


# A carbon footprint assessment of multi-output biorefineries with international biomass supply: a case study for the Netherlands

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**Abstract:** The efficient use of lignocellulosic biomass for the production of advanced fuels and bio-based materials has become increasingly relevant. In the EU, regulatory developments are stimulating the mobilization and production of bio-based chemicals / materials and biofuels from lignocellulosic biomass. We used an attributional life-cycle assessment approach based on region-specific characteristics to determine the greenhouse gas emissions (GHG) performance of different supply-chain configurations with internationally sourced lignocellulosic biomass (stem wood, forest residues, sawmill residues, and sugarcane bagasse) from the USA, the Baltic States (BS), and Brazil (BR) for the simultaneous production of lactide and ethanol in a biorefinery located in the Netherlands (NL). The results are compared with a biorefinery that uses locally cultivated sugar beets. We also compared GHG emissions savings from the supply-chain configurations with the minimum GHG saving requirements in the revised Renewable Energy Directive (RED II) and relevant fossil-based counterparts for bio-based materials. The GHG emissions ‘from cradle to factory gate’ vary between 692 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (sawmill residues pellets from the BS) and 1002 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (sawmill chips from the USA) for lactide and between 15 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> (sawmill residues pellets from the BS) and 28 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> (bagasse pellets from BR) for ethanol. Upstream GHG emissions from the conversion routes have a relatively small impact compared with biomass conversion to lactide and ethanol. The use of woody biomass yields better GHG emissions performance for the conversion system than sugarcane bagasse or sugar beets as result of the higher lignin content that is used to generate electricity and heat internally for the system. Only the sugar beet from the NL production route is able to comply with RED II GHG savings criteria (65% by 2021). The GHG savings from polylactic acid (a derivate of lactic acid) are high and vary depending on choice of fossil-based counterpart, with the highest savings reported when compared to polystyrene (PS). These high savings

are mostly attributed to the negative emission credit from the embedded carbon in the materials. Several improvement options along the conversion routes were explored. Efficient feedstock supply chains (including pelletization and large ocean vessels) also allow for long-distance transportation of biomass and conversion in large-scale biorefineries close to demand centers with similar GHG performance to biorefineries with a local biomass supply. © 2019 The Authors. *Biofuels, Bioproducts, and Biorefining* published by Society of Chemical Industry and John Wiley & Sons, Ltd.

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**Keywords:** bio-based economy; GHG emissions performance; multi-output biorefineries; lignocellulosic biomass; bio-based materials

## Introduction

It is crucial to keep the rise in global temperature to well below 2 °C, as specified by the 2015 United Nations Climate Change Conference (COP21) in 2015 (the Paris Agreement), to prevent dangerous impacts from climate change.<sup>1</sup> Meeting this target requires a significant reduction in anthropogenic greenhouse gas (GHG) emissions. These climate-change targets entail decreasing total GHG emissions between 50% and 55% for 2030 and between 80% and 95% for 2050 in developed countries.<sup>2</sup> The development of a bio-based economy is recognized as crucial for meeting such challenging targets. This will require structural changes across all sectors of the economy<sup>3,4</sup> – in particular in countries with energy-intensive industry, such as the Netherlands.<sup>4</sup> The role of biomass for energy and materials is more essential in sectors that are difficult to decarbonize or have few alternatives to biomass. For example, the substitution from fossil-based products in materials such as plastics and chemicals will likely be based mostly on biomass-derived products.<sup>5</sup> These new conditions can shift the use of solid biomass feedstock from power generation and heat to one that targets bio-based materials and advanced fuel generation.<sup>6</sup> Multi-output biorefineries have emerged as key facilitators for the successful development of the bio-based economy.<sup>7</sup>

Facilitating the development of sustainable biorefineries is one of the key actions in the EU strategy towards the development of a bio-based economy.<sup>8</sup> Lignocellulosic biorefineries are of particular importance for this. The use of lignocellulosic biomass has a crucial advantage over other biomass types by avoiding direct competition with food crops, which can have a detrimental effect on the dynamics of food supply.<sup>9</sup> Regulatory developments are also accelerating the mobilization of such lignocellulosic-based biorefining facilities; for instance, the EU has proposed ambitious targets to reduce the production of biofuels from food crops to 7% by 2030 and has invested close to €80 billion of research

funding to consolidate a lignocellulosic biorefining sector.<sup>10,11</sup> As a result, biorefineries that process lignocellulosic biomass into bio-based materials, advanced biofuels, electricity, and heat are becoming increasingly relevant in different applications for liquid fuels, bio-based chemicals, and bio-based materials.<sup>12</sup> The multi-output / parallel production (co-production) characteristics of biorefineries result in biomass supply-chain optimization by reducing, recovering, or re-using waste, residues, and energy.

The potential benefits from biorefineries will depend to a large extent on the efficiency of the supply chain.<sup>13,14</sup> Most studies investigating the supply-chain design of biorefineries or bioenergy supply chains aim at optimization from an economic perspective, i.e. a minimum cost requirements approach.<sup>15–21</sup> Nevertheless, this approach can diminish potential environmental benefits from biorefineries.<sup>22</sup> Other studies assess supply chains efficiency from a GHG perspective, with recommendations generally targeting logistics.<sup>23–31</sup> There are also concerns regarding the large-range (and sometimes poor) performance in GHG emissions for bioenergy and bio-based materials supply chains.<sup>7</sup> The majority of these studies focus particularly on supply chains with local-sourced biomass, implicitly assuming that these supply chains will have clear GHG benefits over long-distance supply chains. However, in countries such as the Netherlands, the domestic supply potential of biomass available for bioenergy and bio-based chemicals is limited.<sup>32</sup> As a consequence, the Netherlands will depend, to a large extent, on intra-EU and extra-EU imports of sustainably sourced biomass to develop its bio-based economy and biorefining sector.<sup>33,34</sup> Such developments will also have to comply with key sustainability criteria of which GHG emissions savings in comparison to fossil counterparts are considered a crucial aspect,<sup>35</sup> particularly for energy outputs that need to comply with GHG emissions-savings criteria as established in the revised Renewable Energy Directive (RED II).

The main goal of the study is to assess supply-chain options with internationally sourced lignocellulosic biomass for

multi-output biorefineries to identify optimal supply-chain design from a GHG perspective; furthermore, a domestic sugar-beet supply chain is also assessed for comparison purposes. The secondary goal of the study is to indicate the supply chains' GHG emissions savings in comparison with relevant fossil counterparts. This type of assessment can help to maximize GHG savings for the bioenergy and bio-based material sectors in the EU and guarantee sustainable production and efficient use of biomass. It can also ensure a smooth transition from a fossil-fuel-based to a bio-based economy, strengthen the bio-based materials sector from a GHG savings perspective towards future production and demand, facilitate biomass trade, contribute to sound planning for European GHG emissions reduction targets, and avoid the devalorization of biomass streams.

The Netherlands has been selected as a case study given that biomass imports are expected to continue growing in the future to meet national climate targets and potential growth in the demand for bioenergy and bio-based materials.<sup>33,34</sup> The biorefinery is assumed to be located in an existing lounge refinery cluster in the port of Rotterdam. Three lignocellulosic international supply areas relevant for the Dutch context were considered: (1) Most of the European woody biomass imports, especially wood pellets, originate from the southeastern USA.<sup>36</sup> This trade stream, driven by European demand, has increased almost ten times in the last seven years<sup>37</sup> (2) In Europe, the Baltic states' wood exports have recently developed and this area is expected to play an important role in the biomass trade sector.<sup>38</sup> (3) Recently, sugarcane bagasse pellets from São Paulo state (Brazil) are considered to have a high export potential, which meets economic, social, and sustainability criteria.<sup>39</sup> A domestic sugar beets supply stream was also considered for comparison purposes (domestic sourced sugar crops versus internationally sourced lignocellulosic biomass). This crop type is considered relevant for different applications, as in bio-based materials, for the country's bio-based economy transition.<sup>40</sup>

## Methods

### Scope

The output of the biorefinery is to a large extent determined by the characteristics of the different biomass feedstocks. Lignocellulosic biomass consists mainly of lignin, cellulose, and hemi-cellulose.<sup>41</sup> Cellulose is a polysaccharide of glucose (C6 sugar) whereas hemi-cellulose consists mainly of C5 sugars (xylose and arabinose) and some C6 sugars (galactose, glucose, and mannose). In contrast, sugar beets

(SB) only provide C6 sugars (sucrose), and beet pulp.

Currently, industrial fermentation of biofuels and bio-based chemicals is mainly carried out by employing *Saccharomyces cerevisiae* to ferment C6 sugars.<sup>42–44</sup> However, modifications in *S. cerevisiae* have allowed this yeast to ferment C5 sugars on an industrial level.<sup>44–46</sup> The employment of such yeast for industrial / commercial purposes is still under development and mainly focuses on ethanol production.<sup>44,47</sup> To ensure the comparability of the results between the different supply chains it is critical that each of the product systems assessed serves the same function. The simultaneous production of bio-based chemicals from C6 sugars (lactide) and biofuels (ethanol) from C5 sugars is therefore considered the main function of the multi-output biorefinery. Accordingly, two functional units have been defined: 1 kg of lactide and 1 MJ of ethanol.

