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Harnessing the full potential of biomethane towards tomorrow's bioeconomy: a national case study coupling sustainable agricultural intensification, emerging biogas technologies and energy system analysis

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Abstract

Here, we demonstrate the applicability of national strategies towards massive biogas deployment, through a case study Denmark. First, a variety of sustainable agricultural intensification measures to produce additional biomass resources were investigated; as a result, it was found that the biomass currently used in Denmark's biorefineries (including biogas) could be tripled without compromising soil carbon and inducing little to no land use changes. The degree to which these resources could be mobilised for the biogas sector was analysed through examining the extremes, here labelled as LOW and HIGH biomass-to-biogas scenarios. The resulting biomethane production was calculated considering three combinations of biogas production and upgrading technologies: (i) conventional biogas production and upgrading technologies; (ii) plants with prolonged retention time and conventional upgrading technologies and (iii) as in (ii), but upgrading via biological methanation of carbon dioxide in the biogas, using renewable hydrogen. These scenarios revealed a biomethane potential of 24 – 111 PJ y⁻¹. The key finding of our study is that only the extreme deployment measures, in terms of biomass and technology, allowed to fulfill the emerging gas demands, namely buffering the deficits from fluctuating power and transport (light- and heavy-duty vehicles, urban buses, coaches), quantified at 95 PJ y⁻¹. Yet, just harnessing the full sustainable potential of animal manure, straw and perennial grass allows to supply half of this demand. In the LOW and HIGH biomass scenarios, doubling the retention time brought an increased methane production of 20% (energy-wise), while this increase was 87% when methanation was added.

Keywords: bioeconomy; sustainable intensification; fluctuating power; transport; methanation; perennial grasses; straw; hydrogen

1 Introduction

Facing the urgency of avoiding dangerous climatic change [1–3], a number of countries have engaged in a pathway towards a so-called decarbonized economy [4]. A low carbon economy involves an increased reliance upon non-carbon energy sources, and thus renewable electricity (hydropower, photovoltaics, wind). Yet, although it is possible to decouple the energy sector (i.e. transport, heat and electricity) from the use of carbon, this does not apply for chemicals and materials, intrinsically based on carbon. Biomass, being the unique source of renewable carbon on Earth, is thus key to start decoupling the production of future materials and chemicals from the use of fossil carbon, besides being pinpointed as a stepping stone feedstock towards a renewable energy system [5–8].

Being a versatile and storable source of carbon, biogas, i.e. the methane (CH₄)-based gas mixture obtained from the anaerobic digestion of biomass, is seen to have a key role to play in bridging the gap towards a low carbon economy [9,10]. This is reflected, among others, by the various financial support systems established throughout Europe and worldwide for biogas deployment [11]. It is also acknowledged as one of the most cost- and environmentally-efficient mitigation technology for greenhouse gases (GHG) in agriculture, especially when it stems from residual resources like manure and organic wastes [12–14]. In some rural regions of Africa and Asia, biogas significantly contributes to improve human health as it replaces traditional open-fire stoves [15–17], which exposes ca. 40% of the

World population to illnesses attributed to soot particles and pollutants that penetrate the lungs [18]. Unlike other biomass conversion technologies, biogas almost completely preserves plant nutrients in the effluent (digestate)[13,19]. It further allows to return ca. 40% of the initial carbon (C) input back to the most carbon vulnerable soils [13]; such C input from organic material has been shown to be crucial in preventing the loss of native soil organic carbon for conventional staple crops such as wheat, when grown on vulnerable soils [20,21]. Because it can be produced close to the resource and delivered to the consumers via the gas grid, biogas links agricultural and urban areas. The gas grid in fact represents an enormous untapped storage capacity for renewable energy throughout Europe (totalling 2.2 million kilometres; [22]), as the overall gas storage capacity has been growing faster than gas consumption over the last decade [23].

Through this study, both the terms biogas and biomethane are used. The latter refers to biogas from which the carbon dioxide (CO₂) and other impurities (essentially water and hydrogen sulfide) have been removed in order to meet the quality of natural gas used in the gas grid; a process often referred to as biogas upgrading. The term biogas, on the other hand, refers to the raw gas mixture (essentially CH₄ and CO₂, along with some trace gases) obtained from anaerobic digestion. This biogas may or may not be upgraded, depending on its intended use. In this study, biogas is to be seen as an intermediate to produce biomethane.

In spite of the environmental and versatility benefits of biomethane, biogas remains rather unexploited, both in Europe [24,25], Asia [26,27], North America [28,29] as well as in the “developing world” [30]. There was, in 2018, ca. 18 GW of installed capacity in the World (of which 69% in Europe), compared to 2.5 GW installed in 2000 [31]. Despite annual increases averaging 15% between 2005 and 2012, the global average annual increases in installed capacity observed in the last five years was only around 5% [31], as elaborated in the Supporting Information (SI) material.

Whether public investments should be made today for further developing the biogas sector at a much faster rate calls for thoughtful planning. However, little studies have been done documenting how a massive biogas deployment could be implemented at a national scale, along with the biogas increases that could be expected as a result. An analysis of the 500 most cited studies published in English between 2000-2019 obtained with the keywords “biogas strategy” in Web of Science (details in the SI) revealed that the most acknowledged strategical research has focused on biogas technology and its process performance parameters (e.g. [32–34]), on the gas use and consequences of this use on overall sustainable development (e.g. [35–37]), as well as on feedstock type (e.g. [13]). For instance, [32] critically reviewed the mechanisms behind ammonia inhibition as well the possible control strategies to ensure stable biogas production conditions and performance. On the other hand, [33] focused on the pre-treatments needed to enhance biogas production and reviewed their performance according to the type of feedstock. Parawira [34] explored the possibility to produce additional biogas from a given biogas plant, focusing on the specific role of enzymes to release a maximum of fermentable compounds. Borjesson and Mattiasson [38] documented the energy and environmental efficiency of biogas used for transport compared to other biomass-based liquid fuels, while [13] studied the environmental consequences of a large biogas deployment, focusing essentially on the

substrates used for co-digestion with manure. Poeschl et al. [35] analyzed the energy efficiency of a variety of biogas systems including several substrates, mono- vs co-digestion, transport distances per substrate, small- vs large-scale plants, different uses of the biogas, and formulated boundaries outside which producing biogas yields a negative energy return. The work of [35] is certainly amongst the landmark in terms of biogas strategy, but again is restrained to the plant level.

To bridge this gap, we here analyze different strategies for massive biogas deployment at the national level by applying these strategies to a country where the biogas sector is already well developed, namely Denmark. The vision is to imagine how biogas production could be increased well-beyond what is currently done. As any strategic work, this study is oriented towards the future, here the medium-term one, namely 2035. This was chosen as a time point where biogas is envisioned to have a key role to play in meeting the World's climate [39] and overall sustainability [40] ambitions as a renewable hydrocarbon supplier, until the deployment of technologies with lower readiness level is possible (e.g. direct air capture; electrified heavy-duty transport).

Denmark is used as an example of a high agricultural density country that has massively invested in developing its biogas sector in the recent years. This is among others due to a public policy (the Energy Agreement; [41]) launched in 2012, where important feed-in subsidies for injection into the gas grid were introduced to encourage the construction of biogas plants and the upgrading of biogas to biomethane, as further detailed in [42]. In 2019, Denmark was reported to count with ca. 210 biogas plants [43], based on data for 2017/18. In 2016, it produced ca. 1.6 GJ biogas capita⁻¹ [44,45], slightly behind Europe's largest producer, Germany (4 GJ capita⁻¹; [31,46]). In parallel, Denmark is also one of the leading nations in terms of wind power capacity installed per inhabitant (0.91 kW capita⁻¹; [44,45]). Between 2014 and 2017, the number of biogas upgrading plants went from zero to 25 [47,48], making the gas supply more and more decentralized. In 2017, the produced biomethane corresponded to ca. 5% of the country gas consumption [47]; there were, further, 17 gas filling stations throughout the country [49], and 4 more were expected by the end of 2019.

The key questions investigated in this study are: under the premise that a country is very serious about deploying biogas (i) what are the production levels that could be reached; (ii) how and (iii) what does this production represent when contrasted to the current and future demands for the services provided by methane gas? The vision is to provide a framework to support future large investment decisions within the biogas sector, as well as to quantify the minimum and maximum amount of additional biogas that can result from these decisions. Three main pillars are analyzed: (i) the maximal feedstock that could be sustainably mobilized; (ii) the type of biogas technologies used and (iii) the demands for methane gas. The key novelty of our study lies from its analysis standpoint at the interface of these three pillars, combining agricultural sustainable intensification with emerging biomethane production technologies and energy system analysis.

2 Sustainable feedstock mobilization

2.1 Sustainable agricultural intensification

Sustainable agricultural intensification consists to integrate a variety of upstream agricultural and/or silvopastoral practices with the production of the services to be supplied by the biomass (e.g. biorefineries; food for humans, etc.). The vision is to maintain or increase biomass production while enhancing environmental outcomes. A variety of sustainable intensification systems have been elaborated and implemented globally [50], in the United States (Billion-Ton study; [51]) as well as in Denmark (+10 Million tonnes study; [52–54]).

