C123

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Methane oxidative conversion and hydroformylation to propylene

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Deliverable Report

VALORISING METHANE RESOURCES

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Description of the deliverable content and purpose

Propylene production is classified as the fourth largest emitter of greenhouse gases among the major chemical compounds. As the polypropylene market is huge and still growing, it is essential to find alternatives to current, energy-intensive production processes to meet the European environmental challenges. Other C3 derivatives, more specifically propanol and propanal, are also very high added-value chemicals with growing markets. They are obtained via waste-generating and energy-consuming processes. Today, unused carbon resources such as biogas and stranded gas are widely available and most of the time wasted. The C123 project's main goal is the validation in a relevant environment (TRL5) of an efficient and selective transformation of these currently generally accessible, unexploited and cheap methane resources (stranded gas (CH_4) and biogas (CH_4+CO_2)) to propylene in particular and C3 products in general.

To this aim, C123 develops new catalytic materials in novel process configurations and related operating procedures allowing the conversion of these resources to propylene through Oxidative Conversion of Methane, leading to an ethylene, carbon monoxide, and hydrogen mixture with an optimized composition for further Hydroformylation to propanal and/or propanol, ultimately being dehydrated into propylene, either in an integrated manner or as a stand-alone step.

C123 adopts an integrated approach, not studying each step separately but considering the process as a whole, optimizing recycling, avoiding separation, using variable feedstocks and increasing resource and carbon efficiency. The process is evaluated and validated for implementation both as decentralized localized units, the modular route (~10 kt C3 product/yr) and in existing large facilities as the add-on route (200 – 500 kt propylene/yr). Throughout the development and thanks to the complementarity of the partners and the very strong industrial commitment, emphasis is put to maximize further exploitation of the results through industrial implementation.

This Deliverable D.5.2 reports on the development and analysis of industrial baseline scenarios for the implementation of the C123. Five different scenarios for implementation of the C123 technology are presented, according to the type and feedstock of the process.

The attractiveness of each of these five scenarios was analysed from the optimal plant location and the environmental and economic impact assessments. This analysis indicated the potential economic benefit of modular C123 for conversion of biogas. A preliminary LCA on the three modular scenarios indicated that the greenhouse gas emissions and human health impact was lowest for the modular, biogas-based route. The modular routes using other natural gas sources (marginal gas and associated gas) suffered in comparison from the need for O_2 production to feed the oxidative conversion of methane step.

Table of Contents

Table of Figures

Table of Tables

1 Introduction

This public report defines the different C123 scenarios and their feedstock sources, provides locations for each of the scenarios and discusses their preliminary environmental impact and economic assessments.

2 C123 Scenarios

In the C123 scenarios, biogas, stranded gas from marginal gas fields, and associated gas are valorized using oxidative conversion of methane (OCoM) and hydroformylation (HF) for the production of propanal and propanol. Dehydration of propanol delivers propene (propylene). Two process designs are envisioned for the C123 process. The first is an add-on process for an industrial production scale of ca. 200 – 500 kt/yr propylene, where the C123-process will be annexed to an existing petrochemical facility, using marginal gas or associated gas as feedstock. The second is a modular process (~10 kt/yr), where the C123-process will be located remotely with a natural gas feedstock from marginal gas fields or associated gas reserves, or a biogas source. From MS11, three C123 scenarios were selected, shown in Figure 1:

- **C123 Scenario A** (Modular process): Biogas to propanal (and propanol)
- **C123 Scenario B** (Modular process): Natural gas from marginal or associated gas sources to propanal (and propanol)
- **C123 Scenario C** (Add-on process): Natural gas from marginal or associated gas sources to propylene

From these three scenarios, Scenario B and C are split into two respective scenarios, namely

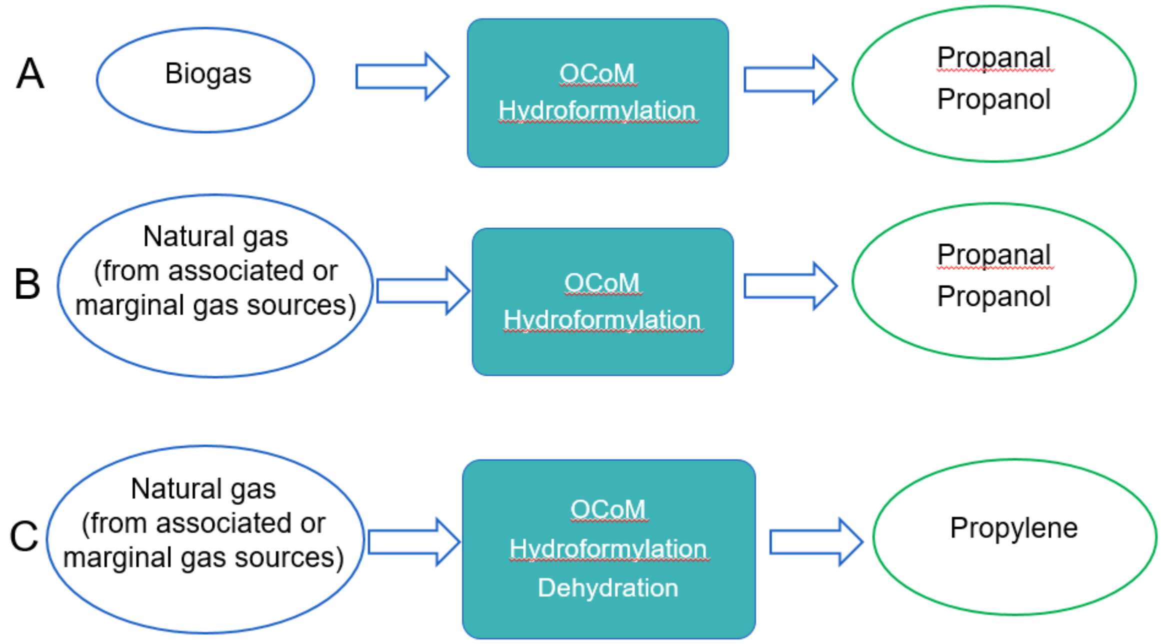


Figure 1: Selection of C123 Scenarios A, B and C.

B1 and B2 and C1 and C2 for using marginal gas and associated gas, respectively.

2.1 C123 feedstock sources

In the text of the original call, the scope of the feedstock was defined as ‘currently unexploited light hydrocarbons resources, biogas or stranded gas, and energy production’. The C123 project summary defines the feedstock as ‘generally accessible, unexploited, cheap methane resources (stranded gas (CH_4) and biogas (CH_4+CO_2))’. To this end, biogas and stranded gas are mentioned as the natural gas feedstocks. The choice of appropriate industrial baseline scenarios (C123 scenarios) requires a definition of these sources.

Biogas is produced by the anaerobic digestion of organic matter such as sewage sludge, cow manure, waste from the agro-food industry and organic fraction of municipal solid waste. It consists primarily of methane (CH_4) and carbon dioxide (CO_2), but it will also vary on the feedstock type and digestion system. The composition of the gas depends on the biogas source, as biogas from landfill sites can also include N_2 and O_2. Also, hydrogen sulfide (H_2S) is usually found in biogas. This compound causes corrosion of installations and therefore it must be removed (by absorption, adsorption or biological conversion into elemental sulfur or sulfate by sulfide oxidizing microorganisms).

Natural gas is a fossil energy source formed deep beneath the earth’s surface and consists primarily of CH_4. Natural gas reserves can be located on-shore or off-shore and can be associated or non-associated (conventional gas fields) with oil. Stranded gas is defined as natural gas that cannot be delivered to market, either for economic or logistical reasons. It is also defined as natural gas that is wasted, unused or under-utilized. Sources of stranded gas are:

- Associated gas reserves. Associated gas is natural gas found in association with oil within an oil reservoir and accounts for 25 % of the worldwide proven reserves of natural gas. [1]
- Deep offshore gas reserves (where access is difficult)
- Marginal gas or remote fields, mall reservoirs that may be distant from markets. Marginal fields have had at least one exploration well drilled and reported, but without any follow-up assessment or development effort for more than 10 years. [2]

Although most stranded gas reserves are completely underdeveloped, it is stated that the C123 feedstock should be generally accessible. Therefore, deep offshore gas reserves and remote fields without extraction infrastructure are excluded from consideration. The stranded gas sources of interest are associated gas and marginal gas fields.

Associated gas is often wasted or under-utilized. As mentioned previously, associated gas is natural gas found in association with oil within an oil reservoir. Associated gas can be wasted or under-utilized by flaring it or using it for low-value applications. Low-value applications may include using it for on-site electricity generation, selling it, or reinjecting it for enhanced oil recovery. High-value applications on the other hand include GTL (gas to liquids) applications or using it as feedstock for the C123 scenario to produce C3-products (propene, propanol and propanal).

Marginal fields can have the following characteristics:

i) Fields not considered by license holders due to limited economic viability;
ii) Fields with at least one exploration well drilled without further development for more than 10 years;
iii) Fields with unfavourable crude oil characteristics;
iv) Fields with high gas and low oil reserves;
v) Fields abandoned by leaseholders for more than 3 years due to operational or economic reasons [2].

Marginal gas reserves account for approximately 15 % of the world's proven gas reserves, of these reserves 20 % is stranded [1]. Marginal gas fields that have existing extraction infrastructure will be considered.

