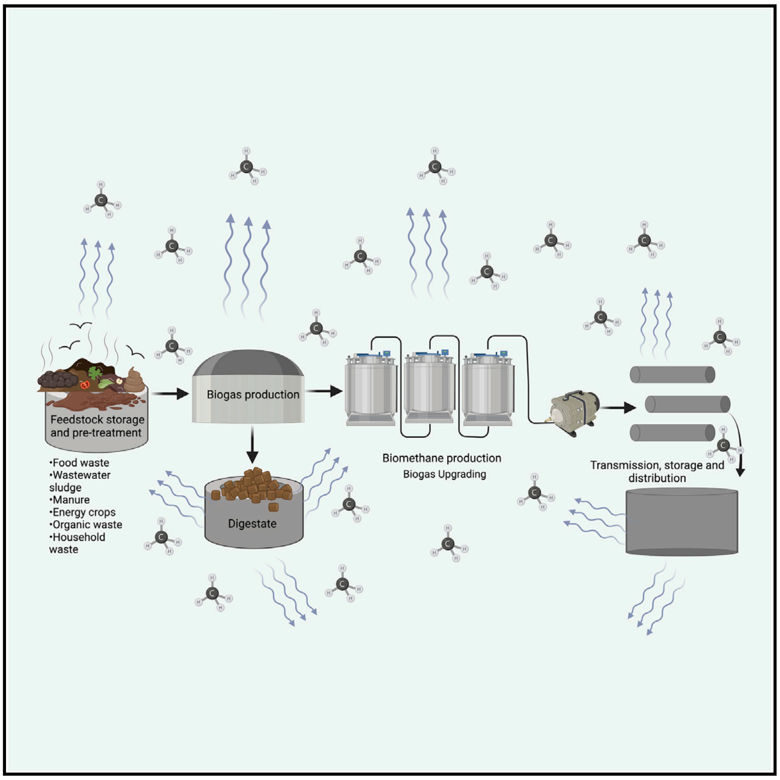
One Earth

Article

# Methane emissions along biomethane and biogas supply chains are underestimated

## Graphical abstract



## Authors

Semra Bakkaloglu, Jasmin Cooper,
Adam Hawkes

## Correspondence

s.bakkaloglu@imperial.ac.uk

## In brief

Biomethane and biogas have emerged as cleaner alternatives for natural gas, as they generate fewer greenhouse-gas emissions. However, their production and distribution can still result in methane emissions, the magnitude of which remains unclear. Here, we evaluate methane emissions throughout the biomethane and biogas supply chains and show that emissions are greater than previously estimated. The digestate stage generated the most $CH_4$, and 62% of total emissions were released by just 5% of emitters.

## Highlights

- The biomethane and biogas supply chain may emit up to 18.5 Tg $CH_4$ per year
- Biomethane and biogas emit much less $CH_4$ than oil and natural gas
- $CH_4$ loss rates in biomethane and biogas supply chain exceed those in oil and natural gas
- The top 5% of emitters account for 62% of $CH_4$ emissions

Bakkaloglu et al., 2022, One Earth 5, 724–736
June 17, 2022
https://doi.org/10.1016/j.oneear.2022.05.012

CellPress
OPEN ACCESS

One Earth

Article

# Methane emissions along biomethane and biogas supply chains are underestimated

Semra Bakkaloglu,[1,2,3,*] Jasmin Cooper,[1,2] and Adam Hawkes[1,2]
[1]Sustainable Gas Institute, Imperial College London, SW7 1NA London, UK
[2]Department of Chemical Engineering, Imperial College London, SW7 2AZ London, UK
[3]Lead contact
*Correspondence: s.bakkaloglu@imperial.ac.uk
https://doi.org/10.1016/j.oneear.2022.05.012

**SCIENCE FOR SOCIETY** An immediate shift away from coal and oil for energy is necessary to limit rising temperatures but is challenging due to energy needs, particularly in areas like heating and cooling that require substantial energy supply all year round. Natural gas is presently being used as a bridging fuel. It delivers the same performance as coal and oil but has lower $CO_2$ emissions. However, natural gas releases methane ($CH_4$), which is a more powerful warming agent than $CO_2$. Biomethane and biogas have emerged as strong candidates to replace gas and lower $CO_2$ and $CH_4$ emissions. However, these replacement fuels are not $CH_4$ emission free. Indeed, $CH_4$ is released at various points during production and distribution, but a thorough understanding of where, when, and how much $CH_4$ is released remains absent. A synthesis and analysis of existing biomethane and biogas $CH_4$ emission data reveal that $CH_4$ emissions throughout the supply chains have been underestimated. The majority of $CH_4$ comes from just a few super-emitters and mainly at the digestate stage. Mitigating $CH_4$ throughout biomethane and biogas supply chains is urgently needed if we are to limit global warming to 1.5°C.

## SUMMARY

Although natural gas generates lower $CO_2$ emissions, gas extraction, processing, and distribution all release methane, which has a greater global warming potential than $CO_2$. Biomethane and biogas that use organic wastes as a feedstock have emerged as alternatives to natural gas, with lower carbon and methane emissions. However, the extent to which methane is still emitted at various stages along biogas and biomethane supply chains remains unclear. Here, we adopt a Monte Carlo approach to systematically synthesize the distribution of methane emissions at each key biomethane and biogas supply chain stage using data collected from the existing literature. We show that the top 5% of emitters are responsible for 62% of emissions. Methane emissions could be more than two times of greater than previously estimated, with the digestate handling stage responsible for the majority of methane released. To ensure the climate benefits of biomethane and biogas production, effective methane-mitigation strategies must be designed and deployed at each supply chain stage.

## INTRODUCTION

As we move further into the 21st century, energy systems must move away from fossil fuels and grow in renewable energy capacity if Paris Agreement temperature targets are to be met. However, due to challenges in adopting low-carbon technologies, certain areas of global energy systems are difficult to decarbonize. These include heavy industry, transport, and heating and cooling systems, which together account for a significant portion of carbon dioxide ($CO_2$) emissions.[1] Natural gas has therefore been used as an important alternative fuel, which can offer large-scale energy supply, especially for domestic space heating and hot water needs, electricity generation, and industrial applications, with much lower $CO_2$ emissions compared with oil and coal. Although replacing oil and coal with natural gas reduces $CO_2$ emissions, fugitive emissions from the supply chain of natural gas—gas extraction, processing, and distribution—can all release $CH_4$. Around 39.6 million tonnes of $CH_4$ were emitted in 2021,[2] representing 61% of oil and gas emissions and 30% of total-energy-sector $CH_4$ emissions. Since $CH_4$ has a much stronger global warming potential than $CO_2$ and is currently responsible for at least one-quarter of global warming, there are strong calls for natural gas use to be reduced by at least 35% by 2050 and 70% by 2100 relative to 2019;[3]

therefore, alternative clean-energy methods are vital to replace natural gas to limit global warming to 1.5°C.

An alternative method of decarbonizing natural gas is via replacing it with biomethane or biogas, which is a mixture of gases (mostly $CH_4$ and $CO_2$) produced from biodegradable materials. Biomethane and biogas production and use have been put forward as part of mitigation efforts,[4] with up to 37 exajoule (EJ)/year of biomass-based gases in Intergovernmental Panel on Climate Change Special Report on Global Warming of 1.5°C (IPCC SR1.5C) scenarios,[5] which limits temperature rises to below 2°C. The International Energy Agency (IEA)[6] reported that global biomethane and biogas production could satisfy nearly 20% of global gas demand if its sustainable potential was fully utilized.[6] Because biomethane is similar to natural gas, it can be easily stored and injected into the existing natural gas infrastructure, potentially providing reliable and affordable energy.[7] At the time of writing, Europe is the world leader in biomethane production by upgrading biogas, followed by the United States, China, and Canada.[8] According to the World Biogas Association (2019), 700 biogas-upgrading plants are operating worldwide, with 195 in Germany (the largest producer), with biogas currently dominating biomethane production. Biomethane and biogas production are expected to grow further, with demand predicted to grow 9-fold by 2040 compared with 2018 levels,[6,9] driven by increases in the volume of organic waste generated by modern societies, changes in waste practices, and the phasing out of fossil fuels aimed at reducing greenhouse gas (GHG) emissions and meeting government targets. Given this host of commitments, investments, and developments, biomethane and biogas could be crucial in helping to establish a clean, reliable, and affordable global energy system.

However, large quantities of $CH_4$ can still be emitted from the biomethane and biogas supply chains, including digestate handling, anaerobic digesters, upgrading units, feedstock storages and transmission, and storage and distribution stages.[4] $CH_4$ is a relatively short-lived GHG but has a global warming potential (GWP) 27.2 ± 11 times larger than $CO_2$ over a 100-year horizon and 80.8 ± 25.8 times larger over a 20-year time horizon for biogenic sources.[10] The importance of reducing $CH_4$ emissions to meet Paris Agreement[11] targets has been demonstrated by Rogelj et al.,[12] as it is an important GHG in terms of potential overshooting of Paris Agreement targets, where warming exceeds "well below 2°C" and then returns to the target level by 2100,[10] leading to potential tipping points in physical and socio-economic systems. The IPCC (Intergovernmental Panel on Climate Change) Sixth Assessment Report (AR6) (Working Group III)[13] highlighted $CH_4$ as playing a significant role in determining whether or when 1.5°C is achieved, as reducing $CH_4$ emissions will offset global temperature increase much more quickly than $CO_2$, due to its relatively short lifetime and higher GHG potency. The AR6 report also noted that reductions to $CH_4$ emissions will need to occur more rapidly than $CO_2$ and that reducing $CH_4$ (and other non-$CO_2$ GHG) emissions is essential for lowering warming.[13] As the AR6 scenarios predict biomethane capacity to increase by up to 200-fold between 2020 and 2050,[14] understanding where $CH_4$ emissions occur and how much is emitted is crucial.

There are some emissions-measurement studies to date focusing on specific biomethane facilities,[4,15–22] which have measured on site (measurement of emissions at each individual point source) and off site (measurement of emissions based on observations made away from the site). These can also be referred to as bottom-up (on-site) and top-down (off-site) studies. These have found that emissions from biomethane facilities can be up to 97 kg $h^{-1}$ $CH_4$.[4,16–24] However, a comprehensive evaluation by characterizing the distribution of $CH_4$ emissions at each biomethane and biogas supply chain stage remains unclear.

