Energy & Environmental Science



PAPER

View Article Online



Cite this: Energy Environ. Sci., 2024, 17, 1482

Received 1st August 2023, Accepted 8th January 2024

DOI: 10.1039/d3ee02516k

rsc.li/ees

A comparative study of biogas and biomethane with natural gas and hydrogen alternatives†

Semra Bakkaloglu ** and Adam Hawkes

Biogas and biomethane are renewable fuels that can help to reduce greenhouse gas (GHG) emissions. However, their supply chains emit methane (CH₄), a potent GHG. This study explores the role of biomethane and biogas supply chains in decarbonising the energy system by comparing their life cycle emissions to alternative scenarios. These alternatives consider the entire life cycle emissions from different ways of treating biodegradable waste rather than anaerobic digestion (upstream), producing natural gas or hydrogen instead of biogas and biomethane (midstream), and using synthetic fertiliser instead of digestate (downstream). We present 22 life cycle assessment (LCA) GHG intensity models for biomethane and biogas and compare them with three primary counterfactual scenarios based on various midstream stages aimed at compensating for biogas and biogas generation. Our findings reveal that biogas and biomethane supply chains achieve an average of 51-70% and 42-65% GHG savings compared to midstream natural gas and all hydrogen production routes, respectively. Conversely, low-carbon green hydrogen at the midstream stage counterfactual scenario outperforms biomethane and contribute to an average of 13-24% GHG savings. Overall, the study suggested that biogas and biomethane have the potential to play a role in reducing GHG emissions as a cleaner and sustainable alternative to fossil fuels, but they are not the lowest-emission option.

Broader context

Biomethane and biogas are renewable energy sources that can be produced from organic waste. They are a clean and sustainable alternative to fossil fuels, and they can help to reduce greenhouse gas (GHG) emissions. However, biomethane and biogas also emit methane, which is a potent greenhouse gas. This study assesses the emissions from biomethane and biogas supply chains, comparing them to three counterfactual scenarios. The counterfactuals consider the total GHG emissions that would occur if biodegradable waste were not converted into biomethane, and if the utilisation of biomethane were replaced by natural gas or hydrogen, and if the biproduct digestate did not substitute mineral fertilisers. The study found that biogas and biomethane supply chains can contribute to significant GHG savings compared to natural gas and hydrogen. However, low-carbon green hydrogen generation scenarios outperform biomethane generation and can contribute to even greater GHG savings.

1. Introduction

Biomethane and biogas already play a significant role in energy systems as countries strive to move away from fossil fuels to achieve the Paris Agreement's well below 2 °C temperature rise target.1 In order to limit greenhouse gas (GHG) emissions and increase the share of renewable energy, the utilisation of biogas and biomethane utilisation is expected to rise in future. According to the SR1.5 report of the Intergovernmental Panel on Climate Change (IPCC) AR6, in scenarios that limit warming

Department of Chemical Engineering, Imperial College London, SW7 2AZ, UK. E-mail: s.bakkaloglu@imperial.ac.uk

† Electronic supplementary information (ESI) available. See DOI: 10.1039/ d3ee02516k

to 1.5 °C with low or no overshoot biomass-based gases could provide up to 28 exajoules of energy by 2050. According to the International Energy Agency (IEA), if biomethane and biogas production were to reach its sustainable potential, it could meet nearly 20% of global gas demand.3 In extreme scenarios, biomethane potential could even reach up to 1300 EJ by 2050, with the availability of aquatic biomass such as macroalgae (seaweed).4

Biogas, consisting of CH₄ (45-75% by volume) and CO₂, is primarily used for generating heat and electricity, as well as for household purposes such as cooking and heating in developing countries. Upon conversion into biomethane through upgrading processes (CH₄ content > 96%⁵), it can easily be stored and/ or injected into the gas grid or used as a transportation fuel. These applications are expected to be its primary future uses to

meet the need for reliable and affordable energy. The biogas and biomethane generation pathways start with the decomposition of organic wastes, such as food, garden waste, sewage sludge, manure, and energy crops, through anaerobic digestion (AD)³ to generate biogas, which is a mixture of gases consisting primarily of CH₄ and carbon dioxide (CO₂). Subsequently, a biogas upgrading system eliminates impurities and produces a gas comprising 95-98% CH₄. The digestate, a by-product of this system, can be used as a soil conditioner, compost, or fertiliser. However, it is important to note that fugitive CH₄ emissions may be produced at each stage of the biomethane and biogas supply chain, 6,7 and under certain conditions, these emissions could even surpass those from the natural gas supply chain per unit of energy delivered. Therefore, minimising fugitive CH₄ emissions is critical for sustainability.^{6,8}

Recent studies indicate that estimates of GHG savings from bioenergy pathways are influenced by system definitions, data inputs, and methodologies.9 The UK's SUPERGEN Bioenergy Hub¹⁰ suggests that many bioenergy pathways have the potential to produce energy with lower GHG emissions compared to fossil fuels. These estimates are calculated by considering alternative bioenergy life cycle assessment (LCA) scenarios, such as the use of land and resources if not allocated to bioenergy production. LCAs are the most frequently applied methodology for evaluating the GHG performance of biogas and biomethane generation pathways. 11-13 The main purpose of LCA is to assess the full impact of a pathway, taking account of emissions avoidance by the substitution of natural gas and synthetic fertiliser with biogas and biomethane and solid digestate. Bakkaloglu et al. 11 show that fugitive CH4 emissions play a significant role in LCAs of biomethane and biogas generation systems. Estimating the credits for CH₄ emissions reduction resulting from the conversion of feedstock into biomethane is a complex task, contingent on establishing relevant counterfactual cases as stated by IEA.3 Therefore, to estimate net GHG savings, emissions from whole biogas and biomethane production life cycles must be compared with alternative waste treatment, energy, and fertiliser production systems.

Although recent studies demonstrate that AD emits considerable amounts of CH₄, 8,14 there is a paucity of information on what would happen to biowaste material if not utilised for biogas generation and if gas were obtained from other sources, such as natural gas or hydrogen. Without this comprehensive counterfactual, it is difficult to fully assess the merits of biomethane production in the context of climate change mitigation. While some studies address counterfactuals to AD usage from various feedstocks^{15,16} and land utilisation^{13,17} and compare GHG savings from biomethane generation and natural gas production, 18,19 none evaluate counterfactual scenarios comprehensively across all stages of the biogas and biomethane supply chain. The purpose of this study is to assess the actual GHG emissions of various biogas and biomethane supply chains. These supply chains begin with the treatment of biodegradables by AD through to biogas and biomethane production, together with digestate as a fertiliser. A set of counterfactual scenarios are then constructed to estimate GHG emissions resulting from alternatives, corresponding to each stage of biogas and biomethane supply chains. These alternatives must capture different biodegradable waste treatment routes, energy generation options, and mineral fertiliser production to provide valuable insights.

The study aims to provide a better understanding of the GHG impact of generating biogas and biomethane from organic compounds, relative to alternatives, by examining the whole supply chain impact and comparing this to the option of not using the organic waste for biomethane generation (i.e. leaving it as is or treating it with various waste treatment technologies), and obtaining energy from an alternative source, and using mineral fertiliser instead of produced digestate. This life cycle emissions (LCE) comparison will explicate the potential role of biogas and biomethane supply chain in decarbonising energy systems, achieving the net-zero goal for waste management, and increasing renewable energy generation.

