHP
12	CHP	127	2.6 ± 0.4	2.1 ± 0.35	11.0 ± 1.9	–
13[b]	Biogas upgrade unit	815	21.2 ± 3.3	2.6 ± 0.40	90.0 ± 13.8	–
Plant average CH_4 loss, **agricultural**: 2.4%						
Production weighted average CH_4 loss, **agricultural**: 1.7%						
14[c]	–	440	9.8 ± 0.7	2.2 ± 0.15	41.7 ± 2.8	Biogas upgrade unit
15[a]	CHP	162	13.5 ± 0.5	8.3 ± 0.33	57.3 ± 2.3	–
16[c]	CHP	100	2.6 ± 0.4	2.6 ± 0.39	11.1 ± 1.6	–
17	Biogas upgrade unit	96	12.3 ± 1.2	12.8 ± 1.29	52.0 ± 5.2	–
18	CHP	88	8.1 ± 0.5	9.1 ± 0.60	34.2 ± 2.3	–
19[c]	CHP	85	2.6 ± 0.1	3.0 ± 0.16	11.0 ± 0.6	–
20	CHP	262	10.0 ± 1.0	3.8 ± 0.38	42.5 ± 4.2	–
21[d]	–	525	33.5 ± 0.6	6.4 ± 0.12	142.3 ± 2.6	Biogas upgrade unit
22[c]	Biogas upgrade unit	83	10.0 ± 0.6	12.0 ± 0.78	42.3 ± 2.7	–
23[c]	Biogas upgrade unit	58	8.6 ± 0.4	14.9 ± 0.72	36.5 ± 1.7	–
Plant average CH_4 loss, **WWTP**: 7.5%						
Production weighted average CH_4 loss, **WWTP**: 5.8%						
All biogas plants						
Plant average CH_4 loss, all: 4.6%						
Production weighted average CH_4 loss, all: 2.5%						

[a] Results were partly (first measurement) reported in Fredenslund et al. (2018).
[b] Results were reported in Reinelt et al. (2017).
[c] Results were reported in Delre et al. (2017).
[d] Results were reported in Samuelsson et al. (2018).
[e] Considering an estimated revenue of 0.7 €/Nm^3 CH_4.
[f] About 20–30% of the gas is used in a CHP unit, while the remaining is transported off site.

the average agricultural biogas plant was thus not identified. In general, the CH_4 emission rate relative to production seemed to correlate with the size of the biogas plant (Fig. 1), in that units with the highest gas production emitted proportionally less CH_4 compared to plants with relatively low output. One reason for this finding may be that the larger facilities have more economical resources for maintenance, re-investment and employment of highly proficient plant operators. Another reason may be that the number of potential emission sources (number of process units, pipes, joints, valves, etc.) is not necessarily proportional to the rate of biogas production. There was also the tendency that the larger agricultural plants were built more recently and thus may better represent the most up-to-date technology. CH_4 emission from biogas plants is not regulated directly in Denmark, so no regulatory explanation for the difference in methane loss for small biogas plants compared to larger plants was found. CH_4 losses from plants built within approximately the last 5 years (plants 1, 2, 5, 6 and 8) were relatively low (0.4, 1.8, 0.6, 2.6 and 0.6%, respectively). Two of the 13 agricultural plants were solely energy plants, where the input was mainly crops grown specifically for energy production, and both had CH_4 losses lower than the average for all plants.

As mentioned previously, agricultural biogas plants rely mostly on revenue from energy production for their existence, whereas energy production is a secondary activity in the case of wastewater treatment biogas plants. The economic incentive to maximise energy production, and therefore minimise leaks, may therefore be stronger for agricultural biogas plants. Finally, it should be noted that in this study total CH_4 emissions from the plants were measured. Wastewater treatment plants are more complex in structure than agricultural biogas plants, as they also have a water treatment operation in addition to sludge management and biogas production. Therefore, CH_4 emission rates measured at wastewater treatment biogas plants could also encounter CH_4 emissions from the water treatment line and from the open storage of sludge, which is more common at WWTPs in comparison to agricultural plants. However, at WWTPs, the main CH_4-emitting source will be biogas activities, even though CH_4 emissions can also occur from the plant inlet and from aeration tanks. Samuelsson et al. (2018) quantified CH_4 emissions from various unit processes at a WWTP and found that overall, about 81% of the CH_4 emissions quantified on site were released from the sludge treatment line. Delre et al. (2017) came to a similar conclusion based on on-site screenings of atmospheric CH_4 concentrations, where the highest elevated intensities were seen in the vicinity of sludge treatment activities. Sludge (un-digested or digested) storage in open tanks or basins can be a potential source of CH_4, which is challenging to quantify due to the large open surface area. At some of the WWTPs, open digestate storage of sludge could explain (but only partly) the higher emission rates. As an example, the average CH_4 loss at WWTPs with open storage was 9.2% in comparison to plants without on-site open storage (6.1%).

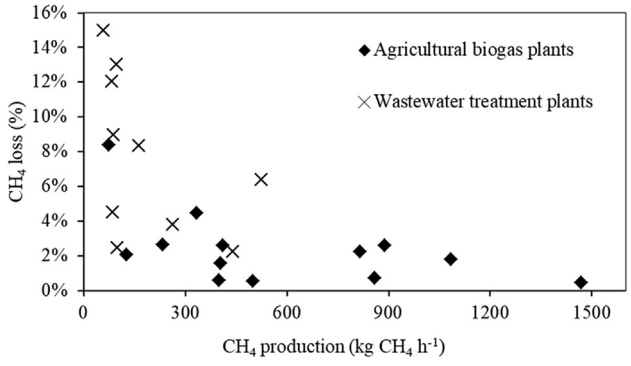

Fig. 1. Average CH_4 loss as a function of the average gas production at biogas plants.

Finally, it should be noted that at four of the plants (5, 11, 14 and 21) all gas utilisation occurs off site and at three of the plants part of the gas utilisation (~70–80%) occurs off site (plants 3, 4 and 7). For these plants, any CH_4 emission from the off site utilisation was therefore not included in the measured total CH_4 emission and thus the total CH_4 emission from the combined production and utilisation could be higher than the values reported in Table 4. At two of the plants (WWTPs 14 and 21) the generated biogas is routed off site for biogas upgrading. CH_4 emission factors from biogas upgrading units vary depending on technology applied. An average CH_4 slip of 0.81% was recently reported based on measurements of nine biogas upgrading units located in Denmark (Kvist and Aryal, 2019). The highest (1.97%) CH_4 slip was detected in the water scrubber methane upgrading technology, while the lowest (0.04%) CH_4 loss was detected in an amine based chemical scrubber (Kvist and Aryal, 2019). At five of the plants (agricultural plants 3, 4, 5, 7 and 11) the generated biogas is used (or partly used) in a CHP located off site. Liebetrau et al. (2013) found biogas co-generation units to emit on average 1.74% of the utilized methane with losses ranging from 0.40 to 3.28% (based on measurements at 10 biogas plants).