The latter showed, through two sustainable intensification scenarios (labelled “bioenergy-optimised” and “environmentally-optimised”), how a variety of measures could increase Denmark’s 2009 net primary production (NPP) without (i) reducing food production; (ii) increasing the amount of land allocated to agriculture; and (iii) reducing the soil organic carbon content of arable lands, biodiversity, and aquatic environment quality. In the present study, the sustainable intensification measures and environmental constraints presented in the environmentally-optimized scenario of the +10 Million tonnes study [52–54] are considered. These are here updated for 2035.

The exact sustainable intensification measures considered for all biomass feedstock are presented in Table 1, and calculations details to estimate the available potential of each of these streams as feedstock for biogas are presented in the SI. In a nutshell, the amount of a resource available in the future is estimated from the known 2009 production, through the use of indexes for changes in arable land, crop yield and number of live animals, reflecting the development forecasted to happen independently of sustainable intensification. In the +10 Million tonnes study [49–51], these indexes were withdrawn from the study of Dalgaard et al. [55], for 2020. In the present study, the potentials are also estimated using the indexes presented in Dalgaard et al. [55], but here the indexes for 2035 are used. Out of the 2035 estimated potential, a utilisation rate is applied, in order to reflect the technical or practical constraints preventing the full stream to be accessible. This discounting technique is commonly applied in biomass assessment studies to derive so-called technical, economic, or sustainable biomass potentials [56,57]. The discounted potential is then boosted according to the specific sustainable intensification measure considered (e.g. selective breeding). Finally, the considered demand for other non-energy services (e.g. bedding) is subtracted, in order to only use the amount in excess of current utilisation. Similarly, the eventual environmental constraints considered are also subtracted (e.g. grain and oilseed straw; Table 1). The potential thereby calculated thus reflects the additional sustainable biomass available for the biogas sector. Equation 1 demonstrates an example of calculation for straw:

$$Pot_{2035, straw, ci} = \left((A_{2009, straw, ci} - A_{constrained, straw, ci}) \times Y_{2009, straw, ci} \times U_{straw} \times I_{AL} \times I_{CY} \times (1 + \Delta_b) \times (1 + \Delta_h) \times 10^{-3} \right) - D_{2035, straw, ci} \quad (1)$$

Where:

$Pot_{2035, straw, ci}$: Straw potential from sustainable intensification in 2035, for crop i (Tg DM y^{-1})
 $A_{2009, straw, ci}$: Area planted with crop ci in 2009 (ha), from which straw can be obtained

$A_{\text{constrained, straw, ci}}$: Area where the sustainable intensification measures should not be applied because of environmental constraints (ha)

$Y_{2009, \text{straw, ci}}$: Straw yield of crop ci in 2009 (Mg DM ha⁻¹)

U_{straw} : Utilisation rate (or technical potential) limit for straw (%) (here 87%; Table 1)

I_{AL} : Index for arable land in 2035, reflecting the change in available arable land between 2009 and 2035 (%). The value used herein is 0.880.

I_{CY} : Index for crop yield in 2035, reflecting the change in overall crop yield between 2009 and 2035 (%). The value used herein is 1.083.

Δ_b : Expected yield increase, selective breeding (%) (sustainable intensification measure 1)

Δ_h : Expected yield increase, improved harvesters (%) (sustainable intensification measure 2)

D_{2035} : Straw demand from crop i to supply selected non-energy services in 2035 (Tg DM y⁻¹)

It should be highlighted that the specific case of straw involves interactions with other measures, here the conversion of 157,000 ha and 78,000 ha of land used for cereals and rapeseed production (in 2009), respectively, towards perennial grass production (Table 1). This implies a decreased straw production from these areas in 2035, and was taken into account as detailed in the SI.

Table 1. Sustainable intensification measures considered to increase the overall biomass available for biogas use, compared to the amount of biomass available for bioenergy in 2009. Measures and constraints (second column) stem from the environmentally-optimized scenario of Denmark's "+10 M tonnes study" [52–54], while potentials are updated for 2035^a.

Measure, generic	Description of specific sustainable intensification measure and constraints considered, if any	Bioenergy use 2009	Potential 2035, streams suitable for biogas only ^b	Does potential 2035 embeds a maximal utilisation rate?
Increased recovery of grain and oilseed straw	<ul style="list-style-type: none"> • Measure 1: Conversion to cereal varieties producing 15% higher straw yield without impairing grain yield through selective breeding for varieties with thicker and stronger stems, see e.g. [58,59]. Result: +15% yield increase. • Measure 2: By changing the design of combine harvesters or by using whole-crop or stripper harvest, the amount of straw recovered from cereals & oilseed rape (including grain chaff and leaves) can be increased by 12 to 30 % [60]. An increased recovery of 15% is here considered. Result: +15% yield increase. • Constraint 1: No straw removal is considered for identified cereal areas with a critically low soil carbon content^c (63,000 ha in 2009), assumed to remain unchanged till 2035. This "no changes" assumption was also applied for the amount of straw used for bedding and feeding in 2009, assumed to remain unchanged in 2035. 	Oilseed and cereals straw used for energy or left unharvested: 1.47 Tg DM y ⁻¹	Total potential use of oilseed and cereals straw for biorefineries: 2.70 Tg DM y ^{-1 d}	Yes; 87% (reflecting the maximum harvestable, technology-wise)
Recovering grass seed straw	<ul style="list-style-type: none"> • Measure 1: Maximal harvest (and use) of herbaceous residue from the threshing of grasses intended for seed production (e.g. ryegrass, red fescue grass), here called grass seed straw. The amount of grass seed straw used for feeding in 2009 is assumed to remain unchanged in 2035. 	Bioenergy from grass seed straw: 0.15 Tg DM y ⁻¹	Use of grass seed straw for biorefinery: 0.394 Tg DM y ⁻¹	Yes; 80%

Shifting selected oilseed rape and cereals areas towards perennial grass production	<ul style="list-style-type: none"> Measure 1: Enhanced photosynthesis efficiency by growing productive grasslands, i.e. either C3 or C4 grasses at an average yield of 15 Mg DM ha⁻¹e,f, by converting: <ul style="list-style-type: none"> (a) 78,000 ha oilseed rape areas (2009)^g (b) 157,000 ha cereal areas (2009) with nitrate retention below 35 % 	Rapeseed oil to bio-refineries: 0.13 Tg DM y ⁻¹	Rapeseed areas (2009) converted to perennial grass: 1.03 Tg DM y ⁻¹ Cereal areas (2009) converted to perennial grasses: 2.07 Tg DM y ⁻¹	No
Harvesting biomass from wetlands	<ul style="list-style-type: none"> Measure 1: Harvesting 100,400 ha (2009) of permanent grass on wetlands (soils with poor drainage) Constraint 1: Without fertilization. Resulting yield (2035) taken at 3 Mg DM ha⁻¹ 	No harvest of permanent grass	Harvesting permanent grass on wetlands: 0.20 Tg DM y ⁻¹	Yes; 75%
Improved utilisation of animal manure ^h	<ul style="list-style-type: none"> Measure 1: A maximum of animal slurry (excreted in stables) is used for biogas production other than on-farm biogas Measure 2: As measure 1, but for solid manure Constraint 1: 5% of all manure will be used for on-farm biogas, thus not available for the biorefineries. 	Manure use for bio-refineries: 0.18 Tg DM y ⁻¹	Manure use for biorefineries: 2.24 Tg DM y ⁻¹	Yes; 90% for slurry and 50% for solid manure
Harvesting road verges and water weeds	<ul style="list-style-type: none"> Measure 1: harvesting cuttings from road verge areas (7,100 ha; 2 Mg DM ha⁻¹) Measure 2: harvesting water weeds along shores (0.7 – 1.2 Mg DM km⁻¹ stream) 	No harvest of road verges and water weeds	0.014 Tg DM y ⁻¹ from road verges 0.007 Tg DM y ⁻¹ from streams	No Yes; 60% for medium-size shore and 80% for large shore
Using and harvesting cover crops	<ul style="list-style-type: none"> Increase in area with cover crops (190,000 ha in 2009 vs 343,000 ha in 2035), and use of the cover crops as biomass feedstock. By harvesting the main crop earlier, a yield of 1.5 Mg DM ha⁻¹ is reached for cover crops. 	No harvest of cover crops	Harvest of 343,000 ha of cover crops with yield of 1.5 Mg DM ha ⁻¹ : 0.39 Tg DM y ⁻¹	Yes; 75%
Total (Tg DM y ⁻¹)		1.93 ⁱ	9.05	

^a See SI (sheet biomass) for details. Most quantified figures are retrieved and described from/in [61], an appendix of the +10M tonnes plan study; ^b Potential after considering: the changes that would have happened anyway (crop yield, animal production, arable land available), potential environmental constraints, utilisation rate, the existing non-energy demand for the feedstock and the effect of the sustainable intensification measure; ^c Areas with a Dexter index (ratio soil clay: soil organic C; [62]) above 10; ^d Accounts for the loss resulting from oilseed rape and cereal areas conversion presented in this Table; ^e This could trigger the need for an import of 1 Tg DM feed [54], see footnote g. However, if the potential of biorefineries is fully developed (e.g. fractionation of the grass biomass into liquid and fibrous parts), converting 10-15% of the grass in biorefineries would be enough, according to [52,53], to compensate for the reduced amount of cereals produced; ^f Plants use either C3 or C4 photosynthesis to fix C from the atmosphere [63,64]; under optimal conditions of temperature and light, C4 plants convert up to 30% more solar energy into chemical energy stored in the biomass [65]; ^g of which 41% formerly used for biodiesel; 59% formerly used as a protein source for animal feed ^hThe index considered for the number of animals in 2035 ($I_{NA, 2035}$) is 0.835, a mix effect of fewer animals and increased feed efficiency [55]; ⁱ To this, there were also 1.70 Tg DM y⁻¹ of woody biomass used for bioeconomy in 2009, namely 0.70 Tg DM y⁻¹ hedgerows, 0.96 Tg DM y⁻¹ energy wood from forestry and 0.04 Tg DM y⁻¹ from poplar. These are not represented herein as not foreseen suitable for biogas, although not impossible, as shown in e.g. [66].