To this end, the three natural gas sources identified for the C123 project are:

1. Biogas (CH_4+CO_2)
2. Associated gas (stranded gas, CH_4)
3. Marginal gas fields (stranded gas, CH_4)

3 C123 scenario locations

The definition of the system boundaries and geographical location of the C123 scenario is important for the techno-economic assessment (e.g. transport considerations, local costs) as well as for an accurate sustainability assessment. The C123 scenarios have been expanded to distinguish Scenario B into B1 and B2, and Scenario C into C1 and C2 depending on the feedstock:

- **Scenario A** (Modular process): Biogas to propanal (and propanol)
- **Scenario B1** (Modular process): Stranded natural gas from a marginal gas reserve to propanal (and propanol)
- **Scenario B2** (Modular process): Stranded natural gas from associated gas to propanal (and propanol)
- **Scenario C1** (Add-on process): Stranded natural gas from a marginal gas source to propylene
- **Scenario C2** (Add-on process): Stranded natural gas from associated gas to propylene

3.1.1 C123 Scenario A

Scenario A utilizes biogas in a modular unit. The selected location is Germany, because it is a big biogas producing country (50% of Europe's biogas production) [3]. Biogas is well spread across Germany; therefore, the specific location will not have a major impact on the attractiveness of the scenario.

3.1.2 Location C123 Scenario B1

Scenario B1 utilizes marginal gas in a modular unit. Many locations are possible in principle, but the selected location is in Russia, where large marginal gas fields are available. The Angara-Lena Terrace basin, indicated in Figure 2 by a shaded red area, can be used for Scenario B1. The closest refinery, owned by Angarskneftorgsintez, is in Angarsk, shown by a black pointer in Figure 2. This refinery has a steam cracker with a capacity of 95,000 and 240,000 metric tonnes per year, respectively (data from the year 2000).

3.1.3 Location C123 Scenario B2

Scenario B2 utilizes associated gas in a modular unit. Two location options have been identified for Scenario B2. The first is in North Dakota (USA) due to the high concentration of flaring in this remote region (Figure 3). There are two spots specifically, 8 km apart, that flare 30 million m^3/yr. This is the preferred location as first calculations show that the natural gas available is sufficient for a production facility producing approximately 10 kt/yr of propanal or propanol. If this cannot be sustained, Russia, where large amounts of flaring are taking place at highly remote areas (Figure 4), is considered as a backup location.

3.1.4 Location C123 Scenario C1

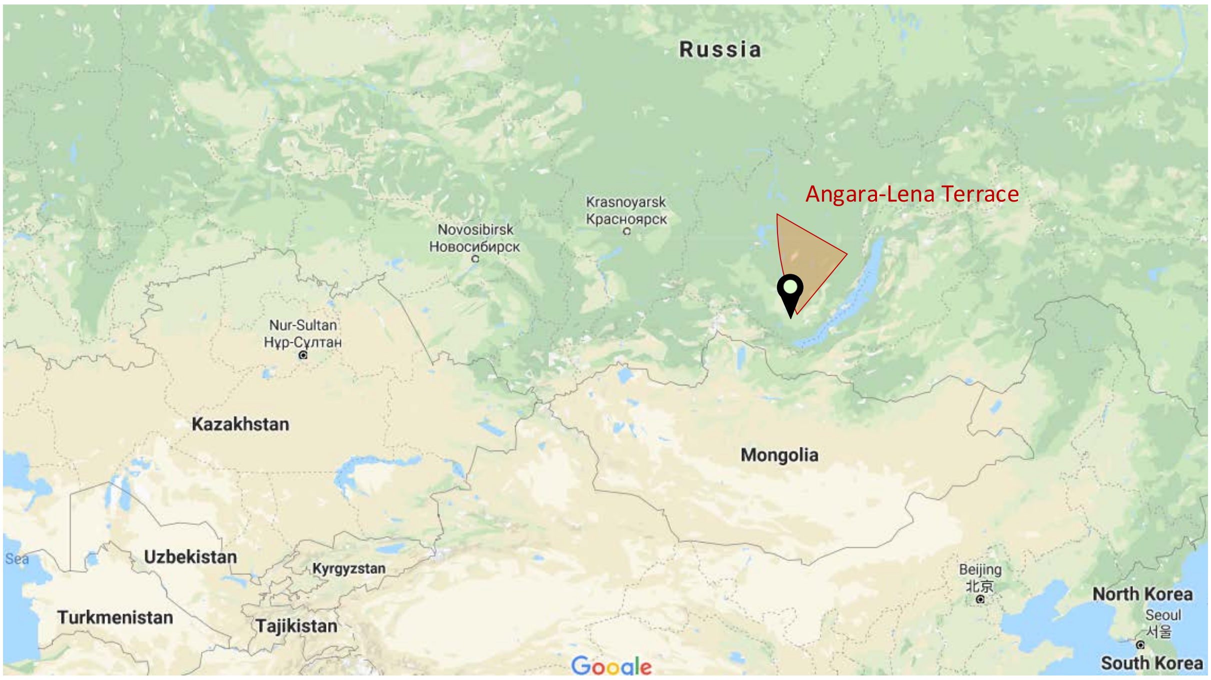


Figure 2: Geographic location of the Angara-Lena Terrace stranded natural gas field in red, and the town Angarsk. indicated by a black pointer (Redrawn from [4]).

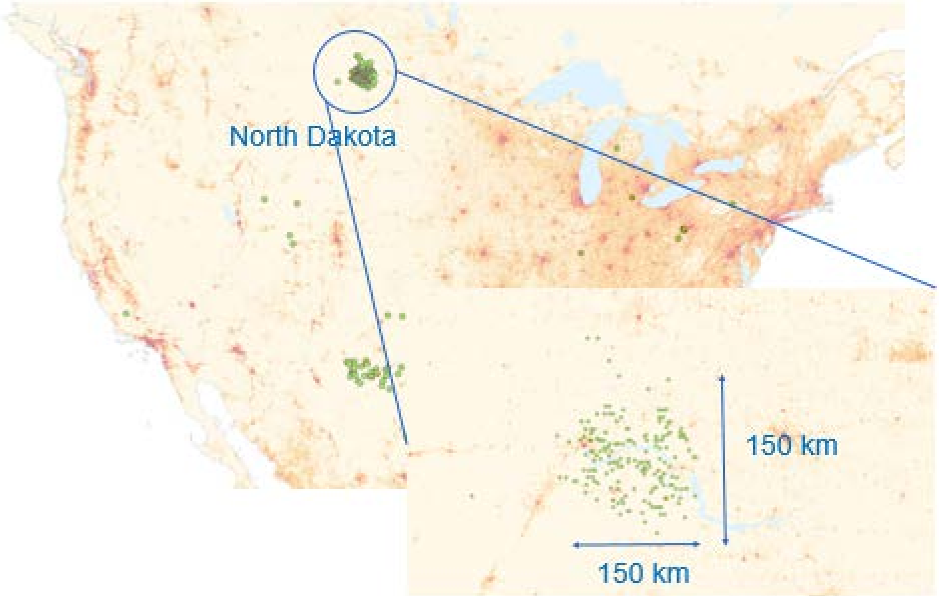


Figure 3: Flaring in North Dakota where each green spot represents a flare. The colour of the map indicates the population density. The darker the colour, the higher the population density.

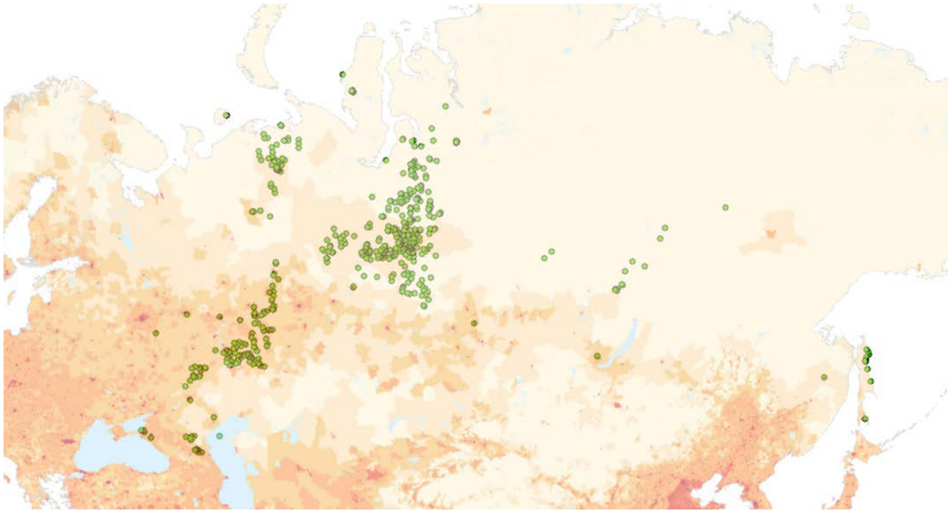


Figure 4: Flaring in Russia where each green spot represents a flare. The colour of the map indicates the population density. The darker the colour, the higher the population density.

Scenario C1 utilizes marginal gas in a large-scale add-on unit. For this scenario, the Absheron gas field in the South Caspian Basin close to Azerbaijan has been selected (Figure 5). It is estimated to contain 350 billion m^3 of gas and 45 tonnes of gas condensate, and it covers an area of 270 km^2, 500 m under water [5,6]. Partners of the Absheron field are Total S.A. (40 %), SOCAR (40 %) and GDF Suez (20 %). It is operated by Total and is located approximately 100 km from Azerbaijan capital Baku where C123 partner ANAS (Azerbaijan National Academy of Sciences) is located. Currently, Total plans to start production of gas to supply to the domestic market in 2021. Since the extraction of the gas is underway, the C123 modular unit can be positioned well to valorise the gas from this basin.