3.2. Contribution of methane emissions to the overall CO_2 footprint

Applying the methodology described in Section 2.4, the importance of various levels of CH_4 loss on the environmental performance of an agricultural biogas plant was assessed (Fig. 2). The impact in terms of GHG emissions (reported in CO_2-eqvivalents) of the different levels of CH_4 loss was assessed for three scenarios. In scenario A, biogas is upgraded to biomethane and substitutes for natural gas. In scenario B, biogas is utilised in a CHP unit, whereby electricity is supplied to the grid, and heat is used for district heating. In scenario B, the average emission factor regarding the production and consumption of electricity was used, as described in Section 2.4. In scenario C, biogas is also used in a CHP unit, but here the marginal emission factor for the production and consumption of electricity (Section 2.4) was used, meaning in this case that electricity production replaces more fossil fuel.

In all scenarios, CH_4 losses from biogas plants had a significant effect on the overall CO_2 footprint (Fig. 2). At 5% loss, CH_4 emissions make a greater contribution to the CO_2 footprint burden (positive CO_2 emission) compared to the other individual positively contributing emissions, namely energy consumption and the transportation of feedstock and digestate in all scenarios.

In scenarios A and B, a CH_4 loss of 20% caused the net GHG emissions to be positive, meaning that the biogas plant can be considered a net emitter of GHG, despite the substitution of fossil fuels, the reduction of GHG from manure storage and the substitution of chemical fertiliser. This is seen similarly in Table 5, where emission factors are listed for the three scenarios and five levels of CH_4 loss. These emission factors are the calculated net GHG emissions of the biogas production per one tonne of feedstock (wet weight) derived from the calculation example described in Section 2.4. The emission factors vary significantly in cases where CH_4 loss is relatively low (1–2%), to cases where the loss is relatively high (10–20%). The results also show that the emission factors in scenario B (CHP, average) vary highly in comparison to scenario C (CHP, marginal). The cause of this difference is the much lower electricity emission factor in the average mix of electricity sources (0.053 kg CO_2-eq. MJ^{-1}) compared to the marginal emission factor (0.24 kg CO_2-eq. MJ^{-1}) (Table 3).

The average CH_4 emission from the 13 agricultural biogas plants equated to 2.4% of the daily plant production (Table 4). Comparing this average CH_4 emission to implications on the total CO_2 foot-

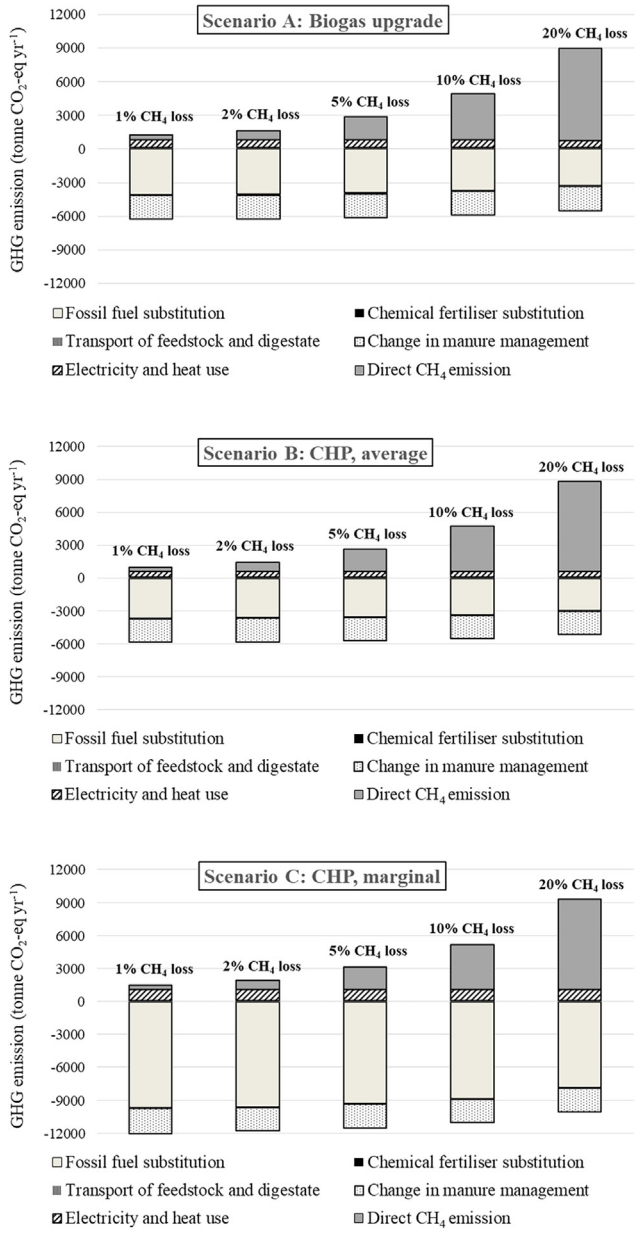

Fig. 2. Greenhouse gas (GHG) emissions calculated for an agricultural biogas plant, considering different biogas utilisation scenarios and five levels of methane (CH_4) loss. CHP: Combined heat and power.

print shown in Fig. 2, this relatively low loss indicates that the production of biogas is a net benefit with regards to GHG emissions. Since CH_4 emission rates compared to production varied greatly between biogas plants (Table 4), it is also likely that the CO_2 footprint of each individual plant will do so, too. Biogas plants, where the loss is particularly high (more than ~15%), may be net emitters of GHG, which underlines the importance of minimising CH_4 emissions from these facilities.

In this study, the carbon footprint of WWTPs was not determined, mainly because the primary purpose of a WWTP is not biogas generation but wastewater treatment, which implies that the services provided by the two types of plants are not comparable. Furthermore, not only CH_4 but also N_2O (another potent GHG) is emitted from WWTP,s primarily from the water treatment line, which needs to be included in footprint calculations. For an evaluation of the carbon footprint for biogas plants located at WWTPs, we instead refer to a recent study by Delre et al. (2019), which assessed carbon footprints for seven Scandinavian WWTPs, including some of the plants in this study. The study showed net carbon footprint values between 0.15 and 0.66 kg CO_2 eq. (Mg of input material)$^{-1}$, depending on the treatment facility. Direct CH_4 and N_2O emissions were the main contributors to the carbon footprint, accounting for between 44 and 71% of the total emission burden (Delre et al., 2019).

