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Towards sustainable methane supply from local bioresources: Anaerobic digestion, gasification, and gas upgrading

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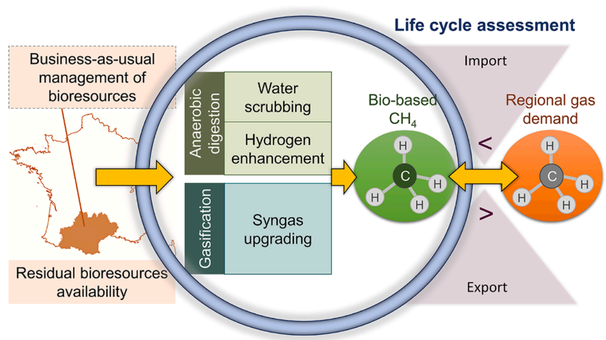
HIGHLIGHTS

- Significant potentials for regional export of methane based on local bioresources were uncovered.
- Intercrops are an important unexploited resource for biomethane provision but should be preferred for gasification.
- Emissions from manure & green wastes current uses are larger than those associated with their use in anaerobic digestion.
- For relevant bioresources, gasification increases methane output up to five times over anaerobic digestion.
- Maximizing CH_4 production benefits six impact categories, including climate, but worsen other ten.
- Systematic life cycle assessment (LCA) is critical for identifying improvement potentials.

GRAPHICAL ABSTRACT



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ABSTRACT

Methane is a versatile and storable energy carrier, which is likely to play an important role in the European transition towards a low fossil carbon energy sector. We investigate the potentials for meeting regional methane demands through conversion of local residual bioresources for supply of bio-based CH_4. We have developed a tiered assessment framework involving i) allocation of residual and constrained bioresources to conversion pathways based on physical and biochemical properties, ii) life cycle assessment (LCA) of technology conversion pathways through process-oriented parameterisation of the LCA model, and iii) LCA modelling of system-level technology implementation scenarios for quantification of regional potentials for bio-based CH_4 supply and environmental savings, in view of current uses of the same bioresources. Two main technology conversion pathways are included: gasification and anaerobic digestion, both with hydrogen enhancement. The latter was also considered with water scrubbing upgrading. The framework is implemented for the French region, Occitania, with a residual bioresource potential of 48 TWh·y^{-1} (distributed on 41 different bioresources), and an annual methane demand of 17.5 TWh·y^{-1}, currently supplied by natural gas. The assessment results clearly demonstrate that utilisation of available residual bioresources has tremendous potential both for covering

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current gas demands in the region (up to about seven times in some scenarios) but also for a reduction in climate change impacts from the region (up to about 37%).

1. Introduction

In the European Union (EU), the recent Green Deal [1] has promoted several policy instruments intended to push towards achievement of the Paris Agreement goal of limiting global mean surface temperature increases below 2 °C, including a Climate Law [2]. This involves dramatic cuts in use of fossil fuels and reductions in fossil carbon dioxide (CO_2) emissions throughout Europe [1]. Natural gas is a major feedstock in the EU energy supply, especially for provision of power and heating in buildings and industrial processes (natural gas is the largest energy source for heating in the residential sector, and the second largest energy source after oil in industry), making up half of EU's total final energy consumption for heating and cooling [3]. In France, for instance, natural gas is the most important energy source used within the district heating network (~35% of consumption), representing about 16% of overall national primary energy consumption (after nuclear and oil) [4]. As gas distribution infrastructure is well-established throughout Europe, methane gas may play a key role also in the future defossilisation by introduction of bio-based methane (CH_4), i.e. CH_4 produced from biomass through biological (anaerobic digestion) or thermochemical (gasification) conversion pathways. This transition, however, has to be assessed in view of the regionally available bioresources and the environmental performance of the involved conversion pathways, to ensure the intended effects on climate change in a systemic perspective.

In its "net zero by 2050" study [5], the International Energy Agency (IEA) forecasts that global bio-based CH_4 demand will increase to 8.5EJ in 2050. In this context, bio-based CH_4 is considered introduced primarily into the natural gas distribution grids through existing pipelines for application directly in end-user equipment (by 2050 average blending is predicted to increase to above 80% in many regions). The IEA [5] further predicts that by 2050, half of CH_4 consumption will be in the industrial sector, replacing natural gas as the primary source for process heat, while the building and transport sectors each will consume 20% of CH_4. To accommodate the transition from fossil to renewable gas supply, a range of strategic actions and investments are needed at regional level to support: i) utilisation of available bioresources located a significant distance away from the gas grid, ii) increasing network injection capacity (e.g., storage units throughout the distribution network), and iii) providing cost reduction initiatives supporting increased competition of bio-based CH_4 with natural gas [6]. In addition to reducing or preventing fossil gas imports, renewable gas supports valorisation of local bioresources, in particular low-value residual bioresources, while increasing local energy production. To promote such a local bioeconomy for bio-based CH_4 production, analysis of local bioresource availability is needed with respect to quantity, accessibility and quality (chemical, physical, and nutritional properties) [7–9]. As different bioresources are associated with different physico-chemical properties, this has profound importance both for the selection of conversion technology and for the environmental consequences associated with use of these bioresources for energy purposes.

The production of bio-based CH_4 from thermochemical and biological processes has been addressed in several studies (e.g., Wang et al. [10], Leonzio [11]), some questioning whether bio-based CH_4 produced from different technologies contribute with environmental benefits in comparison with natural gas. Tagliaferri et al. [12], for instance, carried out a life cycle assessment (LCA) to support decisions on the most environmentally-efficient technology (from both thermochemical and biological processes) to produce a given amount of CH_4, and on the most competitive waste management option for a given amount of waste input. Hahn et al. [13], who performed an LCA of biogas plant configurations, included in the environmental analysis a demand-oriented biogas supply solution for flexible power generation, together with comparison of primary energy supply and GHG balance. Similarly, Hamelin et al. [14] investigated strategies, including sustainable intensification of agriculture, methanation and longer retention times in biogas plants, to boost bio-based CH_4 supply at a national level while also addressing potential future changes in gas demands, such as for transport and flexible power supply. Ardolino and Arena [15] analysed and compared the environmental performance of producing biomethane from both anaerobic digestion and gasification for use in road transport.

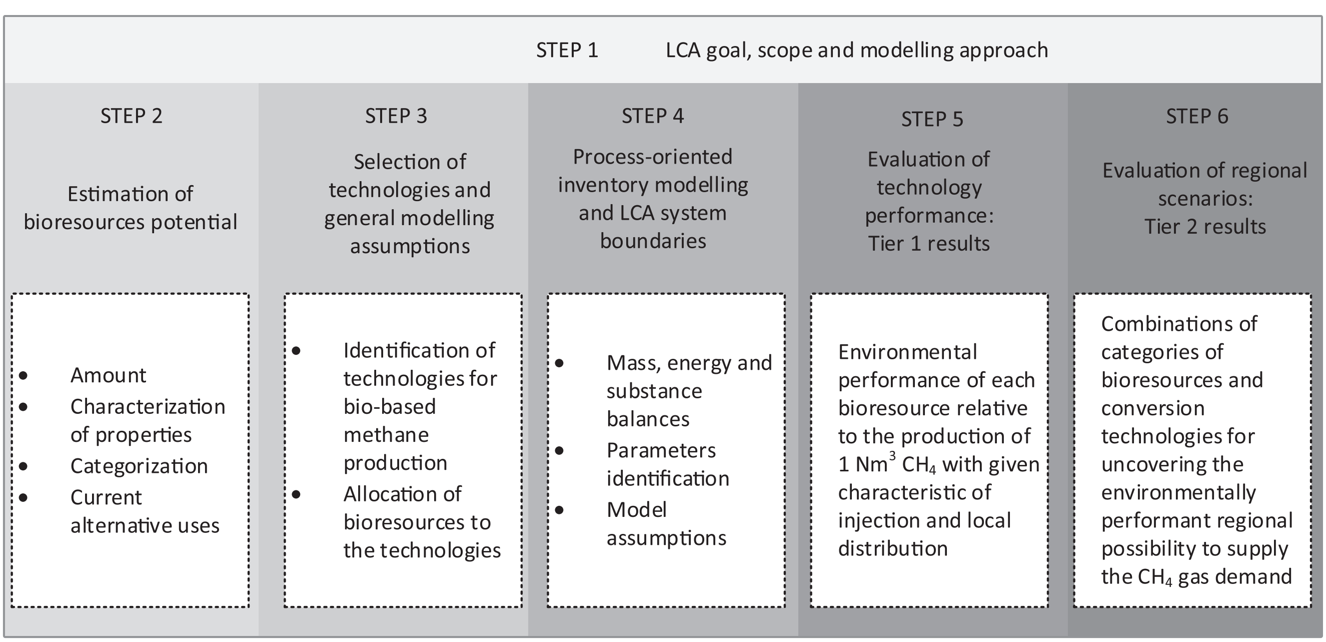


Fig. 1. Overview of tiered assessment framework for regional implementation of bio-based solutions.

While the above studies are valuable, none simultaneously addressed regional constraints in bioresource availability with systematic assessment of the environmental consequences from employing different conversion technologies and technology configurations for production of bio-based CH_4. Yet, waste and residual bioresources need to be considered on a regional basis, due to logistics (storage of large volumes and expensive transport given by their relatively low energy density compared to other fuels) and bioresource properties [16]. Most existing LCA studies addressing prioritisation of bioresources for energy purposes, involve the use of "standard" data inventories for the selected technologies (e.g., Tagliaferri et al. [12]), thereby neglecting to reflect the importance of the bioresource characteristics (physical and biochemical properties) for the environmental performance of the selected conversion technologies [16]. Without systematically evaluating both bioresource availability, bioresource properties and technical configuration of the conversion technologies, LCA studies may not provide relevant quantification of the environmental impacts. Ultimately, this may lead to poorly justified decision-making and implementation of bio-based solutions not leading to net climate benefits.

The aim of this study is to investigate the potentials for regional supply of CH_4 based on locally available residual bioresources, while at the same time providing the largest reductions in climate change impacts. Through a tiered LCA modelling framework, bioresource and conversion technology pathways are assessed for identification of technology parameters critical for the overall environmental performance. Based on analysis of regional bioresources, physico-chemical properties, technology operational parameters, 19 bioresource conversion pathways are evaluated both at technology and system levels for regional supply of bio-based CH_4. While the tiered assessment framework may be applicable for a variety of regional supply-demand situations, the framework is implemented here for the French region of Occitania, with concrete ambitions for bio-based CH_4, local energy transition, and climate savings.

2. Method

The tiered assessment framework (Fig. 1) is based on the life cycle assessment (LCA) methodology, described in ISO 14040 and ISO 14044 [17,18]. While the framework is applied to a specific case study region, it may be implemented for any area (a region can range from a city, to a country or several countries). All calculations are transparently documented in Electronic Supplementary Information (ESI1). The selected case study is Occitania (72,700 km^2, 5.8 M inhabitants), an administrative region in southwest France (one of the 92 European NUTS1 Regions of the territorial units for statistics' nomenclature) [19] with the ambition to become the first "positive energy" region in Europe, meaning that 100% of its energy consumption will be provided by the local production of renewable energy (an ambition locally referred to as "REPOS") [20]. Currently, 41% of the region's electricity production is from renewable energy. Annual gas consumption is 17.5 TWh, and total energy consumption amounts to 128 TWh [21]. This region is known to have a potentially high availability of bio-based residual resources (Karan and Hamelin [9]), but it is also one of the least developed in France in terms of biomethane injection sites, [22] indicating that large-scale investments have to be made in the near future regarding potential phase-in of bio-based CH_4.