3.1.5 Location C123 Scenario C2

Figure 5: Location of marginal gas resource Absheron in the Caspian Sea close to Azerbaijan.

Scenario C2 utilizes associated gas in a large-scale add-on unit. For this scenario, three locations have been identified and evaluated: Venezuela, the Middle East (Iraq) and Russia (Figure 6). These three locations were compared on the total gas flaring emissions, estimated carbon dioxide emissions, proven oil reserves, oil production rate, remaining operating years and near-by existing petrochemical refineries (Table 1). Due to the proven oil reserves and close proximity of Bandar Imam and Kuwait petrochemical complexes, the Middle East was selected as the preferred location for Scenario C2 (Figure 7). Although Venezuela is currently economically and politically instable, this scenario could also be applied to this country in the future, due to the similar gas flaring emissions (1.74 and 1.83 billion m^3/yr), and thus plant capacities, and distance to existing infrastructure (130 km and 150 km).

Figure 6: Locations with the largest gas flaring emissions on a yearly basis indicated with the blue circles (0.2 - 1 billion m^3 per year): Venezuela, Iraq and Russia.

Table 1: Comparison table for selection of location C123 Scenario C2.

Possible locations for Scenario C2	Venezuela	Iraq	Russia
Gas flaring emission (billion m^3/yr) [7]	1.83 (2 spots)	1.74 (4 spots)	Spot 1: 1.0 Spot 2: 0.58
Estimated CO_2 emissions by flaring (Mt/yr)	3.88 (2 spots)	3.68 (4 spots)	Spot 1: 2.12 Spot 2: 1.22
Proven oil reserves	Big oil reserves	4.5 billion barrels [8]	1.5 billion barrels [9]
Oil production rate Years remaining	Not known	360 000 barrels/d [8] ~ 34 years (Zubair oil field)	442 000 barrels/d [9] ~ 9 years (Spot 1: Vankor oil field)
Existing petrochemical refineries or operations	Puerto La Cruz refinery located 130 km away	Bandar Imam Petrochemical complex located 160 km away in Iran OR Kuwait petrochemical complex located 150 km away in Iraq	No existing petrochemical refineries/operations close by. Closest is Angarskneftorgsintez refinery, located 900 km from gas flare 2

Figure 7: Middle East location for C123 Scenario C2, with distances indicated from the flared emissions locations (4 flares) to the closest petrochemical refineries.

4 Environmental impact assessment

In this section, the C123 scenarios will be evaluated from the environmental perspective. A preliminary LCA is done for the modular C123 Scenarios A, B1 and B2. First, the goal, scope and functional unit of the LCA will be discussed. Afterwards, an overview will be given with all inputs and outputs of the C123 scenarios together with their source or destination and with the associated data collection. The assumptions that were made for this assessment will also be described. Next, some preliminary results such as the greenhouse gas emissions or water consumption related to these scenarios will be discussed. Finally, possible improvements of the process design will be proposed. The LCA for the add-on scenarios is not performed yet, but environmental context and perspectives for these scenarios are given. While sustainability assessments are often compared with reference scenarios (conventional technologies), the necessary data on the reference scenarios are not yet available.

4.1 Goal, scope and functional unit of the LCA

The main goal of the LCA is to determine whether the production of the C3 chemicals (propylene, propanal and propanol) via the C123-technology is more environmentally friendly than the reference cases. The preliminary assessment in this document aims to give some preliminary results, to identify which steps in the process design contribute the most to the emissions, e. g. greenhouse gas emissions, to propose improvements for the process design and to get feedback for some assumptions that are made in this analysis.

The LCA is performed from cradle-to-gate. This means that the use and the disposal of the final product are not considered in the assessment. All production steps that precede the C123 production steps are included in the analysis (e.g. production of biogas) since these are part of the life cycle of the final product, e.g. propanol or propylene. The functional unit for this preliminary assessment is 1 kg of propanol (in case of Scenarios A, B1 and B2). So, all data and all preliminary results are presented in function of 1 kg of propanol.

4.2 Data collection and assumptions

4.2.1 C123 Scenario A

In Figure 8, a simplified version of the process design for C123 Scenario A is represented. This scheme consists of five major production steps: the upgrade or purification of the biogas, OCoM to produce a mixture of C_2H_4, CO and H_2, the post-treatment of the OCoM step (removal of water), HF to produce propanal and finally, the post-treatment of the HF step where propanal is converted to propanol and the propanal/propanol mixture is separated via distillation. All inputs and outputs are shown outside the green box. The data for the red boxes in Figure 8 is retrieved from an Aspen simulation by PDC, the data for the production of the inputs and the emissions related to the C123 technology are collected via the literature or via the Eco-invent database (present in the LCA software). In Table 2, the source, the technology and the location (internal or external) of the production of the inputs and the destination of the outputs are presented. The references for the data collection are included as well. Bio-

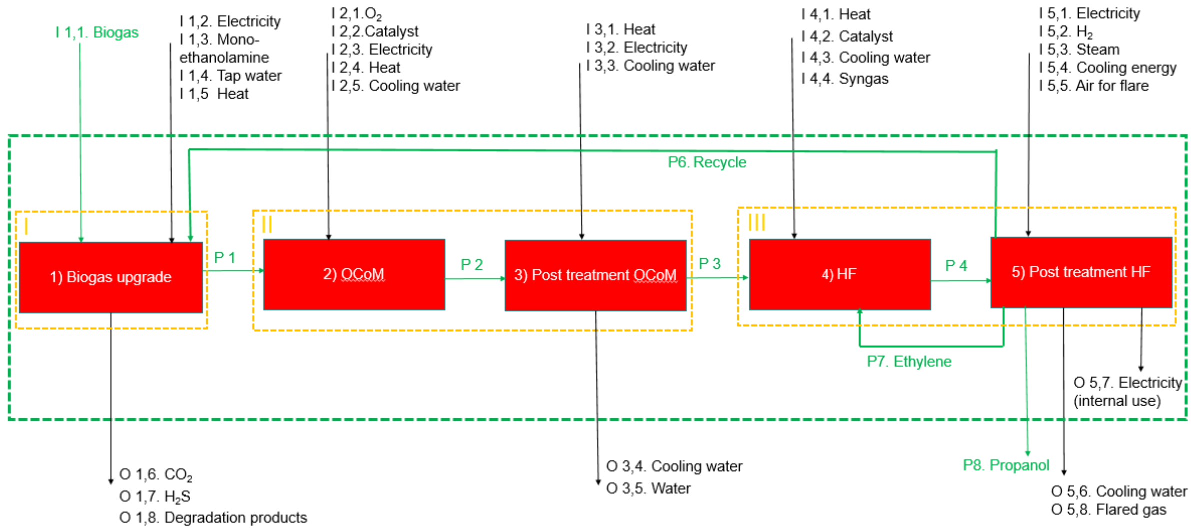


Figure 8: Simplified process design of C123 Scenario A used for LCA analysis. The red boxes represent the five major steps to produce propanol. The inputs (I) and outputs (O) are also mentioned.

based inputs are selected in this scenario where appropriate, to model a full renewables-based value chain.

In the next paragraphs, the assumptions that are made for C123 Scenario A are discussed, starting from the production of the biogas. This resource is produced on-site via an anaerobic digestion of maize silage and cattle manure. These feedstocks are usually co-digested in Germany [10]. The feed consists of 70% maize silage and 30% cattle manure [11]. A co-digestion has typically a higher efficiency than a mono-digestion of both feedstocks. However, it is assumed that both feedstocks will be digested separately because of lack of data for this specific co- digestion [11]. The manure is seen as burden-free input if the storage is not taken into consideration, so the upstream processing related to the milk or meat production is not considered, like in most LCA studies for biogas production from manure [12]. A pre-treatment of energy crops is necessary to prepare a pulp that enters the anaerobic digestion. In this scenario, the chemical treatment of corn silage with sodium hydroxide (NaOH) is selected since this is the most common method [15,16]. Before the feedstock is sent to the anaerobic digestor, it is pasteurized at 70°C for one hour to kill bacteria (pathogens) that would disturb the micro-organisms [17]. After the anaerobic digestion, 0.51 kg biogas and 9.75 kg digestate are obtained from 1.56 kg of maize silage and 4.74 kg manure [18]. The digestate is afterwards used as a fertilizer. Therefore, in the LCA, the avoided burden of the digestate is incorporated as the avoided production of three synthetic fertilizers (55% of weight as N, 20% as P_2O_5 and 25% as K_2O) [18]. The digestate is mostly stored before it is transported; the emissions related with open-air storage are taken into account.

In the next step, the produced biogas is purified by chemical scrubbing. CO_2 and H_2S are removed, and a purer CH_4 stream is obtained. In addition, the removal of H_2S is necessary to avoid corrosion of the installations [19]. Chemical scrubbing is often the preferred technique

Table 2: Summary of data collection for C123 Scenario A. The source of the inputs, the selected technology and the location of the production are highlighted. The destination of the outputs is mentioned as well. Finally, it is shown from which reference the data is collected. Eco-invent is the database in the LCA software that can be used to collect data.

