Process-oriented life cycle assessment modelling of (bio)energy technologies

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Preface

The work presented in this PhD thesis was carried out at the Department of Environmental Engineering of the Technical University of Denmark under the supervision of Professor Thomas Fruergaard Astrup and the co-supervision of Davide Tonini, from December 2016 to July 2020. The work included a two months stay, July and September 2019, at Toulouse Biotechnology Institute (TBI), INSA Toulouse (France) and collaborations with both the Department of Mathematics and Computer Science and the Department of Chemical Engineering of the Technical University of Denmark.

The thesis is organized in two parts: the first part puts into context the findings of the PhD in an introductive review; the second part consists of the papers listed below. These will be referred to in the text by their paper number written with the Roman numerals **I-IV**.

I Lodato, C., Tonini, D., Damgaard a., Astrup, T. F. (2020) A process-oriented life-cycle assessment (LCA) model for environmental and resource-related technologies (EASETECH). The International Journal of Life Cycle Assessment 73–88. **[Paper I]**

II Lodato, C., Zarrin, B., Damgaard, A., Baumeister, H., Astrup, T. F. (2020) A modelling framework for process-oriented life cycle assessment (EASETECH+). Submitted **[Paper II]**

III Ardolino, F., Lodato, C., Astrup, T. F., Arena, U. (2018) Energy recovery from plastic and biomass waste by means of fluidized bed gasification: A life cycle inventory. Energy, 299-314. **[Paper III]**

IV Lodato, C., Hamelin, L., Tonini, D., Astrup, T. F. (2020) Framework for assessing environmental performance of methane gas supply in the context of local bioeconomy. Manuscript **[Paper IV]**

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In this online version of the thesis, paper **I-IV** are not included but can be obtained from electronic article databases e.g. via www.orbit.dtu.dk or on request from DTU Environment, Technical University of Denmark, Miljoevej, Building 113, 2800 Kgs. Lyngby, Denmark, info@env.dtu.dk.

In addition, the following publications, not included in this thesis, were also concluded during this PhD study:

- Lodato, C., Tonini, D., Astrup, T. F. (2017) An Advanced LCA-model targeted to bioenergy systems and technologies: Recent developments of the EASETECH LCA-model. *Abstract from 25th European Biomass Conference and Exhibition - Stockholmsmässan, Stockholm, Sweden, 12-15 June 2017.*

- Lodato, C., Tonini, D., Damgaard, A., Astrup, T. F. (2017) Advanced life cycle assessment modelling of organic waste refineries. *Abstract from Sardinia 2017, 16th International Waste Management and Landfill Symposium - S. Margherita di Pula, Italy, 2-6 October 2017.*

- Lodato, C., Tonini, D., Damgaard, A., Astrup, T. F. (2018) An advanced biorefinery LCA model with a process-oriented approach. *Abstract from Sustain conference 2018: Creating Technology for a Sustainable Society - Technical University of Denmark, Kongens Lyngby, Denmark, 29-30 November 2018*

- Lodato, C., Tonini, D., Damgaard, A., Astrup, T. F. (2018) A bioenergy integrated system with a process oriented LCA modelling approach. *Abstract from 26th European Biomass Conference and Exhibition (EUBCE 2018) - Bella Center, Copenhagen, Denmark, 14-18 May 2018.*

Acknowledgements

It is done! This is last page, the conclusion of an incredible, difficult, amazing, intense, revolutionary experience of my life! I would take the occasion of this page to acknowledge some people that have been part of this journey last few years.
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My co-supervisor, Davide Tonini. I would like to thank him for always supporting, helping, and giving essential recommendations, whenever in the world he was.
I special thanks to Lorie Hamelin who welcomed me in France and made me feel at home. I learned a lot from her in this little time. Thanks for the intense and constructive collaboration.
A great thank to my co-authors Anders Damgaard, Barham Zarrin, Hubert Baumeister whom made me develop the basis of my PhD through the implementation of EASETECH+, as my pen for writing new models. Special thanks to Filomena Ardolino, and Umberto Arena for their important collaboration and all my research group, Residual Resources Engineering -aka – wasters, for the awesome office and working environment.
A great thank to Gazzetta and Tonio, like two big brothers for me, helping from the beginning of my PhD, with suggestions and smiles. A warm thank to Susanna, which supported me "from office to home", to Paola, for being careful, and cheering, and helping me, Zhenhan and Miao for special hotpot and more, Lulu my great officemate always here for me, Rocchino for an amazing friendship, Giorgetta for all the positivity, Trine who translated the Danish summary, Nynne for all the cakes, Emilie for believing in me all the time, David and Inesita for being my second-home roomates, Claudia for being at my side since so long, Amico mio for being here for me from Italy, Paki for last part of my PhD, from Toulouse and over, and Corentin, Carlos, Victor, Monsta, Zhoue, Patrick, and other fantastic friends who made my stay in Toulouse unforgettable, and thanks to other many people and friends that enriched this journey with hugs, smiles, messages, sweets, and joy.
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Summary

In the endeavour of reducing greenhouse gas emissions (GHGs) mainly due to human activities, reaching carbon neutrality by 2050 and maintaining global mean temperatures below political targets, new and efficient solutions are needed. Bio-based energy, or bioenergy, plays an important role in a world where the majority of GHG emissions are from energy. Bioenergy technologies convert biomass to energy. Using biomass as residues from human activities or organic fraction of waste to produce bioenergy has several advantages: i) providing solutions to waste management, ii) promoting material recirculation toward bio-based energy, iii) supplying fossil energy demand, and avoiding for example land use changes of using crops for energy. Bioenergy technologies will contribute to a non-fossil and more sustainable society by transforming bioresources into energy. There are a wide range of challenges associated with this transition such as availability of bioresources, spatial distribution of bioresources, and various conversion technologies. The selection of the environmentally most appropriate technologies to valorise the specific bioresources is also a challenge. Bioresource properties, conversion yields, outputs and rejects, as well as process performance for the bioresources in question need to be systematically evaluated and addressed by assessing the environmental impacts.

Life cycle assessment (LCA) is a standardised method for assessing the environmental performance of technologies and systems. There is a need to expand and improve the modelling of bioenergy technologies, beyond black-box process models in conventional LCA modelling tools. Black-box models typically ignore the links between feedstock characteristics and process outputs. For example, adapting the inventory of a technology to reproduce another one. As such, these models do not reflect changes of operational conditions or conversion efficiencies in a process pathway. Thereby, the reproducibility of a technology and adaptability of the model to specific case studies are limited. The consequence is lack of transparency and limited flexibility from a modelling perspective.

The main goal of this PhD project was to provide a process-oriented LCA modelling framework and apply this to a range of selected bioenergy technologies (e.g. anaerobic digestion, gasification, and upgrading units) and systems of technologies. The framework allowed quantitative and parametrized physical chemical input-output relationships. The generalised principles for process-oriented LCA modelling were developed and implemented into the modelling

framework, EASETECH+, as an extension to the existing LCA model, EASETECH. A range of illustrative examples was used to explain and highlight key features and LCA modelling approaches associated with the framework.

The feasibility of the process-oriented modelling approach was demonstrated upon implementation of technology models within the LCA model EASETECH, including use of all novel operators and functions for model definition in EASETECH+. The new process-oriented framework facilitates LCA modelling of a wide range of conversion processes relevant for bioenergy technologies, including material recirculation, multiple outputs, conditional sequence flows, linear and non-linear responses in conversion pathways.

Based on the PhD, a range of novel process-oriented technology models were implemented into EASETECH as ready-to-use technology templates for new case-studies, including: i) biorefinery, ii) anaerobic digestion, iii) thermal gasification; iv) bio-based methane upgrading.

The consequences of subdividing a technology into unit-processes was given by a second generation biorefinery, managing bioresources with high cellulose, hemicellulose, and lignin content. Pretreatment, hydrolysis, fermentation and distillation, and recovery were the four unit processes identified. Input-output relationships with parameters (e.g. conversion efficiency) were included in each unit-process. Changes of parameters within unit-processes had changes on the mass, substance, energy balance, thus on the intermediate outputs (e.g. simple sugars), final outputs (e.g. ethanol), and environmental performance. For example, increasing the conversion efficiency of cellulose increased the production of sugars and ethanol causing more global warming savings.

A systematic approach accommodating the process-oriented modelling principles was developed and applied on a regional case for bio-based methane supply in the French region of Occitania. This allowed finding environmentally-efficient import/export strategies to supply the gas demand of a region considering: i) availability and properties of bioresources on the region, ii) biological and thermochemical degradation of bioresources, through anaerobic digestion and gasification (both with upgrading), iii) environmental performance of conversion pathways and impacts (induced and avoided) by the current management of the involved bioresources. This can support practical actions toward local bioeconomy and climate goals.

Dansk sammenfatning

Globale og lokale mål for vedvarende energi og reduktion af klimaændringer kræver betydelige forandringer på teknologi- og systemniveau. Bioenergiteknologier anvendes til at konvertere bioressourcer til energikilder, og er dermed essentielle i overgangen til et fossilfrit og miljømæssigt mere bæredygtigt samfund. Der er en lang række udfordringer forbundet med denne overgang, herunder håndteringen af en række rest-råmaterialer fra forskellig oprindelse med forskellig lokal tilgængelighed. Den begrænsede mængde tilgængelig rest-råmateriale udgør i sig selv en begrænsning. Det er også en udfordring at udvælge egnede teknologier til udnyttelse af de specifikke bioressourcer til energiproduktion. Bioressourcernes fysisk-kemiske egenskaber samt energiudbyttet, restfraktioner og driften af processerne varierer imellem forskellige råmaterialer og bioenergiteknologier. Dermed bør der foretages en systematisk vurdering af de miljømæssige fordele og ulemper ved forskellige alternativer.

Livscyklusvurdering er en standardiseret metode til vurdering af teknologiers og systemers potentielle miljøpåvirkninger. Der er behov for at udvide og forbedre modelleringen af bioenergiteknologier, da disse typisk modelleres som aggregerede ”black box”-procesmodeller i konventionelle LCA-modelleringsværktøjer. ”Black box”-modeller udelader typisk forbindelsen mellem et råmateriales egenskaber og outputs fra en proces. Disse modeller afspejler således ikke ændringer i driftsbetingelserne eller effektiviteten på en konverteringsproces. Dette resulterer i en lav reproducerbarhed af teknologien og en begrænset tilpasningsevne til specifikke cases. Konsekvensen heraf er manglende gennemsigtighed og begrænset fleksibilitet set fra et modelleringsperspektiv.

Hovedformålet med dette ph.d.-projekt var at præsentere en procesorienteret LCA-modelleringsramme og anvende denne på udvalgte bioenergiteknologier og -systemer. Modelleringsrammen tillod kvantitative og parameteriserede fysisk-kemiske input-output relationer. De generelle principper for procesorienteret LCA-modellering blev udviklet og implementeret i modelleringsrammen *EASETECH+*, som en udvidelse af den eksisterende LCA-model *EASETECH*. Der blev anvendt en række illustrative eksempler til at forklare og fremhæve de vigtigste elementer og LCA-modelleringsmetoderne i forbindelse med modelleringsrammen.

Den praktiske anvendelse af den procesorienterede modelleringsmetode blev demonstreret ved at implementere de procesorienterede modeller (fra EASETECH+) i LCA-modellen EASETECH. Dette inkluderede alle nye operatører

og funktioner, der indgår i EASETECH+. Den nye procesorienterede ramme letter LCA-modellering af en lang række konverteringsprocesser, som er relevante for bioenergiteknologier og lignende teknologier, herunder recirkulation af materialer, betingede sekvensstrømme samt lineære og ikke-lineære reaktioner i konverteringsveje.

På grundlag af ph.d.-graden blev der implementeret en række nye procesorienterede teknologimodeller i EASETECH som "ready-to-use" teknologiskabeloner til nye case-studier, herunder: i) bioraffinadering, ii) anaerob nedbrydning, iii) termisk forgasning; iv) biobaseret opgradering af metan.

Ydermere blev der udviklet og anvendt en systematisk modelleringsmetode, der var i overensstemmelse med de procesorienterede modelprincipper, for biomethanforsyningen i den franske region Occitania. Dette gjorde det muligt at foretage en ensartet vurdering både på teknologi- og systemniveau med henblik på at identificere relevante råmaterialeteknologier og overordnede scenarier.

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Abbreviations

AD	Anaerobic Digestion
ADH	Anaerobic Digestion with Hydrogen upgrading
EASETECH	Environmental Assessment System for Environmental TECHnologies
GA	Gasification
GHGs	Greenhouse Gases
GW	Global Warming
iLUC	Indirect land use change
MFA	Material Flow Analysis
NB	Net balance
RP	Reference product
UOL	Use on land

1 Introduction

1.1 Background

In the endeavour of supplying solutions for the transition to a more sustainable and resource efficient society, many countries need to make considerable changes and a wide range of new initiatives. There is a need to speed-up and advance the development of integrated solutions for management of locally available resources. These solutions need to fulfil local goals contributing to reach targets imposed by countries and support the global sustainable development (Frizen, K., 2016).

Bioresources are particularly important in this transition. Bioresources may refer to biomass produced from human or animal activities (residual biomass), from crops (produced biomass), from surplus of commercial food products (agriculture surplus biomass), or naturally growing in ecosystems (natural biomass) (Ciria et al., 2016). Organic waste from agricultural, forestry and municipal solid waste are also included.

The transition to a non-fossil economy from a linear to a circular supply chain sees an efficient use of (bio)resources with waste as new resources (Zabaniotou, 2018). The Renewable Energy Directive 2009/28/CE, revised in 2016 has the target of 27% of the total European energy from renewable sources by 2030.Recently, the Green Deal (COM(2019) 640 final) incentivises Europe to become the first carbon neutral continent by 2050. This implies achieving the Paris Climate Agreement goal of preventing increases of the global mean surface temperature to well below 2˚C above pre-industrial levels, and reduce fossil carbon dioxide emissions. This ambitious goal is supported by several instruments, such as Climate Law (EU 2018/1999), the adoption of EU Energy System Integration Strategy toward an integrated energy system with energy and waste recirculation, cleaner power and fuel system.

However, the valorization of bioresources as bioenergy has challenges. The first one regards the quality, quantity and distribution of the available residual bioresources (Van Meijl et al., 2015; Delivant et al., 2015). This reflects national bioeconomy strategies and energy roadmaps (Szarka et al., 2017; Scarlat et al., 2015).

The second challenge of adopting a biobased energy system is the pressure on biodiversity, water scarcity, food security associated with potential unsustainable production of the needed bioresources. “Bio” does not necessarily mean

"environmentally friendly". This is the case for first generation biofuel technologies, which uses feedstock competing with the feed/food sector inducing more environmental impacts than their fossil counterparts especially when the biomass supply causes land-use-change impacts (direct and indirect) (Searchinger 2010; Hamelin et al., 2014; Tonini et al., 2012, and 2016).

A third challenge relates to the conversion technology itself, the scale of operation, and the technology matureness (i.e. different technology readiness level, TRL). Technology can be understood as a mean from bioresource-to-energy and/or materials, fuels. Different technologies with different capacities, operational conditions, and conversion efficiencies can produce the same energy. Some technologies reuse existing infrastructures with new features. For example, Gothenburg biomass gasification plant, GoBiGas, reused two fluidized bed gasifiers of 8 and 16-MWth, respectively 40 and 80 dry tonnes of biomass per day for biomethane production (Alamia et al., 2017; Thunman et al., 2018). Some other technologies are introduced for worldwide renewable targets supported by innovative research studies. An example is given by the increasing numbers of projects worldwide on power-to-gas technologies, which produce storable gas by converting renewable electricity into chemical bound energy (Thema et al., 2019). These emerging technologies can help to stabilize electrical grids and non-fossil economies integrating renewable sources in system of technologies (Bailera et al., 2017). As such, the specific configuration and performance of a technology, as well as the integration of the technology into a system, is important for the overall performance of the technology.

It is fundamental to assess and compare the environmental performance of different solutions for dedicated energy supply, both at a technology and system level. This is relevant to properly choose a technology that fulfils the energy market supply, has environmental savings and lower losses with increased energy conversion efficiencies. Moreover, there is a need for allocating residual bioresources and waste to different technologies based on the (limited) availability of bioresources (Koukios et al., 2017) supporting practical actions and investments in technologies in vision of the global carbon neutrality.

Life cycle assessment (LCA) is a standardised method for quantifying the environmental performance of products, services, or bioenergy pathways (ISO 14040, 14044). The complexity of e.g. enzymatic, microbiological, thermal, catalytic processes of bioenergy technologies need to be reproduced in LCA studies to fully "capture" the environmental performance. However, bioenergy

technologies have several goods and/or services given by co-products, inducing multifunctionality, which needs to be accounted in LCA (EC-JRC, 2010).

The environmental benefits of a non-fossil society toward integration of technologies and material recirculation requires a holistic system perspective and several constrains, such as available bioresources, technology configuration, local context given by e.g. energy mix, climate conditions. Therefore, the design of integrated energy systems with a fast development in technologies is an important aspect that need to be accounted (Directive/EU, 2009). Novel life cycle inventories and advanced modelling tools are needed to improve the sustainability assessment of bioenergy technologies.

LCA modelling tools are mostly available for product and manufacturing, rather than technologies and systems of technologies involving several input feedstock conversion pathways. Most of the LCA tools follow a so-called black-box modelling approach that have embedded data inventories of individual technologies with fixed lists of inputs and outputs (Lodato et al., I). Black-box models cannot represent individual processes within technologies and consequently cannot identify impacts from these processes (Maes et al., 2015). Conversion routes of input properties of bioresources to final bioenergy products is generally not trackable by traditional LCA modelling tools (Blomsma and Tennant, 2020).

In addition, LCA results are highly dependent on differences in modelling assumptions. For example, in Gentil et al., (2010), eight waste LCA models were reviewed and compared with the conclusion that input/output data, parameters, and assumptions reflected the main differences in results, and that consistency across models would require quantification and prioritisation of key parameters. However, LCA of emerging technologies is typically built on available studies for existing technologies by adaption of existing models. This means that new technologies are not appropriately represented by the technology data, performance assumptions, and parameters (Henriksen et al., 2018).

LCA is limited when models reproduce technologies with fixed inventory data and when potential changes in feedstock composition is not reflected by process performance, yields, rejects and emissions. To overcome this, various attempts have been made to integrate material flow analysis (MFA) methods (Mancini et al. 2015; Turner et al. 2016), more advanced process simulation tools (ASPEN 2020; ProSim 2020; ProMax 2020; CHEMCAD 2020) or using the existing LCA software tools in combination with process simulation tools (e.g. MATLAB 2020).

The Danish software tool EASETECH, Environmental Assessment System for Environmental TECHnologies (Clavreul et al., 2014), allows the modelling of technologies based on specific process "templates" (e.g. splitting of mass and substance flows, addition of background processes, specific modules for landfill and anaerobic digestion, etc.) with linear relationships. So far, these process templates have been limited to the operations envisioned when EASETECH was developed originally (Clavreul et al., 2014). In EASETECH, the only way of adding new modelling features in form of new process templates was to change the source code of the software. To remedy this limitation, this project extends EASETECH by developing, validating and applying a process-oriented modelling framework, EASETECH+, to allow more flexible modelling of complex and multi-functional technologies.

1.2 Research objectives

The overall aim of this PhD thesis is to provide an improved basis for advanced modelling of bioenergy technologies within a process-oriented framework. The intention is further to advance the reproducibility of bioenergy technologies and systems. This is finalised by the implementation of process-oriented modelling in EASETECH with new processes and a reproducible framework for assessing energy supply strategies toward sustainable local bioeconomy and a non-fossil society. This is useful to support practical actions of exploiting the carbon potential of local bioresources and technology investments. The research involves the following specific objectives:

- Establish the principles of process-oriented life cycle modelling. This includes elaborating the collected data to establish relationships between operational conditions, process efficiencies and outputs (Lodato et al., **I**)
- Further develop the existing EASETECH modelling framework to facilitate process-oriented LCA modelling (Lodato et al., **II**)
- Implement the process-oriented modelling at a technology level, establishing LCA unit-processes for bioenergy technologies (Lodato et al., **III**)
- Implement the process-oriented modelling at a system level focusing on regional bioenergy supply (Lodato et al., **IV**)

The synopsis part of this PhD thesis is structured as follow:

Section 2 provides an overview of the principles of the process-oriented life cycle assessment modelling

Section 3 presents the EASETECH+ process-oriented framework to facilitate process-oriented LCA models

Section 4 presents the results of process-oriented modelling at a technology level to identify the technology hotspots and comparison among different feed-stock conversion pathways

Section 5 presents the results of process-oriented modelling at a system-level. These results combine different solutions for energy supply

Section 6 discusses the implications of the process-oriented framework

Section 7 summaries the key conclusions of the thesis and provides recommendations for advanced modelling of bioenergy technologies toward a non-fossil society

Finally, **Section 8** offers future perspectives.

2 Principles of process-oriented life cycle assessment modelling

This section addresses the following questions: *What is a process-oriented LCA? What are the elements to account while modelling the process structure?* It is based on Lodato et al., (**I**).

The process-oriented LCA considers "*quantitative and parametrised physical-chemical relationships between input material composition, conversion process units and subsequent output products, promoting mass and substance balanced conversion modelling and environmental assessment*" (Lodato et al., **I**).

The input material is the point of departure of a process-oriented LCA and represents the input flow entering a unit process (e.g. forest residues), which can have more fractions (e.g. softwood, hardwood, poplar) with associated substances as properties, both mass-based and energy-based. These properties are involved in the conversion pathway of a technology unit-process and can be characterized as chemical (e.g. carbon, hydrogen, oxygen), physical (e.g. lower heating value), biochemical (e.g. cellulose, lignin, lipids), and nutritional (e.g. digestible energy). The conversion pathway is output-oriented and it reflects the marked supply of the product demand, e.g. related to biomass-based energy targets. In a process-oriented LCA, the model structure is based on principles of substance and material flow analysis (SFA and MFA) (Allesch and Brunner, 2015; Brunner and Rechberger, 2017). The input material undergoes the conversion pathway established by mathematical relationships reproducing technology operational conditions, conversion efficiencies, chemical reactions, kinetics, respecting the overall mass and energy balance (Lodato et al. **I**).

The output of a process-flow can be intermediate or final. The intermediate output is generated during the modelling stage and it is used as input of another process. The final output is the "last" output of the process representing the final product obtained after the execution of the entire conversion pathway (e.g. bioethanol) (Lodato et al., **I**).

In a conversion pathway, the material flow allows for transitions and transformation of substances or fractions. Transition means "movement". A substance can transit when it is involved in a conversion pathway within a process (i.e. intra-process transition). A transition is between two processes when a substance of the output of a process is the input of the following process (i.e. intra-process transition). Transformation means "changing". A substance is used in

a mathematical relationship to determine another substance or contribute to produce an intermediate or final output. In a transformation, a substance ceases to exist in its original form. Transition and transformations can be partial (less than 100% is transferred or converted to another property/product) or total (100% transferred or converted); this usually depends on the technology conversion efficiencies.

Parameters, such as conversion efficiencies and operational conditions, can be part of mathematical relationships between inputs and outputs of a process. Case-specific conditions can be reproduced by changing parameters in modelled process templates. Therefore, the quality and quantity of outputs are affected by parameters' changes. For example, in a multioutput technology some outputs are preferred to others according to the goal of the LCA and the product to be supplied. Hence, increasing efficiencies or changing some operational conditions, such as the temperature of a reactor, changes the characterization of the outputs produced. Environmental consequences reflect changes of increasing an efficiency within a process and based on the uses of the output products. These changes can have linear and non-linear responses on mass and energy balance in EASETECH. This depends on the mathematical relationships used in the modelling.

In a process-oriented LCA, it is possible to have multiple "if functions," i.e. (multi-)conditional sequence flows (Lodato et al., **II**). When there are multiple outputs, the process has corresponding multiple flows. These flows are in sequence and can be enabled or disabled through the condition met based on the available data (Figure 1). In a conditional process, the process pathway depends on parameters and constrains. While parameters affect the mass/energy balance in conversion pathways, constrains define the enabled pathway during the process execution (Lodato et al., **II**).

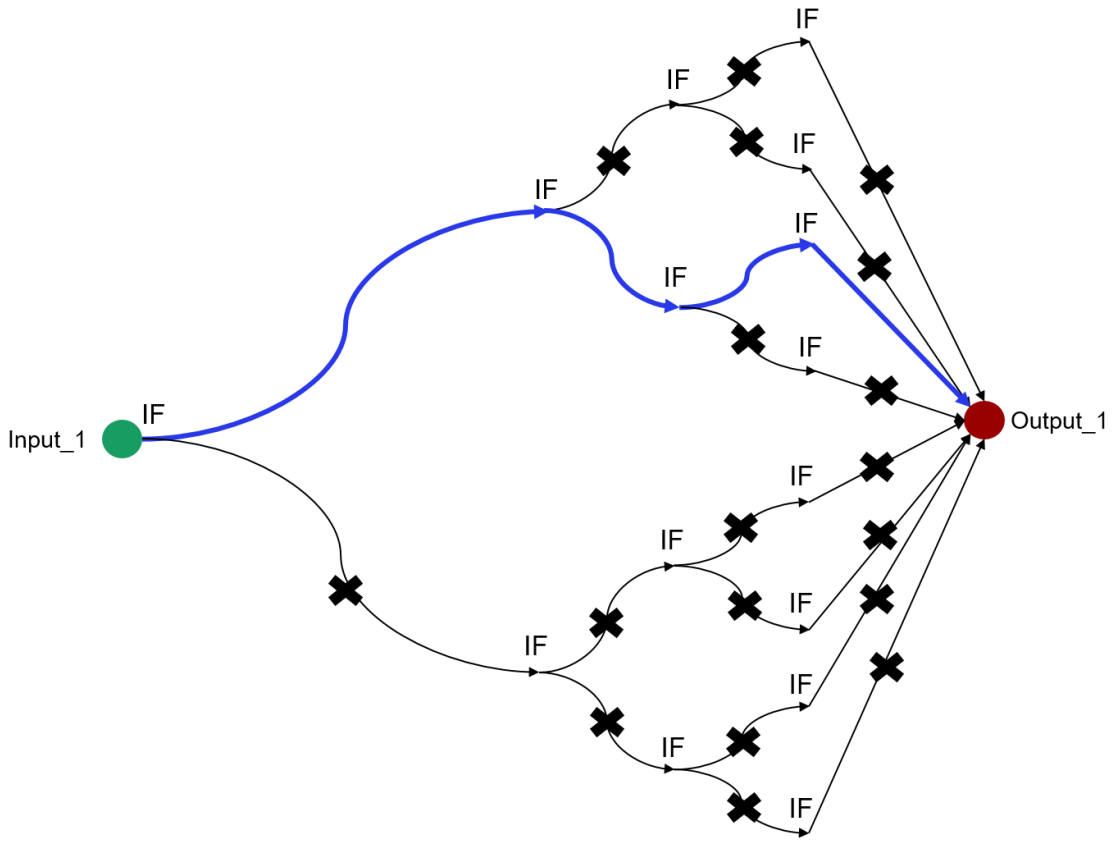

Figure 1: Illustrative example of (multi-)conditional sequence flow. Based on the outcome of "if functions" ("IF"), the blue flow is the one that generates the *output_1* from the *input_1*.

An illustrative example from Lodato et al., (**IV**) of process-oriented modelling is in Figure 2 representing a gasification process. Crop residues, forest residues, green waste, pruning residues, and wood waste represent six input materials (one kilogram each). The conversion-process takes the carbon, hydrogen, oxygen, and nitrogen content of each input based on the content of the biochemical properties of each input. For example, hydrogen (H) is a chemical element and hemicellulose ($C_5H_8O_4$) is a compound; both are properties of the input material: the hydrogen content is calculated based on the hydrogen content of hemicellulose, representing a part of the total hydrogen in the input material. Additionally, syngas as H_2, CO, H_2O, CO_2, CH_4, and N_2, is determined (in Nm^3) based on two independent equilibrium reactions, a global gasification reaction and four material balance equations (Figure 2) (Lodato et al., **IV**). For all input materials, the gasifier temperature, pressure, and equivalent ratio were respectively 850˚C, 1 bar, and 0.33. While the amount of molar moisture and air was different for each input material (Table 1), based on one

mole of dry input material (Lodato et al., **IV**). These relationships quantify the main syngas characteristics, such as volumetric and mass flow rate, composition, low heating value (LHV), and the syngas-specific and process-specific air emissions, together with consumptions of chemicals, such as catalysts, and residues sent to disposal.

Table 1: Molar moisture and air for five dry or semi-dry feedstock. Calculations are based on input feedstock properties, and operational conditions, such as the equivalence ratio (set as 0.33)

Category	**Molar moisture (mol_H2O/mol_in)**	**Molar air (mol_air/mol_in)**
Crop residues	0.00093	0.00773
Pruning residues	0.00105	0.00654
Green waste	0.00184	0.00439
Wood waste	0.00027	0.01530
Forest residues	0.00034	0.01333

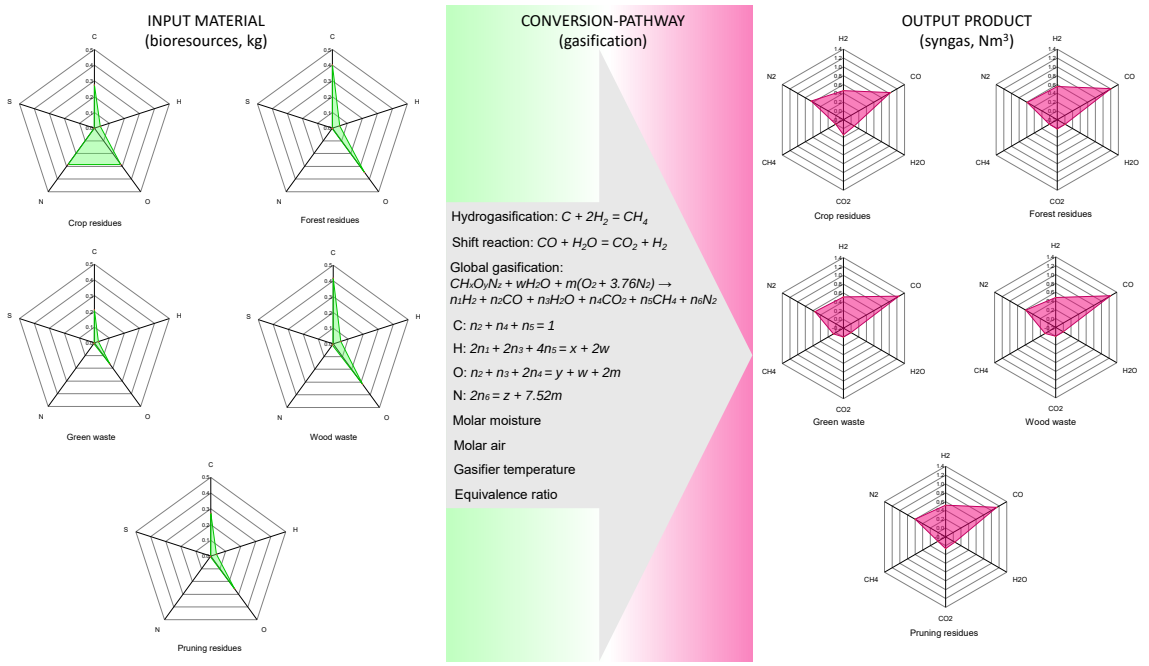

Figure 2: Illustrative example of process-oriented approach applied to a gasification process; to the left, six inputs feedstock with different composition, showed as C, H, O, N, S, and to the right, the syngas produced express as H_2, CO, H_2O, CO_2, CH_4, and N_2. The conversion pathway is represented by the arrow showing some input-output relationships (with parameters)

With this example, syngas characteristics can be determined based on input-specific composition, and technology configuration and performance. The input-to-syngas conversion relationships reflect the reactions of biomass thermal degradation and gas formation. Particularly, it approximates a stoichiometric equilibrium model for the chemical composition of the syngas by mass balance, explained in Ferreira et al., (2019).

3 EASETECH+ process-oriented modelling framework to facilitate process-oriented LCA models

This section presents the framework and method used to model bioenergy processes implemented into EASETECH. It is based on Lodato et al., (**II**).

3.1 A new process template: From modelling in EASETECH+ to implementation into EASETECH

"*Modelling in EASETECH+ starts with a "white canvas" where LCA practitioners can "draw" the process flows with the level of detail needed...*," Lodato et al., **(II)**.

To drawing the process flow, operators, and functions of EASETECH+ are needed. Operators are used to build the structure of the process flow, e.g. as building blocks, while functions are used in the properties of operators and allow for calculations, set up the quantity of substances, fractions, or material in general, to circulate within the process. There are four categories of operators, one is used only for substances (substance operators), one only for fractions (fraction operators), one for input, output and material (process operators), and one for transition among operators creating material flows (flow operators). These operators were introduced when explained the process-oriented modelling approach in Lodato et al., (**I)**, as EASETECH modelling features to support the approach. Additional attention to these operators was given in Lodato et al., (**II)**. The latter was more on the application tool for implementation of process-oriented modelling. Hence, the focus was on the entire modelling framework of EASETECH+ with keys examples and an exhaustive application to understand how to use the software. This is relevant for LCA practitioners who want to model any type of operation they could envision and to implement these as new process templates in EASETECH, without the need for recompilation and redistribution of the EASETECH software itself, which was not possible to do by an LCA practitioner (Lodato et al., **II**).

In EASETECH+, a process is sequential, e.g. input → intermediate output 1 → intermediate output 2 → final output. Linear and non-linear flows are allowed, and they can have some conditions (e.g. multi-conditional sequence flows).

EASETECH+ generates a process flow diagram, i.e. "*a visual representation of all linear and non-linear input-output relationships that may subsequently be imported into EASETECH as a new process template*" (Lodato et al., **II**). In EASETECH, the process flow diagram can represent a process within a technology, such as hydrolysis within the biorefinery, or it can represent an entire technology, such as anaerobic digestion. The level of modelling details is according the goal and scope, and case study to assess in the LCA. Another important feature added to the modelling is the possibility of having iterations in the process flow. This means that transitions or transformations can be repeated several times until the condition is met. Uncertainty propagation is also allowed in EASETECH+, with parameters used to control substance and fraction balances and uncertainty assessment via data distribution.

A sequence of three steps for the application of EASETECH+ modelling framework to produce a process flow diagram is followed presented (based on Lodato et al., **II**):

1. **Identifying the content of the process.** Relationships between input material composition, conversion process and output products
2. **Establishing the intra-process transitions and transformations.** Based on 1, EASETECH+ is applied to model the process flow diagram using operators and functions
3. **Validating the results.** The process flow diagram is modelled based on 1 and 2 and it reproduces the process as expected. The process flow diagram is implemented into EASETECH as a new process template, ready-to-be-used in LCA scenarios.

All the functions and operators, together with key examples, and applications are in Lodato et al., (**II**).

An example of applying EASETECH+ modelling framework is given in Figure 3. This example does not reproduce an existing process. It is merely used for illustrative purposes. This process has an input and two outputs. One of the two outputs, *Feedback output1*, has a condition (*SubC > 0*) which allows for iterations until this condition is not met. Only one output has properties associated, due to the enabled sequence flow. This process has parameters, such as: i) transfer parameters, to transfer substances; ii) conditional parameters, when

parameters are in a condition (e.g. *if parameter_1 = 0*); iii) conversion parameters, when parameters are in an equation (e.g. S*ub_A * X*, *X* is the parameter); and operational parameters (e.g. temperature of a reactor).

Once that the process is validated, it can be implemented into EASETECH, ready-to-be-used in LCA scenarios (Figure 3). The integration with existing processes of EASETECH is allowed. The new processes need to have the same nomenclature as EASETECH, for conformity. Modelling of new substances with new names is allowed too.

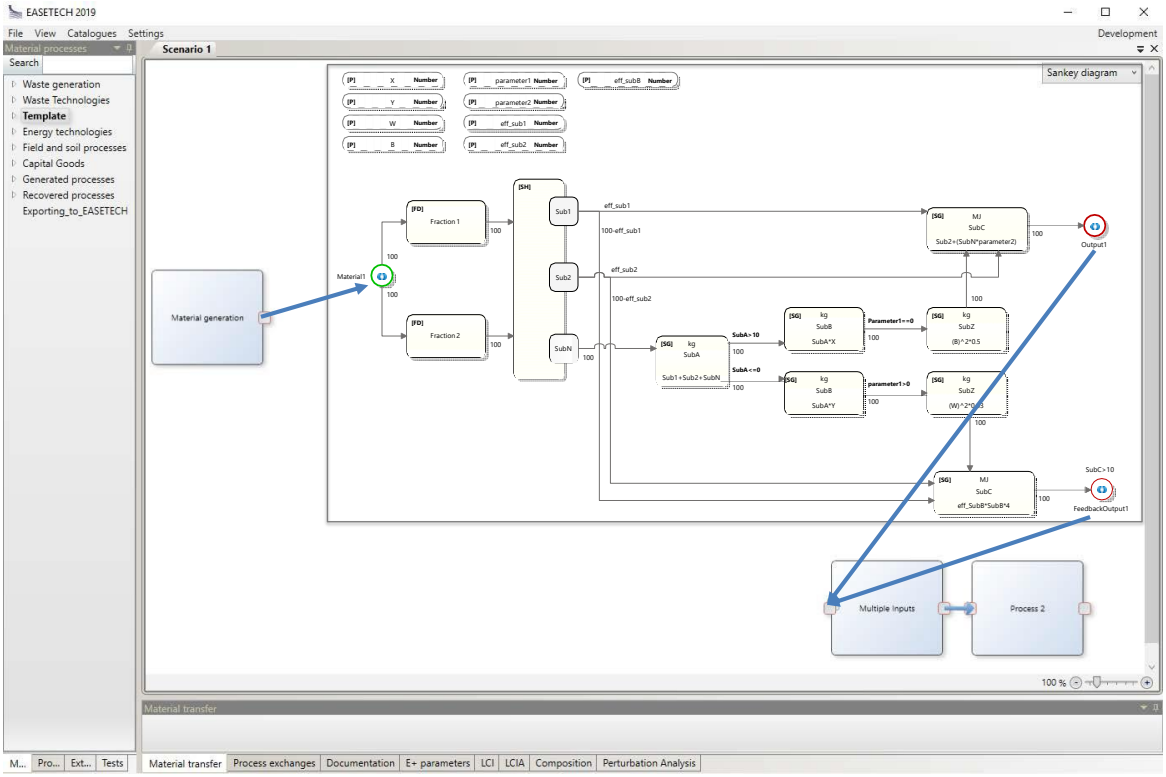

Figure 3: Schematic illustration of implementation into EASETECH of a process modelled in EASETECH+ with the process-oriented modelling approach. The process is integrated with existing processes of EASETECH

3.2 Framework for environmental assessment in the context of local bioeconomy

This section reports the concepts of the stepwise framework presented in Lodato et al., (**IV)**.

For the management of local available resources to supply the bio-based energy and to find strategies for a decarbonized society, a stepwise approach was found based on the principles of the process-oriented and EASETECH+ modelling framework. This represented an integration of the process-oriented modelling with a practical application to support decisions for local management strategy solutions. The approach provides a consistent evaluation of both technology and system perspectives and has the following six steps:

1. **Goal and scope definition.** According to the overall goal of the LCA, definition of functional unit, system boundaries, impact assessment methods, key impact categories, data quality according to ISO standards (ISO, 2006 a, b).
2. **Estimation of bioresource potential.** Reflecting step 1, identification of relevant bioresources with associated physical-chemical and biochemical properties and current management.
3. **Selection of technologies.** Based on step 2, identification of relevant conversion pathways and unit-process chains.
4. **Process-oriented inventory modelling.** Based on step 2 and 3, establishment of relevant process models for both the selected conversion pathways and the counterfactuals (i.e. alternative management or fate) associated with each bioresource.
5. **Evaluation of technology performance.** Evaluating the environmental performance of individual conversion pathways for all relevant bioresources. Calculation of environmental net balances by comparison with counterfactuals. Identification of appropriate conversion pathway and feedstock combinations. Identification of potential environmental trade-offs.
6. **Evaluation of system-level scenarios.** Based on all previous steps, identification of relevant system-level scenarios involving combinations of bioresources and conversion pathways for fulfillment of functional unit and assessment goal.

Some results of applying this stepwise approach are given in Section 5.

4 Process-oriented modelling at a technology level

To provide a comprehensive evaluation of the process-oriented approach, key modelling aspects are highlighted based on a range of questions subsequently addressed through selected application examples. This section reports results of Lodato et al., (**I**), (**II**), and (**III**).

4.1 Technology as unit-process modelling: Second-generation biorefinery

What are the consequences of subdividing a technology into unit-processes? Based on Lodato et al., (**I)**.

A second generation biorefinery (or lignocellulosic) was modelled in EASETECH+, implemented into EASETECH and applied in an LCA scenario (Lodato et al., **I**), where: i) bioethanol, was used as vehicles-fuel, substituting gasoline, ii) solid residues, were used in a power plant, substituting the production and combustion of natural gas; while, iii) liquid residues (molasses) were used in a biogas plant also substituting the production and combustion of natural gas. The modelling started with the identification of five unit-processes:

- **Bio-material generation.** Mathematical equations determined some properties such as C, H, O, N, S based on the quantity, in kilograms, and chemical formula of the biochemical properties of a selected feedstock (e.g. hydrogen (H) and hemicellulose ($C_5H_8O_4$) are properties of the input material: the hydrogen content is calculated based on the hydrogen content of hemicellulose, representing a part of the total hydrogen in the material).
- **Pre-treatment.** The feedstock is heated to break down its lignocellulosic structure and separate lignin from cellulose and hemicellulose to allow an efficient conversion into fermentable sugars (Lee et al., 2015). It accounts for some losses e.g. when the conversion efficiency to the pre-treatment output is less than 100%.
- **Hydrolysis.** Polysaccharides or complex sugars, such as cellulose, hemicellulose, starch, pectin, sucrose, are hydrolyzed into simple sugars with 5 and 6 carbon atoms (C5 and C6 sugars) representing hydrolysis

products. What is not hydrolyzed forms hydrolysis residues, which are transferred to the recovery process.

- **Fermentation and distillation.** Ethanol is produced based on the C5 and C6 sugars during the fermentation, while during the distillation the unconverted sugars (fermentation residues) were separated and transferred to the recovery process.
- **Recovery.** Residues from both hydrolysis, and fermentation and distillation were collected; solid and liquid residues were separated for further energy and material recovery, e.g. through a power plant and an anaerobic digestion plant.

Mathematical relationships between each unit-process were used to reproduce the biorefiney model and implement it into EASETECH. In addition, each unit-process had parameters associated: pre-treatment and recovery allowed mainly material transitions, while hydrolysis, and fermentation and distillation allowed material transformation to simple sugars and to bioethanol, respectively. Figure 4 (Lodato et al., **I**) provides a schematic overview of the unit-processes within the modelled biorefinery and the link among them in terms of material flows represented by the arrows.

The performance of subsequent unit-processes is dependent of the previous step. The interconnection between unit-processes is beyond linear modelling and appropriately represents this stepwise dependency among unit-processes. The subdivision into unit-processes is needed: i) to assess the environmental performance of unit-processes; ii) to control the mass, substance and energy balance of intermediate outputs (together with rejects); and iii) to be able to combine unit-processes in order to integrate systems of technologies in different conversion cycles for material recirculation.