To determine the GHG emissions from the different supply chains and the emissions hotspots, an attributional life-cycle assessment (LCA) approach is used. A 'cradle-to-factory-gate' scope is adopted in this study, i.e. including all stages from biomass production to conversion of bio-based chemicals and biofuels. The use phase and end-of-life (waste management) are excluded from the carbon footprint. This scope is sufficient to identify the best performing supply-chain design from a GHG emission perspective in line with the study goal. Upstream emissions from fuels and chemical / agriculture inputs were included for every supply chain. The distinctive conditions from the conversion routes according to regional characteristics were considered. The technical scope of the study, system conversion to ethanol and lactide, is represented in Fig. 1. It is assumed that the biorefinery will be operational by 2022. Greenhouse gas emissions other than CO<sub>2</sub> (CH<sub>4</sub> and N<sub>2</sub>O) are expressed in CO<sub>2</sub> equivalent for a global-warming potential (GWP) impact calculated over 100 years (GWP100), consistent with the characterization factors used in RED II.

The scope was extended to calculate GHG savings for ethanol and lactide. For ethanol, the use phase was included to allow a comparison with the fossil fuel counterpart as described in RED II (annex V). However, RED II assumes carbon neutrality for biofuels and CO<sub>2</sub> emissions released from the use phase are considered to be zero. For lactide, the scope is still up to factory gate but the impacts from the conversion from lactide to polylactic acid (PLA) were considered (as a proxy indicator) for the comparison with the relevant fossil-based counterparts. In addition, and for a consistent comparison with relevant fossil based counterparts, carbon emissions derived from biomass need to be addressed in terms of carbon neutrality.<sup>48</sup> The carbon uptake from plants and embedded in the materials

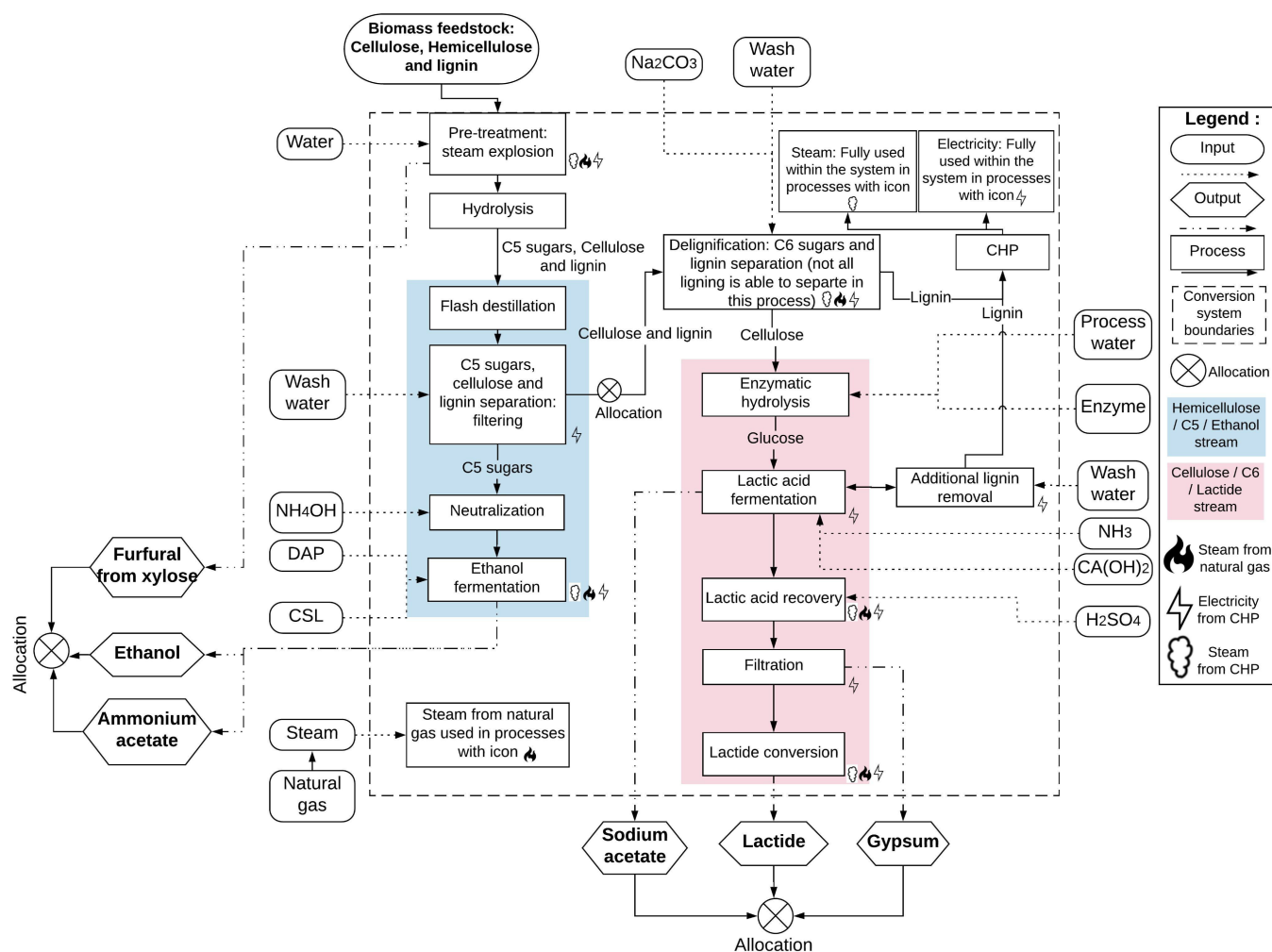


Figure 1. Biorefinery conversion system from biomass to ethanol and lactide.

is therefore considered and included as negative emission through the LCA as discussed in Kikuchi *et al.*<sup>48,49</sup>

## Investigated supply chains

The main combinations of supply chains analyzed in this study are displayed in Fig. 2 and summarized in Table 1. For the base-case supply chains, stem wood (SW) is harvested, seasoned, and collected at roadside or collected at the sugar cane mill in case of bagasse (BG) in the corresponding sourcing country. Lignocellulosic biomass is transported to a pellet plant for pelletization, domestically sourced SB is transported directly from the collection site to the biorefinery. After pelletization, pellets are transported to a terminal and shipped with a dry bulk carrier to the Netherlands. In the port of Rotterdam, pellets are processed in the biorefinery. First, pellets undergo a conversion process that separates hemicellulose, cellulose, and lignin. Hemicellulose and

cellulose are processed in C5 and C6 sugars while the lignin is used as a fuel in a combined heat and power (CHP) plant to generate heat and electricity used by the biorefinery. All the generated heat and electricity are utilized within the biorefinery (no surplus). The C5 and C6 sugars are then processed separately into ethanol and lactide respectively. In the case of SB, ethanol and lactide are both produced from C6 sugars in two separate streams.

Primary forest residues (FR) and sawmill residues (SR) are considered as feedstock for the alternative forest biomass feedstock supply chains. These feedstock types are collected and transported to a pellet facility. The energy intensity of the pelletization process from this type of feedstock was considered. For instance, grinding of SR requires less energy given their structural composition.<sup>50</sup> After pelletization, the alternative forest biomass feedstock supply chains follow the same pathway and conversion to the lactide / ethanol route as the base cases.

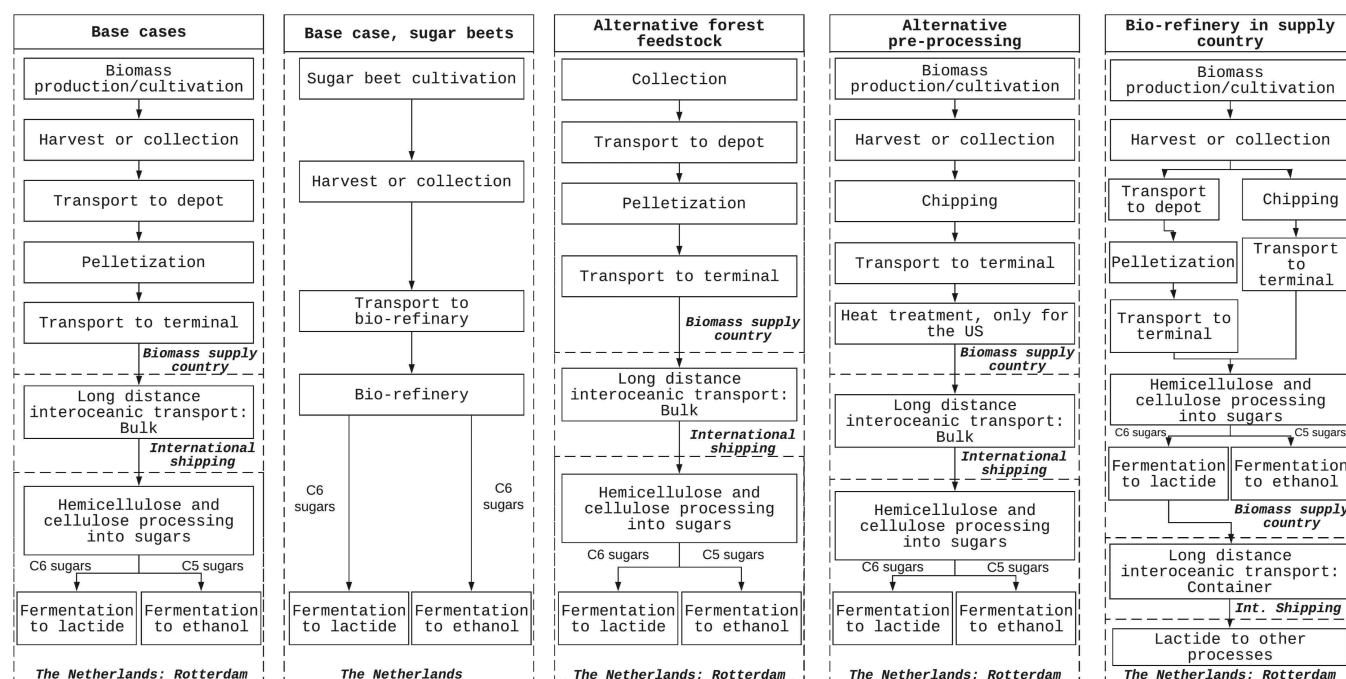


Figure 2. Flow charts of internationally sourced lignocellulosic biomass and locally sourced sugar beets processed into lactide and ethanol in the Netherlands. Conversion to ethanol and lactide in the biomass exporting country is only assessed for the sensitivity scenarios.