2.1. Step 1: LCA goal, scope and modelling approach

The functional unit, in this case the service to be supplied in all scenarios, is "*to fulfil the annual demand for CH_4 gas in the French Occitania region, to the extent possible with local residual bioresources or else with imported natural gas*." The focus in this study included only the gas demand for power production. Residual bioresources are understood as those that can be supplied without generating additional demand for land. In other words, so-called "first-generation energy crops" are not considered.

A consequential approach was applied in the study [23–26]. Accordingly, multi-functionality was addressed using system expansion, i.e. we considered in which markets and for which applications the generated co-products were used, and we accounted for the potential displacement and substitution effects from additional supply of these co-products in the market. This was under the assumption that from a long-term perspective, markets are unconstrained and fully elastic, wherein an increase in demand translates into an equivalent increase in supply (1:1 substitution) [27]. Moreover, only marginal suppliers were considered, i.e. those capable of reacting to a change in demand.

Marginal energy mixes were calculated based on variations in annual supply derived from European and French energy forecasts to 2050 (details in Section S19, ESI1): accordingly, the marginal electricity considered is composed of 55% wind (offshore), 25% solar, 7% biomass, 5% natural gas, 5% hydro and 2% geothermal, while the marginal heat supply considered for the case study region is composed of 91% heat pumps, 8% geothermal and 1% solar thermal [28–30].

The LCA modelling of the technology conversion pathways followed a process-oriented approach: mass and energy balances were established through parametrised mathematical relationships considering the detailed composition of input bioresources, in order to model their conversion-to-output products, both within technologies and throughout the entire system [31]. For example, the carbon content of a specific bioresource was quantified based on the associated biochemical properties (e.g., sucrose, cellulose), while the produced bio-based CH_4, co-products and rejects were quantified on the basis of technology configurations, process conditions and stoichiometry of the involved conversion reactions. Additionally, we addressed the environmental consequences of diverting the residual biomasses from their conventional (current) management routes, herein defined as "counterfactuals," to the selected bio-based CH_4 production and utilisation. The environmental performance of each technology conversion pathway, for each bioresource, was compared with the impacts of conventional natural gas production (reference product, RP). The inventoried flows were then calculated into environmental impacts using the Environmental Footprint EF3.0 life cycle impact assessment (LCIA) method [32] considering all mid-point impact categories. Accordingly, biogenic CO_2 emissions were considered with a global warming potential (over a 100 y horizon: GWP_{100}) of 0 kg $CO_{2\text{-eq}} \cdot kg^{-1}$ CO_2, while long-term sequestration of biogenic CO_2 translated into a GWP_{100} of -1 kg $CO_{2\text{-eq}} \cdot kg^{-1}$ CO_2. It should be noted, however, that the framework can be adapted to any other LCIA method.

The modelling was carried out with the EASETECH [33] life cycle assessment model, using background data from Ecoinvent 3.6 [34]. The model was parametrised, thus giving an uncertainty distribution to each parameter (e.g., representing technology performance and configuration, system framework conditions), based on estimations for minimum and maximum values of individual parameters. Whenever possible, these ranges were literature-based; otherwise, we assumed a variation around the average value for example of 20% or 10%, depending on the type of parameter and associated process (see details in ESI1). Uncertainties were propagated with a Monte Carlo analysis considering 10,000 iterations. The contribution from each of the 507 parameters involved in the assessment to the overall uncertainty of the results (i.e. the impact metrics for each of the 16 environmental impacts considered) was assessed with the methodology provided by Bisinella et al., [35] as further detailed in the ESI1.

2.2. Step 2: Estimation of bioresource potentials for the selected region

Residual bioresources available in Occitania were identified based on regional inventories [36,37] commissioned within the framework of the French national bioeconomy strategy. These inventories provided the quantity, nature and accessibility of residual resources linked to economic and industrial constraints. We identified 41 different residual bioresources, which we categorised within ten categories (Table S1, in ESI1). These bioresources categories were generated from agro-industrial processes, urban and commercial activities, household consumption and primary forestry and agricultural activities. Out of the ten categories, three represent more than 80% of total wet weight, namely: manure (39.7%), intercrops (27.12%, modelled as *Trifolium alexandrinum* with a yield of 20,000 $kg_{ww} \cdot ha^{-1}$ (Section S11, ESI1), used instead of catch crops and grown and harvested between two main crops), and crop residues (23.3%). The other categories included wood waste (2.45%), biowaste (2.16%), agrofood residues (1.87%), sludge (1.87%), green waste (0.923%), pruning residues (0.263%) and forestry residues (0.104%). All 41 bioresources were integrated into a residual bioresource database reporting the available quantities of each stream (Table S1, ESI1), along with their detailed characterisation in terms of physical, chemical, biochemical and nutritional composition and properties (Table S2, ESI1). All calculations related to the establishment of the residual bioresource database are provided in Tabs. S1.1 to S1.8, ESI1.

2.3. Step 3: Selection of technologies and general modelling assumptions

Two technology conversion pathways were considered for production of bio-based CH_4: biological degradation through anaerobic digestion (producing biogas), and thermochemical degradation through gasification (producing syngas).

The two types of gas produced are mixtures of CH_4 and other gases that must be upgraded, i.e. purified, to meet the requirements for injection into the natural gas grid – among other properties, a CH_4 content of 98.3%$_{vol}$ [38] (details in Section S6, ESI1). Several upgrading technologies exist for biogas, as thoroughly described in Angelidaki [39]. Biogas is essentially a mixture of CH_4 and carbon dioxide (CO_2), along with trace gases, and its overall composition depends upon the bioresource properties as input to the digestion process [40]. Here, two types of upgrading processes are considered for the biogas, namely i) the simple removal of CO_2, based on conventional physico-chemical absorption methods, i.e. water scrubbing, and ii) ex-situ hydrogen enhancement, where the CO_2 portion of the gas is converted into CH_4 following the Sabatier reaction (i.e. $CO_2 + 4H_2 \leftrightarrow CH_4 + H_2O$) [40]. For syngas, which is essentially a mixture of hydrogen and C-gases (CO, CH_4, CO_2), only hydrogen enhancement upgrading is considered. When bio-based CH_4 is injected into the natural gas grid afterwards, the gas must have the same pressure as the target network to which it is connected (i.e. connection point). In this specific case study, we consider a transmission network at 40 bars [41]. A CH_4 slip of 1% was considered during the injection step (according to the Ecoinvent 3.6 process "*market for natural gas, low pressure*", consequential).

2.3.1. Methane production pathway: Anaerobic digestion

Anaerobic digestion converts the biodegradable fraction, such as lipids, proteins and carbohydrates of bioresources, into biogas and digestate. The latter is the liquid residue that contains what has not been degraded anaerobically. The first upgrading process considered herein, namely water scrubbing, is an absorption process exploiting CO_2 solubility in water, and it is amongst the most commonly used upgrading technologies [41]. Inventory data from an industrial plant were used, as further detailed in Section S6.1, ESI1. The resulting output of the process was two gaseous streams: recovered CH_4 (100% of the CH_4 in the input biogas) at an output pressure of 40 bar, as well as a gaseous reject (essentially CO_2), emitted into the atmosphere. A loss of 0.1% of the input biogas was considered, as well as a CH_4 slip of 0.1% of produced bio-based CH_4 [41]. Electricity consumption of 0.13 $kWh \cdot Nm^{-3}$ CH_4 (corresponding to 2.16% of biogas input) was assumed for the upgrading process, along with 0.062 $kWh \cdot Nm^{-3}$ CH_4 injected, based on Tyra et al. [38].

The second upgrading process included in this study, namely ex-situ hydrogen enhancement, involves production of hydrogen through water electrolysis, powered by electricity. There are currently several electrolyser technologies and important ongoing research developments available to produce so-called "green hydrogen" [1]. Herein, conventional alkaline electrolysers were considered, [42] as they offer the most commercially available electrolysis technology for hydrogen production to date (Section S6.2, ESI1). We did not include the management of oxygen produced after an alkaline electrolyser. While we considered this oxygen a loss in the system, it may be used for replacing pure oxygen potentially leading to savings of ca. 0.8 $kgCO_{2\text{-eq}}$ per kg O_2 production avoided (Ecoinvet 3.6, market for oxygen, liquid, RER). For hydrogen production, in terms of energy, we considered that 100% input electricity was used to produce 68.0% of hydrogen and 19.6% heat loss, of which 5% is unrecoverable heat loss and 14.6% is recoverable through district heating [41]. A water input of 184 $kg{\cdot}MWh^{-1}$ was considered, [42] corresponding to 0.35 $kg{\cdot}m^{-3}$ H_2. The consumption data for the electrolysis unit is reported in Table S3, ESI1. Electricity consumption was estimated as 20.9 $kWh{\cdot}kg^{-1}$ H_2, corresponding to 0.11 $kgCO_{2\text{-eq}}{\cdot}m^{-3}$ H_2 on the basis of the marginal electricity mix. This hydrogen is then injected, along with raw biogas, into an ex-situ unit where, in the presence of a nickel-based catalyst, hydrogen reacts with carbon dioxide in the biogas to produce additional CH_4 (Sabatier reaction). The upgrading occurs at temperatures between 200 and 550 °C [43]. Our life cycle inventory considers the use of 93 mg of a nickel-based catalyst with 19% nickel and 81% aluminium alloy to produce 1 Nm^3 of CH_4 [44]. Ex-situ upgrading was preferred over in-situ H_2 injection, because the ex-situ method better supports a stable CH_4 yield not affected by the microbial community in the digester, such as hydrogenotrophic methanogens [45]. The resulting bio-based CH_4 also had a pressure of 4 bar. Electricity consumption considered for the Sabatier reaction was 1% of the total input energy based, i.e. 0.11 $kWh{\cdot}Nm^{-3}$ CH_4, while heat recovered into district heating was 10% of the total input energy, corresponding to 3.82 $MJ{\cdot}Nm^{-3}$ CH_4.

The anaerobic digestion process was modelled as a continuous stirred tank reactor maintained at a thermophilic temperature (56 °C). The produced biogas, in terms of both volume and composition, was quantified based on the degradation of carbon, lipids, proteins and polysaccharides, such as cellulose, sucrose, starch and hemicellulose, in the input residual bioresources, as explained in Lodato et al. [46]. We considered an electricity and heat consumption of respectively 0.049 kWh and 0.089 MJ per kilograms wet weight of the mass in input, as well as a CH_4 slip of 2% of total CH_4 generated in the biogas [41]. The digestate was assumed temporarily kept in covered storage and then applied on land, thereby substituting mineral fertilisers – as further detailed in the ESI1 (Section S4). Fertiliser substitution was modelled according to Evangelisti et al. [47], considering different substitution rates depending on the type of organic material to be applied, as well as nutrients. For bioresources where the counterfactuals also involve fertiliser substitution, the net difference between induced and avoided mineral fertiliser substitution is considered.