Here, we bring together the published emissions data from $CH_4$-measurement studies to assess and synthesize the distribution of emissions from each supply chain stage in order to characterize the emissions profile of the biomethane and biogas supply chain (see experimental procedures and Figure S1 for the selected supply chain route). A Monte Carlo aggregation examines the distribution of supply chain emissions. This allows for the emission profile of biomethane and biogas supply chains to be characterized. We find that, while the biomethane and biogas supply chain emits less $CH_4$ than the oil and natural gas supply chain, the emission rate is higher. Furthermore, we find that 62% of cumulative emissions are released by just the top 5% of emitters. We also find that methane emissions could be more than two times higher than previously estimated, and the digestate-handling stage contributed to the largest $CH_4$ emissions along the supply chain. Our results will allow for a greater understanding of how to improve the sustainability of biomethane and biogas production by providing plant operators, investors in the supply chain, and policymakers with information on where improvements can be made in biomethane and biogas supply chains to reduce $CH_4$ emissions, as well as whether existing or proposed $CH_4$ regulations are sufficient or need to be revised.

## RESULTS

### Method summary

To assess overall supply chain emissions, the biomethane supply chain is divided into five major stages: (1) feedstock; (2) biogas production; (3) biogas upgrading; (4) transmission, distribution, and gas storage; and (5) digestate storage. This study was compiled from several published studies and the data from on-site (taken at each individual emission source) and off-site measurements (reported for the entire site). The kernel density estimation (KDE) function was used to assess the characteristics of the data distribution gathered from individual sources for each stage of the supply chain. Following that, a Monte Carlo simulation was performed to estimate total supply chain emissions, which were then compared with the off-site emissions reported from whole-site measurements in previously published studies (see the experimental procedures for further details).

### Total supply chain emissions

The cumulative distribution of the supply chain $CH_4$ emissions is shown in Figure 1A. Median and mean emissions are 40.0–42.3 g $CO_{2\text{-eq.}}/MJ_{HHV}$ (41.1–41.3 at the 95% confidence interval [CI]) and 51.4–52.7 g $CO_{2\text{-eq.}}/MJ_{HHV}$ (52.2–52.4 at the 95% CI), respectively, with a 5th percentile of 11.0–16.3 g $CO_{2\text{-eq.}}/MJ_{HHV}$ (15.6–15.7 at the 95% CI) and a 95th percentile between 118.2 and 144.0 g $CO_{2\text{-eq.}}/MJ_{HHV}$ (131–133 at the 95% CI) using

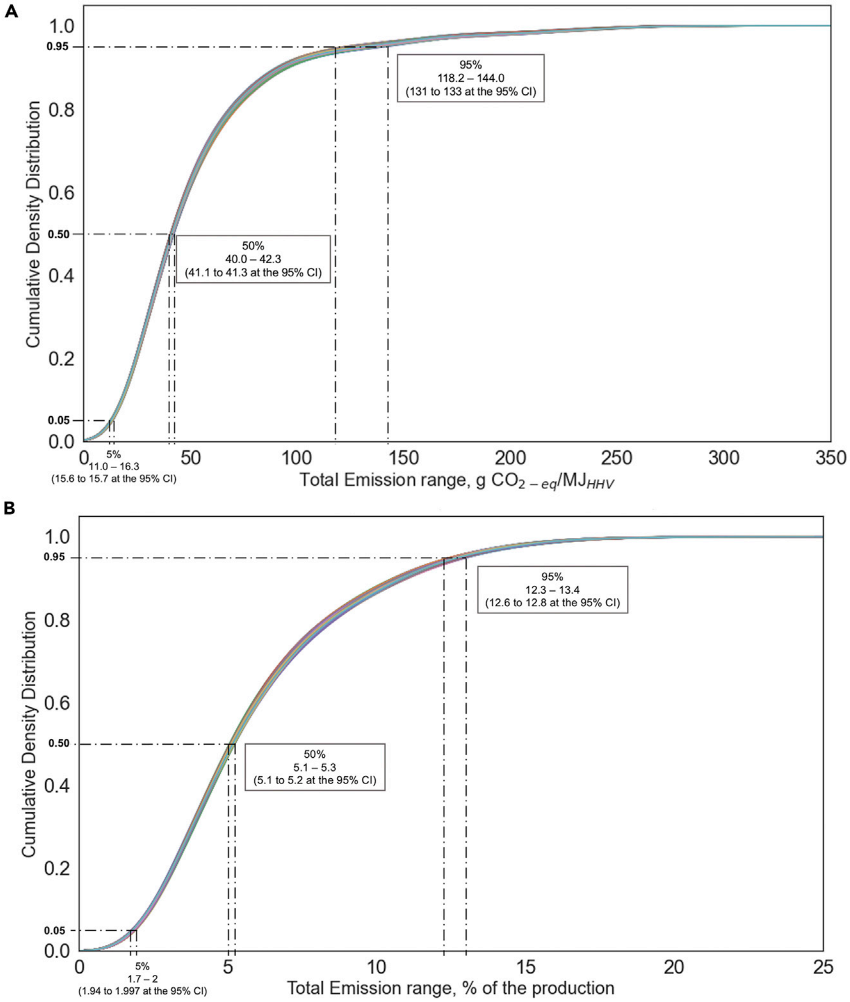


**Figure 1. Cumulative distribution of $CH_4$ emissions from the total supply chain**

(A) Cumulative distribution of total supply chain $CH_4$ emissions for the 10,000 Monte Carlo runs and 100 curves described in the experimental procedures, expressed as g $CO_{2\text{-eq.}}/MJ_{HHV}$.

(B) Cumulative distribution of total supply chain $CH_4$ emissions for the 10,000 Monte Carlo runs and 100 curves, expressed as percentage of total $CH_4$ production.

The range of 5th, 50th, and 95th percentile estimates are shown as dotted black lines. CI: confidence interval.

$GWP_{100}$ values. Each curve defines the cumulative distribution for a single Monte Carlo simulation and shows that total supply chain emissions range from 2.5 to 343 g $CO_{2\text{-eq.}}/MJ_{HHV}$. The emissions distribution is highly upward skewed (Figure 1A), which is indicative of disproportionately high emitting sites referred to as "super-emitters" (see the identification of super-emitters section for details). Our findings are consistent with those observed for oil and natural-gas supply chains.[25–30] Using global biogas and biomethane production of 35 megatonnes of oil equivalent (Mtoe) ($1.47 \times 10^{12}$ MJ) in 2018,[6] our model-based estimate of 2018 biomethane supply chain emissions may account for up to 18.5 teragram (Tg) $CH_4$ per year (6.4–7.8 Tg $CH_4$ year$^{-1}$ at the 95th percentile and an average of 2.8–2.9 Tg $CH_4$ year$^{-1}$), which is more than two times greater than the International Energy Agency's (IEA's) estimate of $CH_4$ emissions from bioenergy (9.1 Tg in 2021).[2] Our estimate of global biogas and biomethane $CH_4$ emissions is significantly lower than in the global oil and natural-gas supply chain (82.5 Tg in 2021);[2] on the other hand, it is comparable to the production segment of the US oil and natural-gas supply chain (6.1–7.1 Tg year$^{-1}$)[31] based on site measurements.

The cumulative distribution of emissions as a percentage of total $CH_4$ produced is shown in Figure 1B. The 5th percentile is 1.7%–2.0% (1.94%–2.0% at the 95% CI) of $CH_4$ production, and the 95th percentile is 12.3%–13.4% (12.6%–12.8% at the 95% CI) of total gas production. The ranges in minimum, median, mean, and maximum values were fairly consistent across all estimates (Figure 1). While the low and median estimates are nearly identical, the disparity between biomethane and natural gas varies widely in the highest estimates. The median ranged from 5.1% to 5.3% (5.1%–5.2% at the 95% CI), with mean emission rates of 5.90%–6.04% (5.9%–6.0% at the 95% CI) of total $CH_4$ production, which is higher than natural gas (0.8%–2.2% of $CH_4$ production).[25,26] Rutherford et al.[31] found $CH_4$ emissions in the oil and natural-gas-production segment to be 1.3% (1.2%–1.4% at the 95% CI), which is significantly lower than our findings. On the other hand, despite declining gas production, one of the highest reported $CH_4$ emissions from oil and gas production (Uinta Basin from a multi-year record of in-site observations) reveals a higher emission rate than our results (6%–8%).[32] Although emissions from the biomethane supply chain are comparable to oil and natural-gas production in terms of Tg $CH_4$ year$^{-1}$, the production-normalized emission rate is considerably higher. This could be due to a variety of factors, including poorly managed production facilities; a lack of attention to the biomethane industry resulting in lower investments for modernization, operation, and monitoring; and employment of highly skilled plant operators[16,21] when compared with oil and natural gas. In addition, poor design and management of feedstock and digestate storage units[33] as well as a limited interest in infrastructure emissions may result in higher emission rates compared with the amount of gas produced. Because oil and natural-gas supply chains have been primarily operated by large companies for decades, they have invested more in leak detection and repair.[34,35] On the other hand, given the growth in biomethane generation due to national decarbonization strategies, more urgent efforts are also needed for the biomethane supply chain to address not only $CH_4$ emissions but also the sustainability of biomethane.

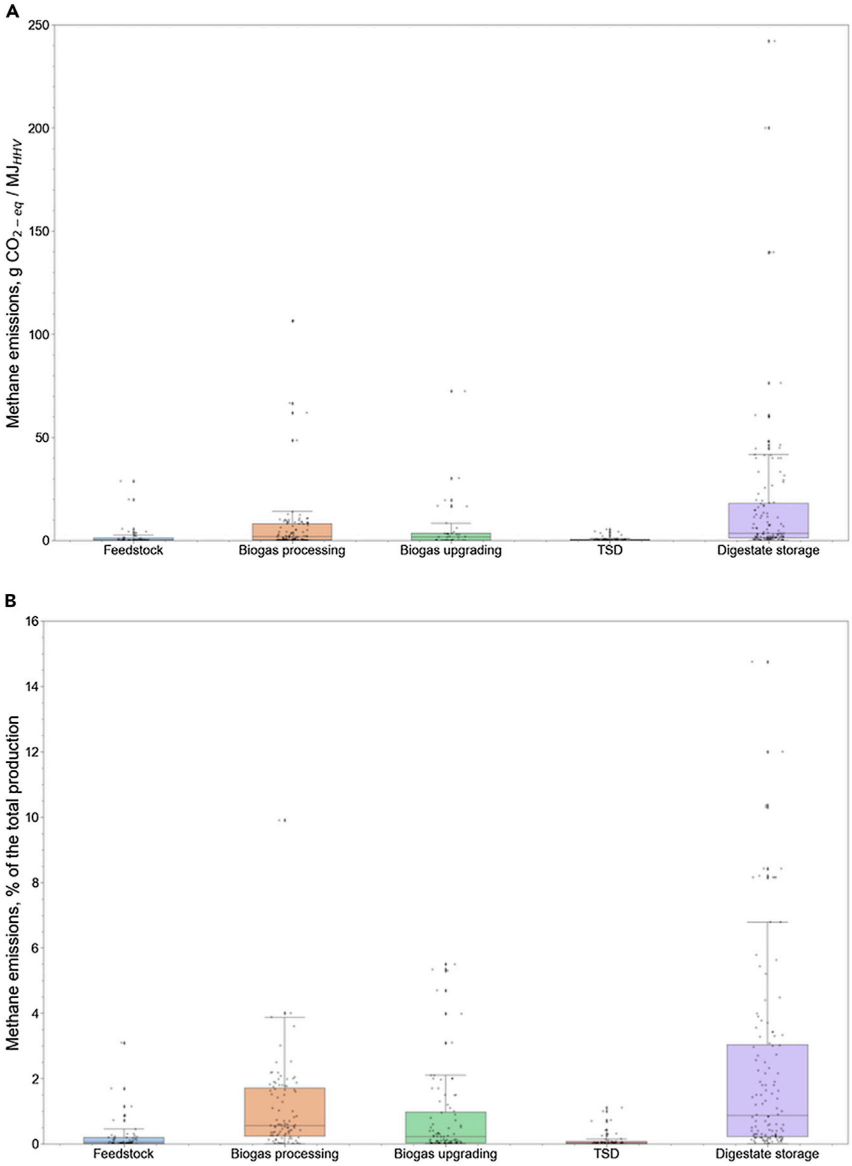


**Figure 2. Literature $CH_4$ emissions from different stages of biomethane supply chain**

(A) Emissions range of feedstock (n = 49), biogas processing (n = 100), biogas upgrading (n = 35), TSD (n = 44), and digestate storage (n = 119) stages in g $CO_{2\text{-eq.}}/MJ_{HHV}$.