2.2 Competing demands for biomass in the emerging bioeconomy: defining the extremes for the biogas sector

To the 9.05 Tg DM y⁻¹ potential for 2035 shown in Table 1, we added the technical potential of three feedstock that were not considered in [52–54], namely sewage sludge, industrial and household organic waste, as shown in Table 2.

These 2035 biomass potentials are considered to be available for all the new biomass demands emerging as the bioeconomy is developing. In other words, biogas is likely to be, towards 2035, in competition with other bioeconomy-driven demands for these additional biomass potentials. Therefore, it is considered unlikely that 100% of the 2035 potential reported in Table 2 will be fully accessible to, or prioritized for, the biogas sector. In order to reflect the full range of possibilities, two end-of-intervals scenarios, namely LOW and HIGH, have been built. Table 2 details the availability considered for each stream in these LOW and HIGH scenarios, along with the rationale behind it.

Table 2. Definition of the LOW and HIGH biomass scenarios for biogas production in 2035

Biomass	Potential 2035 (Tg DM y ⁻¹) ^a	% use in LOW biomass scenario (%)	% use in HIGH biomass scenario (%)	Rationale
Manure	2.24	50	100	Due to its high water content, and high content in macronutrients, it is quite obvious that manure fits very well to the biogas technology. Yet, manure from small or remote farms may be difficult/expensive to access, while acidified manure stemming from e.g. NH ₃ /CH ₄ mitigation technologies may be problematic to ensure a stable digestion process, and emerging competing technologies (e.g. hydrothermal liquefaction) may impair the access to manure.
Straw (grain, oilseed & grass seed)	3.10	20	60	Straw has so far been used mainly for combustion but there are both technical and environmental arguments for using it for biogas instead [67]. However, competing demands for straw (e.g. 2 nd generation bioethanol) emerge too [68,69]. The EU climate policy demands, for Denmark, a 39% decrease in GHG emissions by 2030, compared to the 2005 levels ([70]). This applies for all IPCC sector of activities, including agriculture. If straw is e.g. ploughed down compared to a situation where it was left on soil, the resulting reduction in GHG emission is accounted for in the agricultural sector. However, if straw is delivered for the energy sector (e.g. via biogas, for the natural gas grid) it is accounted for in the energy sector. This impairs the incentive for farmers to export their straw.
Green biomass ^c	3.71	20	60	We argue, based on e.g. [71,72] that this resource (except for cover crops) should primarily be used for green biorefineries to extract proteins for fodder, as well as high-value components when economically relevant. The fibre fraction left may be used as cattle feed, for paper-making, or can be utilized for biogas.
Organic waste ^d	0.45 ^{e,f}	50	100	Organic waste is also an obvious substrate for biogas, and in Denmark, legislation allows for its mixing with agricultural biomass. Yet, current waste incineration plants are dimensioned to include a certain share of organic waste, and the recovery of organics from household waste is still not 100% [73].
Sludge from wastewater, treated at wastewater	0.25 ^{f,g}	60	80	The sludge generated from the anaerobic and aerobic stabilization processes performed at wastewater treatment plants on wastewater collected in the sewage system is a resource for producing biogas. A utilisation

treatment plant			rate of 80% is applied on the total sludge generated from today's plants; the LOW and HIGH amounts are subsequently calculated from this. The LOW and HIGH scenario consider the Chemical Oxygen Demand (COD) partition in today's wastewater plants where anaerobic digestion is performed. In these, only 60% of the COD goes to biogas [74], as Danish wastewater plants are not optimized for biogas production but for the quality of the rejected water effluent. The HIGH scenario considers plants optimized for biogas production.
Total ^h (Tg DM y ⁻¹)	2.9	7.0	

^a Values are from Table 1, except otherwise specified; ^c Includes the perennial grasses grown on converted oilseed rape and cereal areas, cover crops, the permanent grass grown on wetlands, the cuttings from road verges and the collected water weeds; ^d Includes organic industrial and household waste. Details on the specific wastes considered are available in the SI (tab biomass); ^e Retrieved from [75]; ^f This technical potential incorporates a maximal utilization rate of 80%; ^g Based on [76]; ^h Calculation details in the SI, tab Case-Biogas.

3 Biogas (biomethane) conversion technologies

Three different biogas technology scenarios are considered. They all produce biomethane (upgraded biogas) from the LOW and HIGH quantities of the streams presented in Table 2. These are: (i) "State-of-the-art" biogas plant where gas is upgraded to biomethane by conventional upgrading technologies; (ii) As in (i), but with prolonged retention time and (iii) a plant with prolonged retention time and methanation (e.g. [77–80]), where the CO₂ in the gas is reduced with hydrogen (H₂) to generate additional CH₄ and produce upgraded biogas. This concept is also referred to as power-to-gas (P2G), when the H₂ is produced from e.g. water electrolysis via the input of electricity from fluctuating renewable resources (e.g. wind and sun).

In an endeavour to ensure the model tractability, the manure input is, for all scenarios, assumed to consist of 58% cattle manure and 42% pig manure, based on Denmark's national register of biomass used for biogas production (BIB) (see SI for additional details). This simplification thus excludes the 10% of other manure types used in Danish biogas plants (e.g. mink, poultry, deep litter, etc.), as further detailed in the SI. The biogas is assumed to be composed of 62% CH₄ and 38% CO₂ (trace gases, e.g. H₂S, NH₃, and H₂O are neglected) which is representative for manure-based biogas [13,81,82]. The upgraded gas is considered to meet the future quality and safety criteria reported by the Danish TSO (Transmission System Operator) for the gas grid: Wobbe index ranging between 14.1 – 15.5 kWh MJ⁻¹, relative density between 0.555 – 0.700 (dimensionless), and CO₂ content of the gas not exceeding 1.3 vol. %, among others [83]. The lower heating value (LHV) used for methane under normal conditions (0°C; 1 bar) is 35.2 MJ Nm⁻³ CH₄ [13].

Based on the technology data published by the Danish Energy Agency for future biogas plants [84], average overall fugitive losses of 2% of the gross CH₄ produced are considered (accounting all losses occurring up to, and including, grid injection). This may be seen as representing the best plants rather than an average plant. The International Energy Agency (IEA) [85] recently reported overall leaks varying from 0.2 to 13.7 % of the CH₄ produced for upgrading biogas plants (in Germany), while measurements at 10 Danish biogas plants (including upgrading plants) revealed losses varying from 0% to 10% (average 4.3%), which

were however reduced to 0.1% to 4.4% (average 0.8%) after minor repairs [86]. Yet, there are still huge uncertainties related to the measurements of these fugitive losses [85]. This is for example reflected by the extensive campaign on measuring the leakages from the Linköping biogas (upgrading) plant in Sweden [87] where total losses varying between 0.64% and 3% were measured for the same plant (the variation is only due to the team and method to perform the measurements, and to some extent the precise measurement time in the day).

3.1 State-of-the-art (SOTA) biomethane scenario

The state-of-the-art (SOTA) scenario is based on a centralized model plant representing an average Danish installation [88]; a plant processing ca. 200,000 Mg biomass per year, operating at thermophilic conditions (50-55°C), with a total hydraulic retention time (HRT) of ca. 30 days (details on operating conditions at agricultural Danish biogas plants are provided in the supplementary material).

The use of conventional upgrading technologies, i.e. those simply removing CO₂ from the gas (as well as other impurities, depending on the technology) is considered in this scenario, without distinguishing the specific technology being used. These technologies include physical and chemical absorption methods dissolving the unwanted gases in a liquid medium (water scrubbing, organic solvents scrubbing, amine solutions or aqueous alkaline salts), adsorption methods (adsorbent materials selectively retaining CO₂ and H₂S, among others, as compressed biogas at ca. 4-10 bars flows through it) and separation methods (membrane-based, cryogenic) [89]. According to Aryal and Kvist [48], all of the 25 upgrading biogas facilities in Denmark (2018) used these conventional technologies (9 used water scrubbers, 8 were amine-based and 8 membrane-based), though there are projects and demonstration units of biogas upgrading technologies based on hydrogen methanation, as described in [79] and below. In this study, the focus is on the quantification of the renewable methane produced from the available biomass (rather than environmental impacts or energy balance); the choice of the exact conventional upgrading technology therefore only matters to the extent that the biomethane production is affected. For the SOTA scenario, the only difference the type of upgrading technique could make, with regards to the present study, would come from the fugitive losses, since they influence the total amount of biomethane produced. These, however, were fixed independently of the specific technology used.

