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Towards alternative solutions for flaring: Life cycle assessment and carbon substance flow analysis of associated gas conversion into C3 chemicals

Jordy Motte [a,*], Pieter Nachtergaele [a], Mohamed Mahmoud [b], Hank Vleeming [b], Joris W. Thybaut [c], Jeroen Poissonnier [c], Jo Dewulf [a]

[a] *Research Group Sustainable Systems Engineering (STEN), Department of Green Chemistry and Technology, Ghent University, Coupure Links 653, 9000, Gent, Belgium*
[b] *Process Design Center (PDC), Paardeweide 7, NL-4824 EH, Breda, the Netherlands*
[c] *Laboratory for Chemical Technology (LCT), Department of Materials, Textiles and Chemical Engineering, Ghent University, Technologiepark 125, 9052, Ghent, Belgium*

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ABSTRACT

Gas flaring has many environmental impacts at global and local scale. Conversion of associated gas into 1-propanol (scenario PRL) and propylene (scenario PRE) via the C123 process can be a potential solution to prevent combustion. This paper aims to evaluate the environmental performance of C3 production from associated gas compared to flaring and to identify the preferred C3 chemical for associated gas conversion. A carbon substance flow analysis (CSFA) and a life cycle assessment (LCA) were conducted. CSFA was used to map all carbon flows and to calculate the carbon emission savings and carbon efficiency. The LCA focused on the impact categories climate change, fossil resource use, human toxicity and the cumulative exergy extraction from the environment. The results of the CSFA indicate that 2.89 kg CO_2 per kg associated gas could be saved in scenario PRL, when including the avoided conventional C3 production in the analysis. The LCA shows that scenario PRL outperforms flaring for climate change and human toxicity. Consequently, 1-propanol production from associated gas is the preferred alternative at the selected location. Heat integration and renewable electricity production can drastically decrease the impact of C3 chemicals production on climate change and enable CO_2 emissions savings compared to flaring.

1. Introduction

In 2021, the oil and gas sector flared 143 billion m^3 gas worldwide, equivalent to 380 million tons of CO_2 (World Bank, 2022). These emissions amount to 1% of the global CO_2 emissions linked to the burning of fossil fuels (International Energy Agency, 2020). In some countries, flaring can account for a large share in the total CO_2 emissions across all sectors (e.g., 19% of the total greenhouse gas emissions in Iran) (Khalili-Garakani et al., 2021). Most gas is combusted in the upstream processing of crude oil for economic and technical reasons (Elvidge et al., 2016). This gas fraction associated with oil in an oil reservoir is called associated gas. Flaring has several environmental impacts at both global and local scale (Soltanieh et al., 2016). First, the emitted greenhouse gases contribute to global warming. Second, toxic emissions, such as H_2S, CO, SO_2, particulate matter and non-methane volatile organic carbons (NMVOC), cause serious health effects, especially when flaring occurs close to communities (e.g., Niger Delta) (Ajugwo, 2013).

Several associated gas conversion technologies were already invented to utilise this gas fraction and to avoid flaring, namely reinjection of the gas in the oil reservoir, transportation via pipelines, liquefied natural gas, gas to liquids, gas to chemicals, gas to power, etc. A detailed description of these utilisation processes can be found in Khalili-Garakani et al. (2021). Various constraints hinder the big scale implementation of these gas recovery technologies such as high capital and operational costs for gas sweetening and collection, poor economies of scale for low gas volumes, lacking infrastructure for utilities production (electricity), no long term pricing policy for associated gas, etc. In contrast, multiple projects with the successful application of associated gas conversion technologies were reported in Iran. Reinjection and the production of natural gas liquids (NGL) are the most chosen technologies in this country (Khalili-Garakani et al., 2021).

In the C123-project (EU Horizon 2020 project), the industrial production of C3 chemicals such as propylene and 1-propanol from

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* Corresponding author. Research Group Sustainable Systems Engineering (STEN), Department of Green Chemistry and Technology, Faculty of Bioscience Engineering, Ghent University, Coupure Links 653, 9000, Gent, Belgium.
E-mail address: jordy.motte@ugent.be (J. Motte).

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associated gas is studied (European Commission, 2022; Fonseca et al., 2021; Motte et al., 2022; Sintef, 2022). This unconventional methane source is processed via a new process concept, named C123 process throughout this paper, consisting of several chemical reactions. An oxidative conversion of methane followed by a hydroformylation and hydrogenation are needed to obtain 1-propanol. An additional dehydration step transforms 1-propanol into propylene (Motte et al., 2022; European Commission, 2022). This process concept could be added to existing refinery infrastructure (add-on unit) and it may have several advantages compared to flaring. Carbon emissions may be avoided for a longer time due to their storage in chemicals instead of instant combustion, resulting in a reduced impact on climate change. Next, the creation of harmful emissions because of flaring can be prevented which may have a beneficial impact on human health when flaring takes place close to cities.

Apart from waste valorisation, this new technology may be a more sustainable option for the production of C3 chemicals. Currently, steam cracking is the predominant technology to manufacture propylene from naphtha and other fossil feedstocks such as vacuum gas oil (Amghizar et al., 2017). This is a very energy intensive process which causes huge greenhouse gas emissions (Ren et al., 2006). The production of 1-propanol also relies on steam cracking of naphtha and syngas production as ethylene and CO are used as an intermediate (Nanda et al., 2020). Consequently, its current manufacturing is energy demanding. Via hydroformylation and propanal hydrogenation, 1-propanol is obtained from ethylene and CO. The C123 process could be an interesting and novel production pathway for C3 chemicals which may be less energy intensive and may have a lower impact on the environment.