For the alternative pre-processing supply chains, biomass is cultivated / collected at the biomass source location. Wood is debarked and chipped on site. The bark is assumed to be left on site while wood chips are transported directly to the terminal. When wood chips are exported from the USA to the EU, phytosanitary measures (heat treatment) are required.<sup>51</sup> Dry bulk carriers are used to transport wood chips from biomass sourcing country to the Netherlands. These supply chains follow the same conversion to the lactide / ethanol routes as the base cases. Note that these supply chains are technically not feasible for BG and SR. The physical characteristics and handling properties of, for example sawdust, make it very difficult to transport over long distances in its raw form.

For sensitivity scenarios with the biorefinery located in the country of biomass supply, the biorefinery is considered to be located at the terminal from which biomass is exported in other supply chains. This is carried out to indicate the difference in GHG performance from locating the biorefinery close to biomass supply but remote from existing refinery infrastructure and demand. For these scenarios, biomass is pelletized or chipped on site and converted to lactide / ethanol as described in the other cases. After conversion, lactide is transported to Rotterdam in containers and ethanol stays in the biomass supply country. In Rotterdam, lactide is assumed to be an intermediate used for other processes.

## Inventory analysis

### Harvest and collection

For the southeast USA US-SE, wood was assumed to be supplied from a 25-year rotation softwood plantation with medium intensive forest management practices as described in Jonker *et al.*<sup>52</sup> Forestry activities include raking, spot piling, bedding, fertilizer application, thinning, clear-cut harvest and forwarding. Wood from the Baltic states is assumed to come from natural managed forest. Only diesel consumption from forestry activities related to clear cut harvesting and forwarding were therefore included. For the Netherlands, it was considered the agricultural management practices for Dutch SB cultivation presented in Smit *et al.*<sup>53</sup> In addition, agricultural equipment is used during soil preparation, pesticides / fertilizers application, and SB collection. Fertilizer-induced N<sub>2</sub>O emissions were taken into account for all scenarios and calculated in accordance with IPCC, (2006) Tier 1 methods.<sup>54</sup>

In accordance with RED II calculation methods, residues are considered to have zero GHG emissions before the process of collection.<sup>35</sup> Forest residues and sugarcane bagasse residues are therefore considered to have zero GHG emissions upstream of collection. The forest residues collection for the southeast USA and Baltic states contemplates the operations

**Table 1. Overview of the base-case supply chains and supply-chain alternatives.**

Description	Supply chain name <sup>a</sup>	Feedstock	Treatment	Biomass sourcing region	Biorefinery location
Base cases	Pellets SW US - NL C	Pulp grade stem wood	Pelletization	US-Southeast (Georgia)	The Netherlands (Rotterdam)
	Pellets SW LV - NL C	Pulp grade stem wood	Pelletization	Baltic states (Latvia)	The Netherlands (Rotterdam)
	Pellets BG BR - NL C	Bagasse	Pelletization	Brazil (São Paulo)	The Netherlands (Rotterdam)
	SB NL C	Sugar beet	n/a	The Netherlands	The Netherlands (Rotterdam)
Alternative forest biomass feedstock	Pellets FR US - NL C	Primary forest residues	Pelletization	US-Southeast (Georgia)	The Netherlands (Rotterdam)
	Pellets FR LV - NL C	Primary forest residues	Pelletization	Baltic states (Latvia)	The Netherlands (Rotterdam)
	Pellets SR US - NL C	Sawmill residues	Pelletization	US-Southeast (Georgia)	The Netherlands (Rotterdam)
	Pellets SR LV - NL C	Sawmill residues	Pelletization	Baltic states (Latvia)	The Netherlands (Rotterdam)
Alternative pre-processing: Chipping and heat	Chips SW US - NL C	Pulp grade stem wood	Chipping/heat	US-Southeast (Georgia)	The Netherlands (Rotterdam)
	Chips SW LV - NL C	Pulp grade stem wood	Chipping/heat	Baltic states (Latvia)	The Netherlands (Rotterdam)
	Chips FR US - NL C	Primary forest residues	Chipping/heat	US-Southeast (Georgia)	The Netherlands (Rotterdam)
	Chips FR LV - NL C	Primary forest residues	Chipping/heat	Baltic states (Latvia)	The Netherlands (Rotterdam)

<sup>a</sup>Biomass acronyms: SW = stemwood, BG = Bagasse, SB = Sugar beets, FR = Forest residues, SR = Sawmill residues.

of forwarding, lifting, loading, and unloading.<sup>55</sup> Table 2 includes the data used for the collection and cultivation processes. Dry matter percentage losses were also included for each step and are included in Table S1 in File S1.

## Road transport

A spatial explicit approach based on actual infrastructure and biomass availability was used to calculate aggregated road transport distances for all scenarios. This process was carried out on a geographic information system (GIS) tool. The weighted average distances are summarized in Table 3. These distances from biomass supply locations to the pellet facilities and to the export port were calculated based on the optimized total delivery routes when considering the capacity of each pellet facility and surrounding biomass availability. Pellet facilities were therefore selected based on the priorities mentioned above (i.e. distance from biomass supply locations to pellet factory gate, which is cumulative for multiple transport loads) until total cumulative biomass to be utilized at the biorefinery is achieved. When pellet facilities are not within the system (i.e. wood chips and SB scenarios), then

transport distances from the feedstock origins are optimized for and consider the local feedstock type capacity until total biorefinery demand is met.

Biomass potentials from stem wood and residues (including FR and SR) at the county level in the state of Georgia were obtained from Fingerman *et al.*<sup>37</sup> combined with the locations and capacities of actual pellet plants in Georgia.<sup>56</sup> For the Baltic states, sustainable forest biomass potentials were derived from Dees *et al.*<sup>57</sup> and combined with actual pellet plant locations and capacities of Graanul Invest, the largest pellet producer in the Baltic states.<sup>58</sup>

The sustainable potentials of SB in the Netherlands and BG in Brazil were not available. For the Netherlands SB, the geographic potential was determined based on SB suitability maps for the Netherlands. A mean suitability value was calculated,<sup>59</sup> taking into account only SB land cover area in the Netherlands for 2015.<sup>60</sup> This value was combined with the average yield for SB in the Netherlands for 2015.<sup>61</sup> The average suitability / yield relation from SB was used to derive SB yield from each corresponding suitability value for each production area. This approach resulted in a supply map (proxy indicator)

**Table 2. Harvest and collection activity data.**

Process	US southeast (US-SE)	Baltic states (BS)	Netherlands (NL)
Cultivation (kg/ha)			
N-fertilizer	582 <sup>a</sup>	-	149 <sup>d</sup>
P <sub>2</sub> O <sub>5</sub>	103 <sup>b</sup>	-	50 <sup>d</sup>
K <sub>2</sub> O	-	-	40 <sup>d</sup>
CaO	-	-	72 <sup>d</sup>
Pesticides	-	-	15 <sup>d</sup>
Seeds	-	-	3.6 <sup>d</sup>
Diesel	1105 <sup>a</sup>	1023 <sup>a</sup>	98 <sup>d</sup>
Yield	280000 <sup>c</sup>	280000 <sup>c</sup>	81390 <sup>d</sup>
Forest residues collection (kg/ha)			
Diesel	0.0052 <sup>e</sup>	0.0052 <sup>e</sup>	-

<sup>a</sup>N-fertilizer is composed of diammonium phosphate (DAP) and urea.<sup>106</sup>  
<sup>b</sup>46% of DAP.  
<sup>c</sup>52.  
<sup>d</sup>N-fertilizer is composed of synthetic fertilizer and manure.<sup>53</sup>  
<sup>e</sup>55.

