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Assessing butanol from integrated forest biorefinery: A combined techno-economic and life cycle approach

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Abstract: The life cycle assessment (LCA) methodology is increasingly used to ensure environmental sustainability of emerging biofuels. However, LCA studies are usually not performed at the process design stage, when it would be more efficient to identify and control environmental aspects. Moreover, the long-term economic profitability of biofuels depends on future energy and climate policies, which are usually not considered in techno-economic feasibility studies. This paper proposes a holistic approach, combining the LCA method and a TIMES energy system model, to offer a novel simultaneous assessment of potential environmental impacts and market penetration under different energy and climate policy scenarios of emerging energy pathways. The approach is applied to butanol produced from pre-hydrolysate in a Canadian Kraft dissolving pulp mill. Indeed, the integration of biorefinery processes into existing pulp and paper mills has been identified as a promising avenue to maintain mills activities. It could increase and diversify revenues, keep the forestry-based communities alive, and potentially mitigate climate change by replacing fossil-based fuels or products. Results show that 1) the energy efficiency of the butanol production process is a critical aspect to consider in future design and implementation steps in order to make butanol a competitive fuel among all other alternative fuels, 2) with a 50% internal heat recovery, butanol has a role to play in the transportation sector under climate policy scenarios, and may have a lower carbon footprint than gasoline as estimated by a 2010 US EPA study, and 3) higher supply costs for feedstock might undermine the competitiveness of butanol on the medium term (2030), but probably not on the long-term (2050). This novel combination of assessment methods is replicable to assess any types of emerging energy pathways in Canada and in other countries, and to help designing more sustainable forest biorefinery processes in other countries with important forest sector.

Keywords: Life cycle assessment, TIMES model, prospective modeling, biofuel, butanol, climate change

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1 Introduction

Biofuels are often claimed to be better alternatives than their fossil-based counterparts in terms of environmental impacts, climate change, and non-renewable resource depletion. Several jurisdictions, such as the United States and the European Union, have implemented renewable fuel policies to increase energy security and mitigate climate change [1, 2]. However, recent literature has shown that biofuels may also lead to economic, environmental and social issues which should be carefully taken into account to improve their sustainability. For instance, biofuel production may have adverse effects on biodiversity and ecosystem services [e.g. 3-5], or on climate change if land-use change emissions are substantial [e.g. 6]. Issues with climate change and resource depletion may also arise if supply chains and transformation processes require large amounts of fossil fuel [e.g. 7].

The life cycle assessment (LCA) method, supported by the ISO 14040/44 standards [8, 9], is the preferred approach to assess biofuels potential environmental impacts while considering the entire supply chain, and to quantify the potential reduction of greenhouse gas (GHG) emissions compared to substituted fossil-based fuels. Indeed, the LCA method is a life cycle-based and multi-impacts approach. It can therefore identify shifting of potential impacts toward other life cycle stages or other environmental issues that could be missed if only a portion of the value chain (e.g. biofuel combustion) or one single impact indicator (e.g. GHG emissions) was considered. For these reasons, LCA has often been used in recent literature to assess the environmental sustainability of biofuels [10] and to guide policies such as the Renewable Fuel Standard in the United States [11].

Several factors affect biofuels competitiveness, and associated uncertainties create challenges for potential investors [12]. For example, economic profitability of biofuels highly depends on the availability of low-cost feedstock, and on the energy prices that usually account for a substantial part of operating costs for biofuel production. Biofuels competitiveness also depends on fossil fuel prices, as increasing prices stimulate the market for alternative fuels and vice versa [13]. Moreover, future market penetration of biofuels will be affected by future policies such as renewable fuel regulations, carbon taxes or cap-and-trade systems. These factors are not all taken into account in traditional techno-economic calculations performed during the process design, which usually only include capital cost analysis, as well as revenue and operation costs estimation [14]. Moreover, the evolution of the energy sector should be analyzed on a long-term horizon to improve decision-making, given the long-lived nature of the capital stock to which energy production and consumption is tied.

A prospective approach is especially needed to evaluate emerging technologies such as advanced biofuels because technological innovations and political environment play a determining role [15]. Environmental challenges add yet another layer of complexity when assessing sustainability and viability of emerging biofuels. The complexity of these interrelated dimensions suggests the use of a holistic approach that would combine several methodologies such as energy system modelling and life cycle assessment. Today, decision makers are lacking this type of approach that would improve their understanding of how the energy system would react to the widespread deployment of new energy pathways. Indeed, consequences of such structural changes are traditionally assessed by analyzing technological, environmental and socio-economic aspects using separate approaches in separate studies.

There is a growing interest for combining techno-economic models with LCA to generate prospective scenarios and identify potential indirect impacts of policies or widespread deployment of new energy pathways. However, the combination of such approaches is still in its infancy [16]. For instance, Eriksson and colleagues have used scenarios coming from a dynamic optimization model of electricity and district heat production in Nordic countries (NELSON model) to identify marginal technologies for electricity production to be used in an LCA study [17]. These authors have identified several limitations regarding the suitability of these scenarios for their study, but they have shown that the use of a bottom-up energy system model can be effectively used in LCA. Other studies have discussed the importance of using energy system models to identify marginal technologies for electricity production in LCA [18, 19]. As another example, Earles has combined LCA with the US Forest Products Module, an existing partial equilibrium model, to look at potential environmental and economic impacts of emerging forest bioenergy pathways [20]. However, this model is specific to the US forest sector and does not consider the rest of the energy system. Choi and colleagues have combined LCA with a techno-economic model of the energy system (US MRM under the MARKAL framework) to look at potential environmental impacts associated with electricity generation in the US under different policy scenarios [15]. The US MRM model provided prospective electricity mixes that were then combined with life cycle inventory data for different energy generation technologies.

Menten and colleagues have proposed to integrate LCA GHG emissions data to the French MIRET model (under the TIMES framework) to look at the impact of the introduction of the BTL technology ("biomass-to-liquid", i.e. synthetic biodiesel). [21]. TIMES (The Integrated MARKAL-EFOM System) is a bottom-up model representing the entire energy system of a country or region over a long-term horizon. This typically includes extraction, transformation, distribution, end-uses, and trade of various energy forms. Each step of the energy value chain is described by specific technologies represented with their techno-economic characteristics (e.g., cost and efficiency). TIMES also computes GHG emissions from fuel combustion and processes. Emission reduction is brought about in particular by technology and fuel substitutions. TIMES is cast as a dynamic linear programming model. Under the assumption that energy markets are under perfect competition, a single optimization, which searches to meet the exogenously defined demand for energy services at minimum cost, simulates energy market equilibrium. From this perspective, it computes a perfect foresight partial-equilibrium for energy goods, obtained through the optimization of energy uses, while respecting (in some cases) specific policy constraints such as GHG emission reduction targets [22].

The work performed by Menten and colleagues [21] presents a first attempt to use a TIMES model in combination with LCA. The TIMES model was used to determine changes in the French energy system caused by the introduction of BTL fuel, as well as associated GHG emissions. In this paper, we propose a holistic approach combining 1) the life cycle assessment (LCA) method and 2) a TIMES model (NATEM-Canada) to offer a novel simultaneous comprehensive assessment of potential environmental impacts and market penetration of energy pathways. We then apply this new approach to the assessment of butanol produced from pre-hydrolysate in a Canadian Kraft dissolving pulp mill, a biofuel from a process that is still at the design stage.

In recent years, the Canadian forest sector has undergone a substantial decline, as also observed in other developed countries with important forest sector [23]. In response to this decline, the forest sector is currently experiencing major transformation through the development of new products and processes [24]. The current climate change context is also driving this transformation since forest products are often seen as preferable alternatives to non-renewable materials, chemicals, and energy sources. Therefore, an increasing number of policies and programs aim to promote the use of forestry- and agricultural-based biomass to replace fossil fuels [e.g. 25].