Methodology

2.1. Business as usual (BAU) scenarios for biogas and biomethane generation

2.1.1. Goal, scope and boundary definition. The study estimates the GHG savings achieved through the life cycles of current biogas and biomethane production, allowing for comparison with a comprehensive set of "counterfactual" scenarios. These counterfactuals consider the level of LCE (or GHG intensities, kg CO2-eq. per MJ) that would have occurred if the biomass had not been converted into biomethane and the energy had been replaced by natural gas or hydrogen. To achieve this goal, we have developed 22 various LCA models to gain a comprehensive understanding of the business-asusual (BAU) LCE associated with biogas and biomethane supply chains. These 22 LCA models have been constructed to encompass a range of feedstock types, biogas generation and upgrading technologies, and end-use scenarios for biogas and biomethane to enable a comprehensive assessment. The detailed list of 22 LCA models is provided in the ESI† Table S1. These models include five main feedstock types (manure, biowaste, sewage, vegetable cooking oil, and maize silage), two biomethane generation technologies (AD and gasification), three biogas upgrading technologies (amine washing, pressure swing adsorption (PSA) and membrane) to produce to biomethane (see Fig. 1a). Out of these models, five consider biogas generation and its utilisation through CHP systems. Thirteen LCA models focus on biomethane generation, encompassing three upgrading systems (see Fig. 1a). Additionally, four aggregated LCA models were developed to simulate wood-chips biomass gasification to produce biomethane (Fig. 1b and Table S1, ESI†). Given the comprehensive dataset available for the biogas and biomethane supply chain, these 22 LCA models meticulously include all main supply chain routes.

The boundary of this study encompasses the gate to grave life cycle, starting from the point of biomass availability at the treatment facility and ending at the final usage of biomethane

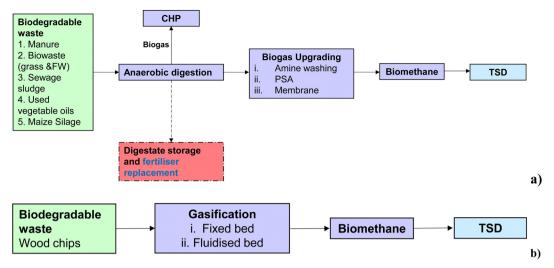


Fig. 1 (a) Biodegradable waste treatment in various AD scenarios for different end-use purposes. Green boxes represent the upstream stage, purple boxes represent the midstream stages, including transmission, storage and distribution (TSD) stages (blue box), and the red box demonstrates the downstream stage. (b) Wood biomass gasification route. Notes: system credits are only given in the LCA models where the digestate is as a fertiliser replacement.

or biogas. GaBi V10 software was used to build the LCA models,20 and the LCA was conducted following the steps outlined in ISO 14040/14044.21,22 The functional unit was defined as follows: the treatment of 1 kg of biodegradable feedstock input for biomethane generation from AD, and the production of cubic meters (m³) of biomethane from gasification processes. The life cycle includes the treatment of biodegradable waste and the recovery of valuable resources, including associated GHG emissions and environmental impacts. The construction of AD and upgrading units were also within the scope of the study but their decommissioning was not included due to missing data. Various higher heating values (HHV) are considered for each feedstock for unit conversion, as listed in Table S2 (ESI†). The HHV for biomethane is assumed to be 38.1 MJ per m3 for the purpose of unit conversion.

2.1.2. Inventory data, sensitivity analysis, environmental impact assessment, and uncertainty analysis for biogas and biomethane LCA models. Our goal is to include a comprehensive range of feedstocks by using all available data from the literature to represent global state of biogas and biomethane LCA models. Inventory data were adapted from Bakkaloglu et al. 11 for biowaste, and the Thinkstep and Ecoinvent v.3.8 available databases for all biodegradables including manure, sewage sludge, and used vegetable oil AD treatment systems²³ (see Table S3, ESI†). The maize silage LCA model is adopted from Thyø et al.24 The inventory data for CHP and biogas upgrading units are assumed to be the same for all types of feedstocks, as specified in Tables S4 and S5 (ESI†), respectively, and were obtained from the Ecoinvent v3.8 database, 23 except for CH₄ emissions. The 18 LCA models considering AD have been characterised using the most recent CH₄ emissions data for each stage of biogas and biomethane supply chains, including different feedstocks, AD and biogas upgrading technologies as well as digestate storage unit based on Bakkaloglu et al.'s

findings. 6 We defined CH₄ emissions as minimum, average and maximum ranges given in Table S6 (ESI†) for each stage to perform a sensitivity analysis on the entire LCEs. The composition of the different feedstocks is considered to calculate emissions in terms of grams of CO_{2-eq.} per megajoule of energy based on HHV values of each feedstock type (Table S2, ESI†). Additionally the data for the gasification process used in this study is sourced from Ecoinvent v.3.8,23 and it is available for two specific regions: Switzerland and the rest of the world. While these representations may not cover regional variations, they provide a consistent dataset to compare the different supply chain configurations. The CH₄ emissions from transmissions, storage and distribution stages have been adopted from Bakkaloglu et al.6

In this study, we have considered the global warming potential (GWP) as the key performance indicator to compare the different scenarios. GWP is the standard metric commonly used in LCAs to quantify the global warming impact of GHG emissions. The GWP₁₀₀ and GWP₂₀ indices are based on the time-integrated radiation forcing of fully mixed GHGs relative to that of CO2, over a 100-year and 20-year horizons, respectively. IPCC AR6 impact category has been used to estimate impacts on global warming and climate change (CC) including biogenic CO₂ as it uses more recent CO₂ equivalences.¹⁹

In order to examine the uncertainty of LCE from biogas and biomethane supply chain routes, we performed the Monte Carlo (MC) method²⁵ using Python. We used kernel density estimation to calculate the non-parabolic probability density function (PDF) of LCE emissions from the LCA model obtained by GaBi software and TSD stages from the literature,6 then summed up those stages for biomethane supply chains. We carried out a random simulation of 10 000 runs to determine cumulative supply chain emissions. The MC method is recommended by the IPCC to integrate uncertainties across various probability distributions, ranges, and correlation structures²⁶ and it is widely used in GHG accounting.6,11,27 Thus, it can capture uncertainties in input data, such as different processes for each stage and regional representation of gasification technologies, and was employed in this study.

2.1.3. Biogas and biomethane generation routes. Biodegradable waste is treated by AD to generate biogas for CHP generation and/or to upgrade biogas to biomethane for injection into the gas grid (see Fig. S1, ESI†). In our 22 LCA models, we considered five major biodegradables (manure, biowaste, sewage, vegetable oil and maize silage) as feedstocks to be treated in AD (see Table 1). The first stage of biogas and biomethane generation is feedstock storage, followed by AD, biogas upgrading, and digestate storage and handling. To ensure a consistent comparison with different counterfactual scenarios in the biogas and biomethane supply chain, we excluded the transportation distance of biowaste collection from residential, service, or industrial sectors to the AD facility. As explained in Section 2.2, we considered various alternative waste treatment methodologies in the counterfactual cases. Excluding collection and transportation to treatment facilities ensures consistency in our comparison. We assumed the source-separated collection of biodegradable waste for each feedstock type based on the Ecoinvent database. 11,23

This study considers single-stage continuous mesophilic AD reactors operating at 35 °C with a capacity to process 10 000 tonnes of waste per year and a projected lifespan of 25 years, which are typical parameters for commercial municipal organic waste treatment facilities.²⁸ The biogas produced through AD is utilised in a CHP unit to simultaneously generate heat and electricity, with assumed efficiency rates of 59% and 41% for heat and electricity, respectively.²³ The data inventory for each type of feedstock treatment in the AD processes is given in Table S3 (ESI†). Additionally, we considered that the digestate resulting from the AD process would be used as fertiliser, requiring it to be heated to 70 °C for 1 hour to meet the British Standard Institution's Publicly Available

Specification (BSI PAS 110).²⁹ We assumed a 100% uptake rate for potassium and phosphorus and 40% and 50% for organic nitrogen for manure and biowaste type feedstocks, respectively.³⁰ The details of the digestate properties to displace fertiliser depend on the feedstock type, which are provided in the Table S3 (ESI†). It is important to note that the digestate from sewage type feedstock treatment in AD is not used as fertiliser based on the Ecoinvent v3.8 dataset.23 This vields additional results that help us better understand scenarios where digestate is not used as fertiliser. Fugitive CH₄ emissions for feedstock storage, AD, upgrading methods and digestate stage were revised based on Bakkaloglu et al.6 (see Table S6 for the details, ESI†). The emission data for CO₂, ammonia, hydrogen sulphide and nitrous oxides for each stage were taken from Ecoinvent.23

At the upgrading stage, based on data availability in the Ecoinvent v.3.8 dataset, 23 three major biogas upgrading systems were considered: amine washing, pressure swing adsorption (PSA), and membrane (see Table S5, ESI†). The CH4 emissions from each upgrading processes are also revised based on findings from Bakkaloglu et al.6 (see Table S6, ESI†). In addition to biogas and biomethane generation from AD, we have also included the biomass gasification route for biomethane generation (Fig. 1b). Wood biomass chips are sent for gasification to produce biomethane by fixed and fluidized bed gasification processes.