To analyse the environmental performance of a process concept, material flow analysis (MFA) and life cycle assessment (LCA) are useful tools. MFA provides an overview of all flows and stocks of a material or substance in a system. This analysis can give insights into the resource use of a technology and its impact on the environment when combined with sustainability assessment. In addition, it can serve as a good basis for process or product design (Huang et al., 2012). This paper focuses on carbon flows within the C123 process concept and its supporting processes. Carbon substance flow analysis (CSFA) was already applied by Zhang et al. (2013) to compare different steel production processes and to recommend carbon mitigation strategies for the steel sector. Ma et al. (2012) estimated the future carbon emissions at country level (China) through this method. In several studies, indicators were developed to measure CO_2 emissions based on CSFA whether or not compared to a reference (Liu et al., 2022; Wang et al., 2016; Zhang et al., 2013). In LCA, the environmental impact of a product is quantified based on all life cycle stages, from resource extraction to product disposal (ISO, 2006a). Only a limited number of environmental assessments on associated gas conversion technologies are available in the literature. Rajovic et al. (2016) investigated the production of heat and electricity from this gas fraction. Khalili-Garakani et al. (2022) studied various flare gas recovery technologies (e.g., gas-to-power, LNG production, etc.) for different gas samples. Most flare gas recovery technologies lead to a CO_2 emission reduction compared to flaring, except gas-to-power. However, no full life cycle approach was used, no other impact categories were considered besides climate change and the preceding oil extraction phase was not taken into account. Consequently, the literature lacks comprehensive environmental sustainability assessments on associated gas conversion into (C3) chemicals.

This paper aims to assess the environmental performance of propylene and 1-propanol production from associated gas via the preliminary C123 process concept at technology readiness level (TRL) 4 compared to flaring. Moreover, the best associated gas conversion route (propylene or 1-propanol) was identified for one specific location. A CSFA and an LCA were conducted for this purpose. Based on the CSFA, all carbon flows within the system were mapped and the carbon efficiency and the CO_2 emission savings compared to flaring were calculated. Next, the LCA focused on global warming, fossil resource use, human toxicity and exergy extraction from the natural environment. Finally, based on these assessments, some improvements to the C123 process concept were suggested for process developers.

2. Methodology

2.1. Description case study: preliminary production process and selected location

Fig. 1 illustrates the preliminary C123 process concept for the conversion of associated gas into 1-propanol and propylene via an unit added to existing infrastructure at TRL 4. A more detailed process flow diagram can be found in Appendix A. First, a pretreatment of associated gas was needed to remove propane and butane as some impurities (e.g., butanol and pentanol) could otherwise be formed during this conversion process. The extracted liquefied petroleum gas, consisting of higher hydrocarbons (C_{3+}), can be used for local heat and electricity production through a combined heat and power installation (CHP). Heat integration was not included yet in the process design. It was assumed that no H_2S or mercaptans are present in the gas at the selected location (Ghadhban and Al-Fathi, 2001). After pretreatment, a mixture of ethylene, CO and H_2 was obtained in the oxidative conversion of methane (OCoM), which includes an oxidative coupling of methane (OCM) and a post-bed cracking (PBC) unit. For the former, 1.13 kg and 1.64 kg oxygen gas, obtained from cryogenic air separation, were added per kg propanol and propylene, respectively. Afterwards, water and CO_2 were partially removed via several compression steps and absorption with mono-ethanolamine. These components originated from the feed or were created in the OCoM section. Cooling between the compression stages was provided through an absorption chiller. The ethylene-CO-H_2 mixture was converted into propanal via hydroformylation. A hydrogenation and dehydration reaction were required to obtain propylene via 1-propanol as intermediate. After hydroformylation, the excess of methane, ethane, CO_2 and hydrogen gas was separated from the produced propanal through extractive distillation. The subsequent pressure swing adsorption unit recovered H_2 from the remaining fraction for its use in the hydrogenation section. However, additional hydrogen (0.026 kg per kg 1-propanol and 0.038 kg per kg propylene) must be added for complete conversion. The required hydrogen was locally produced via water electrolysis. Methane, ethane and CO_2 were sent back to the OCoM section. Finally, in case of propylene production, water formed in the dehydration was again removed through phase separation. More details about the modelling of the C123 process concept can be found in Appendix B. Iraq, especially the Zubair oil field, was the preferred location to apply this process concept due to the large availability of associated gas (1.74 billion m^3 per year), the proven oil reserves for approximately 35 years and the proximity of petrochemical refineries (Carboline, 2017; World Bank, 2020). In addition, the desired production scale of 300 ktonnes per year can be met at this place. It was assumed that all energy is produced from fossil resources as this is mostly the case in the oil and gas sector, especially at a remote location (Halabi et al., 2015).

2.2. Data collection

Process simulations were performed in Unisim to derive data, such as weight fractions, temperatures and pressures of different process flows for the conversion of associated gas into 1-propanol and propylene. Chemical process information for the removal of natural gas liquids, water electrolysis and oxygen production was retrieved from Jiang et al. (2018), Dufour et al. (2012) and the Ecoinvent database version 3.8, respectively. For the utilities, heat and electricity production from fossil fuels was considered and data was obtained from Ecoinvent. Mass and energy balances were composed based on this data (see Appendix C for entire life cycle inventory). Data such as mass flows, utilities, emissions, etc. for the conventional 1-propanol and propylene production, which