3.3. Fugitive methane emissions from Danish biogas production

Danish biogas producing facilities can be divided into four categories: agricultural (centralised and farm-scale) biogas plants (mainly treating manure), industrial biogas plants, wastewater treatment biogas plants (treating sewage sludge) and landfill gas. In total, 165 biogas-producing plants exist in the form of 82 agricultural (28 centralised and 54 farm-based), 51 wastewater treatment biogas, five industrial biogas and 27 landfill gas facilities (Danish Energy Agency, 2017b). The production of biogas has increased from 266 TJ (~5328 tonnes of CH_4) in 1990, to 7899 TJ (~157,985 tonnes of CH_4) in 2016 (Nielsen et al., 2018). In 2016, 86% of the generated biogas was based on manure/organic waste, 12% on sludge from wastewater treatment and only 2% came from landfills (Nielsen et al., 2018). Biogas production at the plants from which emissions were measured in this study represented about between 41% (agricultural) and 45% (WWTPs) of the annual Danish total (in 2016). National CH_4 emissions from Danish biogas production were estimated by applying the measured CH_4 emission factors to nationally generated CH_4 production, distinguishing between emission factors from agricultural and WWTP biogas plants, respectively. Two sets of CH_4 emission factors were used: a plant average and a weighted production average. The plant average was an average of CH_4 losses measured at the plants (sum of CH_4 losses divided by the number of plants), whereas the weighted production average was the sum of all CH_4 emission rates divided by the sum of all plants' CH_4 production rates (cf. Table 4). The plant average represents the biogas technology, whereas the weighted production average represents the combined biogas production in Denmark. Table 6 shows the estimated national CH_4 emissions (tonnes CH_4) for agricultural and WWTP biogas plants, and the total. The total estimated CH_4 emissions are between 3409 and 4683 tonnes, with emissions from agricultural biogas plants making up 6870%, while 30–32% originate from WWTPs (Table 6).

The 2006 IPCC Guidelines consider emissions from biogas plants (anaerobic digestion) as part of the waste sector. According to the IPCC Guidelines, emissions of CH_4 from biogas facilities, due to unintentional leakages during process disturbances or other unexpected events, will generally be between 0 and 10% of the

Table 5
Greenhouse gas emission factors (kg CO_2-eq tonne feedstock^{-1}) calculated for different biogas utilisation scenarios and five levels of CH_4 loss. A negative value implies an overall benefit to the environment, while a positive value implies an overall burden to the environment.

Scenario	1% loss	2% loss	5% loss	10% loss	20% loss
	(kg CO_2-eq tonne feedstock^{-1})				
Scenario A: Biogas upgrade	−44.6	−40.7	−29.0	−9.4	29.7
Scenario B: CHP, average	−42.1	−38.2	−26.5	−7.0	32.0
Scenario C: CHP, marginal	−89.7	−85.3	−72.0	−49.9	−5.7

Table 6
Estimated national CH_4 emissions from the anaerobic digestion of organic waste in agricultural biogas plants and biogas plants at wastewater treatment plants (WWTPs) in 2016 (excluding landfill gas). Numbers in brackets give the percentage out of total CH_4 emissions (excluding landfill gas).

Biogas plant type	Agricultural biogas plants	WWTP biogas plants	Total
CH_4 production, tonnes	135,867	18,958	154,825
CH_4 emission, tonnes (Plant average; $EF_{Agricultural}$ = 2.4% and EF_{WWTP} = 7.5%)	3261 (70%)	1422 (30%)	4683
CH_4 emission, tonnes (Production average; $EF_{Agricultural}$ = 1.7% and EF_{WWTP} = 5.8%)	2310 (68%)	1100 (32%)	3409

amount of CH_4 generated. In the absence of further information, a default value of 5% for the CH_4 emissions should be used (Eggleston et al., 2006).

CH_4 emissions from biogas production are reported in the Danish national greenhouse gas inventory as being a part of the waste sector's GHG emissions (Nielsen et al., 2018). CH_4 emissions were reported at 6635 tonnes in 2016, using an average adopted emission factor (EF) set equal to 4.2% for all types of biogas plants. This emission factor was based on a Danish project where CH_4 leakages were measured at nine biogas plants in Denmark, using on-site point measurement methods (Danish Energy Agency, 2015). Five of the plants were small, single-farm plants, while the other four were larger, centralised agricultural plants. The results were that the CH_4 losses varied from nil to 10% of production, resulting in a weighted average of 4.2%, which was adopted in the national inventory reporting for biogas production independently of the type of biogas plant. Our study shows a lower emission factor from agricultural plants, whereas the emission factor from biogas plants at WWTPs is higher than 4.2%. However, as the share of biogas generated at WWTPs is lower (12%) in comparison to agricultural plants (86%), the combined CH_4 emissions from these two types of facilities are almost comparable, resulting in a national emission of 3409 to 4683 tonnes of CH_4, which is close to the nationally reported figure.

The Danish Biogas Association is a trade organisation representing the Danish biogas sector, with members including plant owners, suppliers, agriculture and energy companies. Within the last few years, this organisation has initiated a voluntary measurement programme with the aim of keeping CH_4 loss at a minimum via a target of 1% loss for the sector. Our results indicate that some improvements are needed to reach this goal. However, the production weighed average loss was just 1.7% for the agricultural biogas plants, where most gas is produced and where production capacity is expanding, and thus the 1% target for the sector as a whole seems to be within reach. However, at plant level, emission rates are higher.

4. Conclusions

Methane losses were measured at 23 biogas plants and found to vary between 0.4 and 15.0% of the production total. Comparing those measured losses to an evaluation of the impact of methane loss on the overall carbon footprint of biogas production, it may be the case that methane loss is the largest positive contributor to greenhouse gas emissions for many biogas plants compared to other factors, such as energy use and the transportation of biomass.

Acknowledgements

This paper presents results from several research projects as well as measurements performed to develop a voluntary measurement programme to monitor CH_4 emissions from biogas plants in Denmark. The Danish Energy Agency, the COWI foundation and the owners of some the biogas plants included in this study are acknowledged for funding the measurements. We thank plant managers and personnel for their cooperation.