3.2 Longer retention time (scenario LRT)

This scenario is exactly as the SOTA scenario, but considers an overall solids retention time (SRT) of 60 days; this measure has the potential to increase the biochemical methane production (BMP) of ca. 10 - 40% compared to the SOTA scenario, depending on the feedstock (Table 3). As a result of a longer retention time, the amount of volatile solids (VS) that degrades to produce biogas is increased (Table 3).

3.3 Methanation with LRT (scenario MET+)

The methanation scenario (MET+) considers the same plant as in the LRT case, but without the conventional upgrading unit. Here, instead, hydrogen is injected to the produced biogas, where the CO₂ portion of the gas is converted into CH₄ following the Sabatier reaction,

either biologically or chemically with the use of catalysts [89]. In this reaction, four moles of H₂ are required per mole of CO₂ to produce one mole of CH₄. The Sabatier reaction is highly exothermic releasing 165 kJ per mole of CH₄ produced [78], and is carried out at a temperature between 250 – 550°C, typically in the presence of a nickel (Ni)-based catalyst [90–92].

In this scenario, the needed H₂ is produced through water electrolysis (e.g. alkaline technology), powered by excess electricity produced from fluctuating power, i.e. the solar or wind power produced in excess of conventional demand. The rationale for this is that the original driver for an increase in renewable gas is climate change mitigation. It is here considered that only 90% of the CO₂ in the biogas will be converted to CH₄, i.e. that 10% of the biogas CO₂ will fail to react with H₂.

3.4 Calculation of the biomethane potential

The amount of biomethane produced in each of the scenario was calculated according to Equation 2:

$$AMP_{s,b,int} = (P_s \times A_{int} \times VS \times BMP_{s,b} \times LHV_{CH_4}) - F_{b,s,int} \quad (2)$$

Where:

AMP_{s,b,int}: Amount of CH₄ produced annually by substrate s, with biogas technology b, at end-of-interval int (PJ y⁻¹)

P: Annual potential of the substrate (technically available estimate; Table 2) (Tg DM y⁻¹)

A_{int}: Availability of the substrate at the LOW or HIGH end-of-interval (Table 2) (%)

VS: Percentage of volatile solids out of total dry matter (Table 3) (%)

BMP: Biochemical methane potential (Table 3) (Nm³ CH₄ kg⁻¹ VS)

LHV_{CH₄}: Lower heating value of methane under normal conditions (MJ Nm⁻³ CH₄)

F: fugitive losses for biogas technology b, prior to injection, considering substrate s and end-of-interval int (PJ y⁻¹)

s: biomass substrate

b: biogas technology scenario

int: interval (represented by scenario LOW or HIGH).

Fugitive losses from the anaerobic digesters (prior to injection) are calculated similarly (Equation 3):

$$F_{b,s,int} = (P_s \times A_{int} \times VS \times BMP_{s,b} \times LHV_{CH_4} \times pct_{fb}) \quad (3)$$

Where:

pct_{fb}: Percentage of CH₄ losses from the digester, out of the gross CH₄ produced (%). This was here fixed to 2%.

The key data considered for all involved biomass streams are presented in Table 3. These are further detailed in the SI.

Table 3. Methane potential production per stream and scenario, key background data^a

Biomass	Biomass scenario, resource available based on Table 2 ($P_s \times A_{int}$) ($Tg DM y^{-1}$)		VS (% of DM) ^d	Biogas scenarios, Biochemical Methane Potential (BMP _{s,b}) ($Nm^3 CH_4 kg^{-1} VS$)		
	LOW	HIGH		SOTA ^d	LRT ^e	Met+ ^f
Manure						
Animal manure, mix ^b	1.12	2.24	80	0.25	0.30	0.47
Straw						
Straw, cereals & rape	0.54	1.62	90	0.22	0.30	0.47
Straw, grass seed	0.08	0.24				
Green biomass						
Grass, perennial ^c	0.62	1.86	95	0.30	0.33	0.51
Grass, from wetlands	0.04	0.12		0.25	0.31	0.48
Grass, from road verges	2.8×10^{-3}	8.5×10^{-3}		0.25	0.31	0.48
Water weed cuttings	1.4×10^{-3}	4.2×10^{-3}		0.25	0.31	0.48
Cover crops	0.08	0.23		0.28	0.33	0.51
Organic waste						
Fatty wastes from oil separation	1.0×10^{-3}	2.0×10^{-3}	90	0.70	0.80	1.24
Consumable oil and fatty wastes	4.3×10^{-4}	8.6×10^{-4}		0.80	0.90	1.40
Household biowaste, source-separated	0.09	0.18		0.40	0.50	0.78
Household biowaste, non-separated	0.12	0.25		0.35	0.40	0.62
Household biowaste, unclassified	7.7×10^{-3}	1.5×10^{-2}		0.40	0.50	0.78
Biowaste, from institutions	3.5×10^{-3}	7.0×10^{-3}		0.40	0.50	0.78
Wastewater sludge						
Wastewater sludge	0.15	0.20	80	0.20	0.25	0.39

^a Values presented with a maximum of 3 significant digits, inconsistencies due to rounding; ^b Taken as a mix of pig (42% share; VS/TS: 80%; BMP: $0.31 Nm^3 CH_4 kg^{-1} VS$) and cattle manure (58% share; VS/TS: 80%; BMP: $0.20 Nm^3 CH_4 kg^{-1} VS$), see text; ^c Stemming from both former cereals and rapeseed areas; ^d Own unpublished data and data from [10,93–96]; ^e Defined according to the data presented in [88]; ^f Stems from the fixed CO₂ content of the biogas (38%), and the assumption that only 90% of this CO₂ is converted to CH₄, see text and SI for details.

For a given biomass category (e.g. organic waste), the AMP_{s,b,int} can be summed, as shown in Equation 4:

$$AMP_{cat,b,int} = \sum_{s=1}^{s=n} AMP_{s,b,int} \quad (4)$$

Where:

AMP_{cat,b,int}: Annual methane produced for category cat, biogas technology b and end-of-interval int (PJ y⁻¹)

n: number of substrate involved in category cat

cat: category of substrates

4 Quantification of the demand for selected biogas roles in 2035

4.1 Selected biogas roles

The importance of the boosted biogas production for the case study is analysed in comparison to the demand for methane gas in the analysed country. This study focuses on two selected sectors of activity where biogas complements a growing deployment towards carbon-free renewable energy technologies. These are the transport sector, as well as the electricity production sector.

Electricity is considered a key sector to the extent biogas can buffer the fluctuating power production. The key approach of today's traditional electricity system consists to constantly adjust the supply in order to match the electricity demand at any time. This allows maintaining a constant frequency on the grid, which is required to ensure the stability of the electricity network and prevent damages to equipment. Although an increasing share of fluctuating power in the electricity mix involves periods of excess electricity, it also involves deficit periods where another energy source must immediately supply the demand. Increasingly, intermittency will be addressed through improved storage technologies and regional integration of renewable energy sources. For the near term, however, biomethane is an obvious complementary energy source to buffer fluctuating power that could assist the expansion and adoption of renewable energy. This is because biomethane can be stored in the natural gas grid and fed to (peak load) gas turbines that are easily and quickly switched on and off in order to balance fluctuations. In this way, the renewable biomass (but not unlimited in supply) is used efficiently by not competing with the wind and sun resources, when they are available.

Domestic transportation (total of 212 PJ y⁻¹ in 2013) includes road (158 PJ y⁻¹), domestic navigation (5 PJ y⁻¹), railway (3 PJ y⁻¹), domestic aviation (4.0 PJ y⁻¹) and other transport (manufacturing & construction industries, off-road agriculture, national fishing, household gardening; etc.) [97]. However, only road transport is considered herein. Although small airplanes running on compressed natural gas (CNG) do exist, and though research efforts are on-going towards the use of liquefied natural gas (LNG) for commercial airplanes, it will likely take decades to be implemented in commercial airplanes. As highlighted by [98], biofuels with properties closer to those of current jet fuels are more likely to be prominent in the transition towards low fossil carbon than other fuels. Railway, on the other hand, is excluded given Denmark's national strategy and plan to shift towards electrified railway (currently mostly based on diesel). Domestic navigation constitutes about 3% of the total energy consumption from the domestic transport sector. Distances between used ferry ports are relatively short. Smaller ferries commuting short distances have the potential of being electrified, as already seen by on-going pilot projects [99]. The vision is that the batteries of the ferry are charged during port time as part of the time table. Domestic navigation was thus excluded, although it is acknowledged that one project is currently on-going (Danish island of Samsø) to have a domestic ferry running on liquefied biomethane (LNG) [100].

For both electricity and transport, the biomethane used is the one supplied directly from the gas grid. As the pipelines in Denmark are relatively new and made of plastic, most fugitive losses post-injection are due to leaks occurring during construction and maintenance events.

The losses occurring in the last 10 years have tended to be around $100 \text{ Mg CH}_4 \text{ y}^{-1}$, which corresponds to ca. 0.005 PJ [101]. Using this figure, coupled with the natural gas consumed in Denmark in 2015 [102], a fractional loss rate of 0.01% for the gas distribution is here taken into account (SI).