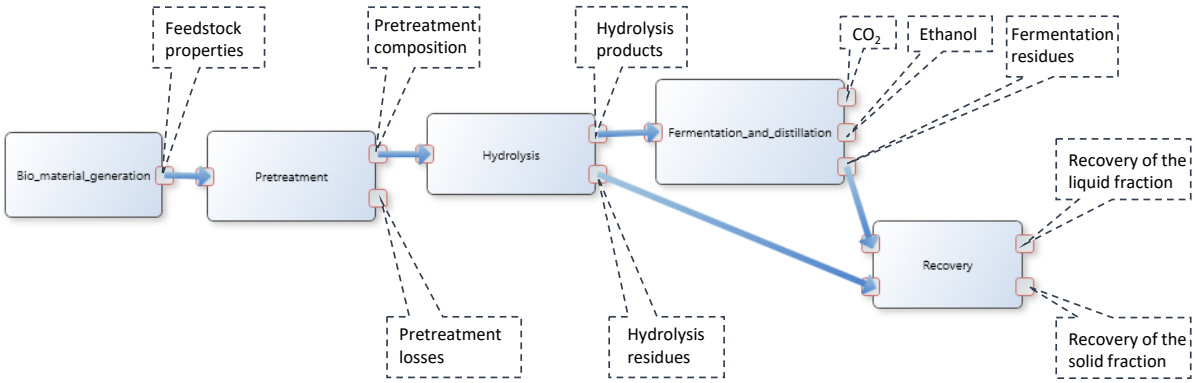

Figure 4: Biorefinery unit processes with input, intermediate and final outputs (Modified from Lodato et al., **I**)

4.1.1 Process performance and choice of parameter

In Lodato et al., (**I**), the stepwise dependency among unit-processes was tested by changing unit-process parameters in two biorefinery unit-processes (hydrolysis, and fermentation and distillation). Mass, substance and energy balance reflected these changes.

In hydrolysis, sucrose and starch were always totally converted into simpler sugars, while cellulose and hemicellulose were partially converted.

In fermentation and distillation, C5 and C6 sugars are diverted between bioethanol and CO_2 according to some efficiencies, e.g. 88% of C5 sugars contributes to the production of bioethanol, while 100% - 88% = 12% to CO_2. The two equations (Eq.1 - 2) used to determine C5 and C6 sugars (Tonini et al., 2016) were:

$$C5\ sugars = Hemicellulose * eff\_Hemicellulose * 1.136 \quad \text{(Eq. 1)}$$

$$C6\ sugars = Sucrose * eff\_Sucrose * 1.05 + Starch * eff\_Starch * 1.1 + Cellulose * eff\_Cellulose * 1.1 + Pectin * eff\_Pectin * 1.1 \quad \text{(Eq. 2)}$$

Where the constants represented the correction factors for the mass intake due to the hydrogenation, meaning that a molecular hydrogen reacts with another compound in the presence of a catalyst (acid or enzymatic) (Aslanzadeh et al., 2014).

Beet top, wild grass, and wheat straw were used independently as input of the biorefinery. Cellulose conversion efficiency and C6 sugars conversion efficiency were changed one-at-a-time during hydrolysis and fermentation and distillation respectively, as 0%, 25%, 50%, 75%, and 100%. These percentages were used for illustrative purposes and to have some extremes (e.g. 0% and 100%). Analysing the effect of changing hydrolysis and fermentation and distillation's parameters, it is possible to notice that: i) when the conversion parameter is changed during hydrolysis, the process-pathway from the hydrolysis on reflects the change. This affects the production of C5 and C6 sugars, together with the output products of the fermentation and distillation, and recovery; ii) with changes in fermentation and distillation, only the output products of this process respond together with the recovery one; iii) the not hydrolysed polysaccharides contribute mainly to solid residues; iv) the not fermented sugars contributed mainly to liquid residues. On this purpose, changing the cellulose conversion efficiency in hydrolysis affected the production of C6 sugars. Producing less or more C6 sugars had a consequence on the solid residues production and this is visible in Figure 5, in output products of the three graphs on the top: with the increased conversion of cellulose there was an increased production of ethanol and a decreased production of solid residues, while the liquid residues were not affected. Additionally, when the conversion efficiency regarded the C6 sugars (Figure 5, three graphs on the bottom), increasing the conversion efficiency produced more ethanol and less liquid residues, while the solid residues were not affected.

An LCA at a technology level with a focus on the unit-process parameters, sees the effects of the output products on the technology environmental performance. This depends on the products' uses. In this case, an increased production of ethanol generated more savings for the global warming impact category, due the substitution of gasoline having an emission factor of 0.097 kg CO_{2_eq} MJ^{-1} vs 0.067 kg CO_{2_eq} MJ^{-1} for the natural gas. Thus, considering the same amount of ethanol and solid/liquid residues, there are more savings from ethanol compared to natural gas (Lodato et al., **I**).

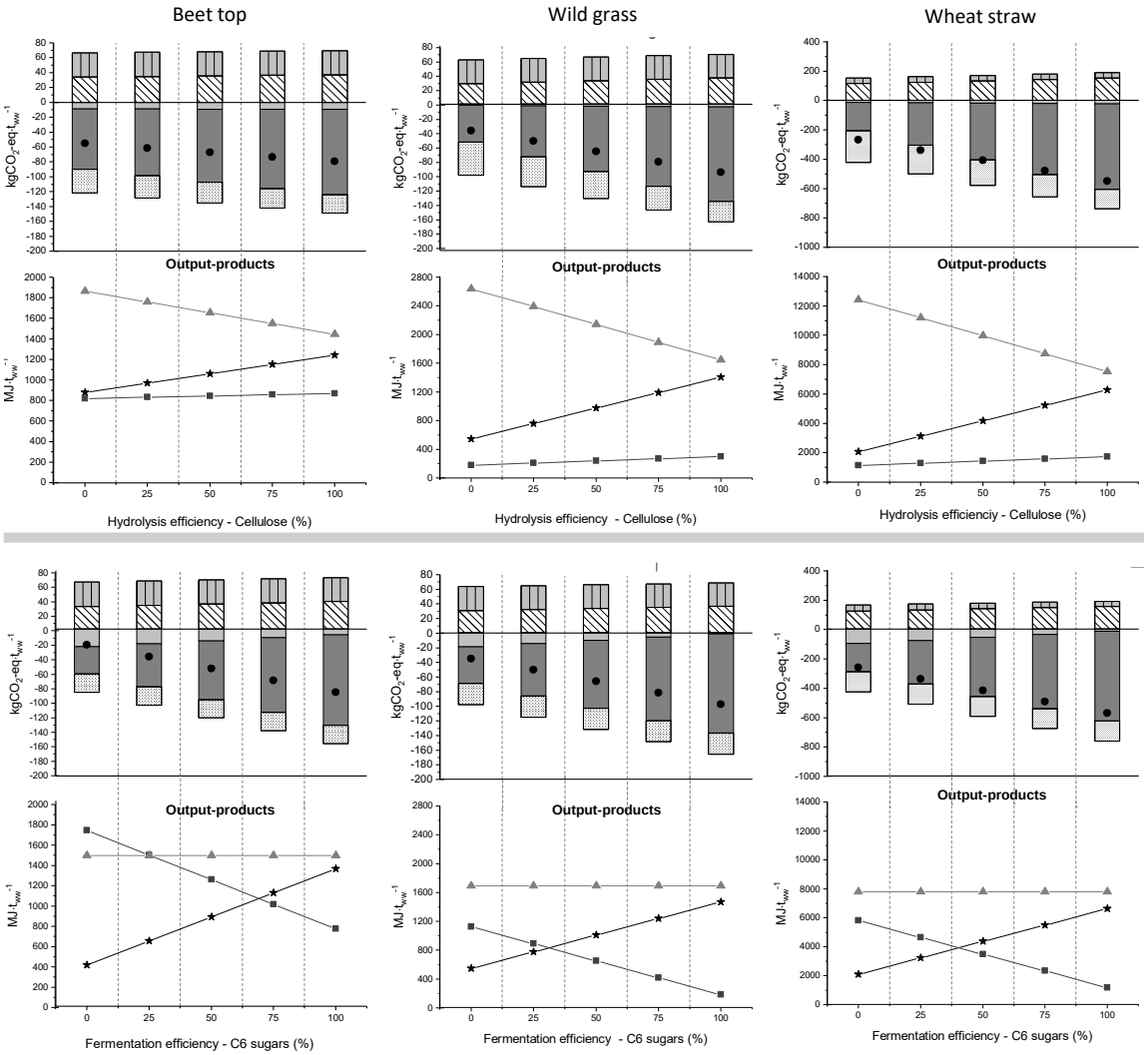

Figure 5: Illustrative example of changing two parameters in two unit-processes (cellulose in hydrolysis, three graphs on the top, and C6 sugars in fermentation and distillation, three graphs on the bottom) of a process-oriented biorefinery. Beet top, wild grass, and wheat straw are the three input biomasses (modified from Lodato et al., **I**)

4.1.2 Feedstock properties at fixed parameters sets

Different input feedstocks have different properties and a different carbon pools to be converted into final technology outputs.

In Lodato et al. (**I)**, the effect of the input feedstock was tested in terms of output products and global warming emissions considering the same biorefinery unit-process parameters (Figure 6). Three feedstocks were the inputs of the biorefinery, selected based on the cellulose hemicellulose and lignin content. The first two, cellulose and hemicellulose, are sugar based and contribute to the ethanol production, while the last one, lignin, contributes to produce the solid residues. Thus, it is expected that a feedstock with more cellulose and hemicellulose produces more ethanol and more global warming savings, compared to the one with more lignin. In addition, cellulose and hemicellulose conversion efficiency to C5 and C6 sugars are important to produce the solid residues, because a low efficiency generates high residues. Therefore, Miscanthus was selected for a high cellulose content (CE = 47.6%DM), brewer's grain for a high hemicellulose content (HC = 29.5%DM), and willow for a high lignin content (LG = 31.6%DM). The unit-process parameters were fixed for the three feedstocks and were: 95% and 75% for cellulose and hemicellulose respectively during hydrolysis, and 88% for both C5 and C6 sugars during fermentation and distillation. The feedstock that produced more ethanol was Miscanthus with 7400 $MJ \cdot t_{ww}^{-1}$, followed by willow (3500 $MJ \cdot t_{ww}^{-1}$) and brewer's grain (1400 $MJ \cdot t_{ww}^{-1}$). Willow produced more ethanol than brewer's grain even if it has a higher lignin content, due to the higher cellulose content, which contributed to the mass balance. In addition, Miscanthus provided the largest solid residues (7200 $MJ \cdot t_{ww}^{-1}$), even if willow had a higher lignin content. This was due to the due to the larger amounts of unconverted sugars (dry basis). The fermentation and distillation process influences the liquid fraction. For this reason, Miscanthus provided the highest liquid output (1600 $MJ \cdot t_{ww}^{-1}$) followed by brewer's grain (1000 $MJ \cdot t_{ww}^{-1}$) and willow (960 $MJ \cdot t_{ww}^{-1}$) (Lodato et al., **I**).

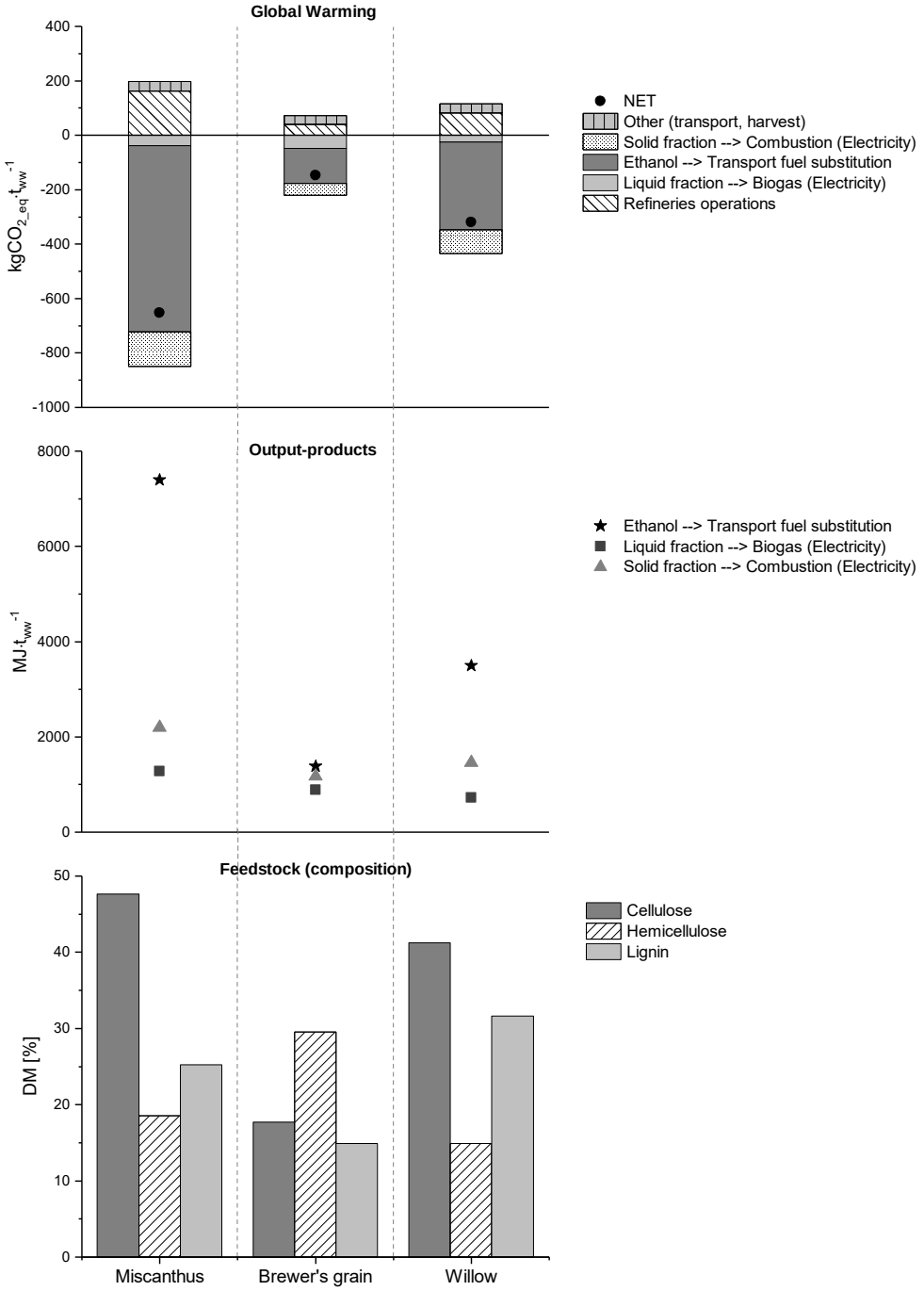

Figure 6: At fixed unit-process parameters, cellulose, hemicellulose, and lignin content of different feedstocks (Miscanthus, brewer's grain and willow, at the bottom) affected the output products (middle) and global warming emissions (top) (Lodato et al., **I**)

Therefore, environmental savings (or burdens) depend not only on the quality and quantity of the output, but also on the management and uses of solid and liquid biorefinery by-products.

4.2 Technology as single process modelling: Anaerobic digestion

Which level of process details can be modelled in EASETECH+?

EASETECH+ modelling framework can accommodate different types of input data and different (multi-)conditional sequence flows to determine expected process outputs. Flows are established based on the data available and the level of detail required to fulfil the functional unit and the goal of the study.

An anaerobic digestion (AD) process was modelled in EASETECH+, imported into EASETECH and applied in an LCA scenario, where: i) biogas was used in a combined heat and power plant for heat and electricity production, and ii) digestate used as fertilizer.

For the identification of the AD, it was considered: i) a simple system of stirred reactors having a constant temperature and a perfect mixing, ii) an input-material of carbon, hydrogen, oxygen, nitrogen and sulphur (C, H, O, N, S), iii) two outputs, biogas as methane (CH_4) and carbon dioxide (CO_2), and digestate or liquid fraction, as non-degraded biochemical properties, water, and inorganics from the input-material.

The AD was modelled as a single process template, with only one process template representing the entire anaerobic digestion (Lodato et al., **II**). The sequence of calculations to get to the two final outputs of the process had conditions based on data available. During the establishment of the intra-process transition and transformations, multiple flows were identified based on:

- **Input feedstock**. Two flows subdivided the model into two cases: *Case A*, when the input modelling started directly by C, H, O, N, S content, and *case B*, when the input modelling started with biochemical properties of the fractions to be digested anaerobically.
- **Output products**. Two flows were identified based on the two final outputs: *gas-flow* for the biogas and *digestate-flow* for the digestate.

- **Methane potential.** Two flows were identified according to the nature of methane potential: if it was calculated from the feedstock's organic matter or experimentally (from measurements).
- **CH_4 measured in the gas** or **CO_2 in the digestate.** Once the biogas is generated, part of CO_2 goes to the liquid phase. On this purpose, the mass balance needed to be adjusted. Therefore, there were two flows to determine CO_2 in the biogas (in m^3): one considered the fraction of CO_2 to the liquid phase, and another one considered the methane measured in the biogas.

In EASETECH+, both cases A and B with all the (multi-)conditional flows were modelled in the same process having flows enabled or disabled according to the available data. When the AD process template is used in EASETECH, a decision tree can be drawn, which defines the path to follow in order to obtain the two final outputs, as illustrated in Figure 7.

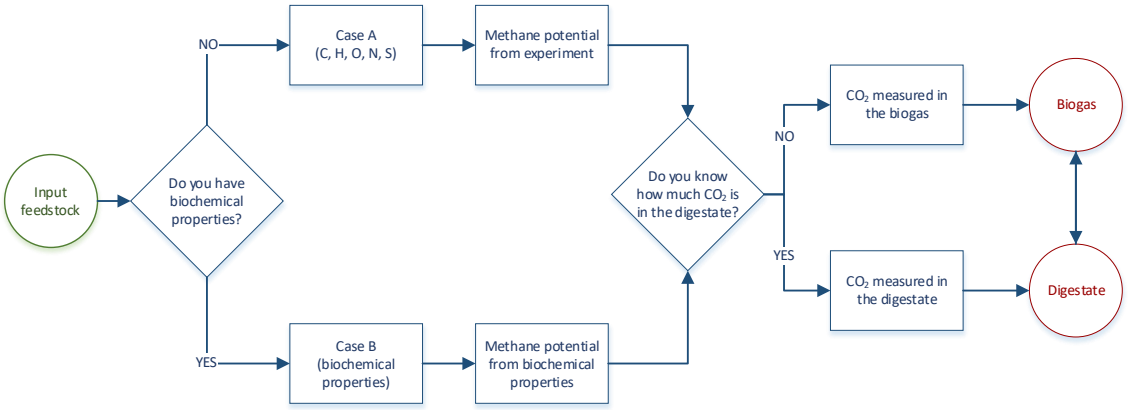

Figure 7: Decision tree to be considered when using the anaerobic digestion (AD) process in EASETECH. It starts from the input feedstock and ends with to outputs products (i.e. biogas and digestate). The modelling flow is based on available data

4.2.1 AD process flow diagram: details of the model

In this section, selected details of the AD model are highlighted to show the complexity that can be reached by modelling a process that can accommodate different types of input data. It is based on Lodato et al., (**II**).

Intra-process transitions and transformations allow to convert properties of the input feedstock (here called “input-material”) into output products. Only selected properties are involved in the conversion pathway. For example, the AD model considers properties of the organic fraction of a feedstock and bases all the calculations in EASETECH considering either C, H, O, N, and S of the input feedstock (case A) or biochemical properties (case B). Thus, substances considered in the conversion pathway are known and are based on the predefined list of material properties in EASETECH. It is important to bear in mind that if the model takes C in the calculations, only C is considered. Thus, adding new properties does not mean that they are included in the calculations. Therefore, the AD model gives a possibility of adding more biochemical properties than the one in the predefined list of EASETECH, such as lactose, fructose, here called “organic molecules” (OM) and includes them in the calculations. In Figure 8 (taken from Supplementary information of Lodato et al., **II**), the process flow diagram of the AD input is showed. The organic molecules can eventually be added (a, Figure 8) together with their properties, such as C, H, O, N, S, methane yield, energy content, VS (b, Figure 8) and diverted between the gas-flow (c, e, Figure 8) or the digestate-flow (d, Figure 8). While, the input feedstock with the predefined list of properties is the input-material (Figure 8). In f of Figure 8, the input-material is diverted between the gas and digestate-flow. Subsequently, the flow from the input-material is enabled when the data available are C, H, N, O, S, or biochemical properties of the predefined list of EASETECH and the flow associated to the organic molecules is disabled. While, when biochemical properties are considered, the one from the input are enabled together with the organic molecules. The latter are included in the model only if the LCA practitioner adds the atoms of the organic molecule in the table of parameter in EASETECH, together with the mass in kg and the efficiency in % defined the mass available to the biogas-flow after the pre-treatment. On the contrary, the default values are set to zero and the model takes only the biochemical properties of the input-material.

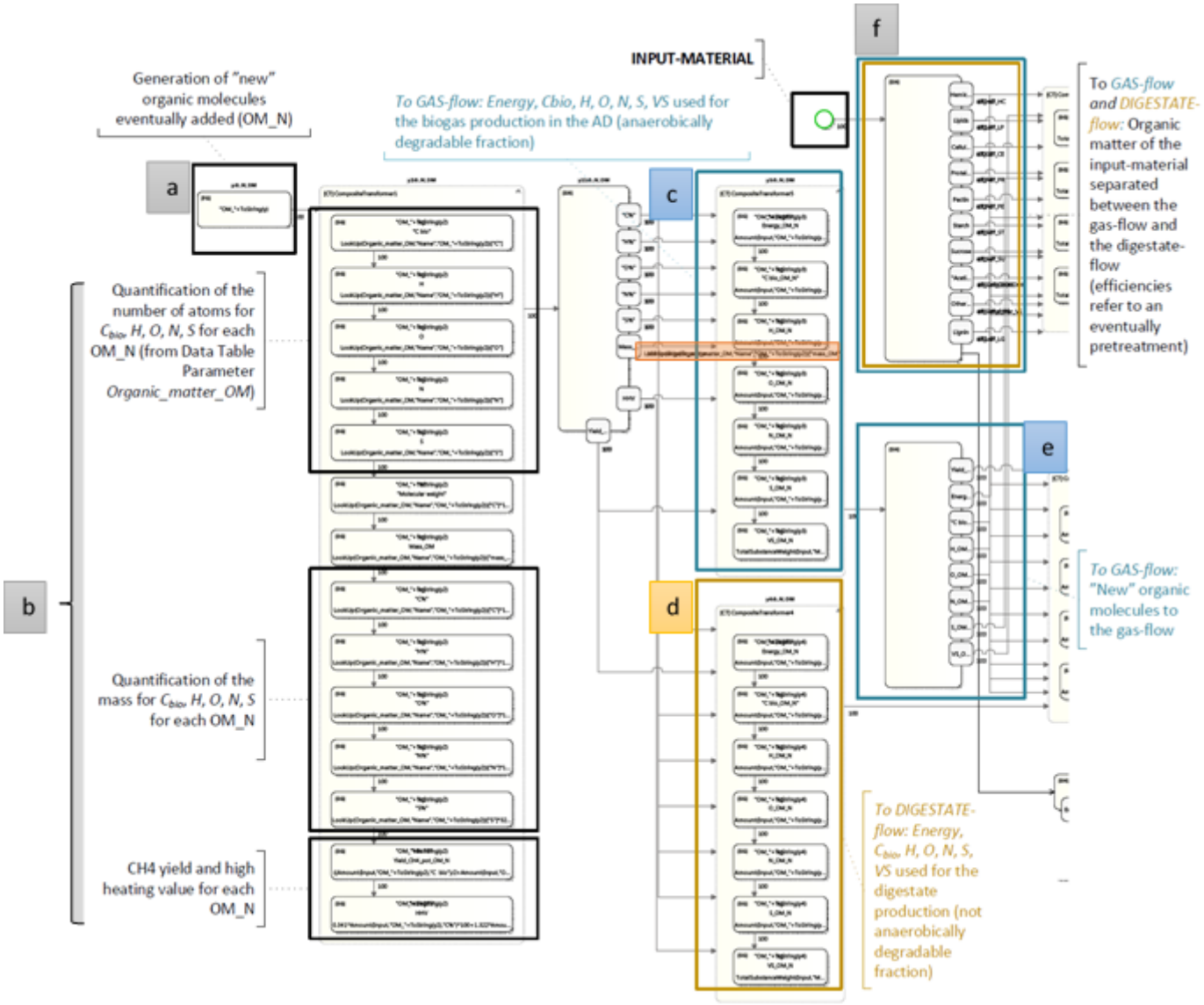

Figure 8: Anaerobic digestion (AD) process flow diagram. It represents the AD input. A, b, c, d, and e are for the eventually added organic molecules, while f is for C, H, O, N, S and the biochemical properties from the input-material (from EASETECH'S material properties) (from Lodato et al., **II**)

4.3 Technology combined with experimental data: Fluidizing bed gasifier

How can a technology model be established based on experimental data?

This section addresses the case of having large amount of high-quality data on a technology with different input feedstock and operating conditions. It is based on Ardolino et al., (**III)**.

A fluidizing bed gasifier (FBG) was modelled in EASETECH following the process-oriented framework, where syngas was burned in a combustor for electricity production and ashes were sent to disposal.

For the identification of the type of reactor and its configuration three unit-process were considered: i) a bubbling fluidized bed gasifier having olivine as bed material, air as fluidizing agent (fluidizing velocity range 0.67 - 0.74 m/s), equivalence ratio (ER) (range 0.20 - 0.31), maximum thermal output of about 400 kW, input capacity between 30 and 100 kg/h, size of the reactor 0.381 to avoid scale-up effect for commercial facilities; ii) a cleaning section formed by a cyclone, for syngas dedusting, and wet scrubber with a mild combustor, chosen for limited formation of dioxins and furans (PCDD/F) and products of incomplete combustion (i.e. soot, polycyclic aromatic hydrocarbons), and iii) an air pollution control system to clean the obtained flue gas through: pulverised activated carbon for absorbing Hg, Cd, low-boiling heavy metals and dioxins, and hydrated lime, for neutralizing HCl, H_2S, SO_x.

The intra-process transition and transformations of the FBG started by considering the feedstock feeding the gasifier and their properties. There were seven waste-derived fuels, two biomasses and a co-gasification (mix of two feedstocks). In addition, the parametrization was used to determine air emissions, electricity recovery and consumption, residues to be disposed, and was based on:

- **Waste-specific parameters.** These two types of parameters refer to the gasifier and the cyclone, resulted from experimental tests: i) transfer coefficient (TC) defined as *the ratio between the mass flow rate of each element in an output stream to the mass flow rate of the same element entering the reactor* (Ardolino et al., **III**). They can range between 0 and 1 and are partitioned between the two outputs of the gasification, syngas and ashes (i.e. TC-to-syngas and TC-to-ashes). The second waste-specific parameter is the ii) substance-to-compound conversion coefficients (CCs), defined as *the amount of C and H transferred into specific syngas compounds (i.e., CH_4, CO_2, CO, tar, H_2, H_2O and C_nH_m with n equals to 2 or 3 and m equals to 2, 4 or 6)* (Ardolino et al., **III**). Firstly, carbon-to-compound were established *dividing the carbon mass flow rate in each syngas compound with the total carbon flow rate in the syngas* (Ardolino et al., **III**). Secondly, for hydrogen the experimental results were coupled with the atomic balances considering the amount of hydrogen present in the compounds already quantified by carbon-to-compounds.

- **Process-specific parameters**, based on operational data (e.g. net electrical efficiency of the organic ranking cycle turbine 17.7%, activated carbon consumption 0.5 kg/t_feedstock, NOx emissions 0.0074 ng/kg_syngas).

Results were validated with the comparison with those evaluated during experimental activity. For example, it was quantified the cold gas efficiency (CGE) as *the ratio between the chemical energy of the produced syngas and that of the feedstock fed to the reactor* (Ardolino et al., **III**) with an average error of 3.5%. Also, *the volumetric flow rate of the dry syngas compounds* (i.e. dry syngas yield) was quantified and compared with the experimental results having an average error of 6.1%. Both were in a good agreement especially considering average values of ER, fluidizing velocity etc. for the model and different values for the experimental one. In this modelling, syngas composition and CGE were sensitive to changes of ER, while the energy production was affected by changes of net electrical efficiency.

5 Process-oriented modelling at a system level

How can process-oriented LCA modelling be applied in a regional context to assess energy supply and associated environmental performance of technological solutions at system level? Based on Lodato et al. (**IV**).

LCA is applied to assess the consequences of producing methane from residual bioresources. A stepwise modelling approach is introduced aiming to support strategies for bio-based methane production on a region to supply the fossil gas demand in an environmental efficient way.

The first step set the basis for the LCA, with the goal and scope, bounderies, impact categories, assessment method, and identification of the annual gas demand.

The second step identifies and quantifies residual bioresources available on a region based, for example, on regional inventories or on advanced geographical information system-based models (e.g. Jayarathna et al., 2020). Subsequently their characterization is defined through the chemical, biochemical, physical, and nutritional properties (e.g. dry matter, moisture content, cellulose, inorganic matter).

The identification of technologies for methane production is part of the third step. Both biological and thermochemical degradation of bioresources are considered to be able to use all the carbon available. For example, dry and semi dry bioresources, with less than 40% of moisture content (e.g. Thunman et al., 2018), are used in the gasifier, and bioresources with a moisture content preferable higher than 70% (Weiland, 2010) are digested anaerobically. This represents a requirement for allocation of bioresources toward one or the other technology. Both AD and GA include the upgrading of methane to reach suitable conditions for injection into natural gas grid, such as CH_4 > 98% (Tyra et al., 2020). For AD two upgrading technologies are considered: i) removal of carbon dioxide in the biogas through water scrubbing, the most commonly used upgrading (Angelidaki et al., 2018), ii) ex-situ upgrading with hydrogen, where hydrogen produced through alkaline electrolyzer (Thema et al., 2019) is coupled with carbon dioxide from the biogas in a separate unit in presence of a

nickel-base catalyst (Zhang et al., 2017). In the gasification, upgrading with hydrogen was also considered.

For the step four, the process-oriented modelling is applied in EASETECH. LCA scenarios are built. Here, the conversion of syngas to methane was modelled following the EASETECH+ modelling framework. The input-output relationships considered hydrogenation reactions, i.e. methanation ($CO + 3H_2 = CH_4 + H_2O$) and Sabatier ($CO_2 + 4H_2 = CH_4 + 2H_2O$). Details regarding the modelling are in Lodato et al., (**IV**).

The environmental assessment is accounting results for all selected impact categories. The environmental evaluation is done through two sequential layers.

For step five, layer 1 results are presented at technology level, relative to the production of 1 Nm^3 of CH_4 with injection and gas grid distribution. The environmental evaluation at a technology level considers: i) the technology performance with the management of co-products and rejects; ii) alternative routes for methane production accounting for the current management of the residual bioresources and the induced effects. These are different for each category of residual bioresource. For example, the counterfactual of cheese whey is animal feeding. This needs to be modelled together with the avoided crops produced for feeding involving land use changes effects. iii) The conventional production of gas, i.e. natural gas production. Thus, at a technology level, the environmental performance is useful for identification of environmental hotspots at unit-process within each conversion pathway (which is not possible with traditional approaches), comparing technology pathways, and with the associated counterfactuals. Trade-off for each impact categories are also evaluated. Net balance of LCA scenarios (i.e. *conversion pathway – counterfactual*) (Tonini et al., 2019) is also compared with the conventional production of gas. LCA scenario with a performance less than the conventional production of gas may be excluded as potential solutions for fulfilling the functional unit.

Subsequently, the last step, step six, provides layer 2 of results at a system level. Here, combinations of scenarios for each category of bioresource is done, considering the high methane production and the lower global warming emissions. Evaluation at a system level need to fulfil the goal of the study, supplying the gas demand of the selected region. However, there are two possible cases: i) a higher production of bio-based methane than gas demand, thus the gas demand is met and bio-based methane can be exported, ii) a lower produc-

tion of bio-based methane than the gas demand, thus methane need to be imported. Conversion technologies and carbon pool are important factors to be accounted in the evaluation at a system level.

The example in Lodato et al., (**IV**), provides strategies for bio-based methane gas supply in the context of a French region Occitania. 41 residual bioresidues were identified based on regional inventories (Couturier et al., 2019; Vinel at al., 2019) divided in 10 categories (Figure 9). The potential amount of residual bioresources was of about 25 Tkg wet basis.

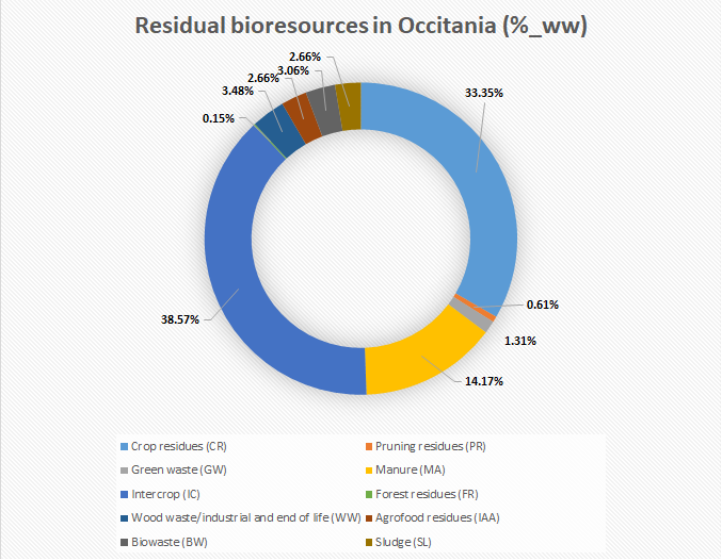

Figure 9: Residual bioresources available in Occitan region of France in 10 categories and its share (%_ww: percentage wet weight)

The functional unit was: "to *fulfil the annual demand for methane gas in the French Occitan region, to the extent possible with local residual resources or else with imported natural gas*" (Lodato et al., **IV**). Three conversion pathways were identified: I) *ADH*, anaerobic digestion with hydrogen upgrading; ii) *AD*, anaerobic digestion with water scrubber upgrading; and iii) *GA*, gasification with carbon to methane (C-to-CH4) upgrading with hydrogen (Lodato et al., **IV**).

Examples at technology level (performance of individual combinations of feedstock and conversion processes) and at system level (potential environmental impacts at system level fulfilling the supply of gas demand): Figure 10 (Lodato et al., **IV**) shows, on the left, contribution to global warming (GW)

results (other impacts categories were also accounted) for each category, conversion pathway, counterfactual, and conventional gas production. On the right, net balances for each category. At a technology level, savings were given mainly from material and energy substitution, while emissions were mainly from gas production and use on-land, storage, and ploughing. In Lodato et al., (**IV**), there are results interpreted also for the other considered impact categories.

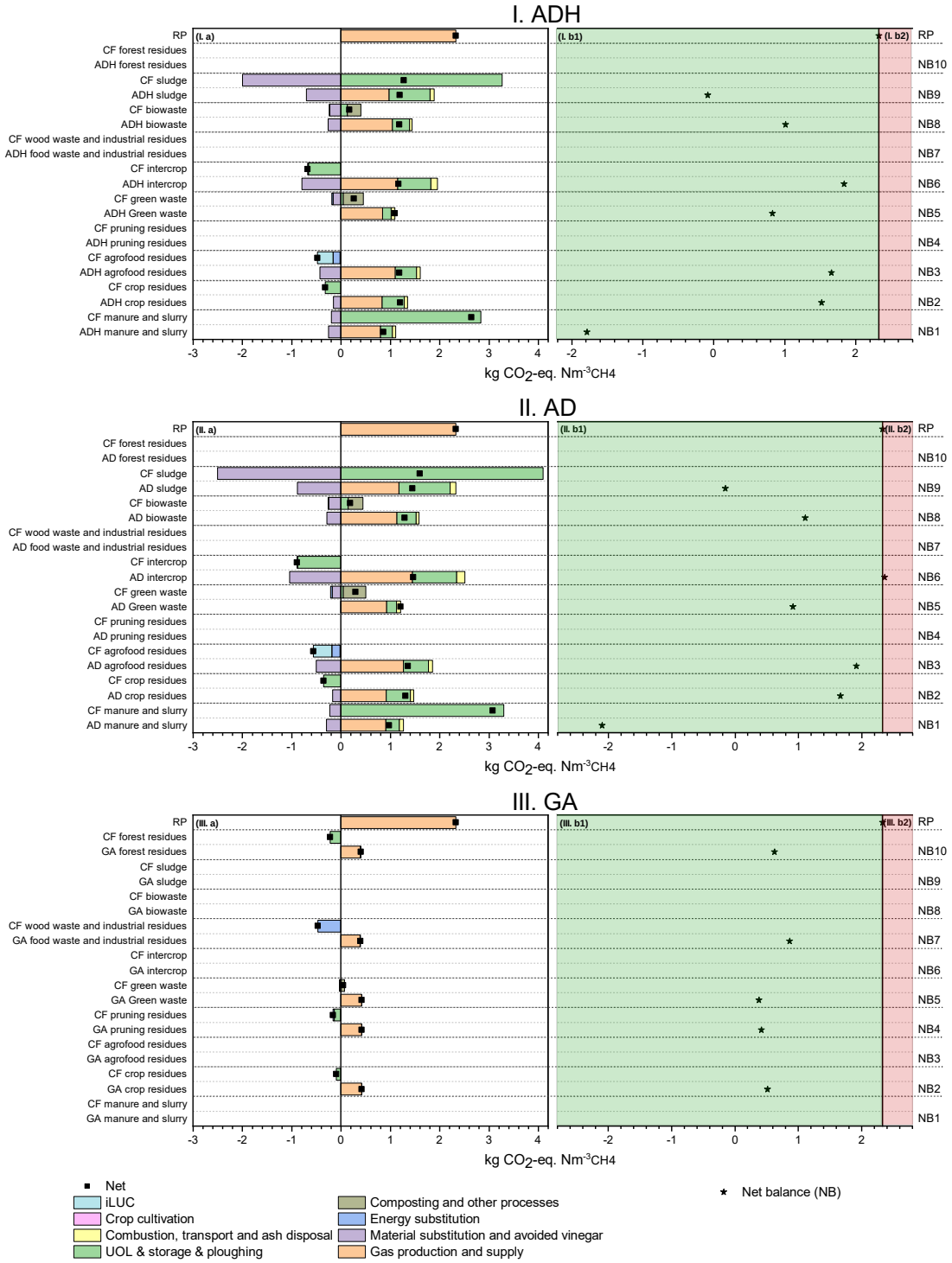

Figure 10: Global warming (GW) results at a technology level. On the left, contribution results for three conversion pathways (ADH, anaerobic digestion with hydrogen upgrading, AD, anaerobic digestion with water scrubber upgrading; GA, gasification with C-to-CH4 upgrading). On the right, net balance (NB) for each scenario compare to the reference product (RP). In the red area there are scenarios that have a worse performance than the fossil RP, while in the green are there are scenarios with a better performance. (iLUC = indirect land use change; UOL = use on land)

At a system level, bio-based methane and global warming results from each category were accounted (Table 2) fulfilling the functional unit of 17.5 TWh annual based and addressing the assessment goal. In general, GA had a higher methane yield and GW emissions compared to AD and ADH. Due to high based methane production in Occitania, exports to neighbouring region are possible. Finally, gasification is seen as a promiscuous technology. More details and results on the combined scenarios are in Lodato et al., (**IV**).

Table 2: Bio-based methane and global warming (GW) results for each category of bioresources in Occitania

		Crop residues (CR)	Pruning residues (PR)	Green waste (GW)	Manure (MA)	Intercrop (IC)	Forest residues (FR)	Wood waste (WW)	Agrofood residues (IAA)	Biowaste (BW)	Sludge (SL)
ADH	CH_4 in the gas grid (TNm^3)	1.51	-	0.038	0.23	0.27	-	-	0.04	0.14	0.02
	GW net ($Mt\_CO_{2\_eq}$)	2.29	-	0.03	-0.41	0.50	-	-	0.07	0.15	-0.002
AD	CH_4 in the gas grid (TNm^3)	1.37	-	0.03	0.20	-	-	-	0.03	0.12	0.02
	GW net ($Mt\_CO_{2\_eq}$)	2.28	-	0.03	-0.42	-	-	-	0.06	0.14	-0.003
GA	CH_4 in the gas grid (TNm^3)	5.26	0.01	0.23	-	-	0.03	0.61	-	-	-
	GW net ($Mt\_CO_{2\_eq}$)	2.70	5.72	0.09	-	-	0.02	0.53	-	-	-

6 Discussion

In this section, selected aspects of the PhD thesis are discussed.

The importance of input material properties throughout different conversion pathways is accounted by the process-oriented technology models. Input material properties contributing to product yields are linked to process performance. For example, the biogas yield depends on origin of the feedstock, contents of organic substrate, composition. amd degradability. This is in accordance with Weiland, (2010), which showed that a higher lipids content provides a higher biogas yield, compared to proteins and degradable carbohydrates. While lignin is not degradable anaerobically, it is preferable for thermochemical degradation together with low moisture content (Kataki et al., 2015). Allowing flexibility for LCA practitioners to adjust input material feedstock composition to specific case-studies, without changing the inventory model itself, and at the same time be able to apply different types of input data (e.g. theoretical or experimentally based) in the same model, potentially offers a more user-friendly approach to modelling. While the underlying technology models may be more complex than before, naturally the models should be documented and validated thoroughly before deployment.

Typically, LCA of bioenergy technologies are more product-oriented than process-oriented. A process-oriented approach reflects different conversion pathways with different process configurations to produce a determined product. EASETECH+ modelling framework allows the modelling of unit processes for further implementation into EASETECH. Therefore, the LCA model EASETECH can have ready-to-use bioenergy technology models. To reproduce chemical processes and evaluate environmental impacts of operation conditions other studies coupled LCA tools with process simulation software (e.g. Morales-Mendoza et al., 2018). However, performing the process simulation and the LCA separately does not allow a direct analysis of the influence of parameter uncertainty on the LCA results. Integrating the process-oriented technology models in EASETECH considerably expands the possibilities of evaluating process parameters and extends the scope of the uncertainty analysis. Ultimately, this can provide much more robust and industry-relevant LCA results.

Unit processes modelled following the process-oriented approach may have parameters in both inputs and process flows. Parametrization of conversion

processes linking input properties to output products can include both correlation and uncertainty propagation among different input parameters (Bisinella et al., 2017; Groen et al., 2016) and uncertainty propagation from technical parameters (Bisinella et al., 2016; Clavreul et al., 2012). This may allow identification of new parameter types and process performance aspects that previously were not realised although these parameters and aspects may critically influence the LCA results. This may assist technology development and upscaling activities in pinpointing key process steps of importance for the overall environmental performance of the technology.

The subdivision into unit-processes promotes material recirculation, e.g. in the perspective of power-to-X technology systems often involving a variety of processes with multiple outputs and interdependence between unit-processes. As such, the process-oriented modelling approach can support specifically the conversion of renewable energy into chemical energy in combination with other production routes for biofuels, biomaterials, and biochemicals (Buffo et al., 2019; Khalilpour, 2018).

Technologies and processes modelled with the EASETECH+ framework inherently require detailed formulation of the involved process relationships, input material conversions, process dependencies, output generation, etc. Links between input and output flows for unit-processes have to be specified by mathematical expressions before implemented into EASETECH+. While these mathematical expressions and relationships in the model have to be documented, this approach supports the establishment of the common understanding of multi-output and multi-functional processes that is necessary to make the process reproducible (Kuczenski et al., 2018). As such, the process-oriented modelling approach is in accordance with the ISO 14048 (ISO, 2006 c), which highlights the important requirement of having a complete documentation of databases and unit-processes.

7 Conclusions and recommendations

The PhD project focused on developing, validating and implementing the modelling framework EASETECH+ to establish detailed biochemical and thermochemical LCA models relating feedstock composition with expected products (biogas, digestate, solid fuels, ethanol, syngas, etc.) and consumptions (for operations) for appropriate representations of actual technologies. The implementation of state-of-the-art models into EASETECH was realised by applying process-oriented LCA modelling on selected energy technologies for quantification of the associated environmental impacts. Ready-to-use bioenergy technology models allowing flexible adjustment to specific case-studies were established for further use within the LCA community. The main findings of the research can be summarized as follow:

(1) The process-oriented modelling framework can be applied on a technology level or a system level, according to the available data and details needed in respect of the LCA goal and scope. Results at technology level identify technology hotspots, evaluate and compare the performance of individual conversion pathways. Results at system level identify combined scenarios for fulfillment of the scope of the study.