(B) Emissions range of feedstock (n = 52), biogas processing (n = 95), biogas upgrading (n = 84), TSD (n = 48), and digestate storage (n = 120) stages in terms of total $CH_4$ emissions as a percentage of the total gas production rate.

Individual estimates are shown as circles in the same color at each stage of the supply chain, with median and 25th and 75th percentile boxes. Sample sizes for each stage are demonstrated in the Figure 3.

## Identification of super-emitters

A small proportion of facilities or equipment with disproportionately large emission rates are labeled super-emitters,[36,37] causing the heavy-tailed distribution (see Figure S4). A small number of high emitters may cause under- or overestimations of emissions rates[38] if they have intermittent emissions patterns, insufficient process equipment usage, or inadequate operations and maintenance strategies. In this study, super-emitters have been investigated at various stages across the supply chain, including feeding systems; substrate storage; runoff ponds; pressure relief valves on the anaerobic digesters and gas holders; exhausts and aeration lines of upgrading units; ventilation of units, such as compressors or closed digestate tanks; open digestate storage; and flaring. Within the heavy-tail distribution (Figures 1A and 1B) and the boxplot of each stage's emissions (Figure 2), the mean emission rate is higher than the median because of super-emitters (see Table S1 for details). Since we lack information on on-site $CH_4$ sources, we identify them as the top 5% of highest emissions based on the cumulative density function of $CH_4$ emissions, parallel to natural-gas production sites.[29,36,39] The highest 5% of total emissions (199–224.8 g $CO_{2\text{-eq.}}/MJ_{HHV}$) account for 62% (CI: 58%–66%) of cumulative emissions, with a threshold of 211.9 g $CO_{2\text{-eq.}}/MJ_{HHV}$. The characteristics of super-emitters in the biomethane supply chain are similar to those of super-emitters in the oil and natural-gas supply chain (the largest 5% of leaks contribute to 50%–60% of total emissions).[29,36] Since super-emitters are unlikely to remain constant over time, continual monitoring will be required to detect intermittent emission patterns or unpredictable leaks from the biomethane supply chain. Future work is necessary to understand the characteristics of individual super-emitter sites in the biomethane supply chain. The efficiency of mitigation efforts could be improved by investing in the underlying cause of preventable operational conditions at a component level.[30]

## Contribution of each supply chain stage

The contribution of each stage of the biomethane supply chain is illustrated in Figure 2A in g $CO_{2\text{-eq.}}/MJ_{HHV}$ and as a percentage of total production in Figure 2B. The distributions are almost identical. Emissions are mainly from digestate storage, followed by production and upgrading stages. Similar results were observed by Reinelt et al.,[17] where the highest emissions are from open digestate storage and pressure-release valves. Similarly, Alvarez et al.[39] found production and gathering units to be the main emission source in the US oil and natural-gas supply chain. Overall, the lowest emissions are exhibited in the transmission, storage, and distribution (TSD) stage, similar to the US oil and natural-gas supply chain.[39]

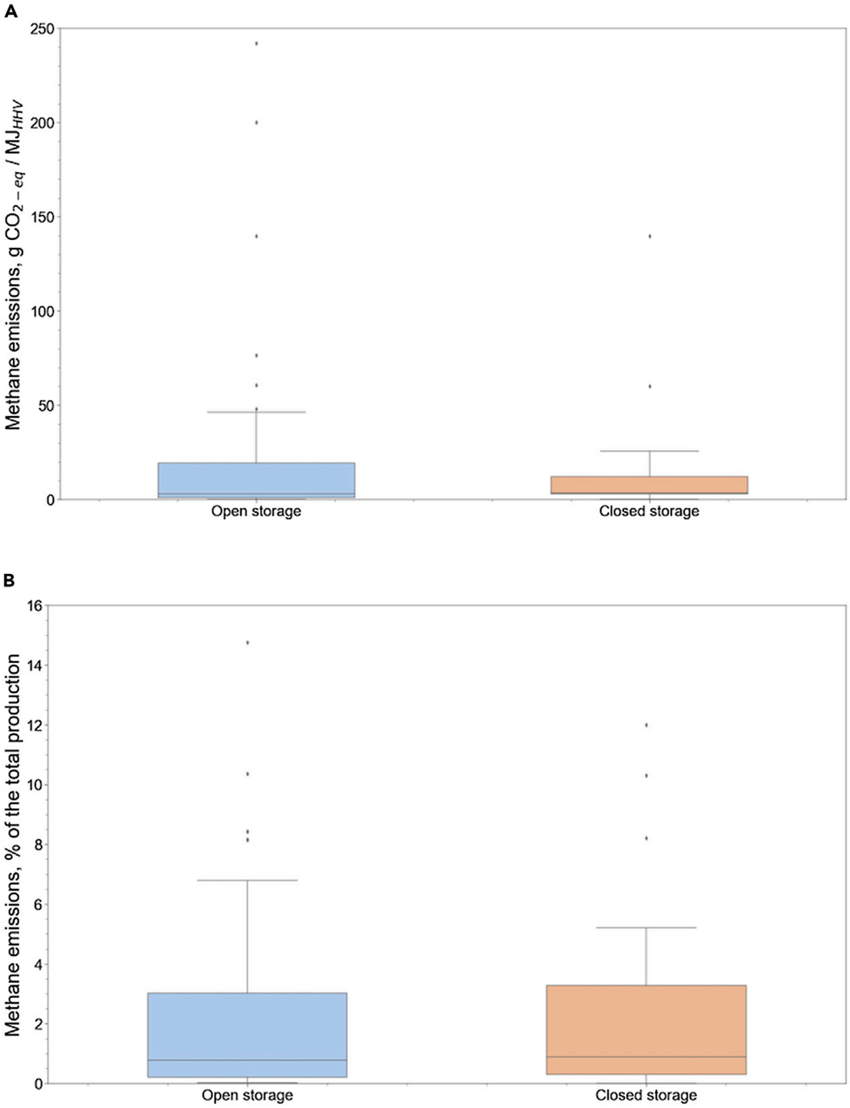


**Figure 3. Literature $CH_4$ emissions of open and closed digestate storage**

(A) Emissions from digestate handling from open (n = 98) and closed (n = 21) storage tanks in g $CO_{2\text{-}eq.}/MJ_{HHV}$.

(B) In terms of the percentage of total biomethane produced, illustrating emissions from the open (n = 95) and closed (n = 25) storage tanks.

Closed tank emissions originate mainly from leaks of covered material and ventilation of stockpile building.

Of particular note here is that the digestate storage stage is a significant source of $CH_4$, ranging between 0.05 and 242.1 g $CO_{2\text{-}eq.}/MJ_{HHV}$ (Figure 2A) or 0.005% and 14.8% of the total biomethane produced (Figure 2B). Sources of emissions are open or covered digestate (liquid and solid) storage tanks and lagoons. The emissions from digestate handling, such as post-composting processes, application of digestate, thickening exhaust, dewatering units, and leaks from centrifuges, were excluded from the biomethane supply chain in this study. Our analysis revealed that $CH_4$ emissions from this stage are 23% higher than previously reported,[40] while they still form a substantial portion of previous studies.[19,40]

The biogas production stage is the second biggest emission source, ranging from 0.002 to 106.5 g $CO_{2\text{-}eq.}/MJ_{HHV}$ (Figure 2A) or 0.001% to 9.9% of $CH_4$ production (Figure 2B). Biogas-production emissions are mainly from the anaerobic digester and hygienization tank. Hygienization tanks represent a relatively small fraction of the emissions from this stage. Emissions from the anaerobic digester are highly variable, depending on fugitive emissions from different cover types and control of venting from pressure-release valves. In addition, Zeng et al.[41] found that the fermentation temperature and quality of the feeding material have an effect on the $CH_4$ emissions from anaerobic digesters. Following biogas production, estimates of emissions from biogas upgrading are 0.002–72.4 g $CO_{2\text{-}eq.}/MJ_{HHV}$ (Figure 2A) or 0.001% to 5.5% of $CH_4$ production (Figure 2B), which are slightly higher than what was reported by Dumont et al.[40] The emissions from the biomethane production stage arise from the exhaust or aeration of units, ventilation ducts, booster pumps, safely valves on upgrading facilities, water or chemical scrubbers, and membranes.

In addition, feedstock emissions, resulting from fugitives and vents from substrate storage, are the fourth highest contributor to the supply chain, accounting for 0.0003 to 28.8 g $CO_{2\text{-}eq.}/MJ_{HHV}$ (Figure 2A) or 0.0003% to 3.1% of the total $CH_4$ production (Figure 2B), and represent the smallest proportion of total supply chain emissions. Higher emissions are associated with substrate storage. Dumont et al.[40] reported a larger range in $CH_4$ emission (0.2%–0.5% of $CH_4$ production) for feedstock storage.[40] However, their results were based on a smaller dataset than ours, and emissions may have reduced through technology improvements since their study was published.