The integration of biorefinery processes into existing pulp and paper mills, to convert lignocellulosic biomass into a broad spectrum of products, has been identified as a promising avenue to maintain mills activities in Canada but also in other countries such as the United States, Brazil, Finland and Sweden [26]. These products could increase and diversify revenues, and keep the forestry-based communities alive. Three main classes of wood components, i.e. cellulose, hemicellulose and lignin, can be chemically or biologically transformed into different molecules to serve as biofuels, building block molecules, or biomaterials [27]. Research is ongoing to develop and optimize these processes, as well as to reduce associated environmental impacts and production costs. However, as these technologies are emerging, there are still very few published LCA studies assessing potential environmental impacts of integrated forest biorefineries [e.g. 20, 28]. Moreover, to our knowledge, there is no LCA study available for the production of butanol in an integrated forest biorefinery.

In Canada, major energy system modeling developments were reached through several successive research projects, culminating into the development of a TIMES model for Canada [29]. The most advanced model for Canada today is part of the NATEM (North American TIMES Energy Model) platform developed by ESMIA Consultants used to derive minimal cost solutions for reaching ambitious GHG emission reduction targets by 2050 in Canada [30]. We therefore use the NATEM-Canada model for the application of our novel approach to this case study, but other TIMES models could be used following the same approach in other geographical contexts.

The assessed butanol production process is added to the NATEM-Canada model to complement other first- and second-generation biofuel pathways. This new information could enhance the representation of the Canadian energy sector in global TIMES models such as TIAM-WORLD [31, 32]. Moreover, the proposed approach is replicable to assess other types of emerging energy pathways, and to help designing more sustainable and viable forest biorefinery processes in Canada and in other countries with an important forest industry sector (e.g. United States, Finland, Sweden). Observations from assessments performed at the process development stage using the holistic approach we have developed can be used to help designing more sustainable new generation biofuels.

2 Methodological approach

Butanol can be produced through biological conversion of hemicelluloses in existing Kraft pulp mills that produce dissolving pulp, using the ABE (Acetone-Butanol-Ethanol) fermentation. The properties of the bio-based butanol are similar to those of conventional gasoline [33], and better than ethanol (e.g. higher energy content, less corrosive, lower volatility) [34]. A process simulation, developed using the Aspen Plus software, is used to model the production process and provide data for both the LCA and the NATEM-Canada models. Consequently, sensitivity analysis can be performed by modifying easily input parameters common to both models and evaluate the impact of these modifications in a consistent manner.

2.1 Description of the butanol production process

In the Kraft process, hemicelluloses are usually burned, together with the black liquor, but can be partly diverted without affecting pulp production [35]. In a Kraft dissolving pulp process, hemicelluloses are already extracted from wood chips, prior to pulping, forming a pre-hydrolysate. This stream can then be diverted and used as a feedstock in a biorefinery. The hydrolysis step, transforming oligomeric sugars derived from hemicelluloses into monomers using sulfuric acid, is followed by concentration and detoxification steps using membrane filtration and flocculation. The goal of this step is to remove part of the water and the inhibitory compounds that are toxic for the fermentation microorganisms, such as furfural and phenolic compounds. Water is added to additionally decrease the phenolics concentration as well as xylose from an external source [36]. A fermentation step then converts sugars into acetone, butanol and ethanol, with recycling of non-fermented sugars. A two-column distillation step follows to separate ethanol and acetone from butanol. Further optimization work has been undertaken to ensure detoxification of the hydrolysate to the feasible thresholds for the ABE fermentation and to optimize the fermentation conditions. Figure 1 shows the process diagram for the production of butanol from hemicellulose extracted in a Kraft dissolving pulp mill.

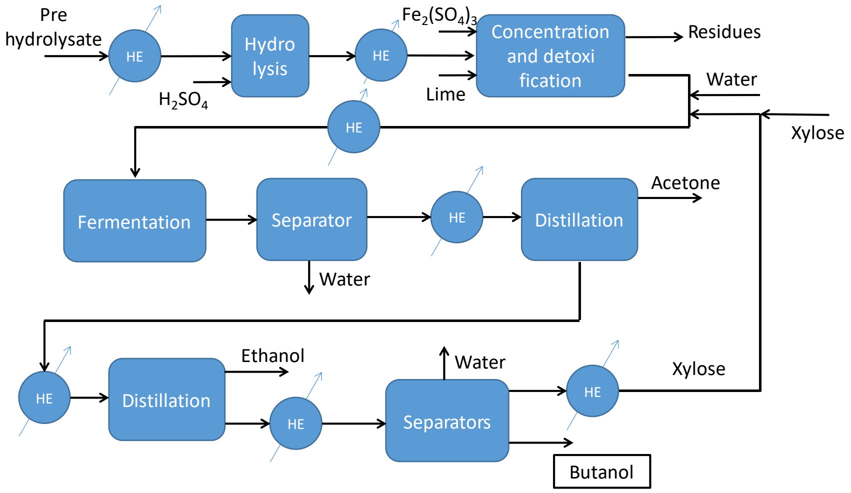

Figure 1: Process diagram for the production of butanol from hemicelluloses extracted in a Kraft dissolving pulp mill

The process simulation, performed using the Aspen Plus software, provides values for material and energy flows that are then used as input data for the LCA model and the NATEM technology database.

Table 1 presents a summary of this data. The design is based on the pre-hydrolysate flow coming from a Kraft pulp mill that produces dissolving pulp and is fed with 2,000 odt/d (oven-dried tons per day) of wood, which is representative of a Canadian pulp mill.

Table 1: Summary of process data based on a process simulation performed using the Aspen Plus software

	Material flows			**Energy flows**	
Inputs	Pre-hydrolysate	56.7 kg/s	Cooling	HE-1	13,800 kW
	H_2SO_4	0.015 kg/s		HE-2	143,300 kW
	Lime	0.012 kg/s		HE-3	3,600 kW
	$Fe_2(SO_4)_3$	0.54 kg/s		HE-5	2,700 kW
	Water	200 kg/s		HE-7	150 kW
	Fresh xylose	2.7 kg/s			
Outputs	Butanol	0.76 kg/s			
	Acetone	0.12 kg/s	Heating	HE-4	84,200 kW
	CO_2	1.5 kg/s		HE-6	15,600 kW
	Ethanol (42%)	0.63 kg/s		Distillation	36,500 kW
	Water	255 kg/s			
	Residues	1.0 kg/s			

2.2 Description of the LCA model

The background data for this LCA study is taken from the version 3.1 of the ecoinvent database with default allocation. This version of the database uses the technique of allocation at the point of substitution for which burdens are attributed proportionally to specific processes. The approach used by the database developer to model product systems may have an influence on the results. Therefore, a sensitivity analysis is performed using the cut-off allocation database which is built on the idea that a producer is fully responsible for the disposal of its waste and that he does not receive any credit for the provision of any recyclable materials [37]. Inputs of material and energy from the process simulation shown in *Table* 1 are scaled to the functional unit, i.e. the production of 1 kg butanol, dividing them by the flow of 0.76 kg/s butanol. The most recent IMPACT 2002+ life cycle impact assessment method updated in 2011 [38], and the SimaPro software are also used for this LCA study.

We assume that the Kraft mill is located at a distance of 500 km from the nearest important city from which chemicals are bought and where butanol is sold to consumers. This distance is an estimated average for Quebec Kraft pulp mills. Acetone and ethanol are considered to be sold as by-products. Therefore, the production of an equivalent amount of each of these products is considered avoided in the LCA model and associated environmental impacts are credited. The CO_2 emissions (see Figure 1) are considered to be released in the atmosphere. Residues are mainly composed of lime, $Fe_2(SO_4)_3$, water, phenol, and acetic acid and are considered treated and landfilled as pulp and paper sludge. The transportation distance between the pulp mill and the waste treatment facility is assumed to be 250 km on average.