**Table 3. Aggregated distances.**

Description	Supply chain name <sup>a,b</sup>	Feedstock supply location to pellet mill (km)	Pellet mill to port (km)	Feedstock supply location to port (km)	Port
Base cases	Pellets SW US - NL C	67.8	182.1	-	Savannah
	Pellets SW LV - NL C	59.3	119.2	-	Riga
	Pellets BG BR - NL C	-	364	-	Santos
	SB NL C	-	-	190	Rotterdam
Alternative forest biomass feedstock	Pellets FR US - NL C	62	182.1	-	Savannah
	Pellets FR LV - NL C	112.5	119.2	-	Riga
	Pellets SR US - NL C	73.7	182.1	-	Savannah
	Pellets SR LV - NL C	99.9	119.2	-	Riga
Alternative pre-processing: Chipping and heat	Chips SW US - NL C	-	-	111.3	Savannah
	Chips SW LV - NL C	-	-	26	Riga
	Chips FR US - NL C	-	-	77.5	Savannah
	Chips FR LV - NL C	-	-	94.3	Riga

<sup>a</sup>Biomass acronyms: SW = stemwood, BG = Bagasse, SB = Sugar beets, FR = Forest residues, SR = Sawmill residues.  
<sup>b</sup>Country-dependent emissions factors for heavy duty 30–32 ton truck for the US, Europe and Brazil were included.<sup>69,107,108</sup> In addition, empty return trips with no load were assumed.

with location and yield of SB spatially explicit defined for the Netherlands. Finally, an optimized service area was calculated that considered the distance required to supply the biomass demands at the biorefinery when considering the calculated yields. For the Brazil sugar cane BG, existing pellet facility locations were used.<sup>62</sup> Note, however, that sugarcane BG pellets in São Paulo state will likely be produced at the sugarcane mill sites.<sup>63</sup> Sugarcane BG availability (total supply) was assumed from the nearest

sugar cane mill site to the existing pellet plant.<sup>64</sup> The availability of BG (a side product of the sugar cane mills) was calculated based on the production statistics of sugar cane mills and a ratio of 26% sugarcane to BG.<sup>65,66</sup> Only 35% of the BG is estimated to be available to produce BG pellets. The remaining fraction is required to meet the internal energy demand of the sugar cane mill (mostly for drying).<sup>67</sup> Thus, 9.1% of total raw feedstock that enters each sugar cane mill is available to produce BG pellets.

## Pre-treatment (chipping, drying, pelletization)

The pelletization process of round wood includes debarking, chipping, grinding, densification, and cooling.<sup>50</sup> Pelletization of BG requires fewer processing steps than woody biomass. It includes conditioning, grinding, densification, cooling, and some miscellaneous procedures.<sup>68</sup> The energy requirements of the individual pre-processing steps at the pellet plant are summarized in Table 4. Biomass was assumed to be used as fuel for the drying process. A reduction of moisture content from 30% to 10% for wood pellets and from 50% to 10% for BG pellets was considered. The heat demand and associated biomass fuel is calculated from the required evaporation of water  $0.28 \text{ kg}_{\text{water}}/\text{kg}_{\text{pellets}}$  for wood pellets and  $0.8 \text{ kg}_{\text{water}}/\text{kg}_{\text{pellets}}$  for BG pellets and boiler efficiency of 85% based on the method and values from Thek and Obernberger<sup>50</sup> and Edwards *et al.*<sup>69</sup> Country-specific electricity mixes were considered to calculate the GHG emissions of electricity use. On-site chipping (diesel powered) for chip pathways includes debarking, chipping, and loading of feedstock.<sup>70,71</sup> For phytosanitary measures, chips are dried at a temperature at which the organisms cannot survive. The drying was assumed to be carried out in the export port. For the drying process, biomass was used as fuel and the same methodology was used as was applied for pellets. A reduction of wood chips' moisture content from 30% to 10% was considered.

## Long-distance sea transportation

Methods from Edwards *et al.*<sup>69</sup> were used to calculate GHG emissions from maritime transport. Bulk carriers are used for oceanic transports of wood chips and wood pellets. These carriers are constrained by the bulk density of the transported goods and the design load is volume limited.<sup>69</sup> The following bulk densities were assumed:

- Wood chips:  $223\text{--}328 \text{ kg}/\text{m}^3$
- Wood and BG pellets:  $650 \text{ kg}/\text{m}^3$ .

Supramax bulk carriers were considered for maritime transport with fuel (heavy fuel oil) efficiency depending on transported feedstock.<sup>69</sup> Return trips with 30% load were also taken into account (30% of total distance under ballast / empty). Electrical grab cranes are used for loading and unloading pellets / chips in Dutch terminals.<sup>72</sup> Greenhouse gas emissions from electric grab cranes were calculated based on work from Tilke *et al.*<sup>73</sup> and depend on feedstock bulk density ( $0.00097 \text{ MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$ ,  $0.0028 \text{ MJ}_{\text{electricity}}/\text{kg}_{\text{chips}}$ ). Distances between ports of export and the port of Rotterdam were based on actual maritime routes.<sup>74</sup>

**Table 4. Pelletization / chipping activity data.**

Process	Unit	Wood	Bagasse
Pelletization			
Conditioning	$\text{MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$	-	$0.022^b$
Debarking	$\text{MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$	$0.038^a$	-
Chipping	$\text{MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$	$0.13^a$	-
Grinding	$\text{MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$	$0.067^a$	$0.076^b$
Densification	$\text{MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$	$0.14^a$	$0.14^b$
Cooling	$\text{MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$	$0.005^a$	$0.022^b$
Additional	$\text{MJ}_{\text{electricity}}/\text{kg}_{\text{pellets}}$	-	$0.065^b$
Heat requirement	$\text{MJ}_{\text{heat}}/\text{MJ}_{\text{pellets}}$	$0.066^a$	$0.207^b$
Chipping and debarking on site			
Chipping	$\text{MJ}_{\text{diesel}}/\text{kg}_{\text{biomass}}$	$0.13^c$	-
Debarking	$\text{MJ}_{\text{diesel}}/\text{kg}_{\text{biomass}}$	$0.065^d$	-
a50 b68 cBased on efficiency from drum and disc chippers. <sup>70</sup> dBased on chain flail debarker. <sup>71</sup>			

## Transportation of lactide in containers (only valid for sensitivity scenarios) from biomass sourcing country

Transportation of lactide in containers is valid only for the sensitivity cases when the biorefinery is located in the country of biomass supply. Lactide is assumed to be transported in a solid state in twenty food equivalent (TEU) containers with a load volume of  $33.2 \text{ m}^3$  per container. These containers are transported in 3000–4999 TEU-type container ships. The bulk density of lactide was assumed to be similar to calcium lactate ( $700 \text{ kg}/\text{m}^3$ ). Greenhouse gas emissions from container transport by ship are calculated following Eqn 1:<sup>75</sup>

$$\frac{\text{GHG emissions}}{\text{container}} = \text{MCR} * \text{LF} * \text{AT} * \text{EF} \quad (1)$$

where

MCR = maximum continuous rating of the combustion engine in use (kW). Typically, this value corresponds to 80% of the installed engine power;<sup>76</sup>

LF = load factors,  $\text{actual speed}_{\text{km/h}} / \text{maximum speed}_{\text{km/h}}$  3 (unit less);

AT = activity time (h);

EF = power-based emission factor ( $\text{kg}/\text{kWh}$ ) for the greenhouse gas.

Maximum speed, TEU capacity, and MCR data were gathered from Rickmers Holdings group.<sup>77</sup> An actual speed of  $35 \text{ km/h}$  was assumed, which is the common navigation speed of these type of ships.<sup>78</sup> The activity time was calculated based on distance from port to port and the

actual speed. Power-based emissions factors were based on Veidenheimer.<sup>75</sup> It was assumed that 65% of the total capacity of the vessel was used for lactide transportation given that container ships carry different products at a time and 70% of container volume utilization (limited by container payload capacity). The containers are loaded using electrically powered cranes at the biomass source country terminal and unloaded at Rotterdam biorefinery. Electrical crane energy consumption (5,26 kWh/TEU) was calculated following Tran *et al.*<sup>79</sup> The GHG emissions from loading and unloading are country specific as a result of variations in the carbon intensity of electricity generation in each country.

## Conversion to lactide and ethanol

To split lignocellulosic biomass into cellulose, hemicellulose and lignin at the biorefinery, biomass undergoes a steam explosion process (Fig. 1). Furfural ( $C_5H_4O_2$ ) is formed during the steam explosion as a result of small share of xylose degradation. A share of the hemicellulose is then processed into 5-carbon sugars by a hydrolysis process. A split-up process is carried out to separate xylose from lignin and cellulose. The liquid filtered part contains the xylose and the solid filtered part the cellulose and lignin. The filtered liquid is neutralized with ammonium hydroxide ( $NH_4OH$ ). Ammonium acetate ( $C_2H_7NO_2$ ) is formed from the neutralization process. Diammonium phosphate (DAP) and corn steep liquor (CSL) are then added into the neutralized liquid where xylose is fermented into ethanol. A chemical delignification process with sodium carbonate ( $Na_2CO_3$ ) is used to remove the lignin from the filtered solid. Lignin is burned in a combined heat and power (CHP) plant to generate electricity and steam at the biorefinery. After another separation procedure, enzymes are used to process cellulose into glucose by means of enzymatic hydrolysis. Then, glucose is fermented into lactic acid.

For the lactic acid fermentation process, ammonia ( $NH_3$ ) is added and calcium hydroxide ( $Ca(OH)_2$ ) is applied to control the pH of the fermentation broth. An additional separation process is carried out to remove additional lignin, which is burned in the CHP plant. The steam and electricity from the CHP are fully used within the system. The steam from the CHP is assigned to each process with the relative share from the total steam demand. We assumed that the extra heat demand needed for each process is supplied from natural gas. The lactic acid fermentation process results in the formation of calcium lactate ( $C_6H_{10}CaO_6$ ) and in low quantities of sodium acetate ( $C_2H_3NaO_2$ ).  $C_6H_{10}CaO_6$  is treated with sulfuric acid ( $H_2SO_4$ ) to recover lactic acid. As a result of this process, lactic acid is formed and calcium sulfate / gypsum ( $CaSO_4$ ) is precipitated. The  $CaSO_4$  is removed by filtration.