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Attachment 3

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At scale, renewable natural gas systems could be climate intensive: the influence of methane feedstock and leakage rates

To cite this article: Emily Grubert 2020 *Environ. Res. Lett.* **15** 084041

Environmental Research Letters

LETTER

At scale, renewable natural gas systems could be climate intensive: the influence of methane feedstock and leakage rates

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Keywords: methane, renewable natural gas, climate change, methane leakage, flaring, environmental assessment

Supplementary material for this article is available online

OPEN ACCESS

RECEIVED
3 February 2020

REVISED
24 April 2020

ACCEPTED FOR PUBLICATION
14 May 2020

PUBLISHED
11 August 2020

Abstract

Renewable natural gas (RNG) is a fuel comprised of essentially pure methane, usually derived from climate-neutral (e.g. biogenic or captured) carbon dioxide (CO_2). RNG is proposed as a climate friendly direct substitute for fossil natural gas (FNG), with the goal of enabling diverse natural gas users to continue operating without substantial infrastructure overhauls. The assumption that such substitution is climate friendly relies on a major condition that is unlikely to be met: namely, that RNG is manufactured from waste methane that would otherwise have been emitted to the atmosphere. In practice, capturable waste methane is extremely limited and is more likely to be diverted from a flare than from direct atmospheric release in a climate-conscious policy context, which means that RNG systems need to be more destructively efficient than a flare to provide climate benefits versus the likely alternative management strategy. Assuming demand levels consistent with the goal of using existing FNG infrastructure, RNG is likely to be derived from methane that is either intentionally produced or diverted from a flare, so essentially any methane leakage is climate additional. Further, in a decarbonizing system, RNG will likely compete with lower-emissions resources than FNG and thus provides fewer net emissions benefits over time. Anticipated leakage is climatically significant: literature estimates for methane leakage from biogas production and upgrading facilities suggest that leakage is in the 2%–4% range (mass basis), up to as much as 15%. Policy makers should consider that under reasonable leakage and demand assumptions, RNG could be climate intensive.

1. Introduction

Climate change motivates an urgent global transition away from the use of fossil fuels for energy (Intergovernmental Panel on Climate Change 2014, Geels *et al* 2017, Mccauley and Heffron 2018, Davis *et al* 2018). Fossil fuels account for 85% of global commercial energy consumption (2018) (BP 2019) and dominate global energy infrastructure. Given the scale, costs, and economic implications of abandoning infrastructure before the end of its useful life, and given the challenge of transitioning energy systems quickly, there is substantial interest in the idea of renewable drop-in fuels (Rye *et al* 2010, Horvath 2016, Lynd 2017) that can use existing infrastructure without creating the problems of fossil fuel use. This interest is particularly salient in the context of end uses that use specific fossil fuels directly. For example, transportation services currently rely primarily on refined oil, and many industrial and other heating applications directly burn natural gas. These direct users of fossil fuels are often unable to accommodate alternative fuels without abandoning functional infrastructure (e.g. internal combustion engine cars and natural gas-fired water heaters) in favor of infrastructure compatible with the new fuels (e.g. electric cars and electric water heaters).

This work assesses renewable natural gas (RNG) (Götz *et al* 2016, Gasper and Searchinger 2018), here referring both to biomethane (Parker *et al* 2017,

Paolini *et al* 2018) and power-to-gas (Götz *et al* 2016, Collet *et al* 2017), as a direct substitute for fossil natural gas (FNG). Drop-in substitutes for FNG specifically are valued due to the diversity of uses, and thus infrastructure, for FNG. In the United States (US), FNG accounts for about 30% of primary energy consumption (EIA 2018), split relatively evenly among power generation (~40%), industrial uses (~30%), and commercial and residential uses (~30%) (Energy Information Administration 2020). RNG has been proposed as a way to decarbonize this system while leveraging existing fossil infrastructure, including pipelines and end use equipment like home and industrial heating devices (Washington State University Energy Program 2018, Bataille 2019). This substitution is particularly relevant for non-electricity uses because they are often more difficult to decarbonize (Davis *et al* 2018, Bataille 2019), though RNG is also valued as an electricity fuel because RNG plants could provide fully dispatchable electricity generation that could reduce the need for costly electricity storage or demand management (Tarroja *et al* 2020). Similarly, like hydrogen, RNG manufacturing has been proposed as a sink for excess variable electricity that can be stored for later use, in the form of power-to-gas schemas (Götz *et al* 2016).

Like FNG, RNG is primarily methane (Gasper and Searchinger 2018), a potent greenhouse gas (GHG) (Intergovernmental Panel on Climate Change 2014) second only to carbon dioxide (CO_2) in its overall contribution to climate change (Weyant *et al* 2006). When RNG is produced from waste methane, converting it to CO_2 by burning it has climate benefits because of methane's much higher climate forcing potential (Intergovernmental Panel on Climate Change 2014) relative to CO_2. If the waste methane were going to be emitted to the atmosphere anyway, any system leakage (i.e. methane emissions) is a lost opportunity but not a climate stressor; otherwise, it contributes to climate change. This analysis shows that 1) RNG from intentionally produced methane, even from climate-neutral CO_2 sources, has substantial climate impacts at methane leakage levels observed in the existing, mature biogas industry (Pertl *et al* 2010, Flesch *et al* 2011, Whiting and Azapagic 2014, Ravina and Genon 2015, Hijazi *et al* 2016, Liebetrau *et al* 2017, Paolini *et al* 2018, Vo *et al* 2018, Ramírez-Islas *et al* 2020); (2) for any meaningful system scale, RNG is likely to be derived from intentionally produced methane; and (3) even RNG from waste methane can have negative climate impacts relative to the most likely alternative of flaring, not venting, the methane when leakage from RNG production and use exceeds flaring loss rates.

2. Methods

This analysis evaluates the GHG intensity of RNG, focused on three methane feedstock pathways for RNG production: (1) from waste methane that would have otherwise been emitted to the atmosphere; (2) from waste methane that would have otherwise been flared; and (3) from intentionally created methane that would otherwise not have existed. The carbon for RNG is assumed to be climate neutral, for example, biogenic or sourced from a carbon capture activity. Reported GHG intensities use IPCC's Fifth Assessment Report (AR5) 20- and 100-year GWPs with climate-carbon feedback, distinguishing between fossil and nonfossil methane GWPs (see Working Group 1, chapter 8, Tables 8.7 and 8.A.1). For comparison, the GHG intensity of FNG, generic resources with life cycle 2050 GHG intensity consistent with 2 °C warming (Pehl *et al* 2017), and zero carbon resources are also included. Full details and calculations are available in the Supplementary Data File.