## Digestate-handling approaches

As discussed above, digestate storage is the largest emission source in the biomethane supply chain. This is because of the accumulation of organic material, which leads to $CH_4$ production from fermentation. According to Döhler et al.,[42] digestate storage may account for nearly 27% of global $CO_{2eq.}$ emissions from anaerobic digestion processes. How digestate is handled has a major impact on emissions, with open digestate storage tanks and lagoons emitting more than closed tanks similar to the results from Paolini et al.[33] (Figure 3). The residual gas potential, digestate temperature, substrate amount, level of filling, and meteorological conditions all have a significant influence on the emission rate from open digestate storage tanks

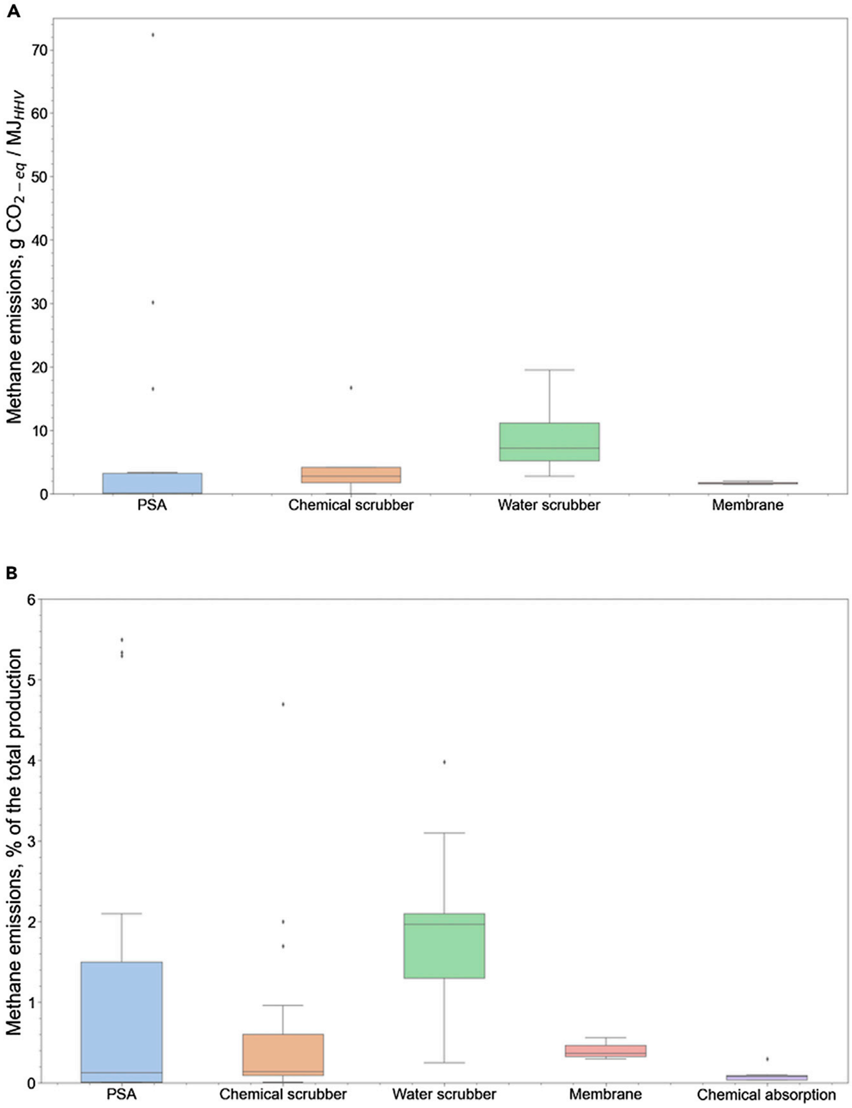


**Figure 4. Literature $CH_4$ emissions from different biogas-upgrading technologies**

(A) Emissions from biogas-upgrading technologies in g $CO_{2\text{-eq.}}$/$MJ_{HHV}$, pressure swing adsorption (PSA) with activated carbon filters (n = 22), water scrubber (n = 4), chemical scrubber (n = 5), and membrane (n = 4).

(B) Emissions from upgrading biogas technologies in terms of the percentage of total biomethane produced, with PSA with activated carbon filters (n = 30), water scrubber (n = 9), chemical scrubber (n = 25), chemical absorption (n = 14), and membrane technologies (n = 6).

Scrubber emissions include chemical (e.g., amine) and water scrubber emissions.

and lagoons.[15,19,43,44] Figure 3 clearly demonstrates that the closed tanks can still emit $CH_4$, although emissions from closed tanks can be avoided with improved covering materials, effective design, and regular maintenance. The facilities should consider becoming accredited under the Publicly Available Specification (PAS) 110 standards,[45] which recommend coverage of digestate to diminish emissions. Therefore, we recommend using closed digestate storage with vapor-recovery systems directed to the upgrading unit where economically viable to address emissions from this stage. Targeting reductions in digestate-handling emissions provides the greatest environmental improvements, though it is noted that detection and mitigation strategies would require additional expense and regulation.

## Impact of biogas-upgrading technologies

Upgrading biogas to biomethane can cause significant emissions. The literature provides scant data on specific upgrading technologies, though available evidence shows that membrane filters and chemical absorption leads to lower emissions than pressure swing adsorption (PSA) and water and chemical scrubbers (see Figures 4A and 4B). Therefore, initial indications are that chemical absorption technology is the best available technology for upgrading to reduce $CH_4$ emissions, which is the line with previously reported values.[46,47] PSA and water scrubber utilization should be avoided, though more measurements should be conducted.

## Total supply chain emission estimates versus whole-site mobile measurements

Alongside the on-site (aggregation of component-based emission) Monte Carlo approach described above, whole-site (off-site) measurements are a useful benchmark. In the literature, $CH_4$ emissions from 792 whole-site measurements varied between 0.1 and 483 g $CO_{2\text{-eq.}}$/$MJ_{HHV}$, with an average of 51.7 and a median of 24.6 g $CO_{2\text{-eq.}}$/$MJ_{HHV}$ (see Figure 5A). This is a larger range with lower median and higher estimate of upper limit than our Monte Carlo simulation of on-site measurements. While the mean of the Monte Carlo runs (51.4–52.7 g $CO_{2\text{-eq.}}$/$MJ_{HHV}$) and whole-site measurements (51.7 g $CO_{2\text{-eq.}}$/$MJ_{HHV}$) are comparable, the median of the Monte Carlo runs (40.0–42.3 g $CO_{2\text{-eq.}}$/$MJ_{HHV}$) are greater than the whole-site measurements (24.6 g $CO_{2\text{-eq.}}$/$MJ_{HHV}$) due to the heavy-tailed distribution of the Monte Carlo runs. Before running the Monte Carlo simulation, the emissions probability density function of each stage is identified to establish a good fit. The heavy-tailed distribution is due to the presence of super-emitters in each supply chain stage in the Monte Carlo runs, while whole-site emissions data did not exhibit this heavy tail.[48] However, super-emitters are certainly observed in the whole-site measurements and the maximum emission is greater than in the Monte Carlo runs, but these are insufficient in quantity and magnitude to raise the median above the mean. This discrepancy between the distribution of whole-site measurements and that observed in the Monte Carlo approach (i.e., from aggregation of measurements from each stage) is

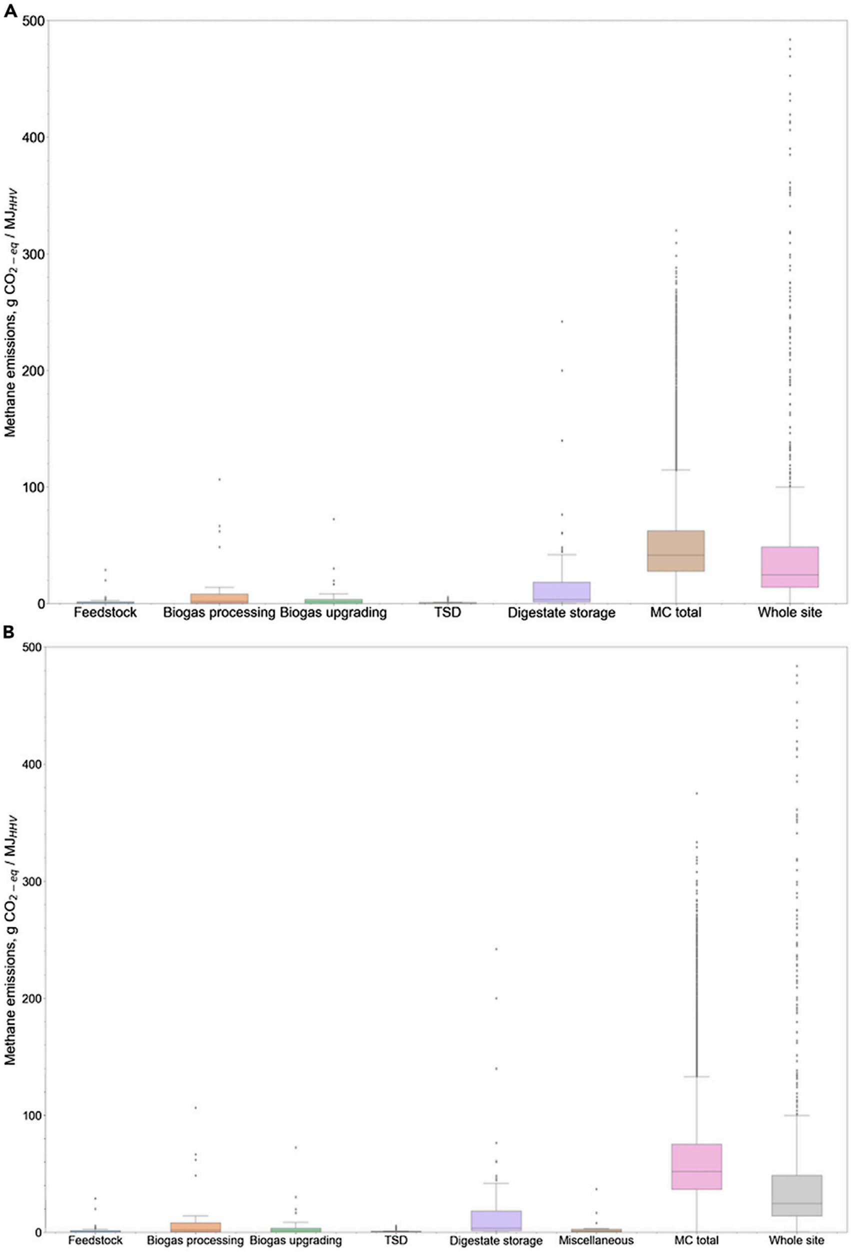


**Figure 5. Each stage of emissions with the whole-site measurements and MC runs**

(A) Literature emissions range for feedstock (n = 49), biogas processing (n = 100), biogas upgrading (n = 35), TSD (n = 44), and digestate storage (n = 119) stages with whole-site measurements (n = 792) and 10,000 Monte Carlo runs (MC total) with respect to g $CO_{2\text{-eq.}}/MJ_{HHV}$.

(B) Miscellaneous sources (n = 19) were added as an additional stage emission and MC total was reassessed to compare with whole-site emissions (off-site measurements).