Since pre-hydrolysate would be used as an energy input if not diverted as feedstock to the biorefinery, an energy production process is added to the modeled system to compensate for this loss. We assume a 50% combustion efficiency when pre-hydrolysate is sent to the mill boiler to have a worst-case scenario in terms of energy efficiency as Kraft pulp mills in Quebec are quite old. An energy content of 0.54 MJ/kg is calculated using the composition of pre-hydrolysate and lower heating value of each compound. The butanol production process still being at the development stage, the configuration of heat exchangers is still unknown. We modeled the worst case, i.e. all the energy needed by the process must be produced in the biorefinery or supplied by the pulp mill. The design of the butanol production process is currently in an early simulation phase. An integration study will be performed later to identify the opportunities for energy and material integration between the pulp mill and the biorefinery process. Such integration should decrease the need for energy production and fresh water supply. In the LCA study, we model two types of energy production often used in pulp and paper mills, i.e. wood in biomass boilers and natural gas. Figure 2 shows all the processes that are considered in the LCA model and Table SM1 in the supplementary material provides additional details such as assumptions and selected ecoinvent processes.

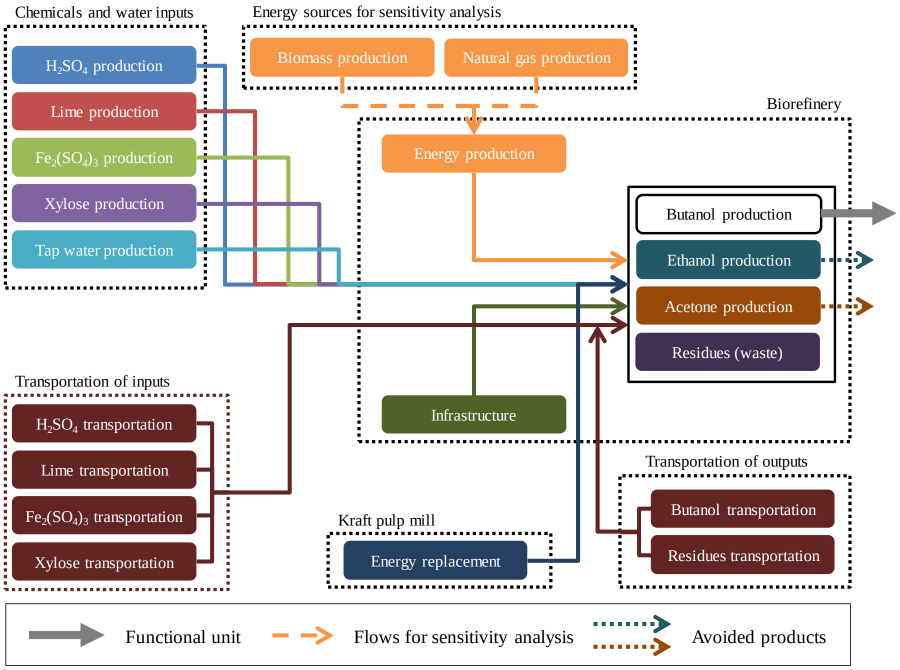

Figure 2: LCA model for butanol production in a biorefinery integrated to a Kraft dissolving pulp mill

2.3 Description of the NATEM-Canada model

NATEM-Canada, which follows a TIMES modeling approach, covers the integrated energy system of the 13 Canadian jurisdictions. It is used to explore the techno-economic potential of butanol production from integrated biorefineries in Kraft pulp mills up to the 2050 horizon in the province of Quebec. The model is driven by a set of 70 end-use demands for energy services in five sectors: agriculture, commercial, industrial, residential and transportation. A large number of technologies are in competition to satisfy each end-use demand, including existing technologies, improved versions of existing technologies, as well as new technologies. The NATEM-Canada database includes all secondary conversion technologies (e.g. power plants, refineries, biofuels/biomass plants, hydrogen production, and liquefaction of natural gas) as well as primary sources (e.g. oil, gas, coal and uranium reserves, renewable potentials and biomass sources). All energy flows between Canadian jurisdictions are optimized by the model while international trade movements are specified exogenously. Finally, NATEM-Canada tracks direct GHG emissions from fuel combustion as well as fugitive emissions from the energy sector.

Butanol production from integrated biorefineries in Kraft dissolving pulp mills was added to the NATEM-Canada database using the assumptions presented in Table 2. This is represented through a technology producing butanol (and heat) from a feedstock of pre-hydrolysate and accounting for all energy flows required during the conversion process (e.g. biomass). The generated heat can be either a) lost, b) re-used within the butanol production process, or c) re-used within the Kraft pulp mill. The butanol becomes a fuel in competition with both conventional fuels and other biofuels for consumption in the transportation sector: as a flexible blend with gasoline (up to 100%) in the various types of road vehicles and/or with jet fuel (up to 50%) for air transportation. The other biofuel pathways are: ethanol fermentation, biodiesel transesterification, synthetic diesel from gasification (Fischer-Tropsch (FT) process), bio-methanol from thermochemical platforms, and renewable natural gas (or upgraded biogas) from anaerobic digesters. Finally, supply curves were built for a large variety of feedstock that are in competition for biofuel production, pellet production, space heating and electricity generation, such as corn, soybeans, canola, greasy residues, fish oil, forest residues, agricultural residues, industrial wastes, dedicated crops for fast-growing trees, municipal organic wastes, manure, etc.

Butanol is produced from pre-hydrolysate that is usually burned to produce heat for the Kraft pulping process. Therefore, biorefinery implementation will lead to a higher external energy demand from the pulp mill, but no feedstock will have to be bought. This additional energy could be provided by biomass, fuel oil or natural gas. Since pre-hydrolysate is a lower quality fuel, we set the feedstock value to half the average price of heavy fuel oil [39]. To test the robustness of the results to this assumption, we also perform a sensitivity analysis using the full average price of heavy fuel oil as a worse case.

Maximum feedstock availability is estimated for 2012, 2020, and 2050 to inform the NATEM-Canada model about the maximum amount of butanol that could be produced in Quebec using the integrated production process. The process has not yet been implemented in a Kraft pulp mill, we therefore set the maximum feedstock availability for 2012 to 0. The process simulation has been developed for an average Canadian Kraft dissolving pulp mill that processes 56.7 kg/s of pre-hydrolysate (see *Table* 1), which corresponds to 0.97 PJ/yr. We assume that a maximum of 2 biorefineries could be implemented by 2020, leading to a feedstock availability of 1.94 PJ/yr. For 2050, we assume that the maximum number of biorefineries implemented is equivalent to the number of Kraft pulp mills currently in operation in Quebec, leading to 11 biorefineries for a total maximum feedstock availability of 10.69 PJ/yr. The increase in feedstock availability is considered linear between these moments.

We calculate investment costs using a classical process design approach [40]. These costs include equipment purchase and installation, engineering, and other indirect costs. The investment costs are divided by the installed capacity, which is the total amount of energy produced in terms of butanol and excess heat. Fixed and variable operating costs are also calculated using the same approach [40]. Annual fixed operating costs include maintenance, administrative overhead, and labor. They are also divided by the installed capacity. Variable operating costs include direct production costs such as chemicals and utilities. They are calculated per unit of energy produced in terms of butanol and excess heat, and do not include energy costs because they are already considered by the NATEM-Canada model when energy is consumed by a given technology.

Table 2: Input data, for the NATEM-Canada model, which describes the production of butanol in biorefineries integrated to Kraft pulp mills in Quebec

Parameters	Values (in primary energy or CAD of 2012)	Comments
Feedstock cost	Base case: 7$/GJ Sensitivity analysis: 14$/GJ	Base case: Half of average heavy fuel oil price in 2011 Sensitivity analysis: Average heavy fuel oil price in 2011
Maximum feedstock availability	2012: 0 PJ/yr 2020: 1.94 PJ/yr 2050: 10.69 PJ/yr	In 2012, the technology is not yet implemented. We assume implementation in 2 plants in 2020, and in 11 plants in 2050.
Investment costs	2020: 327 $/kW 2050: 294 $/kW	10% decrease assumed from 2020 to 2050 from technological improvement
Fixed operating costs	19 $/(yr·kW)	
Variable operating costs	0.31 $/GJ	Excluding energy costs
Annual availability factor	96%	Two weeks of shutdown per year is assumed
First year of availability	2020	
Lifetime	30 years	After 30 years, additional investments are required for modernization, etc.