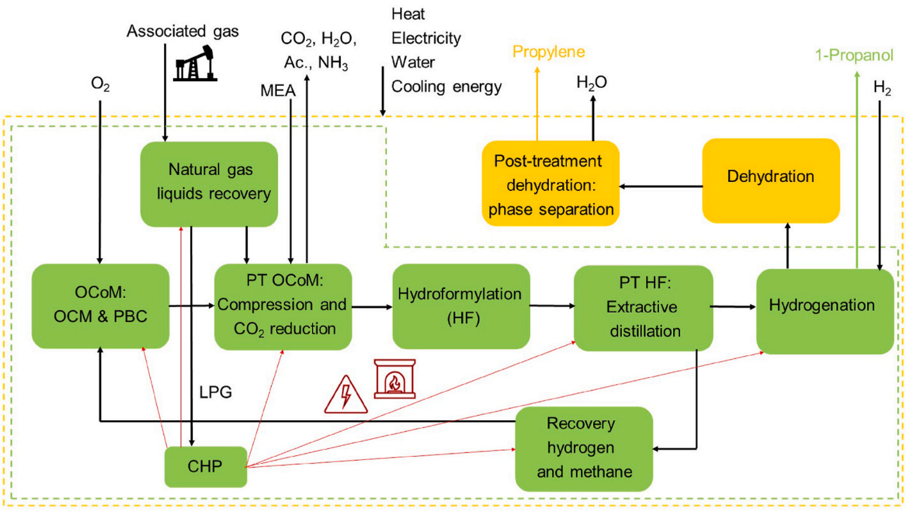

Fig. 1. Preliminary C123 process concept for 1-propanol and propylene production from associated gas. The yellow process blocks are only required for propylene production, while the green process blocks are needed both for 1-propanol and propylene production. The red lines represent heat and electricity produced from the combined heat and power installation. OCoM = oxidative conversion of methane, OCM = oxidative coupling of methane, PBC = post bed cracking, PT = post-treatment, MEA = monoethanolamine, HF = hydroformylation, CHP = combined heat and power installation, LPG = liquefied petroleum gas, Ac. = acetaldehyde. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

are included in the LCA approach (see Section 2.3), was also retrieved from Ecoinvent.

2.3. *Life cycle assessment*

2.3.1. *Goal and scope definition*

The standard LCA methodology was used as specified in the ISO-14040 and ISO-14044 standards (ISO, 2006a; ISO, 2006b). The main goal of this analysis was the environmental performance assessment of producing 1-propanol and propylene from associated gas. The scope of the analysis was from cradle-to-gate, meaning that the distribution, use and disposal of the product was not taken into account. The flaring process was chosen as benchmark because this is still the most common end-of-life scenario for associated gas nowadays. The corresponding flaring emissions were quantified based on the associated gas composition at the location in Iraq and emission factors that could be retrieved from the literature (Environmental Energy Agency, 2019; Kseer and Gzar, 2009; Liousse et al., 2019) (see Appendix D).

2.3.2. *Determination of the functional unit*

It was found that 1 kg associated gas can be converted into either 0.60 kg 1-propanol or 0.41 kg propylene. To enable a fair comparison between C3 chemicals production and flaring, a system expansion was conducted both for the C123 and the reference scenarios as illustrated in Fig. 2. All scenarios include the production of 0.60 kg 1-propanol and 0.41 propylene, either via the C123 process or via conventional production, as it is considered that the market demand for both C3 products must be met. The system expansion for the C123 scenarios contains the conventional propylene production in C123 scenario PRL and the conventional 1-propanol production in C123 scenario PRE. For 1-propanol, the fossil based route, starting from crude oil and including steam cracking of naphtha for ethylene, syngas production for CO, hydroformylation and hydrogenation, was taken into account. In case of propylene, the steam cracking of naphtha was used for conventional production. For the reference scenario, both the conventional 1-propanol and propylene production were accounted for. The environmental impact of the C123 scenarios was compared to the reference scenario by taking the sum of the burden of 1-propanol (or propylene) production and the conventional propylene (or 1-propanol) production. For the reference, the burden of the flaring process and the conventional 1-propanol and propylene were summed. Based on this approach, the functional unit was defined as the treatment of 1 kg associated gas, obtained from oil extraction, and converted into 0.60 kg 1-propanol and 0.41 kg propylene or flared completely.

2.3.3. *Impact assessment*

The Environmental Footprint method was selected for the impact assessment at midpoint level (Saouter et al., 2018). This study focused on climate change (in kg CO$_2$ eq), fossil resource use (in MJ) and human toxicity (in comparative toxic units (CTUh)). The resource footprint was also quantified via the cumulative exergy extraction from the natural environment (CEENE) (in MJex). The first two impact categories were chosen since the C123 project aims to reduce global warming and fossil resource use compared to the current C3 production and flaring via the novel process concept. Human toxicity was also targeted due to the toxic emissions caused by flaring. The C123 process may drastically decrease these emissions as no more combustion reaction takes place. Finally, the CEENE method was selected to evaluate the total resource extraction (in terms of exergy) of the C123 process. The Simapro software version 9.4.0.2 was used to obtain the results. At the crude oil extraction stage, energy allocation was applied between crude oil and associated gas to determine the corresponding burden for associated gas. In this case, 98.8% of the impact of petroleum extraction was attributed to petroleum and 1.2% to associated gas.

2.3.4. *Interpretation of results*

A contribution analysis was performed to identify the C123 production steps with the greatest environmental impact. Finally, a sensitivity and uncertainty analysis were conducted. For the sensitivity analysis, the electricity consumption was varied between 15 and 30 MJ as this parameter affects the results the most. The electricity need may increase if a catalyst can be found that allows higher conversion of methane in the OCoM section. Consequently, more oxygen would be consumed (doubled in the worst case). The production of this chemical requires a significant amount of electricity. A confidence interval of 95% was chosen for the uncertainty analysis.

2.4. *Carbon substance flow analysis (CSFA)*

Based on the acquired mass balance, a CSFA was conducted for C3 chemicals production from associated gas via the C123 process concept. Therefore, 1 kg associated gas was used as starting point and both the conversion of associated gas and the local energy production were considered. First, the carbon content of each flow was calculated. Both feedstocks, process flows and emissions were included. For chemical mixtures, such as associated gas, the mass of the individual components in the mixture was retrieved by means of their mass fractions.

$$Carbon\ content_{flow}\ (kg) = \sum_{i=1}^{N} \frac{wt_i * mass_{flow} * n_i * MW_{carbon}}{MW_i} \qquad \text{(Eq.1)}$$