Individual estimates are shown as circles in the same color at each stage of the supply chain, with median and 25th and 75th percentile boxes. Sample size for each stage and whole-site emissions are depicted. The median of the MC run is substantially higher than the medians of the five stages added together because (1) the medians of the derived KDE functions for each supply chain stage are substantially higher than those of the raw data and, (2) in any case, it is not expected that the sum of medians from each supply chain stage necessarily approximates the median of the sum of stages (see the experimental procedures for details).

worthy of further research in the future. A number of factors are known to affect measurements, including meteorological conditions during the survey, duration of measurements, uncertainties in emissions rate calculation models, the presence of super-emitters, and process conditions of facilities.[18] It is also likely to be influenced by miscellaneous sources (see Figure 5B),[19] such as biofilters used for odor reduction, stored solids causing fermentation, emissions from service opening,[19] or leakages located on top of units,[49] which are not quantified by the on-site measurement studies. We combined the miscellaneous sources reported in a few studies[19,23,50,51] and estimated their impacts on total emissions using a Monte Carlo simulation (see Figure S2) after identifying the associated KDE (see the experimental procedures), which impacts the data distribution.

Figure 5B demonstrated that miscellaneous sources could increase total supply chain emissions by 22%. However, it is noted that, because only four studies reported emissions from biofilters and solid separators, more detailed monitoring surveys are required to better understand the impact of miscellaneous emission sources on supply chain emissions. The high leakage rate reported in off-site measurements could also be due to process disturbances or extensive venting and flaring caused by insufficient infrastructure, which results in an intermittent and highly unpredictable emissions pattern that can overestimate or underestimate significant $CH_4$ sources.[38] The divergence between on-site and off-site measurements is most likely due to abnormal operating conditions resulting in high $CH_4$ emissions primarily from the production segment,[31] which is consistent with that of oil and natural-gas supply chains.[39] We agree with Zavala-Araiza et al.[52] and Rutherford et al.[31] that increasing on-site, component-level emission data through continuous emission monitoring and effective characterization of emission sources can reduce the uncertainty and divergence between on-site and off-site measurements.

## DISCUSSION

Overall, this study showed that the broad features of the biomethane supply chain led to emission profiles similar to those of oil and natural gas, although digestate handling, biogas production, and upgrading are key differentiators. The synthesis of available data here showed that this leads to lower direct $CH_4$ emissions than the oil and natural-gas supply chain but much higher $CH_4$ loss rates than the oil and natural-gas supply chain. This conclusion is pertinent in the context of global efforts to mitigate $CH_4$ emissions, which to date largely focuses on natural-gas supply chains. It is also pertinent to broaden efforts to mitigate climate change, where $CH_4$ emissions are increasingly recognized as a key climate forcer. Given the strong potential role of biomethane in Paris Agreement compliant energy futures, best available technology must be applied to detect and reduce supply chain emissions, policy and regulation[53] must consider these emissions more systematically, and a better understanding of the counterfactual life cycle emissions for waste and by-product biomethane feedstocks must be developed. It should be noted that, even if feedstocks are not used to generate biomethane, they may still emit $CH_4$; in fact, some studies have suggested that treating manure for biomethane production could be a mitigation strategy.[54] We believe that this large amount of $CH_4$ emissions from the biomethane supply chain, on the other hand, can be avoided by taking appropriate emission identification, detection, measurement, and quantification measurements. It is critical to emphasize that, if biomethane is widely used in the future to achieve decarbonization goals, biomethane supply chain emissions should be avoided in order to achieve net zero goals.

Reflecting on these results with respect to the EU Renewable Directive (RED) 2009/28/EC,[55] it is clear that cutting emissions from digestate handling and gas engines could underpin more sustainable biomethane production. According to an EU report[56] on the sustainability of solid and gaseous biomass used for electricity, heating, and cooling, the GHG threshold for biomethane production is 34.8 g $CO_{2\text{-eq.}}/MJ_{HHV}$,[56] excluding digestate emissions. In contrast, $CH_4$ emissions from the biomethane supply chain are estimated in this study to range from 2.5 to 343 g $CO_{2\text{-eq.}}/MJ_{HHV}$ and 0.8 to 182 g $CO_{2\text{-eq.}}/MJ_{HHV}$ (18.3–19.5 g $CO_{2\text{-eq.}}/MJ_{HHV}$ for the median and 64–74 g $CO_{2\text{-eq.}}/MJ_{HHV}$ for the 95th percentile) when digestate emissions are excluded (see Figure S3). In view of $CO_2$ and $N_2O$ ($GWP_{100} = 273 \pm 130$)[10] emissions from biomethane production, total GHG emissions from the biomethane supply chain are likely to exceed this threshold limit unless urgent actions are taken. Given the different lifetimes and GWPs of $CO_2$, $CH_4$, and $N_2O$, future research can focus on integrating emissions across different timescales in order to further expand the impact of the biomethane supply chains on global warming and climate change. Clearly, under these operating conditions and in light of the wide diversity of biogas production pathways, biomethane production may lose its advantages as a clean-energy technology and may jeopardize Paris Agreement targets if used extensively. This study would serve as a guideline for the emission ranges associated with each stage while also recommending appropriate measures for each stage to cut emissions and make progress toward Paris Agreement goals. Therefore, emission-minimizing technologies and techniques and more specific regulations on emissions and leak detection and repair are essential to significantly reduce supply chain emissions. We are also aware of the counterfactual case for what level of $CH_4$ emissions would occur if the feedstock had not been converted into biomethane. Future studies should focus on counterfactual analyses to assess the GHG credits under various counterfactual scenarios.

Our biomethane supply chain emissions model presented in this study represents the most common technologies used in the industry, but it has some limitations regarding data availability and resolution. Firstly, input data were taken only from measurement surveys, and the sample size is not large enough to determine a probability distribution model for each supply chain. As such, the KDE function was used rather than goodness of fit since the data could not be fitted to certain distribution functions, due to the lack of data and heavy-tailed distribution. Furthermore, much of the literature data were excluded, owing to the use of default emission factors, especially in modeling studies. Secondly, some studies could not be included because they did not report biogas or biomethane production rates despite reporting $CH_4$ emissions.

The most detailed measurement surveys have been conducted in various regions of Europe and mainly at agricultural plants, so this study substantially reflects European agriculture. Further possible research directions associated with this work include adapting the emissions profiles of theoretical supply chain routes, for example, using life-cycle-assessment tools. Future research should target data collection from various biomethane supply chain routes in other countries to reduce uncertainty in the data and improve size and representativeness of the samples, which can help to identify the most sustainable biomethane production routes. This accumulated database can be used to improve equipment, processes design, and operations that would mitigate $CH_4$ emissions.

## EXPERIMENTAL PROCEDURES

### Resource availability

#### *Lead contact*

Further information and requests for resources should be directed to and will be fulfilled by the lead contact, Semra Bakkaloglu (s.bakkaloglu@imperial.ac.uk).

#### *Materials availability*

This study did not generate new unique materials.

#### *Data and code availability*

All original datasets used in this work were made available as part of the publications referenced and described in the text. The Python code used for the Kernel density estimation (KDE) function and Monte Carlo simulation with the data have been deposited at Zenodo Data: https://doi.org/10.5281/zenodo.6550794.

### Methodological approach

This study aims to estimate $CH_4$ emissions from the biomethane supply chain and characterize the emissions sources in the various stages of the supply chain (described in Figure S1). The screening criteria for mobile $CH_4$ emissions measurements required on-site or off-site direct measurements be reported by the study authors, rather than drawing on experimental, lab-scale, and theoretical studies.

The existing literature reports emission rates in different units, and some studies provide insufficient information to allow for unit conversion. In this study, $CH_4$ emission estimates were converted into percentage of total production (volume of $CH_4$ emitted/volume of produced gas) and grams of $CO_2$ equivalent per megajoule of energy based on higher heating value (HHV). These were chosen to allow for comparisons with the oil and natural-gas supply chain without inferring downstream services. The source emissions were divided by the total expected volume of biomethane that would be generated for energy utilization.

Assumptions were applied to convert published emission rates into metric units. It was assumed that the $GWP_{100}$ of $CH_4$ is 27.2,[10] with an HHV of

38.1 MJ/m$^3$ or 55.5 MJ/kg. The average volume percentages of $CH_4$ in biogas and biomethane were taken as 65% and 95%, respectively, unless otherwise stated in a study. We assumed the $CH_4$ content is 55% in biogas for manure feedstock. We neglect uncertainty of measurements and $GWP_{100}$ (±11), although measurement uncertainty exists whenever an emissions rate is quantified. Similar to Brandt et al.,[28] we evaluate all emissions at their reported levels and investigate the impact of emissions distribution. GHG emissions other than $CH_4$, such as $CO_2$, $N_2O$, and $NH_3$, were not included because they are out of the scope of this study. Moreover, $CH_4$ emissions based on $GWP_{20}$ (80.8) were provided in the Figure S4. The three stages of this study are:

1. Building the emissions inventory
2. Assessing the supply chain emissions model
3. Applying Monte Carlo simulations to produce total emissions ranges.

### Emissions inventory

Following systematic reviews of the existing evidence base, 51 papers reporting mobile $CH_4$ emissions measurements were examined, including academic papers as well as governmental and industry reports (see the supplemental experimental procedures). We utilized the data from mobile $CH_4$ measurements using on-site leak detection and ground-based remote sensing methods (off site). $CH_4$ emissions from landfill were excluded because there is a lack of data on the amount of biogas and biomethane generated from landfill-gas-collection system, which is mainly calculated using landfill-modeling tools, and $CH_4$ emissions depend on $CH_4$ oxidation rate as well as top soil cover material rather than infrastructure emissions.[57] In addition, we only considered the emissions from wastewater treatment plants with anaerobic digesters. The details of the chosen biomethane supply chain route employed in this study are described in detail in the Figure S1.

Emission inventory data availability is highly variable, owing to variations in the applied methodologies, differing plant design and operation, and insufficient data for each supply chain. Most emissions data were for Europe. Datasets for each emissions source were divided into subcategories, where there was discernible variation between feedstock materials (see Table S1). The stages in the biomethane supply chain are (1) collecting and storing organic materials (feedstock); (2) converting them to biogas under anaerobic conditions (AD); (3) upgrading biogas to biomethane (upgrading); (4) transportation, gas storage, and distribution of generated gas (TSD); and (5) digestate storage.

#### *Feedstock storage*

Any biodegradable material, such as agricultural residues, maize, crops, sewage sludges, or food and drink waste, utilized in anaerobic digestion is called feedstock. The yield of biogas from a specific feedstock can vary based on the dry-matter content, residence time in the digester, and feedstock purity.[58] Feedstock transported from a third party to the production facility is stored in the facility and pre-treated before being sent to the biogas-production stage. This stage covers four major components: runoff ponds, screw conveyor, mixing tank (homogenization tank), and substrate storage. Substrate storage tanks and biomass-receiving units, such as feedstock piles, runoff ponds, screw conveyors, and mixing (homogenization) tanks, are the main sources of emissions, although few studies have investigated emissions from this stage.[15,17,19,22,46,49,59–61] Feeding system emissions are included in substrate storage emissions (see Table S1). These emissions are mainly fugitives, predominantly from open storage tanks and feeding units.