3 Results and discussion

In this section, we present the results of the combined environmental and techno-economic assessment. We first analyze potential environmental impacts from the butanol production process. We then identify hot spots in production and potential for environmental improvement to guide future design steps. We also compare the butanol carbon footprint to those of corn butanol and gasoline from a US EPA study [11]. Finally, we analyze potential market penetration for butanol from integrated forest biorefinery under different GHG emission reduction scenarios, and identify sensitive aspects to guide future design steps.

3.1 Potential environmental impacts

Figure 3 shows endpoint LCA results for the production of 1 kg butanol from pre-hydrolysate from Kraft pulp mill producing dissolving pulp. These results are for the base case in which 100% of the energy needed by the production process is coming from biomass. Absolute results are 3.9 kgCO$_2$-eq for climate change, 1.2x10^{-4} DALY (disability-adjusted life years) for human health, 26 PDF·m^2·yr (potentially disappeared fraction of species per square meter area and per year) for ecosystem quality, 28 MJ primary for resources, and 8x10^{-4} m^3 for water withdrawal.

Energy consumption is the main contributor to potential impacts on climate change, ecosystem quality and resources, followed by xylose consumption. For climate change, the treatment and landfilling of detoxification residues is the third contributor. For potential impacts on human health and water withdrawal, the first and second main contributors are reversed. Negative potential impacts are observed from avoided ethanol and acetone production for climate change, resources, and water withdrawal categories. These negative impacts are negligible for human health and ecosystem quality.

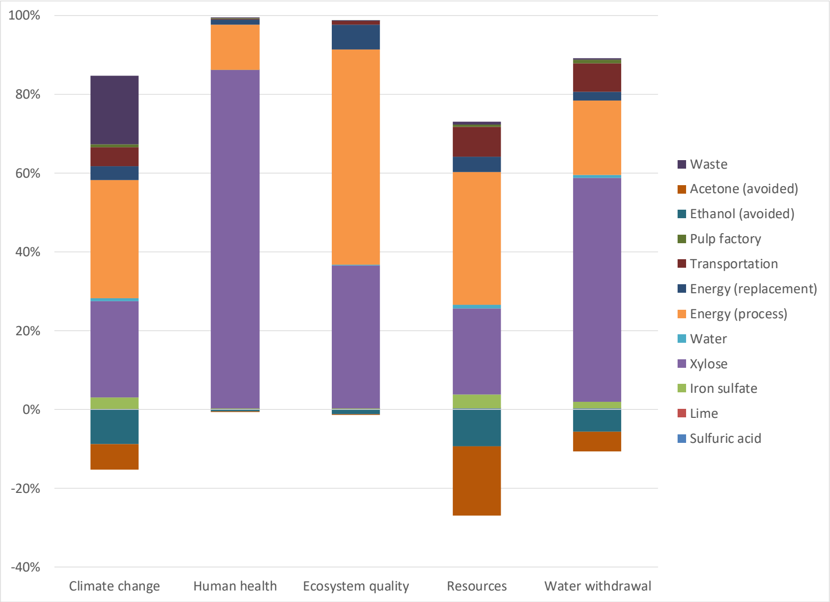

Figure 3: Endpoint LCA results for the production of 1 kg of butanol from pre-hydrolysate from a Kraft pulp mill producing dissolving pulp for the base case (100% energy is produced from biomass) showing the contribution of the different processes

The preliminary results shown in Figure 3 are based on a process simulation performed with the ASPEN software at a very early stage of the design process. Therefore, substantial uncertainties are associated with some process data. However, the contribution analysis performed is already providing some insights to guide further design and implementation in order to achieve better environmental performance. Of course, energy optimization will be a key issue. Indeed, we considered that 100% of the energy needed by the process was to be produced at the biorefinery or pulp mill, and that all the heat removed from streams that need to be cooled in the process was lost. This worst case is not realistic. In a future design stage, an analysis will be performed to determine the best configuration for heat exchangers to maximize energy recovery in the butanol production process. This analysis should include the Kraft pulp mill in which the biorefinery will be integrated using a recently developed methodology [41].

To show the impact of improving energy efficiency on LCA results, we performed a sensitivity analysis considering that only 50% of the energy needed by the process would have to be produced. The other 50% is recovered from other process streams that have to be cooled. This 50% value is an arbitrary choice as it is difficult to estimate the energy integration level that will be achieved at this design stage because it may vary significantly depending on the mill configuration. Moreover, we added two other cases to show the impact on LCA results of using natural gas instead of biomass with the same sensitivity analysis. Figure 4 shows the results of this sensitivity analysis. Having to produce 50% of the energy instead of 100% decreases potential impacts by around 22 to 39% for climate change, ecosystem quality and resources. For the human health and water withdrawal categories, the reduction in impact is lower, i.e. 3 and 6% respectively. If natural gas is used to produce energy instead of biomass, potential environmental impacts for climate change and resources increase by 250 to 800% depending on the energy integration level. However, changing fuel does not affect potential impacts on human health significantly, and leads to a reduction for potential impacts on ecosystem quality.

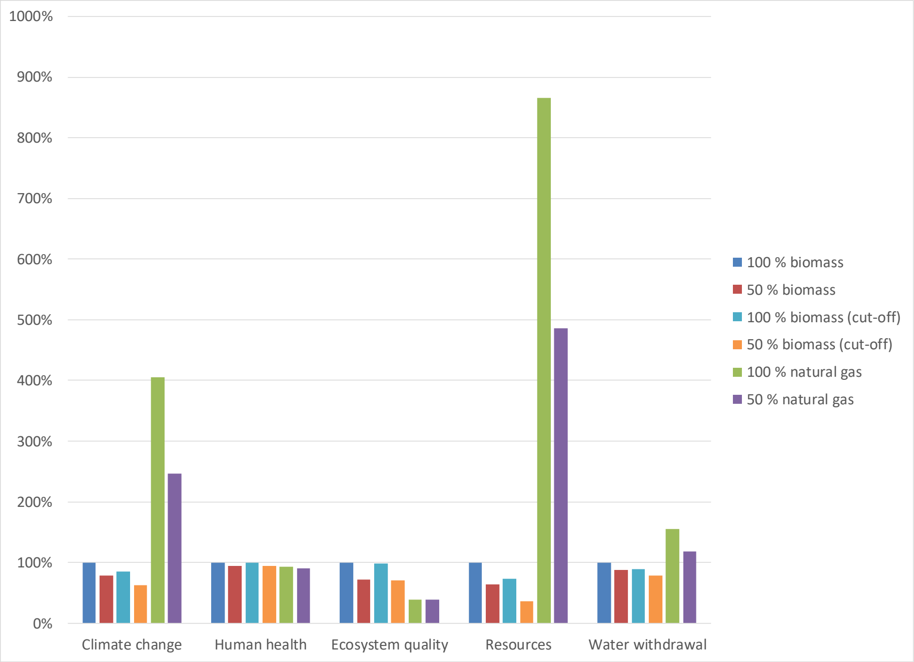

Figure 4: Endpoint LCA results for the production of 1 kg butanol from pre-hydrolysate from a Kraft pulp mill producing dissolving pulp for different assumptions regarding energy source, integration level, and ecoinvent system modelling approach

Xylose production contributes substantially to potential impacts for all the endpoint categories. However, no xylose production processes are available in version 3.1 of the ecoinvent database. Therefore, we used a proxy of sugar production from sugar cane. Xylose production may have more or less impact than sugar from sugarcane. However, these preliminary results do give an indication that we should pay attention to the amount of xylose needed in the process and look for ways to decrease it to get a better environmental profile.

We compared the LCA results for the climate change impact category to those coming from the life cycle greenhouse gas (GHG) assessment performed by the US EPA for the application of the Renewable Fuel Standard published in 2010 [11]. Figure 5 shows the results in gCO$_2$-eq/MJ of fuel so that we compare functionally equivalent options. We used a lower heating value of 34.4 MJ/kg for butanol as a conversion factor [42]. Tailpipe emissions for butanol from integrated forest biorefineries are considered equivalent to tailpipe emissions from corn butanol and are taken from the US EPA study [11].