2.3.2. Methane production pathway: Gasification

Biomass gasification produces syngas (i.e. a gas mixture of H_2, CO, CO_2, CH_4, N_2 and H_2O, plus impurities), char and tar. Here, gasification process conditions and configuration were selected to allow production of a syngas with the highest possible CH_4 content. We selected a fluidised bed gasifier with a cleaning and conditioning system, combined with a methanation unit, to convert the carbon pool in the syngas into CH_4. Li et al. [48] demonstrated that i) gasification efficiency is optimised with an optimum range of equivalence (or air–fuel) ratio (ER) between 0.25 and 0.33; ii) CH_4 content is not expected to be higher than 3% at temperatures above 727 °C (1000 K); for example, with an ER = 0.30 and a temperature of 827 °C (1100 K) the CH_4 concentration is 0.02%; iii) high temperatures do not require high pressures and iv) temperature has an effect on tar yield with higher temperatures decreasing production due to increased tar-cracking rates. On this basis, we considered a gasifier temperature of 850 °C with a pressure of 1 bar, air as a fluidising agent and an ER of 0.33. The modelling of the feedstock and gaseous flows in and out the gasifier was based, to the extent possible, on data from a pilot-scale installation (GoBiGas), [49] which at the time of writing is the only operational unit where syngas-to-CH_4 is produced at scale. Other inputs were based on published literature [49–51]. Initial activation is carried out by adding calcine and potassium while heating the system through the combustion of natural gas (details in Table S13, ESI1). During stopped and initial start phases, the gasification reactor is fed with pure nitrogen, before it is transformed into steam. The bed material takes up the ash components that provide the catalysts for the gasifier in the combustor, which also includes supplemented ash components such as potassium, sulphur and calcium. The choice of this specific configuration is reflected in the LCA model inventory and associated emissions. Considering these process conditions, we estimated, for each individual residual bioresource suitable for gasification (see selection in the following section), output syngas composition through an equilibrium model that we implemented into EASETECH, based on the equilibrium equations presented in Ferreira et al. [52]. From this point onwards, the equilibrium model is referred to as the "GA model". In brief, our stoichiometric GA model (ESI 2) considered the following three principles:

(i) A global gasification reaction (Eq. 1).

$$CH_xO_yN_z + wH_2O + m(O_2 + 3.76N_2) \rightarrow n_1H_2 + n_2CO + n_3H_2O + n_4CO_2 + n_5CH_4 + n_6N_2$$

Where:

x, y, z: number of atoms of hydrogen, oxygen and nitrogen per number of atoms of carbon in the biomass.

w: molar moisture amount in the biomass.

m: molar air amount.

n_1 to n_6: stoichiometric coefficients defining syngas composition (GA_results, ESI3).

(ii) Four material balance equations for C, H, O and N (Eq. 2–5);

$$C: \ n_2 + n_4 + n_5 = 1$$

$$H: \ 2n_1 + 2n_3 + 4n_5 = x + 2w$$

$$O: \ n_2 + n_3 + 2n_4 = y + w + 2m$$

$$N: \ 2n_6 = z + 7.52m$$

(iii) Two independent equilibrium reactions (Eq. 6, 7) and the two-kinetics associated therewith [53]:

$$C + 2H_2 \leftrightarrow CH_4 - 74.8 \ \ MJ/kmol \ \ \text{(Hydrogasification)}$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 - 41.2 \ \frac{MJ}{kmol} \ \text{(Shift reaction)}$$

The GA model itself is provided in ESI3 (GA_model), where the input composition and the operation conditions are implemented as parameters and can be changed to model different biomass types or process conditions. On average, for 1 kg_{ww} of bioresources from a given bioresource category, the GA model provided 2.4 Nm^3 of syngas, which is consistent with published experimental data (e.g., Mustafa et al. [53]).

The composition of gas post-upgrading was also estimated through a stoichiometric model (C-to-CH4_model, ESI3), referred to as the "C-to-CH_4 upgrading model". The upgrading process is based on ex-situ catalytic H_2 enhancement, following the same process and assumptions as mentioned previously for anaerobic digestion. According to Mustafa et al. [53], a CO_2 conversion of almost 98% is needed to achieve CH_4 content higher than 90% in the upgraded gas output, and a CO conversion of 99% is required to achieve a CH_4 content of 95%. Thus, we assumed a conversion rate of 99% for both CO and CO_2 in the methanation (C-to-CH_4 upgrading) model used in this study.

Catalytic methanation reactors have operating temperatures ranging between 200 and 550 °C, with a pressure range between 1 and 100 bar [54]. Göts et al. [54] demonstrated that in order to reach a CO_2 conversion rate of at least 98%, it is recommended to have a temperature below 225 °C (1 bar) or 300 °C (20 bar). In addition, Giglio et al. [55] considered a methanator inlet temperature of 220 °C. Therefore, we considered an operating temperature of 220 °C and a pressure of 1 bar.

The bio-based CH_4 in the output had a pressure of 30 bar [41]. Electricity consumption considered for the C-to-CH_4 upgrading was 63% of the input energy, corresponding to 6.68 kWh · Nm^{-3} CH_4. The heat produced and used in district heating was estimated at 22% of fuel input energy, corresponding to $8.39 \cdot 10^{-5}$ MJ · Nm^{-3} CH_4 [41]. The C-to-CH_4 upgrading model (ESI2) needs:

(i) Syngas characterisation as H_2, CO, CO_2 and CH_4 from the GA model. Water and nitrogen are neglected, because they do not react during upgrading and can also be recirculated in the gasification process.

(ii) Hydrogenation reactions, respectively, methanation and Sabatier (CO_2 methanation), to convert carbon monoxide and CO_2 to CH_4 (Eq. 8, 9).

$$(xsi1)CO + 3(xsi1)H_2 \leftrightarrow (xsi1)CH_4 + (xsi1)H_2O \quad -206 \ \ MJ/kmol \ \ \text{(Methanation)}$$

$$(xsi2)CO_2 + 4(xsi2)H_2 \rightarrow (xsi2)CH_4 + 2(xsi2)H_2O \ -165 \ \ MJ/kmol \ \ \text{(Sabatier)}$$

(iii) External hydrogen needed in mol·s^{-1} (in addition to the one supplied from the syngas itself), which is three times the moles of CO and four times the moles of CO_2 from the input syngas. The

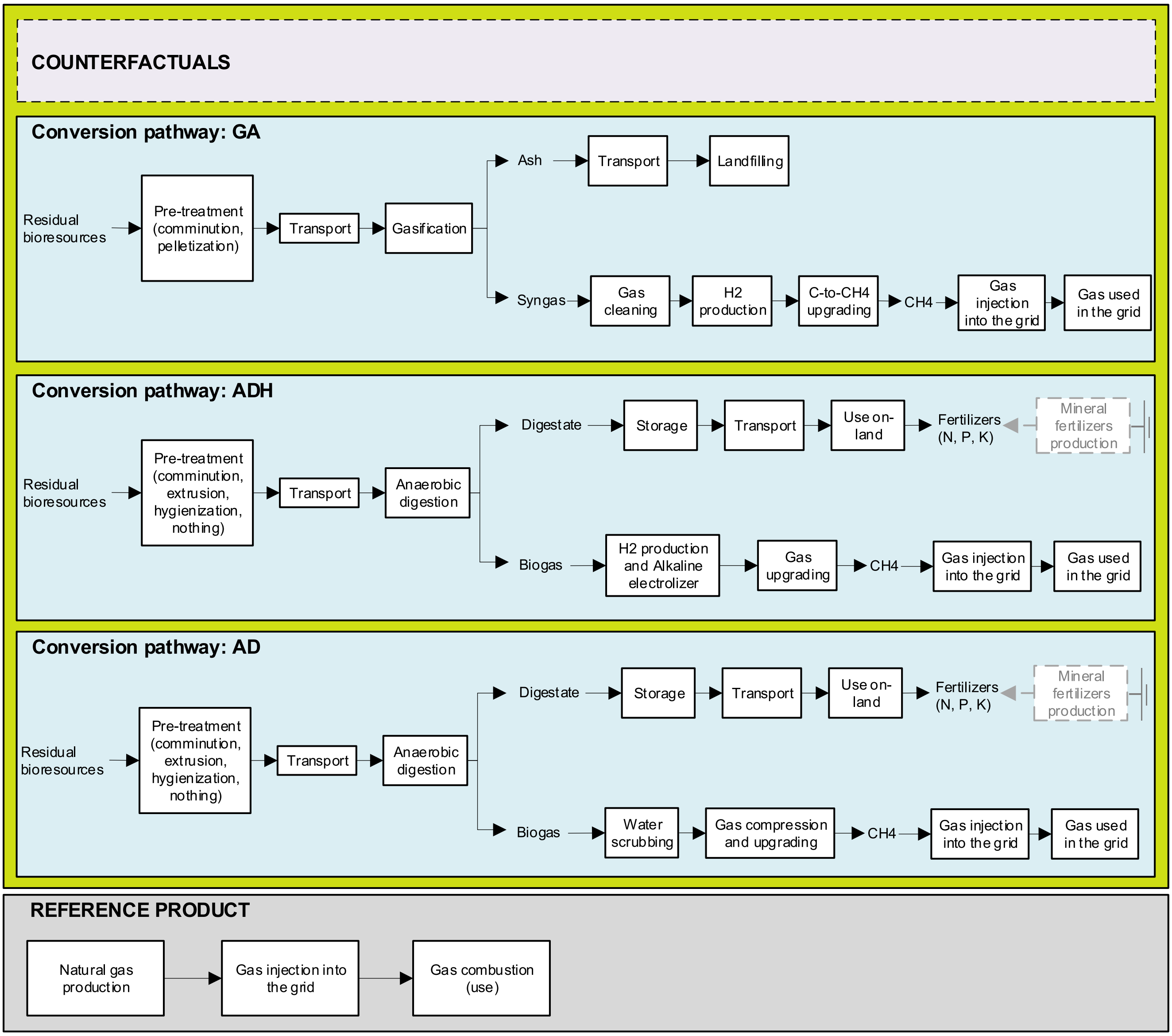


Fig. 2. Schematised process flow diagram of residual bioresource conversion to bio-based methane (CH_4). Dotted lines represent avoided processes. On the top, in dots, are the counterfactual uses of the bioresources; in light blue, main processes involved in the selected CH_4 production pathways: i) gasification with C-to-CH_4 upgrading (*GA*); ii) anaerobic digestion with hydrogen enhancement (*ADH*) and iii) anaerobic digestion with water scrubbing (*AD*). In green, the representation of an LCA scenario. The performance of each LCA scenario is compared with the reference product (in grey)(i.e. fossil-based CH_4 production and use). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

hydrogen needed to convert CO and CO_2 to CH_4 for each category of the gasified bioresource is 0.49 Nm^3 for crop residues, forest residues, pruning residues and wood waste, 0.51 Nm^3 for green waste and intercrops and 0.43 Nm^3 for biowaste (C-to-CH4_results, ESI3).The kmol of H_2 needed for C-to-CH_4 upgrading for each bioresource category is in Table S19 of ESI1.

From the C-to-CH_4 model, two stoichiometric coefficients were determined for estimating the gas in output: i) xsi1 (the stoichiometric coefficient of methanation in Eq. 8), and ii) xsi2 (the coefficient of the Sabatier reaction in Eq. 9). The coefficients applied for each gasified bioresource category are shown in Table S18 of ESI1.

Emissions due to C-to-CH_4 upgrading are modelled based on the GoBiGas plant and grouped according to nine sub-processes: i) hydration of olefins and carbonyl sulphide (COS); ii) H_2S removal; iii) removal of trace components through a guard bed; iv) water-gas shift reaction; v) pre-methanation; vi) CO_2 removal; vii) four-stage methanation; viii) drying and ix) compression before feeding the natural gas grid. The inventory is presented in Section S6.4, ESI1.