#### *Biogas production*

The physically treated material is pasteurized and delivered to an anaerobic digester to generate biogas. The $CH_4$ concentration in the biogas depends on the type of digestate feedstock, type of anaerobic digester, and conditions in the digester, such as mesophilic and thermophilic. The biogas production stage consists of a buffer (hygienization) tank and a reactor (anaerobic digester). Previously reported $CH_4$ emissions from hygienization tanks, anaerobic digesters, and post-digesters are included in this stage (see Table S1).[15–17,19,41,49,50,59,62–76] These emissions are fugitives and venting.

#### *Biomethane production: Biogas upgrading process*

The biogas can be upgraded into biomethane by removing impurities. Depending on the biogas quality and the end use, different upgrading technologies can be used. Currently, water scrubbing is the most common commercial technology, followed by chemical scrubbers, membrane, PSA, organic physical scrubber, and cryogenic separation.[77] $CH_4$ emissions from various upgrading processes, such as carbon filters, chemical and water scrubbers, and membrane technologies, were reported in previous studies[17,19,46,47,49,50,71,78–81] and have been included in the upgrading processes stage (see Table S1). These are fugitives and vent from PSA exhausts and aeration and ventilation ducts.

#### *Transmission, storage, and distribution*

Biogas can be utilized to generate heat, electricity, or both. Biomethane can be injected into a gas grid or used as a renewable transport fuel in vehicles. Previous studies reported exhaust $CH_4$ from cogeneration, electricity production, heat utilization, combined heat and power units, gas engine slip, flare, and gas holder.[17,19,23,41,59,62,67,71,82–85] Emissions from pipeline, flare, compressors, and pressure-relief valves (PRVs) are considered in the transmission, storage, and distribution stage (see Table S1) in order to compare with natural-gas supply chain. End-use emissions mainly coming from incomplete combustion from combined heat and power (CHP), as well as fugitive leaks and vents from energy production units, were not included into emissions from this stage.

#### *Digestate storage*

Digestate can be used as is or can be further processed through different methods to be used as fertilizer. The PAS 110 for digestate quality specification is designed to ensure that digestate is no longer classified as waste and is safe and reliable to use as a fertilizer, soil improver, or conditioner, and it recommends that all types of digestate be covered.[45] Although some facilities follow the PAS 110 scheme, none of the published papers address whether the digestate complies with the standard. The solids-liquids separators, such as centrifuge and screw-press separators, membrane filters, biofilters, aerobic treatment, and composting, are widely used to process digestate.[86] The processing and storage of liquid and solid digestate can also cause emissions, depending on the temperature, wind, atmospheric pressure, plant process parameters, and storage tank filling level.[19] Emissions data by digestate types (e.g., solid or liquid) and storage properties (closed or open tanks) were collected from previous studies (see Table S1).[15–17,19,23,24,43,44,49,50,59,62,67,71–73,85,87–92] The emissions are mostly fugitives and venting from open PRVs. The emissions associated with digestate use, such as fertilizer application and post-composting, were excluded from the supply chain because digestate is not always used in the operation area and their emissions are only reported in a few studies.

#### *Miscellaneous emissions*

An additional emission stage has been added to account for a variety of sources that are not necessarily present in every biomethane supply chain. The $CH_4$ emissions from biofilters for odor reduction, compost filters, and separators have been reported in a few studies[19,23,50,51] and are included as miscellaneous emissions.

### Whole-site mobile measurements

Whole-site mobile measurements were included for comparisons to the modeled of total emissions from each supply chain.[17,19–23,46,49–51,61–65,71,73,75,88,93] Various emission-measurement techniques were used to quantify emissions and their sources in the whole supply chain, which caused a large variation in emissions.

### Supply chain emission models

It is important to determine the probability density function (PDF) of emissions in each stage before running the Monte Carlo simulation.[48] The PDF establishes a good fit of the emissions for each stage, including an uncertainty assessment. Because the emissions in each stage exhibit unique characteristics, particularly with respect to various super-emitters, their data and probability distributions differ (see Table S2 for the characteristics of PDFs). Due to the heavy-tailed distribution (Figure S3) and lack in the sample size, the nonparametric PDF ,which is the KDE, were generated to investigate the PDFs of the emissions from each stage using Python. KDE typically provides more accurate estimates of data distributions than parametric approaches.[94–96] The bandwidth of KDE for each stage was determined automatically in Python's SciPy library using Scott's Rule,[97] which is dependent on the number of data points.[97] The total sample size is 347 and 399 for g $CO_{2\text{-eq.}}/MJ_{HHV}$ and production normalized data, respectively.

### Monte Carlo simulation

The total supply chain emissions were estimated using Monte Carlo simulations,[98] which has been widely applied to estimate $CH_4$ emissions[25,30] with uncertainty assessment. Each supply chain's PDF, obtained from the KDE, was defined in the simulation to sample from each stage, followed by summing up each stage. Rather than separating the data by feedstock type, we divided it by stage of the supply chain, as shown in Figure S1. The total $CH_4$ emissions were assessed 10,000 times with random draws from the distributions for each stage. The 10,000 estimates were then used to assess the cumulative $CH_4$ distribution across the supply chain models by the Python code.

## SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j.oneear.2022.05.012.

## ACKNOWLEDGMENTS

The authors would like to acknowledge the funding from the Sustainable Gas Institute, founded by Imperial College London. We thank Thomas Flesch and Anders Michael Fredenslund for their cooperation. Graphical abstract is created with BioRender.com.

## AUTHOR CONTRIBUTIONS

Conceptualization, S.B. and A.H.; methodology, S.B., J.C., and A.H.; software, S.B.; validation, S.B. and J.C.; formal analysis, S.B. and J.C.; investigation, S.B., J.C., and A.H.; writing – original draft, S.B.; writing – review & editing, J.C. and A.H.; funding acquisition, A.H.; resources, S.B. and A.H.; supervision, A.H.

## DECLARATION OF INTERESTS

The authors declare no competing interests.

Received: February 7, 2022
Revised: April 17, 2022
Accepted: May 23, 2022
Published: June 17, 2022

## REFERENCES

1. The Royal Society (2021). Low-carbon Heating and Cooling: Overcoming One of World's Most Important Net Zero Challenges. https://royalsociety.org/-/media/policy/projects/climate-change-science-solutions/climate-science-solutions-heating-cooling.pdf.
2. IEA (2022). Global Methane Tracker 2022 (Paris: IEA). https://www.iea.org/reports/global-methane-tracker-2022.
3. Speirs, J., Dubey, L., Balcombe, P., Tariq, N., Brandon, N., and Hawkes, A. (2021). The Best Uses of Natural Gas within Paris Climate Targets (Sustainable Gas Institute, Imperial College London).
4. Nisbet, E.G., Fisher, R.E., Lowry, D., France, J.L., Allen, G., Bakkaloglu, S., Broderick, T.J., Cain, M., Coleman, M., Fernandez, J., et al. (2020). Methane mitigation: methods to reduce emissions, on the path to the Paris Agreement. Rev. Geophys. *58*. e2019RG000675. https://doi.org/10.1029/2019rg000675.
5. Huppmann, D. (2018). IAMC 1.5° C Scenario Explorer and Data Hosted by IIASA (Integrated Assessment Modeling Consortium & International Institute for Applied Systems Analysis). https://doi.org/10.22022/SR15/08-2018.15429.
6. IEA (2020). Outlook for Biogas and Biomethane: Prospects for Organic Growth (Paris: IEA).
7. Marc-Antoine, E.M., and Mathieu, C. (2019). Biogas and Biomethane in Europe: Lessons from Denmark, Germany and Italy (Études de l'Ifri). https://www.ifri.org/en/publications/etudes-de-lifri/biogas-and-biomethane-europe-lessons-denmark-germany-and-italy.
8. The World Biogas Association. (2019). Global Potential of Biogas (World Biogas Association). https://www.worldbiogasassociation.org/wp-content/uploads/2019/07/WBA-globalreport-56ppa4_digital.pdf.
9. ADBA (2019). Anaerobic Digestion Policy Report (Anaerobic Digestion & Bioresources Association).
10. IPCC (2021). AR6 Climate Change 2021: The Physical Science Basis (Cambridge University Press).
11. Paris Agreement (2015). United Nations Framework Convention on Climate Change (Paris Agreement).
12. Rogelj, J., Meinshausen, M., Schaeffer, M., Knutti, R., and Riahi, K. (2015). Impact of short-lived non-CO2 mitigation on carbon budgets for stabilizing global warming. Environ. Res. Lett. *10*, 075001. https://doi.org/10.1088/1748-9326/10/7/075001.
13. IPCC (2022). Climate Change 2022: Mitigation of Climate Change. Contribution of Working Group III to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change (Cambridge University Press). https://report.ipcc.ch/ar6wg3/pdf/IPCC_AR6_WGIII_FinalDraft_FullReport.pdf.
14. Edward, B., Volker, K., Elmar, K., Keywan, R., Roberto, S., Jarmo, K., Robin, L., Zebedee, N., Marit, S., Chris, S., et al. (2022). AR6 Scenarios Database Hosted by IIASA (International Institute for Applied Systems Analysis).
15. Reinelt, T., McCabe, B.K., Hill, A., Harris, P., Baillie, C., and Liebetrau, J. (2022). Field measurements of fugitive methane emissions from three Australian waste management and biogas facilities. Waste Manag. *137*, 294–303. https://doi.org/10.1016/j.wasman.2021.11.012.
16. Reinelt, T., and Liebetrau, J. (2020). Monitoring and mitigation of methane emissions from pressure relief valves of a biogas plant. Chem. Eng. Technol. *43*, 7–18. https://doi.org/10.1002/ceat.201900180.
17. Reinelt, T., Delre, A., Westerkamp, T., Holmgren, M.A., Liebetrau, J., and Scheutz, C. (2017). Comparative use of different emission measurement approaches to determine methane emissions from a biogas plant. Waste management *68*, 173–185. https://doi.org/10.1016/j.wasman.2017.05.053.
18. Liebetrau, J., Reinelt, T., Agostini, A., and Linke, B. (2017). Methane Emissions from Biogas Plants (IEA bioenergy).
19. Liebetrau, J., Reinelt, T., Clemens, J., Hafermann, C., Friehe, J., and Weiland, P. (2013). Analysis of greenhouse gas emissions from 10 biogas plants within the agricultural sector. Water Sci. Technol. *67*, 1370–1379. https://doi.org/10.2166/wst.2013.005.
20. Bakkaloglu, S., Lowry, D., Fisher, R.E., France, J.L., Brunner, D., Chen, H., and Nisbet, E.G. (2021). Quantification of methane emissions from UK biogas plants. Waste Manag. *124*, 82–93. https://doi.org/10.1016/j.wasman.2021.01.011.
21. Scheutz, C., and Fredenslund, A.M. (2019). Total methane emission rates and losses from 23 biogas plants. Waste Manag. *97*, 38–46. https://doi.org/10.1016/j.wasman.2019.07.029.
22. Flesch, T.K., Desjardins, R.L., and Worth, D. (2011). Fugitive methane emissions from an agricultural biodigester. Biomass Bioenergy *35*, 3927–3935. https://doi.org/10.1016/j.biombioe.2011.06.009.
23. Daniel-Gromke, J., Liebetrau, J., Denysenko, V., W S, K., and J, H. (2015). The humelock hemiarthoplasty device for both primary and failed management of proximal humerus fractures: a case series. Open Orthop. J. *9*, 1–6. https://doi.org/10.2174/1874325001509010001.
24. Balde, H., VanderZaag, A.C., Burtt, S.D., Wagner-Riddle, C., Crolla, A., Desjardins, R.L., and MacDonald, D.J. (2016). Methane emissions from digestate at an agricultural biogas plant. Bioresour. Technol. *216*, 914–922. https://doi.org/10.1016/j.biortech.2016.06.031.
25. Balcombe, P., Brandon, N., and Hawkes, A. (2018). Characterising the distribution of methane and carbon dioxide emissions from the natural gas supply chain. J. Clean. Prod. *172*, 2019–2032. https://doi.org/10.1016/j.jclepro.2017.11.223.
26. Balcombe, P., Anderson, K., Speirs, J., Brandon, N., and Hawkes, A. (2017). The natural gas supply chain: the importance of methane and carbon dioxide emissions. ACS Sustain. Chem. Eng. *5*, 3–20. https://doi.org/10.1021/acssuschemeng.6b00144.