Results show that the carbon footprint of butanol is 20% higher than that of gasoline for the worst case scenario (i.e. 100% of energy is produced), and 5% lower for the sensitivity analysis case (i.e. 50% of energy integration). These results are valid as long as biomass is used as an energy source without any natural gas. The carbon footprint of corn butanol is lower than that of butanol from a Kraft pulp mill pre-hydrolysate for both scenarios, being 23% lower than that of gasoline. Of course, these results still have to be refined to decrease uncertainty as the design process goes on. This preliminary study shows that efforts are still needed to improve the process before butanol produced from Kraft pulp mill pre-hydrolysate replacing gasoline becomes an interesting choice in terms of GHG emissions reduction. The three main aspects to pay attention to are energy efficiency and integration, fresh xylose consumption from an external source, and detoxification residues.

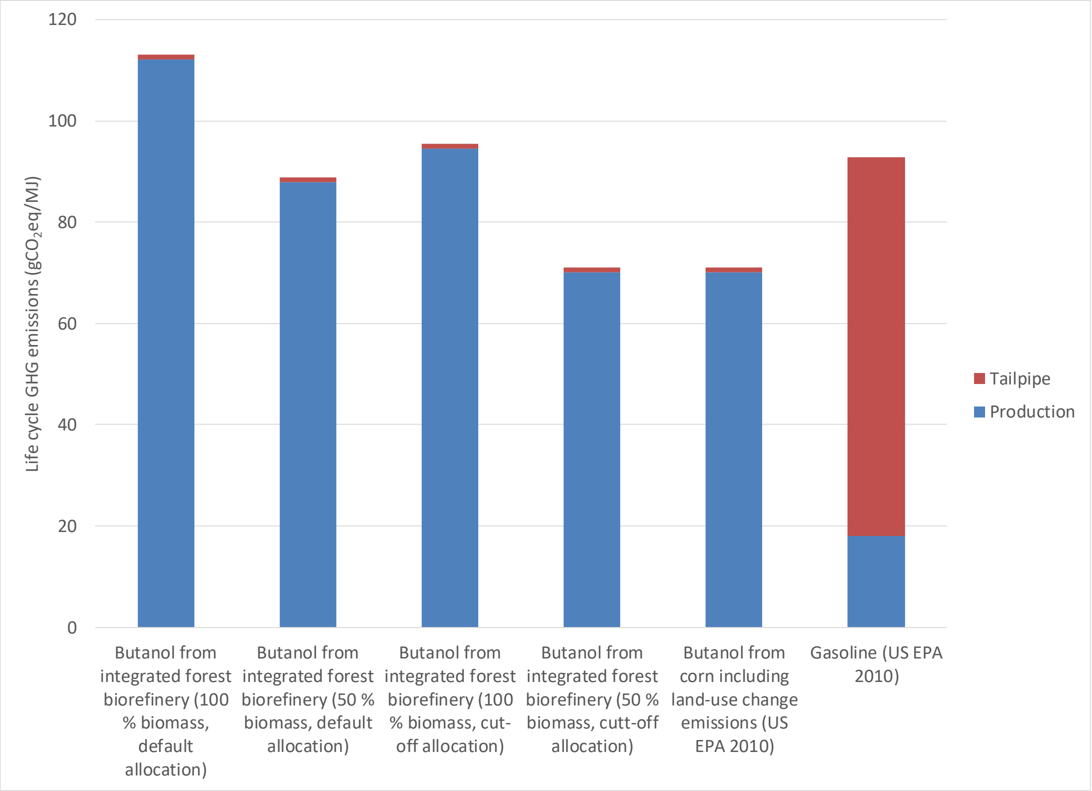

Figure 5: Comparison of life cycle GHG emissions for different fuels

3.2 Techno-economic assessment

This section presents the results computed by the NATEM model. Optimization models provide a rigorous analytical basis for deriving minimum cost solutions that meet both growing demands for energy-related services and reductions in GHG emissions. Capturing the whole energy system, they are commonly used to assess the techno-economic potential of emerging technologies or fuels on energy markets under various socio-economic or environmental conditions in a given country. The approach is applied here to the butanol produced from integrated forest biorefinery and the results show to what extent this new biofuel can compete with other transportation fuels. The figures discussed below represent relevant portions of the optimal configuration of the energy system under different conditions (scenarios).

Optimization runs were carried out with the NATEM-Canada model for three main scenarios, i.e. one reference case and two climate policy cases:

- REF: The reference scenario represents a business-as-usual case for Quebec including policies already in place, but without any GHG emission reduction targets.
- GHG1: A scenario with GHG emission reduction targets for 2020, 2030 and 2050 for Quebec, with access to a carbon market with other jurisdictions (e.g. California); a portion of the target is thus achieved through emission credit purchases (exogenously assumed, that yield lower emission reduction targets for Quebec, see Table ***3***).
- GHG2: A scenario with GHG emission reduction targets for 2020, 2030 and 2050 for Quebec, without access to a carbon market with other jurisdictions (e.g. California); all reductions thus have to be achieved on the Quebec territory.

Policies already in place for the Province of Quebec in the reference scenario include: measures from the Electrification Plan to replace 66 million liters of conventional fuels consumed annually [43], CAFE (Corporate Average Fuel Economy) standards, the federal regulation on the minimum renewable content in the gasoline (5%) and diesel (2%) sold in Canada, and a minimum carbon price going from 10 \$/ton in 2012 to 66 \$/tonne in 2050 (\$2011) to account for the existing carbon market in Quebec (not optimized within the model).

In addition to these existing policies, additional constraints are imposed in the climate policy scenarios to limit GHG emissions from the energy sector, representing 66% of all GHG emissions in 1990 (and 69% in 2013) [44]. Two series of targets are considered [45] depending on the possibility to achieve a fixed portion of the reductions through emission credit purchases (GHG1) or if all reductions need to be achieved in Quebec (GHG2). NATEM-Canada computes the optimal solution for each of these scenarios by identifying the technology and the commodity mixes that minimize the total energy system cost.

Table 3: GHG emission reduction targets in the two climate policy scenarios

Target year	GHG1 – with emissions trading		GHG2 – without emissions trading	
	% reduction from 1990 level	Cap in Mt	% reduction from 1990 level	Cap in Mt
2020	-14,8 %	49.9 Mt	-20,0 %	46.8 Mt
2030	-25,9 %	43.4 Mt	-37,5 %	36.6 Mt
2050	-68,4 %	18.5 Mt	-80,0 %	11.7 Mt

The result analysis was carried based on the assumption that 50% of the heat generated by the butanol production process could be recovered and re-used for its own production. The optimal solution shows indeed that heat is recovered and re-used in replacement of the biomass consumed in the process (natural gas is not used since its combustion generates GHG emissions). The optimization runs without heat recovery show a very marginal production of butanol in 2050 (2.9 PJ). A first insight of this analysis is that heat recovery will be a critical aspect to make butanol a competitive fuel among all other alternative fuels.

In addition, a sensitivity analysis was performed in both climate policy scenarios on the supply cost of feedstock for butanol production, an important factor affecting the economic profitability of producing butanol. As indicated in Table 2, the cost has been set at 7$/GJ in the original scenarios (GHG1 and GHG2), but doubled in the new scenarios (GHG1-HI and GHG2-HI).

More than half of the GHG emissions in Quebec are coming from the transportation sector, since the electricity sector benefit from the availability of abundant and clean hydraulic resources. Due to the dependence of the transportation sector on gasoline and diesel to meet most of the growing demand in a business-as-usual case, this trend is not expected to change over 2050 without incentives. However, under carbon constraints, most of the reductions are coming from the transportation sector where GHG emissions decrease from 37.7 MtCO$_2$-eq in 2050 in the reference scenario (REF) to 6.8 MtCO$_2$-eq and 2.8 MtCO$_2$-eq respectively in 2050 in the climate policy scenarios (GHG1 and GHG2). Figure 6 shows the evolution of emission reductions by sectors in the most stringent scenario (GHG2). This reduction counts for 56% of total reductions required to reach the target in 2050.