(2) The process-oriented modelling framework provides results linked to actual process parameters and to the quality of the input material (physical, chemical, biochemical and nutritional properties). Mass, substance and energy balances are preserved with linear and non-linear responses defined by mathematical relationships. The process flow diagram with associated mass, substance and energy balances can be verified while building the process-oriented models to ensure correct implementation into EASETECH. (Multi-)conditional sequence flows are allowed, enabling and disabling flows according to data availability. Material recirculation and loops are allowed.

(3) State-of-the-art, ready-to-use process-oriented bioenergy technology models developed during this PhD project include: i) a biorefinery (i.e. pretreatment, hydrolysis, fermentation and distillation, separation of solid and liquid fraction for recovery) (Lodato et al., **I**); ii) a bubbling fluidized bed gasifier (i.e. gasifier, gas combustor an air pollution control system) (Ardolino et al., **III**); iii) a circulating fluidizing bed gasifier (with a stoichiometric equilibrium model) (Lodato et al., **IV**); iv) an anaerobic digestion (Lodato et al., **II**); v) a C-to-CH4 upgrading (Lodato et al., **IV**).

A stepwise modelling approach for assessing environmental performance of regional bio-based energy supply, applied to the Occitania region in France, for supply of bio-methane involving assessment both at technology and system levels. This provided practical support to import/export strategies towards non-fossil gas and local use of residual bioresources. Based on the included case-studies, the following recommendations are provided:

(1) Feedstock properties and interdependencies between unit-processes of bioresource conversion technologies may be critical for LCA results. For example, the properties of the input feedstock (e.g. moisture content, hydrocarbon content), and the type of reactor with operational conditions (e.g. temperature, fluidizing agent) provide different rejects, emissions, and product yields with contributions to the environmental performance. It is recommended that LCA models appropriately reflect this in the future, e.g. by using the modelling framework provided here.

(2) LCA modelling of technologies prior to commercialization and full-scale implementation should involve appropriate process-oriented modelling and selection of operational process parameters. This analysis should be done to reproduce the technology with a higher level of details aiming at identifying process hotspots within the technology. Sensitivity and uncertainty propagation can be carried out at the level of the detail of the unit-process.

(3) As with any other LCA model, also process-oriented LCA models should be appropriately documented and validated to ensure transparency. Applying an existing and well-documented process-oriented model on new case-studies may expedite this considerably for LCA practitioners by saving time identifying process relationships and formulate associated mathematical expressions for the model.

(4) In bioenergy technologies the management of co-products and rejects should be accounted, together with current management of residual bioresources with its induced effects. For example, for biogas savings and burdens associated with digestate use, and effects from diversion of biomass residue from their current management should be accounted.

(5) Net balances should be calculated to provide direct comparison between case-study LCA scenarios and the associated reference products.

8 Future perspectives

Based on the knowledge and experiences gained during the PhD project, the below topics represent a range of suggestions for future research activities to further develop the process-oriented LCA modelling:

- Improving EASETECH+ and extending the process-oriented modelling approach to a wider range of technologies, e.g. thermal pyrolysis, biochemical, biomaterial, and biofuels production facilities. This requires identification and appropriate implementation of relevant process relationships between input resources and materials (e.g. chemicals, energy, etc.) and process outputs and emissions. Expanding LCA process-oriented technologies facilitates the environmental analysis of more integrated systems of technologies, such as power-to-X.
- Uncertainty evaluation of bioenergy technologies to provide more robust results, and to identify the most critical parameters in respect to the input feedstock, operational conditions, conversion efficiencies within (multi-)conditional sequence flows.
- Applying the process-oriented modelling approach to technology development and upscaling activities, enabling early process optimization, considering upstream and downstream production to meet changes in supply and demand, and documentation of environmental performance.
- Applying EASETECH+ modelling framework to reproduce technologies and systems of technologies promoting material loop to reduce input materials and waste outputs.

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10 Papers

I A process-oriented life-cycle assessment (LCA) model for environmental and resource-related technologies (EASETECH)

II A modelling framework for process-oriented life cycle assessment (EASETECH+)

III Energy recovery from plastic and biomass waste by means of fluidized bed gasification: A life cycle inventory model

IV Framework for assessing environmental performance of methane gas supply in the context of local bioeconomy

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I

A process-oriented life-cycle assessment (LCA) model for environmental and resource-related technologies (EASETECH)

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LCA OF WASTE MANAGEMENT SYSTEMS

A process-oriented life-cycle assessment (LCA) model for environmental and resource-related technologies (EASETECH)

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Abstract

Purpose In life-cycle assessment (LCA), environmental technologies are often modelled as "black-box processes", where inputs and outputs are typically not linked through physical and/or (bio) chemical relationships. This limits transparency and usability of environmental modelling of resource systems for which the conversion of materials and chemical substances in the materials is essential for the environmental performance. We introduce an advanced "process-oriented" modelling framework allowing quantitative and parameterised physical-chemical relationships between input material composition, conversion process units and subsequent output products, promoting mass and substance balanced conversion modelling and environmental assessment.

Methods A dedicated LCA model, EASETECH, has been used to provide a user-friendly platform for performing advanced LCA of complex technologies, without the need for additional software/tools. In the modelling framework, the technology is subdivided into individual unit processes. In each process, the characterisation of the input feedstock material into biochemical, physical, chemical and nutritional properties is taken into consideration in each multi-output production flow. For each unit process, the processes governing the mass/energy/substance transition and transformation are described by mathematical equations (i.e. relationships between inputs and outputs) through the use of parameters. A range of new operators were developed to establish these relationships that allow for non-linear responses whereby changes in one flow can give a non-linear response in other flows. The modelling framework and the involved operators are explained and applied to a biorefinery case study.

Results and discussion The model facilitates "tracking" of the feedstock material properties from the input to the final products, by establishing mass, substance and energy balances for each conversion unit process. In addition, the process-oriented modelling framework appropriately represents material/substance transition and transformations. The choice of process parameters has considerable importance for the overall results. This was illustrated by one-at-a-time changes in parameter values in two different biorefinery unit processes (i.e. hydrolysis, and fermentation and distillation). In addition, the relevance of feedstock characteristics for the performance of the individual unit processes was proved with fixed parameter sets with different feedstocks. The biorefinery case study demonstrated that the LCA model can be applied to technology cases with different process configurations (e.g. different efficiencies) and different input feedstock properties, where it automatically adjusts to these changes in properties.

Conclusions The advanced process-oriented modelling framework offers more flexible modelling of the conversion technology than previously available, improved options for technology development in view of environmental performance, and potentially more accurate results. This provides a significantly improved basis for environmental modelling and decision-making in relation to resource systems.

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Abbreviations

CT	Composite transformer
FD	Fraction distributor
FG	Fraction generator
FH	Fraction hub
FT	Fraction transformer

GW Global warming
LCA Life-cycle assessment
LCI Life-cycle inventory
MD Material distributor
MF Material flow
MG Material generator
NG Natural gas
RED Renewable Energy Directive
RF Residue flow
SD Substance distributor
SG Substance generator
SH Substance hub
ST Substance transformer

1 Introduction

Life-cycle assessment (LCA) represents a standardised and systematic methodology for assessing the environmental performance of technologies and technology systems (ISO 2006a, b; EC-JRC 2010). In the transition to a more resource efficient and sustainable society, e.g. represented by circular (bio) economy initiatives (European Commission 2017; Zabaniotou 2018) and the European sustainability targets (e.g. European Parliament and the Council of the European Union 2009), appropriate management and utilisation of waste materials and residual resources in society are critical in order to minimise losses, maximise environmental savings and avoid suboptimal solutions at societal level. Waste and residual resources represent complex and heterogeneous materials with a wide range of physical and (bio) chemical properties. Recovery and conversion of such materials into secondary raw materials and new valuable products rely on the specific characteristics of these materials, and the environmental benefits associated with potential management solutions are highly affected by the material properties themselves (Bisinella et al. 2017). LCA modelling of residual resource systems, therefore, should not only account for the resource characteristics but also reflect relationships between input material properties and the output products for a wide range of different conversion technologies and process configurations. This puts considerable demands on LCA modelling of resource systems to ensure transparency and flexibility in modelling.

A wide range of (non) commercial LCA models is available for environmental assessment (e.g. SimaPro 2019; Thinkstep Gabi 2019; TEAM 2019; Umberto NXT LCA 2019; for a more complete list, see EPLCA 2019). While most of these modelling tools are primarily targeted environmental assessments of products and manufacturing, rather than systems comprising several technologies involving material flows and conversion of material resources through physical, chemical and biological processes, the majority of these tools follow a so-called black-box modelling approach where embedded data inventories represent individual technologies with a fixed list of inputs and outputs. This means that the user is limited to the technology assumptions "embedded" in the inventories. As differences in modelling assumptions (e.g. technical assumptions, technology type and the inventories used) lead to differences in LCA results (e.g. Gentil et al. 2010), this is a crucial aspect that has particular importance in relation to resource systems and when the technologies themselves are in focus (e.g. Astrup et al. 2018; Henriksen et al. 2018). A few LCA models are specifically designed to evaluate material and resource flow systems (e.g. Jain et al. 2015), with EASETECH being a notable example for LCA of environmental technologies (Clavreul et al. 2014). Using principles from material flow analysis (MFA), EASETECH keeps track of mass, substance and energy flows throughout a system of processes and technologies represented by a scenario (Clavreul et al. 2014). However, EASETECH is focused on modelling of linear material and substance flows, but does not allow accounting of interactions between individual materials and substances nor the transformation of substances themselves. This interaction is needed in case of technologies involving conversion of substances and materials, and where flows and transformations are linked to the amount of specific materials entering a process. As such, there is a need for LCA modelling frameworks allowing constraints, non-linear relationships and new substances to be created as a result of biological and chemical reactions, while maintaining the overall mass, substance and energy balance of the model.

Black-box models can be defined as a combination of one or more single-operation unit processes aggregated into a fixed list of inputs (energy, materials and chemicals) and outputs (products, emissions and residues) with no direct relationship between inputs, outputs and process operations (EC-JRC 2010). The evolution from product LCA to process LCA has taken time seeing the process as black-box, thus limiting the analysis of unit processes within complex systems (Jacquemin et al. 2012). Recently, this challenge has been highlighted by Maes et al. (2015) who explained how black-box modelling approaches present considerable limitations to application of the EU renewable energy guidelines (European Parliament and the Council of the European Union 2009) when applied on complex production sites, mainly because black-box models cannot appropriately represent the individual unit processes and therefore do not identify the impacts associated with these unit processes. For resource conversion technologies such as biorefineries, this means that no specific links exist between the input feedstock composition, the subsequent transformation of feedstock properties occurring within the individual unit processes, and the final outputs and emissions from the biorefinery. This is in contrast to real processes in which all these aspects are directly interlinked. As such, the LCA models cannot account for potential changes in feedstock composition between case studies, nor for changes in

performance of the involved unit processes. Limiting LCA models to fixed technology aggregations and inventory data, thereby significantly limits the applicability of the LCA model, but also reduces the transparency of the model and requires new inventory datasets to be developed for each case study.

To overcome the need for implementing inventory datasets according to the specific technological, geographical and temporal scope of an assessment, several approaches have been applied in literature: (a) relatively simple MFA methods for determination of material flow and emission partitioning within technologies and across a system of technologies (e.g. Mancini et al. 2015; Turner et al. 2016), and (b) more advanced process simulation tools (ASPEN 2019; ProSim 2019; ProMax 2019; CHEMCAD 2019) to evaluate individual biological, physical and chemical unit processes within a technology (e.g. Tumilar et al. 2016). While these approaches and tools certainly have merits, the definition of the technology inventories remains separated from the LCA modelling itself. A few studies (e.g. Arora et al. 2016; Brunet et al. 2012; Gaha et al. 2017) have attempted to combine LCA modelling with the process simulation tools mentioned above and/or with mathematical programming tools (e.g. MATLAB). While this potentially allows a more detailed process-oriented approach (as opposed to black-box datasets), these models are typically not integrated with the LCA tool and need to be run separately, often requiring specific insights in the programming itself (i.e. limited user-friendliness) (Asprion and Bortz 2018). While such integration is desirable, so far, we are not aware of tools that allow modelling of unit processes of complex technologies and concurrently performing a full LCA.

To further advance and facilitate LCA modelling of more complex and integrated resource management technologies and systems, LCA models should allow the establishment of quantitative relationships between input feedstock composition, unit processes, and subsequent outputs of products and emissions. This means "opening-up" the black-box models and allows the definition of useful relationships between inputs, outputs and process configurations. While subdivision of complex technologies into unit processes is supported by current LCA guidelines (EC-JRC 2010), such a modelling approach is here termed "process-oriented" LCA modelling. Modelling of residual resource technologies like biorefineries requires detailed data of the input material (e.g. water content, energy content), the transformations of materials or substances during processing, and the transition of mass from one flow to another. To enable transparent and flexible adjustment of the model to a specific case study, the involved model parameters should reflect subdivision in relevant unit processes (e.g. for a biorefinery: pre-treatment, hydrolysis, fermentation and distillation, separation and recovery of the solid and liquid fractions). In an integrated technology system with several flows associated to multiple product outputs, working with parameterised unit processes and input-output process relationships allows to change a specific production flow and have a non-linear response in other flows such as increasing or decreasing their production and associated emissions. Currently, no existing publically available LCA model offers such process-oriented modelling approach relevant for resource-centric technologies and systems, although some models enable interaction with external software to allow users some degree of taking these aspects into account.

The aim with this study is to advance LCA modelling of integrated technologies and technology systems targeting environmental assessment of resource management by implementing advanced "process-oriented" LCA modelling. The following specific objectives are addressed: (i) provide a framework for process-oriented LCA modelling of multi-output conversion technologies, (ii) define the needed operators and implement these in the software EASETECH, (iii) demonstrate the applicability of the modelling framework on a simplified biorefinery case study, focusing on global warming impacts in combination with the importance of feedstock characteristics and unit process parameters (e.g. conversion efficiency) under specific operating conditions, and finally on this basis (iv) evaluate the perspectives and implications of the proposed advanced process-oriented modelling approach. The outcome of the study represents the methodological basis for advanced mass, substance and energy balanced LCA modelling to resource technology systems in EASETECH.

2 Material and methods

2.1 Principles of process-oriented LCA modelling

The characterisation of the input feedstock into individual fractions, each with associated biochemical, physical, chemical and nutritional properties, is the point of departure of a process-oriented LCA. Subdividing a material flow according to properties enables modelling of the conversion (or "fate") of these properties within a specific process, technology or an entire system of several technologies, and linking the input feedstock to the associated outputs generated by the involved processes. These material properties thereby represent an extension of the substances used within MFA (Allesch and Brunner 2015; Brunner and Rechberger 2016), e.g. carbon is a chemical element and cellulose is a compound; both of them are properties of the biomass feedstock: the carbon content takes into consideration the carbon content of cellulose, representing a part of the total carbon in the biomass. Conversion of the input feedstock is associated with either transition or transformation of feedstock properties. Transition occurs when a specific amount of a material or fraction or substance (and thereby share of material

properties), usually expressed in percentages, is transferred from an input to an output of a process. The transition within a process can be partial (less than 100% of a material flow is transferred) or total when the entire material flow is transferred. Transformation of the input feedstock material occurs when one or more fractions or one or more substances has a change in its composition within a process. Thus, some fractions/substances may cease to exist, while new ones may be introduced. Also, in this case, the transformation can be total, when a fraction or substance is entirely used in a transformation, or it can be partial when only a defined quantity of a selected substance/fraction is involved in the conversion process. Consequently, the original material prior to the transformation does not exist anymore because a different material is generated departing from it, however maintaining the overall mass, energy and substance balance of the process. Moreover, mass transition and transformation within a system are linked to environmental exchanges that subsequently are converted into environmental impacts. For example, in a process where mass and energy are given by the material conversion of the process itself, considering emission factors during the characterisation phase (after the inventory) allows emissions to be quantified according to the availability of the substance/mass/energy involved in different material flows within the considered unit process. In addition, in the process-oriented model, the technology is subdivided into individual unit processes. For each unit process, the (bio) physical processes governing the mass/energy/substance transition and transformation are identified and described by mathematical equations. These equations allow the establishment of relationships and interdependencies between the input feedstock material properties and the (unit) process outputs. Parameters can be applied to allow adjustments of material flows and process performance to specific cases. If a parameter is in an equation, it can directly affect its result, and thus the conversion process, the substance/mass/energy flow and the respective emission. Furthermore, the proposed framework allows for non-linear responses whereby changes in one flow can give a non-linear response in other flows.

Figure 1 illustrates the generic black-box vs process-oriented technology modelling: in the black-box modelling approach (Fig. 1a), the technology is described by an input and several outputs represented by $Input_1$, the products P_1, P_2, P_3 and the emission E_1.

Any relationships and interdependencies between the input feedstock material and the output products are not represented by the model. In addition, the technology is not subdivided into unit processes and relationships/interdependencies represented by equations containing parameters are not included. On the contrary, in the process-oriented modelling approach, the technology (Fig. 1b) is described through input feedstock material properties (e.g. In_1, In_2, In_3), relevant unit processes (process *A*, *B*, *C*, *D*), and relationships between input feedstock material properties and process outputs using equations with parameters. In Fig. 1b, the output product P_B is partially transferred to process C, i.e. P_B has an associated conversion efficiency (eff_1) in the transition from process B to C. ($1\text{-}eff_1$) represents what is left, i.e. residue, of P_B subsequently transferred to process D. An example of total transition is represented by the product P_C, totally transferred to process D together with the residues of process B. P_2 and P_3 are two products of process D generated through two equations ($P_2 = P_C * par_1/par_2$; $P_3 = Residues * par_3$). These equations are two examples of relationships between inputs and outputs within process D and par_1, par_2 and par_3 are the three associated parameters. As an example, Fig. 1c illustrates this modelling approach implemented on a second-generation biorefinery where the lignocellulosic input feedstock is converted into bioethanol, a solid and liquid fraction, and CO_2. The feedstock is characterised according to relevant biochemical, chemical, physical and nutritional properties (e.g. cellulose, proteins, carbon content, energy content, water content, digestibility, etc.). The overall biorefinery technology is represented by a range of unit processes: (1) pre-treatment, (2) hydrolysis, (3) fermentation and distillation and (4) recovery. In the entire biorefinery system, both transitions and transformations occur, and the relationships between the input feedstock properties and the output products are identified and described by appropriate equations involving adjustable parameters (e.g. conversion efficiency of C6 sugars, eff_{C6}, into ethanol production, thereby facilitating flexible adaption of the model from one case study to another). An example of material transformation is given by the hydrolysis, where polysaccharides, such as cellulose, pectin, hemicellulose, starch and sucrose, are converted into simple sugars with five and six carbon atoms (C5 and C6 sugars). While one substance (polysaccharides) thereby is transformed into another substance (monosaccharides) and thus cease to exist, the overall mass and substance balance of the technology is maintained and the flows are trackable. In the Electronic Supplementary Material (ESM) (Sections S2 to S6), these biorefinery unit processes are thoroughly described including the transformation equations used.

The process-oriented modelling approach allows users to establish models with all the necessary unit processes involved, to clearly define the feedstock conversion and to include appropriate modelling parameters and assumptions. This is useful particularly in studies that wish to base the assessment on pre-developed models reproducing specific technologies such as lignocellulosic biorefineries, but intend to apply case-specific process performance data and/or update the model to reflect assumptions more relevant for the case study in question.

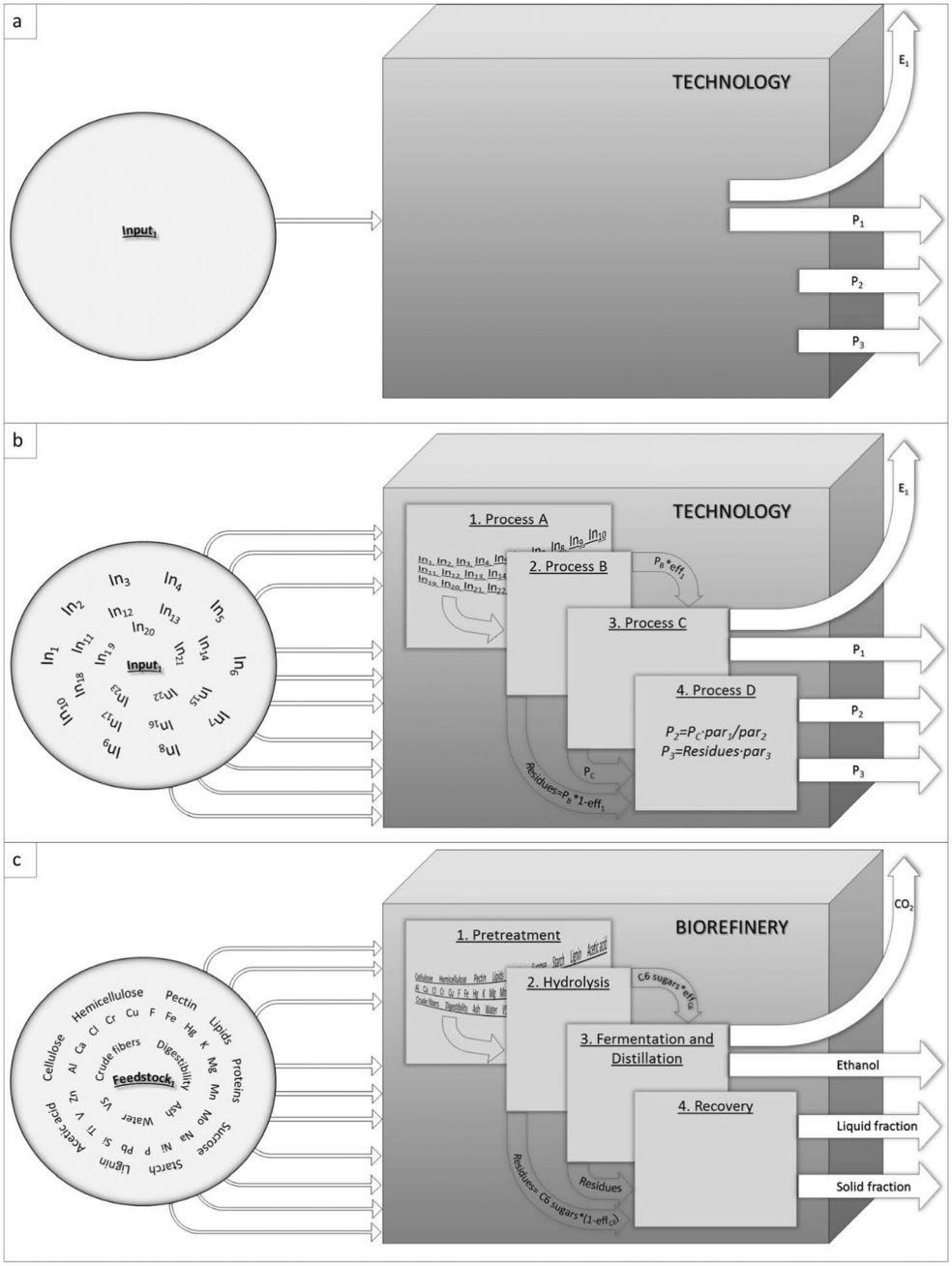


Fig. 1 **a** Black-box modelling approach applied to a generic technology; $Input_1$ is the input process; P_1, P_2 and P_3 are the output products and E_1 is an example of technology emission. **b** Process-oriented approach applied to the same generic technology; same input, final output products and emission (i.e. $Input_1$, P_1, P_2, P_3, E_1); the input properties (e.g. In_1, In_2) are considered. In addition, relationships/interdependencies are established between the technology unit processes (i.e. *Process A*, *B*, *C, D*) and described with equations containing parameters (Eq. 1: $P_2 = P_{A2}par_1/par_2$ and Eq. 2: $P_3 = P_{B1}par_3$, with par_1, par_2 and par_3 as parameters). **c** Process-oriented approach applied to the case of a second-generation biorefinery. The unit processes considered are *pretreatment*, *hydrolysis*, *fermentation and distillation*, *recovery*. The input is $Feedstock_1$, having properties (e.g. cellulose, Ca). Relationships/interdependencies are described through equations with parameters; the final whole-system output products are *ethanol*, *liquid fraction* and *solid fraction*. CO_2 represents an example of emission

2.2 EASETECH modelling features supporting process-oriented modelling

To facilitate process-oriented LCA modelling in EASETECH, a range of new "operators" were developed following the principles of domain-specific language illustrated in Zarrin and Baumeister (2014). The new operators allow a domain expert (a person with the relevant technological and systemic expertise) to establish the relationships between input and output for the individual unit processes, described in the previous section. In EASETECH, LCA scenarios are characterised by a number of "process modules" that are connected with arrows indicating material flows between the processes (see Section 2.3 for further details). Process modules may represent individual unit processes or entire technologies and can be nested, i.e. a number of "unit-process modules" may be "packed" into another process module. As such, the scenario building in EASETECH follows the overall principles of MFA; for details, see Clavreul et al. (2014) and Allesch and Brunner (2015). These principles are also applied to the unit processes modelled involving the new operators and subsequently implemented into EASETECH. Table 1 provides an overview of all new operators, while the remainder of this section explains the key features of the operators. Further

Table 1 Operators available in EASETECH and their application for the modelling of processes within technologies and systems

Operator	Application
Material flow [MF]	MF transfers material from a source element to a target element. It is allowed using more than one MF from the same source element
Residue flow [RF]	RF transfers what is left in a source element (residue). It is allowed using only one RF from the same source element
Fraction distributor [FD]	FD extracts a fraction from a material
Fraction generator [FG]	FG generates a fraction in a material
Fraction hub [FH]	FH groups fractions from an input material
Fraction transformer [FT]	FT transforms a fraction into another one within a material. As a consequence, the previous fraction does not exist anymore
Material distributor [MD]	MD extracts a material
Composite transformer [CT]	CT groups more than one operator. It allows iterating a sequence of transformations and transitions
Primitive parameter	It generates a parameter (numeric or string)
Data table parameter	It generates a table of parameters; it may contain one or more data columns
Data column	It generates columns into a data table parameter; each column refers to a parameter
Material generator [MG]	MG generates a material that may contain one or more material fractions
Input	It contains all the initial inputs (starting point)
Output	It contains all the final outputs (ending point)
Substance distributor [SD]	SD extracts a substance within a fraction
Substance hub [SH]	SH groups substances from fractions
Substance transformer [ST]	ST transforms a substance into another one; consequently, the previous substance does not exist anymore
Substance generator [SG]	SG generates a substance

details describing the individual operators applied for the modelling of the biorefinery case study (see also Section 2.3) unit processes can be found in the ESM, Sections S2.7, S3.1, S4.1, S5.1 and S6.1.

The following three macro levels are considered in the model: materials, fractions and substances. Materials, following the MFA definition, contain both substances and goods. In this case, goods represent fractions, "entities" that share common characteristics, i.e. substances. As such, "grass", "branches" and "wood" may all represent fractions in a material called "garden waste", while substances represent chemical, nutritional, physical and biochemical properties (e.g. cellulose, proteins, lower heating value, methane potential, digestible energy). Some of the substances may be correlated, e.g. the energy content of a fraction is a function of the content of cellulose, proteins, etc. Physical, chemical, nutritional and biochemical properties are assigned to the substance level, although they are not necessarily substances as such (e.g. energy is not a substance, but it is modelled using the same operators as for substances).

For the individual process module, there is at least one input and one output. There are three possible input types: (i) an output from another process, (ii) a material consisting of several fractions or (iii) a single fraction. The anaerobic digestion of organic waste is an example of the first case; it generates biogas and digestate as final outputs: the digestate may then be used as input to a subsequent fertilisation process. For the second case, e.g. a material (e.g. garden waste) with multiple fractions (e.g. grass, wood), the operator that generates the input feedstock material is *material generator* (MG); then, a *fraction generation* (FG) is needed for generating each fraction within the input material. Thus, we are generating the material composition. The last case, when the input is a single fraction (e.g. grass), only an FG is applied to generate the fraction. Lastly, a *substance generator* (SG) is used to specify each input material property, i.e. chemical, biochemical, physical and nutritional. Each of these properties is modelled as substances within a fraction. A range of physico-chemical relationships, represented by mathematical equations, are applied when a substance or a fraction is "transformed" within a process, e.g. one or more substances are converted into a specific product (e.g. glucose to ethanol) that may be the final output of a process or an intermediated product subsequently used in another conversion flow. The following operators are used for this purpose: *substance transformer* (ST) and *fraction transformer* (FT) when the transformation is related to a substance and a fraction, respectively. With these operators, a selected substance or fraction involved in the conversion process can be specified not to exist anymore while another substance or fraction is generated in its place, i.e. transformation from one entity into another. However, a transformation may also represent a modification of the substance or fraction content by changing only its amount while still preserving the substance or fraction itself. It is possible to change the content

of a substance: the substance is the same and its amount is different (e.g. decreasing the water content of 50% of the original value).

Furthermore, each material/fraction/substance within a system or a technology may be transferred from one process to another, i.e. interprocess transition, or within a single process from inputs to outputs, i.e. intra-process transition. The interprocess transition represents cases when a process output is transferred to a subsequent process input, e.g. when sugars produced during hydrolysis are used in fermentation to generate ethanol and CO_2; thus, the transition is from hydrolysis to fermentation. The intra-process transition is when specific properties are involved in the generation of process outputs, e.g. when in hydrolysis, cellulose is depolymerised into C6 sugars and this transition occurs from the hydrolysis input to the hydrolysis output. The carbon content of cellulose (here classified as a substance) contributes to the generation of C6 sugars (classified as substance). To model these two types of transitions, one needs to be able to separate and "extract" a single material/fraction/substance from the remaining materials/fractions/substances. Extracting means isolating the material/fraction/substance and considering it as a single independent element to be subsequently used in other conversion flows. Operators that allow this extraction are *material distributor* (MD), *fraction distributor* (FD) and *substance distributor* (SD). Considering the example of garden waste, an FD may be applied in the example where only grass (a fraction within garden waste) is addressed in a specific (unit) process. Thus, grass may be extracted from the other fractions composing the garden waste and routed to a different flow for modelling purposes. An example of using SD is the separation of non-biodegradable matter such as lignin within an organic feedstock. With SD, the lignin representing a feedstock's biochemical property may be extracted and routed to a combustion process for energy utilisation. In cases when more than one fraction or substance are routed to a new flow, these fractions and substances need to be grouped: a *fraction hub* (FH) is used for grouping fractions while a *substance hub* (SH) is for substances. *Material flows* (MF) are represented by an arrow and are used for the transition of materials, fractions and substances from a source element to a target element. Within a process, conditional statements can be associated with individual MFs, e.g. water content > 0, to ensure a flow continues as long as the given condition is true. A *residue flow* (RF) is applied to close mass balances, i.e. to "catch" and transfer any remaining mass (residues) after transformation operations. Also, RF is represented by an arrow and is used for transitions. While it is possible to have more than one MF from a source element (e.g. an operator), only one RF can be used to close the mass balance. If the residues are transferred to a target element within the process, no other residue exists.

In addition to the above-mentioned methods to transform, divide and group materials, fractions and substances, a range of calculations may be done on these entities by a *composite transformer* (CT). In a CT, calculations may be grouped and if necessary combined with more operators relevant for the material, fraction or substance "level" in question. These calculations are performed using mathematical equations with parameters. *Primitive* parameters represent single values, such as a constant (e.g. conversion efficiency of C6 sugars, eff_{C6} = 88%). *Data table parameter* is used when an element has more than one parameter associated. Each column of the data table represents a parameter, i.e. the values are elements in the table, and each row is a set of parameters. The data table is identified by a name. In order to build this table, columns need to be added for each parameter; this can be done with a *data column* (DC). For each parameter (column), the value type is specified (i.e. a number or string). For example, we model cellulose that has as a parameter mass in kilogrammes and conversion efficiency into sugars in percentage; since it has two parameters associated, we may have a data table parameter with three data columns, one for cellulose (substance), and a further two for the mass and the conversion efficiency. A process finishes with one or more outputs having all properties generated during the process modelling. This involves using one/more *output*(s) representing all the material properties transferred to it/them through MFs and/or RFs.

An example of a combination of more than one operator described in this section is presented in Fig. 2. This represents an illustrative example removing 10% of water (substance) from the grass (fraction) in garden waste (material). A way to accomplish this is first to define and generate the material garden waste through an MG; secondly, the generation of fractions within it, such as grass, wood, plants, branches, tree, and soil, stones and foreign objects, through FGs; thirdly, all these fractions are grouped in an FD, linked to a CT where the substances associated with each fraction are generated through SGs. Subsequently, the grass is extracted through an FD and the other fractions within the garden waste are sent to the final output through an RF. All the substances within grass are grouped in an SH. Water is extracted through an SD and its content is transformed (i.e. − 10%) in an ST. In the final output, water with the different content is sent through an MF linking ST with the final output. Additionally, the other substances (with the same content) are sent to the final output through an RF from SH.

2.3 Application of the process-oriented modelling approach to a biorefinery case

2.3.1 Description of the technology system

The case study evaluates a second-generation biorefinery using the above-mentioned operators within EASETECH. The biorefinery is composed of five main unit processes: *biomaterial generation*, *pre-treatment*, *hydrolysis*, *fermentation*

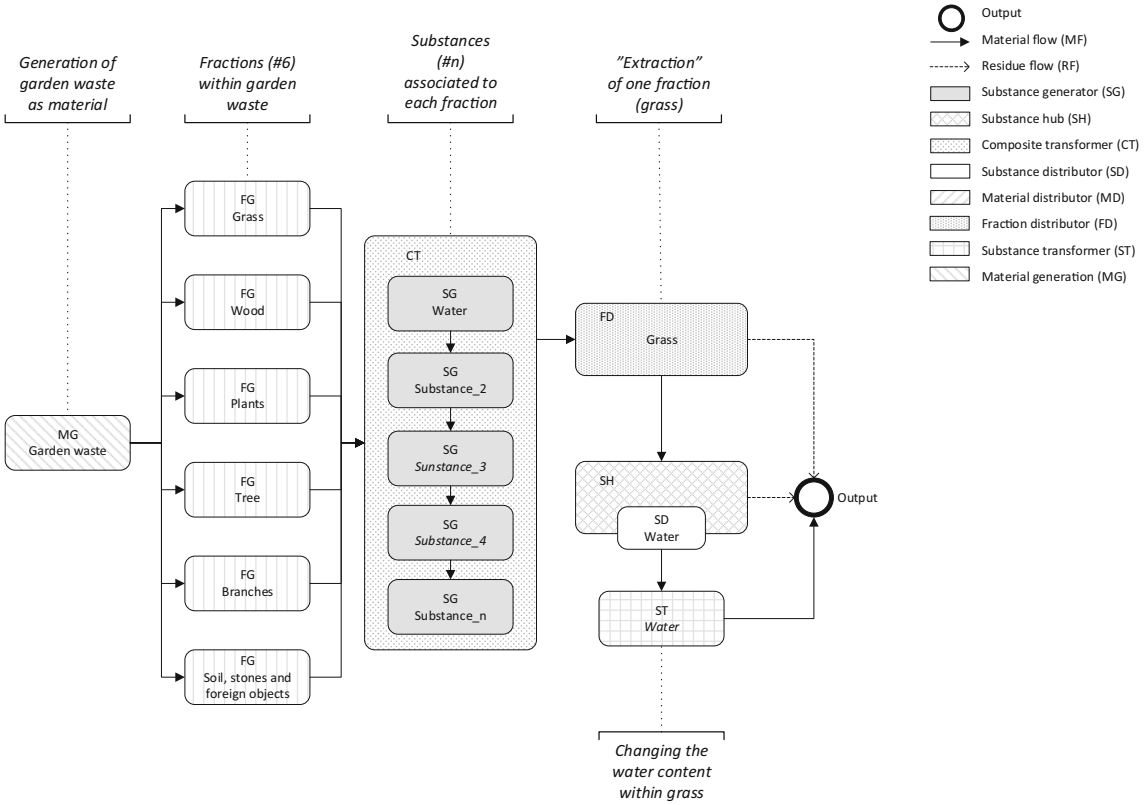


Fig. 2 Example of an application of operators for decreasing the water content (substance) of the grass (fraction) within garden waste (material)

and *distillation*, *recovery*. In biomaterial generation, the input feedstock is modelled considering all its properties (substances), such as biochemical (organic matter content), elemental (inorganic matter content), nutritional (i.e. the "feeding value" calculated based on the feedstock nutritional-energy content) and physical (e.g. water, ash, etc.), see Electronic Supplementary Material, Section S2 for details. For modelling purposes, biomaterial generation is considered as a process, although this does not represent the conversion of the feedstock but merely the relevant calculations of feedstock properties prior to the input to the pre-treatment process. Some properties (e.g. dry matter, nitrogen, oxygen, hydrogen, carbon, sulphur, energy content, methane potential, etc.) are stoichiometrically calculated based on the biochemical and physical contents of the feedstock; as such, these properties are correlated to other properties (Eq. S1 to S15, Electronic Supplementary Material – ESM). In the biomaterial generation, mathematical equations then recalculate some of the properties of the selected feedstock, with the advantage of correlating them (e.g. C with LHV, N with proteins, cellulose/hemicellulose/proteins/etc. with nutritional value and LHV). All the mathematical equations used in the biomaterial composition are explained in the ESM, Sections S2.1 to S2.6. In pre-treatment (Section S2 - ESM), energy in the form of heat is used to pre-treat the feedstock. The structure of the lignocelluloses is broken down to separate the lignin from the cellulose and hemicellulose and allow an efficient conversion into fermentable sugars. Pre-treatment may also result in some losses (e.g. when eventual mass is lost, the conversion efficiency to the pre-treatment output composition is lower than 100%) not routed further to the hydrolysis process. In hydrolysis (Section S4 - ESM), cellulose, starch, hemicellulose, pectin and sucrose are hydrolysed into C5 and C6 sugars. The non-hydrolysed biochemical properties represent the hydrolysis residues. In fermentation and distillation (Section S5 - ESM), the C5 and C6 sugars are converted to bioethanol, CO_2 and liquid molasses. The unconverted sugars are transferred to yet another output and passed on to a recovery process (Section S6 - ESM), which in addition to the fermentation residues receives the mixed solid and liquid residues from hydrolysis (hydrolysis residues); here, all residues are separated to maximise further utilisation.

Regarding the further utilisation of these output products, the liquid fraction was assumed to be converted into biogas, while the solid fraction was assumed to be incinerated with energy recovery. For both fractions, natural gas was assumed to be substituted for simplicity. In order to focus on the technology system modelling, we deliberately neglected the possible impacts from diverting the feedstock from its current use(s) and eventual land-use changes. This should be kept in mind when interpreting the results to avoid inconsistent und unfair comparisons with other studies. We briefly stress the importance of these aspects in Section 4.3.

2.3.2 Assessment scope, functional unit and system boundary

The primary goal with the LCA was to demonstrate the applicability of process-oriented modelling in EASETECH and illustrate potential learnings that can be achieved on this basis. In this perspective, the assessment focus was placed on a single biorefinery scenario without the range of scenario alternatives and sensitivity/uncertainty evaluations otherwise part of an LCA (see Negro et al. 2017; Serra et al. 2017; Wang et al. 2016). As such, the case study followed the principles of the relevant ISO standards (ISO 2006a, b), while not strictly complying with these. Two perspectives were evaluated with the case study: (i) the importance of unit process performance and choice of process parameters for the overall results, and (ii) the importance of feedstock characteristics for the performance of the individual unit processes at fixed parameter sets. For the first perspective, three types of input feedstock were considered: wheat straw, beet top and wild grass, while the second perspective was proved based on *Miscanthus*, brewer's grains and willow. The first set of biomasses was selected based on their different composition to test the biorefinery model and the expected different results. Table 2 presents key characteristics and properties. The second set of biomasses was selected according to their cellulose, hemicellulose and lignin content. These three organic molecules have high importance for the carbon pool available in the biorefinery; *Miscanthus* has the highest cellulose content, brewer's grain has the highest hemicellulose content and willow has the highest lignin one.

The functional unit represented "the valorisation within a biorefinery of one tonne (wet weight) of input-feedstock into three main output-products: bioethanol, solid, and a liquid fraction". While results were calculated for all the impact categories included in the IPCC 2013 method (IPCC 2013; 100-year time horizon was assumed), only results for global warming were discussed for the purpose of illustrating the functionality and applicability of the process-oriented modelling approach. Figure 3 illustrates a generic representation of the biorefinery process-oriented model.

To ensure simplicity, a "zero burden" approach was followed and no upstream burdens associated with the input feedstock biomass nor any indirect effects associated with the diversion from alternative uses of the biomass (counterfactual scenarios) were included. System expansion was applied to credit the system for avoided impacts associated with substituting and displacing conventional market products with the biorefinery output products. Ethanol was assumed to be used in vehicles, substituting gasoline; molasses, the liquid fraction from the biorefinery, was used in a biogas plant substituting the production and combustion of natural gas; solid biofuel, the solid fraction from the biorefinery, was used in an incineration plant that substituted the production and combustion of natural gas. The emission factor assumed for gasoline was 0.097 kg CO_2-eq MJ^{-1} and the emission factor for natural gas was 0.067 kg CO_2-eq MJ^{-1} from EASETECH database (Clavreul et al. 2014). The residual digestate after biogas production was assumed to displace conventional NPK fertilisers, according to the content of N, P and K. The substitution efficiency was assumed to be 40% for N according to current Danish legislation (Danish Ministry of Food, Agriculture and Fisheries 2018) and 100% for P and K. Air and water emissions arising from digestate and mineral fertilisers (avoided) spreading on-land were based on the work of Yoshida et al. (2016); particularly, the emission factors used to describe N_2O emissions from digestate and substituted mineral fertilisers were 2.78% and (2.32 × 0.40)%, respectively. The system boundaries included refinery operations, harvest of biomass, transportation (digestate and solid fraction) as well as final utilisation and management of all biorefinery outputs.

3 Results

3.1 Importance of unit process operational efficiencies

Figure 4 presents the results of global warming (GW) in kg CO_2-eq t^{-1}_{ww} for a biorefinery using wheat straw as feedstock. The biorefinery outputs are given in MJ t^{-1}_{ww} as a function of the efficiencies of the hydrolysis (Fig. 4a, b) and the fermentation (Fig. 4c, d) unit processes. Through the selection of parameters (e.g. yield, efficiencies, etc.), the model responds to variations in the performance of the individual unit processes and allows users to adapt a specific biorefinery configuration. Here, the environmental impacts of the entire technology systems were calculated by one-at-a-time changes in parameter values, from a low conversion efficiency (25%) to a complete conversion (100%). For example, for fermentation of C5 sugars, only the fermentation efficiency was changed with all other parameters unchanged; the parameter values (0%, 25%, 50%, 75% and 100%) were selected for illustrative purposes.