4.2 Buffering fluctuating power

The share of fluctuating power (wind and sun) in the Danish electricity mix already represents more than 45% of the domestic electricity supply [102] (further detailed in the SI). This is illustrated in Figure 1, where the electricity demand (classic consumption; i.e. not including fluctuating consumption such as electric cars, heat pumps and electric boilers) is plotted against the fluctuating power production, both for 2017 (Figure 1a) and forecasted for 2035 (Figure 1b). The 2017 fluctuating power production and classic consumption were retrieved directly from the Danish TSO open data portal (see SI). The 2035 forecast was made considering two key hypothesis. The first is that the hourly classic electricity consumption curve of 2017 can be considered representative of 2035, given that only minor increases in electricity consumption are forecasted for 2035 (33.2 TWh in 2017 against 33.7 TWh in 2035; [103]). The second considers that the hours with a fluctuating power production under 500 MWh h^{-1} in 2017 will remain at the same level in 2035 (in other words, considering that the periods of the year with little wind or sun are not affected by the installed capacity). The fluctuating power production profile for the remaining hours was linearly scaled considering that the $6,092 \text{ MW}$ of fluctuating power installed in 2017 will reach the forecasted capacity of $13,409 \text{ MW}$ installed in 2035 [103]. This means that a scaling factor of 2.2 was applied to the 2017 production except when the production was under 500 MWh h^{-1} in 2017 (details in the SI). The deficits, i.e. events where the electricity demand is greater than the production, obtained at each hours of 2035 were then summed to get the total demand for buffering fluctuating power.

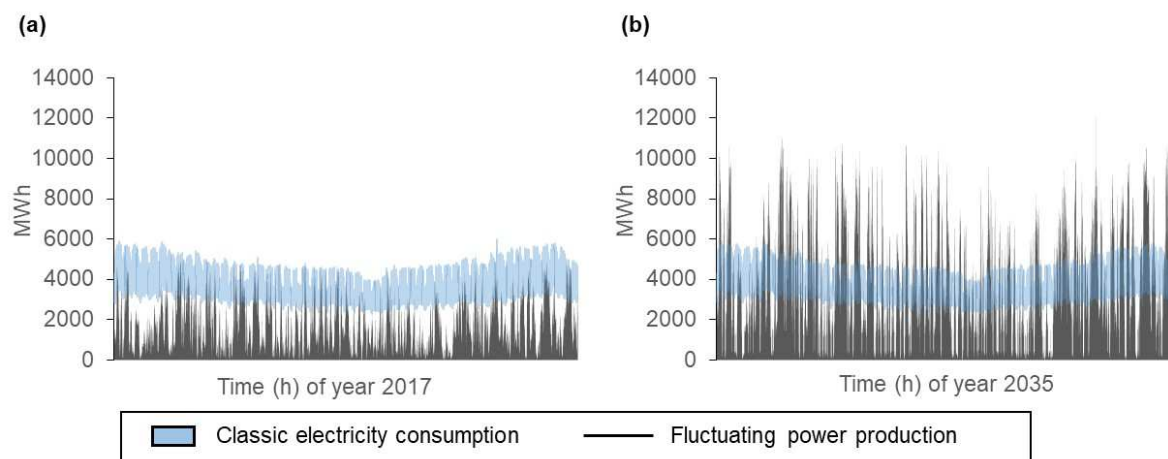


Figure 1. Deficits of electricity that could be supplied by biomethane in (a) 2017 and (b) 2035 (white gaps under the blue curve). Plotted for the 8,760 hours of the year.

4.3 Road transportation

The demands for biogas in various road transportation types in 2035 are presented in Table 4, along with the key rationale for their quantification. Assumptions on the forecasts to 2035 are based on the latest national fuel and emission forecasting [97,101] produced by the institution in charge of Denmark's reporting obligations to the UNFCCC. These studies use statistical data on the Danish vehicle fleet and annual mileage as well as fuel consumption factors provided by the European emission model COPERT 4 [104], among others.

Table 4. Demand for road transportation in 2013, forecasts to 2035, and quantification of the biomethane demand towards 2035 (as final and primary energy demands)

Road transportation type	Final energy demands (PJ y ⁻¹)			Primary energy demand 2035 for biomethane ^d (PJ y ⁻¹)	Rationale
	Demand (all fuels) in 2013 ^a	Demand (all fuels) in 2035 ^a	Demand (biomethane) in 2035		
Passenger cars	95.3	76.8	0	0	Based on the advanced current development of batteries and electric vehicles (some with an autonomy over 400 km), and on the vision that electricity will be the obvious first choice of fuel in the transition towards a decarbonized economy, biomethane is seen to have a very minor role for passenger cars transportation.
Light-duty vehicles	22.4	21.9	11.0	12.0	Based on a recent study [105], it is considered that half the demand from this category could be electrified, leaving the other half for biomethane.
Heavy-duty vehicles	31.8	38.2	38.2	42.0	Based on the long driving distances, heavy-duty vehicles are considered unlikely to electrify, at least before 2035. This demand is here considered fully covered by biomethane.
Buses, urban ^b	5.27	4.73	1.58	1.74	City buses (66% of consumption) may increasingly be battery electric vehicles, charging at terminals or major bus stops (opportunity charging) as part of the timetable. Intercity buses (33% of consumption) are more likely to run on (biomethane-based) LNG and CNG. We here considered a biogas demand for these only (assuming biomethane powers all intercity buses).
Buses, coaches ^c	2.63	2.37	2.37	2.60	The rationale for coaches is as for intercity buses (biomethane covering 100% of the demand)
Mopeds	0.2	0.2	0	0	The same rationale as for passenger cars is considered, where biomethane is seen to have a rather limited role
Motorcycles	0.6	0.6	0	0	The same rationale as for passenger cars is considered, where biomethane is seen to have a rather limited role
Total (PJ y⁻¹)	158	145	53.1	58.4	

^a From [97] and [101] ^b Includes city and intercity buses, ^c Tourist buses, sport club buses, etc., ^d A 10 % increase in fuel consumption is assumed for biomethane-fuelled vehicles compared to diesel-fuelled vehicles [106]

The biomethane demands in Table 4 include a 10% increase in fuel consumption reflecting the greater consumption of gas-fuelled engines compared to diesel-fuelled engines, based on a study from the International Council on Clean Transportation (ICCT) [106]. In addition, ICCT suggests emission factors to reflect CH₄ leaks from natural gas fueling stations. These may occur from valves, pipes and fittings at the tanking facilities and small escapes may occur during nozzle connection and disconnection when tanking the vehicles. Losses may also occur from the compressor at the station (with compressed natural gas; CNG) or as manually vented losses from the vehicle fuel tank prior to refueling (with liquefied natural gas; LNG). To reflect these losses, a fractional loss rate of 0.3% is considered, based on [106].

4.4 Calculating biomethane demands

The demands for biomethane, for each of the selected services, was calculated according to Equation 5:

$$ABD_{se} = D_{se} - L_{se} \quad (5)$$

Where:

ABD_{se}: Annual biomethane demand for service *se* in 2035 (PJ y⁻¹)

D_{se}: Final demand in biomethane for service *se* in 2035 (PJ y⁻¹)

L_{se}: Total CH₄ losses, post injection, for supplying service *se* (PJ y⁻¹)

se: service considered.

The values considered herein for D_{se} and L_{se} are summarized in Table 5.

Table 5. Final demand in biomethane and total losses for the services considered in this study

Service (<i>se</i>)	Biomethane demand for service (D _{se}) (PJ y ⁻¹)	Losses post-injection (L _{se}) (PJ y ⁻¹)		
		At gas grid distribution ^a	At fuel station ^b	Total
Buses (all)	4.34	4.34 x 10 ⁻⁴	1.30 x 10 ⁻²	1.35 x 10 ⁻²
Light-duty vehicles	12.0	1.20 x 10 ⁻³	3.61 x 10 ⁻²	3.73 x 10 ⁻²
Heavy-duty vehicles	42.0	4.20 x 10 ⁻³	1.26 x 10 ⁻¹	1.30 x 10 ⁻¹
Buffering fluctuating power	35.9	3.59 x 10 ⁻³	-	3.59 x 10 ⁻³

^a Taken as 0.01% of the energy content in the fuel delivered, see text; ^b Taken as 0.3% of the energy content in the fuel delivered (transport services only), see text

5 Results

The supply of biogas towards 2035 could, based on our methodology, vary between 23 and 105 PJ y⁻¹, with a range of [23-42 PJ y⁻¹] for the LOW biomass scenario and [56-105 PJ y⁻¹] for the HIGH biomass scenario (Figure 2). These are calculated according to Equations 2-4, as further detailed in the SI.

For all 6 scenarios, more than 80% of this potential is ensured by three major resources: animal manure, straw and perennial grass (grown on converted cereal and rapeseed areas). Moving from SOTA to a LRT biogas production (doubling the retention time) brings an increased methane production of 20% (energy-wise), while this increase is 87% if methanation is added to the LRT biogas production (Met+ scenario), i.e. nearly doubling the potential (Figure 2).