The absolute GHG intensity of RNG is assumed to derive from methane leakage only (because combustion GHG emissions for RNG are climate neutral by assumption), drawing the system boundary at the point when the methane is diverted from the alternative management strategy (venting, flaring, or not existing) and excluding embodied GHGs in infrastructure or any of the production feedstocks. For example, power-to-gas pathways are implicitly assumed to use GHG-neutral power in facilities with zero embodied GHGs. GHG intensity is given as kilograms (kg) of carbon dioxide equivalent (CO_2e) per gigajoule (GJ) of methane consumed—that is, the denominator is the amount of methane that is ultimately delivered to the entity that combusts it, which is less than the amount of methane that is produced or withdrawn if system leakage exceeds 0%. Emissions associated with leakage are thus calculated as follows:

$$\begin{aligned}&\text{absolute leakage – related GHG intensity} = \\ &(\text{mass } CH_4 \text{ produced} \times \text{system leakage} \times GWP_{CH_4}) \\ &= \frac{\text{mass } CH_4 \text{ delivered}}{(1 - \text{system leakage})} \times \text{system leakage} \times GWP_{CH_4}\end{aligned} \quad (1)$$

where system leakage is mass methane emitted/mass methane produced. Net emissions relative to the alternative fate for methane are calculated by subtracting the counterfactual methane emissions. For Path 1 (waste methane would have otherwise been emitted to the atmosphere), counterfactual emissions are that system leakage = 100%. For Path 2 (waste methane would have otherwise been flared), counterfactual emissions are that system leakage = (1-flare efficiency). For Path 3 (methane would not otherwise have existed), counterfactual emissions are 0. Thus, net emissions are given as:

$$\begin{aligned}&\text{net leakage emissions} = (\text{mass } CH_4 \text{ produced} \\ &\times GWP_{CH_4}) \times (\text{system leakage} - \text{counterfactual leakage})\end{aligned} \quad (2)$$

For the three paths, Equation 2 becomes:

$$\text{Path 1, net leakage emissions} = (\text{mass } CH_4 \text{ produced} \times GWP_{CH_4}) \times (\textit{system leakage} - 1) \quad (3)$$

$$\text{Path 2, net leakage emissions} = (\text{mass } CH_4 \text{ produced} \times GWP_{CH_4}) \times (\text{system leakage} - \text{flare leakage}) \quad (4)$$

$$\text{Path 3, net leakage emissions} = (\text{mass } CH_4 \text{ produced} \times GWP_{CH_4}) \times (\text{system leakage} - 0) \quad (5)$$

Note that because of the presentation of results per unit of methane combusted by a user and the fact that methane production = methane delivered/(1—system leakage) (equation (1)), equation (3) reduces to (mass CH_4 delivered $\times -GWP_{CH_4}$) and is independent of leakage rate. For Path 2, emissions upstream of the flare are not considered because waste methane that is vented prior to diversion to the flare is not diverted to the flare, thus falling under Path 1.

Leakage from the RNG system is evaluated as a range because of substantial uncertainty about what leakage levels would be under future conditions, particularly if newer RNG pathways (e.g. power-to-gas) became widespread. This work considers the implications of RNG system leakage between 0%–15% mass leakage/mass produced in order to inform consideration of potential RNG futures. The range is most proximately based on Scheutz and Fredenslund's (2019) evaluation of 23 biogas plants, including seven facilities encompassing production through biogas upgrading to biomethane, where facility leakages from 0.4 to 14.9% of production were observed (see Supplementary Data File for details). Specific leakage sources are not always evident but might be correlated with plant complexity (e.g. number of units), maintenance regimes, and the status of biogas production as a core or non-core function (Scheutz and Fredenslund 2019). Published values in other studies and GHG protocols reflect ranges generally narrower than but consistent with Scheutz and Fredenslund's findings (Flesch *et al* 2011, Liebetrau *et al* 2013, 2017, Hrad *et al* 2015, Vo *et al* 2018, Bartoli *et al* 2019).

Leakage downstream of production and processing (i.e. during transportation, storage, and end use) is assumed to be identical for FNG and RNG. Although it is challenging to assign a value for these processes due to the diversity of end uses, lack of information about leakage during end uses, differential use of transmission, storage, and distribution by end users, and the dependence of transportation leakage on distance, a value of 0.8% (mass leaked per mass withdrawn or produced) was chosen in service of estimating absolute GHG intensities for comparison with zero-GHG systems. This value is based on assumptions and data from the literature (Liebetrau *et al* 2013, Lavoie *et al* 2017, Alvarez *et al* 2018) (see Supplementary Data File for details). For FNG, this value is added to an estimate of production and processing leakage of 2.1% mass leakage/mass withdrawn (Alvarez *et al* 2018), and for RNG, it is assumed to be included in the assessed 0%–15% range for full system potential leakage rates.

3. Results

3.1. GHG intensity of renewable natural gas

Table 1 shows the estimated GHG intensity of RNG for three production pathways as a function of system methane leakage: (1) RNG produced from waste methane that would have otherwise been emitted to the atmosphere (Path 1); (2) RNG produced from waste methane that would have otherwise been flared with 99% destructive efficiency (Path 2); and (3) RNG produced from intentionally created methane (Path 3).

In all cases, this analysis assumes that the CO_2 emitted from burning RNG is climate neutral for the RNG user, e.g. because it is sourced from biogenic or captured carbon that was taken up from and returned to the atmosphere over a period of time that is short from a climate perspective. Further, to emphasize the particular challenge posed by methane leakage, this analysis assumes RNG has no GHG intensity other than that associated with net impacts from methane leakage or destruction—that is, inputs to RNG production like electricity, hydrogen, and support infrastructure are assumed to be climate neutral. This assumption is consistent with the notion that zero-GHG electricity or hydrogen are potential alternatives to RNG. Note that because conversion processes are never 100% efficient, any GHG intensity for RNG associated with electricity or hydrogen inputs would exceed that of the electricity or hydrogen available for use.

As table 1 shows, the GHG intensity of RNG is driven by the counterfactual—that is, what would have otherwise happened to the source methane. Path 1, waste methane diversion from the atmosphere, is highly GHG negative because the counterfactual is that all utilized methane would have been emitted as methane. Leakage is irrelevant to GHG impact per unit of utilized methane because any leaks are methane that would have escaped anyway. Although Path 2 also uses waste methane, the counterfactual is that the waste methane would have been nonproductively burned in a flare, so RNG is GHG negative in this case only if the RNG system's total leakage is lower than leakage from the flare (1%), which is unlikely given that a best-guess estimate of downstream emissions alone is 0.8%. Path 3 uses intentionally produced methane. Here, the counterfactual is that no methane would have been released to the atmosphere, so any system leakage is GHG positive.

Table 1. Renewable natural gas carbon dioxide equivalent intensity by pathway, assuming climate-neutral combustion emissions of carbon dioxide.