As the results demonstrate, process parameters play an important role: the user can modify the mass and energy balances (here represented by the process outputs) with a direct effect on the associated environmental impacts (here represented by GW). In this example, increased efficiency of cellulose hydrolysis leads to better GW performance (Fig. 4b); this is reflected by the increased production of liquid fuel (ethanol) and the decreased production of solid fraction (sometimes called solid biofuel) resulting in the decreased substitution of natural gas combustion. In addition, increasing the fermentation efficiency of C6 sugars leads to better GW performance (Fig. 4d); also in this case, fuel production was increased, but now the liquid fraction (sometimes called molasses)

Table 2 Characteristics and properties of wheat straw (feedstock 1), wild grass (feedstock 2) and beet top (feedstock 3), used as feedstock for the biorefinery case study

Bio_material_generation—parameters						
Subgroup 1	Biochemical properties	Description	Feedstock 1	Feedstock 2	Feedstock 3	Unit
1	Acetic acid*	CH3COOH	0.0	0.0	0.0	$\%_{DM}$
2	Cellulose	Cellulose parameter	34.7	29.1	11.2	$\%_{DM}$
3	Hemicellulose	Hemicellulose parameter	22.4	24.2	16.2	$\%_{DM}$
4	Lignin	Lignin parameter	17.7	3.0	8.2	$\%_{DM}$
5	Lipids	Lipids parameter	2.3	0.5	2.4	$\%_{DM}$
6	Pectin	Pectin parameter	0.0	0.0	8.2	$\%_{DM}$
7	Proteins	Proteins parameter	3.5	5.2	16.9	$\%_{DM}$
8	Starch	Starch parameter	0.0	0.0	3.6	$\%_{DM}$
9	Sucrose	Sucrose parameter	0.0	0.0	11.9	$\%_{DM}$
10	Other VS	Unspecified VS parameter	14.07	33.86	4.9	$\%_{DM}$
Subgroup 2	Elemental properties					
11	Al	Aluminium	0.0168	0.0000	0.0000	$\%_{DM}$
12	Ca	Calcium	0.2435	0.5500	1.3000	$\%_{DM}$
13	Cl	Chlorine	0.3876	0.8000	1.6000	$\%_{DM}$
14	Cr	Chromium	0.0003	0.0000	0.0000	$\%_{DM}$
15	Cu	Copper	0.0004	0.0007	0.0013	$\%_{DM}$
16	F	Fluorine	0.0011	0.0000	0.0000	$\%_{DM}$
17	Fe	Iron	0.0134	0.0220	0.0000	$\%_{DM}$
18	Hg	Mercury	0.0000	0.0000	0.0000	$\%_{DM}$
19	K	Potassium	0.9870	0.3300	4.8000	$\%_{DM}$
20	Mg	Magnesium	0.0439	0.1800	0.4100	$\%_{DM}$
21	Mn	Manganese	0.0020	0.0070	0.0090	$\%_{DM}$
22	Mo	Molybdenum	0.0001	0.0000	0.0000	$\%_{DM}$
23	Na	Sodium	0.0100	0.1500	0.9700	$\%_{DM}$
24	Ni	Nickel	0.0001	0.0000	0.0000	$\%_{DM}$
25	P	Phosphorus	0.0490	0.4000	0.1750	$\%_{DM}$
26	Pb	Lead	0.0003	0.0000	0.0000	$\%_{DM}$
27	S	Sulphur	0.0000	0.2100	0.2000	$\%_{DM}$
28	Si	Silicon	0.9300	0.0000	0.0000	$\%_{DM}$
29	Ti	Titanium	0.0005	0.0000	0.0000	$\%_{DM}$
30	V	Vanadium	0.0001	0.0000	0.0000	$\%_{DM}$
31	Zn	Zinc	0.0034	0.0000	0.0045	$\%_{DM}$
Subgroup 3	Feedstock					
32	Fraction name	Fraction	Wheat straw	Wild grass	Beet top	String
Subgroup 4	Feedstock amount					
33	Quantity	Input amount	1000	1000	1000	kg_{ww}
Subgroup 5	Nutritional properties					
34	Crude_Fibers_input	Crude fibres parameter	45.3	78	82	$\%_{DM}$
35	Digestibility_input	Substrate digestibility	44	24.9	12	$\%_{DM}$
Subgroup 6	Physical properties					
37	Ash	Ash parameter	5.4	4.1	16.5	$\%_{DM}$
38	VS	Volatile solid parameter	94.7	95.9	83.5	$\%_{DM}$
39	Water	Water parameter	12.2	78.8	76.7	$\%_{ww}$

*Acetic acid may be present in some biomasses as degradation product

decreased, thereby resulting in lower biogas production and lower substitution of natural gas combustion. In Fig. 4, for a cellulose hydrolysis efficiency of 0% the associated GW performance was − 269 kg CO_2-eq t_{ww}^{-1}; for an efficiency of 25%, the associated GW performance was − 339 kg CO_2-eq t_{ww}^{-1}; for an efficiency of 50%, the associated GW performance equalled − 409 kg CO_2-eq t_{ww}^{-1}; for 75%, it was − 479 kg CO_2-eq t_{ww}^{-1} and for 100%, it was − 549 kg CO_2-eq t_{ww}^{-1}. Such direct proportionality between the energy/mass balances and the GW impacts may not necessarily have a direct effect on full scenario results as also framework conditions may be important, e.g. type of substituted energy, system boundaries and process configurations. Furthermore, the linear results are due to the equations applied in the case example, the model could just as well have been used for cases with exponential changes, or more scattered results if conditions for flow properties were applied in the model. These aspects can, however, be captured by the process-oriented LCA model either by adjusting parameters, changing the mathematical relationships involving the functions introduced earlier, or choice of background process data and interactions with the background system. For further details of the biorefinery modelling results involving variations in parameter efficiencies and associated GW impacts, please see ESM, Section S7, Table S7.1, S7.2 and S7.3.

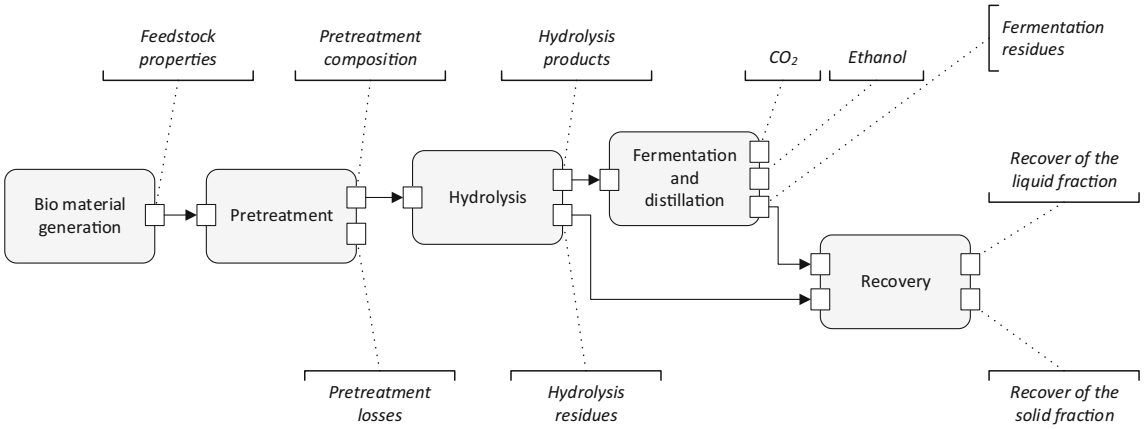


Fig. 3 Generic representation of the biorefinery process-oriented model in EASETECH with the intermediate and final outputs

Overall, similar results and trends were obtained for the two other feedstock types, beet top and wild grass, i.e. higher efficiencies provided larger environmental savings (see ESM, Section S8, Fig. S8.1 and S8.2 for the results). Differences in biochemical and physical properties between wheat straw, beet top and wild grass were reflected in the results by different "levels". With a cellulose hydrolysis efficiency of 0%, the associated GW performance for beet top was − 55 kg CO_2-eq t_{ww}^{-1} and for wild grass − 37 kg CO_2-eq t_{ww}^{-1}; for an efficiency of 25%, the associated GW performance for beet top was − 61 kg CO_2-eq t_{ww}^{-1} and for wild grass − 51 kg CO_2-eq t_{ww}^{-1}; for an efficiency of 50%, the associated GW potential was respectively − 67 and − 65 kg CO_2-eq t_{ww}^{-1}, while for 75%, it was − 73 and − 79 kg CO_2-eq t_{ww}^{-1}, and − 79 and − 93 kg CO_2-eq t_{ww}^{-1} in the case of 100%. While a similar trend in results can be expected, the model demonstrates the relative importance of the hydrolysis and fermentation steps for the three different feedstocks and thereby transparently explains the difference in results between the cases. This demonstrates that the model can be applied to technology cases with different process configurations (illustrated here by different efficiencies of unit processes and subsequent changes in material and substance flows) and can accommodate different input feedstock properties in a flexible manner.

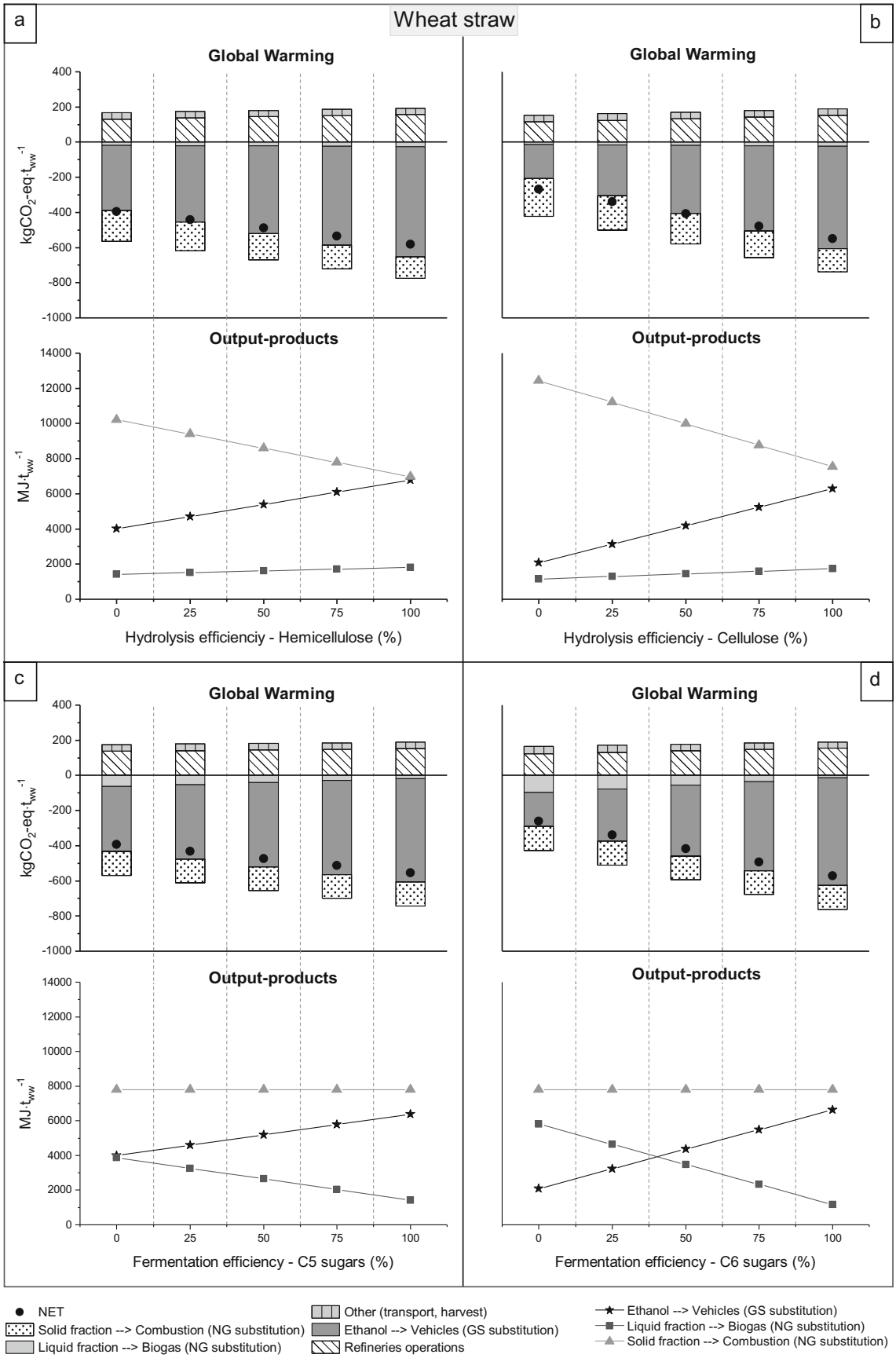
3.2 Importance of input feedstock characteristics

The feedstock characteristics play an important role for the biorefinery performance, both with respect to GW (kg CO_2-eq t_{ww}^{-1}) and output products (e.g. MJ·t_{ww}^{-1}) as illustrated in Fig. 5.

Among the three biomasses addressed here, *Miscanthus* has the highest cellulose content (CE = 47.6%$_{DM}$), brewer's grain has the highest hemicellulose content (HC = 29.5%$_{DM}$) and willow the highest lignin content (LG = 31.6%$_{DM}$). In Electronic Supplementary Material, Section S9, Tab S9.1 presents key characteristics and properties of these three biomasses. The cellulose, hemicellulose and lignin contents for these three biomasses are shown in Fig. 5. The conversion efficiencies considered were 95% and 75% for cellulose and hemicellulose respectively during hydrolysis, and for both C5 and C6 sugars, it was 88% during fermentation and distillation.

Considering the three main products of the biorefinery (ethanol, solid and liquid fraction), cellulose and hemicellulose affect mostly the production of ethanol and the liquid fraction as these molecules can be hydrolysed into chains of monosaccharides (e.g. glucose) used in the fermentation to produce ethanol and CO_2. Lignin represents the carbon pool that in a biorefinery leads to the formation of the solid fraction output unless pre-treatment is applied, together with non-hydrolysed material. With this in mind, based on the composition of the three feedstocks, *Miscanthus* generated more ethanol (7400 MJ·t_{ww}^{-1}), followed by willow (3500 MJ·t_{ww}^{-1}) and brewer's grain (1400 MJ·t_{ww}^{-1}). Considering the solid fraction, although willow has the highest lignin content, *Miscanthus* provided the largest solid fraction (7200 MJ·t_{ww}^{-1}), due to the larger amounts of unconverted sugars (dry basis). The liquid fraction is influenced mainly by the fermentation and distillation process. For this reason, *Miscanthus* provided the highest liquid output (1600 MJ·t_{ww}^{-1}) followed by brewer's grain (1000 MJ·t_{ww}^{-1}) and willow (960 MJ·t_{ww}^{-1}). In this illustrative example, the conversion of all three biomasses provided net GW savings as no upstream activities (e.g. production) and indirect effects (e.g. land-use changes) were included. The largest savings were obtained from *Miscanthus* due to its higher dry matter content. These results were in accordance with Parajuli at al. (2017), who showed that the high dry matter and energy yield of the input feedstock material can contribute to a better environmental performance. In addition, the relevance of conversion efficiencies of feedstock properties (e.g. carbohydrates) in the biorefinery processes was highlighted in Parajuli et al. (2017), in agreement with this study.

While the influence of feedstock choice on the LCA results has been evaluated previously in the literature (e.g. Bernstad


Wheat straw
a
b
c
d
Global Warming
Output-products
kgCO2-eq·tww-1
MJ·tww-1
Hydrolysis efficiency - Hemicellulose (%)
Hydrolysis efficiency - Cellulose (%)
Fermentation efficiency - C5 sugars (%)
Fermentation efficiency - C6 sugars (%)
NET
Solid fraction --> Combustion (NG substitution)
Liquid fraction --> Biogas (NG substitution)
Other (transport, harvest)
Ethanol --> Vehicles (GS substitution)
Refineries operations
Ethanol --> Vehicles (GS substitution)
Liquid fraction --> Biogas (NG substitution)
Solid fraction --> Combustion (NG substitution)

◀ **Fig. 4** An overview of the process-oriented LCA model response, in terms of global warming, GW, (kg CO_2-eq kg_{ww}^{-1}) and mass/energy balance (MJ t_{ww}^{-1}), to (one-at-the-time) unit process performance variations (i.e. 0%, 25%; 50%; 75%; 100%). **a** Hemicellulose conversion efficiency in hydrolysis. **b** Cellulose conversion efficiency in hydrolysis. **c** C5 sugars conversion efficiency in fermentation. **d** C6 sugars conversion efficiency in fermentation. The feedstock considered is wheat straw. NG, natural gas; GS, gasoline

Saraiva 2017; Tonini et al. 2016a, b), the above process-oriented assessment approach demonstrates the added insight of the importance of individual unit processes (and potentially also parameter choices as illustrated in the previous section). Particularly, the inter- and intra-process transition, the material transformation due to the process specificities and the feedstock specificities (e.g. the importance of feedstock properties and their availability to be degraded or converted into different products), and their consequences in terms of environmental impacts.

4 Discussion

4.1 Novel insights from process-oriented modelling

The process-oriented approach focuses on the evaluation of process relationships through subdivisions of technologies into unit processes and appropriate linking of process material inputs with transformation and process outputs. In previous literature (e.g. Tonini et al. 2016a, b), these aspects have been demonstrated as critical for the LCA results and interpretation,

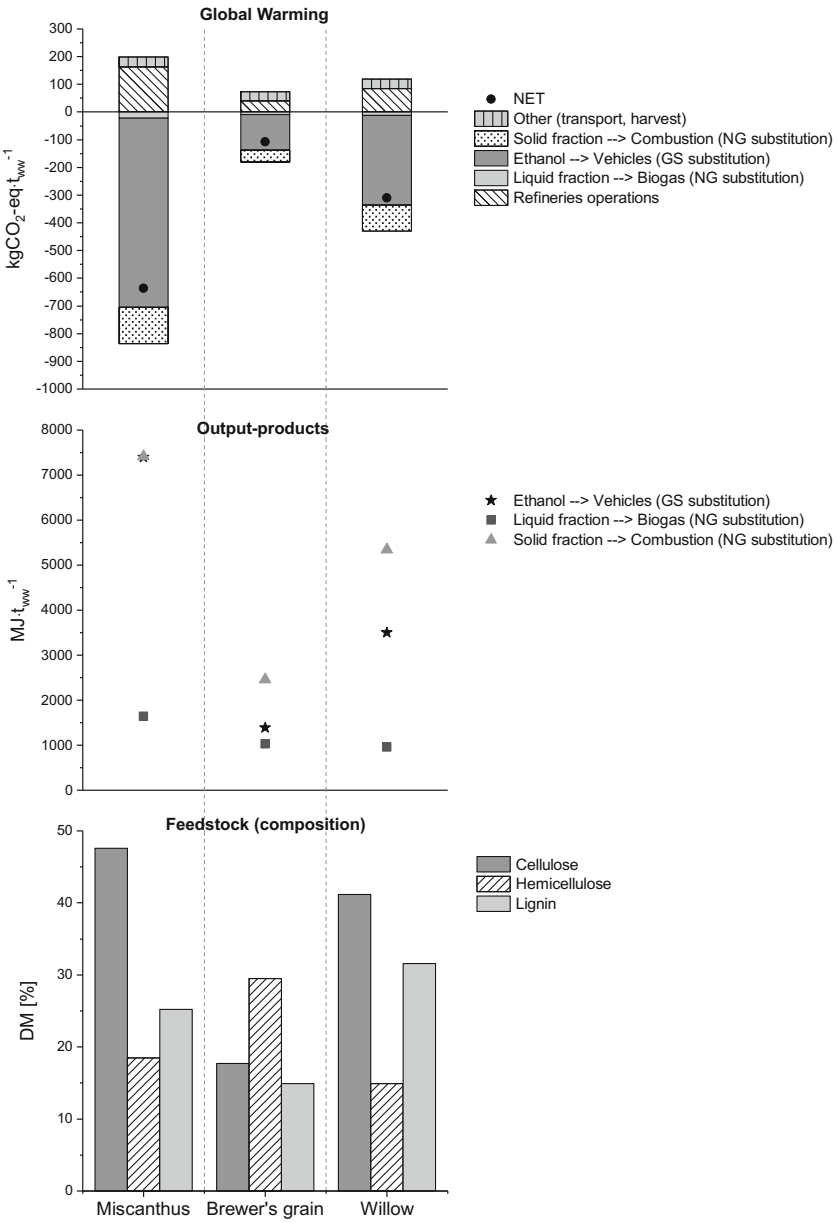


Fig. 5 Process-oriented LCA model response, in terms of global warming, GW, (kg CO_2-eq kg_{ww}^{-1}) and mass/energy balance (MJ t_{ww}^{-1}), to three different feedstocks (i.e. *Miscanthus*, brewer's grain and willow) having different shares of cellulose, hemicellulose and lignin. NG, natural gas; GS, gasoline. For these three biomasses, the values of the parameters used are in hydrolysis, a cellulose and a hemicellulose conversion efficiency of 95% and 75% respectively, and in fermentation and distillation, the conversion efficiency of 88% for both C5 and C6 sugars

in particular in relation to integrated technologies such as biorefineries where the feedstock characteristics and the biorefinery outputs are interdependent and further affect the downstream substitutions (e.g. energy, feed, materials). One of the most notable advantages of the process-oriented modelling approach is the possibility of implementing new (unit) processes by using operators in a single modelling tool such as EASETECH, rather than requiring a combination of several tools as illustrated by previous literature (e.g. in Tonini et al. 2016a, b, and Vadenbo et al. 2018, where a combination of Matlab, Gams, and SimaPro was applied).

Mathematical equations describing the input-output relationships are integrated within the model itself and default parameter values can be further adjusted by the users. The subdivision into unit processes is important for identification, quantification and evaluation of intermediate process outputs within the system. Further, the process parameters and associated mathematical relationships themselves may be selected to appropriately represent operational parameters that can be recognised by users and more easily adjusted to accommodate specific case studies and industry data. Quantification of the intermediate products linking individual unit processes allows evaluation of the environmental performance of these unit processes, which may further allow identification of technology hotspots at a much more detailed level than traditional "black-box" modelling approaches, both in terms of production and emissions. This is fully in line with existing recommendations, e.g. by ILCD guidelines (EC-JRC 2010) and strongly highlighted in Jacquemin et al. (2012).

The process-oriented modelling approach enables more control of the material, energy and substance flows within the analysed technologies. This is particularly important in relation to integrated technologies such as biorefineries or many waste technologies for which intermediate products affect the subsequent processing; an aspect that black-box approaches cannot capture (Maes et al. 2015). Modelling a biorefinery technology within EASETECH following the process-oriented approach offers an "active" material flow system represented by the established input-output relationships and parameters. This material flow system is linked to environmental emissions and output product substitutions associated with the LCA scenarios; thereby a direct link between input feedstock composition, process operation and environmental performance is established. For example, higher hydrolysis and fermentation efficiencies incur larger ethanol production with lower solid and liquid residue quantities, thereby increasing gasoline substitution and lowering natural gas substitution. Although purposely kept simple in this illustrative case study, interactions between the foreground and background systems can be easily modelled with appropriate selection of parameters. The conversion of biochemical properties of the feedstock into the biorefinery products depends on the type of feedstock and its degradability under the specific operating conditions of the technology. All such aspects can be addressed and evaluated by the proposed process-oriented modelling approach.

4.2 Implications for LCA

Subdividing the technology into relevant unit processes and establishing appropriate input-output relationships including operating parameter variations allow a direct response of the LCA model with respect to potential environmental impacts. While subdivision into smaller units has been suggested in previous literature, this has mainly been discussed from the perspective of Maes et al. (2015) rather than with the intention of Götze et al. (2014) and Papadokonstantakis et al. (2016) as suggested here for the process-oriented approach. Only few studies have discussed the potential of establishing operational relationships and more "technology relevant" parameters (e.g. Portha et al. 2010; Kikuchi et al. 2014). As previously indicated, the ability to "track" intermediates and conversion of individual input material fractions is essential for LCA modelling of multi-output technologies (e.g. Astrup et al. 2018), although relatively few LCA studies take this aspect seriously. With a black-box approach, where unit processes may be combined even if they are physically separated, relevant disaggregation of the environmental impacts associated with individual outputs may not be possible (e.g. Jacquemin et al. 2012). As the process-oriented modelling approach attempts to disaggregate technology and process elements into individual units reflecting the actual process flow and conversion steps, the process-oriented approach can facilitate easier compliance with the recommendations provided by current ISO standards and ILCD guidelines with respect to multi-functionality. In the case of LCA modelling of material and resource technology systems, we suggest that the process-oriented approach is a needed development from black-box approaches and that these cannot be considered state-of-the-art for such systems. We envision that further development of process-oriented inventories may offer a route to avoid the current challenges of multi-functionality associated with complex multi-output technologies.

LCA studies are also sometimes used to assess the environmental performance of technologies prior to commercialisation and full-scale implementation, e.g. prospective assessment of emerging technologies (Arvidsson et al. 2017). From a black-box modelling perspective, such assessments pose specific challenges with respect to data uncertainties, process configurations, potential performance improvements, etc. as these aspects are typically aggregated within the technology inventory thereby limiting transparency. Process-oriented modelling, on the other hand, allows disaggregation and establishment of appropriate data relationships. Thereby, the uncertainties and importance of individual process parameters may be evaluated directly and linked to the

environmental performance of the technology in question. This makes process-oriented modelling particularly relevant for LCA assisted technology developments and upscaling activities, as the assessment results allow identification of process hotspots that may otherwise remain un-evaluated. We envision that these aspects are particularly important in relation to integrated and multi-output technologies as part of circular (bio) economy initiatives.

4.3 Further research and perspectives

As developed and implemented in this study, the process-oriented modelling approach represents a first attempt to demonstrate applicability and potential. Future research is intended to focus on improving the existing model and extending the process-oriented modelling approach to a wide range of material-centric technologies, e.g. anaerobic digestion, thermal pyrolysis and gasification, thermal combustion, and biomaterial production facilities. This requires identification and appropriate implementation of relevant process relationships between input resources and materials (e.g. chemicals, energy, etc.) and process outputs and emissions. While EASETECH offers a unique basis for this as the modelling is already based on material flows, implementation of new process-oriented technology models nevertheless requires considerable effort (see Electronic Supplementary Material as an example for a biorefinery). However, once a process-oriented model is established, subsequent adjustments can be achieved simply by changing the appropriate parameters (assuming the fundamental process configuration remains appropriate). As mentioned earlier (Section 2.3.1), to focus on the technology modelling, we deliberately excluded the upstream impacts associated with diverting the feedstock from its current use(s) or with land-use changes. Such impacts have been earlier estimated in the order of 19–88 kg CO_2-eq t_{ww}^{-1} for wild grass and wheat straw, 191–360 kg CO_2-eq t_{ww}^{-1} for perennial energy crops as willow and *Miscanthus,* and 265–287 kg CO_2-eq t_{ww}^{-1} for agro-industrial residues as beet top and brewer's grain (Tonini et al. 2016a, b). These figures should be added to the results quantified in this study to obtain a full picture of the Climate Change impact of the studied scenarios.

5 Conclusions

The study developed a process-oriented environmental life-cycle assessment modelling framework, implemented this in EASETECH and applied this on a biorefinery case study for illustrative purposes. The process-oriented modelling framework provides an improved representation of complex technologies allowing definition and evaluation of process relationships between inputs and outputs. This is particularly important for integrated technologies comprising individual unit processes, e.g. biomass conversion and management of residual resources. Traditional black-box modelling approaches, represented by most existing LCA models, do not offer similar possibilities for detailed evaluation of processes and technologies nor allow the same level of transparency with respect to inventory definition. The process-oriented modelling framework provided by this study allows consistent balancing of material, fraction and substance flows within the technology system and, through mathematical expressions, at the same time establishment of the process relationships that affect these flows through transition and transformation within each single unit process. Based on the biorefinery case study, the advantages of the modelling approach were demonstrated: input feedstocks and key process operational parameters can be adjusted easily in order to evaluate process performance and the importance of feedstock properties. This facilitates quantification of individual/intermediate (bio) product flows within unit processes; this has not been possible previously. The potential implications of process-oriented modelling are considerable, e.g. in relation to novel insights associated with uncertainty evaluation, technology upscaling and process optimisation.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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II

A modelling framework for process-oriented life cycle assessment (EASETECH+)

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Energy recovery from plastic and biomass waste by means of fluidized bed gasification: A life cycle inventory model

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ABSTRACT

The study provides for the first time a life cycle inventory model for the fluidized bed gasification of wastes, based on a large amount of high-quality data. All of them have been obtained from a pilot scale fluidized bed gasifier, fed with ten types of waste and biomass, under a wide range of operating conditions. The model refers to commercial scale gasifiers having a "thermal configuration", where the generated syngas is immediately burned downstream of the reactor. Key relationships between process- and waste-specific parameters have been defined. The model quantifies the main inputs and outputs of the gasification process (emissions, energy recovery, ash disposal, resource consumptions), providing high-quality data that could contribute to improve life cycle assessment modelling of waste gasification. Finally, some case studies have been implemented in the EASETECH software to illustrate the model applicability, evaluate the role of main parameters, and compare the environmental performances of gasification power units with that of the European electricity mix. The performances appear highly affected by metal contents in the waste-derived fuels, while the model results to a limited extent are sensitive to the equivalence ratio and the net electrical efficiency of the energy conversion.

1. Introduction

Gasification is a thermochemical conversion process that allows an efficient resource recovery from a wide range of biomass and waste-derived fuels [1]. Currently, it is utilised in more than 100 commercial-scale waste-to-energy plants, fed with the organic fraction of municipal solid waste (MSW) after source-separation of recyclables as well as with residual from specific recycling chains, as those of plastic wastes [2,3]. Gasification converts solid waste materials into a fuel gas, called "syngas", through a series of heterogeneous and homogeneous reactions, taking place in a reducing atmosphere where the gasifying agent can be air, oxygen-enriched air, steam or carbon dioxide [2,4]. The obtained syngas contains large amounts of incompletely oxidised products, mainly CO and H_2, together with smaller amounts of CH_4. The syngas represents a valuable product that can be used in a wide range of applications, aiming at generation of energy, fuels [5] and drop-in chemicals [6,7]. However, the syngas generally contains a range of organic (tar) and inorganic (H_2S, HCl, NH_3, HCN and alkali metals) impurities that may complicate further its utilisation and cause operating challenges, such as fouling and slagging of heating surfaces, catalyst poisoning, extra costs for maintenance and unplanned plant shut-downs [2,5].

Fluidization is a promising gasification technology as it permits a high quality gas–solid contact, thereby enabling an efficient mass and heat transfer. The process flexibility of fluidized bed gasifiers (FBGs) is able to accommodate variations in fuel quality, to allow the utilisation of different fluidizing agents, reactor temperatures and gas residence times, to add reagents along the reactor height, and to operate with or without a catalyst [2,8].

Only few attempts can be found in scientific literature of fluidized bed gasification modelling in a life cycle assessment (LCA) perspective, based on high quality (operating or experimental) data. In most cases, only the greenhouse gas emissions [9] and the Global Warming Potential (GWP) [10] have been included. The published LCA studies on gasification can be grouped by different

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List of acronyms

APC	Air Pollution Control
BTX	Benzene, Toluene, Xylene
CC	Conversion Coefficient
CCE	Carbon Conversion Efficiency
CGE	Cold Gas Efficiency
CTUh	Comparative Toxic Unit for humans
ER	Equivalence Ratio
FBG	Fluidized Bed Gasifier
GHG	Greenhouse Gas
GWP	Global Warming Potential
HRSG	Heat Recovery and Steam Generator
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LHV	Low Heating Value
MSW	Municipal Solid Waste
ORC	Organic Rankine Cycle
SNG	Substitute of Natural Gas
TC	Transfer Coefficient
TS	Total Solids
VF	Variation Factor
VS	Volatile Solids
ww	Wet weight

criteria, such as the analysed feedstock, specific reactor technology, final products and selected comparative scenarios. Gasification-based waste-to-energy units for MSW have been compared with other thermal treatments [11] or alternative management strategies [12,13]. The environmental performance of biomass gasification has been assessed with reference to different systems for energy production, such as integrated gasification combined cycles [14,15] or combined heat and power plants [16]. In particular, fluidized bed gasification for energy and chemicals production has been assessed for several biomass fuels [17,18] or waste fuels [19], different plant scales [20], or compared with alternative conversion technologies, such as vertical shaft furnace [21] or fixed bed [22]. Other publications have quantified the environmental performance of FBGs aimed at the production and utilisation of gaseous fuels (such as hydrogen [23,24] and SNG [25–27]) or liquid fuels [28]. A few examples of LCA studies quantifying the environmental effects of FBGs integrated in a material recovery facility [29], a bio-refinery [30], and potential future energy systems [31] exist.

None of these studies includes a detailed life cycle inventory (LCI) model for the gasification process. As such, the links between the fuel input, conversion and process outputs may be poorly described. In order to provide high-quality input data for LCA modelling of waste gasification, systematic and transparent definition of the inventory data is needed. While several descriptive or predictive models for fluidized bed gasification of waste exist in the scientific literature [5,8], none of these models is able to appropriately represent the waste gasification behaviour, mainly because it is difficult to quantify the crucial catalytic effect of olivine bed particles. The aim of this study is to provide an inventory model for fluidized bed gasification of waste, in order to improve the quality of LCA studies in this field.

To this end, a large amount of experimental data, collected from a pilot scale bubbling fluidized bed gasifier, with a reactor size sufficiently large to avoid scale-up effects, has been used [32]. These data derive from hundreds of hours of operation of the FBG over 10 years, by feeding 10 different waste and biomass fuels [32–38]. The inventory model has been implemented in the EASETECH LCA model and applied to selected case-studies scenarios to illustrate applicability. The specific objectives were: i) define key relationships between FBG input and outputs with respect to relevant process- and waste-specific parameters; ii) define a set of "Substance Transfer Coefficients (TCs)" and a set of "Substance-to-Compound Conversion Coefficients (CCs)" for all of the analysed fuels; iii) implement the defined waste- and process-specific parameters as well as transfer and conversion coefficients into an inventory model; iv) apply this inventory model to selected scenarios in EASETECH for quantification of the environmental impacts; and v) carry out a sensitivity analysis for identification of the most critical model parameters.

2. Methodology

2.1. The fluidized bed gasifier of reference

The pilot scale bubbling fluidized bed gasifier (FBG) utilised for the experimental activity that generated all the inventory data has a maximum thermal output of about 400 kW, which means an input capacity between 30 and 100 kg/h, depending on the type of feedstock. As mentioned above, the size of the reactor, which has an internal diameter of 0.381 m, allows avoiding any significant scale-up effect, and modelling the performances of larger commercial facilities, as already made in previous studies [38,39]. During each FBG test, the gasifying agent is injected from the reactor bottom into a bed of olivine particles [40] while the waste is fed by means of an over-bed feeding system. The generated syngas is sent to a cleaning section, composed of a cyclone and a wet scrubber. In all the runs, a double system is utilised for on-line measurements of syngas composition: a set of IR analysers (Horiba VA-3115 for CO, CO_2 and O_2, Horiba VA-3001 for CH_4 and Teledyne Anal. Instr.-2000 for H_2), and an Agilent 3000 micro-gas-chromatograph. Each test is carried out under autothermal conditions (i.e. without any thermal assistance by external heaters), and consequently the heat necessary for endothermic gasification reactions is provided by the partial oxidation of the fuel in the gasifier itself and at fixed values of superficial fluidizing velocity and equivalence ratio (ER). The first is defined as the ratio between the volumetric flow rate of the fluidizing agent and the cross-section area of the bed while ER is defined as the ratio between the oxygen flow rate injected into the reactor and that required for the stoichiometric combustion of the solid fuel fed to the reactor. The resulting reactor temperature is a state variable, i.e. the answer of the FBG system to the selected values of design variables, such as ER, waste heating value, and preheating temperature of the fluidizing gas [2].

The implemented thermal FBG model refers to gasification-based waste-to-energy systems of commercial scale. The model assumes air as the fluidizing agent, and olivine as the bed material, due to its good performances as tar cracking catalyst during gasification of polyolefin plastic wastes [32] and biomass [34]. Olivine is a neo-silicate of Fe and Mg with a particle density of 2900 kg/m^3, which is assumed to have a particle size range of 200–400 μm. The model assesses the different behaviours of the selected fuels when the fluidizing velocities and ER range from 0.67 to 0.74 m/s and from 0.20 to 0.31, respectively. A "thermal gasifier" configuration has been chosen: the syngas is dedusted in a cyclone and then

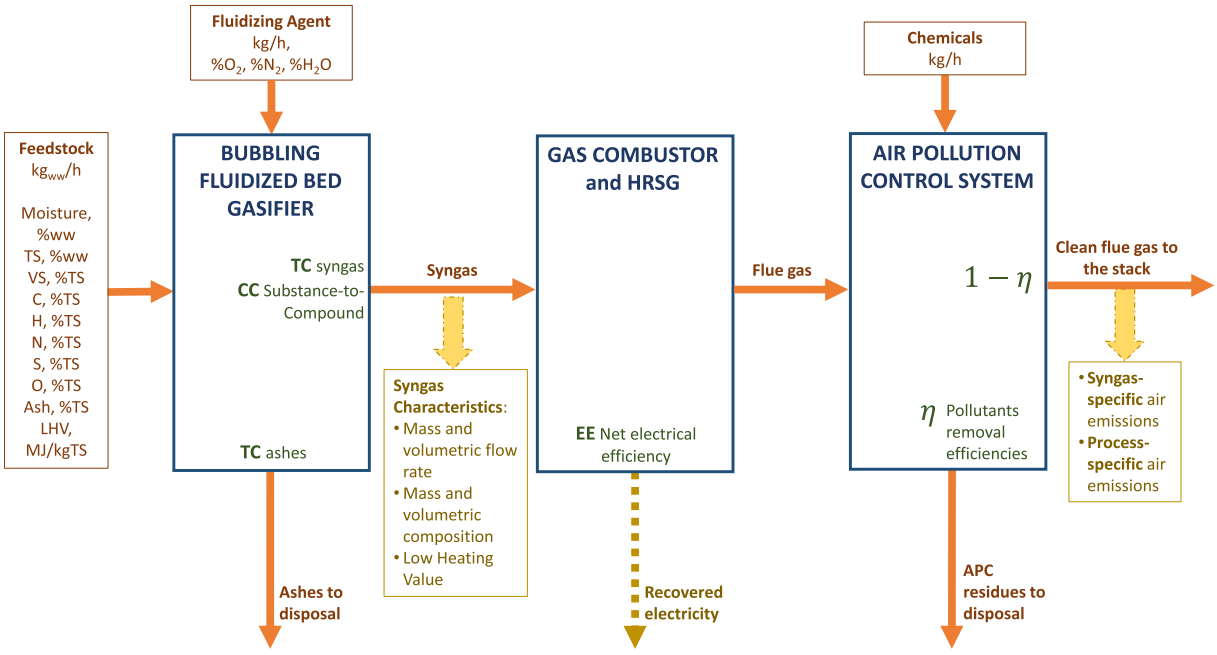

Fig. 1. General overview of the fluidized bed gasifier model with the indication of three main subunits of the gasification-based waste-to-energy unit and the utilised modelling parameters (TC, CC, EE, η). CC = Conversion Coefficient, HRSG = Heat Recovery and Steam Generator, LHV = Low Heating Value, TC = Transfer Coefficient, TS = Total Solids, ww = wet weight.

burned in a combustor for the production of electricity, while the obtained flue gas is cleaned in an air pollution control (APC) system (Fig. 1).

This configuration implies lower electricity conversion efficiencies, but larger technical reliability. It is then suitable for waste or biomass fuels that generate a syngas with a relatively large tar content, since it is able to obtain a potentially complete exploitation of the tar heating content [2]. A mild combustor [41] has been chosen for the syngas utilisation section, due to its limited formation of NO_x, dioxins and furans (PCDD/F) and products of incomplete combustion (i.e., soot, polycyclic aromatic hydrocarbons) [42]. An Organic Rankine Cycle (ORC) has been selected for the energy generation section, assuming a conservative value of the net electrical efficiency of 17.7%, evaluated for a 400 kWe plant [38]. The APC section utilises pulverised activated carbon and hydrated lime. The first is used to adsorb Hg, Cd, low-boiling heavy metals and dioxins, and the second to neutralise HCl, H_2S and SO_x. Both are injected upstream of a fabric filter, which supports the acid gas removal and controls the solid particulate emissions. More details about the gasification-based unit utilised as reference can be found in Ref. [38].

2.2. Selected waste-derived fuels

The experimental tests were carried out with the pilot scale bubbling FBG fed with seven kinds of waste-derived fuels, together with three natural biomass fuels. The waste-derived fuels were obtained from treatments of separately collected materials [32,33]. The general description and the elemental composition of each feedstock tested in the FBG plant are shown in Tables 1 and 2, respectively. Table 3 reports the number of experimental tests carried for each type of feedstock, together with the ranges of tested ER and the main source of data. Some experimental tests have been carried out specifically for this study with two natural biomass (WOOD1 and WOOD2) used as reference fuels, and by co-feeding Plasmix and RIL3, in order to investigate the effects of co-gasification on technical and environmental performances.

2.3. General aspects of the inventory model

Most of the commercial solid waste gasifiers in operation in the world are located in Asia (mainly in Japan and Korea) and utilise an autothermal gasification process [2,3], which implies a sequence of endothermic and exothermic steps: initial drying, devolatilisation, partial oxidation of a fraction of volatiles and char, and gasification reactions. The proposed FBG model, based on a series of material flow analyses, defines the general relationships linking inputs and outputs of the autothermal process. The model identifies the proper waste- and process-specific parameters, reported in Fig. 1 with reference to the gasification subunits of interest. These relationships quantify the main syngas characteristics, such as volumetric and mass flow rate, composition, low heating value (LHV), and the main environmental burdens, such as syngas-specific and process-specific air emissions, together with consumptions of chemicals and amount of residues sent to disposal. The FBG model starts from the feedstock feeding and the indication of its properties, such as composition and energy content, together with the addition of the fluidizing air stream (as quantified in Appendix A).

2.3.1. Waste-specific parameters

Transfer Coefficient (TC) and Substance-to-Compound Conversion Coefficient (CC) are waste-specific modelling parameters utilised for the analysis of the first process subunit, which includes the gasifier and the cyclone. TCs are defined as the ratio between the mass flow rate of each element in an output stream to the mass flow rate of the same element entering into the reactor, so that they can range between 0 and 1. The results of the experimental tests provide a set of TCs of the feedstock elements (all those of the ultimate analysis, ash, moisture, volatile solids, and total solids) for each tested fuel. TCs highlight the partitioning of the input elements (X) between the output streams of dedusted syngas ($TC_{X\ to\ syngas}$) and that of ashes discharged from the cyclone and the bed

Table 1
Description of waste and biomass fuels fed into the FBG plant.

Fuel	Feedstock	Description
Plastics	PE	Recycled polyethylene, derived from separate collection of MSW
	GS3	Mix of recycled polyolefinic plastics obtained from plastic packaging for food and beverages by means of sorting and washing treatments
	Neolite	Mix of plastics obtained from separate collection of post-consumer packaging made of plastic, but containing also ferrous and non-ferrous metals
	Plasmix	Mix of plastics obtained from separate collection of post-consumer packaging made of plastic, but containing also ferrous and non-ferrous metals
	PDF	Mix of different kinds of food packaging, generally consisting of multi-layer packaging of plastic, paper and aluminium
Biomass	RIL1	Wood residues utilised to prepare fuel pellets for domestic heating, which cannot be obtained from contaminated wood
	RIL2	Industrial medium-contaminated wood from recycling chain, made of sawdust from wood packaging industry
	RIL3	Industrial high-contaminated wood from recycling chain, obtained from furniture, doors and window frames
	WOOD1	Mix of different kinds of wood residues, having 22% of moisture content
	WOOD2	Mix of different kinds of wood residues, having 11% of moisture content
Co-fuel	Plasmix + RIL3	Mix of Plasmix and RIL3 with a ratio of 2:1 on mass basis

Table 2
Elemental composition of waste and biomass fuels tested in the FBG plant. TS = Total Solids, VS = Volatile Solids.