The generated biomethane can be injected into the gas grid or used to generate electricity. To compare alternative scenarios for end-use gas utilisation, we considered that natural gas, or hydrogen are the potential energy carriers in the gas grid instead of biomethane.23 Therefore, we considered different combinations of feedstock with different end-use applications, including CHP and upgrading to biomethane through three different upgrading systems for injection into the gas grid (see Table S1, ESI†). We did not apply natural gas and heat and power replacement credits to the biomethane and biogas

Table 1 GHG intensities stages associated with the counterfactual LCA and biomethane supply chain

Emissions stage	Counterfactual case	BAU	
Upstream emissions (feedstock: biowaste, manure, sewage, vegetable oil and maize silage)	(i) Send to landfill with gas recovery(ii) Send to open dumping(iii) Send to incineration(iv) Send to composting(v) Maize silage production	Emissions from feedstock storage and pre- treatment	
Processing emissions (energy generation)	(i) Natural gas production and processing (ii) Hydrogen production and processing	Emissions from anaerobic digestion and upgrading unit.	
TSD¹ of emissions	(i) Natural gas transmission, storage, and distribution	Emissions from biogas/biomethane transmissions, storage, and distribution	
Downstream emissions (fertiliser)	Fertiliser life cycle: production and spreading on landfill as an alternative to digestate	Emissions from digestate storage and credits given to the replacement of fertiliser	
Total emissions	Sum of emissions from all stages	Sum of emissions from all stages	

Note: TSD: transmissions, storage and distribution.

LCA models to allow for direct comparison with the counterfactuals. The biogas LCA models consider on-site CHP generation, excluding emissions from the TSD stage.

In the downstream stage, the digestate is used as fertiliser, therefore system credits were applied for the digestate which replaces an equivalent. The CH₄ emissions related to the transmission, storage, and distribution (TSD) stages were obtained from Bakkaloglu et al.6 (see Table S6, ESI†). It was assumed that the GWP₁₀₀ of 28 for CH₄ would align with the results from IPCC AR6 for LCA studies. These TSD emissions were then integrated into the overall LCE emissions obtained from LCA models by employing MC simulation as explained in the Section 2.1.2.

2.2. Counterfactual scenarios

Because the goal of this study is to understand what would happen if the biodegradable feedstock is not sent to the AD for energy generation, we consider alternative scenarios for each stage of the biomethane generation process. This allows us to scenarios relating to each stage of the supply chain. Upstream and downstream emissions were assumed to be consistent across scenarios C1, C2, and C3. The only distinction among the counterfactual scenarios lies in the midstream stages, specifically gas production for energy generation. C1 considers the complete LCE of natural gas production, C2 covers the entire hydrogen production routes, and C3 focuses solely on the LCE of low-carbon green hydrogen production at midstream stages (Table 2).

After assessing emissions from the counterfactuals at the upstream, mid-stream, and downstream stages, the kernel density estimation (KDE) function was used to assess the characteristics of data distribution gathered for each stage of the counterfactual supply chain. An MC simulation was then performed to estimate total supply chain emissions, which were compared with actual biogas/biomethane supply chain emissions obtained from the 22 LCA scenarios. GHG savings (%) from the BAU scenarios were calculated using the following equation.

Total GHG emissions from counterfactual LCA - Total GHG emissions from BAU scenarios Total GHG emissions from counterfactual LCA

examine the counterfactual case of biomethane and biogas generation, considering the potential outcomes and impacts if each stage of biogas and biomethane supply chain follows an alternative path. Total GHG intensity from the biogas and biomethane supply chains, including fugitive CH4 emissions directly from each stage of supply chains, were compared with the counterfactuals in order to quantify GHG emission savings. Each stage associated with GHG intensities from both the AD plant and the alternatives are depicted in Table 1. Emissions were categorised as upstream (feedstock processes), mid-stream (energy generation processes), or downstream (digestate/fertiliser production process) for the counterfactual and BAU cases (Table 1). Additionally, we considered three alternative scenarios based on the energy generation routes (refer to Table 2). These scenarios were designated as C1, C2, and C3, representing the emissions associated with the TSD stages of natural gas and/or hydrogen. Fig. 2 also illustrates the BAU and counterfactual

2.2.1. Counterfactual scenarios for the upstream stage. GHG intensities from the upstream stage were calculated by assuming that the feedstock is not used in the AD process. The upstream counterfactuals differed on type of feedstock used for biogas and biomethane LCA models. For animal waste, it was assumed that manure is collected and transported to storage tanks, ponds, or lagoons on farms, whereby it is stored for a year in open tanks or lagoons or it is sent to the composting facility. GHG emissions released into the atmosphere are affected by the storage conditions and composition of the slurry. For energy crops and grass, it was assumed that available land is utilised to grow maize silage, and GHG emissions for energy crop counterfactuals included those from crop production, harvesting, processing, and transportation of maize silage. It was also assumed that food and garden (biowaste) waste and

Counterfactual scenario	Emission stages
C1. NG	Upstream emissions (alternative feedstock treatment), process emissions come from natural gas generation; TSD emissions come from natural gas; downstream emissions (synthetic fertiliser production)
C2. HYG	Upstream emissions (alternative feedstock treatment), LCE come from black/blue/grey/turquoise/green and pink hydrogen gas production, storage and distribution (cradle to gate (well to tank)) LCA studies; downstream emissions (synthetic fertiliser production)
C3. Low-carbon green HYG	Upstream emissions (alternative feedstock treatment), LCE come from low-carbon green hydrogen generation (electricity comes from solar and wind); downstream emissions (synthetic fertiliser production)

Note: HYG: hydrogen; LCE: life cycle emissions; NG: natural gas; TSD: transmission, storage, and distribution. Upstream and downstream emissions are identical for C1, C2, and C3 scenarios. The only distinctions between the counterfactual scenarios lie in midstream emissions, signifying gas generation LCE. Green hydrogen is the term used for hydrogen generation through electrolysis using renewable energy, such as wind and solar electricity.

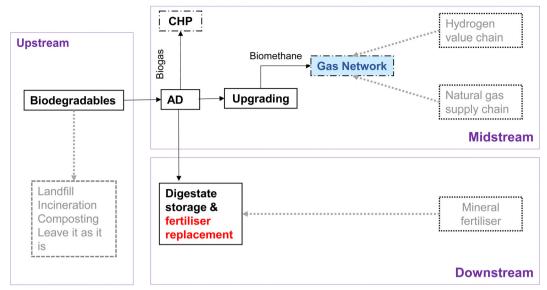


Fig. 2 Representation of counterfactual and BAU scenarios. Note: grey dashed lines show counterfactuals, red lines represent credits given in the LCA models, and purple boxes show the three main stages of the supply chains.

sewage sludge feedstock are sent to landfills with energy recovery, incineration, composting facility or to open dumping sites. We used the Ecoinvent v.3.823 and Thinkstep20 database and to estimate GHG emissions from this stage, using GaBi v.10²⁰ (Table 3). The emission factors of various feedstock types for the composting process are reported in Walling and Vaneeckhaute study.31 Details of feedstock emission factors for composting are given in the Table S7 (ESI†).

The GWP_{100} was obtained from the IPCC AR6 including the biogenic CO2 impact category. To calculate emission intensities in terms of kg CO_{2-eq.} per MJ, we obtained the HHV of different types of feedstocks and considered the lower and higher ranges of HHV in the extant literature (see Table S2, ESI†). These emissions ranges were included at this stage to assess uncertainty in the MC simulation.

2.2.2. Counterfactual scenarios for midstream stage (energy generation). At the mid-stream stage, natural gas or

hydrogen usage was compared to the usage of biomethane. The Ecoinvent v.3.8 database²³ was used to obtain emission factors for natural gas generation at the production, transmission, distribution, and storage (TSD) stages, and LCA literature were reviewed for hydrogen production and TSD stages (see Table 4).