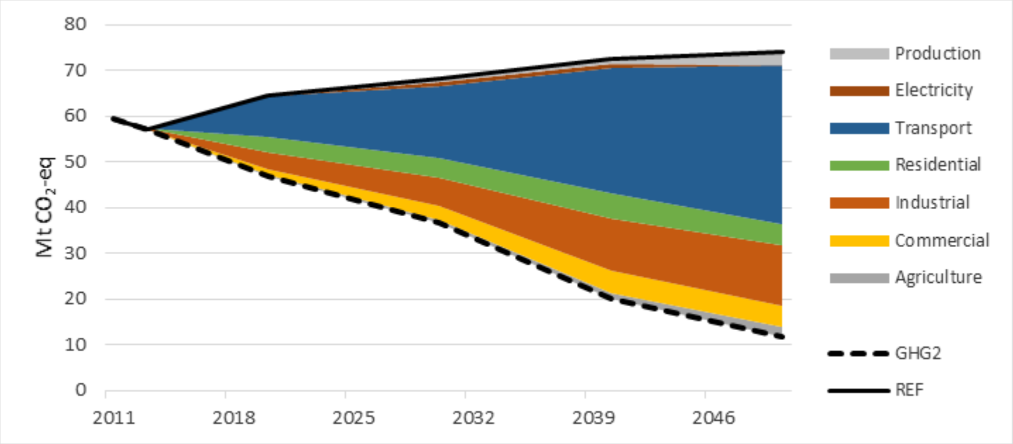

Figure 6: Emission reductions by sectors in the scenario GHG2

Achieving such ambitious reduction targets involves significant transformation in the energy system. For instance, in the GHG2 scenario, we observe: 1) endogenous reduction of the useful demands up to 15% for segments with fewer reduction options such as air transportation, 2) energy efficiency improvements through technology replacements with more advanced versions, 3) higher electricity penetration rate in end-use sectors up to 69% of the final energy consumption in 2050 (compared with 43% in the reference scenario), and 4) a larger proportion of bioenergy rising up to 19% in 2050 (compared with 4% in the reference scenario).

In particular, significant changes occur in the transportation sector (Figure 7) where the GHG reduction targets lead to a much lower level of energy consumption globally for that sector: a 33% decrease in 2050 comparing the GHG2 scenario with the reference scenario. This is mainly due to the electrification of several useful demands for passenger transportation as electric motors are approximately three to four times more efficient than internal combustion engines. Second generation liquid biofuels such as Fischer-Tropsch diesel, biomethanol and cellulosic ethanol are also used in plug-in hybrid vehicles (namely personal cars and light-duty trucks), while butanol is penetrating the market namely for meeting the air transportation demand. Indeed, the road transportation segments benefit from a large variety of alternative fuels, but replacement to conventional aviation fuels are limited.

As for freight transportation, conventional fuels are gradually replaced with a variety of alternative fuels including liquid biofuels, renewable natural gas, liquefied natural gas (LNG), and hydrogen. Liquid biofuels and renewable natural gas are mainly dominant in the road and rail transportation modes for freight, while LNG is replacing most of the heavy fuel oil consumed for marine transportation. Hydrogen is an expensive option which is used for heavy freight transportation when the total amount of biomass available for liquid biofuel production is already totally used such as in 2050 in the GHG2 scenario. In general, most other alternative fuels start penetrating as early as 2020 in the energy mix to replace conventional fuels. However, their penetration rates increase over time with the GHG reduction constraints.

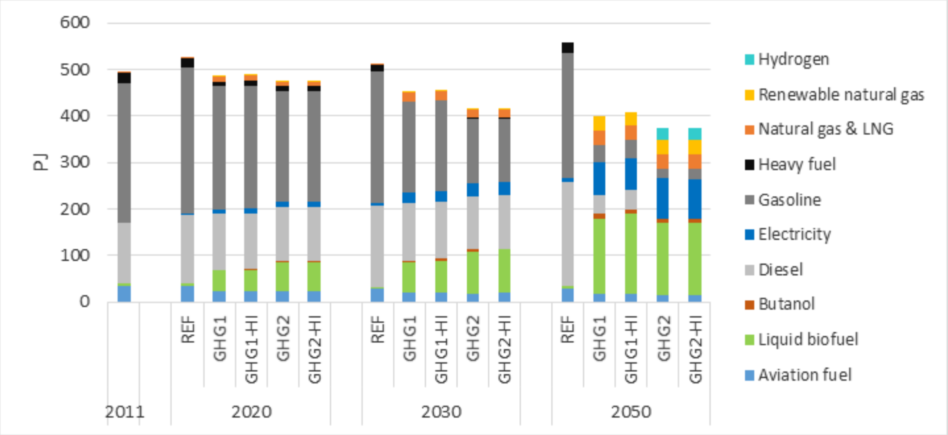

Figure 7: Final energy consumption by type in the transportation sector in Quebec

In a context where ambitious GHG emission reductions are expected, all types of bioenergy play a major role in all sectors and these trends are particularly strong on the long-term 2050 (Figure 8). Many useful demands are in competition for various bioenergy, especially those where the electrification is not possible. The transportation sector requires most of this bioenergy (56%), but the remaining portion is consumed in the commercial (13%) sector and the industrial sectors (25%), although this proportion also includes biomass feedstock used in the pulp and paper industry. While butanol represents a small portion, it has a role to play in the optimal strategies required to achieve ambitious GHG targets.

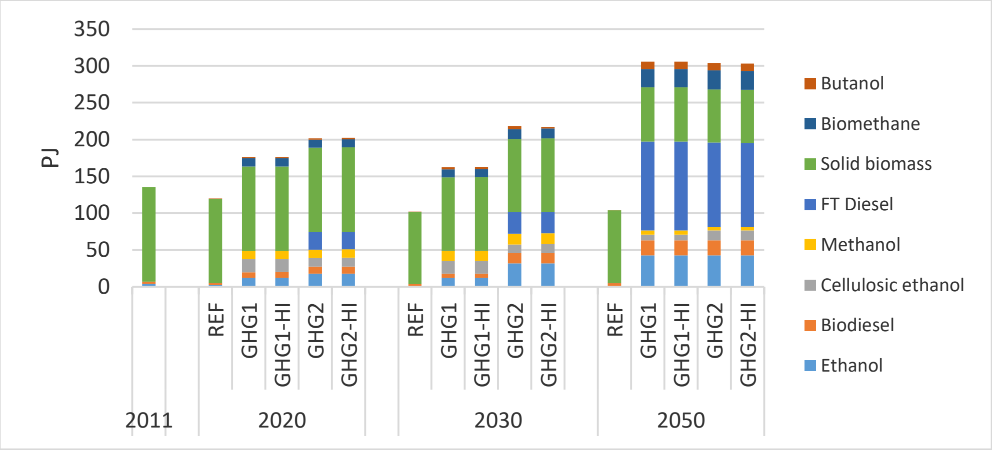

Figure 8: Consumption of various bioenergy by type in all end-use sectors in Quebec

Since biomass conversion processes normally have low efficiency rates, a large amount of feedstock is necessary to produce all these forms of bioenergy (Figure 9). Energy crops allow to produce up to 11 PJ of biodiesel and 15 PJ of ethanol, a maximum reached by 2030 in all climate policy scenarios. A very small quantity of these first-generation biofuels is imported from neighboring jurisdictions. These are the only types of bioenergy allowed for trade in the model due to the federal legislation on renewable content in conventional fuels sold in Canada.[1] Forest residues are available in different quantities at different prices. They are in competition for many usages including second-generation biofuel production, electricity generation, direct combustion for space heating, and feedstock in the pulp and paper industries. A first step of the supply curve is reached by 2020 where the maximum amount of forest residues related to existing harvesting activities (137 PJ) is used. The additional amount of biomass related to existing harvesting activities coming from a more optimistic estimate (totalizing 180 PJ) is used completely by 2030 in the most stringent scenarios (GHG2 and GHG2-HI). By 2050, additional amounts of not yet harvested biomass available at a significantly higher cost is also requested (up to 242 PJ) in all scenarios. While the optimal solutions include the use of lower-cost feedstock, the maximum potential is reached by 2050 for agriculture residues, industrial residues, dedicated crops, organic municipal waste, and biogas.