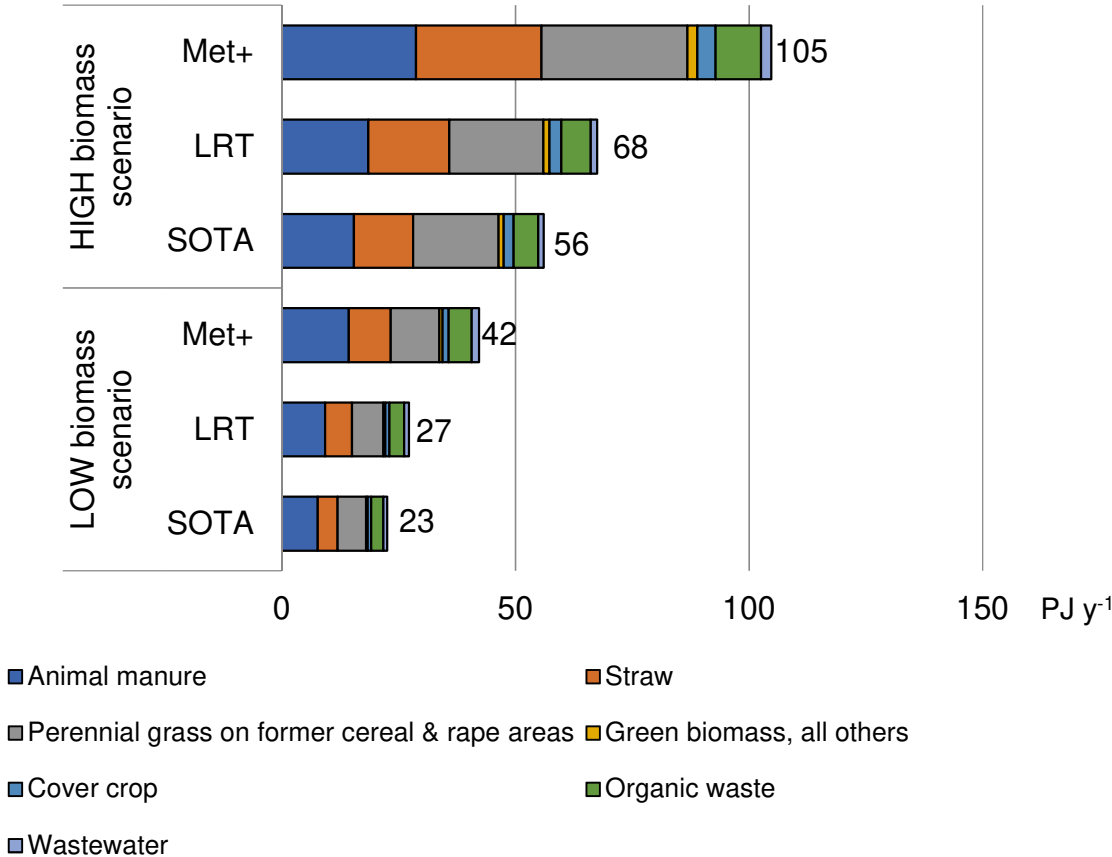


Figure 2. Net biomethane supply (as injected in the gas grid, after subtracting the fugitive losses occurring prior to injection) in 2035 resulting from all scenarios, breakdown per biomass resource. The category green biomass (Tables 2-3) is here broken down into three, namely: “perennial grass on former cereal and rape areas”, “cover crop”, & “green biomass, all others”.

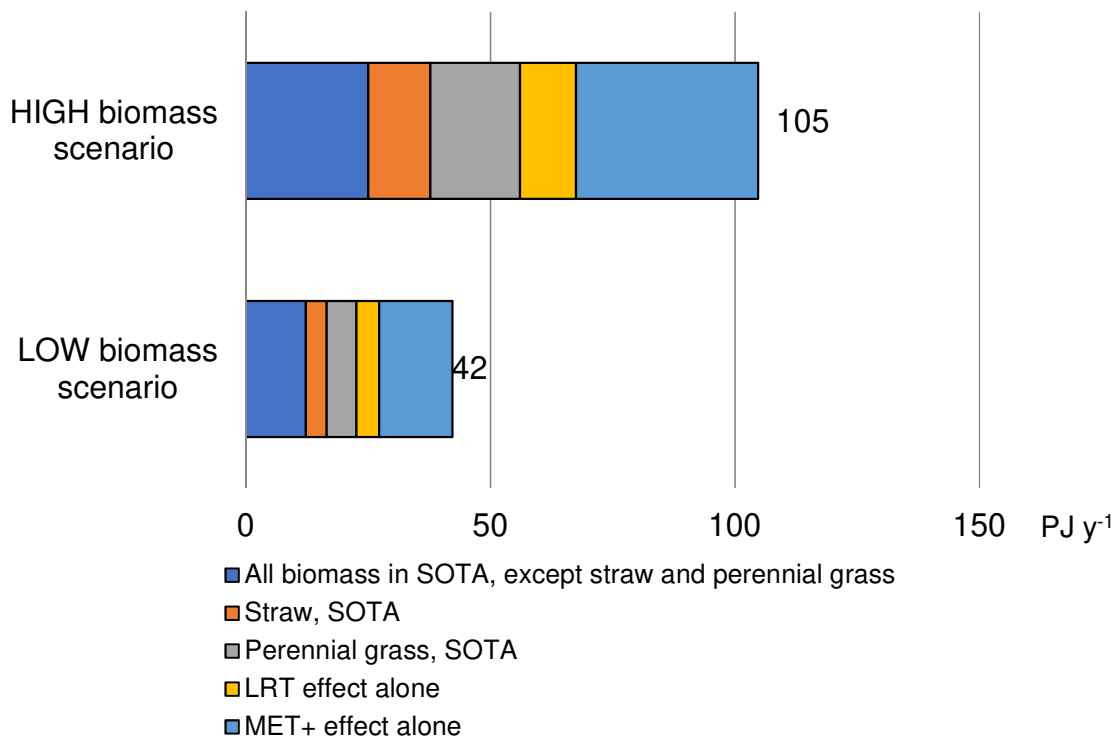


Figure 3. Highlighting the importance of straw, perennial grass, longer retention time and methanation for supplying biomethane in 2035 resulting from all scenarios.

The demand of biogas, in terms of primary energy demand, from the selected key sectors in 2035 varies from 4 PJ y⁻¹ (urban buses) to 42 PJ y⁻¹ (heavy-duty vehicles), as shown in Figure 4. These demands consider the fugitive losses occurring at the fueling station as well as losses from the distribution pipes (Table 5) and were calculated according to Equation 5.

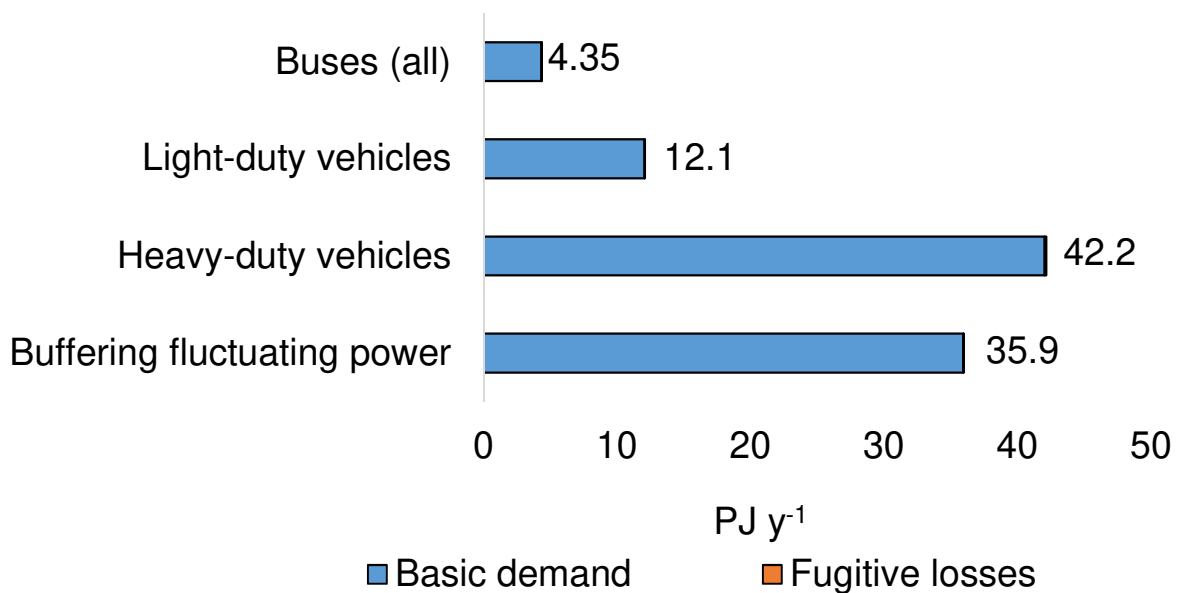


Figure 4. Overview of demands for biomethane, including fugitive CH₄ losses (minor hence barely visible)

6 Discussion

6.1 Only one scenario allows to meet all gas demands

Results from Figure 2 show that under a LOW biomass-to-biogas scenario (i.e. restricted access to biomass resources for biogas production) and a business as usual development of the biogas sector (SOTA scenario), there would be enough biomethane supply (i.e. 23 PJ) to fulfill both the gas demand from buses (intercity and coaches; Table 4) and from light-duty vehicles (here identified as half the total demand from that sector; Table 4), with a slight surplus (Figure 4). Under a LOW biomass-to-biogas scenario, biogas methanation alone (i.e. if the whole 2.9 Tg biomass y⁻¹ of Table 2 was digested in biogas facilities equipped with methanation) would produce 42 PJ y⁻¹, just enough to supply the demand from heavy-duty vehicles (Figure 4). If, however, biomass is totally prioritized for biogas (HIGH biomass-to-biogas scenario), and accordingly 100% of the technically available manure, 60% of the straw, 60% of the green biomass and 80% of the wastewater sludge is used to this end (Table 2), a minimum of 56 PJ biomethane could be supplied (SOTA scenario). This would be enough to cover the light- and heavy-duty transport demands (Figure 4). On the other extreme, if methanation technologies are considered along with a retention time twice as long in digesters, up to 105 PJ y⁻¹ could be produced, which would be enough to cover all demands combined (95 PJ). This highlights that biogas can play a major role in supplying a renewable gas source for services that cannot be immediately electrified, but that this depends on the extent at which resources are made available, and technology deployed.