2.2.2.1. Natural gas and hydrogen generation. Biogas from AD plants is replacing fossil fuels for heat and electricity generation, and biomethane can be injected into the gas grid and used in the same manner as natural gas. Therefore, the natural gas life cycle was the primary counterfactual (C1) in this study. We obtained natural gas production and processing data from Ecoinvent v.3.8 database, 23 including data from various region of the world. This data covers the diversity in natural gas composition and associated purification processes, as well as emissions data. The emissions for the TSD stage of natural gas were collected from Ecoinvent v.3.8 database and Balcombe et al. 32 (see Table 4).

Table 3 1 kg of feedstock GHG emissions obtained from GaBi v.10 considering the aggregated processes and literature review (see Tables S2 and S7, ESI)

Feedstock type	Counterfactual treatment	GHG emissions kg CO _{2-eq.} per kg feedstock	MJ _{HHV} per kg of feedstock type, MJ kg ⁻¹
Biowaste (including food waste and garden waste)	Sent to composting (in-vessel and open windrow composting as well as industrial composting) or incineration or landfill	0.17 to 1.9 (34)	9.0 to 17.1
Manure/animal slurry	Liquid and solid manure spread onto fields with manure spreader, or dried and pelleted or stored or composting	1.1×10^{-5} to 2.8 (30)	4.7 to 11.6
Maize silage	Left as maize silage production or composting	-1.5 to -0.23 (14)	9.5 to 16.2
Sewage sludge	Sent to incineration or landfill or composting	0.001 to 1.5 (22)	9.5 to 17.5

Note: The total of data points for each feedstock treatment are shown in parentheses. All GHG emissions are included. Source of data: Ecoinvent v.3.818 and Thinkstep dataset.2

Table 4 The LCE of natural gas supply chain and hydrogen value chain from the literature

Gas production	GHG intensities, kg $CO_{2\text{-eq.}}$ per MJ_{HHV}	Ref.
Natural gas production and processing	0.002 to 0.4 (15)	18 and 23
Natural gas transmission, distribution, and storage	0.0002 to 0.02 (47)	23, 32 and 36
Hydrogen value chain LCAs	0.003 to 0.24 (80)	23 and 37–52
Low-carbon green hydrogen LCAs	0.005 to 0.018 (18)	37, 39, 53 and 54

Given the UK's net-zero goal, hydrogen utilisation will play a major role in the future gas mix.33 The LCE of various black (coal gasification)/grey (natural gas reforming)/blue (natural gas reforming with carbon capture storage (CCS))/pink (electrolysis driven by nuclear power)/turquois (methane pyrolysis) and green hydrogen (electrolysis driven by renewables such as wind turbine or solar panel)34 value chain methods (C2) were gathered from various literature sources. These LCA models include coal gasification (with and without CCS), natural gas steam methane reforming (SMR) and autothermal reforming (ATR) (with and without CCS), biomass gasification (with and without CCS) and electrolysis with nuclear, solar, wind technologies as well as thermochemical water splitting. The system boundary for the selected routes is "well to tank", and their functional units (kg H2 produced) are adjusted in order to report them as kg CO_{2-eq.} per MJ. The GHG intensity data collected from literature for various hydrogen production routes is presented in Table S8 (ESI†). In addition, we included the low carbon green hydrogen production route for the counterfactual scenario 3 (C3). In this C3 scenario, we solely considered hydrogen production from wind and solar electricity generation for electrolysis based on the UK Low Carbon Hydrogen Standard, which requires meeting a GHG emissions intensity of 20 g CO_{2-eq.} per MJ_{LHV}.35 The selected data for this analysis can be found in ESI† Table S9.

2.2.3. Counterfactual scenario for downstream process and emissions. The digestate produced from anaerobic digestion of biodegradables is normally used for fertiliser or compost. However, if the biowaste is not digested, fertiliser production is required. Therefore, in our counterfactual scenario, we considered the same amount of synthetic fertiliser (nitrogen (N), phosphate (P), potassium (K)) production instead of digestate production to be used for fertiliser. Extensive literature on GHG emissions from the production, storage, transportation, and application of synthetic fertilisers (N, P, K) was reviewed and data were collected (see Table 5). Considering the replacement of N, P and K fertilisers with digestate, the total downstream

counterfactual emissions involve summing the LCE of ammonium nitrate (as N), phosphate (P_2O_5) and potash (K_2O) fertilisers, based on their respective contents in the digestate (see Table 5).

3. Results and discussion

3.1. Biogas and biomethane supply chain LCE

Five LCA models of the BAU case for biogas generation are evaluated with respect to CHP generation, and 17 biomethane LCA models are assessed to produce biomethane. Based on the analysis of 22 LCA results (see Table 6), it was found that biomethane generation yields the lowest GHG intensities (27-29 g CO_{2-eq.} per MJ on average and 18-20 g CO_{2-eq.} per MJ at the median) and CHP generation from biogas (47-50 g CO_{2-eq.} per MJ on average and 36-38 g CO_{2-eq.} per MJ at the median) reveals the highest emissions. Previous studies reported that biomethane generation GHG intensities ranging from -210 to 125 g $CO_{2\text{-eq.}}$ per MJ, 18,67 depending on waste type and credits given to the system. Our findings are in line with these studies, but it is important to note that we did not apply credits for natural gas replacement. The results for CH₄, CO₂, N₂O emissions and GPW₂₀ for each LCA model are also provided in Table S10 (ESI†). Considering CH4's relatively short lifetime (9-12 years²), GWP₂₀ values provide greater insights, as it is recommended to present them in a twohorizon matrix for LCA studies.⁶⁸ However, the discussion is based on GWP₁₀₀ values to facilitate comparison with counterfactual scenarios, as they are generally reported in GWP100 values.

Wood chip gasification for biomethane generation is a promising technology with the potential for technological innovation and cost reductions, but it is still in its early stages of development.³ The LCE from gasification plants are relatively lower than those from AD plants (see Table S10, ESI†). This discrepancy arises because we have had to rely on default

Table 5 Range of life cycle emissions from fertiliser production based on data obtained from the literature

Synthetic fertilizer	GHG intensities, kg $\mathrm{CO}_{2\text{-eq.}}$ per kg fertiliser	Ref.
Ammonium nitrate, as N fertiliser	3.5 to 10.3 (9)	31 and 55–61
Phosphate P ₂ O ₅ , as P fertiliser	0.4 to 8.9 (16)	31 and 55–64
Potash, K ₂ O as K fertiliser	0.14 to 0.25 (2)	31 and 65

Note: it is assumed that 1 tonne of digestate includes 5 kg of ammonium nitrate, 0.5 kg of phosphate as P_2O_5 and 2 kg of potash as K_2O^{30} (number of data samples are represented in the brackets). In total, 288 data points were obtained for the counterfactual downstream emissions considering all possible combination of the available data for N, P and K fertilisers. The calorific value of digestate is taken as 15 MJ per kg of digestate.

GHG intensities, kg CO_{2-eq.} per MJ Mean emissions Median emissions 5th percentile 95th percentile Biogas generation with CHPa 0.047 to 0.050 0.036 to 0.038 -0.011 to -0.0100.141 to 0.145 Biomethane generation^b 0.027 to 0.029 0.018 to 0.020 -0.026 to -0.0250.117 to 0.120 Biomethane generation from AD 0.042 to 0.045 0.030 to 0.032 0.0033 to 0.0040 0.126 to 0.131 Biomethane generation from AD with highest CH₄ emission rates 0.083 to 0.084 0.072 to 0.073 0.017 to 0.018 0.182 to 0.185

Note: all results obtained from 10000 random runs of MC simulation. aCHP engine is considered on-site and TSD stage emissions are not included. ^bBiomethane generation from both AD and gasification are considered.

emission factors from the Ecoinvent data for gasification, primarily because there is a lack of on-site mobile CH₄ measurements from gasification plants to the best of our knowledge. Since AD plants are more mature and widely deployed than wood chip gasification plants, and their LCE reflect realworld operating conditions (as we have revised CH₄ emissions based on mobile measurement studies), we have chosen to focus on the LCE from AD facilities. The average LCE of AD plants to generate biomethane were determined to be 42-45 g CO_{2-eq.} per MJ. The median emissions, lower than the mean emissions, ranged from 30 to 32 CO_{2-eq.} per MJ for biomethane LCA models.