For the last step, water is removed from the lactic acid and processed into lactide. Note that the quantity of chemical inputs used for lactide production in the biorefinery is output dependent.<sup>80</sup> A schematic representation of the conversion process from lignocellulosic biomass to ethanol and lactide is depicted in Fig. 1.

For the conversion of SB to lactide, it was assumed that the energy demand (power and heat) for the fermentation process of SB to lactide is similar to the SB-to-ethanol fermentation process presented in Edwards *et al.*<sup>69</sup> First, SB is processed to extract sugars. The extracted sugar juice is pasteurized and fermented into lactide. As in other pathway conversion processes,  $Ca(OH)_2$  is applied to control the pH of the fermentation broth.  $C_6H_{10}CaO_6$  is formed and is treated with  $H_2SO_4$  to recover lactic acid. Gypsum is formed from the lactic acid recovering process and is filtered. For the final process, purified lactic acid is processed into lactide. Sugar-beet pulp is an additional output generated from the fermentation process. Sugar-beet pulp undergoes a treatment process where it is pressed, dried and pelletized. For the ethanol stream, SB is processed and fermented with only sugar beet pulp as additional output.<sup>69</sup> Table 5 includes the inventory data for the different conversion routes.

## Multifunctionality

The approach chosen to deal with multi-input and multi-output systems in LCA has a strong influence on the results and is, therefore, a debated issue.<sup>81</sup> The lack of harmonization between standards and handbooks and their ambiguity generate inconsistencies when choosing a solution for multifunctionality problems. For instance, the LCA methodology standardized in ISO 14040/14044 requires that allocation should be avoided by subdivision or system expansion when feasible; when these are not possible, allocation should be in proportion to the physical properties of the products such as mass or energy, or economic value. Other standards, such as RED II, allocate the burden of impacts among products and by-products by energy content. However, this specification is frequently challenged because not all products and by-products of biorefineries are meant for energy purposes.<sup>82,83</sup>

The inclusion of the functions of ethanol and lactide provision in a single functional unit would partly avoid allocation but was considered infeasible because the share of lactide and ethanol varies between the studied alternatives as a result of the differences in cellulose and hemicellulose content. This would lead to inconsistent reference flows. System boundary expansion or substitution is not adequate for the attributional model applied in this study. Mass allocation deemed the preferred allocation method because

**Table 5. Input data for the conversion to ethanol and lactide process.**

Feature	Unit	Wood biomass		Sugarcane bagasse		Sugar beet Lactide		Sugar beet ethanol	
		Input	Output	Input	Output	Input	Output	Input	Output
Biomass (dry basis)	kt/year	747 <sup>a</sup>	-	1067.2 <sup>a</sup>	-	588.1 <sup>c</sup>	-	588.1 <sup>d</sup>	-
Steam <sup>e</sup>	kt/year	2168.9 <sup>a</sup>	-	2973.7 <sup>a</sup>	-	2318.2 <sup>d</sup>	-	1189.5 <sup>d</sup>	-
Electricity <sup>e</sup>	MWh/year	54 830.1 <sup>a</sup>	-	65 328.9 <sup>a</sup>	-	76 717.9 <sup>d</sup>	-	49 901.8 <sup>d</sup>	-
Na <sub>2</sub> CO <sub>3</sub>	kt/year	43.2 <sup>a</sup>	-	42.7 <sup>a</sup>	-	-	-	-	-
NH <sub>3</sub>	kt/year	10.2 <sup>a</sup>	-	12.2 <sup>a</sup>	-	12.2 <sup>a</sup>	-	-	-
Enzyme	kt/year	2.7 <sup>a</sup>	-	2.7 <sup>a</sup>	-	-	-	-	-
Ca(OH) <sub>2</sub>	kt/year	119.1 <sup>b</sup>	-	119.1 <sup>b</sup>	-	119.1 <sup>b</sup>	-	-	-
CSL	kt/year	0.08 <sup>a</sup>	-	0.16 <sup>a</sup>	-	-	-	-	-
DAP	kt/year	0.04 <sup>a</sup>	-	0.07 <sup>a</sup>	-	-	-	-	-
NH <sub>4</sub> OH	kt/year	9.9 <sup>a</sup>	-	14.2 <sup>a</sup>	-	-	-	-	-
H <sub>2</sub> SO <sub>4</sub>	kt/year	148.3 <sup>b</sup>	-	148.3 <sup>b</sup>	-	148.3 <sup>b</sup>	-	-	-
C <sub>2</sub> H <sub>7</sub> NO <sub>2</sub> (by-product)	kt/year	-	21.9 <sup>a</sup>	-	31.4 <sup>a</sup>	-	-	-	-
C <sub>2</sub> H <sub>3</sub> NaO <sub>2</sub> (by-product)	kt/year	-	0.01 <sup>a</sup>	-	0.01 <sup>a</sup>	-	0.01 <sup>a</sup>	-	-
C <sub>5</sub> H <sub>4</sub> O <sub>2</sub> (by-product)	kt/year	-	9.4 <sup>a</sup>	-	18.1 <sup>a</sup>	-	-	-	-
CaSO <sub>4</sub> (by-product)	kt/year	-	205.7 <sup>b</sup>	-	205.7 <sup>b</sup>	-	205.7 <sup>b</sup>	-	-
Sugar beet pulp dry (by-product)	kt/year	-	-	-	-	-	36.2 <sup>d</sup>	-	36.2 <sup>d</sup>
Lactide	kt/year	-	256 <sup>a</sup>	-	256 <sup>a</sup>	-	256 <sup>a</sup>	-	-
Ethanol	kt/year	-	63.7 <sup>a</sup>	-	122.3 <sup>a</sup>	-	-	-	181.1 <sup>d</sup>

<sup>a</sup>80.<sup>b</sup>Calculated through a stoichiometry and mass balance equation.<sup>c</sup>109.<sup>d</sup>69.<sup>e</sup>Heat and electricity demand from the lactic acid to lactide conversion process was approximated with a back-end calculation with the results from References 87–100.

(1) the majority of the by-products such as C<sub>2</sub>H<sub>7</sub>NO<sub>2</sub> or CaSO<sub>4</sub> are not energy carriers; and (2) most of the biorefinery outputs, with the exception of ethanol, are not intended for energy purposes. Alternatively, economic allocation could have been applied as demonstrated in the sensitivity analyses (Fig. 9). However, this method is sensitive to price fluctuations that are highly unstable.<sup>84</sup> There is no single correct approach to deal with multifunctionality and it is a major source of method-induced variability. Results vary substantially if alternative methods are used, as shown in Fig. 9.<sup>85</sup>

As illustrated in Fig. 1, a multi-level allocation approach was used at three specific points in the system to isolate the individual production routes of ethanol and lactide that are produced from the shared process of lignocellulosic biomass conversion to fermentable sugars. Lignin is also an intermediate output of this shared process but is used entirely within the system to generate electricity and heat. The allocation factors are displayed in Table 6. First, mass allocation was applied between the C5 and

C6 sugars (intermediate products) to share the burden of GHG emissions up to this conversion to sugars step and to allocate the heat generated by natural gas in the biorefinery and its associated GHG emissions to the individual downstream production routes of ethanol (C5) and lactide (C6). The share depends on feedstock-specific hemicellulose, cellulose, and lignin content (Table S2 in File S1). After this separation, two different process routes can be clearly distinguished: C5 to ethanol and C6 to lactide. Each process route produces by-products. C<sub>2</sub>H<sub>3</sub>NaO<sub>2</sub> and gypsum are by-products of lactide production from C6 sugars. C<sub>2</sub>H<sub>3</sub>NaO<sub>2</sub> is considered a by-product even if it is produced in almost negligible amounts compared to lactide. C<sub>5</sub>H<sub>4</sub>O<sub>2</sub> and C<sub>2</sub>H<sub>7</sub>NO<sub>2</sub> are by-products from ethanol production from C5 sugars. C<sub>5</sub>H<sub>4</sub>O<sub>2</sub> is commonly used for bio-based chemicals applications and C<sub>2</sub>H<sub>7</sub>NO<sub>2</sub> (a salt) as food additive or buffering substance.<sup>86</sup> Mass allocation was applied at sub-level between the individual production outputs of lactide and ethanol and associated by-products.

**Table 6. Allocation factors for main results.**

Feedstock	Process	Mass allocation factor (%)								
		C6 sugars	C5 sugars	Lactide	C <sub>2</sub> H <sub>3</sub> NaO <sub>2</sub>	CaSO <sub>4</sub>	Ethanol	C <sub>2</sub> H <sub>7</sub> NO <sub>2</sub>	C <sub>5</sub> H <sub>4</sub> O <sub>2</sub>	Sugar beet pulp
Wood	Sugars portioning <sup>a</sup> conversion	69.14	30.86	-	-	-	-	-	-	-
	To lactide	-	-	55.43	0.02	44.55	-	-	-	-
	To ethanol	-	-	-	-	-	67.01	23.10	9.89	-
Bagasse	Sugars portioning <sup>a</sup>	53.87	46.13	-	-	-	-	-	-	-
	To lactide	-	-	55.43	0.02	44.55	-	-	-	-
	To ethanol	-	-	-	-	-	71.22	18.27	10.51	-
Sugar beet	To lactide	-	-	51.62	0.02	41.49	-	-	-	6.87
	To ethanol	-	-	-	-	-	84.15	-	-	15.85

<sup>a</sup>Process generates C5/C6 sugars and lignin. The lignin is used in a CHP plant to generate heat and electricity, which is used within the system boundaries (no surplus).