(a) GWP-100, kg CO2e/GJ methane productively consumed							
system leakage (mass CH4 emitted/mass CH4 produced)	0	0.025	0.05	0.075	0.1	0.125	0.15
Path 1: Waste methane diverted from emission to atmosphere	−680	−680	−680	−680	−680	−680	−680
Path 2: Waste methane diverted from a 99% efficient flare	−7	10	29	48	68	89	112
Path 3: Intentionally produced methane used	0	17	36	55	76	97	120
(b) GWP-20. kg CO2e/GJ methane productively consumed							
system leakage (mass CH4 emitted/mass CH4 produced)	0	0.025	0.05	0.075	0.1	0.125	0.15
Path 1: Waste methane diverted from emission to atmosphere	−1720	−1720	−1720	−1720	−1720	−1720	−1720
Path 2: Waste methane diverted from a 99% efficient flare	−17	26	72	121	172	226	283
Path 3: Intentionally produced methane used	0	44	91	139	191	246	304

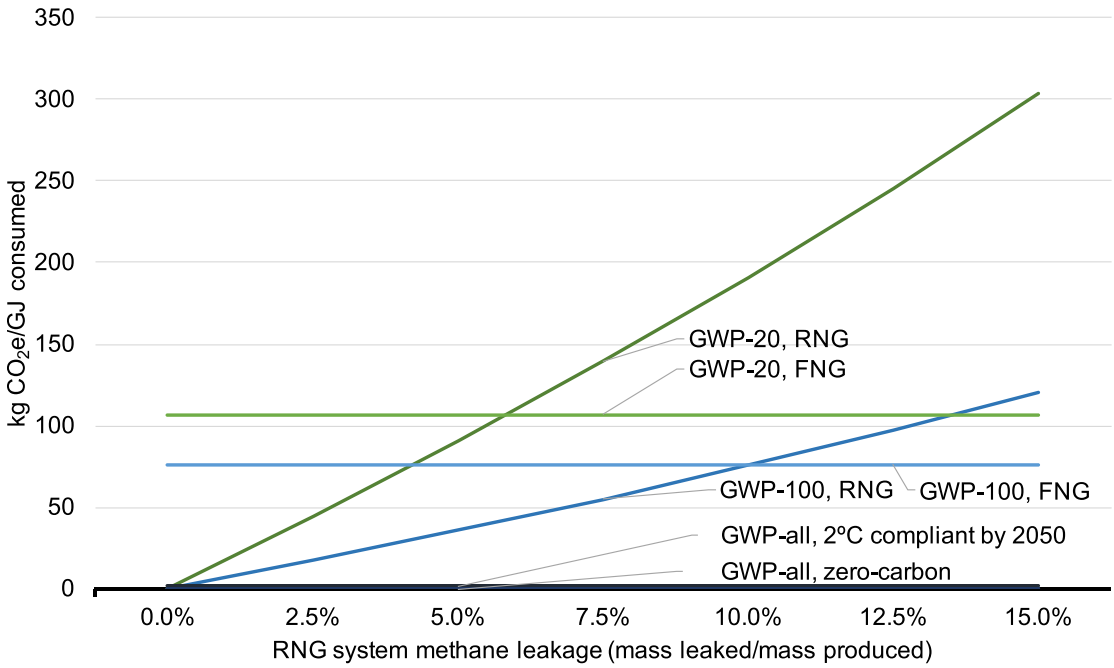

Figure 1. Greenhouse gas intensity of renewable natural gas from intentionally produced methane as a function of system methane leakage. GWP-20 and GWP-100 are shown for intentionally produced RNG for RNG system methane leakage of 0–15%, with GWP-20 and GWP-100 of FNG shown for reference.

3.2. Intentionally produced methane for RNG

A major finding of this analysis is that, as with FNG (Brandt *et al* 2014, Alvarez *et al* 2018, Grubert and Brandt 2019, Zhou *et al* 2019), RNG can have significant climate impacts associated with system methane leakage if the methane is intentionally produced (Path 3; table 1). Figure 1 shows the GHG intensity (kg CO_2e/GJ CH_4 productively consumed, e.g. for heat or electricity generation) of RNG from intentionally produced methane as a function of RNG system methane leakage.

As figure 1 shows, RNG from intentionally produced methane is always GHG positive unless total system leakage is 0. Given demonstrated transportation and end use leakage values on the order of 0.4–0.8% (Liebetrau *et al* 2013, Lavoie *et al* 2017, Alvarez *et al* 2018), RNG from intentionally produced methane cannot outperform zero-GHG hydrogen or electricity systems on GHG intensity. Although this analysis does not consider non-operational, non-methane GHGs, note that both hydrogen and electricity are likely inputs to intentionally produced methane for RNG, which therefore inherits and amplifies embodied emissions. The estimated methane-only GHG footprint of such RNG exceeds the combustion plus methane leakage GHG footprint of FNG when RNG system leakage is higher than about 10% (GWP-100) or 6% (GWP-20) on a mass leaked per mass produced basis. Accounting for IPCC stated uncertainty in the GWP of methane (Intergovernmental Panel on Climate Change 2014), the estimated leakage range within which RNG becomes more GHG intensive than FNG is about 9.1–11.1% (GWP-100) or 5.0–6.6% (GWP-20). Although power-to-gas systems and evaluations remain rare enough that data on leakage are not widely available (though leakage has been discussed (Vo *et al* 2018)), such leakage rates—particularly for a full system—are not uncommon

for biogas to RNG systems (Liebetrau *et al* 2013, 2017, Scheutz and Fredenslund 2019). For electricity, assuming the heat rate of a US natural gas combined cycle power plant and GWP-100, RNG's operational methane GHG intensity surpasses the 15 kg CO_2e/MWh total life cycle 2050 GHG intensity consistent with a 2 °C warming limit (Pehl *et al* 2017) for system leakage of 0.3%, which is less than some observed leakage from power plants alone (Lavoie *et al* 2017). Calculations can be found in the Supplementary Data File.

3.3. At scale, most methane feedstocks for RNG would likely be intentionally produced

How much RNG is likely to come from intentionally produced methane, which includes all power-to-gas RNG and RNG produced from feedstocks that would not degrade anaerobically (i.e. to methane rather than CO_2) absent intentional intervention (Meyer-Aurich *et al* 2012, Börjesson *et al* 2015, Agostini *et al* 2015)? The answer depends on assumptions about total demand and the availability of waste methane for diversion to RNG production. If the goal is to maintain the usefulness of FNG infrastructure, one potential assumption for RNG demand is that it would match current FNG demand. In 2017, US consumer consumption was 27.2 exajoules (EJ) of FNG, including 10.1 EJ for electric power and 8.7 EJ for often difficult-to-decarbonize industrial uses (see Supplementary Data File). The energy content of 2017 US uncontrolled methane emissions was about 1.6 EJ/year, about 0.3 EJ of which were emitted from biogenic sources (as opposed to, say, the FNG system) that could reasonably be captured (wastewater treatment plants and landfills, not enteric fermentation) (US EPA 2019, Grubert 2020), not all of which would become consumable RNG (i.e. due to parasitic energy requirements, conversion losses, etc). Thus, although some capturable waste methane (Paths 1 and 2) clearly exists, the degree to which RNG systems can depend on such resources at scale is low (<1%) relative to current natural gas demand.