Fuel	Plastics					Biomass					Co-fuel
	PE	GS3	Neolite	Plasmix	PDF	RIL1	RIL2	RIL3	WOOD1	WOOD2	Plasmix + RIL3
Water (%)	0.2	0.3	0.6	0.7	5.6	6.7	7	11.3	22	11	1.90
TS (%)	99.8	99.7	99.4	99.3	94.4	93.3	93	88.7	78	89	98.1
Low Heating Value (MJ/kgTS)	45.6	45.6	33.1	40.5	24.1	18.1	25.1	23.1	17.2	16.4	37.8
					%TS						
VS	99.0	98.7	93.3	98.1	93.5	98.9	98.7	94.9	99.5	99.0	97.8
C bio	–	–	–	–	–	48.9	61.7	56.6	49.5	46.7	18.1
C fossil	85.2	84.7	68.5	80.1	57.1	–	–	–	–	–	57.3
H	13.8	14.0	10.3	13.2	8.2	6.0	7.9	7.9	5.9	6.2	12.2
N	–	–	–	0.2	0.5	0.1	0.8	2.6	0.9	1.4	0.61
S	–	–	0.1	0.1	0.1	–	–	–	0.1	0.4	0.1
Cl	0.002	–	1.019	0.052	0.282	0.391	–	0.091	0.052	0.052	0.06
O	–	–	14.4	4.5	27.6	43.9	28.4	27.8	43.1	44.3	9.38
Ash	1.00	1.30	6.74	1.91	6.53	1.07	1.29	5.07	0.51	1.01	2.24
Al	0.02	0.05	0.10	0.10	0.26	0.01	0.01	0.09	0.03	0.03	0.097
As	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	6.0E-06	4.0E-05	2.1E-05	–	–	1.3E-05
Ba	–	–	–	1.2E-05	–	–	–	–	–	–	7.8E-06
Bi	–	–	–	1.2E-03	–	–	–	–	–	–	0.0008
Bo	–	–	–	1.9E-06	–	–	–	–	–	–	1.3E-06
Ca	0.46	0.35	0.08	1.01	2.29	0.18	0.34	1.53	1.32	1.32	1.14
Cd	4.0E-05	3.6E-05	2.0E-04	4.8E-05	2.5E-05	6.0E-06	1.0E-05	7.6E-05	–	–	5.5E-05
Ce	–	–	–	1.4E-04	–	–	–	–	–	–	9.2E-05
Co	3.0E-05	5.1E-05	1.4E-04	1.1E-04	1.4E-04	1.7E-05	3.0E-05	1.0E-04	–	–	1.0E-04
Cr	6.8E-04	2.4E-03	1.0E-02	1.0E-03	6.1E-04	4.4E-05	2.0E-04	6.0E-04	4.5E-03	4.5E-03	8.7E-04
Cu	9.0E-04	1.6E-03	1.3E-01	1.7E-03	1.9E-03	1.0E-05	1.0E-05	3.8E-04	–	–	1.28E-03
Fe	2.8E-03	1.1E-02	4.9E-02	3.1E-02	1.4E-02	9.0E-03	4.3E-02	6.2E-02	6.5E-02	6.5E-02	3.9E-02
Hg	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.3E-05	1.0E-05	1.0E-05	–	–	9.8E-06
K	0.04	0.05	0.16	0.15	0.21	0.03	0.01	0.15	0.27	0.27	1.5E-01
Mg	8.3E-02	–	–	2.5E-02	–	1.8E-02	6.6E-02	1.3E-01	2.8E-01	2.8E-01	5.6E-02
Mn	1.0E-05	2.5E-05	1.9E-03	6.4E-04	7.1E-04	1.5E-03	6.3E-03	3.9E-03	2.2E-02	2.2E-02	1.6E-03
Mo	–	–	–	2.9E-05	–	–	–	–	–	–	1.94E-05
Na	3.5E-02	1.7E-02	7.5E-02	5.3E-02	9.1E-02	1.1E-02	1.9E-02	3.5E-01	–	–	1.40E-01
Ni	3.0E-05	1.4E-04	1.6E-02	1.1E-04	6.7E-04	2.0E-05	2.0E-04	3.8E-04	4.4E-03	4.4E-03	1.9E-04
P	–	–	–	2.6E-03	–	–	–	–	7.4E-02	7.4E-02	1.8E-03
Pb	3.3E-03	3.1E-02	8.7E-02	3.4E-03	8.2E-03	1.2E-04	4.0E-04	3.5E-03	–	–	3.3E-03
Sb	1.4E-04	7.0E-04	6.7E-03	1.0E-05	2.0E-04	2.0E-04	6.0E-05	9.9E-03	–	–	3.0E-03
Si	–	–	–	8.1E-03	–	–	–	–	7.5E-01	7.5E-01	5.5E-03
Sn	3.2E-04	9.9E-04	2.2E-03	1.7E-03	7.1E-04	3.3E-03	4.1E-03	5.0E-03	–	–	2.6E-03
Ti	–	–	–	1.1E-02	–	–	–	–	1.3E-02	1.3E-02	7.2E-03
Tl	3.0E-05	2.5E-05	2.5E-05	2.5E-05	2.5E-05	1.0E-06	3.0E-05	2.5E-05	–	–	2.4E-05
V	–	1.5E-05	1.9E-03	8.0E-05	2.0E-04	1.0E-05	2.0E-05	6.1E-03	–	–	1.9E-03
Zn	–	–	–	1.3E-03	–	–	–	–	5.3E-03	5.3E-03	8.82E-04

bottom ($TC_{X\ to\ ashes}$).

The carbon and hydrogen transfer coefficients are coupled with the related substance-to-compound Conversion Coefficients (CCs), which indicate the amount of C and H transferred into specific syngas compounds (i.e., CH_4, CO_2, CO, tar, H_2, H_2O and C_nH_m with n equals to 2 or 3 and m equals to 2, 4 or 6). Carbon-to-compound CCs have been obtained from experimental results by dividing the carbon mass flow rate in each syngas compound with the total carbon flow rate in the syngas. The same procedure has been applied for hydrogen, by coupling the experimental results with the atomic balances, and taking into account the amount of hydrogen present in the compounds already quantified by carbon-to-compounds CCs. The model then provides the main syngas characteristics, such as mass and volumetric flow rate, composition, and LHV. In particular, the volumetric flow rates of each compound in dry syngas, as well as mass flow rates of tars can be determined by the following two relationships.

$$\textbf{Compound } \boldsymbol{i}[\mathbf{m^3_N/h}] = \text{kg\_X*TC}_{\text{X to syngas}}\text{*CC}_{\text{X to i}}\text{*Volume\_gas}/(\text{MW\_X*n}) \quad (1)$$

Where:

- *i* indicates CH_4, CO_2, CO, tar, H_2, H_2O, C_nH_m, N_2;
- kg_X indicates the input of each element (C, H, and N) expressed as kg/h;
- MW_X indicates the atomic weight of each element (C, H, and N) expressed as kg/kmol;
- Volume_gas is the molar volume of the ideal gases, equals to 22.4 m^3_N/kmol.
- TC and CC for H, C and N are the coefficients for each kind of feedstock obtained from the experiments.

$$\textbf{CO}_{\mathbf{2,\ fossil}}[\text{kg/h}] = \text{kg\_C*TC}_{\text{C to syngas}}\text{*(MW\_CO}_2\text{/MW\_C)*(kg\_C}_{\text{foss}}\text{/ kg\_C)} =$$
$$= \left[(\text{CO}_{\text{syngas}} + \text{CO}_{2\ \text{syngas}} + \text{CH}_{4\ \text{syngas}} + 2\text{C}_n\text{H}_{\text{m syngas}} + 6\ \text{BTX}_{\text{syngas}})/(\text{Volume\_gas})\text{*(MW\_CO}_2)\right. \quad (3)$$
$$\left. + \ (\text{tar}_{\text{syngas}}\text{*\%}_{\text{m}}\text{C}_{\text{tar}}/\text{MW\_C*MW\_CO}_2)\right]\text{*\%C}_{\text{foss}}$$

- n is the number of atoms of the element X in the compound *i*. It is set equal to 2 for C_nH_m, which is an average value, based on the results of experimental tests.

$$\textbf{Tar}_{\text{syngas}}[\text{kg/h}] = \text{kg\_C*TC}_{\text{C to syngas}}\text{*CC}_{\text{C to Tar}}/\%_{\text{m}}\text{C}_{\text{tar}} \quad (2)$$

Where:

- kg_C is the input expressed as kg/h of C;
- TC and CC for C are the coefficients for each kind of feedstock obtained from the experiments.
- $\%_mC_{tar}$ is the mass fraction of carbon in tar compounds and it is assumed to be equal to 0.87, which is an average value based on the experimental tests.

2.3.2. Process-specific parameters

With reference to the process scheme of Fig. 1, several process-modelling parameters have been defined for the syngas combustor, the HRSG unit and the APC system. The identification of proper values for these process-specific parameters has been based on operational data for all the analysed fuels. Table 4 lists these parameters, the utilised values and the related data sources.

2.3.3. Life cycle inventory model

The main inputs and outputs provided by the inventory model, and based on the identified waste- and process-specific parameters, are: air emissions, electricity recovery, residues to be disposed, and material consumptions. Air emissions can be quantified in terms of syngas- and process-specific emissions. Syngas-specific emissions into the atmosphere are strictly related to the syngas composition, and include fossil carbon dioxide, sulphur dioxide, hydrogen chloride, heavy metals and particulate matter. Some of these emissions can be quantified by means of the following relationships.

$$\textbf{SO}_{\mathbf{2}}[\text{kg/h}] = \left[\text{kg\_S*TC}_{\text{S to syngas}}\text{*}(1-\eta_{\text{SO2}})\right]\text{*MW\_SO}_2\text{/MW\_S} \quad (4)$$

$$\textbf{HCl}[\text{kg/h}] = \left[\text{kg\_Cl*TC}_{\text{Cl to syngas}}\text{*}(1-\eta_{\text{HCl}})\right]\text{*MW\_HCl/MW\_Cl} \quad (5)$$

Where:

- kg_C, kg_C $_{foss}$, kg_S, and kg_Cl are the FBG inputs, expressed as kg/h of total C, fossil C, S, and Cl, respectively.
- TC are the coefficients for each kind of feedstock obtained from the experiments.
- MW_CO_2, MW_C, MW_SO_2, MW_S, MW_HCl, and MW_Cl are the molecular weights, expressed as kg/kmol of CO_2, C, SO_2, S, HCl, and Cl, respectively.
- CO $_{syngas}$, CO_2 $_{syngas}$, CH_4 $_{syngas}$, C_nH_m $_{syngas}$, and BTX $_{syngas}$ are the volumetric flow rates of each compound in the syngas, quantified by the relationships 1, expressed as m^3_N/h.
- tar $_{syngas}$ is the mass flow rate of tar in syngas, quantified by the relationship 2, expressed as kg/h.
- η_{SO2} and η_{HCl} are the pollutants removal efficiencies, reported in Table 4.

Table 3
Experimental tests utilised for FBG modelling [32–37].

Fuel	Feedstock	n. of experimental tests	Equivalence Ratio	Source
Plastics	PE	10	0.21–0.31	Arena et al., 2009 [32]
	GS3	5	0.23–0.27	Arena et al., 2010 [33]
	Neolite	4	0.22–0.33	Arena et al., 2012 [35]
	Plasmix	4	0.21–0.27	Di Gregorio and Zaccariello, 2012 [36]
	PDF	2	0.26–0.31	Arena and Di Gregorio, 2014 [37]
Biomass	RIL1	5	0.20–0.30	Arena et al., 2010 [34]
	RIL2	3	0.22–0.30	
	RIL3	3	0.21–0.31	
	WOOD1	3	0.28–0.33	This study
	WOOD2	6	0.26–0.35	
Co-fuel	Plasmix + RIL3	1	0.25	This study

Table 4
Parameters utilised for FBG modelling, for each process stage and valid for all types of waste and biomass fuels (process-specific parameters).

Process parameters	Amount	Short Symbol	Source
Syngas combustor and HRSG section			
Net electrical efficiency of the Organic Rankine Cycle turbine, %	17.7	EE	Arena et al., 2015 [38]
APC system			
Activated carbon consumption, kg/t $_{feedstock}$	0.5	–	Ardolino et al., 2017 [29]
Hydrated lime consumption, kg/t $_{feedstock}$	6.5	–	Ardolino et al., 2017 [29]
Removal efficiency obtained by the APC system (fabric filter + activated carbon), %	99	η_{APC}	Ardolino et al., 2017 [29]
HCl removal efficiency by means of hydrated lime, %	99	η_{HCl}	Ardolino et al., 2017 [29]
SO_x removal efficiency by means of hydrated lime, %	95	η_{SO2}	Ardolino et al., 2017 [29]
NH_3 emissions, mg/kg_{syngas}	0.038	–	Ardolino et al., 2017 [29]
NO_x emissions, mg/kg_{syngas}	73	–	Ardolino et al., 2017 [29] Cavaliere and de Joannon 2004 [41]
PCDD/F emissions, ng/kg_{syngas}	0.0074	–	Ardolino et al., 2017 [29]

- Volume_gas is the molar volume of the ideal gases, equals to 22.4 m^3_N/kmol.
- $\%_mC_{tar}$ is the mass fraction of carbon in tar compounds and it is assumed to be equal to 0.87, which is an average value based on the experimental tests.
- $\%C_{foss}$ is the ratio of mass flow rate of fossil carbon entering into reactor and that of total carbon.

Heavy metals and particulate matter emissions strictly depends on the fate of the ash forming matter. The latter, contained in the fuel as discrete particles or inclusions of the combustible matrix, can be converted into solid, liquid and gaseous compounds, and finally leaves the system as gas effluents, bottom ashes or fly ashes. Conversion of ash-forming material depends on many factors, such as temperature, surrounding gas atmosphere (oxidising or reducing), pressure, fuel particle size distribution, residence time. The form of occurrence of the ash-forming compounds in the fuel is another important factor [43,44]. Upon oxidising atmosphere, discrete mineral particles are quickly isolated at high temperatures, and then condensed during the cooling stage after leaving the furnace. Included minerals become more concentrated in the fuel matrix as the connecting hydrocarbon is consumed. Metal oxides can be reduced by carbon and then partially undergo oxidation, clustering and coalescence, forming a significant part of the particulate to be collected by the dust control system. The gasification environment reduces metal oxides to elemental metals, which have a lower boiling point, in a reacting atmosphere without any re-oxidation and clustering [45,46]. The quantification of the generated solid residues from the process (that is, the ashes from bed bottom and cyclone, and the fly ashes from APC section) together with air emissions of heavy metals and particulates have to take into account these phenomena. A reasonable estimation of these outputs can be done by means of the following relationships within the APC subunit:

$$\textbf{FBG Ashes}[\mathrm{kg/h}] = \mathrm{kg\_X}*\mathrm{TC}_{\mathrm{X\ to\ ashes}} \quad (6)$$

$$\textbf{APC residues}[\mathrm{kg/h}] = \left(\mathrm{kg\_Ashes}*\mathrm{TC}_{\mathrm{ashes\ to\ syngas}}*\eta_{APC}\right) + \mathrm{kg\_Spent\ chemicals} \quad (7)$$

$$\textbf{Heavy metal } \boldsymbol{y}[\mathrm{kg/h}] = \mathrm{kg\_Heavy\_metal\_}y*\mathrm{TC}_{\mathrm{metal}\ y\ \mathrm{to\ syngas}} *(1-\eta_{APC}) \quad (8)$$

$$\textbf{PM2.5}[\mathrm{kg/h}] = \mathrm{kg\_Ashes}*\mathrm{TC}_{\mathrm{ashes\ to\ syngas}}*(1-\eta_{APC}) \quad (9)$$

Where:

- kg_Ashes, kg_X and kg_Heavy_metal_y are the FBG input, expressed as kg/h;
- $TC_{X\ to\ ashes}$ are the specific quantified coefficients for each element entering into FBG with the feedstock (that is inorganic elements but also carbon and hydrogen) transferred to the FBG ashes.
- $TC_{metal\ y\ to\ syngas}$ is the specific quantified coefficient for each metal entering into FBG with the feedstock and transferred to the syngas as elemental metal.
- $TC_{ashes\ to\ syngas}$ is the specific quantified coefficient for ashes entering into FBG and escaping the reactor and the cyclone as particles in the syngas.
- η_{APC} is the removal efficiency obtained by the APC system (fabric filter + activated carbon) reported in Table 4.
- kg_Spent chemicals are chemicals utilised in the APC system, expressed as kg/h.

In particular, the value of $TC_{ashes\ to\ syngas}$ is affected by the design of the specific cyclone as well as by density and, above all, size distribution of particles generated during the FBG process. The latter parameters are in turn related to the above cited fate of the ash forming matter contained in the fuel [44] and the comminution phenomena (attrition, fragmentation, and percolation) occurring in the reactor [2,46]. A reasonable estimation of this value can be obtained based on data coming from the complete series of experimental activity with the pilot scale gasifier. However, the amount of ashes escaping the cyclone as particles in the syngas is generally very low, or negligible, when the gasifier is fed with plastic waste [2,33]. Process-specific air emissions include NH_3, NO_x, and PCDD/F. They are quantified on the basis of the amount of the produced syngas, as indicated in Table 4.

The specific electricity recovery has been calculated considering the chemical energy transferred to the syngas and the net electrical efficiency of syngas utilisation section, as showed by the following two relations, one for the recovered electricity and the second one LHV of the dry syngas.

$$\textbf{Recovered electricity}\left[\mathrm{kWh}/\mathrm{kg_{feedstock}}\right] = \mathrm{LHV_{Drysyngas}}*\text{Dry syngas specific yield}*\mathrm{EE} \quad (10)$$

$$\textbf{LHV}_{\textbf{Dry syngas}}\left[\mathrm{kWh}/\mathrm{m^3_N}\right] = \Sigma(\text{Compounds } i_{\mathrm{syngas}}\text{\_LHV} *\%_{\mathrm{vol}}\text{Compounds } i_{\mathrm{syngas}})/3.6 \quad (11)$$

Where:

- Dry syngas specific yield is the sum of syngas compounds (i.e. CO, CH_4, C_nH_m, H_2, tar, BTX, and N_2), expressed as m^3_N/kg feedstock.
- EE is the net electrical efficiency reported in Table 4.
- Compounds *i* $_{syngas}$_LHV are Low Heating Values of each compound in the syngas (i.e. CO, CH_4, C_nH_m, H_2, tar, BTX, and N_2), expressed as MJ/m^3_N.
- $\%_{vol}$ Compounds *i* $_{syngas}$ are the volumetric fractions of each compound in the syngas (i.e. CO, CH_4, C_nH_m, H_2, tar, BTX, and N_2).
- 3.6 is the conversion factor from MJ to kWh.

2.4. Lca case study

2.4.1. Goal and scope definition

The described FBG model has been used to develop an attributional and standardised LCA case study [47], carried out with the EASETECH software package. The intended application is investigating and quantifying the environmental performances of a FBG process, fed with different waste and biomass fuels. The functional unit is "the production of 1 kWh of electricity from the syngas combustion". The system boundaries (Fig. 2) include all the activities from the delivery of the feedstock at the FBG entry gate until to the management of all process residues. They can be schematised in a foreground system (i.e. the one analysed) and a background system (i.e. that interacting with the investigated system in order to provide the necessary materials and energy).

Data utilised for the foreground system derive from on-field experimental activities, and then are of high quality. Data for background system are provided by the ELCD 3.1 databank. The multi-functionality's allocation issues of the analysed system (i.e. the production of electricity and the waste safe disposal) have been approached with the system expansion methodology [48], by identifying which waste treatments are avoided when waste is instead gasified in the FBG. Life cycle environmental impacts have

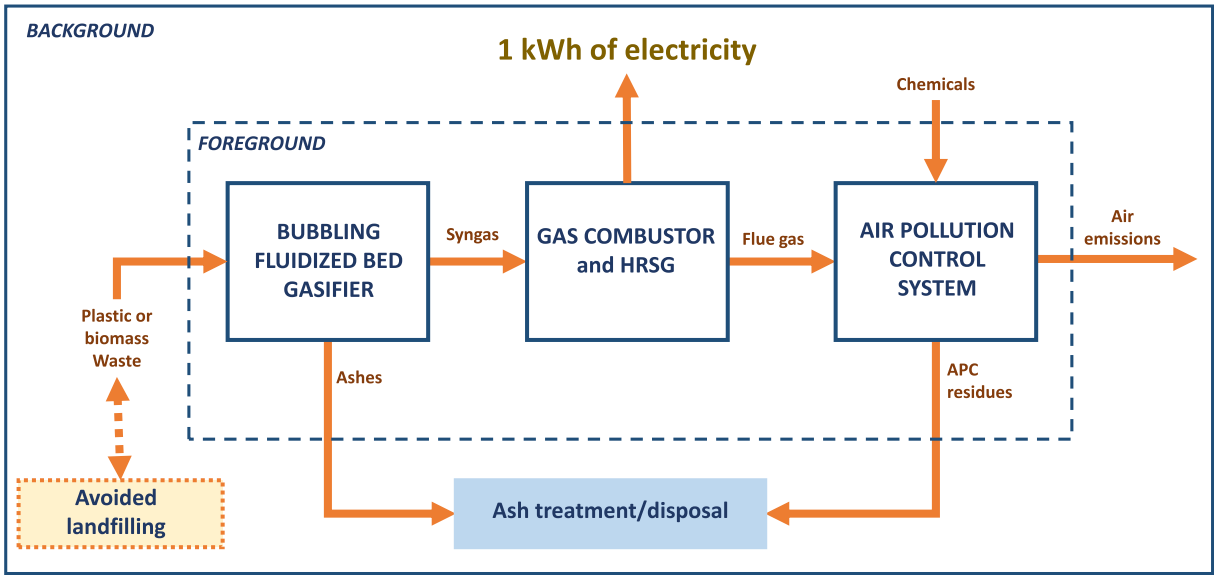

Fig. 2. System boundaries of the analysed fluidized bed gasifier, together with the indication of the foreground and background systems. APC = Air Pollution Control, HRSG = Heat Recovery and Steam Generator.

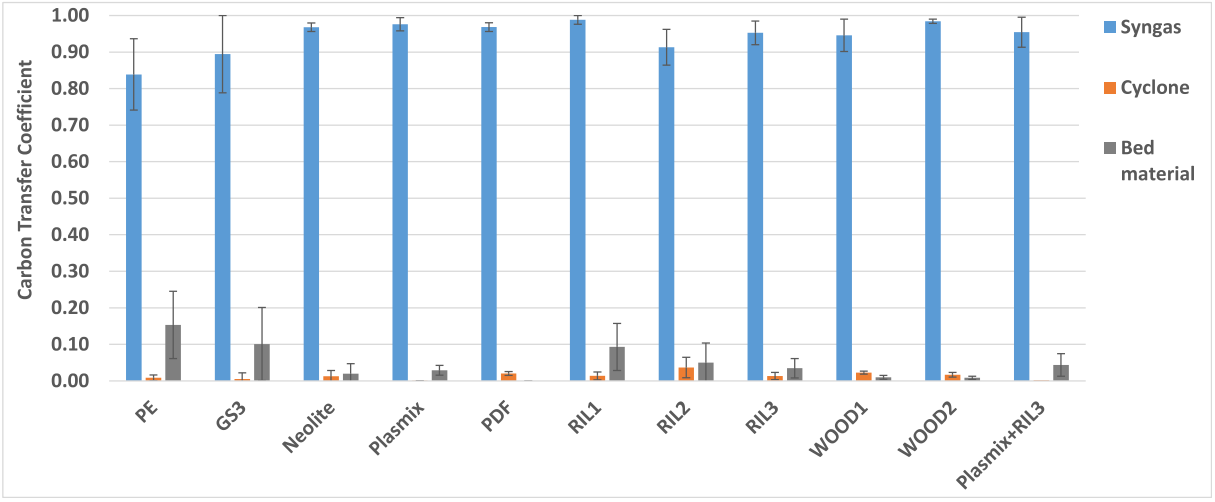

Fig. 3. Carbon transfer coefficients into syngas, cyclone and bed material for the selected fuels. Standard deviations (shown as bars) indicate the variations of TCs with reference to different values of ER.

been assessed by means of the ILCD 2011 methodology, developed by Joint Research Centre of the European Commission, and which includes 16 midpoint categories related to climate change, human health, eco-terrestrial toxicity and resource consumptions [49].

3. Results and discussion

3.1. Determination of waste specific parameters

Carbon transfer coefficients into the syngas ($TC_{C\ to\ syngas}$) for each of the fuels under analysis coincide with the related carbon conversion efficiency (CCE), defined as the flow rate of C converted to gaseous products with respect to that fed to the reactor. The average values of these $TC_{C\ to\ syngas}$ range from 0.84 to 0.98 for the selected waste fuels, as shown in Fig. 3, together with the standard deviation estimated by taking into account the different values of ER.

Hydrogen transfer coefficients into the syngas ($TC_{H\ to\ syngas}$) are generally higher than 0.99, while TCs into the syngas for oxygen, nitrogen, sulphur and chlorine are set equal to 1, then assuming that they are totally transferred into the produced syngas. With reference to the CCs, feedstock materials show different gasification behaviours (Fig. 4).

In particular, carbon contained in polyolefin wastes - the quasi-pure plastic streams, named PE and GS3 - is converted mainly into CO. For these wastes, the carbon conversion into tar is very low or substantially zero, due to the huge catalytic effect of olivine particles utilised as bed materials, which greatly improves the extension of tar cracking (dehydrogenation) and carbonization reactions [50]. On the contrary, this effect is only partially present during the gasification of other plastic fuels derived from the separate collection of MSW or industrial biomass waste, due to the inhibiting effects of metal impurities [33,37]. This catalytic effect is also evident in the high C content inside the bed (Fig. 3), which is a result of the catalysed carbonization reaction [50]. In particular, the larger standard deviation appearing in Fig. 3 for PE and GS3 is likely

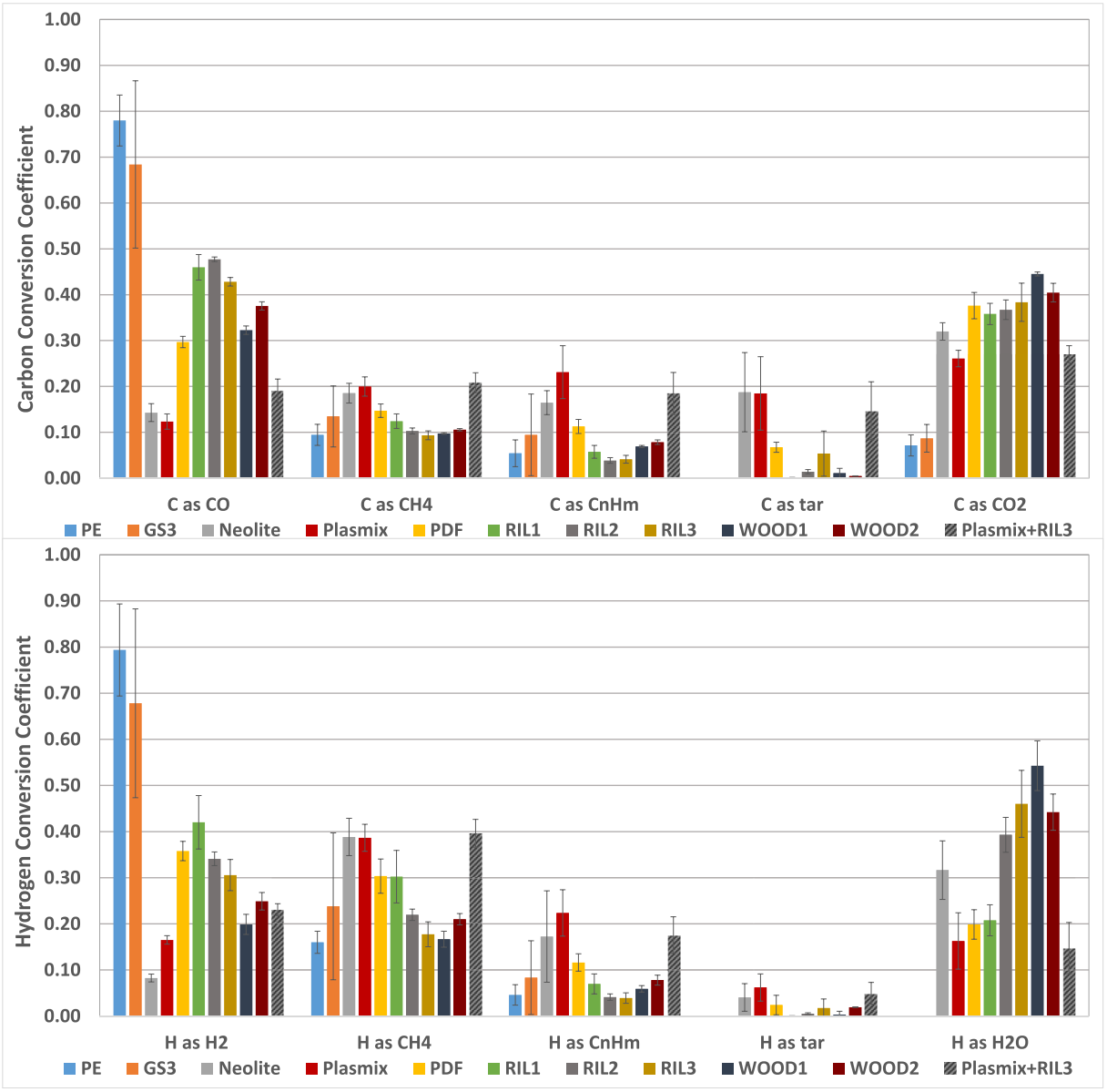

Fig. 4. Carbon-to-compounds (up) and Hydrogen-to-compounds (bottom) conversion coefficients for the selected feedstock materials. Standard deviations (shown as bars) indicate the variations of CCs with reference to different values of ER.

Table 5
TCs of inorganics in the fines collected at the cyclone, for some of tested waste and biomass fuels [32–34].

	TCs of inorganics in ashes from cyclone							*as*
	PE	Plasmix	PDF	RIL1	RIL2	RIL3	Plasmix + RIL3	
As	0.28	0.00	0.00	1.10	0.01	0.10	0.01	***As***
Al	0.89	0.01	1.07	0.91	1.00	0.37	0.04	Al_2O_3
Sb	0.77	0.46	0.41	0.03	0.85	0.03	0.02	***Sb***
Cd	0.70	0.01	0.20	0.40	0.73	0.13	0.02	***Cd***
Ca	0.23	0.00	0.00	0.12	0.30	0.22	0.02	***CaO***
Co	21.5	0.01	1.63	6.86	2.97	0.22	0.03	***Co***
Cr	5.70	0.01	1.18	3.79	2.61	0.34	0.05	***Cr***
Fe	189	0.04	0.01	8.80	3.38	0.68	0.08	Fe_2O_3
Mg	0.02	0.04	0.00	5.97	1.79	0.31	0.05	***MgO***
Mn	527	0.02	1.57	0.39	0.29	0.21	0.04	***MnO***
Hg	0.28	0.00	0.06	0.42	0.03	0.00	0.00	***Hg***
Ni	599	0.76	3.38	35.00	10.4	0.82	0.25	***Ni***
Pb	0.26	0.01	0.06	1.30	1.35	0.69	0.06	***Pb***
K	0.02	0.00	0.06	0.22	0.70	0.12	0.01	K_2O
Cu	1.25	0.01	0.83	16.49	41.5	0.77	0.11	***Cu***
Na	0.12	0.00	0.34	0.05	0.84	0.02	0.01	Na_2O
Sn	8.76	0.00	1.96	0.03	0.00	0.07	0.02	***Sn***
Tl	0.11	0.00	0.06	0.01	0.03	0.00	0.00	***Tl***
V	17.9	0.01	2.70	1.93	1.19	0.01	0.00	***V***
Cl	0.00	0.00	0.00	0.00	0.00	0.00	0.00	***Cl***

related to the uncertainty in the amount of C on the bed particles, which is affected by the quality and quantity of the bed sampling. Carbon and hydrogen entering with Plasmix, Neolite, and PDF are for larger extents converted into CO_2, CH_4, C_nH_m and tar, with lower generation of CO. Among the tested biomass fuels, the differences about carbon and hydrogen conversion coefficients are less evident, even though carbon entering with RIL3 shows the higher conversion into tar, because of the presence of glues and other additives in the biomass waste. As expected, the cogasification of Plasmix and RIL3 contributes to attenuate the disadvantages of the co-fuels, mainly in terms of tar and heavier hydrocarbons generation. These results suggest that the olivine bed particles work as an efficient tar removal catalyst during gasification of PE and GS3, and, even though to a lesser extent, of uncontaminated biomass. This positive effect reduces during operation with other fuels, which derived from biomass or plastics containing impurities. The differences, discussed in detail in Refs. [32,33] are crucial in the FBG modelling for life cycle assessment, since they largely affect the quantification of the main environmental burdens.

TCs of inorganics found in the fines collected at the cyclone have been quantified for PE, PDF, Plasmix, RIL1, RIL2, and RIL3 (Table 5): some elements showed an enrichment phenomenon, which leads to a mass flow rate in output higher than that in input. This phenomenon can be partially affected by the possible uncertainties in the metal content of the collected fines. Anyway, it is particularly interesting for iron, since part of that present on the external surface of olivine particles (and responsible of the catalytic activity for tar cracking) escapes the reactor due to the mechanical attrition between bed particles [50]. Based on these observations and taking into account typical cyclone efficiencies [46,51], TC for the total mass flow rate of ash entering with feedstock and transferred to the fines collected from cyclone has been conservatively set equal to 0.90. Accordingly, TC of ashes transferred to the syngas as particle has been set equal to 0.10.

TCs of inorganics in ashes are utilised also for the quantification of TCs for metals (As, Cd, Cr, Cu, Fe, Hg, Pb, and Sb) entering into the FBG with the feedstock and transferred into the syngas as elemental metals (Fig. 5), assuming negligible their stocks in the reactor [35]. Different values of TCs for metals are mainly related to the composition of feedstock, conversion of ash-forming material, and catalytic activity for tar cracking [43]. An average value of 0.45 for TC of Zn into syngas has been assumed for all the fuels, based on data reported by Arena and Di Gregorio [37].

The FBG model has been utilised for the quantification of two process performances: dry syngas yield and cold gas efficiency (CGE). The first is the sum of the volumetric flow rates of the dry syngas compounds. The second is the ratio between the chemical energy of the produced syngas and that of the feedstock fed to the reactor.

Results obtained from the FBG model, reported in Figs. 6 and 7, appear generally in fair agreement with those evaluated during experimental activity. The average error is, in particular, 3.5% for CGE and 6.1% for dry syngas yields. This rather good agreement was only partially expected, considering that experimental values were obtained for different values of equivalence ratio, fluidizing velocity and air preheating temperature, while the model assumes average

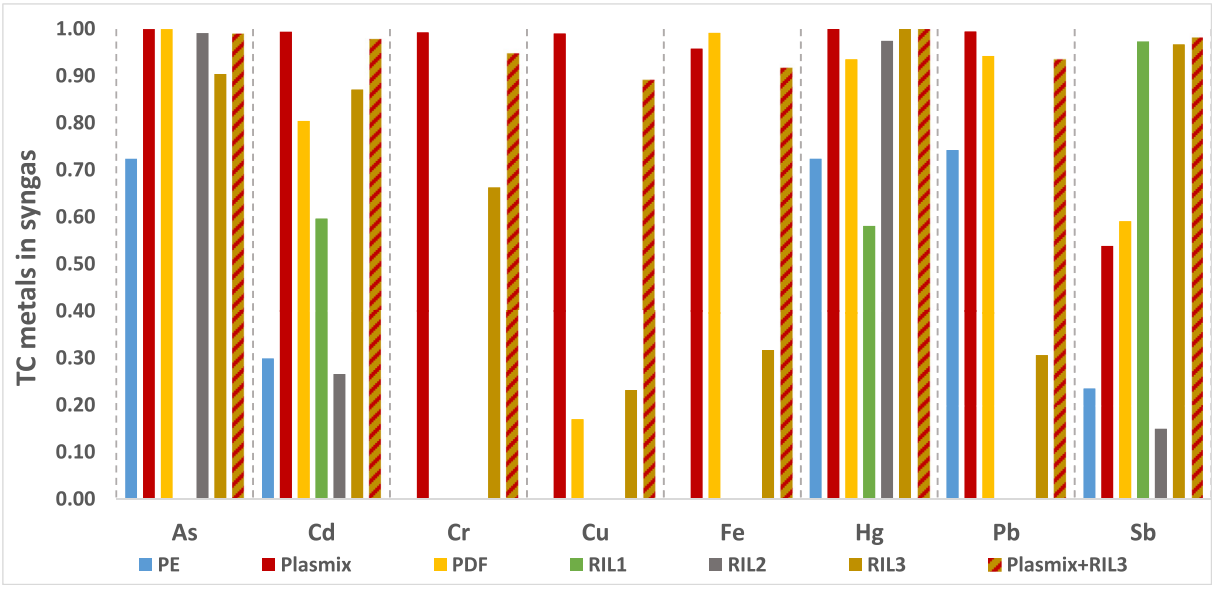

Fig. 5. Estimated transfer coefficients (TCs) for metals (As, Cd, Cr, Cu, Fe, Hg, Pb, and Sb) entering into fluidized bed gasifier with the feedstock and transferred to the syngas as elemental metal.

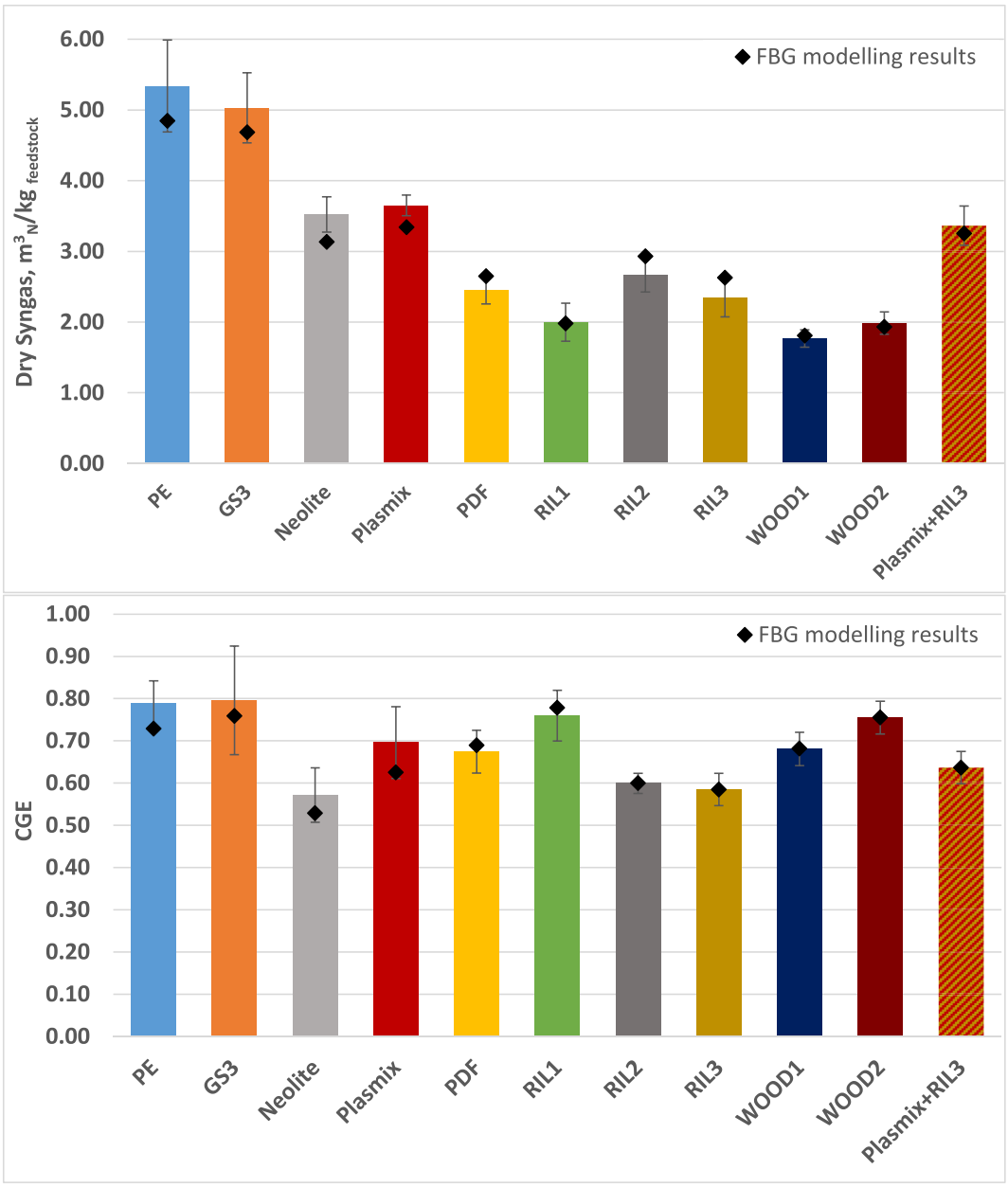

Fig. 6. Comparison between experimental and modelling results, in terms of dry syngas yield (up) and Cold Gas Efficiency (bottom).

values for all these parameters. A sensitivity analysis has been developed to investigate more on their effect.

3.2. LCI and LCIA results

Based on the standard procedures and the methodology described above, the FBG model implemented in EASETECH defines a life cycle inventory for the gasification process of the selected waste-derived fuels. Table 6 shows the LCI table, which also contains the feedstock amount required to obtain the production of 1 kWh of electricity. The direct emissions into the atmosphere for all the waste and biomass under analysis have been quantified by assuming that the TCs of metals (Fig. 5) defined for PE can be utilised also for GS3, those of Plasmix also for Neolite, and those of RIL1 also for WOOD1 and WOOD2, based on their similar chemical compositions and gasification behaviours. Burdens related to landfilling of plastic waste have been chosen as avoided burdens for all the plastic-based wastes, while those related to landfilling of biodegradable waste have been chosen as avoided burdens only for biomass-based wastes, then excluding the natural biomass (RIL1, WOOD1 and WOOD2).

An analysis of normalised results for all the impact categories considered in the ILCD 2011 (version EU-27) methodology suggests focusing the attention on the midpoint categories of Climate Change, Particulate Matter, and Human Toxicity. Fig. 8 reports LCIA results in terms of kg of carbon dioxide equivalent, for the gasification of the main fuels: it is evident the huge impact of the treatment of plastics waste, and the importance of avoided impacts related to the missed utilisation of landfill in the case of biomass waste gasification. PDF shows the worst performance since its gasification has a low energy efficiency (CGE = 0.69), and then higher amounts of feedstock are required for the production of 1 kWh of electricity.

RIL3 indicates the largest savings thanks to the avoided impacts

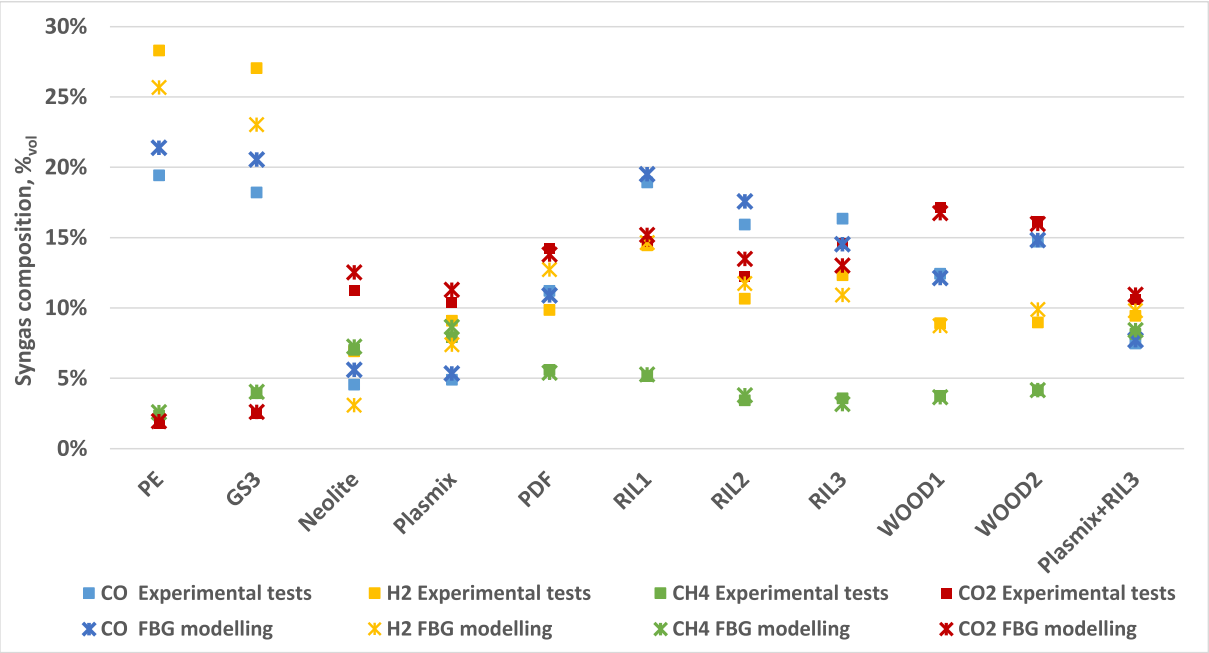

Fig. 7. Comparison between experimental and modelling results, in terms of volumetric concentration of the main syngas compounds.