Inputs	Source and technology	Internal/external production	Data Reference
Feedstock			
Biogas	Anaerobic digestion of maize silage and manure	Internal	[12]
Chemicals			
O_2	Cryogenic air separation	Internal	Eco-invent
MEA[a]	Production from ethylene oxide and ammonia	External	Eco-invent
Syngas	Production from wood in fluidized bed gasifier	Internal	Eco-invent
H_2	Electrolysis of water (electricity supplied via wind turbine)	Internal	[13]
Utilities			
Electricity	From German electricity grid	External	Eco-invent
Cooling water	From natural origin	Water from natural origin close to plant	Eco-invent
Cooling energy	With refrigerant and absorption chiller	Internal	Eco-invent
Tap water	From local water grid	External	Eco-invent
Heat	From natural gas in boiler	Internal	Eco-invent
Steam	From natural gas in boiler	Internal	Eco-invent
Air for flare	Air taken from environment	Internal	Own calculation

Outputs	Destination	Internal/external valorization	Data reference
Product			
Propanol	Market	-	Simulation PDC
By-product			
Electricity	Internal use	-	Simulation PDC
Emissions			
CO_2	Air emission	-	Simulation PDC
H_2S	Air emission	-	[14]
Degradation products	Air emission	-	[14]
Cooling water	Water emission	-	Simulation PDC
Flared gas	Air emission	-	Simulation PDC/ own calculation
Water	Water emission	-	Simulation PDC

[a] Monoethanolamine

for biogas upgrading because membrane technologies such as PSA lead to CH_4 loss [20].

Monoethanolamine (MEA) is the selected solvent that will form a chemical bond with CO_2 and H_2S in the biogas [14]. Afterwards, heat (from natural gas in boiler) is supplied to regenerate the solvent (CO_2 and H_2S are again released). However, the emission of H_2S should be prevented by using this chemical for the production of sulphur, which has several applications in the chemical industry, because H_2S has a negative impact on human health. Carbon capture and storage (CCS) or carbon capture and utilization (CCU) could also avoid the emission of CO_2 [14]. Both options are not considered in this study, but these can be further studied in the future. Other emissions that are taken into account are degradation products such as NH_3 and acetaldehyde which are formed during the regeneration of the solvent. Finally, tap water and electricity from the German grid are needed for the biogas upgrade.

Before the upgraded biogas is sent to the OCoM reactor(s), it is preheated until 350 °C. Pure O_2 is supplied to the reactor to react with the CH_4 in the biogas feed. It is considered (for now) that the pure O_2 is produced via a cryogenic air separation, and further preheated to 550 °C. Overall, cooling is needed to control the combined reactions that take place during OCoM. This assumption can change after additional studies. The catalyst production or regeneration for OCoM is not included in the current LCA study. The heat is always produced in a boiler with natural gas as fuel. Afterwards, water that is formed in the OCoM reaction is removed via a decanter, a multi-compression system and a water trap. 505.9 kg of cooling water is supplied to decrease the temperature after OCoM and 0.68 MJ of electricity from German electricity grid is needed for the compression.

Before the HF step, make-up CO and H_2 (syngas) is added to the C_2H_4 stream and both are heated up to 200 °C. The heat is delivered via an industrial furnace with light fuel oil and the syngas is produced from wood chips in a fluidized bed gasifier that is located on the plant [14]. The wood chips originate from German wood species oak and beech. Thereafter, propanal is produced via the exothermic HF reaction. Cooling water is used to keep the temperature constant. Next, other components such as CH_4 and C_2H_6 are separated from propanal in the first distillation column. A refrigerant cool down the upper fraction in the condenser, steam heats up the lower fraction in the reboiler.

To obtain propanol, propanal is hydrogenated with H_2. It is assumed that H_2 is produced on-site via water electrolysis. Water is deionized before electrolysis to avoid corrosion by salts and fouling of the installation. A small amount of KOH is added to the water and serves as an electrolyte. A high amount of renewable based electricity (17.56 MJ for 1 m^3 of H_2) is needed to split the water in H_2 and O_2 [13]. An efficiency of 80 % is considered for this reaction. O_2 is vented in the atmosphere [21]. After hydrogenation, the unconverted propanal must be removed from the product stream via an additional distillation. 4 MJ of cooling energy is needed in total for the first and the second distillations. 0.85 kg of steam is needed for both distillations and the preheating of the product stream before the last distillation. The purge gas, which consists of CH_4, C_2H_6 and CO_2 and which is not recycled, is sent to a flare. Additional air is supplied for the combustion reaction. The corresponding emissions are taken into account in the LCA. Further research can look into heat recovery from the combustion of the purge gas as a better option. Catalysts for the HF and hydrogenation are again not taken into account in this study. For all inputs that are produced outside the plant, transportation is also considered.

Finally, a turbine is positioned between the two heat exchangers of the OCoM post-treatment to generate electricity from the recycle stream of the HF post treatment. In this way, 0.13 MJ of electricity could be produced for local use.

4.2.2 C123 Scenarios B1 and B2

In Figure 9, a simplified version of the process design for C123 Scenarios B1 and B2 is represented. This scheme again consists of five major production steps: the pre-treatment of the feedstocks, OCoM, post-treatment of OCoM (removal of water), HF to produce propanal and finally, post-treatment of HF to obtain propanol. All inputs and the outputs are shown outside the green box. In Table 3, a summary of the data collection and the assumptions for C123 Scenarios B1 and B2 is shown. A big part of these scenarios is analogous to case A, and only the deviating assumptions between these scenarios will be discussed in the next paragraphs.

In the first step, the extraction of the feedstocks (marginal gas for case B1 and associated gas for case B2) is considered. The extraction from marginal gas consists of several phases. First, the well is installed by drilling of the well and the installation of the casing [13]. The second phase is the well completion whereby the well is further developed [13]. When the well is operational, liquid unloading occurs to remove water and condensates in the well which hinder the flow of natural gas [13]. The related greenhouse gas emissions and the water use of these phases are taken into account. Fugitive emissions from equipment and emissions caused by venting and flaring are also included. In Table 4, the greenhouse gas emissions and

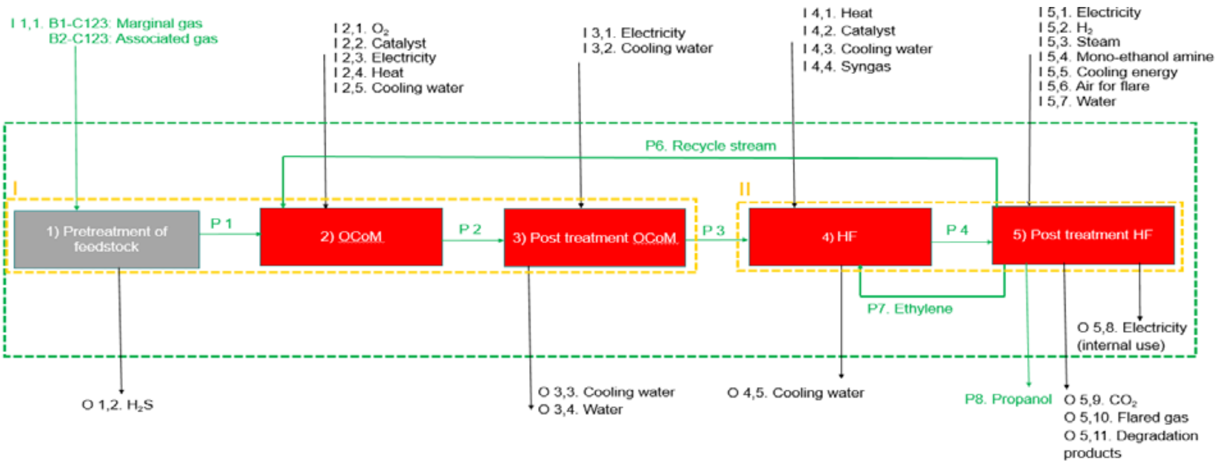


Figure 9: Simplified process design of C123 Scenarios B1 and B2. The red boxes represent the five major steps to produce propanol. The inputs (I) and outputs (O) are also mentioned. The box for the pretreatment of the feedstocks is coloured grey, because this step is not included at this moment. This pretreatment will be considered in the future dependent on the detailed composition of the feedstocks.

Table 3: Summary of data collection for C123 Scenarios B1 and B2. The source of the inputs, the selected technology and the location of the production are highlighted. The destination of the outputs is mentioned as well. Finally, it is shown from which reference the data is collected. Eco-invent is the database in the LCA software that can be used to collect data.

Inputs	Source and technology	Internal/external production	Data reference
Feedstock			
B1-C123[a] B2-C123[b]	Extraction from marginal gas/oil reservoir	Internal or close to plant	[22]
Chemicals			
O_2	Cryogenic air separation	Internal	Eco-invent
MEA[c]	Production from ethylene oxide and ammonia	External	Eco-invent
Syngas	Production from coal via gasification	Internal	[23]
H_2	Via steam reforming of methane	Internal	Eco-invent
Utilities			
Electricity	Production from natural gas in conventional power plant	Internal	Eco-invent
Cooling water	From natural origin	Water from natural origin close to plant	Eco-invent
Cooling energy	With refrigerant and absorption chiller	Internal	Eco-invent
Water	From natural origin	Water from natural origin close to plant	Eco-invent
Heat	From natural gas in boiler	Internal	Eco-invent
Steam	From natural gas in boiler	Internal	Eco-invent
Air for flaring	Air taken from environment	Internal	Own calculation

Outputs	Destination	Internal/external valorization	Data reference
Product			
Propanol	Market	-	Simulation PDC
By-product			
Electricity	Intern use	-	Simulation PDC
Emissions			
CO_2	Air emission	-	Simulation PDC
Degradation products	Air emission	-	[24]
Cooling water	Water emission	-	Simulation PDC
Flared gas	Air emission	-	Simulation PDC/ own calculation
Water	Air emission	-	Simulation PDC

[a] Marginal gas. [b] Associated gas. [c] Monoethanolamine.

the water use and discharge are presented per kg of propanol produced [13]. In general, it can

Table 4: Greenhouse gas emissions and water use and discharge related to the extraction of marginal gas onshore (in function of 1 kg of propanol) [13].