It should nevertheless be highlighted that the biogas potentials found herein are high compared to existing estimates. For example, the Danish Energy Agency estimated in 2014 [107] a biogas potential of 42 PJ (for 2050; SOTA technologies), that could be boosted to 65 PJ via methanation. The lower estimate from [107] corresponds, in our study, to the production obtained from a low biomass prioritization for biogas, while in [107] it translated constraints (economic, infrastructure) regarding the production of hydrogen. Such constraints were not considered herein, as the vision was to reflect the full environmentally sustainable potential.

6.2 Biomass and land prioritization are key to a massive biogas deployment

Figure 3 highlights the importance of ensuring, on top of animal manure, the contribution of straw and perennial grass resources (stemming from the conversion of cereals and rapeseed areas; Table 1) for deploying the full biogas potential. These two resources here represent between 46 and 55% of the overall potential in terms of PJ CH₄ produced. Of course, the use of perennial grass implies the conversion of 157,000 ha of cereals (from nitrate-sensitive areas; 2009) and 78,000 ha of rapeseed (41% formerly used for biodiesel; 59% formerly used as a protein source for animal feed) in order to grow it (Table 1). Under the conventional (fossil-based) economy, the increased demand for this rapeseed and cereals no longer supplied by Denmark would likely trigger an increase in the price for these crops, which in turn is translated by two main reactions: cropland intensification and expansion of nature to

agriculture [108,109]. These land use change reactions were shown to decrease the environmental performance of biogas [13,109] or bioenergy in general [110]. To account for this, Schmidt and Munos [111] proposed an emission factor of $1.7 \text{ t CO}_{2e} \text{ ha}^{-1} \text{ y}^{-1}$ of “global” arable land demanded, and Tonini et al. [109] of $4.1 \text{ t CO}_{2e} \text{ ha}^{-1}$ arable land demanded y^{-1} . However, in the perspective of a high bioeconomy future with developed biorefineries, the grass could be first fractionated into a liquid and a fibrous part; only the latter would contain a significant amount of carbon and end up into biogas plants. The liquid portion would be mostly nitrogen-based, and used to produce a protein concentrate substituting soya and rapeseed cake, among others [71]. In other words, on top of using the liquid portion to produce a grass-based protein concentrate, the fibrous part, with ca. 10% higher energy-corrected milk yield per kg DM than fresh grass [112], is also used for feed. The vision is that it can substitute maize silage, and this with much higher yields than any grain crops [113]. In this “high bioeconomy” future (itself a pre-condition to obtain the biomass potentials shown in Table 1), the hectares of rapeseed and cereals displaced by perennial grass would thus not generate any land use changes, and would even prevent it to happen elsewhere by (over-) supplying high quality protein and fibres for ruminants and monogastrics. Moreover, it should be highlighted that we here only include 20 – 60% of the grass produced on these converted hectares for biogas (LOW-HIGH ranges of Table 2), meaning that there is 40 – 80% left that can be fully dedicated to increasing feed production.

Similarly, Figure 3 highlights that in order to reach 105 PJ y^{-1} of biogas, an increase in overall harvestable straw of ca. 100% (i.e. doubling) compared to the level of 2009 (Table 1) is required, of which 20%-60% is to be used for biogas production (LOW and HIGH scenario, respectively; Table 2). As mentioned in Table 2, straw is increasingly demanded as a feedstock for other biomass conversion technologies (e.g. to produce ligno-cellulosic bioethanol, as a bio-based construction material, as a feedstock for energy, etc.) [63,65,106,111–115]. Whether the technical straw potential (from cereals, oilseed and grass seed) can really be doubled and whether it is achievable to get 20-60% of this straw for biogas production is thus critically important to get to 105 PJ y^{-1} . Moreover, the use of straw as a feedstock for bio-based products and services raises the issue of preserving soil carbon stocks. In fact, soil carbon is acknowledged as a main contributor to healthy ecosystems, food- & water security [119–124], and straw is a key input of carbon to soils [20,125–128]. Here, this was taken into account by excluding the straw from carbon vulnerable areas (Dexter index [62] above 10; Table 1) from the technical potential. Compared to competing straw conversion technologies, biogas conversion presents the advantage that only part of the carbon (the easily degradable one) ends up in the gas; the remaining recalcitrant carbon stays in the digestate [13,19], and can thus be returned to soils while the digestate is used as an organic fertilizer. Through simple mechanical separation technologies, the carbon from this digestate could even be separated in an easily transportable solid fraction, which could be re-distributed to the vulnerable areas where this carbon is most needed. In subsequent studies, a spatial analysis could investigate these concerns and determine to which extent (and where) straw can be removed from agricultural fields without impacting soil carbon (with and without potential redistribution).

In each scenarios animal manure represents about a third of the potential, in terms of overall biomethane energy supply (Figure 2). In Table 2, it was considered that between 50% and 100% of the technically available manure will be used for biogas. As mentioned in Table

2, one concern often pointed out against the feasibility of a 100% manure utilisation for biogas is the increasing use of in-house acidification technologies to reduce ammonia (and methane) emissions from manure. These technologies typically rely on sulfuric acid to reduce manure pH below 6, yet sulphur inhibits, to some extent, the production of methane. According to the projections of [101], 18% of the dairy and heifer production will use this technology by 2030, while this is 15% for pig production. Yet, Moset et al. [129] showed that up to 10% acidified slurry (as a percentage of the total slurry input) can be added to digesters without inhibition. Similarly, Sutaryo et al. [130,131] showed that separating the acidified slurry in a solid and liquid fraction, and using only the solid fraction for anaerobic digestion allows to avoid sulphide inhibition from acidified slurry (the limit of separated solids input being fixed by operational constraints, e.g. respecting a certain dry matter input limit for easily pumping the digestate out of the digesters [19]). In conclusion, a potential close to 100% of the technically available manure does not appear utterly unrealistic.

Organic waste, on the other hand, is much less visible in Figure 2, representing about a tenth of the potential. According to [75], an additional 90,000 t DM from household source-separated biowaste along with an additional 18,750 t DM of biowaste from institutions is likely to be available in Denmark, in the medium-term future (i.e. on top of the amount presented in Table 2). In terms of energy unit (PJ of biomethane produced per year), however, this would only contribute with 1-2 PJ extra, depending on the scenario (LOW or HIGH biomass, respectively). Removing the 10 PJ y^{-1} supplied from organic waste in the HIGH biomass scenario (Figure 3) still allows to meet all the gas demands considered in Figure 4 (95 PJ y^{-1}). This tend to indicate that, although not negligible, this potentially hazardous and more challenging stream is not, to the extent animal manure is highly available, critical for the deployment of biogas, and could instead be prioritized for the circular economy [132,133], with biogas as a second priority.

Similarly, it may be argued that value chains based on grass from road verges and wetlands as feedstock for biogas production are less realistic, being expensive (for example, [75] evaluates it between 0.35 and 0.59 Euros per Nm^3 CH_4 produced with SOTA technologies, for a transport distance of 10 km; twice to three times as much as for pig and cow slurry). Here, these feedstocks are shown to be rather marginal in terms of biogas production, representing at maximum 2.1 PJ y^{-1} (Figure 2).

Beyond showing the importance of using manure, straw and perennial grass, Figure 3 also highlights the importance of fully deploying biogas technologies. The MET+ technology combination itself represents over 36% of the potential biogas supply.

6.3 Comparison of the biomethane production with the gas demand

Although the yearly supply of 105 PJ of biomethane is enough to supply all demands quantified in this study on a yearly basis, it must be ensured that these demands can also be met at all hours of the year. Through the 8,760 hours of the year, the demand for buffering fluctuating power (i.e. the difference, at a given hour, between the electricity demand and the supply of fluctuating power) reaches at maximum 0.020 PJ h^{-1} (SI). To this peak electricity demand (to buffer fluctuating power), the other demands must be added. Roughly assuming the gas demand for transport fuels to be equally distributed through the hours of

the year, a total of 0.007 PJ h^{-1} are required at all hours, based on the data from Table 4. Considering a constant biomethane production through the 8,760 hours of the years (i.e. continuous biogas production, with subsequent upgrading), a maximal hourly gas supply of 0.013 PJ h^{-1} is available. Based on these figures (a peaking demand twice as much as the supply), which do not even consider the efficiency of the gas turbine (electricity production case), meeting the peaking demands appear tight, and whether it is possible or not will depend on the amount of biomethane stored (from the hours with lower needs). Such dynamic analysis was beyond the scope of the present study, but issues regarding these peaking demands should be examined in detail through a dynamic and supply risk assessment study.