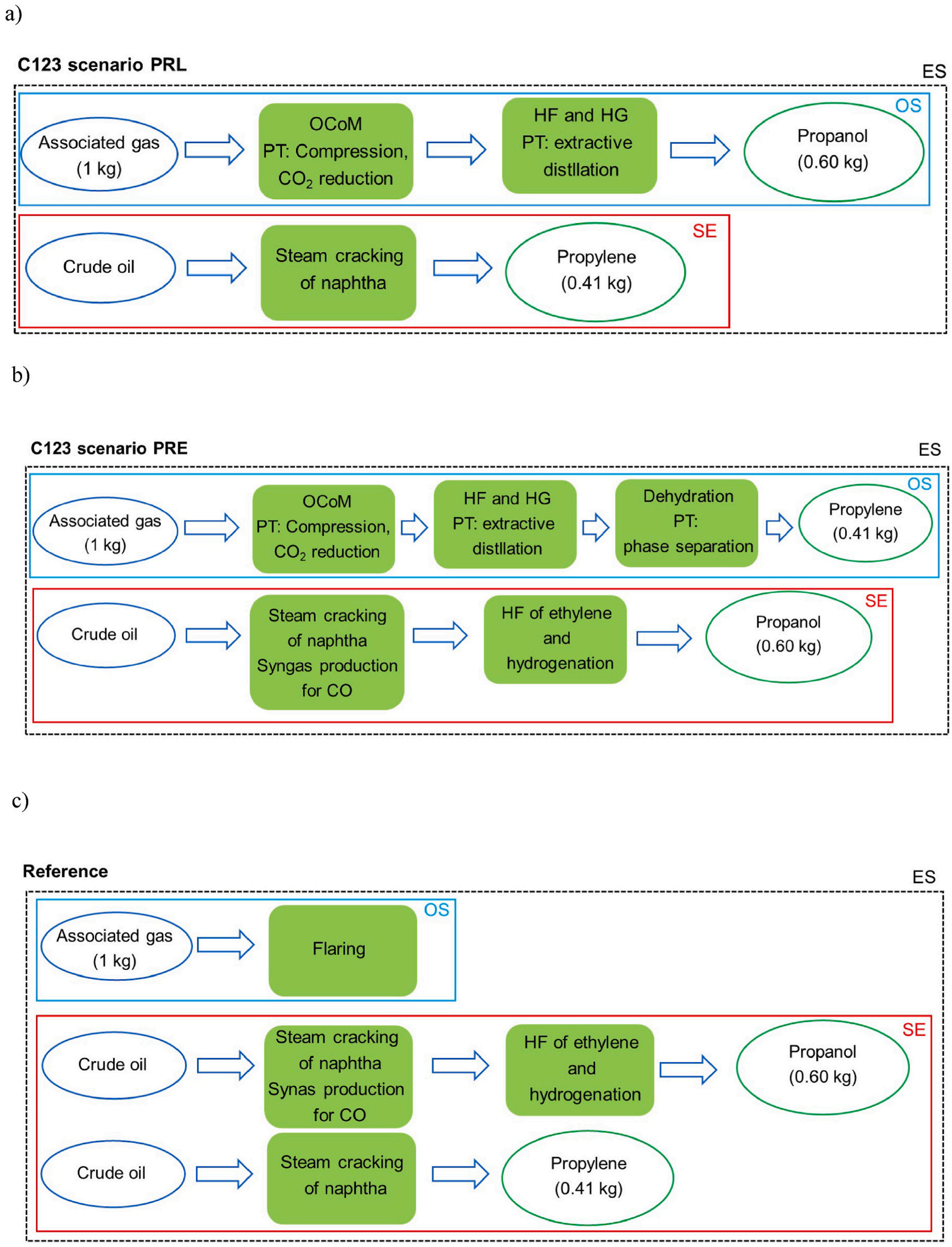

Fig. 2. LCA approach for comparison between associated gas conversion into 1-propanol and propylene via the C123 process and flaring. (a) C123 scenario PRL: associated gas conversion into 1-propanol + conventional propylene production, (b) C123 scenario PRE: associated gas conversion into propylene + conventional 1-propanol production, (c) reference: flaring + conventional propylene production + conventional 1-propanol production. OS = original system, SE = system expansion, ES = expanded system, OCoM = oxidative conversion of methane, PT = post-treatment, HF = hydroformylation, HG = hydrogenation, PRL = 1-propanol, PRE = propylene.

Where $mass_{flow}$ is the mass of a flow in kg, n_i is the number of carbon atoms of a component i in the flow, wt_i is the weight fraction of component i in the flow, MW_i is the molecular weight of a component i in the flow in kg/kmol and N is the total number of components in the flow. Next, the carbon balance was checked based on the law of conservation of mass for the individual process steps and the entire process.

To compare the environmental performance of 1-propanol and propylene production to flaring, the carbon efficiency and the CO_2 emission savings were quantified via the formulas below. These calculations were carried out for scope 1a (excluding local energy production) and scope 1b (including local energy production) to distinguish between the direct and indirect carbon emissions related to C3 chemicals production. Furthermore, these indicators were also calculated for the expanded systems from Section 2.3.

Scope 1a:

$$Carbon\ efficiency\ (\%) = \frac{C_{product}}{C_{associated\ gas}} \tag{Eq.2}$$

$$CO_2\ emission\ savings = \left(C_{flaring} - C_{C123\ direct\ process\ emissions}\right) * \frac{MW_{CO_2}}{MW_{carbon}} \tag{Eq.3}$$

Scope 1b:

$$Carbon\ efficiency\ (\%) = \frac{C_{product}}{C_{associated\ gas} + C_{fossil\ fuels\ for\ energy\ production}} \tag{Eq.4}$$

$$CO_2\ emission\ savings = \left(C_{flaring\ emissions} - C_{C123\ direct\ process\ emissions} - C_{C123\ indirect\ emissions}\right) * \frac{MW_{CO_2}}{MW_{carbon}} \tag{Eq.5}$$

Where $C_{product}$ is the carbon content of the final product in kg, $C_{associated\ gas}$ is the carbon content of associated gas in kg, $C_{flaring\ emissions}$ is the carbon content of the flaring emissions in kg, $C_{C123\ direct\ process\ emissions}$ is the carbon content of the direct carbon emissions excluding the energy production in kg, $C_{fossil\ fuels\ for\ energy\ production}$ is the carbon content of the fossil fuels needed for electricity and heat production in kg and $C_{C123\ indirect\ emissions}$ is the carbon content of the indirect carbon emissions related to the energy production from fossil fuels in kg. The CO_2 emission savings were expressed in kg CO_2 per kg associated gas. All results were visualised via a Sankey diagram (see Section 3.2).