Natural gas type	Marginal gas onshore
CO_2-emission (kg)	0.25
N_2O-emission (kg)	5.89E-06
CH_4-emission (kg)	1.84E-02
Water use (kg)	1.38
Water discharge (kg)	1.2

be assumed that the extraction of marginal gas has lower greenhouse gas emissions compared to conventional natural gas, because the efficiency is higher [13]. Conventional gas reservoirs are operational for a longer time, and the efficiency lowers over time. However, the data collection for the marginal gas extraction must be expanded with other materials/energy requirements beside the greenhouse gas emissions and the water use to have a more accurate result. The input of industrial partners can be helpful. The emissions of the associated gas extraction are not taken into account, because it is assumed that it is just a waste fraction of the oil extraction (burden-free input).

The pre-treatment of the feedstocks is not included (90 mol % CH_4 and 10 mol % CO_2 taken as inlet). However, this can be considered in the future, dependent on the detailed composition of the feedstock. If H_2S is present in the feed, this component must be removed to avoid corrosion of the installations and to protect catalysts. Hence, a detailed composition of the feed is important for an accurate LCA.

The assumptions for the OCoM step and its post-treatment are analogous to C123 Scenario A. The only difference is the supply of electricity and water. In C123 Scenarios B1 and B2, the electricity is produced on-site via a conventional power plant with natural gas as feedstock. Water is always retrieved from a natural source e.g. a river. It is assumed that the plant is located on a remoted place where no electricity or water can be supplied via the electricity or water grid.

In the HF reaction, additional CO and H_2 is delivered in the form of syngas. This chemical mixture is produced from hard coal via gasification because it is an abundant feedstock in Russia and the USA, and it is compatible with the current production technology [22]. 1.43 kg coal is necessary to obtain 1 kg of syngas [23]. Pure O_2 and steam are supplied to the gasifier for the conversion of the carbon to CO and H_2 [23]. Also, nitrogen gas is used as a scavenging gas to purify the syngas after gasification. Consequently, H_2S and CO_2 are emitted by this purification [24]. However, H_2S can be converted into sulphur. This conversion will be further studied and included in an updated version of the LCA. O_2 and N_2 are produced in an air separation unit. The on-site generation of steam, the transportation of coal, the need of electricity from the local electricity grid are also included in this study [24]. Finally, it is assumed that the remaining ash from the gasification is landfilled. All background data is taken from the Eco-invent database.

The H_2 needed for the hydrogenation of propanal to propanol is obtained via steam reforming of methane in Scenarios B1 and B2. This option is selected to have a complete fossil-based case. This choice can be adapted if necessary. The data collection is analogous to that for Scenario A. Catalysts for HF and hydrogenation are again not taken into account in this study.

Finally, electricity is generated, and CO_2 is removed from the recycle stream of the HF post treatment. A turbine is positioned between the two heat exchangers of the OCoM post-treatment to generate electricity from this recycle stream. In this way, 0.45 MJ of electricity could be produced for local use. Chemical scrubbing with MEA as solvent is again selected as the preferred method to remove CO_2. The solvent is regenerated via heating. CO_2 and degradation products such as acetaldehyde and ammonia (formed due to the supply of heat during the regeneration) are released in the atmosphere. CCS and CCU are again not implemented in this scenario. Finally, the transportation of all inputs that are not located on-site is included.

4.3 Impact assessment method

ReCiPe 2016 is selected as impact assessment method both from the midpoint and endpoint level. At the midpoint level, indicators are chosen in the middle of the cause-effect chain, e.g. emission of greenhouse gases for global warming. At the endpoint level, the final damage on the area of protections is measured, namely human health, ecosystems and resources. Hierarchist (H) is selected as the cultural perspective for this LCA. This means that a timeframe of 100 years is used for global warming. The chosen midpoint indicators for the representation of the results are global warming, ozone depletion, human toxicity, particulate matter formation, ionizing radiation, photochemical ozone formation and water consumption. Human health, natural environment and natural resources are the selected endpoint indicators. This first indicator is expressed in DALYs or the number of years that a person loses because of illness caused by human practices or industrial activities. In the next section, the results for global warming, water consumption and human health will be discussed. The other results can be consulted in the Appendix. For this document, a first set of indicators is selected to present the results, but this set can be expanded later on in the project.

4.4 Preliminary results

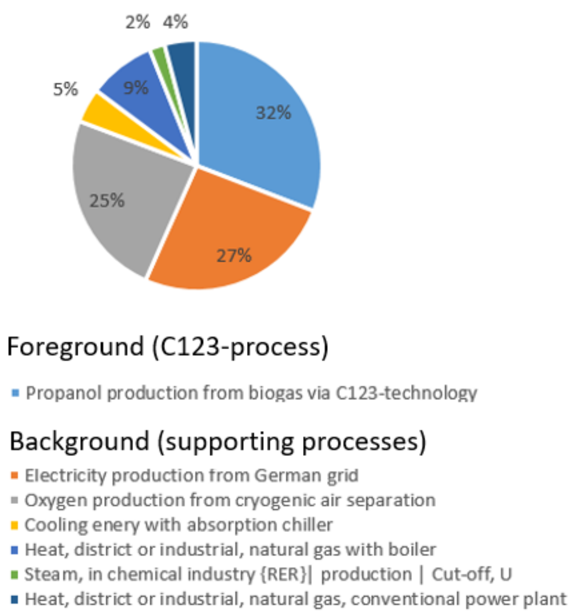
4.4.1 Preliminary results C123 Scenario A

The production of propanol from biogas causes an emission of 5.51 kg CO_2-equivalents per kg of propanol. In Figure 10, the main contributors for these greenhouse gas emissions are presented. Approximately 34 % of the emissions are caused by the electricity production. This can be explained by the high electricity demand for the production of biogas via anaerobic digestion. The production of pure O_2 via air separation also has a high contribution (around 22

%). The greenhouse gas emissions for the conversion of biogas to propanol are not taken into account since these emissions originate from biogenic resources.

The C123 technology in Scenario A consumes 0.064 m^3 of water. After comparison with the reference technologies, it can be decided if this water use is far too high or acceptable. In the first case, recycling of water can be integrated in the process design.

The total impact on human health amounts to 1.01 x 10^{-5} DALYs per kg propanol. In Figure 11, the main contributors are again shown. The production of propanol from biogas via the C123 technology has the highest contribution (32 %). The production of electricity and oxygen also causes a high amount of DALYs (27 and 25 % respectively). This can be explained by the high scores for particulate matter formation and human carcinogenic toxicity of these two inputs. The production of propanol via the C123 technology also causes 32 % of the DALYs by the upgrading of biogas. H_2S and degradation products are emitted by the regeneration of MEA. These compounds have a negative impact on human health (high score for human toxicity).



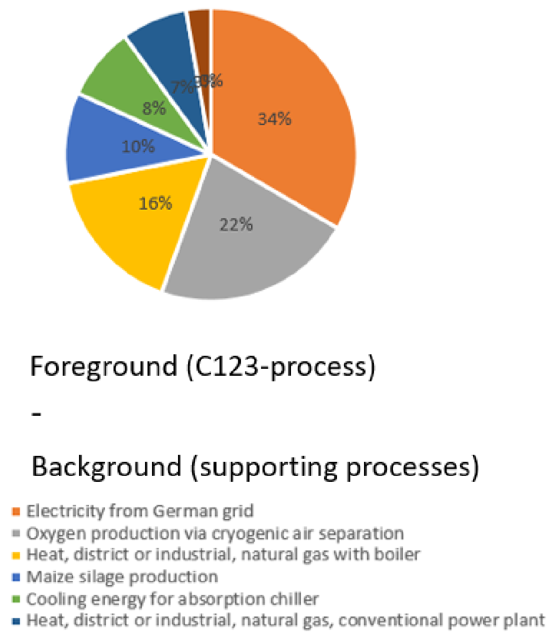


Figure 10: Representation of the main contributors to the total greenhouse gas emissions (in CO_2-equivalents) for C123 Scenario A. The avoided emissions of the synthetic fertilizer production are not included in these results.

Figure 11: Representation of the main contributors to the damage on human health (in DALYs) for C123 Scenario A. The avoided emissions of the synthetic fertilizer production are not included in these results.

4.4.2 Preliminary results C123 Scenarios B1 and B2

The associated greenhouse gas emissions for the production of 1 kg propanol from marginal gas amounts to 9.10 kg CO_2-equivalents. Figure 12 shows which factors influence this result the most. This time, the production of O_2 via cryogenic air separation causes the highest emission of greenhouse gases (35 %). The C123 technology emits 20 % of the total amount.

This emission is linked with the removal of CO_2 from the gas stream. In the production of syngas from coal, a significant amount of CO_2 is also emitted. Finally, the extraction of marginal gas is accompanied with emissions due to its operations (see Table 4). In scenario B1, 0.083 m^3 of water is consumed.