Depending on the feedstock, vegetable cooking oil (VCO) had the highest range of LCE, followed by maize, sewage sludge, biowaste, and manure (Table S10, ESI†). This variation is primarily due to the wide range of CH₄ emissions associated with the VCO and maize systems (Table S6, ESI†). Additionally, the sewage system had higher LCE emissions than the manure and biowaste systems because the system credit for fertiliser usage was not applied to the sewage LCA model. CH₄ emissions have a significant impact on the overall LCE of the system, consistent with findings in the literature. 6,11 Due to CH₄

emissions and the energy consumed in each stage, the biogas upgrading systems had the highest LCE, followed by digestate (which has the highest CH₄ emissions rates⁶), AD, and feedstock storage stages. Among the upgrading methods, the membrane process exhibited the lowest emissions compared to amine washing and PSA. It tends to have lower CH₄ emissions and requires less energy than the amine washing system. The amine washing system requires heat for steam generation and electricity,23 resulting in higher emissions than other upgrading stages.

Due to the significant impact of CH₄ emissions on the overall LCE of the system, we have reported the highest LCEs within the biomethane supply chain from AD facilities, which are based on the highest CH₄ emissions in the supply chain (see Table S6, ESI†). The emissions range from 17.4 to 185 g CO_{2-eq.} per MJ at the 5th and 95th percentiles, respectively. The mean emissions are 83-84 g CO_{2-eq.} per MJ, and the median emissions are 72-73 g CO_{2-eq.} per MJ (see Table 6). This indicates that the GHG intensities of biomethane supply chains are highly dependent on the CH4 emissions range, which can range from as low as −25 to as high as 183 g CO_{2-eq.} per MJ (see Table 6).

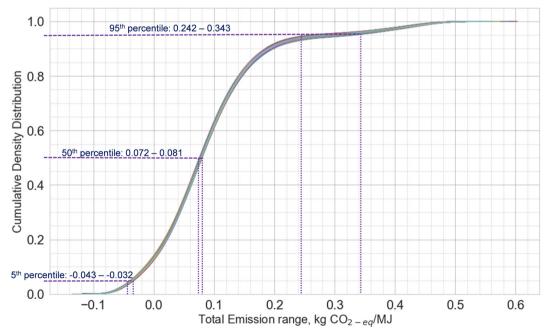


Fig. 3 Results from 10 000 Monte Carlo simulation runs for the first counterfactual scenario (C1, natural gas utilisation at midstream stages). The figure includes 1000 curves.

3.2. C1. Natural gas utilisation instead of biogas/biomethane generation

The first main counterfactual scenario (C1) considers alternative upstream treatments of biodegradables (feedstock), midstream natural gas generation, and synthetic fertiliser production at downstream. In this scenario, there are four main stages: treatment of biodegradables; natural gas generation and processing; TSD of natural gas; and synthetic fertiliser production. The GHG intensity of these four stages were calculated using MC runs. The data characteristics of each stage of emissions are determined by the kernel density function before running 10 000 random runs of the MC model. Total emissions from this counterfactual scenario are up to 0.52 kg CO_{2-eq.} per MJ. The mean and median GHG intensities range are 0.091-0.093 kg CO_{2-eq.} per MJ and 0.072-0.081 kg CO_{2-eq.} per MJ at the 95% confidence interval (CI), respectively (see Fig. 3), with a 5th percentile of -0.033 kg $CO_{2-eq.}$ per MJ at the 95% CI. The 95th percentile of GHG intensity are 0.24-0.34 kg CO_{2-eq.} per MJ using GWP₁₀₀ values. The entire range of results for the C1 scenario can be found in Table 7. Previous research focusing on GHG emissions from the natural gas supply chain has reported mean estimates of GHG intensities ranging from 0.022 to 0.107 kg CO_{2-eq.} per MJ_{HHV}, taking into account both CO2 and CH4 emissions, 36 which aligns with our findings, particularly when considering alternative waste treatment and synthetic fertiliser GHG intensities. As observed in the cumulative distribution in Fig. 3, the 95th percentile of total supply chain emissions falls within a higher range compared to the 5th percentile, mean, and median emissions. Furthermore, the mean emissions are higher than the median emissions, indicating an upward-skewed emissions distribution. This skewness suggests the presence of high-emitting points in the supply chain. In our C1 scenario, we find results consistent with those in the literature for the natural gas supply chain, which also identifies "super-emitters" along the chain. 36,69-71 The highest emissions originate from natural gas processing due to super-emitters, followed by upstream emissions, TSD, and digestate stage emissions. The data for the upstream stage exhibits a much wider range due to the diverse range of feedstock treatments using various technologies. The negative emissions in the C1 scenario result from the maize feedstocks, as well as the lowest emissions range of the natural gas supply chain and synthetic fertiliser production.

Based on the C1 scenario, we found that biogas generation for CHP units and biomethane generation can lead to more significant GHG reductions compared to the C1 scenario. Specifically, biomethane generation from AD and gasification can achieve GHG saving of 69–70% and 75–76% compared to the C1 scenario, based on mean and median emissions, respectively. Excluding the biomass gasification route, it can be concluded that biomethane generation from AD plants can yield GHG savings of 51–53% and 58–61% based on mean and median emissions, respectively, at the 95th CI. Even with the highest CH₄ emissions, biomethane generation route from AD plants still provides an average GHG saving of 3–13% and a median GHG saving of up to 11% compared to the C1 scenario.

3.3. C2. Hydrogen instead of biogas/biomethane generation

In the C2 scenario analysis, we considered the same upstream and downstream emissions as in the C1 scenario, but midstream energy generation is provided from hydrogen generation. It is important to note that our analysis encompasses all hydrogen generation routes. Based on Fig. 4, the GHG intensity of the C2 scenario can emit up to 0.47 kg CO_{2-eq.} per MJ. The mean and median emissions range from 0.079 to 0.082 kg CO_{2-eq.} per MJ and from 0.064 to 0.071 kg CO_{2-eq.} per MJ at the 95th percentile CI, respectively (see Fig. 4 and Table 7). The 5th and 95th percentile of GHG intensities are -0.022 to -0.015 kg CO_{2-eq.} per MJ and 0.212 to 0.221 kg CO_{2-eq.} per MJ, respectively at 95th CI. The entire range of results for the C2 scenario can also be found in Table 7.

In the C2 scenario, the mean and median emissions are closely aligned, with the mean emissions being slightly higher. This suggests the presence of few high-emitting points, although the disparity between mean and median emissions is not as significant as observed in the natural gas supply chain (refer to Fig. 3). The cumulative graph of the C2 scenario exhibits a more normal distribution when compared to the C1 scenario (Fig. 4a), with the majority of data points clustered around the mean and only a few outliers. This distribution pattern arises from mid-stream (hydrogen supply chain) emissions aligning with upstream emissions when considering all hydrogen generation routes. In previous studies conducted by Speirs et al., 18 which calculated GHG emissions from hydrogen production using different technologies, emissions have found to vary between -0.10 to 0.18 kg CO_{2-eq.} per MJ HHV, H2. This range aligns with our midstream range of -0.002 to 0.24 kg CO_{2-eq.} per MJ_{HHV, H2}. To better understand which hydrogen generation routes, provide greater GHG savings compared to biomethane supply chains, we divided the midstream stages in Fig. 4b. If midstream gas production is obtained from pink or green hydrogen, GHG savings can be achieved in terms of mean and median emissions compared to biomethane generation from AD plants. However, blue, turquoise, grey, and black

 Table 7
 Life cycle emissions of counterfactual scenarios

GHG intensities, kg CO _{2-eq.} per MJ	Mean emissions	Median emissions	5 th percentile	95 th percentile
C1	0.086 to 0.096	0.072 to 0.082	-0.039 to -0.027	0.193 to 0.422
C2	0.074 to 0.086	0.063 to 0.075	-0.022 to -0.010	0.197 to 0.230
C3	0.034 to 0.040	0.027 to 0.033	-0.043 to -0.033	0.121 to 0.157

Note: the results include the entire range of data from 10 000 MC runs.