2.3.3. Selection of the technology conversion pathway based on feedstock properties

Moisture content is the main criterion applied for initial allocation of each bioresource category to one of the two CH_4 production pathways. In the literature, a relative wide range of moisture contents are reported for various gasifier configurations with typical values around 15–30%, and in all cases below 35% [56–61]. Consequently, herein we decided on a limit of 35% moisture content for gasification. On the other hand, biological degradation is preferred for bioresources with low lignin content, and it is better suited to handling bioresources with high moisture contents, e.g., higher than 70% [62]. Moreover, it preserves the macronutrients recoverable from the digestate. For bioresources having a moisture content in a range of 35–70%, not preferable to either anaerobic digestion or gasification, more investigation is needed

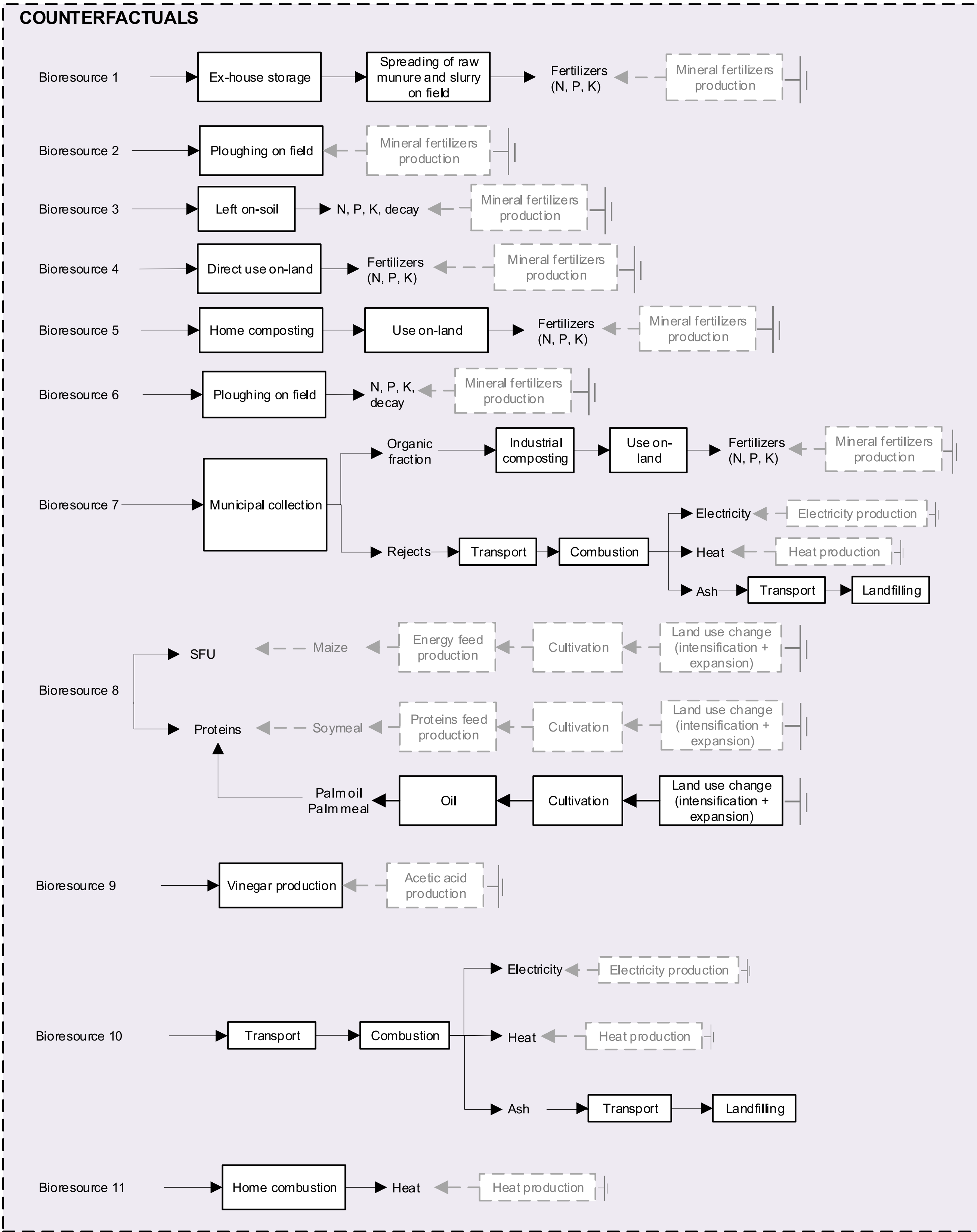


Fig. 3. Process flow diagrams for the counterfactuals considered in the study. (*Bioresource 1*: Cow solid manure, horse solid manure, cow slurry, pig slurry, chicken slurry, chicken manure; *Bioresource 2*: Wheat straw, triticale straw, maize silage, barley straw, rye straw, oat straw, sorghum straw, beet tops, potato tops, sunflower canes, oilseed straw, soybean straw, corn stover; *Bioresource 3*: Hardwood, softwood, poplar, viticulture, fruit pruning; *Bioresource 4*: Sewage sanitation; *Bioresource 5*: Household biowaste; *Bioresource 6*: Intercrops; *Bioresource 7*: Garden and park mowing, green waste, urban garden waste, urban market waste, commercial biowaste; *Bioresource 8*: Cheese products; *Bioresource 9*: Spoiled wine; *Bioresource 10*: Industrial hardwood, industrial softwood, industrial poplar, wood waste, butchery meat, canned fruit and vegetables; *Bioresource 11*: Pruning residues from hedges).

Table 1
"Bioresource category and technology conversion pathway combination" criteria results. In weighted average, values in bold are suitable for GA, while values in italic are suitable for AD/ADH. Classification of bioresources according to the preferred pathway is provided in the right-hand column.

Bioresource category	Fraction name	Share to the category ($\%_{ww}$)	Water ($\%_{ww}$)	VS ($\%_{TS}$)	Lignin ($\%_{TS}$)	Cellulose ($\%_{TS}$)	Proteins ($\%_{TS}$)	Lipids ($\%_{TS}$)	Relevant pathways
Agrofood residue	Butchery meat	5	25	51	0	0	36	4	AD/ADH
	Fruit and vegetable canned	21	85	92	0	1	13	4	
	Cheese products	7	49	94	0	0	47	42	
	Spoiled wine	67	87	98	0	0	2	0	
	Weighted average		*78*	94	0	0	9	4	
Biowaste	Urban markets biowaste	17	22	91	7	17	11	10	AD/ADH, GA
	Commercials biowaste	23	29	95	8	11	21	7	
	Household biowaste	60	73	87	23	0	14	14	
	Weighted average		54	90	**17**	**6**	*15*	*12*	
Crop residues	Wheat straw	30	9	93	7	43	4	1	AD/ADH, GA
	Triticale straw	3	8	95	0	48	3	2	
	Maize silage	8	63	96	3	20	0	7	
	Barley straw general	6	9	93	7	42	4	1	
	Oat straw	0	10	93	7	38	0	2	
	Sorghum straw	1	7	93	7	41	4	1	
	Rye straw, Occitanie	0	8	92	9	41	4	0	
	Beet tops	0	77	84	8	11	17	2	
	Potato tops	0	77	87	0	0	11	4	
	Sunflower canes	15	10	88	0	0	7	2	
	Oilseed straw	3	6	96	18	45	0	0	
	Soybean straw	0	11	92	16	44	7	4	
	Corn stover	34	70	93	6	0	7	2	
	Weighted average		**34**	93	**5**	**20**	5	2	
Forest residues	Hardwood	58	18	98	23	40	0	0	GA
	Softwood	42	21	99	29	44	0	0	
	Poplar	0	10	99	25	46	0	0	
	Weighted average		**19**	98	**26**	**42**	0	0	
Green waste	Garden and park mowing	0	7	89	0	0	0	0	AD/ADH, GA
	Green waste	100	54	89	5	15	0	0	
	Urban garden waste (leaves)	0	33	94	19	27	10	6	
	Weighted average		54	89	**5**	**15**	0	0	
Intercrops	Oat forage	100	*74*	90	**5**	**39**	*11*	*3*	AD/ADH, GA
Manure	Cow solid manure	82	85	81	25	18	14	7	AD/ADH
	Horse solid manure	7	76	89	12	38	6	2	
	Cow slurry	6	89	81	25	18	14	7	
	Pig slurry	3	93	81	19	12	24	14	
	Chicken slurry	2	83	86	5	39	12	3	
	Chicken manure	1	52	83	8	18	24	2	
	Weighted average		*85*	82	**24**	**19**	*14*	*6*	
Pruning residues	Viticulture	85	44	95	17	19	5	2	GA
	Fruit pruning	11	18	96	12	36	0	0	
	Pruning products and hedges	4	7	89	0	0	0	0	
	Weighted average		40	94	**16**	**20**	4	1	
Sludge	Sewage treatment/ sanitation plants	100	*90*	80	0	0	*47*	*11*	AD/ADH
Wood waste/industrial end of life wood	Industrial/Hardwood	0	15	99	0	0	0	0	GA
	Industrial/Softwood	0	24	98	0	0	0	0	
	Industrial/Poplar	0	9	98	0	0	0	0	
	Wood waste	100	16	95	29	44	0	0	
	Weighted average		**16**	95	**29**	**44**	0	0	

considering also other biochemical properties. Buffiere et al. [63], found inverse proportionality between the ligno-cellulosic content and the anaerobic digestion biodegradability, which decreases in bioresources with the sum of cellulose and lignin contents higher than ca. 25–30$\%_{VS}$. However, Møller at al. [64] demonstrated a higher methane productivity based on higher VS contents, corresponding to high protein and lipid contents. Therefore, we classified each "bioresource category and technology conversion pathway combination" into three categories according to the relevance for anaerobic digestion and gasification: desirable, acceptable and unacceptable. The first step accounts for moisture content: gasification was considered desirable for bioresources with moisture contents lower than 35%, and anaerobic digestion was considered desirable for bioresources with moisture contents higher than 70%. If the moisture content was between 35 and 70% (±5%), the bioresources were classified as acceptable or unacceptable, depending on outcome of the second step. The second step accounts for the VS content as lignin, ligno-cellulose, lipids, and proteins: Bioresources were considered acceptable for anaerobic digestion when i) the lignin content was low, ii) the sum of lignin and cellulose content was low, and iii) the proteins and lipids content was high, while acceptable for gasification when i) the lignin content was high, and iii) the sum of lignin and cellulose content was high. Otherwise, the bioresources were considered unacceptable for either technology.

2.4. Step 4: Process-oriented inventory modelling and system boundary definition

System boundaries in the LCA scenarios are shown in Fig. 2 and consist of the following: the main foreground processes involved for each scenario (in light blue) and the counterfactuals (in dotted lines). These CH_4 production scenarios are compared with the reference product, namely natural gas for power production (in grey). The main foreground processes include three CH_4 production pathways: i) Gasification with C-to-CH_4 upgrading (GA), ii) anaerobic digestion with hydrogen upgrading (ADH), and iii) anaerobic digestion with water scrubbing upgrading (AD).