Although the biorefinery is a single facility with interconnected energy flows, the multi-level allocation approach avoids overestimation or underestimation of GHG emissions for the two functions of the system that were considered. The production of lactide from C6 sugars is more resource and energy intensive than the production of ethanol from C5 sugars. If the environmental burden (GHG emissions) would be distributed proportionally over the outputs of the total system, part of the environmental burden resulting from the lactide production process would be allocated to ethanol.

The ethanol results in the section headed ‘GHG emission savings in comparison to fossil-based counter parts’ are presented following RED II methods. For this section and to allow a consistent comparison with the fossil fuel counterpart presented in RED II, energy allocation has been applied considering all the system outputs (without multi-level allocation) and RED II calculation rules. The energy allocation factors applied for that section are present in Table 7.

## GHG emission saving criteria

The RED II has established minimum GHG emissions-saving requirements for bioenergy in comparison with its fossil fuel counterparts. From October 2015 until December 2020, biofuels should save at least 60% of GHG emissions; from January 2021 onwards, biofuels should save at least 65% of GHG emissions. This signifies that biofuel conversion routes must emit no more than 37.6 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> from 2015 (after October) to 2020 and 32.9 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> from 2021 onwards. For bio-based materials, there are no

**Table 7. Energy allocation factors.**

Feedstock	Product	Energy allocation factor (%)
Wood	Lactide	70.32
	Ethanol	26.23
	Sodium acetate	-
	Gypsum	-
	Ammonium acetate	-
	Furfural	3.45
	Total	100
Bagasse	Lactide	55.25
	Ethanol	39.54
	Sodium acetate	-
	Gypsum	-
	Ammonium acetate	-
	Furfural	5.21
	Total	100
Sugar beet	Ethanol	90.80
	Sugar beet pulp	9.20
	Total	100

binding minimum GHG emissions saving requirements or other sustainability criteria at the EU level. Lactide is an intermediate for polylactic acid (PLA) production. The emissions from the ring-opening polymerization of lactide for PLA production reported in Vink and Davies<sup>87</sup> were used and added as a proxy indicator to give a better comparison between the lactide supply chains and fossil-based counterparts. Three fossil-based polymers were considered for the comparison exercise: (1) polystyrene (PS) 2.25

kg CO<sub>2eq</sub>/kg, for polypropylene (PP) 1.63 kg CO<sub>2eq</sub>/kg and for polyethylene terephthalate (PET) 2.2 kg CO<sub>2eq</sub>/kg.<sup>88–90</sup>

## Results

### Carbon footprint of the supply chains

Figure 3 presents the total sum of ‘cradle-to-factory-gate’ GHG emissions for 1 kg of lactide production for the assessed supply chains. The GHG emissions are calculated to be between 692 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (SR pellets from the Baltic states) and 1002 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (SW chips from the USA). The conversion process of lignocellulosic biomass to lactide is the main contributor to the total GHG emissions. Those GHG emissions are mainly related to process energy demand supplied from natural gas and upstream emissions from chemicals used in the process. The use of Ca(OH)<sub>2</sub> for pH control during lactic acid fermentation is one of the main contributors. Conversion routes from the USA and Baltic states make use of the same feedstock (wood biomass) and therefore share the same conversion system and associated GHG emissions from chemicals and heat supply. Differences in conversion to lactide process between conversion routes are mainly attributed to each system’s heat demand and feedstock lignin content. Feedstocks with a higher lignin content (burned in the CHP) can supply net heat to a larger extent than feedstocks with a lower lignin content; this results in lower amounts of natural gas needed to meet total heat demand for lactide production. Thus, the higher lignin content from wood (US and Baltic states conversion routes), in comparison to BG (Brazil conversion route) and SB (no lignin present in SB, Netherlands conversion route), results in better GHG emissions performance for the conversion to lactide step. For all conversion routes, with the exception of

the SB one, the amount of electricity generated at the CHP from burning lignin is enough to cover the system’s electricity demand completely. The absence of lignin in SB determines that heat and electricity requirements are supplied entirely from natural gas and the grid, which results in an additional impact.

For ethanol, the total sum of ‘cradle-to-factory-gate’ GHG emissions varies between 15 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> (SR pellets from the Baltic states) and 27 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> (BG pellets from Brazil) as shown in Fig. 4. The conversion process of lignocellulosic biomass to ethanol is the main contributor to GHG emissions. Those emissions are strongly related to the system’s heat demand and the upstream emissions from NH<sub>4</sub>OH used in the neutralization process; it is worth noting that the SB conversion route omits the use of NH<sub>4</sub>OH. The GHG emissions from the use of DAP fertilizer are also omitted.

The large difference between second-generation (lignocellulosic biomass) production routes is attributed to the variance in biochemical composition between wood biomass pellets and sugarcane BG pellets (see above). These differences mean that the burden of GHG emissions from the Brazil pathway is shared more uniformly between lactide and ethanol production than for other biomass types. Although ethanol results follow a similar trend to lactide, the biophysical composition that determines the share of the burden between ethanol and lactide for the sugarcane BG pathway also results in a slightly higher GHG emissions footprint than the SB conversion route; this is contrary to the results trend for lactide. The first-generation ethanol production route (ethanol from SB in the Netherlands) emits one-third more (fossil) GHG emissions compared to second-generation ethanol from wood biomass. However, compared to the sugarcane BG, the impact is almost identical. High

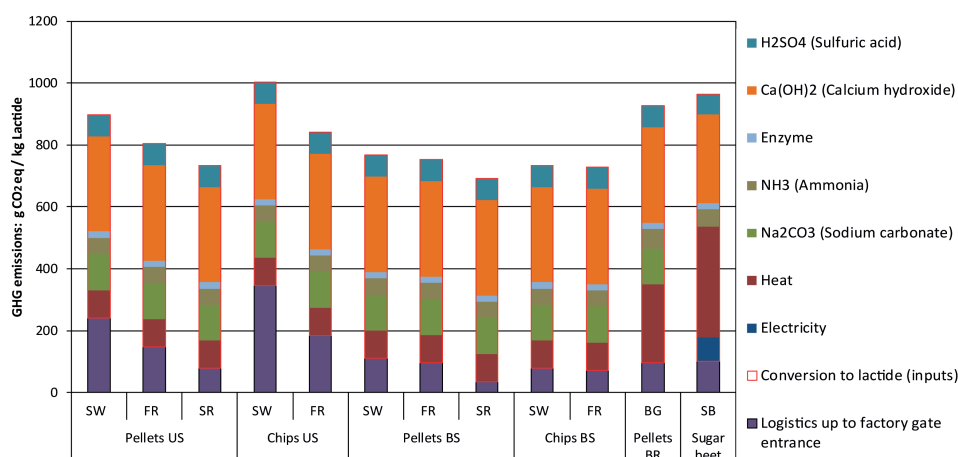


Figure 3. ‘Cradle-to-factory-gate’ GHG emissions of 1 kg lactide production.

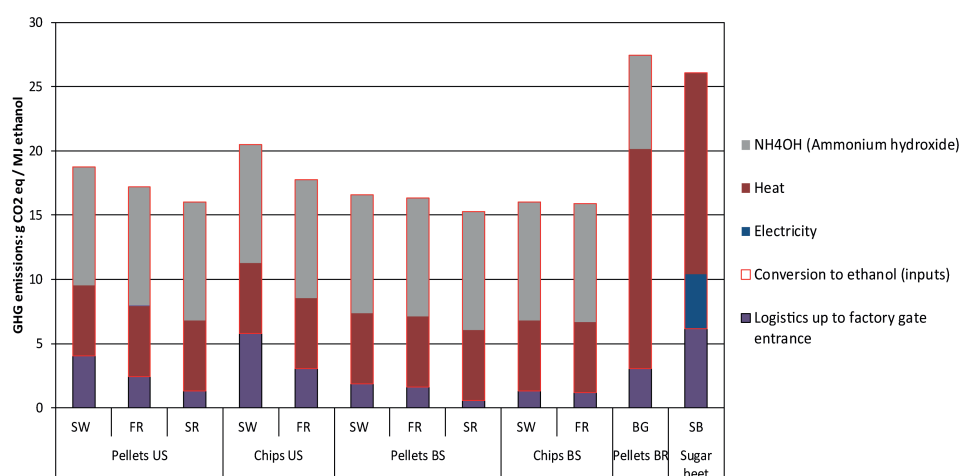


Figure 4. 'Cradle-to-factory-gate' GHG emissions of 1 MJ ethanol production.