Table 6
Life Cycle Inventory table generated by the FBG model, for the gasification process of the selected waste and biomass fuel and with reference to the functional unit.

Functional unit: 1 kWh of recovered electricity											
Feedstock	Plastics					Biomass					Co-fuel
	PE	GS3	Neolite	Plasmix	PDF	RIL1	RIL2	RIL3	WOOD1	WOOD 2	Plasmix + RIL3
IN (kg)	0.61	0.59	0.88	0.64	1.16	1.54	1.42	1.55	2.18	1.84	0.86
DIRECT BURDENS											
Consumptions											
Activated Carbon (g)	0.31	0.29	0.44	0.32	0.58	0.77	0.71	0.78	1.09	0.92	0.43
Hydrated lime (g)	4.0	3.8	5.7	4.1	7.5	10.0	9.2	10.1	14.2	12.0	5.6
Residues											
Cyclone ashes (g)	89.6	59.5	72.4	22.9	79.3	26.2	86.2	99.3	53.2	27.2	46.5
APC residues (g)	4.3	4.1	6.1	4.4	8.1	10.8	9.9	10.9	15.3	12.9	6.0
Direct air emissions											
$CO_{2\ fossil}$ (kg)	1.60	1.63	2.12	1.81	2.23	–	–	–	–	–	1.69
$CO_{2\ bio}$ (kg)	–	–	–	–	–	2.54	2.72	2.72	2.92	2.76	0.54
HCl (mg)	0.11	–	91.7	3.4	31.7	57.9	–	12.8	9.1	8.8	5.2
$PM_{<2.5\ \mu m}$ (mg)	6.1	7.6	58.9	12.1	71.4	15.4	17.0	69.7	8.7	16.6	18.9
SO_x (mg)	–	–	87	63	120	5	–	–	102	622	84.3
Sb (mg)	0.002	0.009	0.31	3.4E-04	0.013	0.03	0.008	1.10	–	–	0.24
As (mg)	0.0004	0.0004	0.001	0.001	0.001	–	0.005	0.001	–	–	0.001
Cd (mg)	0.001	0.001	0.02	0.003	0.002	0.001	0.001	0.006	–	–	0.004
Cr (mg)	–	–	0.87	0.06	–	–	0.02	0.05	–	–	0.07
Fe (mg)	–	–	4.1	1.8	1.5	–	1.81	–	–	–	3.1
Cu(mg)	–	–	11.5	0.11	0.03	–	0.0003	0.02	–	–	0.09
Pb (mg)	0.15	1.36	7.5	0.21	0.84	–	0.02	0.44	–	–	0.26
Hg (mg)	0.0004	0.0004	0.001	0.001	0.001	0.001	0.001	0.001	–	–	0.0008
Tl (mg)	0.002	0.001	0.002	0.002	0.003	0.0001	0.004	0.003	–	–	0.002
Zn (mg)	–	–	–	0.04	–	–	–	–	0.4	0.4	0.03
NO_x (mg)	42.9	42.1	55	44.5	76.6	217	99	101	0.06	119	56.9
NH_3 (mg)	0.02	0.02	0.03	0.02	0.04	0.10	0.05	0.05	3E-05	0.07	0.03
PCDD/F (ng)	0.004	0.004	0.006	0.004	0.007	0.02	0.009	0.01	6E-06	0.01	0.05
AVOIDED BURDENS											
Landfilling (kg)	0.61	0.59	0.88	0.64	1.16	–	1.42	1.55	–	–	0.86

from released biogas from landfilling of biodegradable waste. This scenario also has the best environmental performance in terms of kg of PM 2.5 equivalent, as showed by Fig. 9, even though gasification of this feedstock generates a remarkable direct impact related to the emissions of particulates into the atmospheres (69.7 mg of PM2.5 μm for 1 kWh of electricity). For the same impact category, gasification of plastic wastes shows considerably better performances, both in terms of generated and avoided impacts, mainly because of the avoided release of particulate and sulphur dioxide emissions during landfill operations.

This behaviour is confirmed (Fig. 10) in terms of "Human toxicity, cancer effects", for which biomass waste gasification has

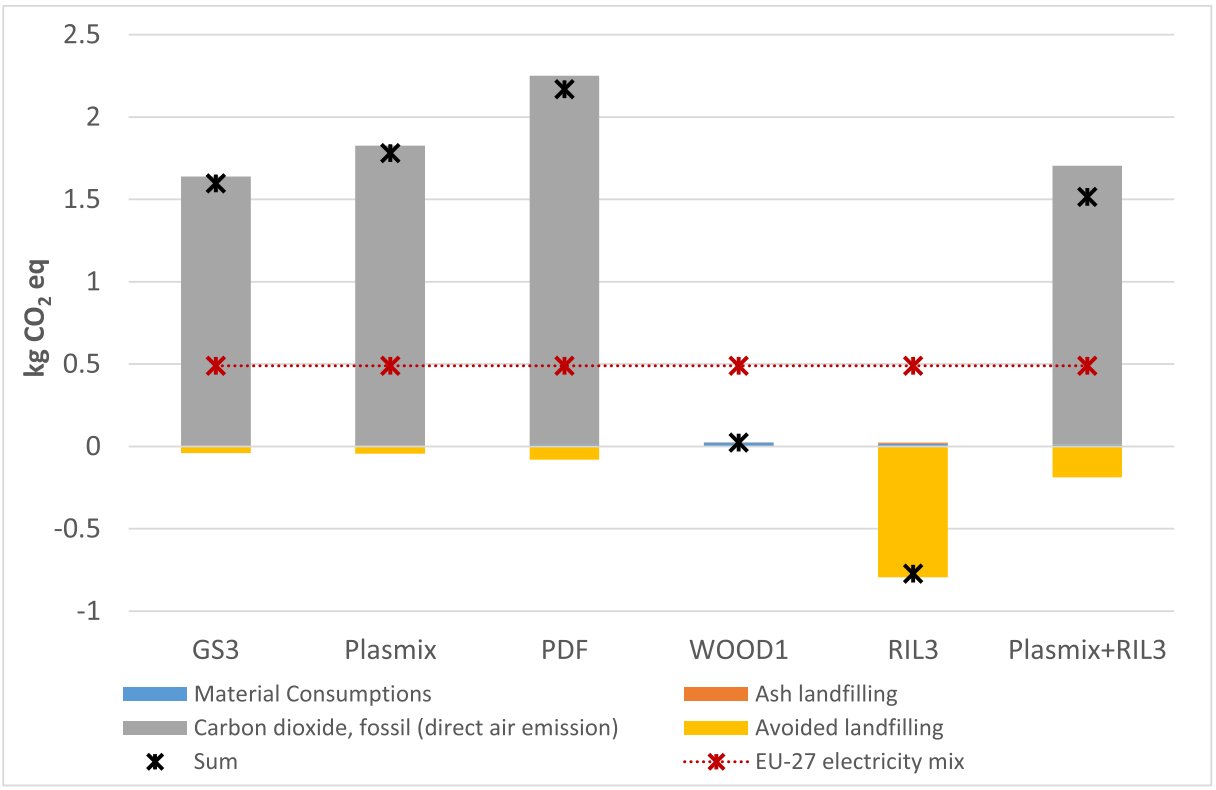

Fig. 8. LCIA results for the gasification of some of the analysed wastes, with reference to the "Climate change" impact category, and compared with those associated to the European electricity mix.

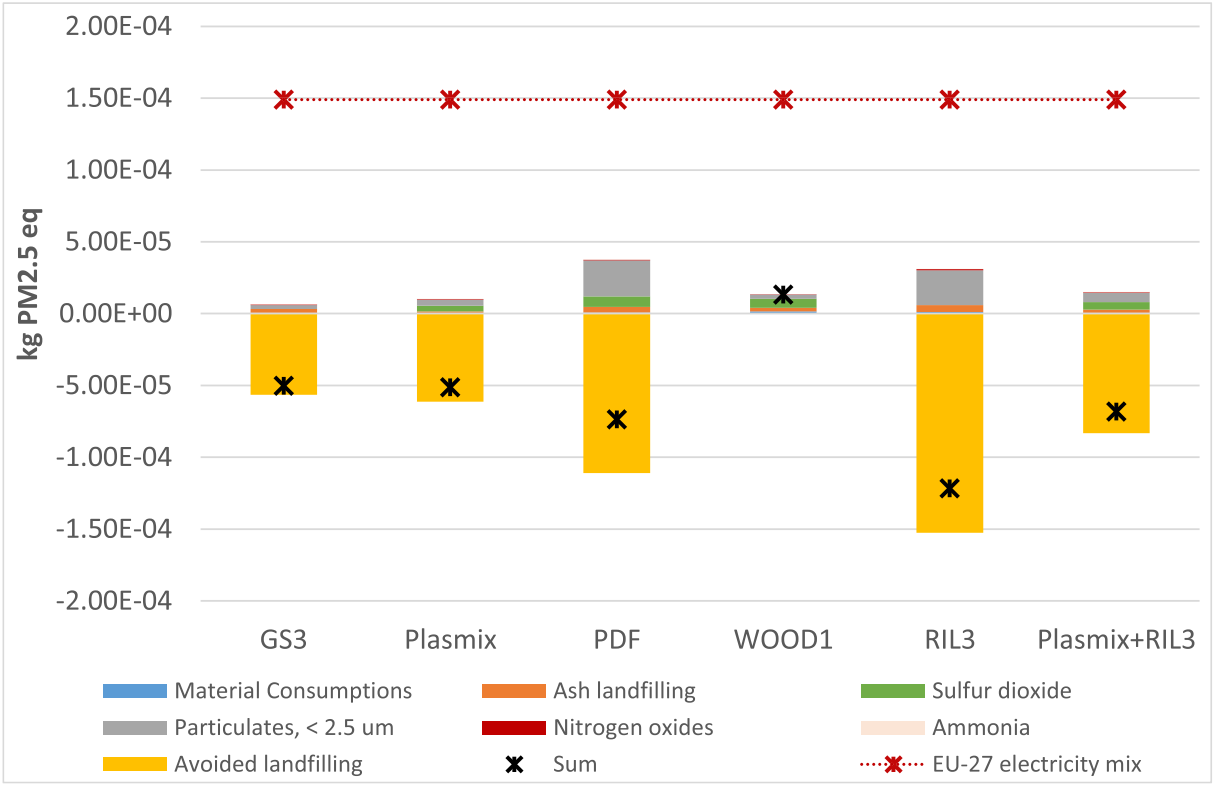

Fig. 9. LCIA results for the gasification of some of the analysed wastes, with reference to the "Particulate matter" impact category, and compared with those associated to the European electricity mix.

Fig. 10. LCIA results for the gasification of some of the analysed wastes, with reference to the "Human toxicity, cancer effects" (up) and "Human toxicity, non-cancer effects" (bottom) impact categories. Data have been compared with those associated to the European electricity mix. CTUh = Comparative Toxic Unit for humans.

the highest impacts. Higher avoided impacts related gasification of plastic waste are associated with the higher avoided emissions of arsenic in water, mercury in air, and zinc in soil during landfilling operations.

For the same impact category, Plasmix and RIL3 are negatively affected by direct air emissions of chromium (with an average value of 0.06 mg for 1 kWh of electricity). Highest lead air emissions of GS3 (1.36 mg of lead for 1 kWh of electricity) negatively affect the

Table 7
Sensitivity analysis on the effect of equivalence ratio on the main parameters of the implemented model.

Feedstock	GS3			Plasmix			PDF			WOOD1			RIL3		
	Min	Base Case	Max	Min	Base Case	Max	Min	Base Case	Max	Min	Base Case	Max	Min	Base Case	Max
ER, -	**0.23**	**0.25**	**0.27**	**0.24**	**0.25**	**0.27**	**0.26**	**0.27**	**0.31**	**0.28**	**0.28**	**0.33**	**0.21**	**0.3**	**0.31**
CGE, -	0.75	0.76	0.88	0.69	0.63	0.59	0.67	0.69	0.60	0.66	0.68	0.73	0.61	0.59	0.60
Feedstock IN, kg	0.6	0.59	0.51	0.57	0.64	0.67	1.2	1.16	1.34	2.26	2.18	2.04	1.36	1.55	1.38
TC Carbon to syngas	0.75	0.89	0.96	0.95	0.98	0.998	0.98	0.97	0.96	0.90	0.95	1.0	0.953	0.954	0.985
CC Carbon as CO	0.73	0.68	0.81	0.11	0.12	0.14	0.29	0.30	0.31	0.32	0.32	0.32	0.418	0.42	0.436
CC Carbon as CH_4	0.12	0.14	0.09	0.21	0.2	0.18	0.16	0.15	0.14	0.10	0.10	0.10	0.103	0.09	0.094
CC Carbon as C_nH_m	0.05	0.09	0.04	0.27	0.23	0.19	0.12	0.11	0.10	0.07	0.07	0.07	0.043	0.04	0.048
CC Carbon as tar	0	0	0	0.13	0.19	0.24	0.08	0.07	0.06	0.01	0.01	0.02	0.097	0.05	0.000
CC Carbon as CO_2	0.1	0.09	0.06	0.27	0.26	0.25	0.36	0.38	0.40	0.44	0.45	0.44	0.339	0.38	0.422

performance for "Human toxicity, non-cancer effects" category. Similar observations can be done for PDF, WOOD1 and RIL3, which show strong contributions due to the air emissions of lead, zinc and mercury. The role of avoided impacts appears less crucial in this impact category. The strong contributions of direct air emissions for human toxicity categories relate to the higher TCs of metals in syngas for PDF but also for Plasmix and RIL3, for both gasification or co-gasification modes (Fig. 5). It is noteworthy that data reported in Figs. 8–10 suggest better environmental performances of the gasification of all waste, when compared to the production of 1 kWh by the European electricity mix. The only exception is that of the GWP, which is strongly affected by the fossil origin of plastic wastes, and, on the other hand, by the increased fraction of non-fossil energy sources (biomass, solar, wind, nuclear, hydroelectric) in the European electricity mix.

A sensitivity analysis for an LCA study is generally carried out by changing different parameters in their reasonable range of variation and/or comparing possible different scenarios [22,52]. In this study, the situation is different since the data set of the reported experimental activity has been used also for implementing the

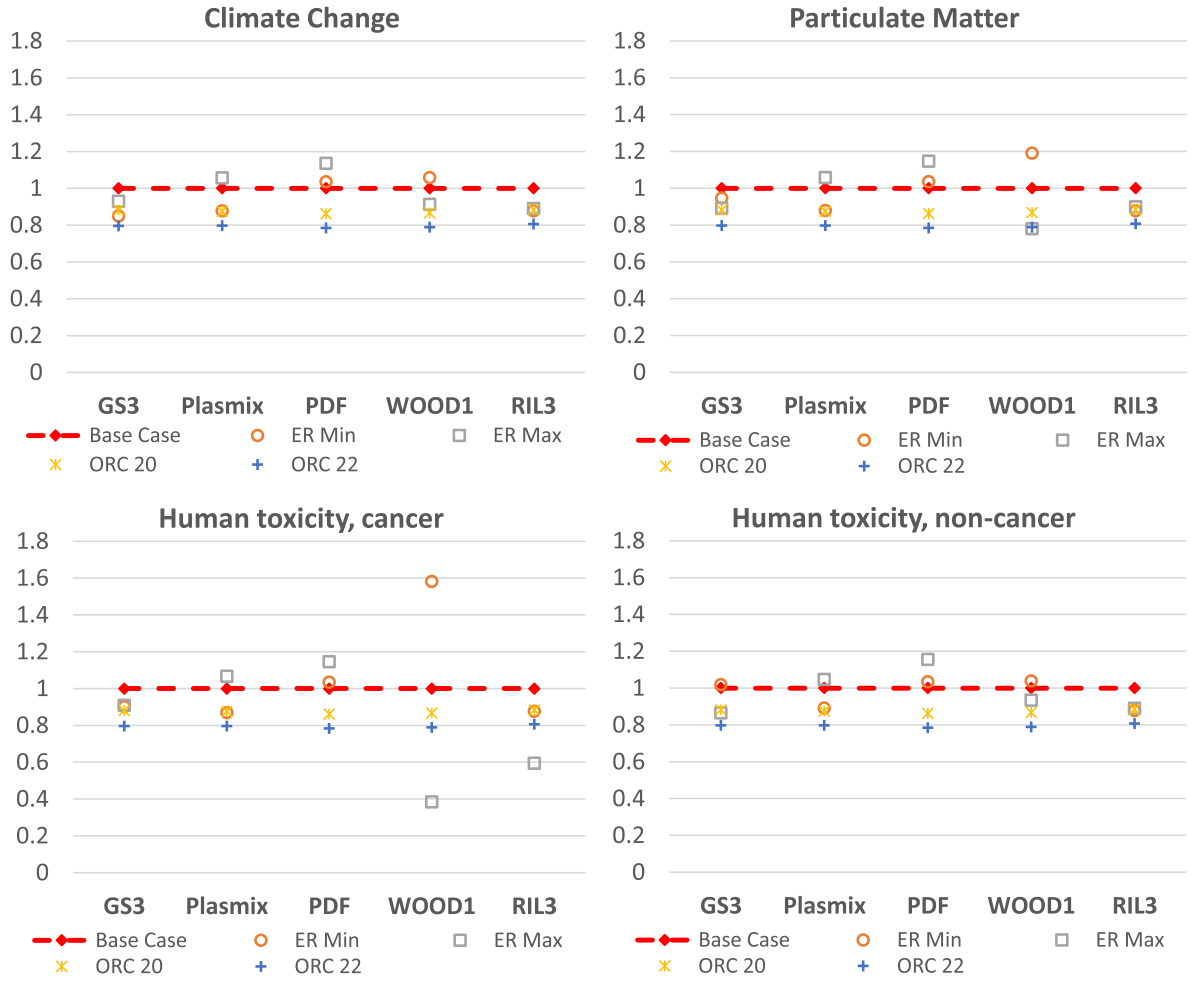

Fig. 11. Sensitivity analysis on the effect of ER and net electrical efficiency of the ORC, in terms of a variation factor VF, which transforms the base case value in that related to the modified parameter.

sensitivity analysis. Therefore, the analysis was limited to the effects associated with only two parameters. The first is ER, the most important operating parameter of waste gasification process [2,4], since it strongly affects the syngas composition (and then, its LHV and tar content) and cold gas efficiency of the process. The second parameter is the net electrical efficiency of the ORC, which relates to the final stage of syngas utilisation for energy production. The effect of ER on the main model parameters is reported in Table 7, which compares the model results as obtained in the base case (when an average value of ER has been considered) with those obtained by means of a re-determination of TCs and CCs based on the specific experimental results for ER values, at the extremes of their range of variation (Table 3). Fig. 11 reports instead the LCIA results in terms of a variation factor VF [11], defined as the ratio between the result for the changed parameter in the sensitivity analysis and that estimated for the base case. Then, VF = 1 indicates no variation; some variations occur when VF is <1 or VF > 1; and a negative value of VF changes the potential impact from positive to negative or vice-versa. Data in Table 7 and Fig. 11 suggest that the assumption of an average value of ER has limited effect on the final results, even though some notable exceptions in terms of Human Toxicity can be seen. The same figure reports also the data obtained by changing the value of the net electrical efficiency of ORC from the conservative value of 17.7% assumed in the base case to those of 20% and 22%, which can be better applied to medium-large commercial scale plants [53]. The effect seems again not remarkable.

4. Conclusions

A life cycle inventory model for the fluidized bed gasification of a set of waste materials has been developed for the first time, based on an extensive collection of experimental data. All data derive from a pilot scale fluidized bed gasifier, operated under auto-thermal conditions with ten types of waste and biomass fuels. The implemented model refers to commercial-scale gasification-based waste-to-energy systems, having a "thermal configuration" where the generated syngas is directly burned in a mild combustor for production of energy, and the obtained flue gases are cleaned in an air pollution control system.

The study does not develop a descriptive or predictive model for fluidized bed gasification of wastes but implement a life cycle inventory model for fluidized bed gasification of waste, intended to improve the quality of LCA studies in this field.

For each of the selected waste-derived fuels, the model quantifies the main inputs and outputs related to the gasification process: syngas- and process-specific emissions, generated electricity, ashes sent to disposal, and resource consumptions. The model appropriately represents the pilot-scale experimental data even though these were obtained for different values of equivalence ratio, fluidizing velocity and air preheating temperature.

The inventory model has been used to assess the environmental performances of some case-studies in an attributional LCA perspective. Potential environmental impacts and performance of the gasification process for the selected waste fuels were assessed with respect to selected impact categories, and compared with those associated with the average European mix for electricity production. The model appears to have limited sensitivity to the values of equivalence ratio and net electrical efficiency of the energy conversion device.

Acknowledgements

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Appendix A

Quantification of the fluidizing air stream

The following relationships quantify the amounts of O, N, and H, expressed in g/h, which enter into the reactor with the fluidizing air stream.

$$\mathbf{O}[g/h] = ER*((kg\_C/MW\_C*2) + (kg\_H/(MW\_H*2)*1) + (kg\_S/M\_S*2))*(MW\_O)*1000 \quad (A.1)$$

$$\mathbf{N}[g/h] = (ER*((kg\_C/MW\_C*1) + (kg\_H/(MW\_H*2)*0.5) + (kg\_S/M\_S*1))/0.21)*0.79*2*MW\_N*1000 \quad (A.2)$$

$$\mathbf{H}[g/h] = (ER*(((kg\_C/MW\_C*1) + (kg\_H/(MW\_H*2)*0.5) + (kg\_S/MW\_S*1))/0.21)*MW\_Air)*\%_{mass}H*1000 \quad (A.3)$$

Where:

- kg_C, kg_H and kg_S, are the inputs expressed as kg/h of C, H, and S, respectively;
- MW_C, MW_H, MW_N, MW_S and MW_Air are the molecular weights expressed as kg/kmol of C, H, N, S and air, respectively;
- 0.21 and 0.79 are the molar fractions in the air of O and N, respectively;
- $\%_{mass}H$ is the mass fraction of the H entering as moisture in the air.

According to the standards of EASETECH software, Volatile Solids (VS) have been defined as the fraction of all the input streams that can be converted in gaseous components (i.e. organic matter). VS can enter the reactor with the solid fuel and the fluidizing gas. In particular, VS_{air} is the mass of volatile solids that enters the reactor inside the air stream fed from the gas distributor at the bed bottom:

$$\mathbf{VS_{air}}[g/h] = O[g/h] + N[g/h] + H[g/h] \quad (A.4)$$

while VS_{fuel} is the mass of volatile solids that enters the reactor in the solid fuel stream. The latter is reported in Table 2, as obtained from the total fuel mass excluding the inorganic components.

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III

Energy recovery from plastic and biomass waste by means of fluidized bed gasification: A life cycle inventory model

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A modelling framework for process-oriented life cycle assessment (EASETECH+)

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Abstract

Life cycle assessment (LCA) modelling of resource related technologies can be challenging due to the complexity and need for appropriate disaggregation into unit-processes, e.g. in the context of circular economy, bioeconomy, recycling and waste management. Existing LCA models are poorly equipped to implement complex material conversion processes with property dependent linking between unit-processes while maintaining consistency of material/substance/energy flows throughout the assessed system. As an extension to EASETECH, an existing user-friendly LCA software for modelling of environmental technologies, EASETECH+ allows LCA practitioners to implement advanced process models, disaggregate technologies into relevant unit-processes, make material, substance and energy conversion dependent of intrinsic material flow properties, and subsequently import these process models into EASETECH for full environmental system level assessment. EASETECH+ thereby provides a fully flexible modelling framework for process-oriented assessment of integrated technologies with the potential for involving precisely the parameters of relevance. EASETECH+ and the associated modelling language is described in detail and applied to an illustrative case of biological processing (via anaerobic digestion) of degradable organic waste for generation of biogas and digestate.

Keywords

Modelling framework; non-linear relationships; mass balance; EASETECH; (multi-)conditional-sequence flow

Highlights

- Establishing a framework for process-oriented modelling: life cycle assessment EASETECH+
- Extension of the existing LCA model, EASETECH, focusing on waste and resource systems
- Implementing an illustrative case of biological processing (via anaerobic digestion) of degradable organic waste for generation of biogas and digestate
- Demonstrating the integration with EASETECH
- Evaluating modelling perspectives and applicability of circular resource systems

Abbreviations

DM: Dry matter; TS: Total solid; ww: wet weight

1 Introduction

The world is facing a wide range of challenges associated with anthropogenic activities, socio-economic growth, urbanization and industrialization. Resource consumption and limited access to resources both lead to competition among individual economic sectors and environmental impacts (IPCC, 2018; Olhoff et al., 2018; World Bank, 2019, 2020). To mitigate future climate impacts, improve resource efficiency in society, and minimize environmental impacts from urban activities, complex decisions involving a wide range of resource-oriented technologies are required (e.g. Beatriz et al., 2019; Capizzi et al., 2019). Circular economy initiatives involving recycling of several materials and biotechnologies are examples of solutions involving ranges of several steps represented by physical flows (Vanhamäki et al., 2020). New technologies create opportunities for more efficient resource management (Böckin et al., 2020), but technology performance depends on the framework conditions, process configurations and implementation. Technologies that appear to improve environmenntal performance may cause adverse effects when evaluated in a system perspective. Without the ability to analyze individual technologies and processes into smaller units, to model the environmental performance of these units, and subsequently to integrate the processes into a full system, decisions may be based on limited insights into process dependencies, importance of technology parameters, and false assumptions about environmental benefits. Ultimately, this may lead to unintended increase in environmental emissions and implementation of new technologies without the promised contributions to sustainability (UN, 2019; UNEP, 2018). As such, robust and flexible modelling tools are needed to ensure appropriate environemental assessments of circular and resource-oriented technologies.

LCA is a standardized methodology for quantifing and analyzing the full life cycle of products, technologies, systems, and services (ISO, 2006a, 2006b; EC-JRC, 2010a, 2010b). LCA can provide decision-support for identification of preferred options in view of the environmental consequences across a wide range of impact types. As such, LCA software tools involve handling and editing of large amounts of data about processes, materials, products, etc. (Cooper et al., 2013). A range of different LCA softwares exists, some of them are topic-specific, such as *GREET,* (2020) (Greenhouse gases Regulated Emission and Energy use in Transportation) for transportation some other are general, such as *Umberto NXT LCA*, (2020), *SimaPro*, (2020) , *openLCA*, (2020), and associated life cycle inventory (LCI) databases, such as *Ecoinvent,* (2020), *Agri-footprint,* (2020) specific for food (please refer to EPLCA, 2020, for a full list of tools). Software tools and databasesinvolve different modelling approaches In Gentil et al., (2010), eight waste LCA models

were identified based on selection criteria, reviewed and compared with the conclusion that input/output data, parameters, and assumptions reflect differences in results with the need for quantify and prioritise key parameters to validate and make them consistent internationally and among different models. However, LCA of new technologies typically builds on available studies for existing technologies and adaption of existing models. This means that technology data, performance assumptions, and parameters may not appropriately represent the new technology in question (Henriksen et al., 2018). This results in loss of information, but also the need for a more process-oriented LCA modelling approach for individual technologies and technology systems having input-output relationships as suggested by Lodato et al., (2020). Such relationships can be linear or non-linear (e.g. linear regression and logarithm, respectively) and are necessary for process modelling where simple mass balances are not sufficient (Unger et al., 2009). So far, no existing LCA models allow implementation of non-linear relationships in a modelling framework where the material properties at substance and fraction levels (e.g. hydrogen or cellulose content as substances, and softwood as fraction) are tracked throughout the individual processes/technologies affected by the material flows. In addition, parametrization increases transparency, representativeness and also a wider applicability of process models by creation of process templates which can be adapted to specific cases, simply by changing the values of individual parameters (Cooper at al., 2012). However, this places demands on both the inventory data and the LCA model structure (Kuczenski et al., 2018). Traditional product-oriented LCA modelling tools are not able to "track" the flow of resources and their properties through technologies and integrated systems of technologies considering all the transformations and transitions at a substance level through intermediate products to the final product state (Blomsma and Tennant, 2020).

In this study, we expand an existing LCA model, EASETECH (Clavreul et al., 2014), with a framework for process-oriented life cycle assessment modelling, EASETECH+, allowing users to model new advanced processes within environmental technologies in view of the circularity of resources. The technology processes developed in EASETECH+ can be directly implemented into EASETECH and applied across different LCA studies. This enables LCA practitioners to flexibly create new processes; something that previously required reprogramming of the software model itself. Thereby, EASETECH+ facilitates modelling of new processes in a more transparent, reproducible, and user-friendly way.

The overall aim is to establish and document the EASETECH+ modelling framework, including: i) description of the graphical user interface together with its operators and functions,

(multi-)conditional sequence flows, iteration and uncertainty propagation (Section 2); ii) demonstration of EASETECH+ modelling framework of an illustrative case-study related to biogas production from organic waste (Section 3), and finally iii) evaluation of novelties and potential implications for LCA modelling of circular resource systems (Section 3).

2 Materials and methods

2.1 EASETECH+ modelling framework

Modelling in EASETECH+ starts with a "white canvas" where LCA practitioners can "draw" the process flows with the level of detail needed according to the available information (e.g. technology configuration) and the requirements of the study (e.g. at technology level involving more details, or at system level with less details and a wider perspective of the assessment). During the modelling, LCA practitioners should adhere to the mass and energy balance principles of material flow analysis (MFA). In EASETECH, there are three macro levels to represent the nature of components in a flow: substances, fractions, and materials (Lodato et al., 2020). Substances are properties: biochemical (representing the elements of the organic fraction, i.e. organic molecules, such as cellulose), physical (e.g. ash content), nutritional (e.g. digestable energy, expressing the level of a biomass to be digested by animals) and chemical (e.g. alluminium, calcium, iron) of a fraction. Fractions are "entities that shares common characteristics, i.e. substances" (Lodato et al., 2020). One or more fractions define a material, for example "municipal solid waste" is a material and its fractions can be "vegetable food waste", "magazines", "plastic bottles", etc. The process flow needs to be clear, i.e. units are expressed, parameters are defined and input-output relationships are documented for reliability and transparency. The modelling can be based either on theoretical calculations, on results from experiments, i.e. empirical values, or a combination of both.

EASETECH+ represents an "evolution" of the LCA software EASETECH (Clavreul et al., 2014). EASETECH allows the modelling of technologies based on specific process "templates" (e.g. splitting of mass and substance flows, addition of background processes, specific modules for landfill and anerobic digestion, etc.) with linear relationships. However, these process templates were limited to the operations envisioned when EASETECH was developed originally (Clavreul et al., 2014). In EASETECH, the only way of adding new modelling features in form of new process templates was to change the source code of the software. This required recompiling and redistributing the full software package (Zarrin et al., 2014). The situation made it impossible for a LCA practitioner to add new processes to EASETECH and represented a barrier for expanding the applicability of the models.

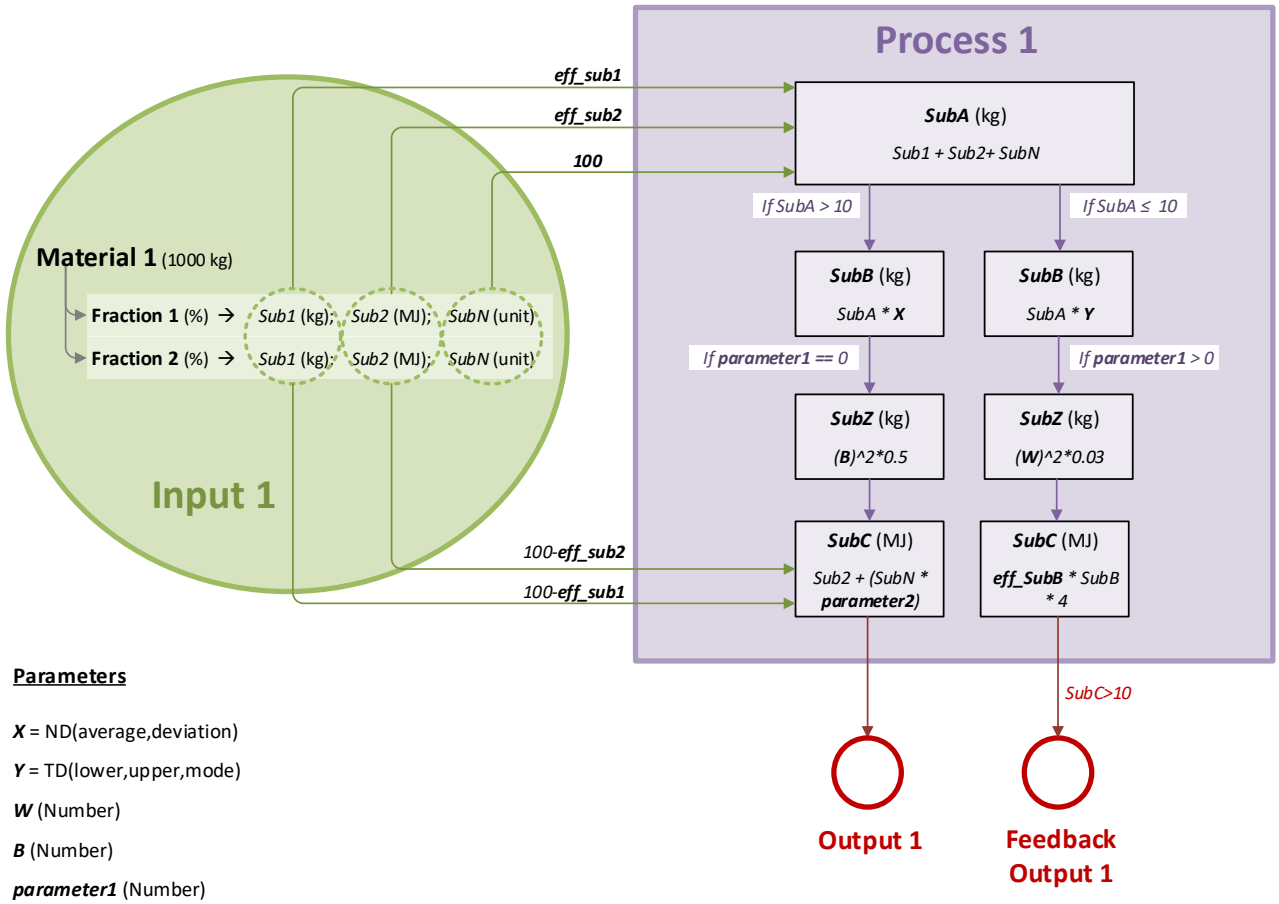
Accordingly there was a need for a new framework, EASETECH+, allowing LCA practitioners to model any type of operation they could envision and to implement these as new process templates in EASETECH, without the need for recompilation and redistribution of the EASETECH software itself. While EASETECH only had linear flow relationships, EASETECH+ enables both linear and non-linear flows. In addition, multiple "if functions*"* with conditions, here called (multi-)conditional-sequence flows are allowed following the conditional process modelling (Hayes and Preacher, 2013), where the process pathways are dependent on the outcome of other processes or calculations involving variables and parameters. In a conditional-sequence flow, the sequence defines the flow, i.e. which calculation steps are performed before other. This involves suitable parameters and constrains (Zavatteri and Viganó, 2018); while parameters depend on the process itself, particularly they affect the mass/energy balance, constrains define the "pathway" (or flow) that is enabled or disabled during the process exectution.

The outcome of EASETECH+ is a process flow diagram, i.e. a visual representation of all linear and non-linear input-output relationships that may subsequently be imported into EASETECH as a new process template. In EASETECH+, a dynamic-link library (*.dll* file) is generated then allowing further distribution to other users as an EASETECH process template. As such, the process flow diagram itself represents a disaggregation of individual processes once imported into EASETECH. The output of a process flow diagram can be intermediate or final. An intermediate output refers to a substance, fraction, or material generated during the modelling stage and further used to generate another output, thus the intermediate output is part of a process flow, but not one of the final outputs of the process. The final output is the (last) output of the process obtained after the execution of all the expected transitions and transformations of substances and/or fractions within the process flow diagram. Transformations refer to changes in substances of fractions and materials and may be partial and full. Substances that are fully transformed into other substances cease to exist e.g. when proteins are transformed into amino acids while still keeping track of the elemental composition such as C, H, O, N, S, etc.

The modelling framework begins with the input characteristics defined by its material, fractions and substances (Input 1, Figure 1) in kg_{ww}, kg_{TS}, m^3, MJ, etc. (where *ww* is wet weight and *TS* is total solid or dry matter, DM). Then, each substance within each fraction of a material in the input is tracked via individual material flows, represented by arrows linking input(s)-process-output(s) sequences. Material flows follow a sequence determined by the process configuration, from the execution of the first component until the last, ending the modelling with the final process

output(s). The modelling allows (multi-)conditional-sequence flows: by including a condition on an ongoing flow, the model execution evaluates the condition and continues only if the condition is met. If a process is parametrized, multiple conditions are possible with parameters, e.g. "if X>Y then B, or else C". This facilitates different calculation pathways depending on the type of input data available, such as in Figure 1 where *parameter 1* equals zero, i.e. *parameter1==0*, the flow on the left is enabled and the flow on the right (the one having the condition *parameter1>0*) is disabled. Additionally, iterative operations can be implemented to enable repetition of transitions and transformations for a defined number of times. Transitions may be iterated to repetitively transfer a material, a fraction or a substance from a source to a target component in a flow . Transformations may be iterated to repeat generating a material, a fraction or a substance within a specific flow. Two different types of iterations are implemented:

- *min/max iterations*, when the number of iterations are specified within a minimum and maximum value;
- *loop iterations*, when a sequence of statements is specified once but may be executed several times in succession. The loop is closed once the condition is met. In Figure 1, feedback output contains a condition that produces a loop; it stops once *SubC* is greather than 10. The example in Figure 1 is only for illustrative purposes (see Zarrin, 2017, for further details).



Parameters

X = ND(average,deviation)
Y = TD(lower,upper,mode)
W (Number)
B (Number)
parameter1 (Number)
parameter2 (Number)
eff_sub1 (%)
eff_sub2 (%)
eff_SubB (%)

Figure 1: Example of the EASETECH+ modelling framework, e.g. (multi-)conditional-sequence flows

2.2 Uncertainty propagation in EASETECH+

Parameters have an important role: they control substance and fraction balances and are used for sentitivity and uncertainty assessment via data distributions. EASETECH currently allow four different distributions (normal, uniform, log normal, and triangular) used for Monte Carlo simulation. Parameters are specific for each process and scenario. Therefore, in (multi-)conditional-sequence flows in EASETECH+ parameters can be added with distributions for propagation of uncertainty. To our knowledge, EASETECH+ is the first dedicated LCA modelling framework that allows such (multi-)conditional-sequence flows in the process modelling with linear and non-linear responses.

2.3 Link between EASETECH+ and EASETECH

The outcome of EASETECH+, i.e. the process flow diagram, is directly implemented into EASETECH as a process template for the LCA modelling and further adjustment of parameter values, if needed. In Figure 2, we provide an illustrative example of how components of a process flow diagram, here called "operators", can be combined and applied in EASETECH+. In this example, a process with the name *Exporting_to_EASETECH* is modelled. When exporting this process to EASETECH, it appears in the list of EASETECH's material processes and it is ready to be used in LCA scenarios. The final outcome in EASETECH is shown in Figure 2 (b) with the combination of operators used in EASETECH+ (a, Figure 2): one input and three outputs (*Residue*, *Water*, and *Chlorine*). The selected fraction has a range of properties. These properties are fully (i.e. 100%) transferred to a *substance hub* through a *material flow*. Subsequently, Chlorine and water are extracted through a *substance distributor* and Chlorine is fully transferred to the output Chlorine through a *material flow*, while water is partially transferred (i.e. less than 100%), only 50% of the input, to the output Water and its residue, 50%, is transferred through a *residue flow* to the output Residue. In general, residues represent the material, fraction or substance left in the operator after their partial or total transition to another operator. In the example in Figure 2, residues represent the remains of the *substance hub* excluding materials that were transferred to the other two outputs.

Further, these residues are fully transferred to the output Residues through a *residue flow*. In EASETECH, *material generation* is used to define the matrial input. In this example, we selected willow with the input composition shown in Figure 2. When Chlorine is fully transferred to one of the final outputs of the process, no Chlorine remains in the residue; however, when water is partially transferred, 50% ends up in the output *water* and 50% in the residue (see the composition of Output 1, Output 2 and Output 3, Figure 2).

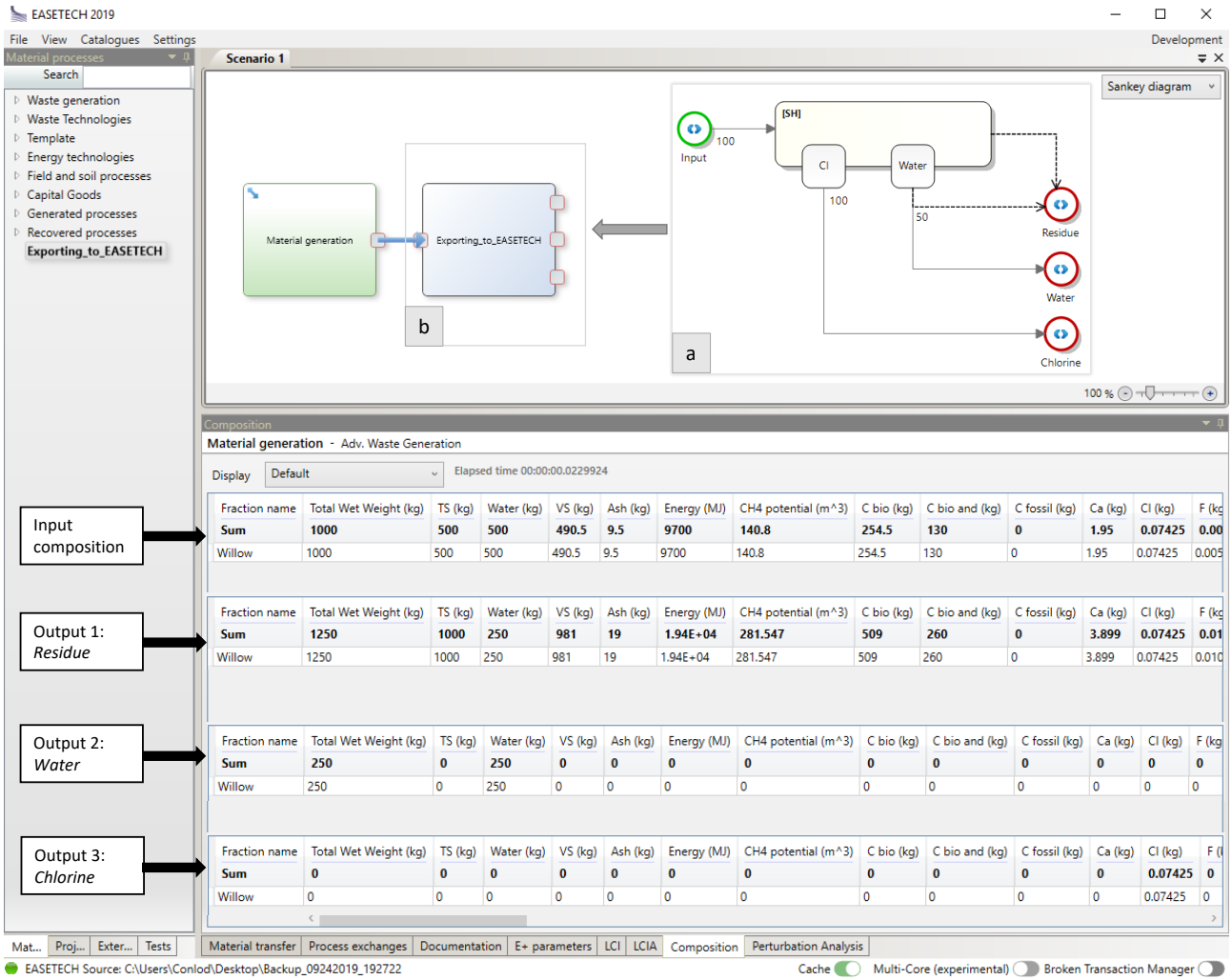


Figure 2: Illustrative representation of a EASETECH process template "*Exporting_to_EASETECH*" developed in EASETECH+ and used in an LCA scenario combined with a process "*material generation*" already in EASETECH. The process flow diagram (process flow diagram) of *Exporting_to_EASETECH* is shown in the figure, but it is only visible in EASETECH+. Below, compositions of: i) input, from *material generation*; ii) output 1, residue; iii) output 2, water; iv) output 3, chlorine. The composition of these three outputs follows the modelling structure defined in EASETECH+

In EASETECH+, the level of detail of process flow diagrams can be defined by the LCA practitioner according to the scope of the LCA following the process-oriented modelling approach. More details can be involved at technology level with specific input-output relationships e.g. involving chemical elements and several intermediate outputs, while a system level focus may require less details.