Counterfactuals refer to the current management of the residual bioresources associated with each selected technology conversion pathway. As shown in Fig. 3, a bioresource category may have several counterfactuals, representing the range of managing pathways for the individual residual bioresources within the category. For example, the category "agrofood residues" comprises four residual bioresources: butchery meat, canned fruit and vegetables, cheese products and spoiled wine (Table S1, ESI1). The counterfactuals associated with these four residual bioresources are incineration with combined heat and power (CHP) production for both butchery meat and canned fruit and vegetables (thereby avoiding the production of marginal heat and power), while cheese products are used as feed (thereby preventing the use of conventional ingredients and their associated land use change, LUC, assessed based on the methodology presented in Tonini et al. [65], and spoiled wine is used for vinegar (acetic acid) production (thereby preventing marginal acetic acid production). The LCA system boundary considered for the counterfactuals of all bioresources is illustrated in Fig. 3, with additional details provided in Table S21, ESI1. In addition, the life cycle inventory of each counterfactual process is presented in Section S9 to S17, ESI1. For all bioresources spread on soil to decay, or applied to land as organic fertiliser, we followed the findings from Pehme et al. [66] suggesting that 10% of the C in the biomass remains in the soil as sequestered carbon (within 100 years), thereby not leading to CO_2 emissions within the same period. For compost and digestate used as organic fertilisers, we considered a soil carbon sequestration of 11.3%, based on Hansen et al. [67].

2.5. Step 5: Interpretation of technology performance, tier 1 results

Step 5 evaluates the environmental performance of technology conversion pathways for each bioresource (independently) relative to the production, injection and use of 1 Nm^3 CH_4^{-1} (for electricity production), here referred to as "tier 1 results." As a technology-by-technology comparison, this step provides the potential of each bioresource and technology conversion pathway to contribute with net environmental benefits, as well as a basis for identification of process hotspots and carrying out an environmental contribution analysis, following the principles of e.g., Albizzati et al. [68], Tonini et al. [69]. The net environmental balance (NB) corresponds to the difference between the emissions of the selected bioresource-technology conversion pathway and the associated counterfactuals for the bioresources. The number of LCA scenarios assessed in step 5 are given by the number of selected technology pathways times the number of bioresource categories following the individual pathways (in this study 21 scenarios). Finally, tier 1 results are used to identify the most promising conversion pathways and evaluate the importance of technology data for the results.

2.6. Step 6: Interpretation of regional scenarios, tier 2 results

Extending tier 1 results, step 6 evaluates selected combinations of bioresource category and technology conversion pathways at system-level reflecting the regional conditions (here referred to as "tier 2 results"). While all impact categories included in the EF LCIA methodology are quantified (results provided in Section S20, ESI1), only climate change impacts are discussed in detail, reflecting the considerable focus on $CO_{2\text{-eq}}$ emissions for regional decisions on the future developments in energy supply (e.g., European Green Deal [1], the REPOS goal of Occitania to become the first European region fully based on renewable energy [20]). Consequently, the system-level scenarios in step 6 includes only technology conversion pathways with climate change impact NB's lower than the reference product (i.e. with potential for net climate benefits). Tier 2 results represent two scenarios for potential regional bio-based supply of 17.5 TWh y^{-1} of CH_4, with low-as-possible GWP_{100} emissions and fossil CH_4 consumption. While these regional scenarios do not represent an exhaustive list of all possible combinations nor the result of formal optimisation, two main criteria were applied for selection of relevant bioresource-technology pathways from step 5: i) pathways providing the largest regional production of bio-based CH_4 (as TWh·y^{-1}), and ii) pathways providing the lowest regional GWP_{100} emissions (as Mt $CO_{2\text{-eq}}$·y^{-1}). Two overall regional scenarios, representing combinations of selected technology pathway for the individual bioresource categories, are provided in step 6: one for each criterion.

Table 2
Amount of biomass expressed in kg wet weight (ww) required to produce 1 Nm^3 of CH_4 with the characteristics of being injected and distributed into the gas grid, for each bioresource category and technology conversion pathway.

kg_{ww} in input to produce 1 Nm^3 of CH_4			
Category	**AD**	**ADH**	**GA**
Crop residues	7.72	3.46	1.32
Pruning residues	–	–	1.38
Green waste	12.5	5.62	1.79
Manure	22.9	10.27	–
Intercrops	18.2	8.16	3.05
Forest residues	–	–	1.16
Wood waste and industrial end of life wood	–	–	1.00
Agrofood residues	25.6	11.49	–
Biowaste	8.34	3.74	0.842
Sludge	54.2	24.31	–

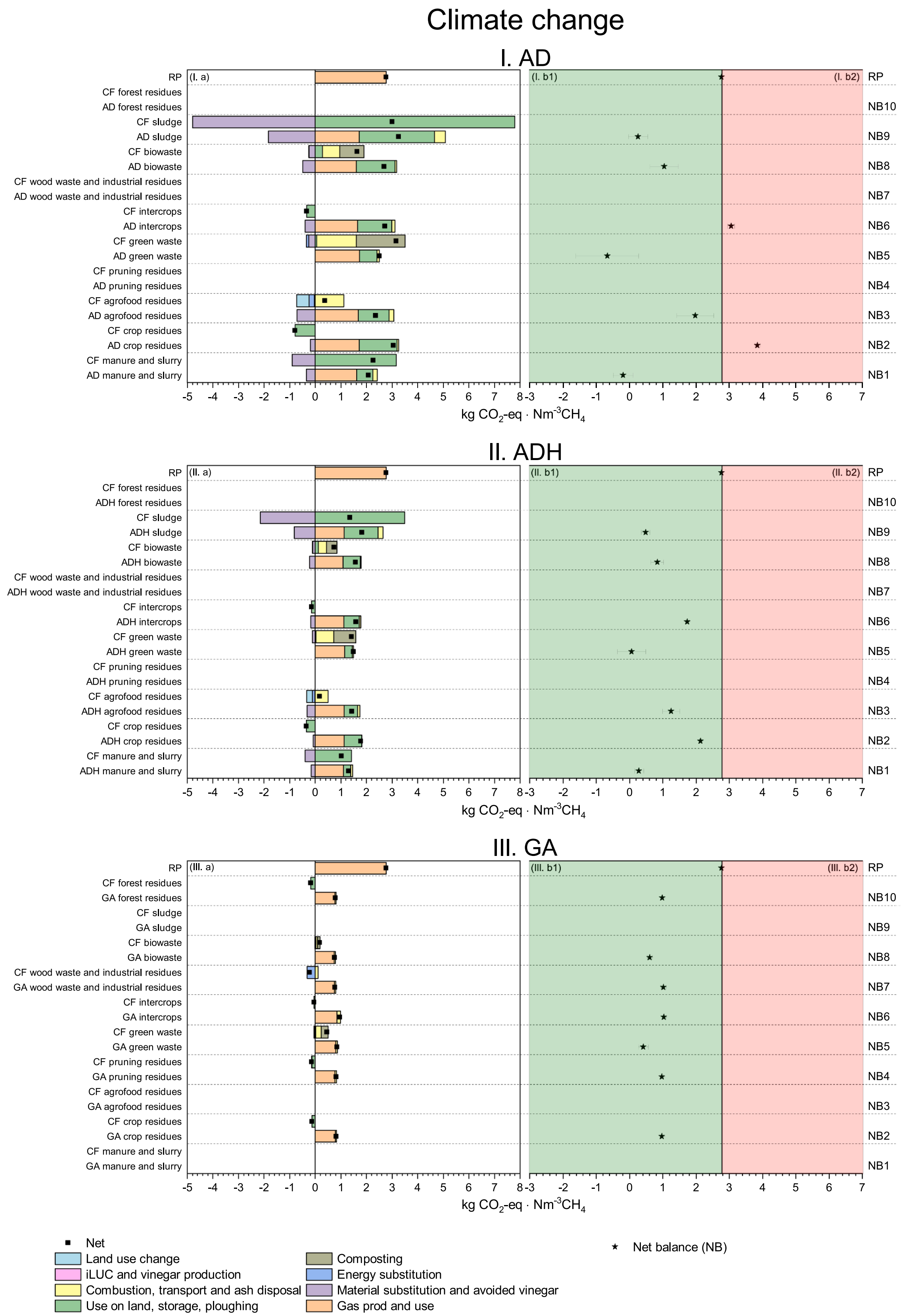
3. Results and discussion

3.1. Allocation of bioresources to individual technology pathways

Moisture and biochemical properties content for each bioresource category were calculated as the weighted average of the included bioresources (see Table 1). The bioresource categories manure, agrofood residues, intercrops and sludge were allocated to AD/ADH, while crop residues, forest residues and wood waste/industrial end-of-life wood were allocated to GA, based on their moisture contents (Table 1). Some categories had a moisture content beyond the desirable range of both AD/ADH and GA (Table 1), namely: green waste, biowaste and pruning residues. Given their relatively high lignin content (16%$_{TS}$), and ligno-cellulosic content (36%$_{TS}$), pruning residues were acceptable for GA only. Similarly, biowaste was evaluated for both pathways: for GA due to their lignin (17%$_{TS}$) and ligno-cellulosic content (23%$_{TS}$), and for AD/ADH based on the protein (15%$_{TS}$) and lipid (12%$_{TS}$) contents (Table 1). The same applied to green waste being acceptable for AD/ADH due to the low lignin content (5%$_{TS}$), while the ligno-cellulosic content did not make green waste unacceptable for GA (Table 1). Two bioresource categories, namely crop residues and intercrops, were found acceptable for both GA and AD/ADH with moisture contents of 34%$_{ww}$ and 74%$_{ww}$, respectively, also in view of the lignin (5%$_{TS}$, crop residues) and ligno-cellulosic centents (44%$_{TS}$, intercrops). The results for each bioresource are illustrated in Table 1.

3.2. Hydrocarbon to CH_4 conversion efficiencies (tier 1)

Gasification required the least amount of residual bioresources (in kg_{ww}) to produce 1 Nm^3 of gas (Table 2). This can also be visualised at the resource level; for instance, green waste generates about six times more CH_4 per wet tonne if gasified compared to anaerobic digestion. GA provides the highest C-to-CH_4 conversion, which is also in line with recent studies (e.g., Ardolino and Arena, [15] Le Quéré, [70]), while for



(caption on next page)

Fig. 4. Results for the climate change impact category associated with the ten bioresidue categories treated in three different CH_4 production pathways (*AD,* anaerobic digestion with water scrubbing upgrading; *ADH*, anaerobic digestion with hydrogen upgrading;.*GA,* gasification with C-to-CH4 upgrading). The left-hand side of the graph (*I. a*, *II. a*, *III. a*) shows the contribution to the net results by the three individual pathways and counterfactuals (*CF*), while the right-hand side (*I. b1 - b2*, *II. b1 – b2*, *III. b1 – b2*) displays the net balance (*NB*) calculated for each scenario. RP is the reference product. The values in the green area (*b1*) have lower impacts than the reference product, while vice versa applies for those values found in the red area (*b2*). With respect to the legend "Land use change," savings from avoided land use changes; "Composting" emissions from composting process, use on-land, and combustion of rejects from screening; "iLUC and vinegar production" indirect land use changes, as well as production of vinegar from spoiled wine; "Energy substitution" from substituting electricity and heat; "Combustion, transport and ash disposal" transportation, combustion and landfill disposal considered during the pathway and counterfactuals; "Material substitution and avoided vinegar" avoided vinegar production, intercrops production and all the mineral fertilisers when use on land is considered; "Use on land, storage and ploughing" digestate/compost use on land, ploughing on fields and the storage of digestate, manure and slurry; "Gas prod and supply" production of methane from the conversion pathways considered in this study, as well as injection into the gas grid and the combustion of this gas. "Gas prod and use" natural gas production, injection into the grid and its combustion, representing the reference product. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

anaerobic digestion a portion of the carbon remains in the digestate. This digestate, however, may be dried and gasified if sufficiently rich in fibres, thus boosting total CH_4 recovery (or directly combusted, though not considered here). ADH provides more CH_4 than AD per unit of biomass input (about45%). As a result, and as shown in Table 2, the amount of bioresource required for 1 Nm^3 CH_4 varies for the individual bioresource and technology conversion pathway.