Regarding the pre-hydrolysate from the pulp and paper industry available namely for butanol production, the results show that the maximum potential is reached by 2020 in all scenarios (Figure 10). Based on the assumptions described in Table ***3***, it would be cost-effective to implement biorefineries in two pulp mills. By 2030, assumptions on the supply cost of feedstock are affecting the economic profitability of producing butanol: the maximal potential is reached in the most stringent scenario GHG2 with lower supply cost (7$/GJ), but higher supply costs (14$/GJ) undermine the competitiveness of butanol over other next-generation biofuels.

The marginal abatement costs increase rapidly with the reduction target level. In 2020, they reached 277 \$/tCO$_2$-eq (GHG1) and 352 \$/tCO$_2$-eq (GHG2). In 2030, they increase to 400 \$/tCO$_2$-eq (GHG1) and 566 \$/tCO$_2$-eq (GHG2), and in 2050, to above 1000 \$/tCO$_2$-eq due to the very ambitious targets combined with the lack of GHG emission reduction options in some sectors and relatively inelastic useful demands. The total net discounted cost of the energy system increases relatively to the baseline by 2.4% (GHG1) and 7.7% (GHG2); the total cost is discounted at 5% to the 2011 base year.

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[1] The availability of feedstock in general and other types of bioenergy is limited to the borders of each jurisdiction. The assumption is made that a jurisdiction cannot increase its biomass imports from neighboring jurisdictions, given the uncertainty on the adoption of targets elsewhere, and consequently, competition for the available biomass and bioenergy. These issues are beyond the scope of this analysis.

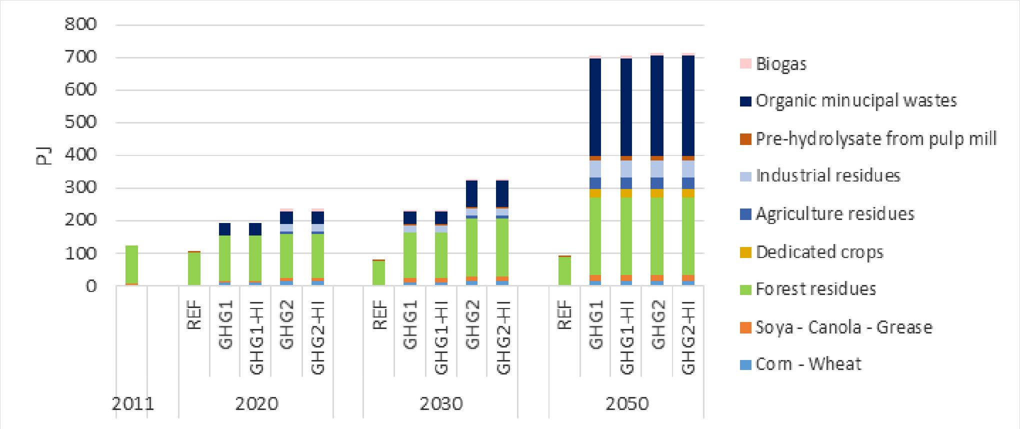

Figure 9: Consumption of feedstock by type for bioenergy production in Quebec

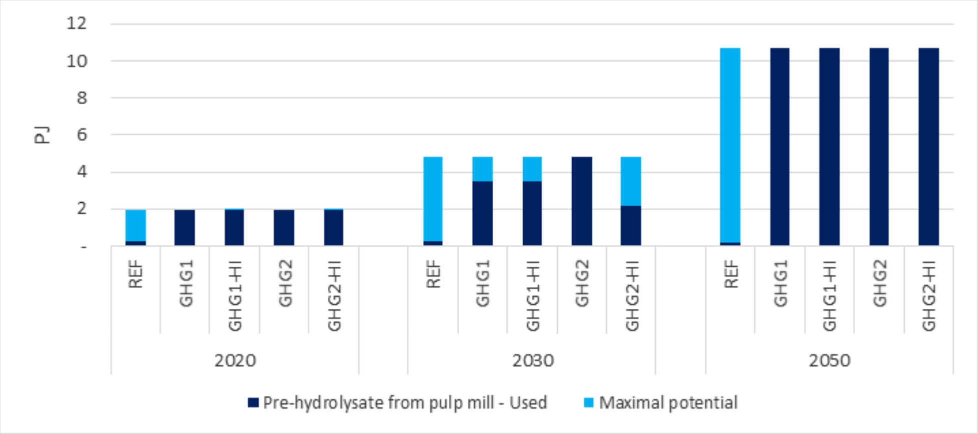

Figure 10: Consumption of pulp and paper residues for butanol production in Quebec

While it is not possible to compare the techno-economic potential of butanol production from an integrated forest biorefinery with other estimates given in the literature due to the novelty of this process, a brief comparative assessment of the role of transportation biofuels in climate mitigation scenarios in general is worth introducing. Indeed, numerous carbon mitigation studies have been carried out using cost-optimization TIMES models in multiple countries. Our results show that liquid biofuels could represent 45% of the transportation fuels in 2050 in order to meet Canadian GHG reduction targets with renewable natural gas accounting for another 7% to 9%. In California, for instance, a scenario analysis for achieving an 80% GHG reduction goal of 1990 levels by 2050 shows that liquid biofuels make up 37% of transportation fuels in 2050 [46]; the rest of the sector being decarbonized through important demand reductions and hydrogen use. In a similar scenario for Ireland, imported biofuels and domestic renewable natural gas account for 82.5% of the transport energy consumption [47]. In Greece, biofuels cover 31% to 34% of transport energy demand by 2050 in scenarios that respectively reduce 60% and 70% of the country's GHG emissions [48]. On the opposite, in India, there is no significant take-up of biofuels in the transport sector in deep decarbonization scenarios, but rather a high penetration of hybrid vehicles, as biomass resources are limited and used for direct combustion in other sectors [49]. The share of biofuels in general and of butanol in particular in the energy mix of a country will depend on the availability of feedstock for their production and other low carbon options for the transport sector (e.g. clean electricity supply, hydrogen, etc.). Based on the literature review, it is realistic to assume that butanol would play an important role for GHG mitigation in several countries as a new option for decarbonization of the transportation sector.

4 Conclusion

The objective of this paper was to propose a holistic approach combining the LCA methodology and a TIMES energy system model to assess potential environmental impacts and possible market penetration of emerging energy pathways. The developed approach was then applied to the case of butanol produced from pre-hydrolysate in a Kraft dissolving pulp mill at an early stage of the design process. Indeed, recent literature has shown that the potential of biofuels to mitigate climate change highly depends on several factors such as the type of feedstock or the energy efficiency of the production process. Biofuels production can also be undermined by other sustainability issues [e.g. 3-7]. LCA is increasingly used to assess biofuel pathways. However, published studies are often limited to GHG emissions [50], leading to risks of burden shifting toward other environmental issues, and are usually not performed at the process design stage, when it would be more efficient to identify and control environmental aspects [51]. Moreover, the long-term economic profitability of biofuels depends on future energy and climate policies [13], which are usually not considered in techno-economic feasibility studies. The holistic approach developed in this paper allows identifying the variables that strongly influence the environmental performance and the market penetration of emerging biofuels, addressing two different aspects of sustainability at the same time. It also allows providing recommendations for the future design and implementation steps. This new approach can be used to help the forest sector of other countries to design more sustainable biorefinery processes. It is also replicable to assess other types of emerging energy sources (e.g. other second-generation biofuels). From that respect, the idea of combining LCA and TIMES approaches is not specific to Canada nor to a particular promising new energy source.