The total impact on human health amounts to 1.45 x 10^{-5} DALYs per kg propanol. This result is higher than for Scenario A. In Figure 13, the main contributors are represented. The production of pure O_2 has the highest share (53 %) followed by the production of propanol via the C123-technology (12 %) and the production of steam (9 %). The impact of the O_2 production is higher than in Scenario A because a higher amount of pure O_2 must be supplied to the OCoM reactor and pure O_2 is needed to produce syngas. The O_2 production also has a much higher score for particulate matter formation and human carcinogenic toxicity then the other processes in Figure 13.

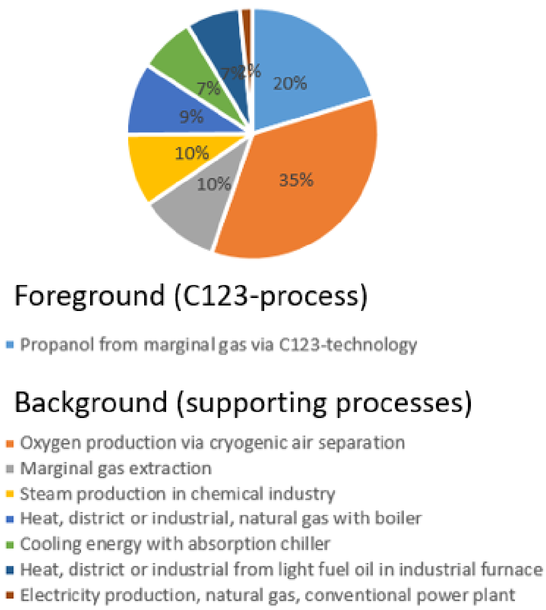


Figure 12: Representation of the main contributors to the total greenhouse gas emissions (in CO_2-equivalents) for C123 Scenario B1.

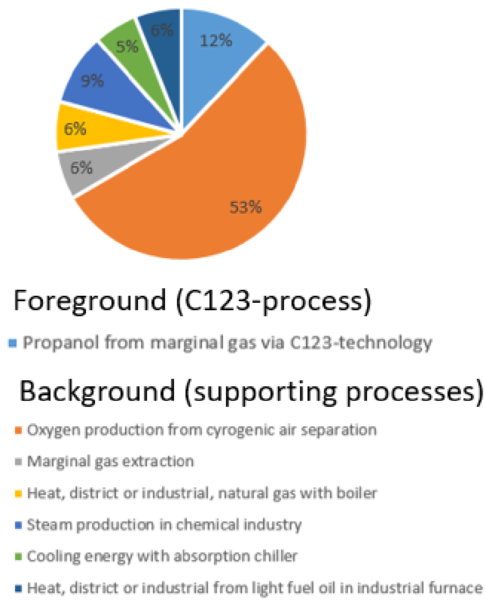


Figure 13: Representation of the main contributors to the damage on human health (in DALYs) for C123 Scenario B1.

In Scenario B2, the analysis is done for two locations, Russia and the USA. If associated gas is selected as feedstock, the total greenhouse gas emissions related to the production of propanol are higher than for Scenario A, but lower than for Scenario B1 namely 8.19 kg CO_2-equivalents in Russia and 8.07 kg CO_2-equivalents in the USA, per kg propanol. The difference between the two results is quite small. It can be explained by the higher amount of greenhouse gases emitted during hard coal production in Russia (compared to the USA). The cryogenic air separation for the production of O_2 again contributes the most to the amount of emitted greenhouse gases, independent of the location. The water consumption in Scenario B2 amounts to 0.0829 and 0.0827 m^3 for Russia and the USA, respectively. These values are approximately the same as for Scenario B1, but lower than for Scenario A. Finally, Scenario B2 causes 1.36 x 10^{-5} DALYs in Russia and 1.35 x 10^{-5} DALYs in the USA. These results are a little bit smaller compared to Scenario B1. The cryogenic air separation causes 53 % of the total DALYs, while this is only 12 % for the conversion of marginal gas to propanol (caused by the removal of CO_2). At this moment, the results between Scenarios B1 and B2 are similar, but the data collection for the extraction of marginal gas must be more complete before a final conclusion can be made.

Aspects that need further attention include:

- Detailed compositions of marginal gas and associated gas including potentially required pre-treatment of the natural gas. Specific attention must be paid to H_2S, because of corrosion issues and potential impact on the catalyst performance.
- Recovery of H_2S from air emission currently included in Scenario A. At this moment, H_2S is considered as an air emission. The release of this harmful compound can be avoided, e.g. by the conversion of H_2S into sulphur.
- Update with the latest OCoM and HF findings and performance results achieved in C123.
- Alternative O_2 production method with lower impact on greenhouse gas emissions and the impact on human health. The current analysis indicates that the production of pure O_2 via cryogenic air separation has a high impact on the final results. The alternative production method should alignment with the process design and local conditions (remote area).
- The remaining gas is flared in the current design. Heat can be recovered from this combustion reaction.
- The water footprint can be lowered by recycling.
- The CO_2 emissions are partially caused by the removal of CO_2 from the recycle stream in Scenarios B1 and B2. A solution to avoid this emission can be implemented in the process design.

4.5 Environmental context for add-on scenarios

The environmental performance analyses for the add-on scenarios (Scenarios C1 and C2), where propylene is produced, are not performed yet, but the environmental benefit that these scenarios can deliver will be highlighted. The current production processes for propylene are energy intensive and these emit a large amount of CO_2 [25]. The C123 technology can be a more sustainable alternative. Marginal gas and associated gas are the selected feedstocks for Scenarios C1 and C2. This last feedstock is today often flared. A world map with detected flaring sites in 2012 is shown in Figure 14 [26]. There is an effort to decrease flaring, because it negatively affects the local air quality and releases CO_2, significantly impacting global warming. Consequently, the application of C123 technology for associated gas from avoided flaring can reduce greenhouse gas emissions and emissions of harmful components for humans such as NOx, CO, SO_2, etc., contributing to a more

Figure 14: World map with red dots indicating the gas flaring spots. (KLM-file in Google Earth [26].)

sustainable process and lower environmental impact.

5 Economic assessment

The first C3 products produced from the HF of C_2H_4 are *n*-propanol and propanal (propionaldehyde). For the current modular designs, the propanal produced is hydrogenated to *n*-propanol and purified to 99.5 mol %. Propanal can also be used to produce propionic acid through oxidation or trimethylolethane through condensation, as summarized in Figure 15.

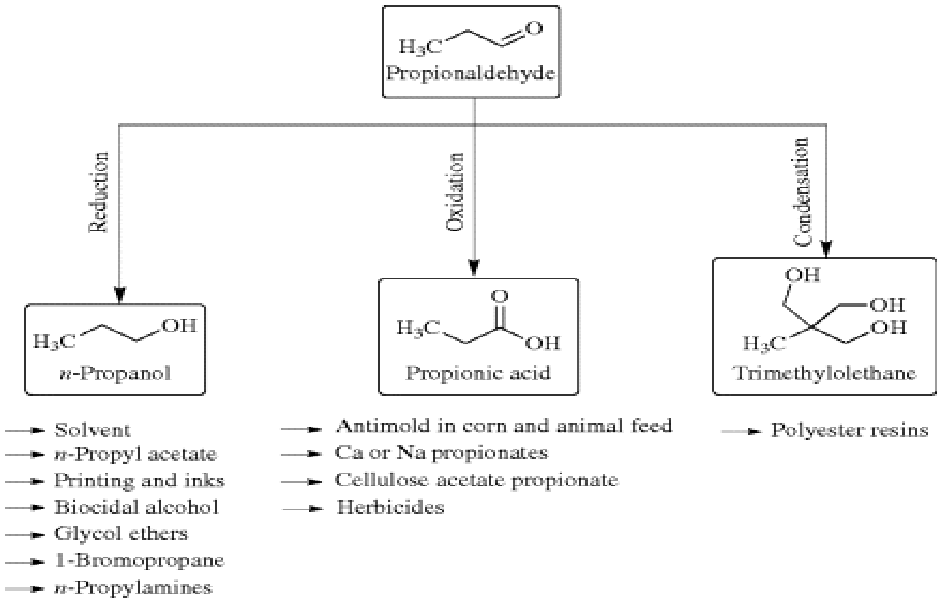


Figure 15: Chemicals that can be obtained from propanal (propionaldehyde) and their uses.

5.1 Economies of scale

The concept of economies of scale refers to the effect where an increase in production volume leads to a decrease in cost per unit produced. This is because indirect costs such as overheads are supported by a larger number of units that can be sold. As a result, an increase in production capacity may often lead to a more economically viable production process due to economies of scale. This is expected to apply to the case for the large scele, add-on C123 Scenario C.

It may seem that small capacity plants such as the modular C123 scenarios cannot compete with large scale processes and therefore will not be viable, or as viable as the large-scale concepts. However, due to production intensification, the operation of multiple parallel plants contributes to a potential overall larger production capacity of containerized C123 plants. By producing a high volume of similar units (e.g. containerized processing plants) economies of scale are obtained during the manufacturing process of capital equipment, thereby lowering the cost per unit, which contributes to the overall economic feasibility of the modular C123 scenarios. Furthermore, the small-scale modular units may benefit from a lower feedstock cost as alternative applications bring no or low added value.

5.2 Market analysis of C3 products n-propanol and propanal

Although it is shown that the derived products propionic acid and trimethylolethane can be produced from propanal, only propanal and *n*-propanol are discussed in this section, since these are currently the main interest of the C123 project.