3.3. LCA contribution analysis for each technology pathway (tier 1)

For each scenario, tier 1 results are shown in Fig. 4 for the climate change impact category (GWP_{100}). Results for all remaining impact categories are shown in Section S19 of ESI1. Positive (i.e. above zero) bars indicate burdens, while negative bars are savings, with the difference between both representing the net GWP_{100}. The GWP_{100} impact of the reference product (RP; natural gas extraction and combustion for power production) is shown to highlight how the CH_4 produced from each bioresource category and technology conversion pathway combination compares. On the left-hand side of Fig. 4, two bars are shown for each bioresource category: a first one (CF) representing the impact breakdown of the counterfactual management of the bioresource, and a second one (e.g., AD) representing the CH_4 conversion pathway.

Note that the impacts associated with the individual bioresource counterfactuals are the same for all pathways: AD, ADH and GA (in Fig. 4, these counterfactuals have different magnitude as they are normalized to the Nm^3 CH_4 produced by each conversion pathway, detailed in Table 2).

For the AD conversion pathway, only two bioresource categories had higher net GWP_{100} emissions than the RP, namely intercrops and crop residues (right hand-side of Fig. 4). Intercrops and crop residues were also the only two bioresource categories providing net negative impacts for the counterfactuals (in particular due to the 10% carbon sequestration within 100 years), meaning that only conversion pathways with net negative emissions (greater than those associated with the counterfactual) could render the conversion attractive for these bioresource. This applies for both AD and ADH. For ADH and GA, all of the suitable bioresources provided lower net GWP_{100} emissions per unit of bio-based CH_4 than the fossil reference. The results illustrate two main drawbacks for AD and ADH in comparison with GA: i) the GHG emissions (N_2O and CH_4) associated with digestate storage and spreading are often critical, albeit the emissions from storage may be limited through use of waterproofed covers (which is in fact required by a recent French law), [71–72] and ii) the larger amounts of bioresources needed to produce 1 Nm^3 of bio-based CH_4 increase the impacts from both counterfactuals and conversion processes. Consequently, only bioresource-technology combinations for which emissions from the counterfactuals are larger than those associated with the bioresource conversion itself should be preferred for AD/ADH over GA (and AD over ADH). Fig. 4 illustrates that this is the case for manure (AD) and green waste (AD). For AD and ADH, mineral fertiliser substitution was found to be the most important contributor to negative emissions for most bioresource-technology combinations.

Based on the net balance GWP_{100} emissions, the tier 1 results suggest that intercrops and crop residues should not be pursued for AD and thus excluded from the system-level scenarios in step 6.

3.4. Critical parameters and uncertainty (tier 1)

For each impact category, the contributions of a total of 507 parameters to the uncertainty of the net result were estimated. Parameters contributing the most to uncertainty in climate change impact for each bioresource category and each conversion scenario are shown in Fig. 5. In each extended pie chart, we display only the parameters whose sum explains at least 80% of the uncertainty. Overall, ten parameters were identified as particularly critical for the tier 1 climate change impact results: 1) *Water_cont*, which is the required water content of the anaerobic digestion tank (set to 90% [88.2–91.8]) [62]. Its contribution to the uncertainty of the net results for AD/ADH ranged from 26% (agrofood residues) to 96.6% (crop residues). 2) *Transport_compost*, which in the counterfactuals represents the transport distance from bioresource collection to the composting plant (set to 30 km [10–50]). This parameter contributed from 7.8% (biowaste) to 37% (green waste) for GA, and from 21.2% (biowaste) to 32.9% (green waste) for AD/ADH. 3) *Transp_comb*, which represents the transportation distance for ashes from combustion of compost rejects to landfill (for both counterfactuals and digested compost; set to 30 km [10–150]) [68]. Contributions ranged from 5.2% (biowaste, AD/ADH) to about 52% (agrofood residues, AD/ADH). 4) *Xsi_1*, the stoichiometric coefficient controlling CO-to-CH_4 conversion in the GA upgrading model (methanation reaction; Eq.8; values in Table S18, ESI1). Contributions to the uncertainty were 20–65% depending on the gasified bioresource category (details in ESI1). 5) *Xsi_2*, the stoichiometric coefficient controlling CO_2-to-CH_4 conversion in the GA upgrading model (Sabatier reaction, Eq.9; values in Table S18, ESI1). Contributions to uncertainty were 5.4–11.5% depending on the gasified bioresource category (details in ESI1). 6) *Distr_TS*, which defines the distribution of total solids (TS) to compost and (100 - Distr_TS) transfers TS to rejects (set to 95% to compost [76–95%]). Contributions to the uncertainties ranged between 5.2% (biowaste, GA) and 20.8 (green waste, AD). 7) *C_field_emission*, which is the transfer coefficient of carbon for use on field of manure and slurry, set to 90%, the rest 10% of initial carbon is considered sequestered (after 100 year). This parameter was relevant for manure in both AD and ADH, with a contribution of about 43%. 8) *Fert_sub_N*, when digestate or residual bioresources are used-on-land and substitute nitrogen mineral fertilizer (set to 48% [0.384–9.576]) [47]. It was about 20% for both sludge AD/ADH. 9) *NG_grid_distr_loss*, which represents losses of upgraded bio-based CH_4 during/after injection into the natural gas grid (1% [0.8–1.2]). For AD/ADH, the contribution to uncertainty ranged from 0.3 to 1.4% and for GA 4.4–16.3% (respectively green waste and pruning residues). 10) *MCF,* which refers to the temperature- and management-dependent methane conversion factor, calculated based on IPCC 2019 guidelines for the storage of organic fractions. For Occitania the MCF was 36% [32.4–39.6] (Section S4, ESI1), contributing from about 1% (biowaste and intercrop) to 20% (agrofood residues) for AD/

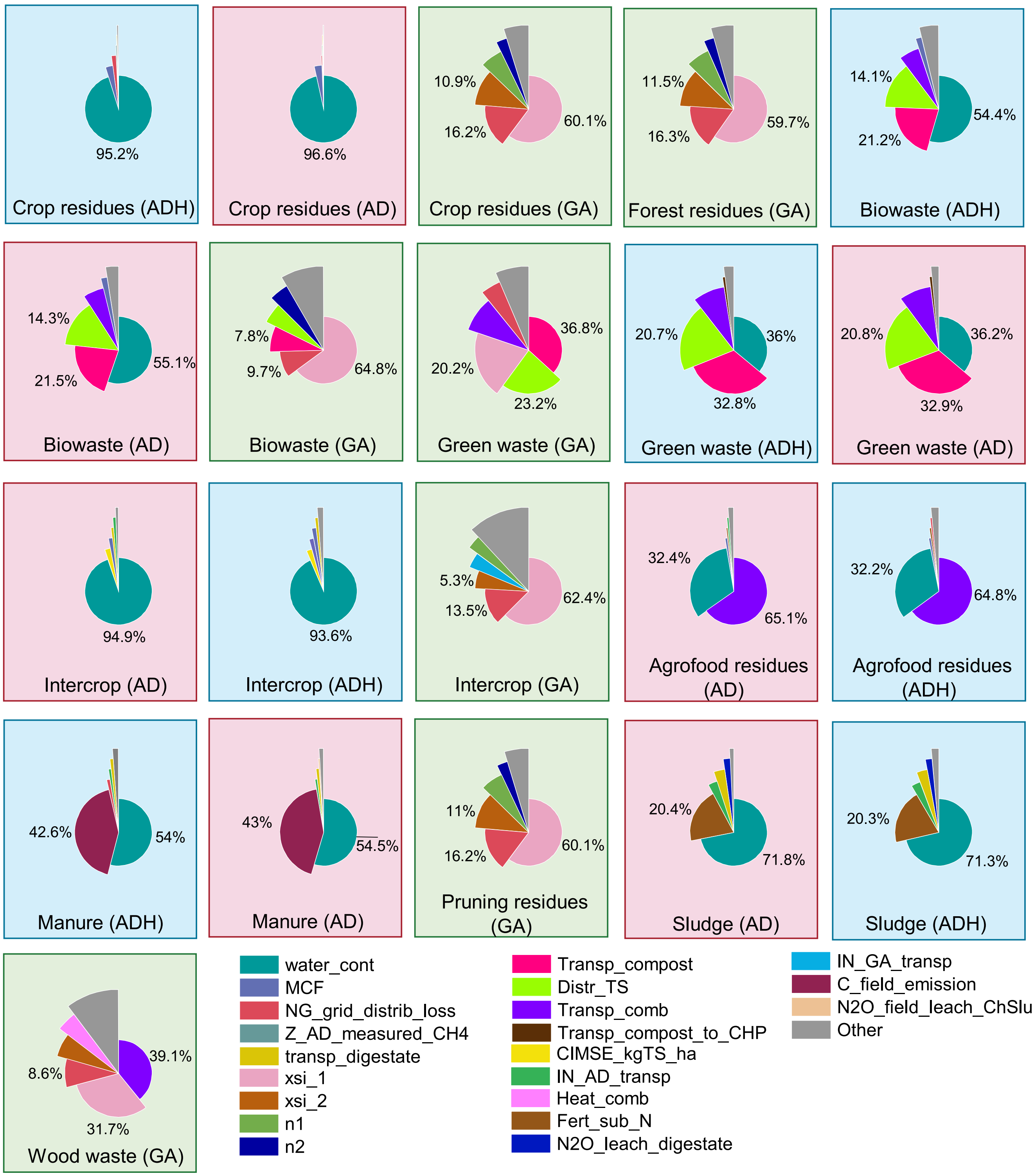


Fig. 5. Extended pie chart displaying parameters contributing the most to uncertainty around the net result for climate change as an impact category, for each biomass category and conversion pathway: AD (light red background), ADH (light blue background) and GA (light green background). *Other* is the complement to 100%. The parameters are explained in Section 3.4. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

ADH. Other parameters represented in Fig. 5 are: *IN_AD_transp*: transportation from the pre-treatment facility to the anaerobic digestion plant; *transp_digestate*: transportation of digestate to the field; *transp_compost_to_CHP*: transportation of compost rejects to a combined heat and power (CHP) plant; *IN_GA_transp*: transportation of residual bio-resources from the pre-treatment to the gasification plant; *CIMSE_kg_TS_ha*: intercrops yield dry basis during harvest; *N2O_leach_digestate*: N_2O emissions from digestate leaching; *Heat_comb*: heat efficiency in the considered CHP plant; *Z_AD_measured_CH4*: methane measured in the biogas; N2O_field_leach_ChSlu: Nitrous oxide leaching after field application of chicken slurry; n1: stoichiometric coefficient for H2 formation in the conversion of biomass to syngas (Eq. S1, ESI);

Table 3
Tier 2 results providing overview of net CH_4 production and net GWP_{100} impacts associated with conversion of the individual bioresource categories via each of the three technology pathways: anaerobic digestion with hydrogen enhancement (ADH), anaerobic digestion with water scrubbing (AD), and gasification with C-to-CH_4 upgrading (GA). For each bioresource, the highest yields of bio-based CH_4 are indicated bold, and the lowest global warming potential in italics (Sd: standard deviation).