3. Results and discussion

3.1. Life cycle assessment

Table 1 shows the LCA results for all selected midpoint indicators, namely climate change, fossil resource use, human toxicity and CEENE. When the associated gas conversion into 1-propanol and propylene are compared, these values do not differ much as the dehydration and the subsequent phase separation do not require much energy in comparison to the shared C123 production steps. The values for propylene production are slightly higher for all indicators.

Next, C123 scenarios PRL and PRE cause less greenhouse gas emissions than the reference scenario looking at the impact category climate change. However, the result for scenario PRL is 1.45 kg CO_2 equivalents and 1.6×10^{-10} CTUh lower than for scenario PRE due to the higher environmental impact of the conventional propanol production compared to conventional propylene production (3.41 kg CO_2 equivalents per kg 1-propanol and 1.57 kg CO_2 equivalents per kg propylene). Next to steam cracking, syngas production is required to obtain CO for ethylene hydroformylation. Additionally, a much lower result is obtained for human toxicity compared to the reference in both cases because of the many toxic components (particulate matter, CO, etc.) emitted during flaring. Consequently, the implementation of the C123 process could have beneficial human health effects when flaring takes place near communities. This is the case for this study as the selected location is situated close to Basra, a major city in Iraq. For instance, 75% of the damage to human health by particulate matter emissions from flaring can be caused at a distance of 256 km from the point source (Goodkind et al., 2019). Moreover, C123 scenario PRL consumes fewer (fossil) resources than the current C3 production and flaring together (−11 MJ for fossil resource use and CEENE). However, C123 scenario PRE would require more of these non-renewable resources than in the current situation, as the common 1-propanol production needs more fossil fuel input than propylene production (e.g., for energy production in both the steam cracking and syngas production).

Table 1

Overview of LCA results for associated gas conversion into 1-propanol and propylene compared to flaring. The standard deviation for each impact category obtained via uncertainty analysis is mentioned between brackets. PRL = 1-propanol, PRE = propylene.

Scenario	C123 scenario PRL	C123 scenario PRE	Reference (flaring)
Fossil resource use (MJ)	119 (± 11)	144 (± 21)	130 (± 21)
Climate change (kg CO_2 eq)	4.58 (± 0.14)	6.03 (± 0.53)	6.49 (± 0.39)
Human toxicity (CTUh) ($\times 10^{-10}$)	6.6 (± 0.9)	8.2 (± 1.2)	44.5 (± 0.9)
CEENE (MJex)	141 (± 14)	179 (± 26)	152 (± 26)

In Fig. 3, the different shares of the C123 production steps contributing to climate change are illustrated. The results for the C123 scenarios and the reference are also shown. The total share of the associated gas conversion via the C123 process in scenario PRL and PRE is almost equal to the share of flaring in the reference scenario. The OCoM section is the largest contributor to climate change due to the required oxygen gas. For oxygen production from cryogenic air separation, a substantial amount of electricity is needed. The CO_2 reduction stage and the CHP also have a significant share mainly because of the CO_2 removal and the direct CO_2 emissions generated via combustion of LPG. Flaring has a high contribution in the reference scenario (53.4%). Moreover, the contribution for the conventional 1-propanol production is almost four times larger than for the propylene production. The acetaldehyde emission related to CO_2 reduction affects the result for human toxicity in the 1-propanol and propylene production via the C123 process the most. This compound is released during regeneration of the absorption solvent. The associated gas extraction accounts for most fossil resource use, due to the upstream extraction of petroleum resources partially attributed to associated gas, followed by local electricity production. Finally, fossil fuels also dominate the results for the CEENE as 90% of the extracted exergy corresponds to fossil energy.

When the electricity consumption for the C123 process is adjusted between 15 MJ and 30 MJ in the sensitivity analysis, the impact on climate change ranges between 6.56 and 9.58 kg CO_2 eq per kg 1-propanol and 8.03 and 11.12 kg CO_2 eq per kg propylene. Consequently, with an electricity usage of 30 MJ, scenario PRL still has a lower impact on climate change than the reference. In this case, scenario PRE exceeds the reference for this impact category. The uncertainty analysis shows that only scenario PRL has a significantly different impact on climate change than the reference (see Table 1). The standard deviations are high for fossil resource use, meaning that the fossil fuel consumption is not significantly different compared to the reference in scenarios PRL and PRE. Both scenarios have a substantially lower impact on human health. All details of the uncertainty analysis can be consulted in Appendix E. For the C123 production steps, data was obtained via simulations in the Unisim software. Data validation must be performed when an industrial installation is built.

3.2. Carbon substance flow analysis

Fig. 4 illustrates the results for the CSFA of associated gas conversion into 1-propanol and propylene. In both cases, 1 kg associated gas contains 0.73 kg carbon. First, natural gas liquids (e.g., propane and butane) are recovered for local energy production through the CHP. In the latter unit, 0.24 kg liquefied petroleum gas with 0.20 kg carbon enters the system. Consequently, 0.76 kg purified associated gas (0.53 kg carbon) is sent to the C123 process. The limited loss of carbon is a big advantage of this methane conversion technology. Most hydrocarbons (e.g.,