2.4 Graphical user interface

For the modelling of a process template, EASETECH+ has operators that can be used and combined according to the specific process in question, e.g. configuration, input properties and parameters. The graphical user interface of EASETECH+ is presented in Figure 3.

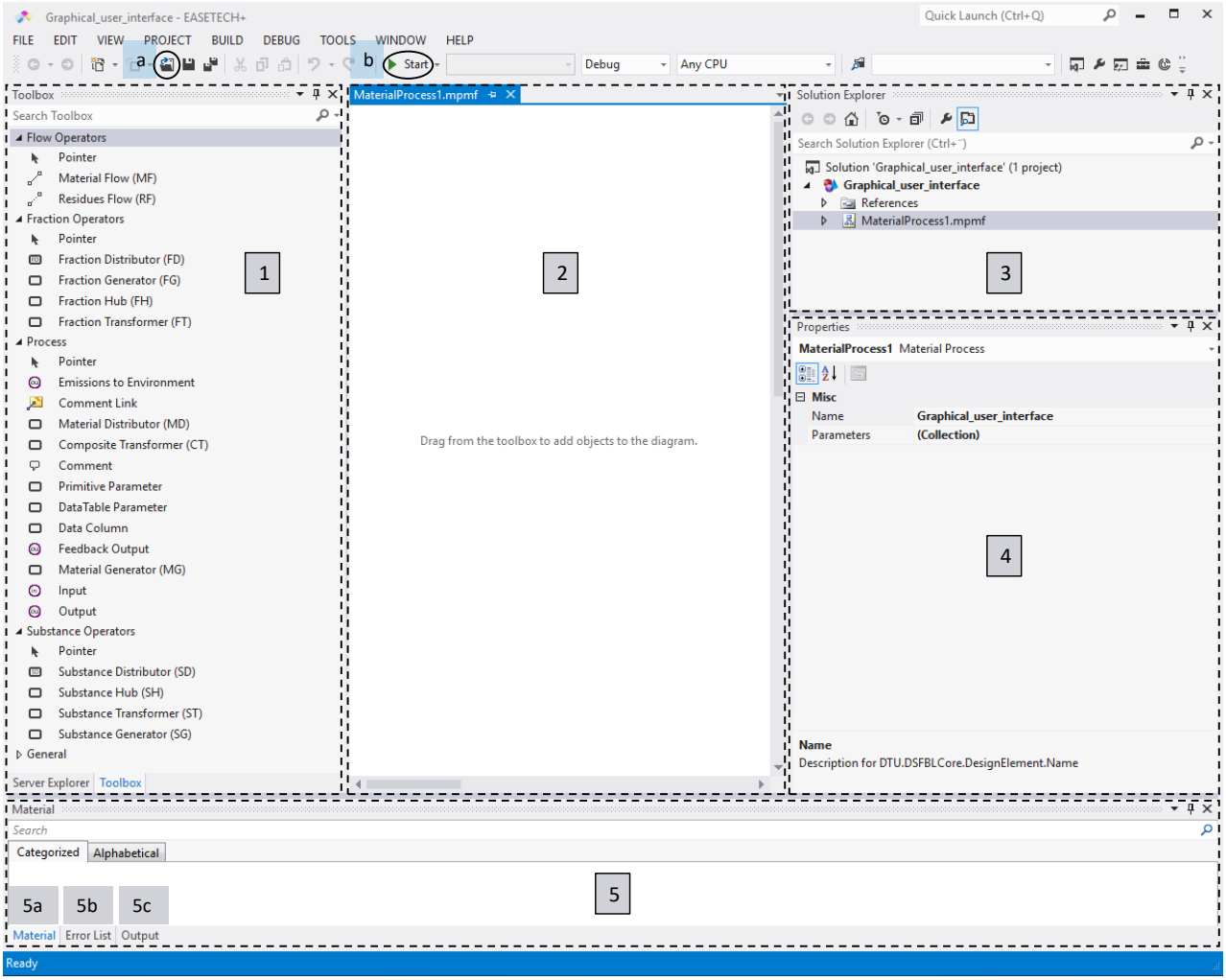


Figure 3: EASETECH+ user interface (1: *toolbox window*; 2: *main window* (process editor); 3: *solution explorer*; 4: *property window*; 5: *simulation results*; 5a: *material window*; 5b: *error list*; 5c: *output window*; a: *open file*; b: *start*)

Five main workspaces are identified: *toolbox window* (1, Figure 3), *main window* (process editor) (2, Figure 3), *solution explorer* (3, Figure 3), *property window* (4, Figure 3) and in *simulation results* (5, Figure 3): *material window*, *error list*, and *output window* (5a, 5b, and 5c respectively, Figure 3). In EASETECH+, operators are the elements defining the process flow diagram.

The *toolbox window* contains all operators, which can be dragged and dropped in the *process editor* to define the process flow diagram. Each operator has properties to be specified by the user. The *Solution explorer* presents the list of files belonging to the ongoing project in EASETECH+. Particularly, the *solution explorer* indicates the full name and path of the project, i.e. the folder where the project is located. In the *property window*, operators are defined and functions can be added to define input-output relationships. This contributes to the outputs of the process being modelled. The process flow diagram can be tested by clicking on "start", and subsequently "build" when the development is complete. By building the process, all calculation steps within the process flow diagram are verified; runtime errors are displayed, otherwise the process is ready to be exported to EASETECH and be applied in LCA scenarios.

2.5 Operators and functions

The principles of operator use were introduced in Lodato et al., (2020), while the operator functionalities were developed by Zarrin et al., (2015). To use operators, their properties need to be specified, such as *Amount* which represents the quantity available of a material, fraction or substance, either expressed through a value or a parameter, or expressed as the result of a calculation e.g. in *Amount*, it is possible to write an equation by using some expressions (i.e. symbol or combinations of symbols to define a input-output relationship). Another example is given by *Deg* (degradation) assigning an arithmetic expression as the degradation value. With material degradation some material is lost, e.g. when the amount of a given material is degraded with a certain ratio. In EASETECH+, not all operators allow degradation, iterations or conditions. A description and graphical representation of each operator together with its properties, and a key example for each operator is provided in Table A1, Appendix A. Four different operator categories are defined: *flow operators*, *fraction operators*, *process operators*, and *substance operators*.

- Flow operators allow the transition of a specific amount of materials, fractions or substances from a source component to a target component of the process flow diagram (from one operator to another). A transition can have conditions which enable or disable some process flows. Flow operators include:

- *Material flows* have specific amounts to transfer, generally set as percentages e.g. espressed through an equation. A source component can have one or more outgoing *material flows*. Conditions and iterations are allowed.
- *Residue flows* transfer only residues of materials, fractions or substances, i.e. remains left after other operations in a process. Only a single *residue flow* is allowed in a process. Conditions and iterations can be included.

- Fraction operators can be used only for fractions. A fraction is a component of a material. Fraction operators include:
  - *Fraction generators* generate fractions within a material. For example, if "garden waste" is defined as a material, all involved material fractions need to be generated such as "grass", "branches", "tree", "stone", "foreign objects".
  - *Fraction distributors* extract one or more fractions from a material, e.g. two *fraction distributors* are needed to extract "grass" and "branches" from "garden waste" for use in other process flows.
  - *Fraction hubs* collect several fractions into a material flow, e.g. "grass" and "branches" can be collected in a *fraction hub* after extraction from the material "garden waste".
  - *Fraction transformers* convert or "transform" partially or fully a fraction into another (existing or new). Transformations change the properties of a fraction. After transformation, the previous fraction does not exist anymore (while all properties such as mass and substances are transferred to the new fraction).
- *Substance operators* represent the properties of a fraction: biochemical, physical, nutritional and chemical. Four operators are provided for modelling substances: *Substance distributor*, s*ubstance hub*, *substance transformer*, and *substance generator*; all follow the same logic as described for fractions (e.g. distributors extract substances, hubs collect, etc.).
- *Process operators* refer to material, input(s) and output(s) and parameters:
  - *Material generators* generate a specific material (e.g. "garden waste"). The associated fractions need to be created via a *Fraction generator*.
  - *Material distributors* extract a material from several others.
  - *Input*s define process inputs which take the chemical, physical, nutritional and biochemical properties from the output of a previous process.
  - *Outputs* refer to a final output of a process containing all properties that have been generated, transformed and sent to the output during the process modelling.

- *Emissions to environment* represent a special final output that directly transforms a substance to emissions into specific environmental compartments, e.g. soil, air, water, etc., which represent the recipient being affected by the emission. The substance in the final output *emission to the environment* is removed from the material flow and cannot be involved in transitions or transformations.
- *Feedback outputs* allow creation of feedback loops within chains of processes after specifing a condition. In this case, the sequence of iterations associated with a loop stops once the condition is met. Therefore, while the condition is met, the *feedback output* works as a normal output because the generated material is not involved in a loop but proceeds as an *output*.
- *Primitive parameters* define parameters in a process.
- *Data table parameters* generate a table of parameters, which can be used when more than one parameter is associated to the same input. Each column of these tables are added through a *data column* operator.
- *Composite transformers* group several operators thereby providing a mechanism that allows transitions and transformations of a dedicated (to be defined) iteration.

Furthermore, it is possible to add comments through the *comment* operator, thereby supporting transparency and documentation. Comments can refer to specific component of the flow diagram through a *comment link*. An example of a *comment* and *comment link* can be found in Table A1, Appendix A: within the graphical representation of a *substance generator* "unit" a comment is added. Comments are visible in EASETECH+. Input-output relationships can be established through mathematical expressions by defining the properties of the operators enabled for calculations, such as *amount*, *iterator*, and *conditions*. This is possible through a set of functions (for further details, see Section A2.2, Appendix A).

2.6 Application of EASETECH+ modelling framework: Illustrative case study

For illustrative purposes, a case example of an anaerobic digestion (AD) process implemented into the library of EASETECH's material processes is included to demonstrate the application of EASETECH+. The intention is to show the application of almost all operators and functions of EASETECH+ within a single process template. EASETECH+ modelling framework applied to model the AD process in EASETECH+ followed these steps:

1. **Identifying the content of the process.**We quantified biogas and digestate produced based on the methane potential, which is the theoretical methane, applying Boyle's formula in case that nitrogen and sulfur is considered (Achinas and Euverink, 2016). The biogas reactor was a simple system of stirred reactor having: i) a constant temperature; ii) a perfect mixing; iii) an input-material of carbon, hydrogen, oxygen, nitrogen and sulphur (C, H, O, N, S) from the biochemical properties of the fractions to be digested anaerobically; iv) two outputs, biogas and digestate. The first, as methane, CH_4, and carbon dioxide, CO_2, while the second, digestate (liquid fraction), as non-degraded biochemical properties, water and inorganics from the input-material. Finally, the process was focus on the carbon conversion to the two final outputs.
2. **Establishing the intra-process transitions and transformations.** We defined mathematical relationships bethween substances of the input-material (please, see Section 3.1) to determine the two final outputs of the process. (Multi-)conditional-sequence flows are included to enable or disable specific flows within the process according to the type of data available.
3. **Validating the results.** We reproduced the process externally (e.g. in Excel) to verify that the mass/energy balance was respected.

3 Results and discussions

3.1 Process flow diagram: Anaerobic digestion model

The AD has the anaerobically degradable fraction of the feedstock converted into biogas, and the non-degradable fraction of the feedstock converted into digestate. In this respect, in EASETECH the feedstock in input (i.e. material fraction) generates two flows, one for the biogas and one for the digestate. In EASETECH+, the AD model follows different pathways according to the data available and the material transition and transformation due to the configuration of the process itself. Therefore, from the input-material we have case A (Figure 4), which directly considers C, H, O, N, S content as input, and case B (Section A3, Appendix A), which considers biochemical properties (e.g. cellulose, hemicellulose) as input. In EASETECH+, both cases are implemented in the same process; the calculation sequence enabled depends on which input data the user defines.

In the descrption of the AD process, we simplify the explanation of its modelling identifing three sub-areas: one for the AD input, "*input-model*" (Section S2, Supplementary material), one for

the gas production, "*gas-flow*" (Section S3, Supplementary material), and one for the digestate production, "*digestate-flow*" (Section S4, Supplementary material).

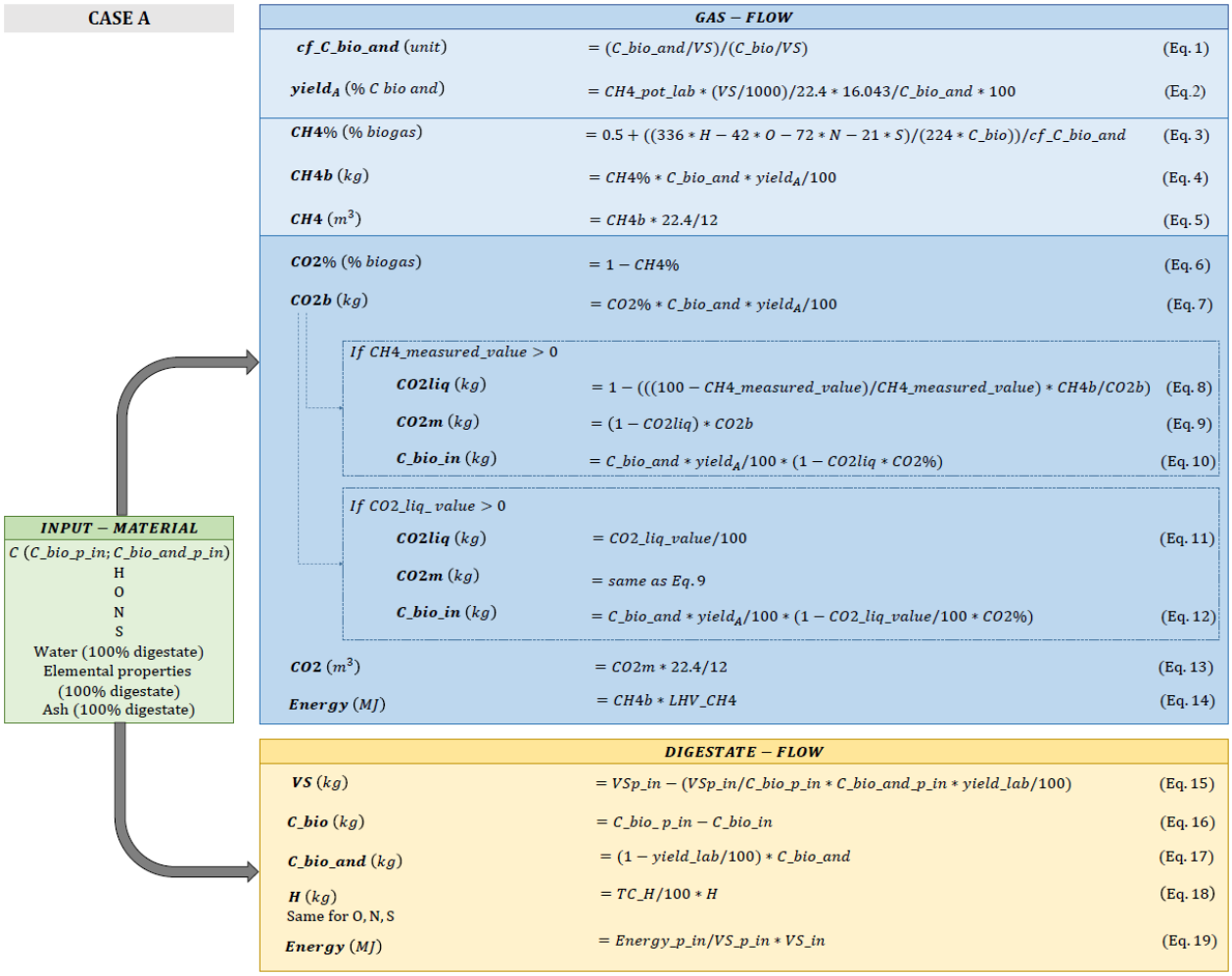


Figure 4: Anaerobic digestion – Case A. Process flow with 19 equations (units are espressed for each of them) from the input-material to the two main outputs, i.e. biogas and digestate. The gas-flow is for the biogas production and the digestate-flow is for the digestate production. Water, elemental properties and ash are totally transferred from the input-material to the digestate

In *case A*, the detemination of C, H, O, N, and S content of the input-material considers the organic fraction of the feedstock, both the degradable anaerobically and not. However, in the AD only the degradable fraction is converted into biogas. Thus, we define a conversion factor "*cf_C_bio_and*" (Eq.1, Figure 4) for the ratio between the degradable and the not degradable fraction of the feedstock. We need to determine methane and carbon dioxide anaerobically produced considering only the degradable fraction of the feedstock in input (input-material). We quantify methane *CH4%* (Eq.3, Figure 4) and carbon dioxide *CO2%* (Eq. 6, Figure 4) in percentage of biogas. Additionally, we need methane and carbon dioxide in m^3. Thus, we determine the methane and carbon

dioxide content firstly in kg, *CH4b* and *CO2b* (Eq. 4 and 7, respectively, Figure A3) and later in m^3, *CH4* and *CO2* (Eq. 5 and 13, respectively, Figure 4). This is done based on *CH4%* and *CO2%*, introduced previously, together with the methane obtained through measurements, here called "*CH4_pot_lab*" (in Nm^3/tVS, in Eq.2, Figure 4). The latter is an input of the process, as parameter used to adjust the ideal conditions of the gas produced to real one. In addition, for carbon dioxide, the theoretical biogas, *CO2b*, is partitioned between the gas phase defined as *CO2m* and the liquid phase as *CO2liq*, which goes to the digestate (Figure 5), both of them are mass-based. Thus, carbon dioxide in the gas (*CO2m*) needs to be determined considering the fraction remaining in the digestate (*CO2liq*). In Figure 5, we conventionally call "theoretical" both, *CH4b* and *CO2b*, and "measured" *CH4m* and *CO2m* when we determine their content considering carbon dioxide in the digestate (i.e. *CO2liq*). There are two possible ways of determining carbon dioxide in the biogas, *CO2m*, according to the measured data available:

- one considers *CO2_liq_value>0* (Figure 4), where *CO2_liq_value* is an input of the process, as parameter representing CO_2 content in the digestate as percentage of the theoretical CO_2 (*CO2b*);
- the other one considers *CH4_measured_value>0* (Figure 4), where *CH4_measured_value* is an input of the process, as parameter representing the methane measured as percentage of the biogas output (i.e. CH_4/CO_2 ratio).

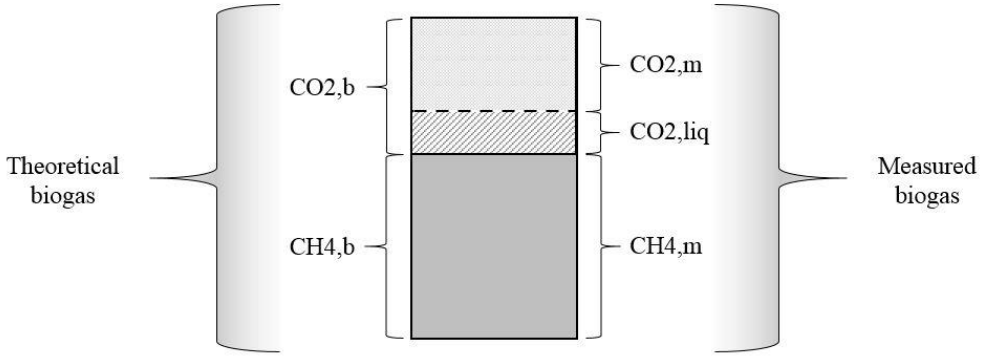


Figure 5: CH_4 and CO_2 share in the biogas. *CO2,b*: theoretical CO2 in the biogas; *CH4,b*: theoretical CH_4 in the biogas; *CO2,m*: CO_2 measured in the biogas (gas phase); *CO2,liq*: CO_2 measured in the liquid phase (digestate); *CH4,m*: CH_4 measured in the biogas (gas phase) (NOTE: *CH4m* is methane in m^3, which in Figure 3 is as *CH4*)

In *case B*, the biochemical properties of the input-material are the starting point of the AD process. We have a predefined list of biochemical properties. However, we give the possibility of

adding and considering in the AD process modelling new biochemical properties that are not in the predefined list in EASETECH. The new biochemical properties, here called "organic matter (OM)", together with the predefined ones, contribute to the determination of C, H, O, N, S, VS content (Eq. 20-23, respectively, Figure A1, Appendix A), and the methane yield for case B, $yield_B$ (Eq. 26, Figure A1, Appendix A). The latter, considers the yield of each biochemical property as *yield_CH4_pot* (Eq.24, Figure A1, Appendix A). Subsequently, in the gas-flow we calculate the biogas ratio (*CH4%* and *CO2%*, Eq. 27 and 30 respectively, Figure A1, Appendix A), the theoretical biogas (*CH4b* and *CO2b*, Eq. 28 and 31, respectively, Figure A1, Appendix A), and *CH4* and *CO2* in the gas phase (Eq. 29 and 36, respectively, Figure A1, Appendix A). Also for case B, we determine *Cbio* (here C_bio_in) in the gas, *CH4* and *CO2* after removing the fraction of carbon dioxide lost in the digestate by following the same procedure as case A (Eq.32-35, respectively, Figure A1, Appendix A).

For both, case A and case B, the final outputs of the gas-flow are *CO2*, *CH4*, *C_bio* and *energy*, while for the digestate-flow, volatile solids (*VS*), *C_bio*, *C_bio_and* (Eq. 15-17 respectively, Figure A1, Appendix A), *H*, *O*, *N* and *S* (Eq. 18, Figure A1, Appendix A), and the energy content (Eq. 19, Figure A1, Appendix A). In addition, the undegraded fraction of the input, such as water and inorganic matter (i.e. ash and elemental properties as Ca, Fe, Mg), remains in the digestate. This example was intended to illustrate the flows where the input-properties are involved (e.g. which flow is enabled or disabled due to the true condition), and their contribution to the final outputs (e.g. material transformation). In Appendix B, the process flow diagram of the AD case study is illustrated in detail.

EASETECH+ allows to add uncertainties and propagation to different types of parameters: process efficiencies, properties, but also all the elements of equations and consitions. We apply the AD process in EASETECH using an input, such as barley straw, from material generation process. In EASETECH, the "*E+ parameter*" window is enabled for processes modelled in EASETECH+. Here, it is possible to add a distribution (normal, triangular, log normal, uniform). We applied a normal distribution of ±10% to the parameter *CH4_measured_value* of the AD process, such as ND(63,10), where 63 is the default value. *CH4_measured_value* represents the methane concentration measured at the gas output. We considered case B of the AD process, where we have biochemical properties as input-material. In addition, we enabled the flow where *CH4_measured_value* is positive. Intra-process transitions and transformations that have the parameter *CH4_measured_value* are dependent on five final outputs: two of the gas-flow, *C_bio* and CO_2, and three of the digestate-flow, *VS*, *TS* and *ash* (Figure 6). Thus, results quantifing these five outputs have the standerd deviation associated due to the normal distribution of

CH4_measured_value. As result, models built in EASETECH+ can be: i) imported and integrated into the library of EASETECH's material processes; ii) used in LCA scenarios, and iii) data distribution is possible.

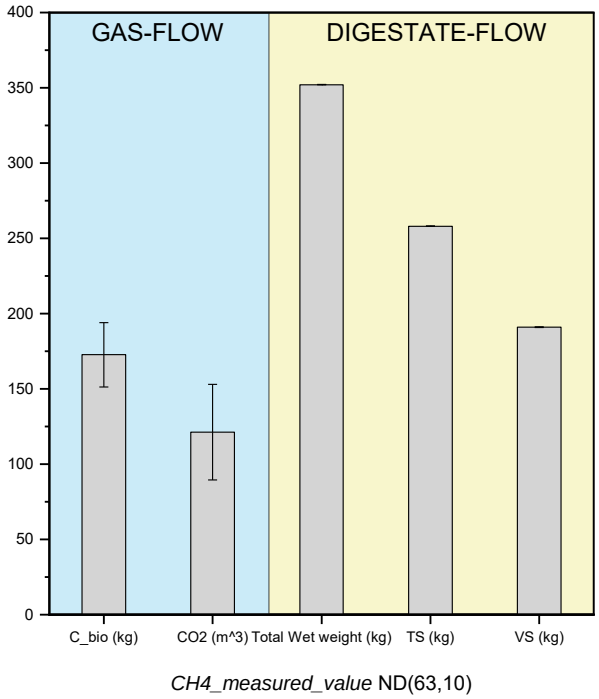


Figure 6: Five final outputs of the AD process are affected by the normal distribution to the parameter *CH4_measured_value*: *C_bio* and *CO2* in the gas-flow and *total wet weight, TS* and *VS* in the digestate-flow

3.2. Other examples of process flow diagram applications

Depending on the modelling scope and perspective (e.g. technology vs. system level), the processes in EASETECH+ are representations and to some extent approximations, i.e. "models", of the actual processes occurring within the system. In case of chemical reactions within processes, models can be stoichiometric or based on other physico-chemical properties. Consequently, process can work without specific experimental restrictions involving theoretical yields (through input-output relationships) and – once imported into and distributed via EASETECH – can be applied by all users. The following offers a few illustrative examples of potential application of the EASETECH+ modelling framework for a range of technology types for which the outputs are depending on the input-material properties:

- **Precipitation of salts**, such as the estimation of salts being produced during a simple step-wise precipitation of salts from a brine solution. The amounts and types of salts

that precipitate vary depending on the dissolved species (i.e. cations and anions) and on the solubility product of salts. The amounts of specific salts being precipitated (e.g. a very soluble salt and a less soluble salt) can be estimated based on molar calculations, provided that there is sufficient knowledge on the process and composition of the brine. In other words, EASETECH+ can be used to make substance flow analyses, calculating the amounts of dissolved species at each step and evaluating whether or to what extent the following salt can be produced.

- **Pyrolysis and gasification** processes, which provides a thermal decomposition of dry feedstock in specific conditions (e.g. temperature, pressure). The input-output relationships reproduce reactions that quantify the intermediate and final outputs. The final outputs of pyrolysis are: condensable vapours, noncondensable (permanent) gases, and solid char; while the outputs of gasification are: syngas, char, and tars. An example is a fluidized bed gasification process which converts the input material (waste and biomass fuels) into syngas (Ardolino et al., 2018); it quantifes through input-output relationships: the volumetric flow rate of the syngas, its mass flow rate, composition, energy content, together with emissions to air, consumptions of chemicals, and residues produced.
- Modelling of **biogas upgrading**, therefore, depends on the composition of the input materials and process conditions, which can be represented by input-output relationships. The upgrading of biogas increases the methane content and converts the biogas to a higher fuel with specificities similar to natural gas (Angelidaki et al., 2018). An example is the biological biogas upgrading via hydrogenotrophic methanogens that convert carbon dioxide to methane by using hydrogen. This upgrading is within the methanogeniesis, final sub-process of the anaerobic digestion.
- **Biorefinery**, which coverts the lignocellulosic input material into bioethanol, a solid and a liquid fraction through a sequence of sub-processes (i.e. pretreatment, hydrolysis, fermentation and distillation, recovery) (e.g. Lodato et al., 2020). This example considers the production of C5 and C6 sugars during the hydrolysis followed by the production of ethanol and CO_2 during the fermentation and distillation.

3.3 EASETECH+: Implications for future modelling

In EASETECH+, LCA practitioners can model in a flexible way their own process by using the provided modelling framework. Flexible due to the freedom of modelling offered by EASETECH+, which does not limit the LCA practitioner to specific process configurations but allow implementation of new configurations with the use of the provided operators. EASETECH+ has operators allow the modelling of materials, fractions and substances, their transitions and transformations within a process in an advanced and user-friendly way. The graphical user interface of EASETECH+ allows definition and visualization of the process flow diagram by the diagram representing the process. We can verify both the mass/substance/energy balance and the intermediate outputs while we are modelling. We can check the modelling structure by building the processes and verify that there are no errors. We encurage checking and validating the process during the modelling stage before the final output(s) to facilitate the identification of errors: errors may occur due to incorrect selection of formulas or due to the way of using operators or writing functions. An error list within EASETECH+ assists the identification of such errors. We can simulate processes directly in EASETECH+ after modelling and validation. When we run a process, we run the sequence of operators and functions, and we determine the output(s) of the process, which represent its results according to the flows enabled or disabled due to the data available. Processes modelled in EASETECH+ can be parametrized using raw data and formulas which represent input-output relationships. In agreement with Cooper et al., (2012), parametrization has several advantages such as employing an increased types of computational methods due to the different types of parameters (e.g. efficiencies, conversion factors). In addition, we use a standard way of modelling (e.g. through functions) which enables more consistency and transparency across projects and between users. However, in EASETECH+, we can model materials, fractions and substances existing in EASETECH's library thereby ensuring backwards consistency of nomenclature, or allow new nomenclature and thereby more flexibility looking forward. This is an important requirement defining clarity, consistency and extensibility as explained in Edlen et al., (2018), here also important to maintain the link with existing processes in EASETECH (as was indicated in Figure 3, Section 2.3).

EASETECH+ provides substance conversions with substance balancing. For example, material properties, such as water, VS, calcium, lipids, follow different process flows and the obtained products are determined based on the input properties involved in the conversion process. Consequently, an input property changes after the conversion based on the process performance, its configuration and the available amount in input. Thus, the final outputs are functions of the input(s)

and the material/substance conversions/transitions within the process, producing a dynamic material/substance-flow-based balance.

Linear and non-linear relationships are allowed through input-output relationships, by means of functions used (e.g. for linear, multiplication with a or more parameters, and for non-linear, exponential, logaritm). In addition, (multi-)conditional-sequence flows and iterations providing loops are allowed. In accordance with Unger et al., (2009), these relationships are necessary for modelling of specific transformation processes. Another important feature provided by EASETECH+ is the possibility of having material degradation, i.e "loss" of materials, e.g. when the amount of a given material is degraded by a certain fraction.

All these features imply to define life cycle inventories of processes modelled in EASETECH+:

- based on high quality data (e.g. modelling of a pyrolysis process based on an existing plant)
- considering specific configurations and operating conditions (important during the identification of input-output relationships)
- considering specific parameters (LCA practitioners can change it according to their case and use the process template in their studies)
- linking input-output flows (e.g. the dynamic material/substance-flow-based balance)
- handling numerical uncertainty and propagation
- handling different input-materials (i.e. different compositions)
- producing high quality results based on the modelling
- applying the same process template in different LCA scenarios (usable from more LCA practitioners, experts and not of the process, and with the intentions of considering in their LCA the process template implemented into the process library of EASETECH)
- evaluating the contribution of parameters and data used to LCA results (important for the interpretation of the results)
- allowing substance recirculation within the modelled system (e.g. through loop and iterations)

3.4 EASETECH+: Model transparency, reproducibility and expandability

When we model a process in EASETECH+, we need to ensure clarity and transparency, e.g. by ensuring that operators are arranged and connected to each other in a logical and sequential way,

allowing the "trace-back" of results and facilitate identification of errors. The process flow diagram should be thoroughly documented with respect to a specific case-study; an example is provided in Supplementary material. The documentation should ensure transparency, provide understanding of involved processes, material flows, conditions etc. and allow reproducibility of the work. This supports a collaborative environment among LCA practitioners in modelling new processes with a wide variety of details and at different process levels. Further, this offers the possibility of updating and improving previous processes or developing new ones having different configurations and features. This is in agreement with Cooper et al., (2012), Kuczenski et al., (2018), Unger et al., (2004), and ISO standards.

5 Conclusion

The concept of process-oriented life cycle assessment (LCA) modelling of resource flows was implemented into the EASETECH+ modelling framework, thereby providing the ability to model complex processes and technologies involving mathematical relationships associated with material flow properties. EASETECH+ allows a variety of modelling scopes, e.g. at process and technology level or at system level. New process templates built in EASETECH+ can be imported and distributed via the already established LCA model EASETECH, where process models can be adapted to new case-studies simply by changing parameter values. A variety of operators are provided in EASETECH+, e.g. iterations, loops and (multi-)conditional-sequence flows are allowed. Based on EASETECH+, LCA studies focusing on resource technologies and system cannot be carried out at an entirely new level: i) material properties such as substances of input material(s) can be involved in transformation processes defined through linear and/or non-linear relationships, providing material conversions while maintaining balancing at mass, energy and (other) substance levels. The EASETECH+ modelling framework was demonstrated through an illustrative case-study, focusing on anaerobic digestion. EASETECH+ has a potential to improve the applicability of new process models within resource systems, increase transparency across studies through adjustment of process parameters rather than establishment of entirely new models, and to easily share process templates between EASETECH users.

Appendix A: Details of i) how to import a process flow diagram into EASETECH; ii) operators and functions of EASETECH+ modelling framework; iii) Input-output relationships of the case B.

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IV

Framework for assessing environmental performance of methane gas supply in the context of local bioeconomy

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Draft manuscript

Framework for assessing environmental performance of methane gas supply in the context of local bioeconomy

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Abstract

Biomass as source of non-fossil carbon is seen as potential for CH_4 production, in the endeavor of contributing to limiting global warming to 1.5 °C above pre-industrial levels and achieving zero CO_2 emissions by 2050, targets of the European Green Deal. In Europe there is an untapped potential to use residual bioresources for CH_4 production, which requires a system perspective and a regional context. We developed a stepwise framework with import/export strategies for bio-based methane production from residual bioresources to supply the gas demand of a region considering anaerobic digestion and gasification, both with upgrading of CH_4. The environmental performance accounted for technology conversion-pathways, the current management uses of the residual bioresource and their allocation to different technologies. The application of the framework on Occitania, an administrative region in South West of France, resulted to support exporting strategies by meeting and exceeding the annual gas demand. The combination of biological and thermochemical degradation of dry, semi-dry, and wet residual bioresources allowed to use all the hydrocarbon available. The reproducibility of this framework on any other region provides support of practical actions toward local bioeconomy and climate goals.

Introduction

In Europe, the recent Green Deal[1] promotes several instruments, including a Climate Law[2], towards achieving the Paris Agreement goal of limiting global mean surface temperature increases to well below 2°C above pre-industrial levels. This involves sharp cuts in the use of fossil fuel carbon dioxide (CO_2)

emissions, these being the main cause of human-induced climate change, counting with ca. 69% of global greenhouse gas (GHG) emissions[3]. Overall, global greenhouse gas emissions are due to 5 key sectors of activity, namely: Energy (73%), Agriculture (12%), Industrial processes (6%), Land Use, Land Use change & Forestry (6%) and Waste management (3.2%).[3]

In this ambition, revisiting the role of natural gas for heating, cooling, for electricity supply or as a feedstock for the chemical industry may play a significant role. Natural gas is the main energy source used in Europe in different sectors (e.g. transformation, final consumption) and any changes in gas demand as a result of Green Deal policies produce an important impact not only at a national level, but also at a regional level.[1] France is one of the European countries having a high natural gas demand. For example, in 2015, the share of natural gas demanded for heating was above 70%.[4]

Bio-based methane is an accessible solution for the decarbonization of the natural gas. Bio-based methane can be produced from an anaerobic digestion (AD) or a gasification (GA) plant, both with an upgrading unit to meet the conditions of methane injection into the natural gas grid.[5] In the short term, France is structuring the stimulation of a demand for renewable gas around "Guarantees of Origin", which certifies the renewable nature of bio-based methane.[6] In medium and long terms, French challenges on creating conditions for: i) exploiting and including bioresources that are located significantly distant from the gas grid in the methane conversion process; ii) increasing the network injection capacity (e.g. backhauling, storage unit on the distribution network); iii) producing cost reduction strategies, such as increasing the carbon tax with a target of 100 €/t CO_2 by 2030, which would make bio-based methane competitive with natural gas (e.g. with 50-60 €/t CO_2 by 2030).[6] In addition, the goal of 10% of renewable gas consumption by 2030 was set by limiting the global warming to 2 ˚C by 2050. However, with the new objective of 1.5 ˚C set under the Paris agreement, an acceleration of the renewable gas development was considered with a realistic target of 60 TWh by 2028.[6] The main purpose of renewable gas production is to reduce or exclude fossil gas imports, while valorizing local bioresources and increasing local energy production in the context of a local bioeconomy. On this purpose, Occitania (72,700 km^2, 5.8 M inhabitants) is an administrative region in South West of France that wants to become the first European region at "positive energy" (REPOS scenario), meaning that 100% of the energy consumption is from local production of renewable energy.[7] Currently, 41% of the regional electricity production is from renewable energy. The annual gas consumption is 17.5 TWh on a total energy consumption of 128 TWh.[8] In addition, 2/3 of the greenhouse gases is from CO_2, with 21.5 Mt CO_2 from energy.[8] However, to promote a local bioeconomy an analysis of the bioresources available on the territory is necessary in terms of quantity, quality (chemical/physical/nutritional properties) and accessibility. The region is rich in bioresources: it is the first French region for viticulture (it contributes to 33% of the production of French wines); the second region for fruit production and for agriculture, the first region for sheep, and the forth for vegetables.[7] Thus, different bioresources with different physico-chemical properties, which define the technology to be used for the methane production, e.g. AD or GA.

The production of bio-based methane from thermochemical and biological processes was addressed in several studies (e.g. [9, 10]). Previous studies have questioned whether bio-based methane gas produced from different technologies provides environmental benefits. Dong et al., [11] evaluated the environmental performance of the production of bioenergy from four commercial operation plants, such as pyrolysis, gasification, gasification-melting, and modern incineration, substituting European mix of heat and electricity. It did not account for biological conversion processes to biomethane. Tagliaferri et al., [12], carried out an LCA to support decision on the best technology (from both thermochemical and biological processes) to produce a given amount of methane, and the best waste management option of a given amount of waste-input. Hahn et al., [13], included in the environmental analysis a demand-oriented biogas supply for flexible power generation together with the comparison with primary energy supply and GHG balance. Ardolino et al., [14] analyzed and compared the performance of producing biomethane from both AD, GA, and upgrading to use bio-based methane in road transport. While the above examples are valuable for assessing the environmental performance of different technologies for bio-based methane production for different scopes, research gap remains when decarbonization strategies need to be adopted based on local available resources. There is a need for tailor-made solutions to identify technologies and systems of technologies that can produce renewable energy exploiting the potential of residual bioresources available on a region. With this study, we will investigate h*ow to supply the demand for methane gas of a region in an environmentally-efficient way and uncover what this environmental performance depends on*. In this endeavor, this study aims to provide a reproducible stepwise framework integrated with the process-oriented modelling approach for regional use of bioresources to supply bio-based methane.

Method

The stepwise framework proposed herein is largely based on the Life Cycle Assessment methodology, as described in ISO 14040 and ISO 14044. [15, 16] It is here applied to a specific case-study region, but is designed to be applied to any areas, with all calculations transparently documented in the ESI. This stepwise procedure for consistent evaluation of both technology and system perspectives has six steps. Each step as applied to a case study for illustrative purposes and described in more details in the following sections.

Step 1: LCA methodology

The functional unit, here the service to be supplied in all scenarios, is "*to fulfill the annual demand for methane gas in the French Occitania Region, to the extent possible with local residual biomass resources or else with imported natural gas*". Residual bioresources are here 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. The temporal scope of the study is 2015-2050, while the geographical scope is the Occitania region.

A consequential approach was applied in the study.[17, 18, 19, 20] Consequently, multi-functionality was handled by means of system expansion, i.e. we considered in which market and for which function the generated co-products were used and accounted for eventual displacement and substitution mechanisms occurring as other suppliers were affected. This was made with the assumption that in a long-term perspective, markets are unconstrained and fully elastic, where an increase in demand translates into an equivalent increase in supply (1:1 substitution).[21] Moreover, only the marginal suppliers were considered, i.e. those reacting to a change in demand.

Marginal energy mixes were calculated based on variations in annual supply derived from European and French forecasts to 2050 (details in Electronic Supplementary Information, ESI): accordingly, the marginal electricity is composed of 36% solar, 52% wind, 5% hydro, 6% biomass, while the marginal heat supply is composed of 1% geothermal, 91% heat pumps, 1% solar thermal. [22, 23, 24]

The LCA modelling of the conversion-pathways followed a process-oriented approach: mass and energy balance were established through parametrized mathematical relationships for the conversion of input bioresource properties to output products within technologies and the entire system. For example, the carbon content of each category of bioresources, quantified based on biochemical properties (e.g. sucrose, cellulose), produced bio-based methane and co-products (and rejects) respecting the technology configuration and reaction's stoichiometry of different conversion-pathways. Additionally, we addressed the environmental consequences of changing the conventional management routes of residual biomasses, herein defined as “counterfactual”, to alternative methane conversion pathways for bio-based methane production and utilization. The environmental performance of each conversion pathway, for each residue input, was compared with the impacts of conventional natural gas production (reference product, RP). To be able to compare the impacts of the proposed methane-conversion pathway (i.e. by using residual biomasses) with the impact of conventional natural gas production, the net balance in terms of induced and avoided emissions was calculated according to [25]. The net balance is the result of producing the service under assessment while avoiding the alternative management routes, (i.e. pathway minus counterfactual).

The life cycle impact assessment was performed with the ReCiPe2016 method[26], considering all the mid-point impact categories it encompasses.

The modeling was facilitated with the EASETECH software[27] using background data from Ecoinvent 3.6, [28]. Finally, we parametrized the model giving an uncertainty distribution to each parameter. In this way, we were able to propagate the uncertainty with a Monte Carlo analysis with 1000 iterations. The contribution of the uncertainty of each parameter to the overall uncertainty was assessed with analytical propagation.[29] (*For the draft’s version of the thesis this aspect was not included, however it will be accounted before submission to a selected journal*).

Step 2: Estimation of bioresource potential

Residual bioresources available in Occitania were identified based on regional inventories[30, 31] commissioned in 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 and quantified 41 residual bioresources, which we categorized within 10 categories (in ESI). These bioresources are generated from agro-industrial processes, urban and commercial activities, household consumption, as well as primary forestry and agricultural activities. Out of the 10 categories, three represents more than 80% of the total wet weight, namely intercrop (39 %), crop residues (33 %) and manure (14 %). The other categories include wood waste (3.48 %), household biowaste (3.06 %), agri-food residues (2.66 %), sludge (2.66 %), green waste (1.31 %), pruning residues (0.61 %), and forestry residues (0.15 %). All 41 streams were integrated within a residual biomass database reporting the available quantities of each stream, along with their detailed characterization in terms of physical, chemical, biochemical, and nutritional composition and properties (ESI).