One important observation for contextualizing these values is that not all methane from waste is waste methane. For example, the National Renewable Energy Laboratory (NREL) estimates that the energy content potential from methanogenic US wastes is about 5% the size of the US natural gas system (National Renewable Energy Laboratory 2013), including a methane potential estimate from wastewater (2.3 million tonnes/year) (National Renewable Energy Laboratory 2013) that is four times the US Greenhouse Gas Inventory (GHGI) estimate for methane emissions from wastewater treatment plants as of 2017 (0.57 million tonnes/year, Table 2–2) (US EPA 2019). Why? Unintentionally produced waste methane typically results from natural anaerobic digestion of wet organic wastes, like animal manure, sewage, and landfilled wastes, but this digestion process does not completely convert carbon wastes to methane. Rather, digestion produces biogas, a blend of methane and CO_2 that can then be upgraded into near-pure biomethane, a form of RNG. Crucially, in part because biogas and biomethane can generate revenue, it is not only possible but expected to intervene in biological systems to increase methane production beyond what would have happened anyway when there is an incentive to do so (Hijazi *et al* 2016, Ferreira *et al* 2019, Garcia *et al* 2019). Thus, a single facility might produce both Path 1 (GHG-negative) and Path 3 (GHG-positive) methane from the same wastes.

Despite its limited availability, Path 1 methane is so GHG negative (table 1) that it is reasonable to investigate whether climate benefits can be retained if small amounts of very climate-negative RNG are blended with RNG from intentionally produced methane. Figure 2 shows GHG intensity of RNG for blends of 0%–100% Path 3 methane (intentionally produced) with Path 1 methane (waste methane diverted from release).

Figure 2 suggests that blending very GHG-negative RNG with GHG-positive RNG can enable a fairly large RNG system that is overall at least somewhat GHG-negative, assuming leakage levels within a typically observed range (see Supplementary Data File for detailed calculations and values). Assuming all 0.3 EJ of uncontrolled methane emissions from landfills and wastewater treatment plants could be captured and converted to consumer-ready RNG, either current industrial demand or electric power demand for FNG could be fulfilled by an RNG system with up to about 3% system leakage and theoretically remain GHG-negative (GWP-100; see Supplementary Data File). As the next section shows, however, such an outcome is unlikely because of the actual nature of waste methane management.

3.4. Capturable waste methane would be flared, not vented

The possible conclusion that sufficient highly GHG negative methane exists to support a large (e.g. FNG electricity system-sized) GHG negative RNG system is based on the assumption that waste methane is diverted from emission to the atmosphere (Path 1). This assumption is flawed if one also assumes that GHG emissions reductions are a policy priority, as existing practice is not the appropriate baseline for determining the counterfactual management practice for waste methane that could be available for RNG production (Haya *et al* 2019). Specifically, if the methane can be captured for RNG production, it can be captured for diversion to a flare, and it is unrealistic to assume that capturable methane would be vented under a GHG conscious policy regime. Even without federal climate regulation, the US regulates methane emissions from new landfills (US EPA 2016),

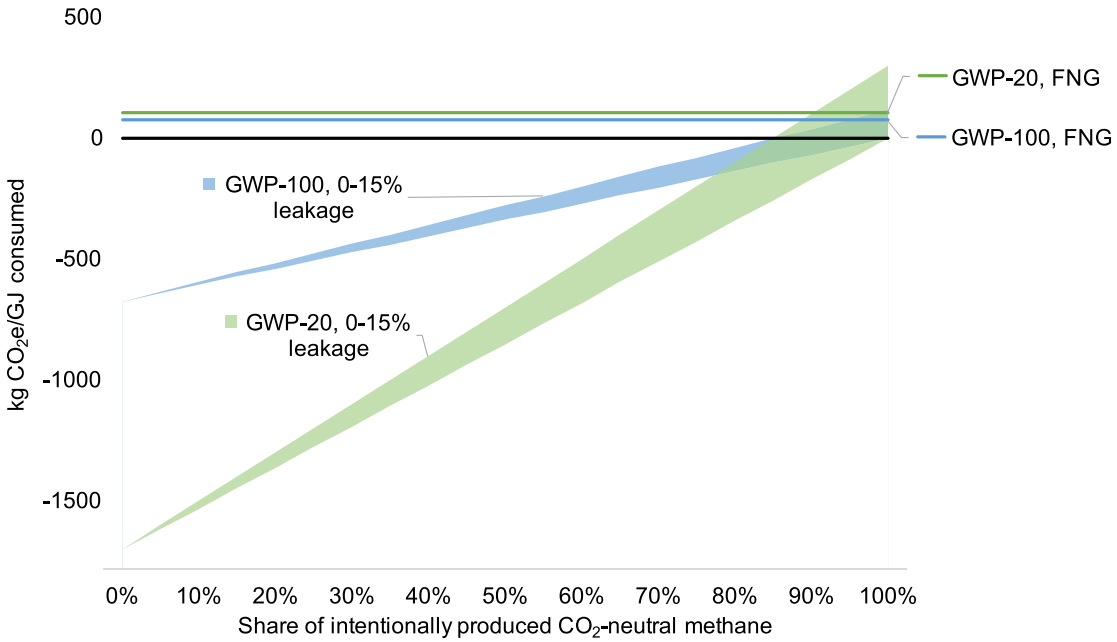

Figure 2. Greenhouse gas intensity of RNG systems that blend waste methane that would otherwise have been released to the atmosphere with intentionally produced methane, for 0%–15% system leakage. Lower bound of shaded areas = 0% system methane leakage for RNG; upper bound of shaded areas = 15% system methane leakage for RNG. GWP-20 and GWP-100 for RNG and FNG are shown for blends of 0% to 100% RNG from intentionally produced methane.