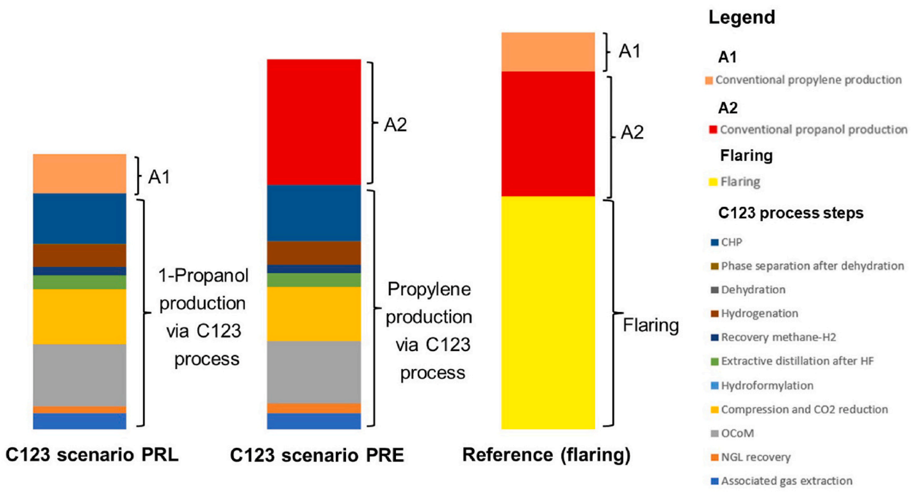

Fig. 3. Contribution analysis of 1-propanol and propylene production from associated gas via C123 for the indicator climate change. The total results for C123 scenario PRL and PRE are shown with the first 2 bars. The result for the reference is illustrated by the last bar. The yellow, red and blue blocks represent flaring emissions, CO_2 emissions associated to conventional 1-propanol production and CO_2 emissions associated to conventional propylene production, respectively. NGL = natural gas liquids, OCoM = oxidative conversion of methane, CHP = combined heat and power, HF = hydroformylation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

methane, ethane, etc.) are recycled within the system. Only in the CO_2 reduction stage, some CO_2 is removed via an amine unit which results in a CO_2 emission. A small amount of purge gas, containing methane, is also released after hydroformylation. Therefore, around 0.12 kg carbon is released in the atmosphere in both cases. The obtained weight of the final products (1-propanol and propylene) amounts to 0.60 kg (0.36 kg carbon) or 0.41 kg (0.35 kg carbon) per kg associated gas. The higher weight for 1-propanol can be explained by the oxygen atom present in the molecule. The CHP causes a CO_2 emission of 0.92 kg (0.25 kg carbon). However, additional electricity and heat production is needed as the energy production in the CHP is not sufficient. In case of 1-propanol, 0.97 kg fossil fuels with a carbon content 0.69 kg are consumed, causing 2.53 kg emitted CO_2. For propylene production, 1.01 kg fossil fuels (0.72 kg carbon) are required for energy production, generating 2.65 kg CO_2.

Based on the CSFA, the carbon efficiency and the CO_2 emission savings are calculated (see Table 2). At scope 1a, the carbon efficiency amounts to 0.49 and 0.48 for 1-propanol and propylene, respectively. Compared to flaring, 1.3 kg CO_2 emissions could be saved per kg associated gas in both cases. The results are similar for 1-propanol and propylene despite the additional dehydration and phase separation that is needed for propylene production. At scope 1b, the carbon efficiency decreases to 0.25 and 0.24 in case of 1-propanol and propylene, respectively, due to the need of fossil fuels for energy production. This also results in no more CO_2 emission savings. The production of 1-propanol emits 1.23 kg more CO_2 than flaring per kg associated gas; propylene production even leads to an increase of 1.38 kg emitted CO_2 per kg associated gas compared to flaring. When the CSFA is applied to the expanded systems from Section 2.3, the carbon emission savings compared to the reference and carbon efficiency for C123 scenario PRL increase to 2.89 kg CO_2 per kg associated gas and 0.37, respectively. However, no carbon emissions could be saved in scenario PRE (plus 0.93 kg CO_2 per kg associated gas). Both results can be explained by the large CO_2 emissions and fossil fuel consumption associated to conventional 1-propanol production (mainly due to steam cracking and syngas production to obtain ethylene and CO, respectively). The carbon efficiency of C123 scenario PRE amounts to 0.23.

3.3. Recommendations for the C123 process

Both the CSFA and the LCA delivered insights to improve the performance of the C123 process. First, the local production of utilities, such as heat and electricity, from fossil resources highly affects the results. The CSFA at scope 1b shows that the combined C123 conversion and the energy production to support the C123 production steps even cause more carbon emissions than those generated by flaring. High fossil resource consumption can be noticed from the LCA impact category fossil resource use, especially for C123 scenario PRE, resulting in more fossil resource extraction than in the current situation. Next, the contribution analysis highlights a high share of OCoM due to the needed oxygen, requiring 20.5 MJ electricity for 1.64 kg oxygen. Full renewable electricity production may drastically reduce the impact of associated gas conversion into C3 chemicals.

Heat integration can slightly drop the impact of local heat production. For instance, heat can be recovered and converted into steam from the exothermic oxidative coupling of methane for utilisation in other production stages with a heat requirement such as the preheater before the OCoM section and the dehydration. In case of propylene and 1-propanol production, 20.1 MJ heat per kg propylene and 14.0 MJ per kg 1-propanol could be used internally.

The combined heat integration and renewable electricity production can decrease the impact on climate change and fossil resource use with 54 and 47%, respectively, in case of propylene production. For 1-propanol production, it drops by 54 and 35%. Due to this intervention, CO_2 emissions at scope 1b can be saved compared to flaring, namely 1.11 kg CO_2 per kg associated gas for propylene. The carbon efficiency at scope 1b can increase to 0.46. For 1-propanol production, the CO_2 emission savings and carbon efficiency at scope 1b would be equal to 1.13 kg CO_2 per kg associated gas and 0.47, respectively.

Apart from the utilities, the used catalysts for the several chemical conversions also influence the results. Other catalysts can increase the carbon conversion efficiency of the C123 technology and can consequently lower the needed amount for some inputs (e.g., associated gas). However, an increased reaction performance may lead to a larger oxygen requirement for OCoM.

Additional hydrogen gas must be produced for hydrogenation. For this purpose, electrolysis of water and electricity generation from fossil resources are assumed at this moment. However, this production method is only interesting in combination with renewable electricity production as it requires a substantial amount of electricity. Steam reforming of methane can be considered to obtain hydrogen when renewable electricity production is not realistic at the selected location. This modification would lower the impact on climate change with 4.35 kg CO_2 equivalents per kg C3 chemical. Finally, carbon capture and storage or utilisation can be considered to avoid the direct CO_2 emissions in the CO_2 reduction stage.