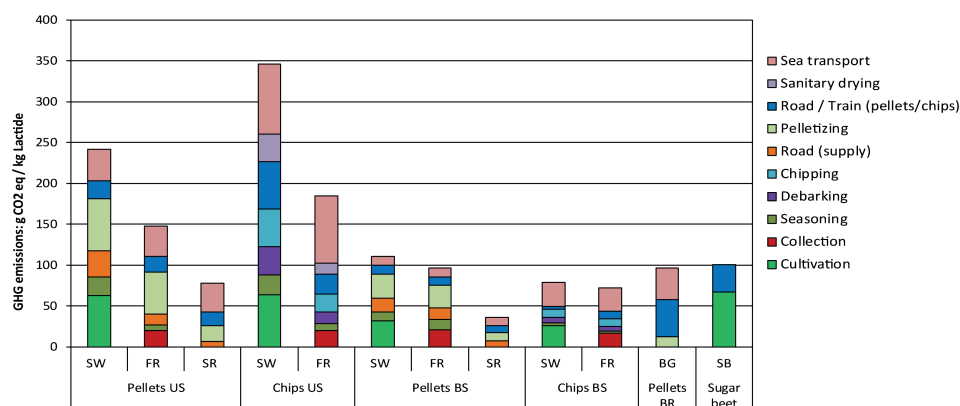


Figure 5. 'Cradle-to-factory-gate' entrance (logistics) GHG emissions of 1 kg lactide production.

emissions from the SB conversion to ethanol process are related to steam and electricity demand, which are provided from natural gas and grid electricity.

### GHG emissions upstream of feedstock supply up to the factory gate

Figure 5 presents the total sum of upstream GHG emissions of feedstock supply up to the factory entrance gate in the Netherlands for 1 kg of lactide production. Greenhouse gas emissions are lowest for SR pellets from the Baltic states ( $36 \text{ g CO}_{2\text{eq}}/\text{kg}_{\text{lactide}}$ ) and highest for SW chips from the USA ( $346 \text{ g CO}_{2\text{eq}}/\text{kg}_{\text{lactide}}$ ). The GHG emissions from the logistics in the conversion routes are considerably lower than the GHG emissions from the conversion to lactide step (Fig. 3), with the exception of SW chips from the USA. For this conversion route, the logistics emissions account for one-third from the total. The higher GHG emissions performance of wood

residues (FR and SR versus SW) pathways can be accredited to the lack of a cultivation phase and the feedstock features advantages for pelletization. For instance, there are no requirements for chipping or grinding SR when pelletization is carried out. There is a tradeoff between benefits from wood on-site processing (debarking and chipping) and pelletization. On-site processing has a slightly lower impact than pelletizing. There is also a shorter transport distance as the travel from pellet facilities is omitted. However, bulk oceanic transport of (heat-treated) chips is less efficient. The lower bulk density of chips in comparison to wood pellets results in higher maritime transport emissions. Differences in GHG emissions from the pelletization process between locations arise mostly from regional characteristics in electricity production. To illustrate, Brazil's electricity carbon mix intensity is lower than the one from Latvia (Baltic states) or the USA. The availability of feedstock types can vary depending on local conditions and it is a key aspect of GHG

emissions performance. For example, the availability of forest residues is comparatively lower than that of other wood biomass sources in the Baltic states.

The GHG emissions from SB are accredited to relatively large agricultural inputs including N-fertilizers, and transport of wet SB (76% moist) from the supply area to the biorefinery. Large amounts of agricultural land area are required to supply the necessary quantity of SB to meet annual feedstock demand for lactide production in the Netherlands. For the Brazil conversion route, there is a high impact for transport given the long distance between pellet plants located inland and the port of Santos. Overall, the superior GHG performance of the Baltic states' pathways is related to shorter maritime transport distances between ports (Riga and Rotterdam), biomass availability, and shorter transportation distances between biomass supply areas and pellet plants.

In terms of upstream GHG emissions before lignocellulosic biomass conversion to ethanol, emissions vary between 0.6 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> (SR pellets from Baltic states) and 6.5 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub> (SB from the Netherlands) as shown in Fig. 6. The upstream GHG emissions before the conversion to ethanol are comparably higher for the Netherlands pathway than for any other conversion route. Like the results for lactide, high emissions from the Netherlands scenario are attributed mainly to high input of N-fertilizers for the SB production. The impact from N-fertilizers includes the direct and indirect N<sub>2</sub>O emissions from managed soils and fossil inputs for fertilizer production. There is also no division of the burden between ethanol and lactide logistics for SB as in other conversion routes. The system design mentioned above results in considerably larger GHG upstream emissions for the SB conversion. The rest of the conversion routes results follow a similar trend to lactide.

## GHG emission savings in comparison to fossil-based counter parts

Figure 7 compares 1 kg lactide production with fossil-fuel counterparts. The error bars for the US and Baltic states supply chains represent the range of GHG emission between the different supply chains composition valid for each conversion route (only valid for woody supply chains). All conversion routes report high GHG savings in comparison with the selected fossil-based polymers. On average, the highest GHG emissions savings are reported when the conversion routes are compared with fossil-based PS; GHG emissions savings are reported to be between 2916 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (Netherlands supply chain) and 3151 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (Baltic states supply chains). The high GHG emissions savings in comparison with fossil-based counterparts are mainly attributed to the carbon neutrality advantage that are accounted as negative emissions from the embedded carbon in the materials. All conversion routes nevertheless report GHG emission savings if the carbon neutrality characteristic is omitted. The performance of the conversion routes is also strongly related to the GHG emissions generated to supply heat and electricity demand from these conversion routes. However, the impact from heat and electricity is bounded to the fossil inputs used to generate them. This impact can therefore be reduced significantly in the future with the reduction of fossil inputs. To illustrate, in 2014, 70% of the electricity used in the Netherlands was produced with fossil inputs; for 2035 it is expected that more than 50% of the electricity will be generated with renewable sources.<sup>91</sup> These changes can reduce considerably the impact from the conversion routes that have a higher electricity and heat demand (Brazil and Netherlands routes).

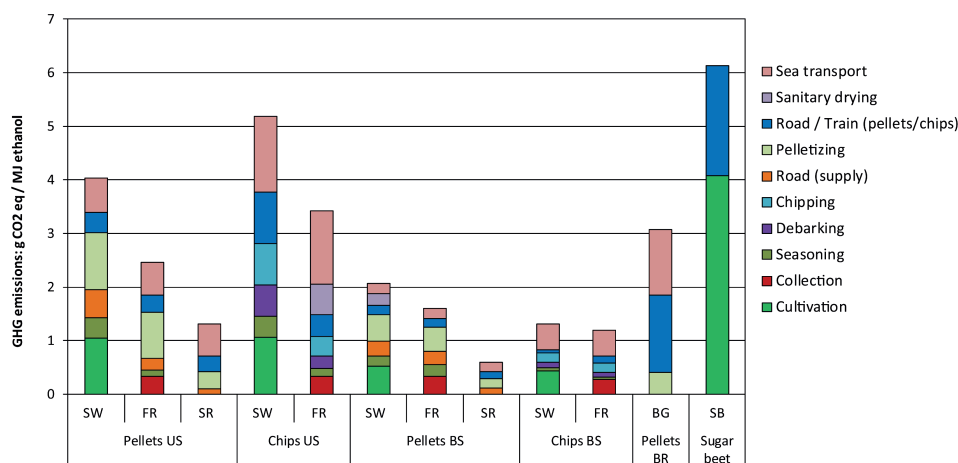


Figure 6. 'Cradle-to-factory-gate' entrance (logistics) GHG emissions of 1 MJ ethanol production.

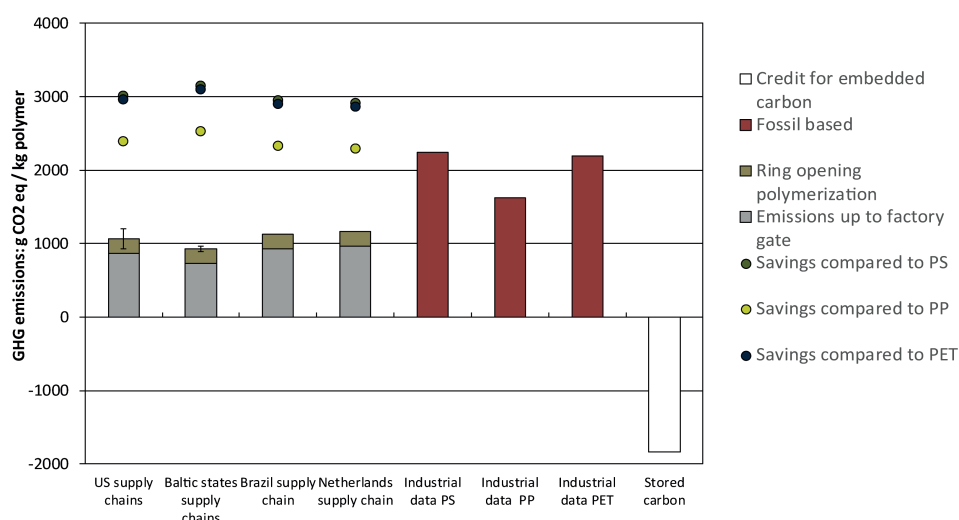


Figure 7. 'Cradle-to-factory-gate' GHG emissions of 1 kg lactide production in comparison with fossil-fuel counterparts.