Scenario	Selection criteria for tier 2	Crop residues	Pruning residues	Green waste	Manure	Intercrops	Forest residues	Wood waste/ industrial end of life wood	Agrofood residues	Biowaste	Sludge
ADH	CH_4 in grid (Nm^3)	$2.31{\cdot}10^9$	–	$5.65{\cdot}10^9$	**$1.33{\cdot}10^9$**	$1.15{\cdot}10^9$	–	–	**$5.61{\cdot}10^7$**	$1.98{\cdot}10^8$	**$2.65{\cdot}10^7$**
	Sd	$\pm 2.81{\cdot}0^7$	–	$\pm 6.86{\cdot}10^5$	**$\pm 1.63{\cdot}10^7$**	$\pm 1.40{\cdot}10^7$	–	–	**$\pm 6.83{\cdot}10^5$**	$\pm 2.42{\cdot}10^6$	**$\pm 3.21{\cdot}10^5$**
	Net GWP_{100} (Mt $CO_{2\text{-eq}}$)	*4.94*	–	0.00390	0.386	*1.99*	–	–	0.0704	0.166	0.0129
	Sd	±*0.09030*	–	±0.0240	±0.186	±*0.0553*	–	–	±0.0141	±0.0378	±0.00347
AD	CH_4 in grid (Nm^3)	–	–	$2.53{\cdot}10^7$	$5.97{\cdot}10^8$	–	–	–	$2.51{\cdot}10^7$	$8.90{\cdot}10^7$	$1.19{\cdot}10^7$
	Sd	–	–	$\pm 2.18{\cdot}10^4$	$\pm 5.15{\cdot}10^5$	–	–	–	$\pm 2.16{\cdot}10^4$	$\pm 7.56{\cdot}10^4$	$\pm 1.02{\cdot}10^4$
	Net GWP_{100} (Mt $CO_{2\text{-eq}}$)	–	–	*−0.0168*	*−0.113*	–	–	–	*0.0498*	*0.0642*	*0.00305*
	Sd	–	–	±*0.0240*	±*0.183*	–	–	–	±*0.0140*	±*0.0373*	±*0.00347*
GA	CH_4 in grid (Nm^3)	**$6.08{\cdot}10^9$**	**$1.06{\cdot}10^8$**	**$1.78{\cdot}10^8$**	–	**$3.07{\cdot}10^9$**	**$3.10{\cdot}10^7$**	**$8.44 \cdot 10^8$**	–	**$8.90{\cdot}10^8$**	–
	Sd	**$\pm 3.61{\cdot}10^8$**	**$\pm 6.38{\cdot}10^6$**	**$\pm 1.10{\cdot}10^7$**	–	**$\pm 1.90{\cdot}10^8$**	**$\pm 1.83{\cdot}10^6$**	**$\pm 5.00 \cdot 10^7$**	–	**$\pm 6.41{\cdot}10^7$**	–
	Net GWP_{100} (Mt $CO_{2\text{-eq}}$)	5.899	*0.103*	0.0738	–	3.19	*0.0306*	*0.864*	–	0.546	–
	Sd	±0.288	±*0.00498*	±0.0257	–	±0.160	±*0.147*	±*0.061*	–	±0.0615	–

n2: stoichiometric coefficient for CO formation in the conversion of biomass to syngas (Eq. S1, ESI). All details related to the uncertainty analysis can be found in ESI1 (Table S33 to S53), as well as for the remaining impact categories.

3.5. System analysis for regional gas demand (tier 2)

Table 3 provides an overview of net bio-based CH_4 production and GWP_{100} impacts for the individual bioresource categories and all three technology conversion pathways, representing the entire potential of each bioresource in Occitania. For each bioresource category, the technology pathways offering both the highest bio-based CH_4 production (bold) and the lowest GWP_{100} impacts (italics) are indicated.

Among all of the bioresource categories, digestion of all manure in Occitania provides both a relatively high CH_4 production and the lowest GWP_{100} impact (Table 3). The high CH_4 production level is essentially due to the large availability of manure in Occitania, which is the most abundant bioresource with a share of about 40% per wet weight. The low GWP_{100} impacts are caused by the non-negligible emissions associated with the counterfactuals (i.e. emissions from raw manure storage and field application), resulting in higher emissions than those associated with the AD/ADH technology conversion pathways, as also observed in several earlier studies [73–76]. For many of the bioresources (Table 3), gasification offers the highest CH_4 production. This is due to the higher production of bio-based CH_4 achievable through the gasification process compared to anaerobic digestion. Both AD and ADH are considered for green waste, biowaste, manure, and sludge. Table 3 highlights that for all these streams, more bio-based CH_4 is produced through ADH, but at the expense of larger GWP_{100} emissions. Crop residues and intercrops are the bioresource categories contributing the most to bio-based CH_4 supply, in particularly via gasification. However, the use of crop residues as input resource can induce competition among several technologies, for example bioethanol, biodiesel, bioplastic, and bioenergy production [77–81]. On the other hand, intercrops represent an unexploited resource that is not yet used for bio-based CH_4 production. In agreement with Slomka and Oliveira, [82] this study demonstrates the potential of intercrops for renewable bioenergy and points out that more focus on intercrops is needed.

Based on a contribution analysis (Table 4) of the bio-based CH_4 production and use via the gas grid, it was observed that "CH_4 distribution losses in the natural gas grid" and "combustion of bio-based CH_4" accounted for the highest contributions to GWP_{100} emissions, corresponding to about 30% and 60% of the total emissions within each technology conversion pathway. The distribution losses should be minimised, since CH_4 has a stronger global warming potential than CO_2 [16,77]. For AD, the process having the lowest emissions was "losses from heat and pipeline infrastructure for gas distribution" with about 1% of the total emissions within AD. For ADH, "bio-based CH_4 production" provided net savings, mainly due to the 10% of heat recovered in district heating and the substitution of marginal heat. For GA, "H_2 production" represented the lowest emissions mainly due to the savings from the 14.6% of heat recovery for district heating and the substitution of marginal heat. These results are conditional to the relatively high marginal energy mix in renewables considered herein. On this purpose, to give an impression of the difference it would make if hydrogen was produced with an energy mix involving more fossil carbon, natural gas was considered for heat, and as a representative, the Malaysian electricity mix (57% natural gas, 17% lignite, 15% hydro, 7% nuclear, 2% geothermal, 2% wood) was considered for electricity. Therefore, 1 Nm^3 of H_2 produces 0.954 $kgCO_{2\text{-eq}}$ with the Malaysian electricity mix and 0.131 $kgCO_{2\text{-eq}}$ with the electricity considered in this study. While, net negative emissions from heat recovery were 0.103 $kgCO_{2\text{-eq}}$ with the heat from natural gas and 0.0209 $kgCO_{2\text{-eq}}$ with the heat considered herein. Consequently, it is relevant to ensure that H_2 is produced from decarbonized electricity.

In Table 4, *total AD/ADH/GA* corresponds to the GWP_{100} emissions given by the sum of the process contributions for each of the three technology conversion pathways, without including counterfactuals, pre-treatment and transport to the anaerobic digestion and gasification plants. *Net balance AD/ADH/GA scenario* represents the net GWP_{100}, including the counterfactuals and the technology conversion pathways.

3.6. Combinations of technology conversion pathways and comparison with regional gas demand (tier 2)

Two overall sets of results combining bioresource categories and technology conversion pathways are provided: i) pathway selection prioritising the highest production and use in the natural gas grid (combustion) of bio-based CH_4 (Table 5, *Combination of technologies with* max *bio-based CH_4*), and ii) pathway selection prioritising the lowest GWP_{100} emissions (Table 5, *Combination of technologies with* min *GWP_{100}*).

Table 4
Global warming potential breakdown by activity in the bio-based methane supply chain (excluding impacts from counterfactual, pre-treatment, and transport to the anaerobic digestion and gasification plants). Absolute net results, per bioresource category, for all conversion pathways (total $CO_{2\text{-eq}}$ including all impacts throughout the full bioresource pathway) (numbers are rounded).

Conversion pathway	Process	Crop residues	Pruning residues	Green waste	Manure	Intercrops	Forest residues	Wood waste/ industrial and end of life	Agrofood residues	Biowaste	Sludge
AD	Bio-CH_4 production (water scrubbing)	–	–	0.0020	0.047	–	–	–	0.0020	0.0070	$9.4{\cdot}10^{-4}$
(Mt CO_{2_eq})	Losses from heat and pipeline infrastructure for gas distribution	–	–	$2.5{\cdot}10^{-4}$	0.0058	–	–	–	$2.5{\cdot}10^{-4}$	0.0087	$1.2{\cdot}10^{-4}$
	CH_4 distribution losses in natural gas grid	–	–	0.0062	0.15	–	–	–	0.0062	0.022	0.0029
	Combustion of bio-CH_4	–	–	0.012	0.27	–	–	–	0.012	0.0041	0.0054
	Total AD	**–**	**–**	**0.020**	**0.47**	**–**	**–**	**–**	**0.020**	**0.071**	**0.0094**
	Net balance AD scenario	**–**	**–**	**−0.017**	**−0.11**	**–**	**–**	**–**	**0.050**	**0.093**	**0.0031**
ADH	H_2 production (with 14.6% of heat recovery in district heating)	0.073	–	0.0018	0.050	0.036	–	–	–	0.0063	$8.3{\cdot}10^{-4}$
(Mt CO_{2_eq})	Bio-CH_4 production (with 10% of heat recovery)	$-9.3{\cdot}10^{-12}$	–	−0.0012	−0.029	−0.025	–	–	–	−0.0043	$-5.7{\cdot}10^{-4}$
	Losses from heat and pipeline infrastructure for gas distribution	0.023	–	$5.5{\cdot}10^{-4}$	0.013	0.011	–	–	–	0.0019	$2.6{\cdot}10^{-4}$
	CH_4 distribution losses in natural gas grid	0.57	–	0.014	0.33	0.28	–	–	–	0.049	0.0065
	Combustion of bio-CH_4	1.1	–	0.026	0.61	0.53	–	–	–	0.091	0.012
	Total ADH	**1.7**	**–**	**0.041**	**0.97**	**0.83**	**–**	**–**	**–**	**0.14**	**0.019**
	Net balance ADH scenario	**4.9**	**–**	**−0.039**	**−0.39**	**2.0**	**–**	**–**	**–**	**0.17**	**0.013**
GA	H_2 production (with 14.6% of heat recovery in district heating)	$3.2{\cdot}10^{-4}$	$5.8{\cdot}10^{-6}$	$1.3{\cdot}10^{-5}$	–	$3.8{\cdot}10^{-4}$	$1.4{\cdot}10^{-6}$	$3.3{\cdot}10^{-5}$	–	$4.9{\cdot}10^{-10}$	–
(Mt CO_{2_eq})	Bio-CH_4 production	0.011	$2.0{\cdot}10^{-4}$	$4.5{\cdot}10^{-4}$	–	0.013	$4.9{\cdot}10^{-5}$	0.0012	–	$2.1{\cdot}10^{-8}$	–
	Losses from heat and pipeline infrastructure for gas distribution	0.059	0.0010	0.0017	–	0.030	$3.0{\cdot}10^{-4}$	0.0082	–	$1.6{\cdot}10^{-7}$	–
	CH4 distribution losses in natural gas grid	1.5	0.026	0.044	–	0.75	0.0076	0.21	–	$4.1{\cdot}10^{-6}$	–
	Combustion of bio-CH_4	2.8	0.049	0.081	–	1.4	0.014	0.39	–	$7.6{\cdot}10^{-6}$	–
	Total GA	**4.4**	**0.076**	**0.13**	**–**	**2.2**	**0.022**	**0.60**	**–**	**$1.2{\cdot}10^{-5}$**	**–**
	Net balance GA scenario	**5.9**	**0.10**	**−0.074**	**–**	**3.2**	**0.031**	**0.86**	**–**	**0.55**	**–**

The results for both of the two combinations not only fulfilled Occitania's gas demand but actually exceeded this by about 116 and 37.5 TWh CH_4, corresponding to about 7.6 and 3.1 times the current demand, respectively. Assuming that all surplus CH_4 is exported, and that it leads to additional substitution of power from natural gas, the total savings in GWP_{100} emissions are 30.4 and 9.81 Mt $CO_{2\text{-eq}}$ for two scenarios in Table 5, respectively. By moving from the second set of combinations, which provides lower climate impacts, to the first one, with higher bio-based methane supply, Occitania can achieve additional 78.7 TWh CH_4 at the "cost" of 3.22 $MtCO_{2\text{-eq}}$ locally, while in turn avoiding an additional 20.6 $MtCO_{2\text{-eq}}$ from natural gas substitution outside the region. Consequently, the net difference between these scenarios when natural gas substitution is considered is the avoidance of 17.4 $MtCO_{2\text{-eq}}$, equivalent to annual emissions from 2.68 M French citizens (4.0% of population) [83–84] This demonstrates a considerable potential for improving the climate performance of bio-based CH_4 from the Occitania region.