3.4. Future research

3.4.1. Scope of the analysis

In this paper, only a cradle-to-gate LCA of C3 chemical production was conducted. Transportation of C3 chemicals to the market, its use

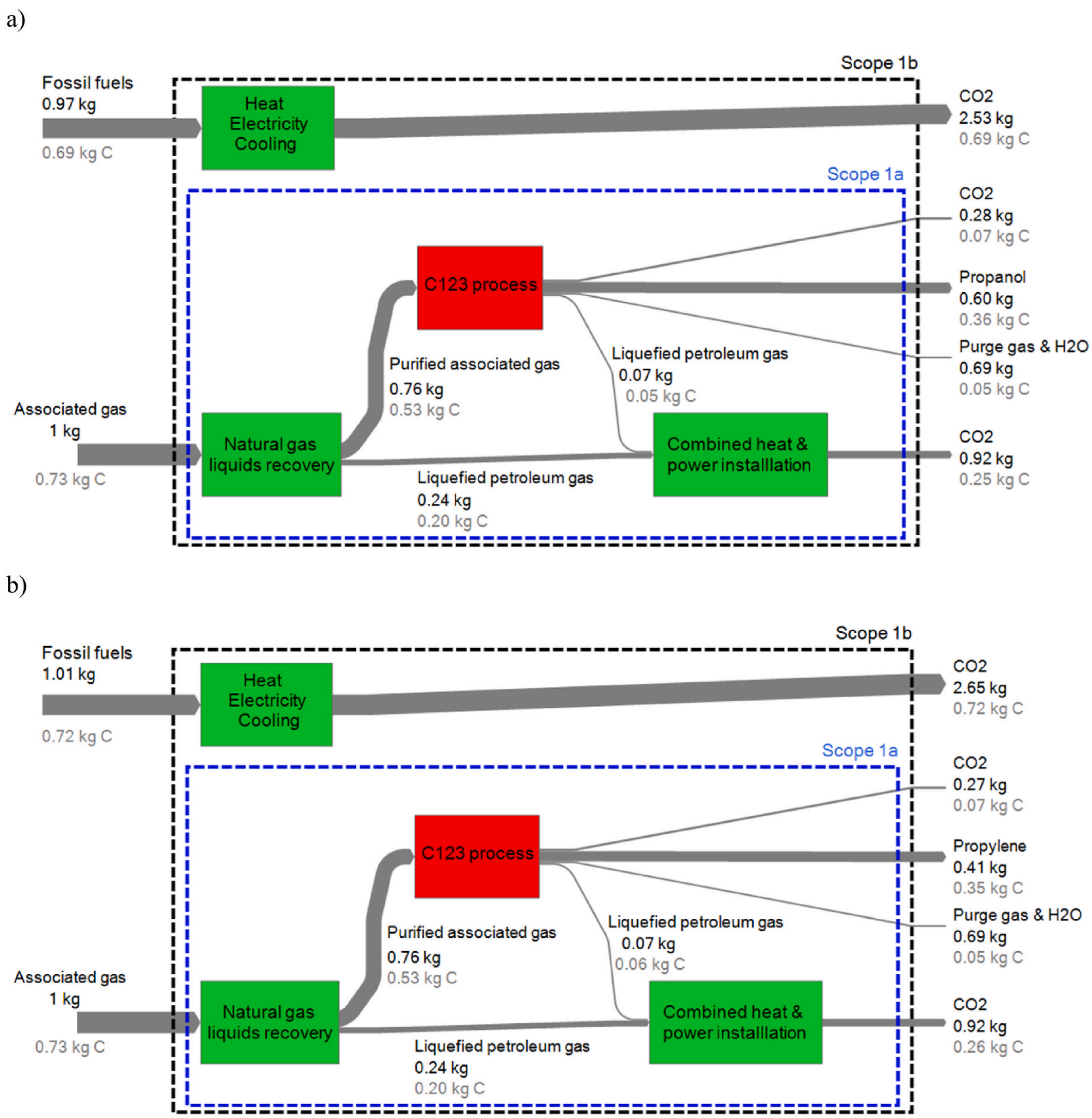

Fig. 4. Sankey diagrams of carbon substance flow analysis for 1-propanol (a) and propylene (b) production from associated gas via the C123 process including scope 1a and 1b. The grey lines represent carbon flows. The wider the flow, the higher the carbon content.

Table 2
Results for carbon efficiency and CO_2 emission savings based on carbon substance flow analysis.

Indicator	Scope 1a		Scope 1b		Expanded system	
	1-Propanol	Propylene	1-Propanol	Propylene	C123 scenario PRL	C123 scenario PRE
Carbon efficiency (%)	0.49	0.48	0.25	0.24	0.37	0.23
CO_2 emission savings compared to flaring (kg CO_2 per kg associated gas)	1.30	1.27	−1.23	−1.38	2.89	−0.93

and disposal were not included as C3 chemicals can be used for a wide range of applications. First, transportation of a liquid product is more favorable than a gaseous molecule due to the smaller volume that must be transported (more cost-effective) (Wood et al., 2012). Therefore, 1-propanol is also the preferred option above propylene. Local transformation of propylene into polypropylene beads (solid material) could be considered to simplify propylene transport, but this requires additional energy and limits its application range.

Second, a big advantage of the C123 process is the carbon retention, avoiding flaring and converting methane into C3 chemicals via several chemical reactions. However, the time frame over which carbon can be stored, depends on the use and disposal phase of the C3 chemical. For example, propylene can be used to produce polypropylene (plastic) which have a long lifetime when this is not burned (Pinaeva and Noskov, 2020; Wei et al., 2021). However, this material must be fully recycled to prevent marine littering and the formation of microplastics after biodegradation (Andrady, 2011). 1-propanol can serve as a solvent in the lab for drug manufacturing, a chemical intermediate or an additive in pharmaceutics (Walther and François, 2016). This chemical has a rather short lifetime in case of lab usage as it is immediately incinerated due to the presence of contaminants (Chea et al., 2019). Further research is needed to analyse all possible applications of 1-propanol and propylene, their disposal and the related impact on the environment.