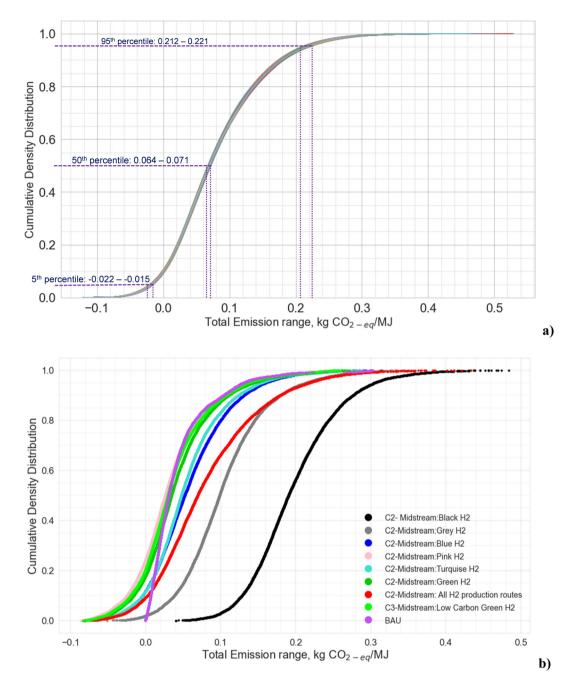


Fig. 4 (a) Cumulative density distribution of Monte Carlo simulation 10 000 runs for the second counterfactual scenario including 1000 curves. (C2, hydrogen utilisation at midstream stages); (b) representation of each hydrogen production route separately at midstream stages in the C2, as well as BAU and C3 scenarios. In the BAU, biomethane generation from AD is depicted, while gasification is excluded.

hydrogen generation at the midstream stage does not yield any GHG savings. Black and grey hydrogen may exhibit lower emissions within the 5th percentile of cumulative emissions due to maize feedstock, revealing negative emissions that offset GHG emissions from those high-emitting hydrogen production routes. The greatest GHG savings can be achieved using pink hydrogen at the midstream stage among all hydrogen routes. Notably, green hydrogen at the midstream stage (mean emissions: 0.041-0.044 kg CO_{2-eq.} per MJ; median emissions: 0.034-0.036 kg CO_{2-eq.} per MJ) can compete with biomethane from the AD supply chain (see Fig. 4b and Table 6).

In comparison to the C2 with BAU scenario, biomethane generation can achieve substantial GHG reductions of 63-65% on average and 71-73% in median emissions compared to the C2 scenario. Also, biomethane generation from AD facilities yields 42-45% GHG savings on average and 52-57% in median emissions at the 95th CI compared to the C2 scenario. It's important to note that biomethane generation from AD with the highest CH₄ emissions offers minimal GHG reductions up to 3% in median emissions, compared to C2 scenario, with no GHG reduction in terms of mean emissions. Furthermore,

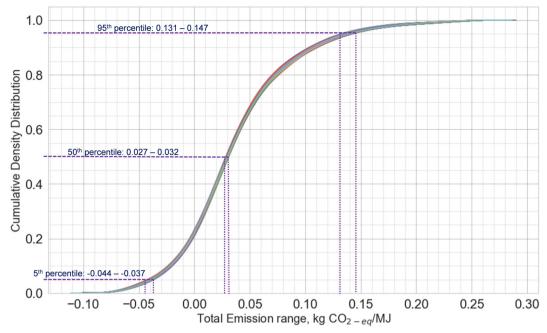


Fig. 5 Cumulative density distribution of Monte Carlo simulation 10 000 runs for the third counterfactual scenario (C3, only low-carbon green hydrogen utilisation at midstream stages). The figure includes 1000 curves.

hydrogen utilisation at midstream stage evidently leads to a 14-16% reduction in mean GHG emissions compared to natural gas usage at midstream stage (C1 scenario).

3.4. C3. Low-carbon green hydrogen utilization instead of biogas/biomethane generation

Similar to C3, we considered to the same upstream and downstream emissions as in the C1 and C2, but energy generation is only provided from low-carbon green hydrogen generation in C3 scenario. Based on Fig. 5, GHG intensity of C3 scenario can emit up to 0.26 kg CO_{2-eq.} per MJ. The mean and median emissions range are 0.036-0.038 kg CO_{2-eq.} per MJ and 0.027-0.032 kg CO_{2-eq.} per MJ at the 95th percentile CI, respectively (see Fig. 5 and Table 7 for whole range). The GHG intensities at 5th and 95th percentile are as follows: -0.044 to -0.037 kg $CO_{2-eq.}$ per MJ and 0.131 to 0.147 kg CO_{2-eq.} per MJ at the 95th percentile, respectively. In the C3 scenario, the mean and median emissions are more closely aligned than C2 scenario, albeit with mean emissions slightly exceeding the median emissions. The primary distinction between the C2 and C3 scenarios lies in the mid-stream stage emissions. Comparing the C2 and C3 scenarios reveals a notable reduction in GHG intensities achieved using the low-carbon hydrogen generation pathway. On average, this decision leads to a remarkable 51-53% decrease, while median emissions experience a substantial 56-57% reduction compared to all available hydrogen generation routes at the midstream stage (C2 scenario).

In the C3 scenario, the utilisation of low-carbon green hydrogen generation at midstream stage leads to significant GHG emission reductions. Conversely, the BAU scenario, which involves biomethane generation, still achieves substantial savings, with average emissions reduced by 21-28% and median

emissions by 33-39% when compared to the C3 scenario. It's worth noting that biomass gasification pathways contribute to an overall reduction in LCE. However, considering the maturity of biomethane generation technologies, AD facilities lose their GHG advantage in terms of average emissions compared to the low-carbon hydrogen generation route. The C3 scenario can reduce average GHG emissions by 13-24%; however, the BAU scenario, which exclusively focuses on AD facilities, can achieve a slight reduction in median emissions of up to 3% compared to the C3 scenario. If we consider the highest CH4 emissions from AD plants, the BAU scenario could potentially lose its advantage over the C3 scenario. This highlights the significance of CH₄ emissions in the decision-making process.

3.5. Limitations and future research

This research aimed to understand counterfactual emissions from each stage of the biogas and biomethane supply chain in order to gain an overall perspective on GHG savings. There are a few limitations to application of the study in the context of current waste treatment and energy system.

- This study specifically concentrated on GWP₁₀₀ and does not encompass other dimensions of sustainability, such as economic and social impacts. We acknowledge that GWP100 results are susceptible to uncertainties and variations arising from the data and assumptions employed in the counterfactual scenarios.
- Setting the boundaries for this study posed a challenge due to the presence of various stages in the life cycle of the examined system occurring in different geographic locations and involving different stakeholders. It is widely acknowledged that the selection of system boundaries can have a substantial

impact on the outcomes of an LCA study and may impose limitations on its scope, especially the LCA of hydrogen generation and TSD from various sources. Different energy and waste treatment systems' LCA results may be hardly comparable, due to differences between the chosen system boundaries, functional units and energy content. For example, sourcesegregated waste treatment is assumed, and the transportation distance of waste from the source to treatment facilities is not considered for either the BAU or counterfactual scenarios. Future studies could further analyse the impact of transportation distance for small local digesters in rural areas and large facilities collecting organic waste from large cities.