The net balances for all the midpoint impact categories considered are presented in Table 6, for the two combinations of technologies. For both combinations, particulate matter and acidificationpresent net

Table 5
Scenarios of technologies selected based on two criteria: i) highest bio-based CH_4 produced and injected (first row); ii) lowest global warming potential (second row). In bold, the sum of bio-based CH_4 achieved, and the sum of the carbon dioxide emitted, both obtained from the selected conversion pathways. In italics, the avoided gas production elsewhere, considering Occitania's gas demand of 17.5 TWh y^{-1} and the associated $CO_{2\text{-eq}}$ emissions (numbers are rounded).

	Bioresource category	Conversion pathway	CH_4 in grid (TWh)	Net balance (Mt $CO_{2\text{-eq}}$)
Combination of technologies with max bio-based CH_4	Crop residues	GA	64.6	5.90
	Pruning residues	GA	1.12	0.103
	Green waste	GA	1.88	0.0738
	Manure	ADH	14.1	0.386
	Intercrops	GA	32.5	3.19
	Forest residues	GA	0.328	0.0306
	Wood waste/ industrial and end of life	GA	8.94	0.864
	Agrofood residues	ADH	0.594	0.0704
	Biowaste	GA	9.44	0.546
	Sludge	ADH	0.281	0.0129
	Total, bio-based CH_4 production and GWP_{100} emissions		**134**	**11.2**
	Avoided CH_4 gas production elsewhere and associated GWP_{100} emissions from the surplus		*116*	30.4
Combination of technologies with min GWP_{100}	Crop residues	ADH	24.5	4.94
	Pruning residues	GA	1.12	0.103
	Green waste	AD	0.270	−0.0168
	Manure	AD	6.33	−0.113
	Intercrops	ADH	12.1	1.99
	Forest residues	GA	0.328	0.0306
	Wood waste/ industrial and end of life	GA	8.94	0.864
	Agrofood residues	AD	0.267	0.0498
	Biowaste	AD	0.943	0.0934
	Sludge	AD	0.130	−0.00305
	Total, bio-based CH_4 production and GWP_{100} emissions		**55.0**	**7.95**
	Avoided CH_4 gas production elsewhere and associated GWP_{100} emissions from the surplus		*37.5*	9.81

negative emissions. While terrestrial eutrophication has net negative emissions only in the first combination, freshwater eutrophication presents net negative emissions only in the second combination. The consequences for emissions for all impact categories by "moving" from the "max bio-based CH_4" scenario towards the "min GWP_{100}" scenario, is provided in Table 7. This indicates that all impacts, with the exception of terrestrial eutrophication and water use, follow the direction of climate change, with decreases observed for all but these two impacts. However, if accounting for the "loss" of the 78.7 TWh CH_4 and the natural gas it would have substituted, it shows that for six out of the sixteen impacts studied, the "min GWP_{100}" scenario is worth the sacrifice of these TWh. Climate change is one of these six impact categories together with ozone depletion, acidification, terrestrial and marine eutrophication, and water use.

4. Future perspectives

For the Occitania region, this study clearly demonstrates that while bio-based CH_4 currently remains under-exploited, considerable production increases may be feasible based on available bioresources, existing conversion technologies, and gas grid infrastructures [79]. Regions such as Occitania may be self-sufficient with bio-based CH_4 by

Table 7
Net balance results by "moving" from the "max bio-based CH_4" scenario towards the "min GWP_{100}" scenario (this implies that 78.34 TWh less are produced). In the last row, a positive sign implies net additional emissions from the move, while a negative sign implies improvements (numbers are rounded).

	Climatechange($MtCO_{2-eq}$)	Ozonedepletion($kgCFC-11_{-eq}$)	Humantoxicity, cancer(kgN_{-eq})	Humantoxicity, non − carcinogenic(CTUh)	Particulatematter(diseaseincidences)	Ionisingradiation($MtBqU-235_{-eq}$)	Photochemicalozoneformation(10^9molH+_{-eq})	Acidification(10^9molN_{-eq})
Difference of emissions (*Emissions_max_bioCH₄ - Emissions_min_GWP₁₀₀*)	3.22	$3.16 \cdot 10^4$	21.0	4239	53.2	0.115	0.0416	0.00622
Emissions credit of 78.34 TWh from the reference product (natural gas) no longer happening	20.6	3538	1.47	21.9	1.47	0.000711	0.0208	0.0158
Net difference when considering the natural gas substitution no longer happening (*Difference of emissions - Emissions_78.3 TWh_natural gas*)	−17.4	−379	19.5	$4.22 \cdot 10^4$	51.8	0.114	0.0208	0.00960

	Terrestrialeutrophication(MtN_{-eq})	Freshwatereutrophication(tP_{-eq})	Marineeutrophication(tN_{-eq})	Freshwaterecotoxicity(10^9CTUe)	Landuse(−)	Wateruse(km^3water_{-eq})	Resourceuse, minerals($kgSB_{-eq}$)	Resourceuse, energycarrier(PJ)
Difference of emissions (*Emissions_max_bioCH₄ - Emissions_min_GWP₁₀₀*)	−0.146	$1.61 \cdot 10^4$	$3.52 \cdot 10^4$	187	$8.52 \cdot 10^{10}$	−0.196	$6.57 \cdot 10^4$	342
Emissions credit of 78.34 TWh from the reference product (natural gas) no longer happening	0.0623	20.09	$3.71 \cdot 10^6$	21.1	$-1.40 \cdot 10^9$	1.27	$8.71 \cdot 10^3$	324
Net difference when considering the natural gas substitution no longer happening (*Difference of emissions - Emissions_78.3 TWh_natural gas*)	-0.208	$1.59 \cdot 10^4$	$-5.68 \cdot 10^6$	−166	$8.38 \cdot 10^{10}$	−1.47	$5.69 \cdot 10^4$	18

utilising residual bioresources, thereby avoiding natural gas import and even supplying bio- based gas for neighboring regions. A deeper analysis of individual bioresources, e.g., as illustrated by the profound potential of intercrops for contributing to bio-based CH_4, shows that potential utilisation of these bioresources is often underestimated. As intercrops are typically harvested only if the yield exceeds a minimum threshold, for Occitania suggested to 4 t_{TS} (ha y)$^{-1}$, [37] intercrops may represent an important opportunity for local gas supply.

The continuous expansion of bio-based CH_4 injection into the natural gas grid [85] is supported by further development of the market for bio-based CH_4 through several European projects (e.g., GreenGasGrid project [86]. Bioresource categories rich in nutrients should be preferred for anaerobic digestion pathways. The reason for this is that nutrients can be recovered from the digestate. In a context of eventual disruption of the fertilizer supply chain (currently highly dependent upon natural gas resources), looping nitrogen and eventually phosphorus in values chains becomes tremendously important. However, regions such as Occitania may consider gasification as a valuable supplement for valorisation of other non-digestible residues and further increase in bio-based CH_4 production (also in agreement with the French eco-industries strategic committee) [87]. On the other hand, from an environmental perspective relatively few in-depth assessments have been carried out focusing on the regional bioresource constraints. For further development of assessment such as this study, consistent technology and full-scale process data, quantification of (fugitive) emissions associated with gasification, and relationships between feedstock properties and process outputs are required. Moreover, we considered one of the highest technology readiness level available for gasification (i.e. the GoBiGas pilot-scale plant), but emerging technologies such as hydrothermal gasification intended to handle substrates with high water content could be added in the next version of this framework [88]. While this study integrated process-oriented LCA modelling and parameterisation beyond previous literature, the modelling may be further expanded through multi-objective mathematical optimisation, as in Vadenbo et al. [89]. The impact coverage may be expanded to include economic aspects (e.g., through life cycle cost modelling as in Albizzati et al. [90]). In Cross, [91] the cost of bio-based CH_4 produced through GA was between five and twelve times more expensive than natural gas, and between two and five times more expensive than CH_4 from landfills. The costs of bio-based CH_4 produced from AD were four times more expensive than natural gas, and less than two times more expensive than landfill CH_4 [90].

5. Conclusions

The study provided a comprehensive tiered framework for assessment of environmental impacts associated with regional supply of bio-based CH_4 for replacement of fossil natural gas through conversion of locally available residual bioresources, thereby contributing to regional renewable energy targets. Implemented for the French region, Occitania, the results demonstrated that utilisation of available residual bioresource has tremendous potential both for covering the current regional gas demands (up to about seven times the gas demand in some scenarios) and for lowering the global warming potential from the region (up to about five times the net GWP_{100} impacts). The tiered assessment framework demonstrated that "process-oriented" LCA modelling of anaerobic digestion, gasification, and gas upgrading technologies can be applied on an actual regional setting while accounting for detailed relationships between bioresource feedstock properties and conversion technology performance, and on this basis offer consistent system-level evaluation of full-scale implementation scenarios for supply of bio-based CH_4. Introducing a tiered approach, the framework provided valuable insights from both technology and system-level perspectives: i) a technology level (tier 1) for evaluation of bioresource-technology performance and identification of critical process parameters in an environmental perspective, and ii) a system-level (tier 2) for identification of relevant bioresource-technology combinations and their potentials for regional supply of renewable energy and environmental benefits. While implemented on a specific regional system, the tiered assessment framework can be expanded and applied on a much wider range of systems and contexts.

CRediT authorship contribution statement

Concetta Lodato: Conceptualization, Formal analysis, Methodology, Investigation, Software, Writing – original draft. **Lorie Hamelin:** Conceptualization, Methodology, Supervision, Validation, Funding acquisition, Writing – review & editing. **Davide Tonini:** Conceptualization, Methodology, Supervision, Validation, Funding acquisition, Writing – review & editing. **Thomas Fruergaard Astrup:** Conceptualization, Methodology, Supervision, Funding acquisition, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

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