5.2.1 Propanal

The annual quantity of propanal that is imported or manufactured in Europe is in the range of 100 – 1000 tons [27]. Most of the manufactured propanal is used internally for the production of other chemicals. According to the trade data, a significant volume of propanal was exported in 2019 [28,29]:

- Germany had 22 % of the world exports (BASF, Oxea).
- The United-States had 22 % (Dow Chemical, Eastman).
- China had 14 % (Zibo Nalcohol Chemistry, which represents a production of 2 million tons per year) [29].

The export volumes of these countries and the average distance travelled by the product (weighted by the trade value) are shown in Figure 16. The concentration value on the x-axis is an indication of the diversity of the importing countries, where a value of 1 indicates a single country. The data illustrate that for propanal there would be a demand for small/local production, with small units, avoiding long distance transports and representing a potential market of 100 M USD or 85 M EUR. It is also seen that propanal produced in China and the USA travel longer distances than propanal produced in Germany. This also supports the selection of Germany as a location for C123 Scenario A (biogas).

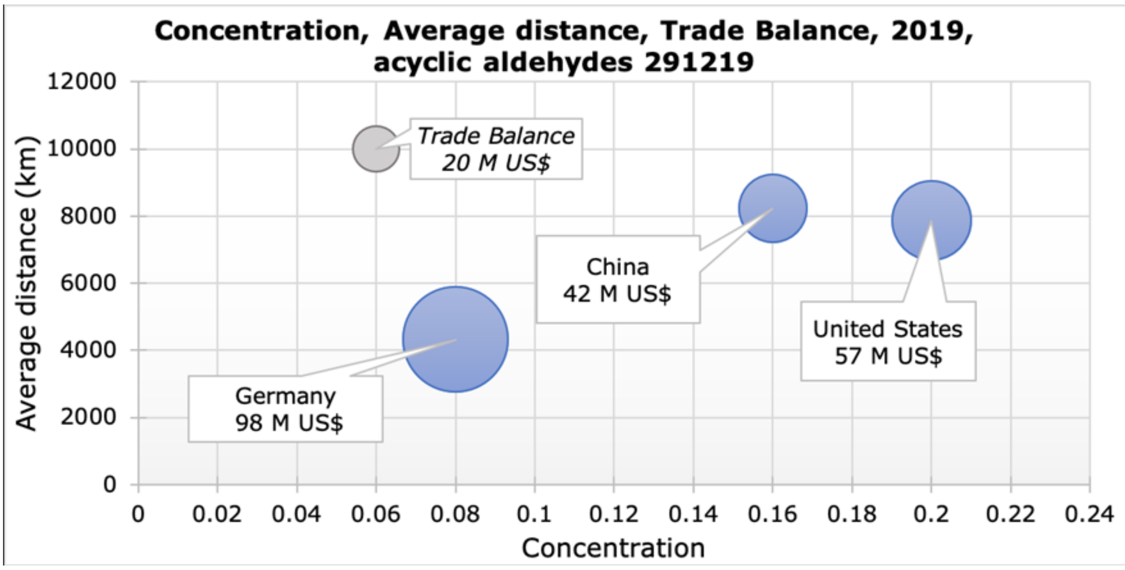


Figure 16: Concentration of exporting countries of products 291219 (acyclic aldehydes) and average distance with their destination countries in 2019. Data have been computed from TradeMap [29].

5.2.1.1 *n-Propanol*

The market for *n*-propanol is more developed, due to a wider range of potential applications. Each year 10,000 – 100,000 tons are produced in Europe [31]. *n*-Propanol contributes to 85 % of the propanol market and isopropanol contributes to the remaining 15 %. Asian countries produce mostly isopropanol (for example by Tokuyama, ISU, LCY, Zhejiang Xinghua Chemical, LG Chem). The net difference between exports and imports has been computed from the annual data available from TradeMap between 2010 and 2018 and is shown in Figure 17. By looking at the net quantity, the main producers could be identified.

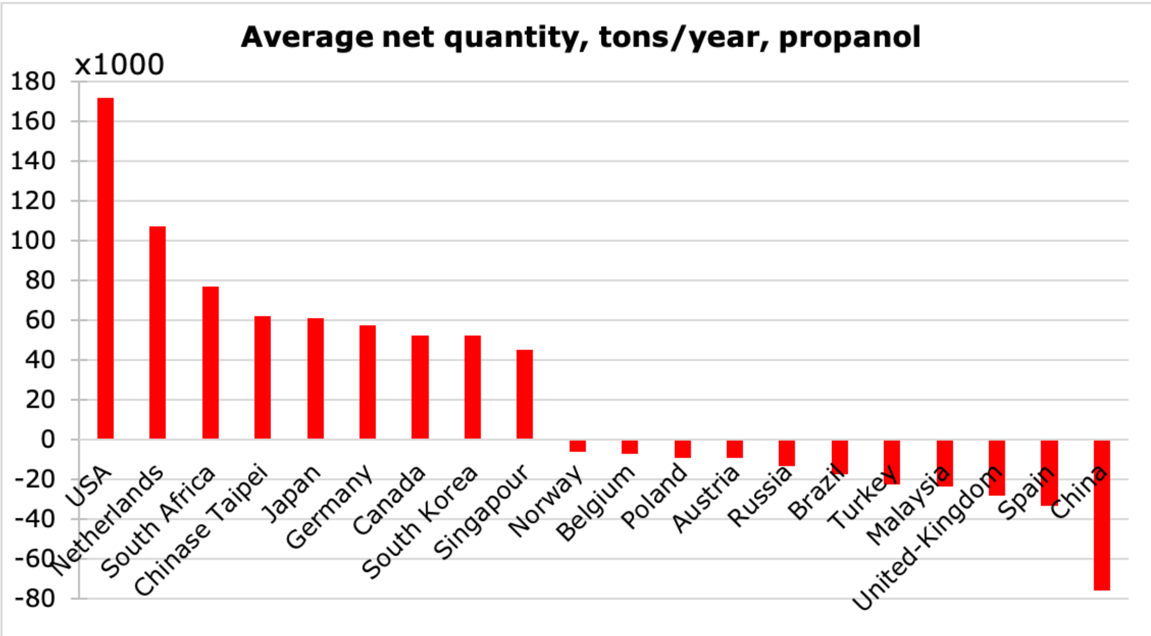


Figure 17: Calculated average of annual differences between exported and imported quantity, in each country, for the 2010-2018 period (tons per year), for products 290512 (*n*- and isopropanol). Data computed from TradeMap database [29].

According to the trade data, a significant volume of propanol was exported in 2019 [29,32]:

- The United-States had 22 % of the world exports (Dow Chemical, Oxea). Oxea produces around 100 000 ton per year [33].
- China had 10 % (Zibo Nalcohol Chemistry which products 1 million ton per year) despite a negative trade balance in 2019 (-2.1 M USD) [30].
- Germany had 9 % (Sasol Germany, Oxea).
- The Netherlands had 7.8 % (Eastman Chemical).
- South Africa had 6.3 % (Sasol, which claims 30 % of the *n*-propanol market) [34].

A corresponding comparison of export volume versus distance travelled for propanol is shown in Figure 18. From the main producing countries (USA, China, Germany, The Netherlands and South Africa), it can be seen by the low concentration factor in the x-axis that they export to many different countries. The average distance is on the same order of magnitude as for propanal. The propanol exported by Germany and the Netherlands do not travel long distances (most likely exported to Europe) compared to those of South Africa, South Korea and the USA. The average exporting distance of the USA is 6000 km. This supports the placement of modular C123 units in locations such as Russia.

5.2.1.2 Influences of prices

The variation of the trading value (import/export prices) for *n*- and isopropanol from the USA [29], the price of crude oil (Brent is selected as a better world price reference than WTI) [35], US ethylene [36] and US propylene [29], the traditional feedstocks for C3 products, is shown in Figure 19. It illustrates the dependency between the prices of the raw material and

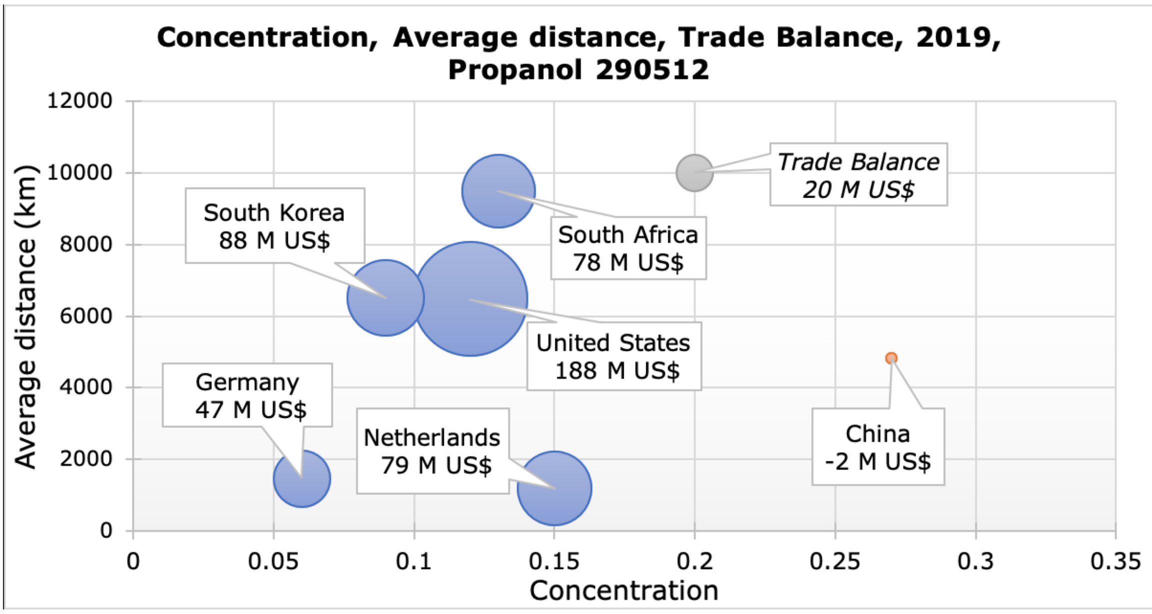


Figure 18: Concentration of exporting countries of products 290512 (*n*- and isopropanol) and average distance with their destination countries in 2019. Computed from TradeMap [29].

products of interest, and that the value of propanol replicates the price of crude oil, ethylene, and propylene. When propanol is at the same price per ton as that for propylene, the value of propanol is larger, since the dehydration to propylene also corresponds to a weight loss in material.