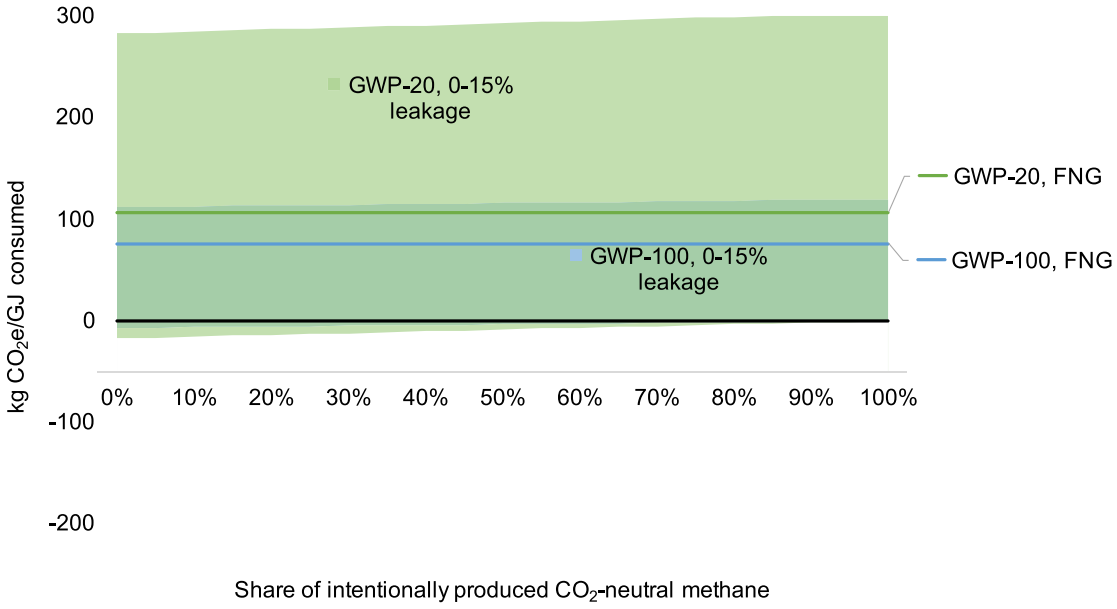

Figure 3. Greenhouse gas intensity of RNG systems that blend waste methane that would otherwise have been flared with intentionally produced methane, for 0%–15% system leakage. Panel A: Flare with 99% destruction efficiency. Panel B: Flare with 86% destruction efficiency. Lower bound of shaded areas = 0% system methane leakage for RNG; upper bound of shaded areas = 15% system methane leakage for RNG. GWP-20 and GWP-100 for RNG and FNG are shown for blends of 0% to 100% RNG from intentionally produced methane.

and many methanogenic facilities use methane capture with flares for safety reasons. Flaring destroys the methane with the same destructive benefit as combusting the methane productively. Figure 3 updates the assumptions used in figure 2 to show the same results, but assuming that RNG using Path 3 (intentionally produced) methane is blended with RNG using Path 2 (waste diverted from flare) rather than Path 1 (waste diverted from release) methane. Figure 3 assumes 99% flare efficiency, described in the GHGI as a median value (US EPA 2019). Figure S1 (available at stacks.iop.org/ERL/15/084041/mmedia) shows results assuming the GHGI's lower flare efficiency bound of 86 (US EPA 2019).

As figure 3 shows, conclusions about the viability of a large, GHG-negative RNG system change radically when the more realistic counterfactual of methane destruction rather than methane venting is applied. RNG system leakage would need to be essentially 0 (that is, lower than the flare's leakage) to be GHG-negative versus typical flare performance. Based on literature values for leakage, including the estimate of 0.8% leakage for processes downstream of production and processing, productive use of waste methane is unlikely to be more destructively efficient than a flare. Although waste methane being diverted for productive use arguably would not have been captured without the financial incentive of energy sales, given that capture infrastructure is not free, flaring is most likely the less GHG intensive alternative for waste methane once it has been captured. In a decarbonized energy system where RNG would be less likely to be replacing GHG-intensive fuels (and thus offsetting their emissions), and when a policy regime requiring or incentivizing destruction of GHG-intensive wastes might reasonably be expected to be in place, expected levels of methane leakage suggest that RNG is unlikely to be a low GHG energy resource relative to alternatives.

4. Discussion

RNG is not inherently climate friendly. Based on consideration of both the source of methane used to produce RNG and the likely alternative fate of that methane, and using reasonable assumptions about likely system methane leakage, it is unlikely that an RNG system could deliver GHG-negative, or even zero GHG, energy at scale. Substantial GHG benefits can be attained when waste methane is genuinely diverted from emission to the atmosphere, but the availability of such methane is low (Liu and Rajagopal 2019, US EPA 2019) relative to potential demand for climate friendly RNG, especially when considering that the alternative fate of capturable methane is more likely flaring than venting in a GHG-conscious setting. Under some system leakage rates that have been observed for biogas systems (Liebetrau *et al* 2017, Scheutz and Fredenslund 2019), RNG might not even meet the less stringent threshold of outperforming FNG from a GHG perspective.

Designing a system that depends on RNG, or delaying transition to a system that does not depend on natural gas because of the promise of RNG, could delay climate mitigation because of induced demand for intentionally produced methane. Particularly given that past experience demonstrates that policy can rapidly drive resource allocation to RNG (Bartoli *et al* 2019), RNG's environmental performance should be carefully compared with that of its likely long-term competitors—not just FNG—before resources are allocated. Current literature on RNG often assumes the context of a fossil-based system (see e.g. the reference systems for papers included in a review of LCA of biogas production (Hijazi *et al* 2016)), which leads to the crediting of lower environmental burden relative to this context (e.g. when RNG is given credit for avoided GHG impacts from FNG consumption (Scheutz and Fredenslund 2019, Ramírez-Islas *et al* 2020)). Such fossil-linked benefits disappear in a context where RNG could be substituting for zero-GHG alternatives like zero-GHG electricity or hydrogen rather than FNG, petroleum fuels, and GHG-intensive electricity.

Even beyond GHG emissions, environmental burdens associated with RNG that are acceptable relative to FNG merit deeper investigation when the alternative is, e.g. zero-GHG electricity. RNG is designed to be effectively indistinguishable from FNG at the point of use, so local combustion impacts are likely to be similar for clean RNG, and potentially worse for less pure RNG (Paolini *et al* 2018). Upstream of use, RNG would likely have different socioenvironmental impacts than FNG. Although RNG can use existing pipeline and user infrastructure, for example, it would obviate the need for FNG's production infrastructures, which have substantial socioenvironmental impacts (Jacquet *et al* 2018). RNG production facilities using primarily waste products (e.g. agricultural wastes, landfill gas, wastewater treatment gas, excess electricity generation) would likely not qualitatively change socioenvironmental impacts from those activities, though making certain practices financially viable could extend their life and extent (Haya *et al* 2019). Relatedly, having access to RNG could extend the life of existing fossil infrastructure, with mixed socioenvironmental outcomes.

To the extent that RNG facilitates lower impact energy systems, e.g. by avoiding the need for mineral (Sovacool *et al* 2020)- and cost-intensive electricity storage to help match supply and demand (Tarroja *et al* 2020), some of the marginal impacts of RNG could be offset by system benefits. These benefits are not guaranteed, however. As demonstrated by experience with renewable drop-in transportation fuels, the potential for drop-in renewable fuel use might not actually lead to renewable fuel use (Pouliot and Babcock 2017), and the renewable fuels themselves might have undesirable environmental characteristics (Liu and Rajagopal 2019). This work shows that RNG needs to be carefully evaluated in the context of expected long-run system conditions before it is adopted as a component of a zero GHG energy system, particularly given its potential for methane leakage-related climate pollution.

Acknowledgments

This research did not receive any specific grant from funding agencies in the public, commercial, or

not-for-profit sectors. The author thanks two reviewers for their constructive comments.

Data Availability Statement

Any data that support the findings of this study are included within the article.

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