27. Balcombe, P., Anderson, K., Speirs, J., Brandon, N., and Hawkes, A. (2015). Methane and CO 2 Emissions from the Natural Gas Supply Chain (Sustainable Gas Institute).
28. Brandt, A.R., Heath, G.A., and Cooley, D. (2016). Methane leaks from natural gas systems follow extreme distributions. Environmental science & technology *50*, 12512–12520. https://doi.org/10.1021/acs.est.6b04303.
29. Omara, M., Zimmerman, N., Sullivan, M.R., Li, X., Ellis, A., Cesa, R., Subramanian, R., Presto, A.A., and Robinson, A.L. (2018). Methane emissions from natural gas production sites in the United States: data synthesis and national estimate. Environmental science & technology *52*, 12915–12925. https://doi.org/10.1021/acs.est.8b03535.
30. Zavala-Araiza, D., Alvarez, R.A., Lyon, D.R., Allen, D.T., Marchese, A.J., Zimmerle, D.J., and Hamburg, S.P. (2017). Super-emitters in natural gas infrastructure are caused by abnormal process conditions. Nat. Commun. *8*, 14012. https://doi.org/10.1038/ncomms14012.
31. Rutherford, J.S., Sherwin, E.D., Ravikumar, A.P., Heath, G.A., Englander, J., Cooley, D., Lyon, D., Omara, M., Langfitt, Q., and Brandt, A.R. (2021). Closing the methane gap in US oil and natural gas production emissions inventories. Nat. Commun. *12*, 4715. https://doi.org/10.1038/s41467-021-25017-4.
32. Lin, J.C., Bares, R., Fasoli, B., Garcia, M., Crosman, E., and Lyman, S. (2021). Declining methane emissions and steady, high leakage rates observed over multiple years in a western US oil/gas production basin. Sci. Rep. *11*, 22291. https://doi.org/10.1038/s41598-021-01721-5.
33. Paolini, V., Petracchini, F., Segreto, M., Tomassetti, L., Naja, N., and Cecinato, A. (2018). Environmental impact of biogas: a short review of current knowledge. J. Environ. Sci. Health A Tox Hazard Subst. Environ. Eng. *53*, 899–906. https://doi.org/10.1080/10934529.2018.1459076.
34. EDF (2019). Methane Mitigation in the Oil & Gas Industry. https://business.edf.org/insights/methane-mitigation-in-the-oil-gas-industry/.
35. IEA (2021). Driving Down Methane Leaks from the Oil and Gas Industry (Paris: IEA).
36. Zavala-Araiza, D., Lyon, D., Alvarez, R.A., Palacios, V., Harriss, R., Lan, X., Talbot, R., and Hamburg, S.P. (2015). Toward a functional definition of methane super-emitters: application to natural gas production sites. Environ. Sci. Technol. *49*, 8167–8174. https://doi.org/10.1021/acs.est.5b00133.
37. Brandt, A.R., Heath, G.A., Kort, E.A., O'Sullivan, F., Pétron, G., Jordaan, S.M., Tans, P., Wilcox, J., Gopstein, A.M., Arent, D., et al. (2014). Methane leaks from North American natural gas systems. Science *343*, 733–735. https://doi.org/10.1126/science.1247045.
38. Duren, R.M., Thorpe, A.K., Foster, K.T., Rafiq, T., Hopkins, F.M., Yadav, V., Bue, B.D., Thompson, D.R., Conley, S., Colombi, N.K., et al. (2019). California's methane super-emitters. Nature *575*, 180–184. https://doi.org/10.1038/s41586-019-1720-3.
39. Alvarez, R.A., Zavala-Araiza, D., Lyon, D.R., Allen, D.T., Barkley, Z.R., Brandt, A.R., Davis, K.J., Herndon, S.C., Jacob, D.J., Karion, A., et al. (2018). Assessment of methane emissions from the US oil and gas supply chain. Science *361*, 186–188. https://doi.org/10.1126/science.aar7204.
40. Mathieu Dumont, N., Luning, L., Yildiz, I., and Koop, K. (2013). Methane emissions in biogas production. In The biogas handbook (Elsevier), pp. 248–266. https://doi.org/10.1533/9780857097415.2.248.
41. Zeng, J., Xu, R., Sun, R., Niu, L., Liu, Y., Zhou, Y., Zeng, W., and Yue, Z. (2020). Evaluation of methane emission flux from a typical biogas fermentation ecosystem in China. J. Clean. Prod. *257*, 120441. https://doi.org/10.1016/j.jclepro.2020.120441.
42. Döhler, H., Niebaum, A., Roth, U., Amon, T., Balsari, P., and Friedl, G. (2009). Greenhouse Gas Emissions and Mitigati-On Costs in Two European Biogas Plants (Gülzower Fachgespräche).
43. Gioelli, F., Dinuccio, E., and Balsari, P. (2011). Residual biogas potential from the storage tanks of non-separated digestate and digested liquid fraction. Bioresour. Technol. *102*, 10248–10251. https://doi.org/10.1016/j.biortech.2011.08.076.
44. Vergote, T.L., Bodé, S., De Dobbelaere, A.E., Buysse, J., Meers, E., and Volcke, E.I. (2020). Monitoring methane and nitrous oxide emissions from digestate storage following manure mono-digestion. Biosyst. Eng. *196*, 159–171. https://doi.org/10.1016/j.biosystemseng.2020.05.011.
45. WRAP. (2014). PAS 110:2014. Specification for Whole Digestate, Separated Liquor and Separated Fibre Derived from the Anaerobic Digestion of Source-Segregated Biodegradable Materials (The BSI). https://wrap.org.uk/sites/default/files/2021-03/PAS110_2014.pdf.
46. Ricardo Energy and Environment. (2017). Methodology to Assess Methane Leakage from AD Plants. Part I: Report on Proposed Categorisation of AD Plants and Literature Review of Methane Monitoring Technologies (Ricardo Energy and Environment). https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/786756/Methodology_to_Assess_Methane_Leakage_from_AD_Plants_final_report_part1.pdf.
47. Paolini, V., Torre, M., Giacopini, W., Pastori, M., Segreto, M., Tomassetti, L., Carnevale, M., Gallucci, F., Petracchini, F., and Guerriero, E. (2019). CO2/CH4 separation by hot potassium carbonate absorption for biogas upgrading. Int. J. Greenh. Gas Control *83*, 186–194. https://doi.org/10.1016/j.ijggc.2019.02.011.
48. McMurray, A., Pearson, T., and Casarim, F. (2017). Guidance on Applying the Monte Carlo Approach to Uncertainty Analyses in Forestry and Greenhouse Gas Accounting (Winrock International).
49. Fredenslund, A.M., Hinge, J., Holmgren, M.A., Rasmussen, S.G., and Scheutz, C. (2018). On-site and ground-based remote sensing measurements of methane emissions from four biogas plants: a comparison study. Bioresour. Technol. *270*, 88–95. https://doi.org/10.1016/j.biortech.2018.08.080.
50. Holmgren, M.A., Hansen, M.N., Reinelt, T., Westerkamp, T., Jørgensen, L., Scheutz, C., and Delre, A. (2015). Measurements of Methane Emissions from Biogas Production–Data Collection and Comparison of Measurement Methods: Energiforsk report 2015: 158 (Energiforsk AB).
51. Jensen, M.B., Møller, J., Mønster, J., and Scheutz, C. (2017). Quantification of greenhouse gas emissions from a biological waste treatment facility. Waste Manag. *67*, 375–384. https://doi.org/10.1016/j.wasman.2017.05.033.
52. Zavala-Araiza, D., Lyon, D.R., Alvarez, R.A., Davis, K.J., Harriss, R., Herndon, S.C., Karion, A., Kort, E.A., Lamb, B.K., Lan, X., et al. (2015). Reconciling divergent estimates of oil and gas methane emissions. Proc. Natl. Acad. Sci. U S A *112*, 15597–15602. https://doi.org/10.1073/pnas.1522126112.
53. European Commission (2020). EU Strategy to Reduce Methane Emissions (European Commission).
54. Kupper, T., Häni, C., Neftel, A., Kincaid, C., Bühler, M., Amon, B., and VanderZaag, A. (2020). Ammonia and greenhouse gas emissions from slurry storage-A review. Agric. Ecosyst. Environ. *300*, 106963. https://doi.org/10.1016/j.agee.2020.106963.
55. European Union (2009). Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the Promotion of the Use of Energy from Renewable Sources and Amending and Subsequently Repealing Directives 2001/77/EC and 2003/30/EC (Official Journal of the European Union).
56. European Commission (2014). SWD(2014) 259 Final, State of Play on the Sustainability of Solid and Gaseous Biomass Used for Electricity, Heating and Cooling in the EU (European Commission).
57. Bakkaloglu, S., Lowry, D., Fisher, R.E., France, J.L., and Nisbet, E.G. (2021). Carbon isotopic characterisation and oxidation of UK landfill methane emissions by atmospheric measurements. Waste Manag. *132*, 162–175. https://doi.org/10.1016/j.wasman.2021.07.012.
58. Bioenergy, I. (2013). The Biogas Handbook (United Kingdom: Woodhead Publishing Limited).
59. Liebetrau, J., Clemens, J., Cuhls, C., Hafermann, C., Friehe, J., Weiland, P., and Daniel-Gromke, J. (2010). Methane Emissions from Biogas-producing Facilities within the Agricultural Sector*10* (Wiley Online Library), pp. 595–599. https://doi.org/10.1002/elsc.201000070.