3.4.2. Impact of location on the analysis

When another location would be chosen for this analysis, the LCA and CSFA results may slightly differ due to the varying associated gas composition worldwide. In Appendix F, the associated gas composition can be found for several locations. For instance, in the south of Iraq, more ethane, propane and butane and less methane is present in the feed compared to e.g., Russia and Tomeslake (USA) (Snytnikov et al., 2018). However, the carbon efficiency of C123 conversion in Russia remains the same and the carbon emission savings only decrease with 0.05 kg CO_2 per kg associated gas both for 1-propanol and propylene. The production of 1-propanol is still preferred based on the LCA results. Nevertheless, a higher value for climate change is observed due to higher greenhouse gas emissions associated to electricity production via a gas turbine in Russia compared to Iraq (4.28 kg CO_2 equivalents versus 3.68 kg CO_2 equivalents per kg associated gas).

The associated gas composition affects the process design and the needed weight for conversion into C3 chemicals. In case of high C_{3+} concentrations, an additional natural gas liquids extraction unit is required to remove this fraction and to prevent the formation of impurities via the C123 process. The more natural gas liquids recovered, the more associated gas must be supplied to meet the needed amount of methane and ethane for C3 chemical production. Furthermore, more energy must be produced from other fossil fuels for associated gas conversion via the C123 process when less natural gas liquids are available from the associated gas at the particular location. Finally, the distance from the oil reservoir, where the associated gas is obtained, to the market and to the nearest cities determines the corresponding transportation cost and impact on human health, respectively. Consequently, a case-by-case approach is highly recommended to find the most suitable associated gas conversion for a specific location.

3.4.3. Alternative associated gas utilisation or conversion technologies

Liquefaction of associated gas could be an alternative for the conversion into chemicals such as 1-propanol and propylene. However, the transportation of LNG is often accompanied with methane leakages (Sun et al., 2020). This should be avoided at all time as methane has a 34 times higher global warming potential (GWP) than CO_2 (IPCC, 2013), while 1-propanol and propylene have no GWP. Furthermore, LNG is mostly utilised for energy purposes (e.g., in transport sector), so carbon is only stored for a short time (Banaszkiewicz et al., 2020). The same holds for NGL as these mostly serve for energy generation (cooking, heating, etc.) (Energy Information Administration, 2012).

Pipeline transportation is only feasible when the distance to the market is lower than 2000 km and when the gas volume is high enough (Khalili-Garakani et al., 2021). Next, reinjection is not interesting from the economic perspective as it will not generate any direct profit unlike 1-propanol and propylene (Khalili-Garakani et al., 2021). The gas is just injected into the reservoir to keep the pressure in the reservoir at the same level. Natural gas hydrates and compressed natural gas technologies are not entirely mature yet and need further consideration (Khalili-Garakani et al., 2021). Finally, gas-to-power is a cost-effective option when used locally. Nevertheless, it is not a good alternative from the environmental point of view due to released greenhouse gases during gas burning (Khalili-Garakani et al., 2022).

Based on the LCA results, the C123 production of 1-propanol and propylene seems an interesting associated gas conversion technology for this location, with a preference for 1-propanol. However, a techno-economic assessment and market analysis must be performed, including CAPEX and OPEX, the price of the final product, etc., to evaluate the economic viability of this technology because this parameter often hinders the big scale implementation of gas-to-chemicals technologies (Khalili-Garakani et al., 2021).

4. Conclusions

This paper evaluates the environmental performance of associated gas conversion into C3 chemicals such as 1-propanol and propylene via the preliminary C123 process. The preferred associated gas conversion (into 1-propanol or propylene) was identified and the C123 performance was compared to flaring from the environmental perspective. Both a CSFA and LCA were applied to assess the C3 chemical production via the C123 process. The CSFA shows that associated gas conversion into C3 products causes more CO_2 emissions than flaring at scope 1b (including local energy production: plus 1.23 and 1.38 kg CO_2 equivalents per kg associated gas for 1-propanol and propylene, respectively, compared to flaring) due to the large amount of CO_2 emissions associated to local energy production. For instance, oxygen gas production requires a substantial amount of electricity. The carbon efficiency amounts to 0.25 both for 1-propanol and propylene production via the C123 process. However, this process must be viewed in a broader context, assuming C3 production via the C123 process can (partially) replace the current C3 production and avoid flaring at the same time. After system expansion, carbon emissions can be saved in C123 scenario PRL compared to the reference (2.89 kg CO_2 per kg associated gas), while C123 scenario PRE causes more CO_2 emissions (plus 0.93 kg CO_2 per kg associated gas).

According to the LCA results, C123 scenario PRL outperforms C123 scenario PRE for all indicators, which means that 1-propanol production via the C123 process would be the best associated gas conversion method among these two options. Only C123 scenario PRL has a significantly lower impact on climate change than the reference. Both scenarios can have a beneficial effect on human health. Moreover, renewable electricity production and heat integration could significantly reduce the environmental impact of the C123 process. The impact on climate change would be halved and a CO_2 emission saving around 1.1 kg CO_2 per kg associated gas can be achieved in both cases compared to flaring. However, a case-by-case approach is highly recommended for the selection of the best associated gas conversion technology as the associated gas composition varies worldwide. Further research includes a techno-economic assessment of C3 chemical production through the C123 process and a cradle-to-grave LCA analysis considering the transport, use and disposal for all possible applications of C3 products.

CRediT authorship contribution statement

Jordy Motte: Data curation, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Pieter Nachtergaele:** Methodology, Supervision, Writing – review & editing. **Mohamed Mahmoud:** Conceptualization, Writing – review & editing. **Hank Vleeming:** Conceptualization, Writing – review & editing. **Joris W. Thybaut:** Funding acquisition, Supervision, Writing – review & editing. **Jeroen Poissonnier:** Writing – review & editing. **Jo Dewulf:** Methodology, Funding acquisition, Supervision, Writing – review & editing.

Declaration of competing interest

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

Data availability

The used data is included in the supplementary material.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.137742.

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