The LCA study identified three main contributors to potential environmental impacts: energy, fresh xylose consumption, and the treatment of detoxification residues. The sensitivity analysis performed on the level of energy integration, which is still unknown at this early design stage, showed that improving energy efficiency of the process is one of the main leverage to increase its environmental performance for the climate change, ecosystem quality, and resource depletion impact categories. Decreasing fresh xylose consumption and reducing detoxification residues would also lead to substantial improvements. Research is ongoing to develop alternative hydrolysate detoxification techniques in order to decrease or eliminate the amount of fresh xylose needed.

The study performed using the optimization energy system model NATEM-Canada also showed that the level of energy integration between the pulp mill and the butanol biorefinery seems to be a critical aspect to consider in order to make butanol a competitive fuel among all other alternative fuels. Indeed, only a very marginal amount of butanol is produced in 2050 when heat is not recovered in the production process or re-used in the Kraft pulp mill, while maximum level of production is reached at most periods in all scenarios with the assumption that 50% of the heat could be recovered replacing biomass. Natural gas is never selected as an energy source for the butanol production process by the NATEM-Canada model because of too high GHG emissions when burned. The LCA study also shows that using natural gas instead of biomass would lead to substantial increases (250 to 800%) in environmental impacts for climate change and resource depletion categories. Moreover, the comparison of butanol produced from pre-hydrolysate in a Kraft dissolving pulp mill with corn butanol and gasoline showed that its carbon footprint may be better than that gasoline if biomass energy is used and a high energy integration level is reached, and equivalent to that of corn butanol depending on the energy efficiency of the process.

Since more than half of the GHG emissions in Quebec are coming from the transportation sector, major changes are required in that sector to achieve ambitious GHG reduction targets. Under these conditions, butanol becomes a fuel in competition with other alternative fuels (electricity, Fischer-Tropsch diesel, biomethanol, cellulosic ethanol, renewable natural gas, liquefied natural gas, and hydrogen) to meet a growing transportation demand while respecting the GHG emission targets. However, in a context where ambitious GHG emission reductions are expected, all types of bioenergy, including butanol, have a role to play in the transportation sector, but also in the commercial and industrial sectors. Butanol is primarily used by the model in the air transportation segments as a large variety of alternative fuels are available for road transportation, while options for aviation are more limited. Most of the feedstock sources available for the production of bioenergy are used at their maximum potential by 2050, including pre-hydrolysate from Kraft pulp mills available for butanol production. In the medium-term, the supply cost of feedstock might affect the economic profitability of producing butanol as higher supply cost (14$/GJ) undermine the competitiveness of butanol over other next-generation biofuels. However, due to high marginal abatement costs, the supply cost of feedstock for butanol production is not a sensitive parameter in the long-term.

We have shown with the butanol case study that the proposed approach, combining LCAand a TIMES energy system model, can provide process designers and investors with very useful information regarding the environmental performance and potential market penetration of emerging energy pathways. The results could be used to focus future efforts on the aspects that highly affect environmental impacts and economic competitiveness in order to decrease the risks associated with their development. As TIMES models are used in more than 70 countries, the approach can be adapted to other geographical contexts.

Although the analysis leads to interesting conclusions, important drawbacks need to be raised. Firstly, there are many uncertainties associated with the estimation of process, techno-economic and environmental parameters for the butanol production modeled in this study, and for other bioenergy production and consumption processes modeled in NATEM-Canada. Considering that bioenergy plays a significant role in climate policy scenarios, it would be important to assess the impact of having different estimations for the most sensitive parameters on optimal solutions from the NATEM-Canada model and on life cycle environmental impacts. Secondly, energy and climate change issues are complex and the analysis did not consider all the factors that could vary and affect the optimal solutions proposed by the NATEM-Canada model. Aspects like the socio-economic growth, energy demand in the United Sates and the rest of the world, adoption of GHG emission reduction targets in neighboring jurisdictions, optimisation of the carbon market, availability of other potential disruptive technologies (e.g. biofuel from algae), and other reduction options in the industrial sector (e.g. carbon capture and storage) should be considered for a more comprehensive assessment. Thirdly, some limits are associated with the status of development of the NATEM-Canada model that does not account at this stage for non-energy GHG emissions (representing 31% of total emissions in 2013), and other air pollutants. Finally, some limits are inherent to the optimization bottom-up energy models in general which do not capture feedback on macroeconomic variable such as the gross domestic product and employment rates. Combination with a general equilibrium model would probably be beneficial. Future works will try to overcome these drawbacks.

5 Supplementary material

Table SM1: Detailed list of processes included in the LCA model (t·km: tons·kilometers, obtained from the multiplication of the mass transported by the distance)

LCA processes	Economic flow per functional unit (i.e. production of 1 kg butanol)	Comments	ecoinvent v.3.1 process
H_2SO_4 production	0.02 kg	From simulation	Sulfuric acid, RoW, production
H_2SO_4 transportation	0.01 t·km	500 km assumed	Transport, freight, lorry >32 metric ton, EURO5, RoW
Lime production	0.016 kg	From simulation	Lime, CA-QC, lime production, milled, loose
Lime transportation	0.008 t·km	500 km assumed	Transport, freight, lorry >32 metric ton, EURO5, RoW
$Fe_2(SO_4)_3$ production	0.71 kg	From simulation	Iron sulfate, RoW, production
$Fe_2(SO_4)_3$ transportation	0.36 t·km	500 km assumed	Transport, freight, lorry >32 metric ton, EURO5, RoW
Water	260 kg	No water recycling is assumed	Tap water, CA-QC, market for
Xylose production	3.6 kg	Fresh xylose input is needed, no xylose process available in ecoinvent	Sugar, from sugarcane, RoW, cane sugar production with ethanol by-product
Xylose transportation	1.8 t·km	500 km assumed	Transport, freight, lorry >32 metric ton, EURO5, RoW
Ethanol (avoided product)	0.37 kg	From simulation, Economic flow adjusted from 42% (simulation) to 95% (ecoinvent) concentration	Ethanol, without water, in 95% solution state, from fermentation, US
Acetone (avoided product)	0.16 kg	From simulation	Acetone, liquid, RoW

Energy needed to operate the process (for the biomass case)	180 MJ	Worst case: all heating must be produced, cooling is not considered	Heat, district or industrial, other than natural gas, RoW, heat production, softwood chips from forest at furnace 1000 kW
Energy needed to operate the process (for the natural gas case)	180 MJ	Worst case: all heating must be produced, cooling is not considered	Heat, district or industrial, natural gas, RoW, heat production, natural gas, at industrial furnace > 100 kW
Additional energy needed by the Kraft mill because pre-hydrolysate is diverted (for the biomass case)	21 MJ	Assumption: combustion efficiency of pre-hydrolysate in the boiler is 50%, energy content of pre-hydrolysate is 0.54 MJ/kg	Heat, district or industrial, other than natural gas, RoW, heat production, softwood chips from forest at furnace 1000 kW
Additional energy needed by the Kraft mill because pre-hydrolysate is diverted (for the natural gas case)	21 MJ	Assumption: combustion efficiency of pre-hydrolysate in the boiler is 50%, energy content of pre-hydrolysate is 0.54 MJ/kg	Heat, district or industrial, natural gas, RoW, heat production, natural gas, at industrial furnace > 100 kW
Residues (waste)	1.32 kg	Sludge from detoxification process (lime, $Fe_2(SO_4)_3$, water and organic compounds) considered treated and landfilled	Sludge from pulp and paper production, RoW (adapted for CA-QC context), treatment of, sanitary landfill
Residues transportation to treatment facilities	0.33 t·km	250 km assumed between the pulp mill and the farm	Transport, freight, lorry >32 metric ton, EURO5, RoW
Infrastructure	$1.4x10^{-10}$ pulp factory	Assumption: 10% of a pulp mill surface for a 30-year lifetime	Pulp factory, RoW, construction
Butanol transportation	0.5 t·km	500 km assumed	Transport, freight, lorry >32 metric ton, EURO5, RoW

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