Step 3: Selection of technologies and general modelling assumptions

Two technology conversion pathways were considered: a biological conversion, i.e. anaerobic digestion (AD) producing biogas, and a thermochemical conversion, i.e. gasification (GA) producing syngas. Thermochemical degradation is suitable for dry or semi-dry feedstocks, e.g. with less than 40% of moisture content (e.g. [32]). On the other hand, biological degradation is preferred for resources having a high sugar, lipids, and proteins content, and is better suited to handle feedstock with a moisture content higher than 70%.[33] These values are here considered as threshold for allocating the streams towards one or the other technology. Therefore, AD and GA can complement each other by distinctively using both wet and dry biomasses in the endeavor to reach the desired levels of bio-based methane production.

The biogas and syngas produced are mixtures of CH_4 and other gases that must be upgraded to meet the requirements for injection into the natural gas grid, which requires a methane content of 98.3 %vol,[5] (details in the ESI). Several upgrading technologies exist for biogas, as thoroughly described in [34]. Biogas is essentially a mixture of methane and carbon dioxide (CO_2), along with trace gases, and its overall composition depends upon the streams being digested.[35] Here, two types of upgrading processes are considered for the biogas, namely a simple removal of the carbon dioxide (CO_2) based on conventional physico-chemical absorption methods, and ex-situ hydrogen enhancement, where the CO_2 portion of the gas is converted into CH_4 following the Sabatier reaction (i.e. $CO_2 + 4H_2 = CH_4 + H_2O$).[34] For syngas, which is essentially a mixture of hydrogen along with C-gases (CO, CH4, CO2), only the latter upgrading is considered. When bio-based methane is afterwards injected into the natural gas grid,

it must have the same pressure as the target network it is connected to (i.e. connection point). In this specific case study, we consider a distribution network at 40 bars. A CH4 slip of 1% was considered during the injection step.[28]

Methane conversion-pathway: Anaerobic digestion

Anaerobic digestion converts the biodegradable fraction, such as lipids, proteins and carbohydrates of bioresources into biogas and digestate, i.e. a liquid residue that contains what has not been degraded anaerobically. The first upgrading process considered, namely water scrubbing, is an absorption process exploiting the CO_2 solubility in water, being amongst the most commonly used upgrading technologies.[34] Inventory data from an industrial plant were used, as further detailed in the ESI. The resulting output of the process is two gaseous streams: CH4 representing 99.5% of the initial biogas input to the scrubber and CO2 representing 0.55 %, here considered to be emitted to the atmosphere. The pressure considered for the output methane was from 4-7 bar, with an electricity of 0.062 kWh Nm^{-3} of methane.[5]

The second upgrading process included, namely hydrogen enhancement, considers the production of hydrogen through water electrolysis, powered by the marginal electricity. There are several electrolyzers technologies and important ongoing research development to produced so-called green hydrogen ([1], hydrogen strategy), but here conventional alkaline electrolyzers were considered[36], being the most commercially available electrolysis technology for hydrogen production to date[37]. This hydrogen is then injected, along with raw biogas, into an ex-situ unit where it can, in the presence of a catalyst, react with the carbon dioxide in the biogas to produce additional methane. The upgrading occurs at temperatures between 300 and 400 ºC and in presence of a nickel-based catalyst. We used 93 mg of a nickel-based catalyst with 19 % nickel and 81 % of aluminum alloy to produce one Nm^3 of CH_4.[38] The ex-situ upgrading was preferred over an in-situ H_2 injection because it secures stability of the methane yield by not affecting the microbial community of the digester, such as hydrogenotrophic methanogens.[39]

The anaerobic digestion process was modelled as a continuous stirrer tank reactor and temperature (thermophilic). Biogas was quantified based on the degradation of carbon of lipids, proteins, and polysaccharides, such as cellulose, sucrose, starch, hemicellulose, of residual bioresources in input. A methane slip of 2% of the total CH_4 in the biogas was considered.

Methane conversion-pathway: Gasification

Biomass gasification converts mainly dry and semi-dry input materials (< 20 - 30 % TS) into syngas (i.e. H_2, CO, CO_2, CH_4, N_2, H_2O plus impurities), char and tar. We selected a fluidizing bed gasifier with a cleaning and conditioning system, and a methanation unit with carbon-to-methane (C-to-CH4) upgrading, aiming to convert all carbon to methane. We considered a gasifier temperature of 850 ˚C and a pressure of 1 bar, with air as a fluidizing agent and an equivalence (or air-fuel) ratio of 0.33. These

conditions were selected as favorable for improving the methane concentration in the syngas. The modeling of the feedstock and gaseous flows in and out the gasifier was based, to the extent possible, on data from the pilot-scale GoBiGas plant, being, at the time of writing, the only running unit where syngas-to-methane is produced at scale. Ancillary materials and other inputs were based on published literature.[32, 40, 41] The initial process activation is considered to be carried out by adding calcine and potassium while heating the system through the combustion of natural gas (details in ESI). During the stop and initial start phase, 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 include supplemented ash components, such as potassium, sulfur, and calcium. Considering these process conditions, we estimated, for each of the five residual bioresources suitable for gasification, the output syngas composition through an equilibrium model that we implemented within EASETECH, on the basis of the equilibrium equations presented in[42]. In a nutshell, our stoichiometric model considered three principles:

i) A global gasification reaction (Eq. 1)

$$CH_xO_yN_z + wH_2O + m(O_2 + 3.76N_2) \rightarrow n_1H_2 + n_2CO + n_3H_2O + n_4CO_2 + n_5CH_4 + n_6N_2 \quad \text{(Eq. 1)}$$

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

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

$$C: n_2 + n_4 + n_5 = 1 \quad \text{(Eq. 2)}$$

$$H: 2n_1 + 2n_3 + 4n_5 = x + 2w \quad \text{(Eq. 3)}$$

$$O: n_2 + n_3 + 2n_4 = y + w + 2m \quad \text{(Eq. 4)}$$

$$N: 2n_6 = z + 7.52m \quad \text{(Eq. 5)}$$

iii) Two independent equilibrium reactions (Eq. 6, 7) and the two-kinetics associated.

$$C + 2H_2 \leftrightarrow CH_4 - 74.8\ MJ/kmol \quad \text{(Hydrogasification)} \quad \text{(Eq. 6)}$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 - 41.2\ MJ/kmol \quad \text{(Shift reaction)} \quad \text{(Eq. 7)}$$

Details regarding the gasification model can be found in the ESI. A schematic representation of the stoichiometric equilibrium model of the methane conversion-pathway for the gasification is given in Fig. 1, for each of the residual biomass categories undergoing gasification, such as crops residues, forest residues, green waste, pruning residues, and wood waste. The GA model provided on average, from 1 kg of bioresources, 2.4 Nm^3 of syngas, which is consistent with published experimental data (e.g. [43]).

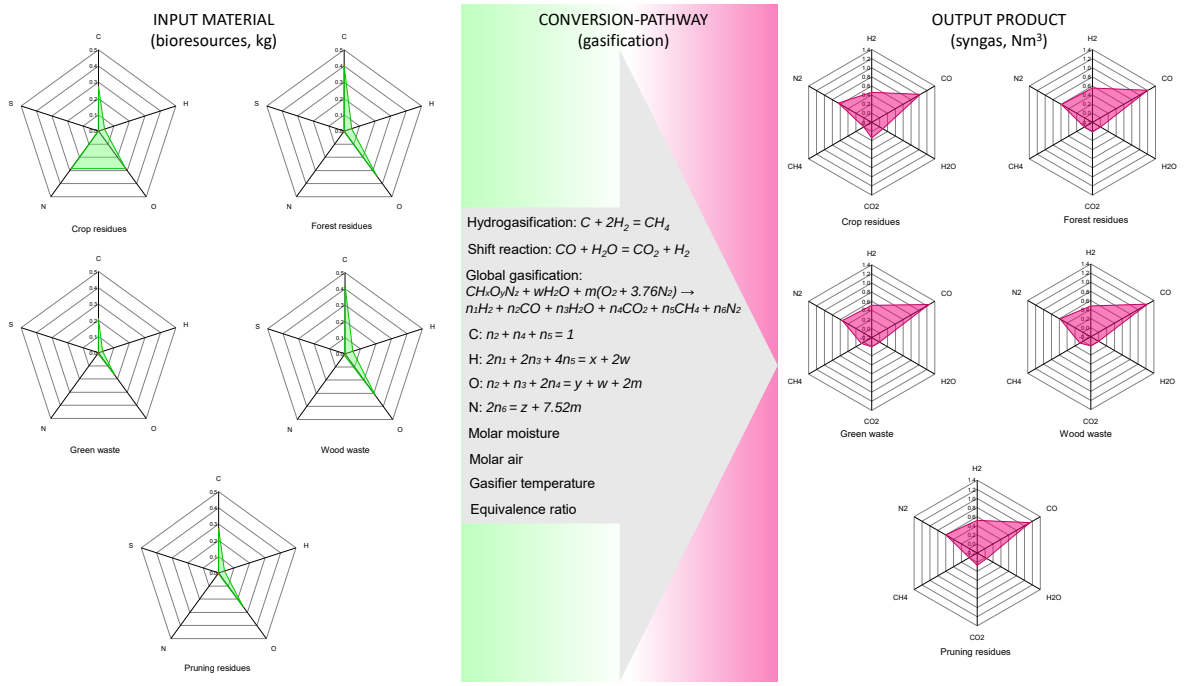

Fig. 1: Schematic representation of the stoichiometric equilibrium model applied to six categories of bioresources as input material expressed as C, H, O, N, S considering 1 kg input each, and corresponding syngas output products as H_2, CO, H_2O, CO_2, CH_4, and N_2, in Nm^3

We considered hydrogenation reactions (respectively, methanation and Sabatier) to convert carbon monoxide and carbon dioxide to methane (Eq. 8, 9). According to [44], a CO_2 conversion of almost 98% is needed to achieve a methane content higher than 90% in the upgraded gas output, and a CO conversion of 99% to achieve a methane content of 95%. Thus, we set a conversion of 99% for both CO and CO_2 in the methanation (C-to-CH4 upgrading) model used in this study. The catalyst considered for the CO_2 methanation is a nickel-based catalyst, as in the AD upgrading. Catalytic methanation reactors have a range of operating temperature between 200 and 550 ºC with a pressure range between 1 and 100 bar.[44] We considered an operating temperature of 350 ºC and a pressure of 1 bar. More details can be found in ESI.

The methanation section, modelled on the basis of the GoBiGas plant consists of a series of 10 processes: i) hydration of olefins and carbonyl sulfide (COS); ii) H_2S removal; iii) removal of trace component through a guard bed; iv) water-gas shift reaction; v) pre-methanation; vi) CO_2 removal; vii) four-stage methanation; viii) drying; ix) compression before feeding the natural gas grid.

Step 4: Process-oriented inventory modelling and LCA system boundary

Two technology conversion pathways were considered: a biological conversion, i.e. anaerobic digestion (AD) producing biogas, and a thermochemical conversion, i.e. gasification (GA) producing syngas. Thermochemical degradation is suitable for dry or semi-dry feedstocks, e.g. with less than 40% of moisture content (e.g. [32]). On the other hand, biological degradation is preferred for resources having a high sugar, lipids and proteins content, and is better suited to handle feedstock with a moisture content higher than 70%.[33] These values are here considered as threshold for allocating the streams towards one or the other technology. Therefore, AD and GA can complement each other by distinctively using both wet and dry biomasses in the endeavor to reach the desired levels of bio-based methane production.

The methane conversion-pathways are: i) *ADH*, anaerobic digestion and its upgrading with hydrogen (i.e. alkaline electrolyzer); ii) *AD*, anaerobic digestion and its upgrading without hydrogen (i.e. water scrubbing); and iii) *GA*, gasification with C-to-CH4 upgrading. Fig. 2 shows the system boundaries of the studied technologies (i.e. methane conversion-pathways), the distribution to the possible conversion-pathway per type of residue inputs and the counterfactual (in dotted lines).

The 10 categories can follow a specific methane production route, one for the anaerobic digestion (AD and ADH pathway) and one for the gasification (GA pathway) (Figure XXXX). Bioresources that can be both digested and gasified are: crop residues, green waste, intercrop, and biowaste; the one that can be only digested are: manure and slurry, agrofood residues, and sludge; those that can be only gasified are: pruning residues, forest residues, wood waste and industrial end-of-life (Fig. 2, in blue). Finally, the bio-based methane produced from each conversion-pathway was compared to the natural gas demand of Occitania, equaling 17.5 TWh per year (34).

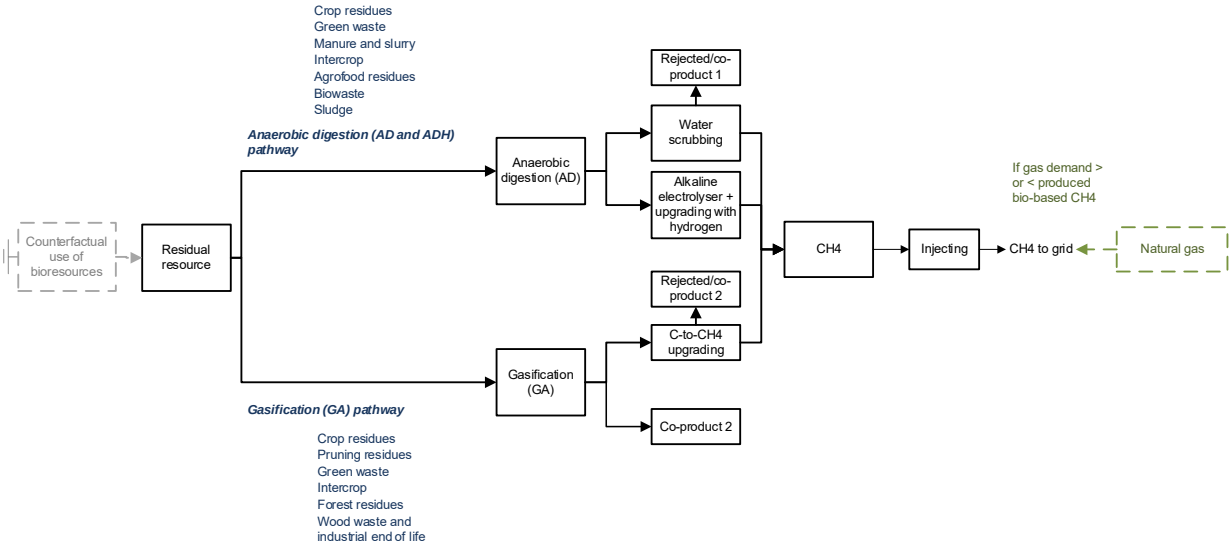

Fig. 2: Process flow diagram of residual resource conversion to bio-based methane (CH_4) in a general overview. In grey, residual resources identified with associated counterfactuals (conventional treatment or alternative route); in blue, selection of technologies to the two methane conversion routes; in black, conversion pathway and injection of methane to the natural gas grid; in green, comparison with the regional gas demand

Step 5: Evaluation of technology performance

Step 5 provides the environmental performance of each bioresource (independently) relative to the production of 1 Nm^3 CH_4 including injection into the gas grid and local distribution, here referred to as layer 1 results. We calculated the net balance (NB), i.e. emissions minus savings for each scenario, in order to be able to compare the impacts of bio-based methane production with the reference product (fossil natural gas). Finally, residual bioresource conversions having an impact higher than the natural gas were identified and excluded from further interpretation. Therefore, NB is useful for a preliminary evaluation of scenarios' performance allowing the exclusion of some pathways before analyzing their contributions to the net impacts.

Step 6: Evaluation of system-level scenarios

The system-level scenarios are selected combinations of bioresources and conversion technologies for fulfilling the functional unit and uncovering environmentally-performant regional possibilities to fully supply 17.5 Twh y^{-1} of bio-based CH_4, I.e. the region annual gas demand. Thereby the scenarios presented herein are not an exhaustive list nor an optimization of all possible options, but handpicked ones as judged relevant for the region. We identified four cases: i) combined *ADH-AD-GA scenario*, with the selection of the stream categories and technology producing a maximum of CH_4 while emitting a minimum of CO_{2_eq}; ii) *AD1 scenario*, as in (i), but considering only anaerobic digestion without hydrogen upgrading; iii) *AD2 scenario*, anaerobic digestion without hydrogen upgrading as the only conversion technology; iv) *ADH scenario*, anaerobic digestion with hydrogen upgrading as the only involved technology.

Results and discussion

AD vs GA for methane production

Gasification required the least amount of feedstock per unit of gas. It indeed provides a maximum of C-to-CH4 conversion, also in line with recent studies (e.g. [14, 45]), while with anaerobic digestion a portion of C remains undegraded in the digestate. The digestate however may be gasified, if it is rich in fiber, boosting the total CH_4 recovery. However, the biological degradation was fed with higher amounts of bioresources having a low organic matter content, such as intercrop, sludge, agrofood residues, manure. Based on the biochemical properties and the conversion process, the amount of kilograms input for each category of residual bioresources was different (Tab. 1).

Tab. 1: Amount of feedstock required to produce 1 Nm^3 of CH_4 for each combination of biomass category and technology pathway.[1]

Category	kg input to produce 1 Nm^3 of CH_4		
	ADH	AD	GA
Crop residues	5.4	5.9	1.6
Pruning residues	-	-	1.5
Green waste	8.5	9.5	1.4
Manure	15.0	17.5	-
Intercrop	35.0	46.3	-
Forest residues	-	-	1.4
Wood waste/industrial and end of life	-	-	1.4
Agrofood residues	16.5	19.3	-
Biowaste	5.6	6.1	-
Sludge	27.1	34.0	-

[1] Figures are presented with a maximum of 3 significant digits; not to be seen as reflecting precision, but for replicating the calculations.

Layer 1: Technology performance

Bio-based methane produced

Results for layer 1 are shown in Fig. 3, 4, and 5 for climate change, terrestrial acidification, and freshwater eutrophication, respectively. Results for the other impact categories are available in ESI. In the area with the white background (i.e. I. a, II a; III a), the reference fossil-based gas production (RP) is compared with the pathway and associated counterfactual. Positive results indicate burdens while negative are savings. While scenarios that perform better than the RP fall in the area with green background (i.e. I. b1, II. b1, III. b1), those with a worse performance situate in the area with red background (i.e. I. b2, II. b2, III. b2).

The RP had an impact of 2.33 kg $CO_{2\_eq.}$ Nm^{-3} CH_4, 3.13E+04 kg SO_{2_eq} Nm^3 CH4, and 1.23E-06 kg P $_{eq}$ Nm^3 CH4, respectively for the three impact categories in Fig. 3, 4 and 5.

For global warming (GW) (Fig. 3), only intercrop (NB6) in the AD pathway slightly exceeded RP (Fig. 3, II. b2) with 2.36 vs 2.33 kg CO_{2_eq} Nm^{-3} CH_4. For terrestrial acidification (Fig. 4), three categories, such as sludge (NB9), biowaste (NB8), and agrofood residues (NB3), exceeded RP in ADH and AD, while in GA only pruning residues (NB4) did. Finally, for freshwater eutrophication (Fig. 5), two categories, sludge (NB9) and intercrop (NB6), exceeded RP in both ADH and AD pathways, while only wood waste and industrial residues (NB7) did in the GA pathway. The NB in freshwater eutrophication for each category and pathway was almost as the RP, except for the manure and slurry (NB1) in the ADH and AD pathway that was lower.

Contribution of counterfactuals and pathways to environmental results

Savings given by the counterfactuals and the three pathways are different for each impact category, however there are some trends. AD and ADH have savings mostly from *material substitution* caused by the mineral fertilizers' substitution (for sludge, manure and slurry, green waste, and household biowaste after composting), together with the avoided intercrop production (e.g. due to removing intercrop from the field when mineral fertilizers are used) and vinegar production induced in the specific case of the waste wine used in the agrofood residues category. For GA, energy substitution when residues were burned, was the main source of savings together with mineral fertilizer substitution when residues were left on-soil (e.g. *use on-land, storage, and ploughing* of pruning, crop residues, forest residues, and intercrop). Emissions given by the counterfactuals and the three pathways are mostly given by *use on-land, storage, and ploughing* for manure and slurry, sludge, biowaste, intercrop, green waste, agrofood residues, and crop residues. However, for GW *gas production and supply* contributed mainly from the three pathways. .

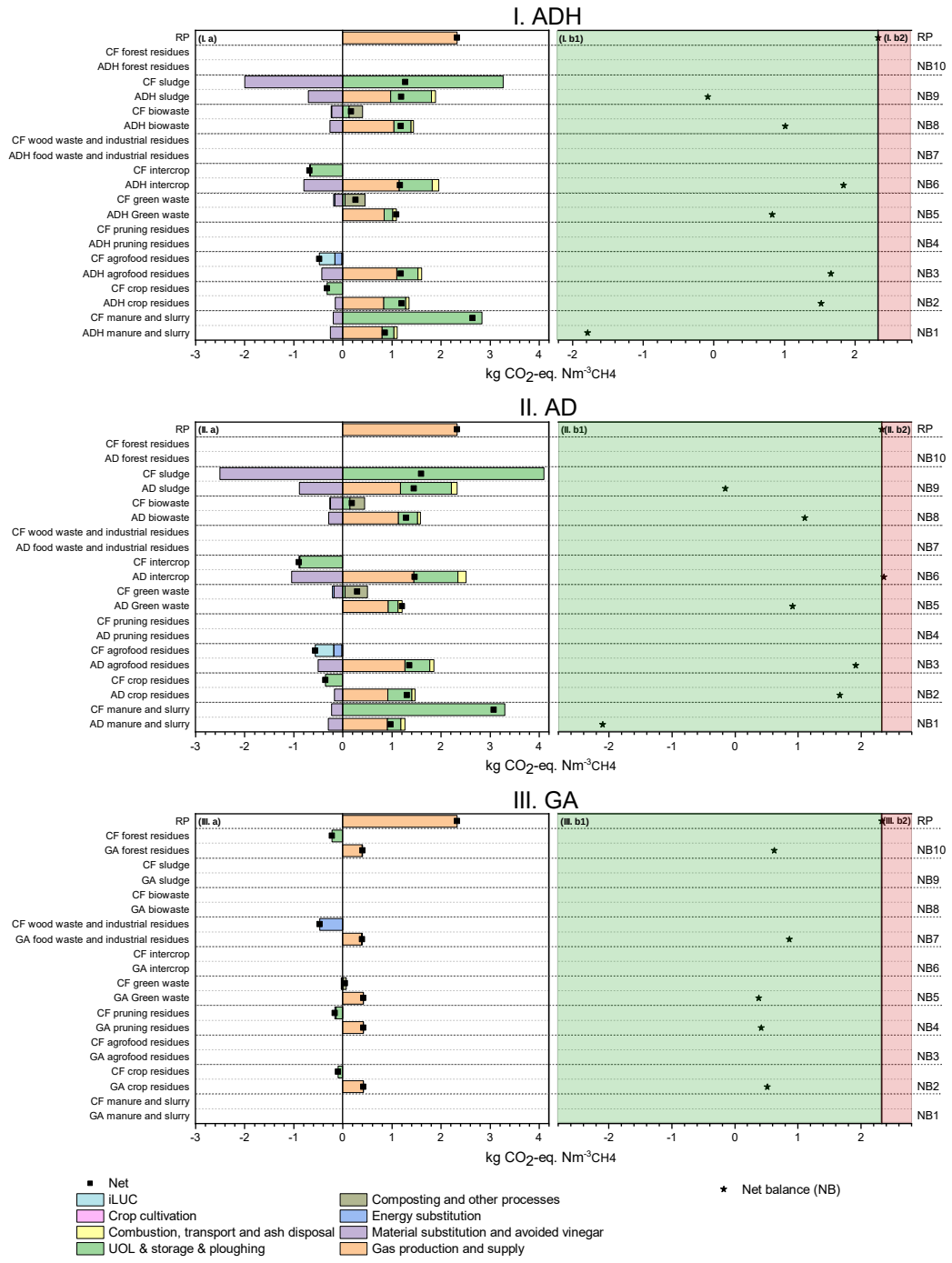

Fig. 3: Results for the climate change impact category associated to the ten residue categories treated in three different methane conversion routes: *I. ADH* (anaerobic digestion with upgrading with hydrogen), *II. AD* (anaerobic digestion with upgrading without hydrogen), *III.GA* (gasification with C-to-CH4 upgrading). On the left (*I. a*, *II. a*, *III. a*), contribution results for the three pathways together with counterfactuals (*CF*) and reference (*RP*). On the right (*I. b1 - b2*, *II. b1 – b2*, *III. b1 – b2*), net balance (*NB*)

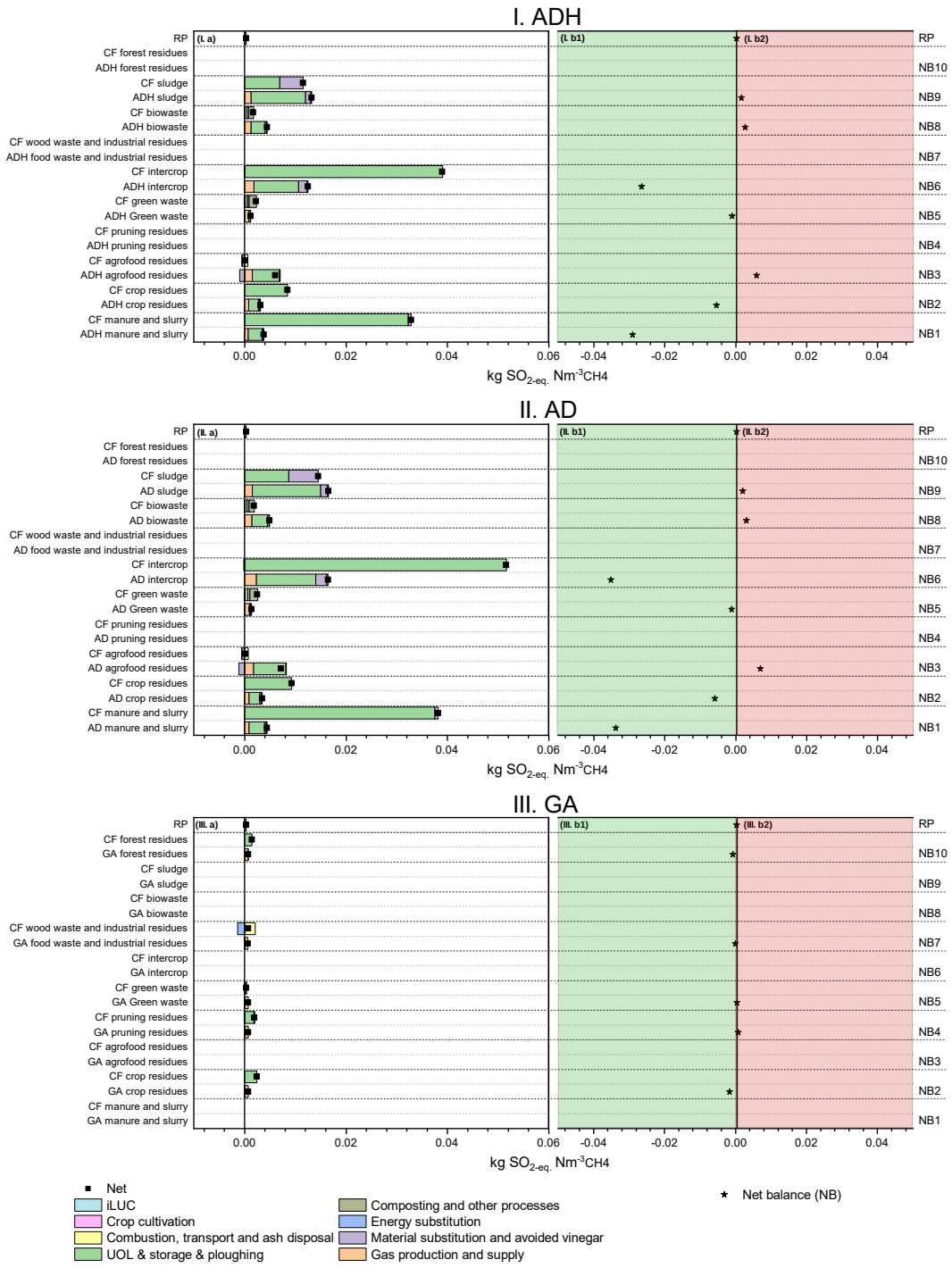

Fig. 4: Results for the terrestrial acidification impact category associated to the ten residue categories treated in three different methane conversion routes: *I. ADH* (anaerobic digestion with upgrading with hydrogen), *II. AD* (anaerobic digestion with upgrading without hydrogen), *III.GA* (gasification with C-to-CH4 upgrading). On the left (*I. a*, *II. a*, *III. a*), contribution results for the three pathways together with counterfactuals (*CF*) and reference (*RP*). On the right (*I. b1 - b2*, *II. b1 – b2*, *III. b1 – b2*), net balance (*NB*)

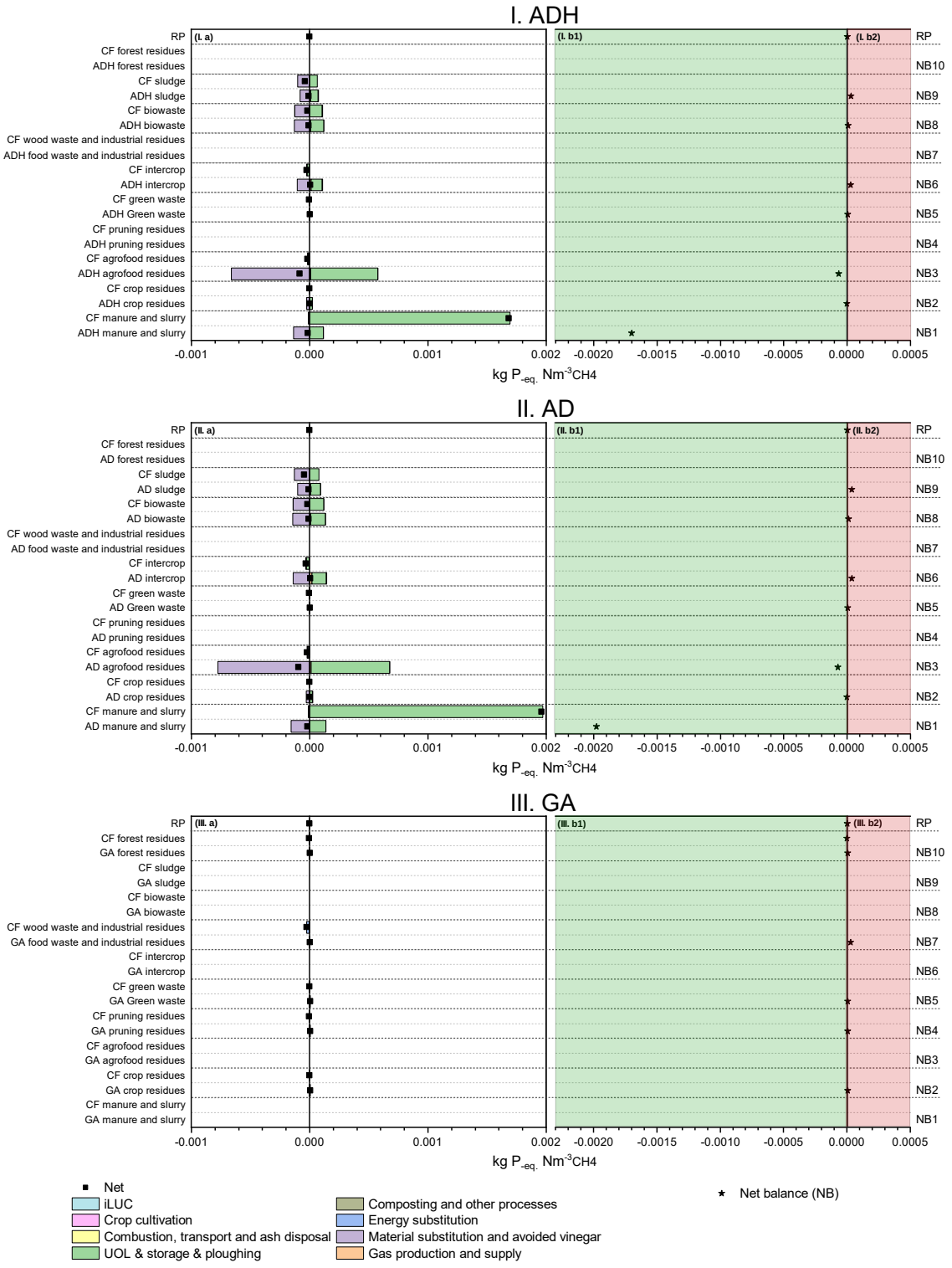

Fig. 5: Results for the freshwater eutrophication impact category associated to the ten residue categories treated in three different methane conversion routes: *I. ADH* (anaerobic digestion with upgrading with hydrogen), *II. AD* (anaerobic digestion with upgrading without hydrogen), *III.GA* (gasification with C-to-CH4 upgrading). On the left (*I. a*, *II. a*, *III. a*), contribution results for the three pathways together with counterfactuals (*CF*) and reference (*RP*). On the right (*I. b1 - b2*, *II. b1 – b2*, *III. b1 – b2*), net balance (*NB*)

Layer 2: System-level scenarios

Bio-based methane production and GW are the two elements to consider for the results of layer 2. We quantify CH_4 supplied to the gas grid as produced by the three conversion-pathways (ADH, AD, and GA), and the GW for each scenario resulting from the NB for each combination of category of bioresources. Results are shown in Tab. 2.

Tab. 2: Results overview in terms of GW and CH4 production for each combination of category of bioresources (column) and technology pathway (row). Highest yields of methane are in green, and the lowest GW emissions are in blue.

		Crop residues	Pruning residues	Green waste	Manure	Intercrop	Forest residues	Wood waste	Agrofood residues	Biowaste	Sludge
ADH	CH_4 in the gas grid (Nm^3)	1.51E+09	-	3.76E+07	2.31E+08	2.70E+08	-	-	3.94E+07	1.35E+08	2.40E+07
	CO_2 in the gas grid (Nm^3)	-	-	-	-	-	-	-	-	-	-
	GW net ($Mt\_CO_{2\_eq}$)	2.29	-	0.03	-0.41	0.50	-	-	0.07	0.15	-0.002
AD	CH_4 in the gas grid (Nm^3)	1.37E+09	-	3.36E+07	1.99E+08	-	-	-	3.36E+07	1.23E+08	1.91E+07
	CO_2 in the gas grid (Nm^3)	1.24E+07	-	9.47E-01	1.80E+06	-	-	-	3.04E+05	1.11E+06	1.73E+05
	GW net ($Mt\_CO_{2\_eq}$)	2.28	-	0.03	-0.42	-	-	-	0.06	0.14	-0.003
GA	CH_4 in the gas grid (Nm^3)	5.26E+09	9.9E+07	2.27E+08	-	-	2.50E+07	6.08E+08	-	-	-
	CO_2 in the gas grid (Nm^3)	-	-	-	-	-	-	-	-	-	-
	GW net ($Mt\_CO_{2\_eq}$)	2.70	5.72	0.09	-	-	0.02	0.53	-	-	-

Combined ADH-AD-GA scenario

In the combined scenario, the highest methane yield was given by GA (for 5 categories) and ADH (for the other 5) (Tab. 3). For the highest GW emissions, AD had lowest emissions for six categories, followed by GA (with three), and ADH (one). The combined scenario not only met the annual Occitania gas demand, but also exceeded it of three times with a total production and utilization of 73 TWh of bio-based methane (vs 17.5 TWh). Thus, this combined scenario allows exports of bio-based methane.

Tab. 3: ADH-AD-GA combined-scenario in terms of bio-based methane production and GW

ADH-AD-GA scenario	CH_4		GW	
Category	**Tech**	**CH_4** (TWh)	**Tech**	**GW** (Mt CO_{2_eq})
Crop residues	GA	55.71	AD	2.29
Pruning residues	GA	1.05	GA	0.06
Green waste	GA	2.41	AD	0.03
Manure	ADH	2.45	AD	-0.42
Intercrop	ADH	2.86	ADH	0.50
Forest residues	GA	0.26	GA	0.02
Wood waste/industrial and end of life	GA	6.44	GA	0.53
Agrofood residues	ADH	0.42	AD	0.07
Biowaste	ADH	1.43	AD	0.14
Sludge	ADH	0.25	AD	-0.003
	Total CH_4	73.28	Total GW	3.20

AD1 scenario: minimum set of bioresources

In AD1 scenario (Tab. 4), we selected only AD with water scrubber upgrading and the minimum set of categories of bioresource to reach the exactly amount of the Occitania gas demand. The realization of this scenario was possible due to the high potential of bio-based methane in Occitania. This allowed to select only residual bioresources biologically degradable and with the pathway having less methane yield. In addition, we did not consider categories of bioresources, such as intercrop, excluded preliminary, due to by their higher NB than the fossil RP for GW emissions. Moreover, bio-based methane was still higher than the Occitania gas demand after all these exclusions. Further exclusion can be done by considering the methane yield of each category and the gas demand. Therefore, we did not include 1.30 TWh of CH_4, and 0.1 Mt CO_{2_eq} from biowaste. With this scenario we equaled the Occitan gas demand without any import/export, and we had ca. 2 Mt CO_{2_eq}.

Tab. 4: AD1 scenario with the minimum set of bioresource to fulfill the gas demand of Occitania

AD1 scenario	CH_4		GW	
Category	**Tech**	**CH_4** (TWh)	**Tech**	**GW** (Mt CO_{2_eq})
Crop residues	AD	14.54	AD	2.28
Pruning residues	-	-	-	-
Green waste	AD	0.36	AD	0.03
Manure	AD	2.11	AD	-0.42
Intercrop	-	-	-	-
Forest residues	-	-	-	-
Wood waste/industrial and end of life	-	-	-	-
Agrofood residues	AD	0.36	AD	0.06
Biowaste	-	-	-	-
Sludge	AD	0.20	AD	-0.003
	Total CH_4	17.56	Total GW	2.09

AD2 scenario: only anaerobic digestion without hydrogen upgrading

AD2 scenario (Tab. 5) follows the same principle of AD1. However, AD2 has a different purpose than AD1: Instead of equaling the Occitania gas demand, AD2 evaluates the effects on the methane yield and GW of using only AD as a widespread technology, which does not require many investments. The bio-based methane production was ca. 19 TWh, meeting the Occitania gas demand and with ca. 1 TWh of exportable gas. AD2 had ca. 2 Mt CO_{2_eq}.

Tab. 5: AD2 scenario, only anaerobic digestion with upgrading (water scrubbing)

AD2 scenario	CH_4		GW	
Category	**Tech**	**CH_4**(TWh)	**Tech**	**GW** (Mt CO_{2_eq})
Crop residues	AD	14.54	AD	2.28
Pruning residues	-	-	-	-
Green waste	AD	0.36	AD	0.03
Manure	AD	2.11	AD	-0.42
Intercrop	-	-	-	-
Forest residues	-	-	-	-
Wood waste/industrial and end of life	-	-	-	-
Agrofood residues	AD	0.36	AD	0.06
Biowaste	AD	1.30	AD	0.14
Sludge	AD	0.20	AD	-0.003
	Total CH_4	18.86	Total GW	2.09

ADH scenario: only anaerobic digestion with hydrogen upgrading

In ADH scenario (Tab. 6), we selected only ADH with alkaline electrolyzer for hydrogen production and further ex-situ upgrading. The goal was to evaluate the effects on the bio-based gas and GW emissions by using only ADH. The realization of this scenario was possible due to the potential of bio-based methane given by the bioresources available in Occitania. In ADH, we considered only biologically degradable bioresources as for AD1 and AD2. On the contrary, we included intercrop because its NB had a better GW performance than the fossil RP. For the ADH scenario, the bio-based methane gas was about 24 TWh, exceeding of about 6 TWh the Occitan gas demand. These 6 TWh can be exported. For ADH, GW emission was about 3 Mt CO_{2_eq}.

Tab. 6: ADH scenario, only anaerobic digestion with hydrogen upgrading

ADH scenario	CH_4		GW	
Category	**Tech**	**CH_4**(TWh)	**Tech**	**GW** (Mt CO_{2_eq})
Crop residues	ADH	15.98	ADH	2.28
Pruning residues	-	-	-	-
Green waste	ADH	0.40	ADH	0.03
Manure	ADH	2.45	ADH	-0.41
Intercrop	ADH	2.86	ADH	0.50
Forest residues	-	-	-	-
Wood waste/industrial and end of life	-	-	-	-
Agrofood residues	ADH	0.42	ADH	0.65
Biowaste	ADH	1.43	ADH	0.14
Sludge	ADH	0.25	ADH	-0.002
	Total CH_4	23.78	Total GW	2.60

Framework insights and main learnings

The stepwise framework here presented allows to use the hydrocarbon source of local available residual bioresources for versatile bio-based methane, as it can be stored in the existing gas grid infrastructure. Thus, providing solutions for waste management and non-fossil source of CH_4.

It supplies a tailor-made substrate-based decision for the selection of the biological or thermochemical degradation. This is relevant for the technology conversion performance, which is affected by the nature of the input and the technology configuration. Furthermore, this framework includes the process-oriented modelling of the syngas upgrading to CH_4, providing the first LCA inventory on this technology. Additionally, the results are given in two sequential layers: i) at a technology for preliminary exclusion of scenarios and evaluation of the contribution to environmental performance, and ii) at a system level, for identification of combination of scenarios for fulfillment of the functional unit. By applying this framework, it is possible to give practical investments' solution, in terms of technologies, while ensuring the environmental and economic performance. Finally, it is possible to reproduce this framework on other regional contexts.

In France, injection of biomethane into the natural gas grid is in a constant progression and development.[46] The promotion of the market for biomethane through several European projects (e.g. GreenGasGrid project, [47]), supports the methane conversion route of the anaerobic digestion. However, Occitania may consider investing in the gasification to be able to use and valorize other not digestible residues and expand the production of bio-based methane. This agrees with [48].

The main learning from the above case studies include:

(1) Bio-based methane is very little and unexploited, even if its production is feasible through existing technologies, available biomasses, and gas grid infrastructures. Regions as Occitania, with a lot of residual bioresources, can help to supply the gas demand of neighboring regions not self-sufficient.
(2) Gasification is a promising technology, particularly C-to-CH4 upgrading, which converts all the carbon in input bioresources.
(3) All combined scenarios at a system level fulfil the Occitania gas demand.
(4) Benefits or burdens given by counterfactuals are relevant for the contribution to LCA results at both, layer 1 to compare with the conversion pathway and the reference product, and at layer 2 included in the GW performance of each scenario, important for the selection of combined scenarios.

Conclusions

The stepwise framework here proposed allows finding environmentally-efficient import/export strategies to supply the fossil gas demand of a region with the bio-based methane produced from local available residual bioresources. The application on the French region Occitania resulted in meeting and exceeding the annual gas demand, thus supported export strategies. The scenario with the higher

production was the combined ADH-AD-GA scenario with ca. 73 TWh of bio-based methane (against ca. 18 TWh demanded in Occitania, annual basis) and a GW associated of 3 Mt $CO_{2\text{-eq}}$. This scenario included thermochemical and biological biomass degradation. Results showed the potential of investment in gasification and chemical upgrading with hydrogen.

Future perspectives

For future studies special focus should be on the following topics:

(1) Combing technologies, recirculating material/energy. For example, the hydrogen in the syngas can be used in the ex-situ unit for the Sabatier reaction converting carbon dioxide of biogas to biomethane.

(2) Optimization for the identification of the optimal combined scenario that supply the gas demand of a region under given system constraints, such as: i) minimization of environmental emissions for all impact categories, ii) parameter uncertainty in LCI and propagation; iii) selection of the conversion route for each input residual bioresource; iv) comparison between net balance of scenarios and fossil RP; v) maximization of bio-based methane.

(3) Exploiting the potential of intercrop for bio-based methane, which at moment is underestimated.

(4) C-to-CH4 upgrading from syngas.

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