- The availability of data for biogas and biomethane LCA are limited in the Ecoinvent and Thinkstep databases, particularly when it comes to specific feedstock types, and countries. Variations in the types of grass, maize silage, and different ratios of feedstock can potentially affect the upstream emission profiles.
- We acknowledge that organic waste can be subjected to hydrothermal liquefaction and pyrolysis processes to produce biofuels. However, our study intentionally excludes the consideration of biofuel generation because our primary objective is to gain insights into counterfactual scenarios for the biogas and biomethane supply chain within the context of gas grid network decarbonisation. Therefore, we have not addressed transportation fuel usage in this research. Future studies could explore counterfactual scenarios related to biofuels.
- We standardised the representation of GHG intensities in terms of kg CO_{2-eq.} per MJ to ensure uniformity across the upstream, midstream, and downstream stages, enabling the aggregation of emissions from various waste types, energy generation processes, TSD, and fertiliser production. In future research, emissions may also be reported as kg CO_{2-eq.} per year, contingent on the generation of diverse biodegradable waste types, the utilisation of natural gas and hydrogen, and the application of mineral fertilisers. This approach offers an alternative perspective on the scale of emissions when considered on an annual basis.
- We excluded carbon capture, usage, and storage (CCUS) from biogas upgrading in this study to be consistent with the C1 scenarios and because it is not economically feasible⁷² Further research is needed to assess the scale-up feasibility,⁷³ despite its commercial availability.⁷⁴ It is important to note that CCUS can reduce GHG emissions in the biomethane supply chain, and this should be explored in future studies.
- In this study, we did not include 'gold hydrogen' production, also known as 'natural hydrogen' or 'white hydrogen.' Gold hydrogen, which is hydrogen that occurs naturally in the Earth's crust, is still in the early stages of development. We would like to highlight the possibility that the landscape of hydrogen production may change significantly in the future due to emerging technologies or unconventional sources of hydrogen. This shift could play a significant role in the transition to a low-carbon economy.

Despite the limitations associated with the counterfactual scenarios and data implementation, our objective was to

incorporate the most representative data available for a comprehensive LCA of the counterfactual and BAU cases. The MC simulation also took account of uncertainties arising from the data, and we are confident that the emissions ranges used accurately represent the counterfactual and BAU scenarios.

4. Conclusion

In this study, we assess 22 different LCA models of biogas and biomethane, including the injection of biomethane into the gas grid. We compare their GHG intensities with three main counterfactual scenarios to understand the GHG savings offered by biomethane. Our results demonstrate that biomethane generation offers greater GHG savings compared to biogas generation. Furthermore, the injection of biomethane into the gas grid yields GHG savings of 51-70% for natural gas (C1) and 42-65% for hydrogen (C2) generation at midstream stages. Notably, the GHG savings from low-carbon green hydrogen production counterfactual scenario (C3) at midstream stage provides lower average GHG intensities (13-24% lower), resulting in significantly greater GHG emissions savings compared to the BAU scenario.

While recent studies have indicated that CH₄ emission rates from biomethane supply chain routes could be greater than natural gas supply chains,6 when considering the credits given to the system (such as replacement of conventional fertiliser with digestate) and comparing it with other alternative biodegradable waste treatment methods, the biomethane utilisation remains a favourable option over natural gas even in scenarios with the highest CH4 emission levels. They are a clean and sustainable alternative to fossil fuels. In cases where CH4 emissions from AD plants reach the upper range, it may be advisable to consider alternative options such as energy generation from hydrogen and different waste treatment routes to reduce overall GHG emissions. These findings emphasise the importance of considering the entire life cycle of biogas and biomethane production and their associated emissions when evaluating their GWP impact. The results of this study contribute valuable insights into the GHG savings and environmental aspects associated with biomethane generation, supporting its role as a sustainable energy solution.

Conflicts of interest

The authors declare no conflict of interests.

Acknowledgements

The authors would like to acknowledge the Chemical Engineering Department at Imperial College London, United Kingdom. We extend our gratitude to Dr Jasmin Cooper for her valuable review of the methodology section.

References

- IPCC, Climate Change 2022: Mitigation of Climate Change. Contribution of Working Group III to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, ed. J. S. P. R. Shukla, R. Slade, A. Al Khourdajie, R. van Diemen, D. McCollum, M. Pathak, S. Some, P. Vyas, R. Fradera, M. Belkacemi, A. Hasija, G. Lisboa, S. Luz and J. Malley, 2022.
- 2 IPCC, AR6 Climate Change 2021, The Physical Science Basis, 2021.
- 3 IEA, Outlook for biogas and biomethane: Prospects for organic growth, 2020.
- 4 D. Nelissen, Availability and costs of liquefied bio-and synthetic methane, 2020, CE Delft: Netherlands.
- 5 EBA, EBA's Biomethane Fact sheet, 2013.
- 6 S. Bakkaloglu, J. Cooper and A. Hawkes, Methane emissions along biomethane and biogas supply chains are underestimated, *One Earth*, 2022, 5(6), 724–736.
- 7 D. P. Moore, et al., Underestimation of Sector-Wide Methane Emissions from United States Wastewater Treatment, Environ. Sci. Technol., 2023, 57(10), 4082–4090.
- 8 S. Bakkaloglu, *et al.*, Quantification of methane emissions from UK biogas plants, *Waste Manage.*, 2021, **124**, 82–93.
- 9 H. Haberl, *et al.*, Correcting a fundamental error in greenhouse gas accounting related to bioenergy, *Energy Policy*, 2012, 45, 18–23.
- 10 P. Adams, *et al.*, Understanding greenhouse gas balances of bioenergy systems, 2013.
- 11 S. Bakkaloglu, J. Cooper and A. Hawkes, Life cycle environmental impact assessment of methane emissions from the biowaste management strategy of the United Kingdom: Towards net zero emissions, J. Cleaner Prod., 2022, 376, 134229.
- 12 P. C. Slorach, *et al.*, Assessing the economic and environmental sustainability of household food waste management in the UK: Current situation and future scenarios, *Sci. Total Environ.*, 2020, **710**, 135580.
- 13 A. Welfle, *et al.*, Generating low-carbon heat from biomass: life cycle assessment of bioenergy scenarios, *J. Cleaner Prod.*, 2017, **149**, 448–460.
- 14 T. Reinelt, *et al.*, Field measurements of fugitive methane emissions from three Australian waste management and biogas facilities, *Waste Manage.*, 2022, **137**, 294–303.
- 15 D. Tonini, *et al.*, GHG emission factors for bioelectricity, biomethane, and bioethanol quantified for 24 biomass substrates with consequential life-cycle assessment, *Bioresour. Technol.*, 2016, **208**, 123–133.
- 16 D. Styles, et al., Climate mitigation efficacy of anaerobic digestion in a decarbonising economy, J. Cleaner Prod., 2022, 338, 130441.
- 17 D. Styles, E. M. Dominguez and D. Chadwick, Environmental balance of the UK biogas sector: An evaluation by consequential life cycle assessment, *Sci. Total Environ.*, 2016, 560, 241–253.
- 18 J. Speirs, et al., A greener gas grid: What are the options, Energy Policy, 2018, 118, 291–297.

- 19 S. J. Richards and J. Al Zaili, Contribution of encouraging the future use of biomethane to resolving sustainability and energy security challenges: The case of the UK, *Energy Sustainable Dev.*, 2020, 55, 48–55.
- 20 Thinkstep, GaBi Software-System and Database for Life Cycle Engineering, BOKU, Editor, 2021.
- 21 ISO 14040, Environmental Management, Life Cycle Assessment, Principles and Framework, 2006, Geneva: International Organization for Standardization.
- 22 ISO 14044, Environmental management, Life cycle assessment, Requirements and guidelines., in First edition, 2006, Geneva: International Organization for Standardization.
- 23 Ecoinvent, Ecoinvent Database v3.8. 2021: Zurich and Lausanne, Switzerland.
- 24 K. A. Thyø and H. Wenzel, Life cycle assessment of biogas from maize silage and from manure: for transport and for heat and power production under displacement of natural gas based heat works and marginal electricity in northern Germany, 2007.
- 25 R. Y. Rubinstein and D. P. Kroese, *Simulation and the Monte Carlo method*, John Wiley & Sons, vol. 707, 2011.
- 26 J. Penman, *et al.*, Good practice guidance and uncertainty management in national greenhouse gas inventories, 2000.
- 27 A. McMurray, T. Pearson and F. Casarim, Guidance on applying the Monte Carlo approach to uncertainty analyses in forestry and greenhouse gas accounting, Winrock International, Arlington, VA, USA, 2017:26.
- 28 ADBA, Anaerobic Digestion Policy Report, 2019, Anaerobic Digestion & Bioresources Association.
- 29 BSI, PAS 110:2014, Specification for whole digestate, separated liquor and separated fibre derived from the anaerobic digestion of source-segregated biodegradable materials, 2014.
- 30 WRAP, Field experiements for quality digestate and compost in agriculture, 2016.
- 31 E. Walling and C. Vaneeckhaute, Greenhouse gas emissions from inorganic and organic fertilizer production and use: A review of emission factors and their variability, *J. Environ. Manage.*, 2020, 276, 111211.
- 32 P. Balcombe, *et al.*, The natural gas supply chain: the importance of methane and carbon dioxide emissions, *ACS Sustainable Chem. Eng.*, 2017, 5(1), 3–20.
- 33 HM Goverment, UK Hydrogen Strategy, 2021.
- 34 National Grid, The hydrogen colour spectrum, 2023 [cited 2023 December 20]; available from: https://www.nationalgrid. com/stories/energy-explained/hydrogen-colour-spectrum.
- 35 DESNZ, UK Low Carbon Hydrogen Standard, 2023.
- 36 P. Balcombe, N. Brandon and A. Hawkes, Characterising the distribution of methane and carbon dioxide emissions from the natural gas supply chain, J. Cleaner Prod., 2018, 172, 2019–2032.
- 37 S. Kolb, *et al.*, Renewable hydrogen imports for the German energy transition–A comparative life cycle assessment, *J. Cleaner Prod.*, 2022, **373**, 133289.
- 38 E. Cetinkaya, I. Dincer and G. Naterer, Life cycle assessment of various hydrogen production methods, *Int. J. Hydrogen Energy*, 2012, 37(3), 2071–2080.