The use of methane gas for the transport sector, and for buffering the fluctuating power, can be considered as new demands, i.e. on top of current demands for natural gas. In 2015, the gross natural gas consumption in Denmark was 133 PJ y^{-1} [102]; the maximum biomethane supply found in this study represents between 32% and 79% of that (LOW and HIGH biomass scenario, respectively). Yet, whether the vision is to use biomethane to supply new demands (as investigated in this study) or to first supply the current ones, our results highlight the importance of scale and investment.

Considering the major impact methane has on global warming (global warming potential 28-34 times greater than CO_2 , on a 100-y basis; [134]), fugitive losses from biomethane are a rather important concern if biogas is to play a big role in the path towards low fossil carbon. In this perspective, the overall losses considered herein may be seen as “optimistic”. For example, distribution losses of 0.01% of the fuel delivered were considered to reflect the fact that the Danish gas grid consists of plastic pipes that are rather new. In comparison, McKain et al. [135] found losses of 2.7% for the (cast iron and bare steel) distribution pipes of the Boston urban area. In [136], the losses from the Russian natural gas long distance network were found to be around 0.6% of the gas delivered.

This study only focused on specific roles for biogas and biomethane. There are other ways to use biogas, for example as a feedstock for H_2 production for e.g. the fertilizer industry (as for current natural gas cracking), as a carbon feedstock for green chemistry, to produce industrial process heat, etc. There are also some concomitant roles; biogas is produced along with a nutrient-rich digestate where all nutrients and non-degraded carbon remains. This digestate is a valuable source of organic fertilizer, and even enhanced in comparison to non-digested organic fertilizers because the nitrogen is then under the ammonium (NH_4^+) form and thus readily available for plant uptake [13]. The latter aspect of nutrients & partial carbon preservation and return to soils is often used as an argument for prioritizing resources to biogas [21].

6.4 Eventual policy applications, limitations and perspectives

The analysis of specific public policies that could lead to investments in the technical solutions presented herein was beyond the scope of the present work. Here, we only aimed at analyzing the efficiency of these selected solutions in terms of biomethane supply. Yet, energy and climate challenges are only some of the grand challenges we are up against [40]. Denmark, as other countries, is also severely challenged by improving the quality of surface

water in lakes, streams and in the sea as regulated by the European Water Framework Directive. Agricultural land may have to be set aside in the coming years in the most nitrate sensitive areas, if no measures are implemented to significantly reduce nitrate leaching. One of the most efficient measure to this end is to convert the production from annual crops to perennial crops, but current agricultural markets do not allow for more grass production. In this study, we showed that biogas could be a market for this grass. In this case, the combination of biorefinery processes to extract high-value components such as protein concentrate, with biogas production to provide an important carbon source for the low-fossil economy, can represent a strategy for co-fulfilling a number of goals. The support towards such a development may be obtained from modifications of the EU Common Agricultural Policy (CAP) to further support climate and environmental perspectives of crops produced and exploit the potential of sustainable perennial grasses. Besides supporting markets for grass, the key policies that could be supported by the findings of this study would go in the direction of supporting methanation, and prioritization of straw for biogas. Both were found necessary in order to fully deploy the production of biomethane, at a level that reaches ca. 80% of current natural gas consumption.

The key underlying hypothesis of this study is that the development of the biogas sector and infrastructure allows, in the short- to medium-term, to mitigate dangerous global warming. Therefore, it should be kept in mind that biogas is likely to play a key role in the energy sector in the short-term (to offset fuels stemming from fossil carbon), and in other sectors in the long-term (as a source of hydrocarbon for chemistry). This study focused on the short-term and revealed the important demand from the heavy-duty transport (45% of the quantified demands herein). In the long-term, however, alternatives should be considered, such as direct electrification (so-called eHighways where trucks with a pantograph can connect to wires installed over the inner lane of highly used highways; [137]) or battery electrification (e.g. Tesla Semi, with a reported autonomy of 800 km and a charging time of 30 minutes for 650 km; [138]). The vision, thus, is not to consider whether investments should support biogas or electrification. Both have a role to play for the energy sector, but at different time scales, given their readiness level. Further, investments in biogas plants, upgrading technologies and the biomethane grid are to be seen as investments in supplying the hydrocarbons needed in the future low fossil carbon economy.

Although the issue of prioritization is unavoidable (of the resource first, of the biomethane produced second), it should be discussed in the light of techno-economic or multi-criteria analysis (keeping climate change mitigation as the main driver for renewable gas production), which was beyond the scope of this study. Electrolyzers, for example, require important amounts of energy input. In the methanation scenario, ca. 7.8 kWh of electricity is required per Nm³ of biogas to be upgraded (considering 58% P2G efficiency, excluding heat recovery). This is the equivalent of 0.83 PJ electricity input per PJ biomethane produced in the MET+ scenario. This emphasizes the importance of ensuring that surplus electricity from fluctuating power is used to power the electrolyzers. Other issues, such as the potential efficiency gains in the energy system of having decentralized gas-powered CHP units or grids could also be examined in such techno-economic analysis, along with a dynamic analysis. Moreover, the methodologies demonstrated in this study could be supplemented by integrated and spatial assessments allowing to identify and quantify all potential cross-

cutting issues and rebound effects (e.g. competing demands for the feedstock, additional land demand, interactions with the district heating system to absorb the waste heat, etc.).

Moreover, only longer retention time has been addressed as a technology to increase the biogas yield from biomass (methanation increases the biomethane production, but not the biogas yield). There exists several other technologies in the form of pre-treatment of the biomass prior to anaerobic digestion to increase the amount of biogas that can be produced from the process. However including pre-treatment technologies would complicate our study since the various biomass streams considered herein require different types of pre-treatment. Moreover, the energy consumption associated with the pre-treatment will vary from one biomass to another. Since pre-treatment mainly has the impact of speeding up the process, prolonged retention time will have the same effect as pre-treatment technologies, in terms of biomethane produced per feedstock used. Furthermore, the energy consumption associated with prolonged retention time is very limited and therefore this has been chosen as the technology to represent enhanced biogas yields.

Besides the framework it provides, the key result of this study lies in quantifying the national potential supply of biomethane, here for Denmark, in the framework of applying serious measures for increasing biogas production. This quantification builds upon 3 aspects: agricultural intensification (see Equation 1 and Table 1), degree of biomass mobilization (Table 2), and type of biogas technology (Equation 2). For these 3 aspects, two main types of data uncertainties were involved: those related to the individual data themselves (parameter uncertainties; e.g. current yield of winter wheat straw) and those related to the assumptions (scenario uncertainties; e.g. degree of biomass mobilization). The importance of individual data uncertainty on the final biomethane supply result can be captured through techniques such as error propagation or Montecarlo analysis [139,140], while sensitivity analyses are typically used to assess the importance of scenario assumptions. One limitation of our study is that parameter uncertainty was not incorporated. However, it is our claim that scenario uncertainty, in particular for biomass mobilization, greatly overcomes the overall parameter uncertainty induced by the individual data uncertainties. This can be seen by the important total range presented in Table 2, which varies from simple to double. Our approach, based on justified min/max ranges (Table 2), is the equivalent of a sensitivity analysis considering the extreme ranges of possible values.

7 Conclusion

This study provided a framework for quantifying how key measures combining sustainable intensification of agriculture and new biogas technologies could massively increase the supply of biomethane, and to which extent this increased supply matters in comparison to emerging demands for methane gas. This framework was illustrated through a national case study applied to Denmark. This combination of sustainable agricultural intensification measures, with biogas technology improvements and energy system analysis is the key novelty of the study.

Besides the framework itself, the key insights that can be derived from our national case study can be summarized as follows:

- Through a variety of sustainable intensification measures, the supply of agricultural biomass suitable for anaerobic digestion can be increased more than 4-fold. The 2 most significant measures include harnessing a greater share of the excreted animal manure, and converting rapeseed areas and cereal areas from nitrate-vulnerable soils to high-yielding perennial grasses (>15 Mg DM ha⁻¹).
- Biogas can play a major role in supplying a renewable gas source for services that cannot be immediately electrified, but this strongly depends on the extent at which resources are made available, and technology deployed. The gas demand for these services (95 PJ y⁻¹) could only be met under a high prioritization of the biomass potential (here corresponding to 72% of the resources) combined with biogas technologies allowing to produce a maximum of biomethane (doubling the retention time of anaerobic digestion combined with biological methanation upgrading), which allowed for the supply of 105 PJ biomethane y⁻¹. This corresponds to ca. 85% of the current natural gas consumption.
- Mobilizing animal manure is, in a country with high animal density, the most important resource for increasing biomethane production. Here, manure alone represented between 27% (low mobilization) and 34% (high mobilization) of the biomethane produced, in terms of PJ y⁻¹.
- The mobilization of straw and perennial grass resources (stemming from the conversion of cereals and rapeseed areas) was also shown to be of importance; these two resources combined represented between 46% and 55% of the biomethane supply, in terms of PJ CH₄ produced. Organic waste, on the other hand, represented no more than 12% of the biomethane supply. Removing it from the best case scenario decreased the biomethane production from 105 to 95 PJ y⁻¹, i.e. still allowing to meet the gas demand for services that cannot be immediately electrified.

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