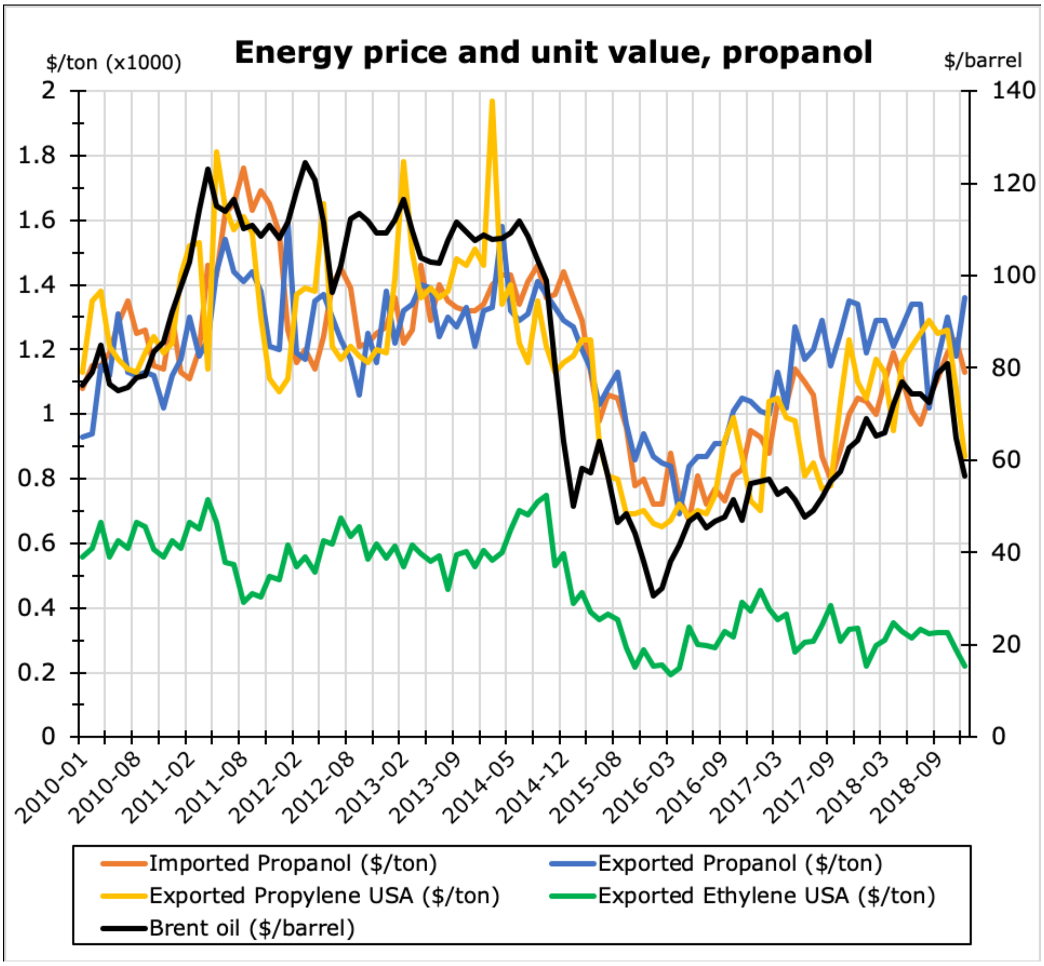


Figure 19: Evolution of energy (crude oil), ethylene, propylene and propanol prices in the USA between 2010 and 2018. Data from TradeMap [29] and IHS Market [36].

It is important to note that the price of the product depends on the production process as well. In South Africa, propanol is produced by Sasol via the Fischer-Tropsch process, which results in an average export price of 765 USD/ton [29], compared to the United States where propanol is produced through the hydrogenation of propionaldehyde, resulting in an export price at around 1008 USD/ton [29].

The influence of raw material costs on final product prices in a process can be analysed with the help of a correlation matrix. In a correlation matrix, values range between -1 and +1, where a positive value indicates that two parameters vary in the same direction. A value close to 0 indicates that two parameters are relatively independent, while a value close to 1 indicates that the two parameters are highly dependent on each other. A correlation matrix has been made for USA exports (Table 5), since the country produces C3 chemicals of interest.

To this end, the price fluctuations of the geo-economic environment can be isolated to reflect the connections to raw materials.

Table 5: Correlation Matrix between unit values for exports from the USA, monthly data in the period 2010-2018. Data from TradeMap [29] and IHS Market [36].

	Propanol	Propanal	Propionic Acid	Crude Oil	Ethylene	Propylene
Propanol	1					
Propanal	0.16	1				
Propionic Acid	0.04	0.21	1			
Crude Oil	0.67	0.48	0.07	1		
Ethylene	0.53	0.52	0.17	0.76	1	
Propylene	0.71	0.36	0.09	0.82	0.71	1

As a result, the value of exported ethylene correlates less with the value of crude oil (value of 0.76) than with the value of propylene (value of 1). This is most probably due that ethane crackers have recently increased ethylene production in the US. Propanal, which is produced by ethylene HF, correlates poorly with ethylene (value of 0.52), because most of the production is consumed internally, and only part of it is sold on the market. Propanol correlates with the price of crude oil (0.67) and ethylene (0.53), as this is the major market; while propionic acid, used in feed additives, has a rather small market and therefore poorly correlates with feedstocks (values of 0.07 - 0.17). Products correlate poorly between themselves (e.g. propanol and propionic acid have a correlation value of 0.04). This shows that there are opportunities for each product, and that a balanced product portfolio is probably a wise strategy. In addition, the targeted products have a high growth potential, of around 6-8 %. Therefore, propylene, propanal, *n*-propanol and propionic acid are deemed good C3 candidates for the C123 technology.

6 Conclusion

This report on the most attractive scenarios for the implementation of the C123 concepts. As a first step the different C123 scenarios were identified. The major differences between the scenarios are the feedstock used (biogas, marginal gas or associated gas) and the production scale (modular, containerized or large scale, add-on processes). The process development of these scenarios was discussed in Section 3. The geographic location, environmental impact, economic- and technical performance are the parameters that were used to determine the 'attractiveness' of a scenario. Each of these parameters are discussed for each C123 scenario in Section 4. The C123 scenario, feedstock, production scale, key product and location are summarized in Table 6.

A preliminary LCA was performed for C123-scenarios A, B1 and B2. This assessment

Table 6: Overview of the C123 Scenarios' feedstock, production scale, key products and location.

C123-scenario	Feedstock	Production scale	Key products	Location
A	Biogas	Modular	n-Propanol	Germany
B1	Marginal gas	Modular	n-Propanol	Russia
B1	Associated gas	Modular	n-Propanol	USA and/or Russia
C1	Marginal gas	Large scale	Propylene	Azerbaijan
C2	Associated gas	Large scale	Propylene	Middle-East

already delivered interesting findings. The greenhouse gas emissions and the impact on human health were the lowest for Scenario A. THe production of pure O_2 had a high contribution to the total greenhouse gas emissions and caused a high impact on human health in comparison with the other production steps in the analysed scenarios, but especially for Scenarios B1 and B2. Also, the removal of CO_2 from the recycle stream (Scenarios B1 and B2) had a relative high score for these two indicators.

The partners in the C123 project will continue with the further development of the technology and collaborate on the further definition and detailing of the C123 scenarios. The preliminary techno-economic and environmental impact assessment with comparison to reference scenarios are considered of high value for improving the feasibility, cost effectiveness and sustainability of the C123 scenarios under development.

Nomenclature

Symbol	Explanation
ANAS	Azerbaijan National Academy of Science
C1/C2/C3	Refers to chemicals with 1, 2 or 3 carbon molecules, respectively. C1 (e.g. carbon monoxide, methane), C2 (ethane, ethylene), C3 (propane, propanal, propanol, propylene)
CCS	Carbon capture and storage
CCU	Carbon capture and utilization
DALY	Disability adjusted life years
GTL	Gas to liquids
HF	Hydroformylation
KLM-file	Keyhole markup language file
kt	Kilotonne
LCA	Life cycle assessment
MEA	Monoethanolamine
OCoM	Oxidative conversion of methane
PDC	Process Design Center
TRL	Technology readiness level
UGent	University of Ghent
USA	United States of America
WP2	Work Package 2
yr	year

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