- 39 A. Ozawa, et al., Assessing uncertainties of Well-to-Tank greenhouse gas emissions from hydrogen supply chains, Sustainability, 2017, 9(7), 1101.
- 40 C. Bauer, et al., On the climate impacts of blue hydrogen production, Sustainable Energy Fuels, 2022, 6(1), 66-75.
- 41 O. Siddiqui and I. Dincer, A well to pump life cycle environimpact assessment of some production routes, Int. J. Hydrogen Energy, 2019, 44(12), 5773-5786.
- 42 Z. Li, et al., Coal-derived methanol for hydrogen vehicles in China: energy, environment, and economic analysis for distributed reforming, Chem. Eng. Res. Des., 2010, 88(1), 73 - 80.
- 43 A. Verma and A. Kumar, Life cycle assessment of hydrogen production from underground coal gasification, Appl. Energy, 2015, 147, 556-568.
- 44 A. Mehmeti, et al., Life cycle assessment and water footprint of hydrogen production methods: from conventional to emerging technologies, Environments, 2018, 5(2), 24.
- 45 J. Incer-Valverde, et al., "Colors" of hydrogen: Definitions and carbon intensity, Energy Convers. Manage., 2023, 291, 117294.
- 46 Y. K. Salkuyeh, B. A. Saville and H. L. MacLean, Technoeconomic analysis and life cycle assessment of hydrogen production from natural gas using current and emerging technologies, Int. J. Hydrogen Energy, 2017, 42(30), 18894-18909.
- 47 A. Oni, et al., Comparative assessment of blue hydrogen from steam methane reforming, autothermal reforming, and natural gas decomposition technologies for natural gas-producing regions, Energy Convers. Manage., 2022, 254, 115245.
- 48 M. Hermesmann and T. Müller, Green, turquoise, blue, or grey? Environmentally friendly hydrogen production in transforming energy systems, Prog. Energy Combust. Sci., 2022, 90, 100996.
- 49 A. Ozbilen, I. Dincer and M. A. Rosen, A comparative life cycle analysis of hydrogen production via thermochemical water splitting using a Cu-Cl cycle, Int. J. Hydrogen Energy, 2011, 36(17), 11321-11327.
- 50 A. E. Karaca, I. Dincer and J. Gu, Life cycle assessment based sustainable nuclear production options, Int. J. Hydrogen Energy, 2020, 45(41), 22148-22159.
- 51 Z. Khila, et al., Thermo-environmental life cycle assessment of hydrogen production by autothermal reforming of bioethanol, Energy Sustainable Dev., 2017, 37, 66-78.
- 52 M. R. Kabir and A. Kumar, Development of net energy ratio and emission factor for biohydrogen production pathways, Bioresour. Technol., 2011, 102(19), 8972-8985.
- 53 M. Al-Breiki and Y. Bicer, Comparative life cycle assessment of sustainable energy carriers including production, storage, overseas transport and utilization, J. Cleaner Prod., 2021, 279, 123481.

- 54 C. Wulf, et al., Life Cycle Assessment of hydrogen transport and distribution options, J. Cleaner Prod., 2018, 199, 431-443.
- 55 M. Skowroñska and T. Filipek, Life cycle assessment of fertilizers: a review, Int. Agrophys., 2014, 28(1), 101-110.
- 56 M. Elsayed, R. Matthews and N. Mortimer, Carbon and energy balances for a range of biofuels options, 2003
- 57 G. Kongshaug, Energy consumption and greenhouse gas emissions in fertilizer production, in IFA Tech. Conf., Marrakech, Morocco, 1998, 1998, Int. Fertilizer Industry Assoc.
- 58 J. Davis, Life cycle inventory (LCI) of fertiliser production: fertiliser products used in Sweden and Western Europe, 1999: SIK Institutet för livsmedel och bioteknik, Göteborg, Sverige.
- 59 K. J. Kramer, H. C. Moll and S. Nonhebel, Total greenhouse gas emissions related to the Dutch crop production system, Agric., Ecosyst. Environ., 1999, 72(1), 9-16.
- 60 A. G. Williams, E. Audsley and D. L. Sandars, Environmental burdens of producing bread wheat, oilseed rape and potatoes in England and Wales using simulation and system modelling, Int. J. Life Cycle Assess., 2010, 15(8), 855-868.
- 61 F. Brentrup, A. Hoxha and B. Christensen, Carbon footprint analysis of mineral fertilizer production in Europe and other world regions, in Conference Paper, The 10th International Conference on Life Cycle Assessment of Food (LCA Food 2016), 2016.
- 62 T. J. Albaugh, et al., Carbon emissions and sequestration from fertilization of pine in the southeastern United States, Forest Sci., 2012, 58(5), 419-429.
- 63 F. Zhang, et al., Life cycle assessment of diammonium-and monoammonium-phosphate fertilizer production in China, J. Cleaner Prod., 2017, 141, 1087-1094.
- 64 G. A. da Silva and L. A. Kulay, Environmental performance comparison of wet and thermal routes for phosphate fertilizer production using LCA-A Brazilian experience, J. Cleaner Prod., 2005, 13(13-14), 1321-1325.
- 65 W. Chen, et al., Life cycle assessment of potash fertilizer production in China, Resour., Conserv. Recycl., 2018, 138, 238-245.
- 66 M. Kratzeisen, et al., Applicability of biogas digestate as solid fuel, Fuel, 2010, 89(9), 2544-2548.
- 67 Y. Zhou, et al., Life-cycle greenhouse gas emissions of biomethane and hydrogen pathways in the European Union, 2021.
- 68 P. Balcombe, et al., Methane emissions: choosing the right climate metric and time horizon, Environ. Sci.: Processes Impacts, 2018, 20(10), 1323-1339.
- 69 D. Zavala-Araiza, et al., Super-emitters in natural gas infrastructure are caused by abnormal process conditions, Nat. Commun., 2017, 8(1), 1-10.
- 70 M. Omara, et al., Methane emissions from natural gas production sites in the United States: Data synthesis and national estimate, Environ. Sci. Technol., 2018, 52(21), 12915-12925.

- 71 A. R. Brandt, *et al.*, Methane leaks from North American natural gas systems, *Science*, 2014, **343**(6172), 733–735.
- 72 T. Johansson and M. Knutsson, Techno economic assessment of CCUS for a biogas facility in Sweden: Evaluating the economic feasibility for three CCUS concepts, 2022.
- 73 S. Fu, I. Angelidaki and Y. Zhang, In situ biogas upgrading by CO2-to-CH4 bioconversion, *Trends Biotechnol.*, 2021, **39**(4), 336–347.
- 74 E. Esposito, *et al.*, Simultaneous production of biomethane and food grade CO2 from biogas: An industrial case study, *Energy Environ. Sci.*, 2019, **12**(1), 281–289.