60. Hrad, M., Piringer, M., Kamarad, L., Baumann-Stanzer, K., and Huber-Humer, M. (2014). Multisource emission retrieval within a biogas plant based on inverse dispersion calculations—a real-life example. Environ. Monit. Assess. *186*, 6251–6262. https://doi.org/10.1007/s10661-014-3852-0.

61. Hrad, M., Vesenmaier, A., Flandorfer, C., Piringer, M., Stenzel, S., and Huber-Humer, M. (2021). Comparison of forward and backward Lagrangian transport modelling to determine methane emissions from anaerobic digestion facilities. Atmos. Environ. X *12*, 100131. https://doi.org/10.1016/j.aeaoa.2021.100131.

62. Daelman, M.R., van Voorthuizen, E.M., van Dongen, U.G., Volcke, E.I., and van Loosdrecht, M.C. (2012). Methane emission during municipal wastewater treatment. Water Res. *46*, 3657–3670. https://doi.org/10.1016/j.watres.2012.04.024.

63. Delre, A., Mønster, J., and Scheutz, C. (2017). Greenhouse gas emission quantification from wastewater treatment plants, using a tracer gas dispersion method. Sci. Total Environ. *605-606*, 258–268. https://doi.org/10.1016/j.scitotenv.2017.06.177.

64. Delre, A., Mønster, J., and Scheutz, C. (2014). Quantification of Fugitive Methane Emissions from the Biogas Plant in Linköping (SE) (Technical University of Denmark, DTU Environment), p. 15.

65. Groth, A., Maurer, C., Reiser, M., and Kranert, M. (2015). Determination of methane emission rates on a biogas plant using data from laser absorption spectrometry. Bioresour. Technol. *178*, 359–361. https://doi.org/10.1016/j.biortech.2014.09.112.

66. Harper, L.A., Flesch, T.K., Weaver, K.H., and Wilson, J.D. (2010). The effect of biofuel production on swine farm methane and ammonia emissions. J. Environ. Qual. *39*, 1984–1992. https://doi.org/10.2134/jeq2010.0172.

67. Jonerholm, K., L.H. (2012). Sweco Environment AB Methane Losses in the Biogas System (Baltic Biogas Bus). https://docplayer.net/29996723-Methane-losses-in-the-biogas-system.html.

68. Paredes, M., Güereca, L.P., Molina, L., and Noyola, A. (2019). Methane emissions from anaerobic sludge digesters in Mexico: on-site determination vs. IPCC Tier 1 method. Sci. Total Environ. *656*, 468–474. https://doi.org/10.1016/j.scitotenv.2018.11.373.

69. Noyola, A., Paredes, M., Güereca, L., Molina, L., and Zavala, M. (2018). Methane correction factors for estimating emissions from aerobic wastewater treatment facilities based on field data in Mexico and on literature review. Sci. Total Environ. *639*, 84–91. https://doi.org/10.1016/j.scitotenv.2018.05.111.

70. Reinelt, T., Liebetrau, J., and Nelles, M. (2016). Analysis of operational methane emissions from pressure relief valves from biogas storages of biogas plants. Bioresour. Technol. *217*, 257–264. https://doi.org/10.1016/j.biortech.2016.02.073.

71. Ricardo Energy and Environment. (2017). Methodology to Assess Methane Leakage from AD Plants Part:2 Monitoring Methodology (Ricardo Energy and Environment). https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/786757/Methodology_to_Assess_Methane_Leakage_from_AD_Plants_final_report_part2.pdf.

72. Sax, M., Schick, M., Bolli, S., Soltermann-Pasca, A., and Van Caenegem, L. (2013). Methanverluste bei landwirtschaftlichen Biogasanlagen (Methane losses from agricultural biogas plants), (Bern: Bundesamt für Energie BFE).

73. Samuelsson, J., Delre, A., Tumlin, S., Hadi, S., Offerle, B., and Scheutz, C. (2018). Optical technologies applied alongside on-site and remote approaches for climate gas emission quantification at a wastewater treatment plant. Water Res. *131*, 299–309. https://doi.org/10.1016/j.watres.2017.12.018.

74. Tauber, J., Parravicini, V., Svardal, K., and Krampe, J. (2019). Quantifying methane emissions from anaerobic digesters. Water Sci. Technol. *80*, 1654–1661. https://doi.org/10.2166/wst.2019.415.

75. Yoshida, H., Mønster, J., and Scheutz, C. (2014). Plant-integrated measurement of greenhouse gas emissions from a municipal wastewater treatment plant. Water Res. *61*, 108–118. https://doi.org/10.1016/j.watres.2014.05.014.

76. Dhingra, R., Christensen, E.R., Liu, Y., Zhong, B., Wu, C.-F., Yost, M.G., and Remais, J.V. (2011). Greenhouse gas emission reductions from domestic anaerobic digesters linked with sustainable sanitation in rural China. Environmental science & technology *45*, 2345–2352. https://doi.org/10.1021/es103142y.

77. Angelidaki, I., Boe, K., and Ellegaard, L. (2005). Effect of operating conditions and reactor configuration on efficiency of full-scale biogas plants. Water Sci. Technol. *52*, 189–194. https://doi.org/10.2166/wst.2005.0516.

78. Wolf, D., and Scherello, A. (2013). Messung der Methanemission an der Biogasanlage Einbeck mittels CHARM®(Methane Emission Measurement at Biogas Plant Einbeck using CHARM®) (Gwf GasErdgas, Deutscher Industrieverlag GmbH), pp. 1–7.

79. Kvist, T., and Aryal, N. (2019). Methane loss from commercially operating biogas upgrading plants. Waste Manag. *87*, 295–300. https://doi.org/10.1016/j.wasman.2019.02.023.

80. Westerkamp, T., Reinelt, T., and Liebetrau, J. (2014). Scientific Measurements of Methane Emissions with Remote and On-Site Methods in Comparison (Presentation at the 2nd IBBA workshop in Kiel, Germany).

81. Holmgren, M.A., Hellström, H., Petersson, A., and Blom, A. (2012). The Swedish Voluntary Agreement for Control of Methane Emissions from Biogas Plants (SP Technical Research Institute of Sweden).

82. Aschmann, V., Kissel, R., and Gronauer, A. (2006). Exhaust emissions and performances of biogas-driven combined heat and power plants. Agrartechnische Forschung *12*, 46–52.

83. de Zwart, M., van Dijk, G., and Klimstra, J. (2012). Methane emissions from gas engines driving combined heat and power installations. J. Integr. Environ. Sci. *9*, 113–125. https://doi.org/10.1080/1943815x.2012.691885.

84. Woess-Gallasch, S., Bird, N., Enzinger, P., Jungmeier, G., Padinger, R., Pena, N., et al. (2011). Greenhouse Gas Benefits of a Biogas Plant in Austria (Joanneum Research Forschungsgesellschaft mbH. Resources – Institute of Water, Energy and Sustainability).

85. Woess-Gallasch, S., Enzinger, P., Jungmeier, G., and Padinger, R. (2007). Treibhausgasemissionen aus Biogasanlagen (Graz).

86. Al Seadi, T., Drosg, B., Fuchs, W., Rutz, D., and Janssen, R. (2013). Biogas digestate quality and utilization. In The biogas handbook (Elsevier), pp. 267–301. https://doi.org/10.1533/9780857097415.2.267.

87. Czubaszek, R., and Wysocka-Czubaszek, A. (2018). Emissions of carbon dioxide and methane from fields fertilized with digestate from an agricultural biogas plant. Int. Agrophys. *32*, 29–37. https://doi.org/10.1515/intag-2016-0087.

88. Hrad, M., Piringer, M., and Huber-Humer, M. (2015). Determining methane emissions from biogas plants–Operational and meteorological aspects. Bioresour. Technol. *191*, 234–243. https://doi.org/10.1016/j.biortech.2015.05.016.

89. Jensen, M.B., Møller, J., and Scheutz, C. (2017). Assessment of a combined dry anaerobic digestion and post-composting treatment facility for source-separated organic household waste, using material and substance flow analysis and life cycle inventory. Waste Manag. *66*, 23–35. https://doi.org/10.1016/j.wasman.2017.03.029.

90. Maldaner, L., Wagner-Riddle, C., VanderZaag, A.C., Gordon, R., and Duke, C. (2018). Methane emissions from storage of digestate at a dairy manure biogas facility. Agric. For. Meteorol. *258*, 96–107. https://doi.org/10.1016/j.agrformet.2017.12.184.

91. Oshita, K., Okumura, T., Takaoka, M., Fujimori, T., Appels, L., and Dewil, R. (2014). Methane and nitrous oxide emissions following anaerobic digestion of sludge in Japanese sewage treatment facilities. Bioresour. Technol. *171*, 175–181. https://doi.org/10.1016/j.biortech.2014.08.081.

92. Awiszus, S., Meissner, K., Reyer, S., and Müller, J. (2018). Ammonia and methane emissions during drying of dewatered biogas digestate in a two-belt conveyor dryer. Bioresour. Technol. *247*, 419–425. https://doi.org/10.1016/j.biortech.2017.09.099.

93. Bühler, M., Häni, C., Ammann, C., Brönnimann, S., and Kupper, T. (2022). Using the inverse dispersion method to determine methane emissions

from biogas plants and wastewater treatment plants with complex source configurations. Atmos. Environ. *13*, 100161. https://doi.org/10.1016/j.aeaoa.2022.100161.

94. Senga Kiessé, T., Corson, M.S., and Eugène, M. (2022). The potential of kernel density estimation for modelling relations among dairy farm characteristics. Agric. Syst. *199*, 103406. https://doi.org/10.1016/j.agsy.2022.103406.
95. Čížek, P., and Sadıkoğlu, S. (2020). Robust nonparametric regression: a review. Wiley Interdiscip. Rev. Comput. Stat. *12*, e1492. https://doi.org/10.1002/wics.1492.
96. Selingerova, I., Katina, S., and Horova, I. (2021). Comparison of parametric and semiparametric survival regression models with kernel estimation. J. Stat. Comput. Simulat. *91*, 2717–2739. https://doi.org/10.1080/00949655.2021.1906875.
97. Scott, D.W. (2015). Multivariate Density Estimation: Theory, Practice, and Visualization (John Wiley & Sons). https://doi.org/10.1002/9781118575574.
98. Rubinstein, R.Y., and Kroese, D.P. (2011). Simulation and the Monte Carlo Method (John Wiley & Sons).