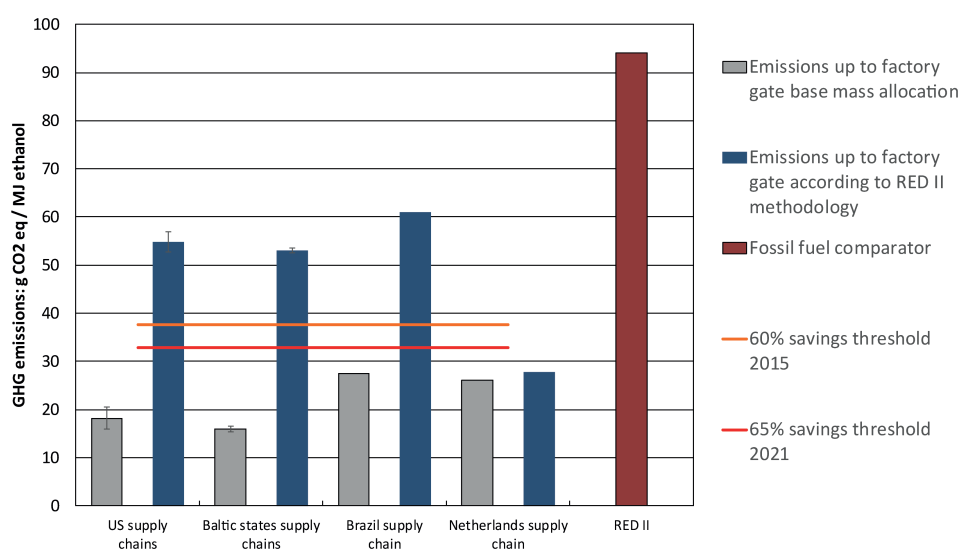


Figure 8. 'Cradle-to-factory-gate' GHG emissions of 1 MJ ethanol production (in accordance with RED II GHG calculation rules) in comparison with fossil-fuel counterpart.

Figure 8 displays the comparison of 1 MJ ethanol production in accordance with RED II GHG calculation rules and the fossil-fuel counterpart. All conversion routes report GHG emissions savings in comparison with the fossil fuel reference established in RED II. Unlike bio-based materials, there are already established mandatory savings criteria for GHG emissions in biofuels. None of the conversion routes with the exception of SB from the Netherlands are able to comply with RED II GHG savings criteria for 2015 and 2021. The difference in performance between conversion routes is strongly related to the energy allocation rules in agreement

with RED II and the different system designs of the SB and lignocellulosic conversion routes. For lignocellulosic conversion routes the burden between lactide, ethanol and by-products is spread with energy allocation. However, several by-products are allocated zero emissions as they are not energy carriers. This characteristic in combination with the high energy content of ethanol (in comparison with other outputs such as lactide) dictates that a large share of the total generated GHG emissions are allocated to ethanol. Considerable large improvements along the lignocellulosic biomass conversion routes are needed to reduce GHG

emissions and meet the GHG emissions savings criteria from RED II.

## Discussion

For illustrative purposes, the figures present in the discussion section exclude the net GHG emissions reported for lactide.

### Implications of methodological choices

The LCA attributional approach adopted in this study is static, independent of a policy and economic context.<sup>92</sup> This approach is unsuitable for assessing the possible consequences of policy choices including the potential GHG mitigation impact of developing a biorefinery in the Netherlands. Important aspects that are not included are counterfactual and marked-mediated impacts including emissions from (indirect) land-use change and possible carbon debt.<sup>93,94</sup> (Indirect) land use change is most relevant for production systems that are land intensive, such as the SB supply chain. Carbon debt and payback time are most relevant for the systems that use slow-growing biomass (SW and FR).

The fossil-based counterpart comparison exercise was based on 'cradle-to-factory-gate' emissions per kg of polymer. Nevertheless, the different density and the end-of-life of plastic materials should also be incorporated in such exercise. Different material densities can lead to different weights within the same plastic application (e.g. a plastic cup). It can be more significant to compare bio-based materials using a volume reference and account for plastic items produced by volume filling techniques, such as injection molding. In terms of end of life, the incorporation of this stage in such comparator exercises should be in line with policy legislation targets. An adequate comparator should include at least the GHG emissions from incineration to account for the carbon neutrality advantage of the bio-based polymer, as in Kikuchi *et al.*<sup>48</sup> To illustrate, if a policy has a target in terms of GHG emissions savings for 2030 and the percentage of plastic materials incinerated is forecast to be, for example, 25% in the same year, the fossil fuel comparator should be 'cradle-to-gate' emissions plus 25% of the emissions that would be released by burning that polymer. This suggestion can be an adequate proxy even if it does not consider other possible end-of-life options but the following consideration must be accounted for: (1) The near future infrastructure may not allow PLA recycling;<sup>95</sup> (2) there are environmental trade-offs in shifting from a recyclable (i.e. PP, PS and PET) to a compostable polymer;<sup>96</sup> and (3) there are large uncertainties about the landfilling emissions of PLA.<sup>97,98</sup>

### Impact of allocation methods

Different allocation procedures can be applicable to deal with multifunctionality and could significantly impact the results.<sup>94,99</sup> In this study, a mass balance allocation with an intermediate allocation step approach was determined as the most suitable allocation method for the system composition and is defined as base allocation (see above). To understand the impact of allocation procedures in multifunctional systems and determine the robustness of the results, a sensitivity analysis was carried out. For this approach, the burden of GHG emissions was allocated to the biorefinery outputs based on physical or other types of relationship between products; considering the conversion to lactide / ethanol as a unified process and not as separated streams (after C5 and C6 sugars division) as illustrated in Fig. 1. Allocation was applied based on (1) the total mass outputs from the main products and by-products (mass allocation); (2) the energy content of products and by-products as suggested in RED II (energy allocation); and (3) the market value of the products and by-products (economic allocation). For illustrative purposes, the sensitivity allocation analysis was only carried out for the base cases displayed in Fig. 2. The resulting allocation factors are reported in Table 8.

Figure 9 displays the 'cradle-to-factory-gate' GHG emissions for 1 kg of lactide and 1 MJ of ethanol production for the base conversion routes when applying mass, energy, and economic allocation. The results show that the chosen allocation method has a strong impact on the GHG emissions of both lactide and ethanol production. The use of mass allocation leads to a better GHG emissions performance for lactide and slightly worse performance for ethanol in comparison with the base approach used in the base cases. When mass allocation is applied, a larger share of GHG emissions is allocated to gypsum. Large quantities of gypsum are produced from the lactic acid recovery process, which results in a considerable mass output of gypsum from the system. This allocation procedure entails that the burden of GHG emissions is spread out more evenly between gypsum and lactide, and, to a lesser extent, to ethanol. This characteristic also applies for the base allocation method. However, with the base approach, emissions upstream from the factory gate are only allocated to the main products of the system. The production of gypsum is not considered a primary function of the system. Mass allocation can lead to inappropriate conclusions as large part of the GHG emissions burden from the lactide production and before conversion are allocated to gypsum.

Applying energy allocation as suggested by RED II leads to considerably higher GHG emissions for ethanol in all

**Table 8. Mass, energy and economic allocation factors.**

Feedstock	Product	Mass flow (kg/year)	Mass allocation factor (%)	LHV (MJ/kg)	Energy allocation factor (%)	Price (€/kg)	Economic allocation factor (%)
Wood	Lactide	255 987 048.9	45.97	18.72 <sup>a</sup>	70.32	2.47 <sup>d</sup>	89.65
	Ethanol	63 747 939.9	11.45	26.81 <sup>b</sup>	26.23	0.66 <sup>e</sup>	6.03
	Sodium acetate	11 689.6	0.00	-	-	0.79 <sup>f</sup>	0.00
	Gypsum	205 757 911.6	36.95	-	-	0.008 <sup>g</sup>	0.23
	Ammonium acetate	21 976 441.3	3.95	-	-	0.84 <sup>h</sup>	2.61
	Furfural	9 410 719.4	1.69	23.98 <sup>c</sup>	3.45	1.1 <sup>i</sup>	1.47
	Total	556 891 750.8	100	-	100	-	100
Bagasse	Lactide	255 987 048.9	41.00	18.72 <sup>a</sup>	55.25	2.47 <sup>d</sup>	83.00
	Ethanol	122 332 237.7	19.31	26.81 <sup>b</sup>	39.54	0.66 <sup>e</sup>	10.71
	Sodium acetate	16 692.7	0.00	-	-	0.79 <sup>f</sup>	0.00
	Gypsum	205 757 911.6	32.48	-	-	0.008 <sup>g</sup>	0.22
	Ammonium acetate	31 382 358.1	4.95	-	-	0.84 <sup>h</sup>	3.45
	Furfural	18 059 161.9	2.85	23.98 <sup>c</sup>	5.21	1.1 <sup>i</sup>	2.61
	Total	633 535 410.9	100	-	100	-	100
Sugar beet	Lactide	255 987 048.9	51.62	18.72 <sup>a</sup>	90.29	2.47 <sup>d</sup>	99.47
	Sodium acetate	16 692.7	0.00	-	-	0.79 <sup>f</sup>	0.00
	Gypsum	205 757 911.6	41.49	-	-	0.008 <sup>g</sup>	0.26
	Sugar beet pulp	34 111 554.2	6.88	14.43 <sup>b</sup>	9.71	0.05 <sup>j</sup>	0.27
	Total	495 873 207.5	100	-	100	-	100
Sugar beet	Ethanol	181 144 115.4	84.15	26.81	90.80	0.66 <sup>e</sup>	98.62
	Sugar beet pulp	34 111 554.2	15.85	14.43 <sup>b</sup>	9.20	0.05 <sup>j</sup>	1.38
	Total	215 255 669.6	100	-	100	-	100

<sup>a</sup>Assumed from PLA.<sup>110</sup><sup>b</sup>69.<sup>c</sup>111.<sup>d</sup>Price of raw lactide.<sup>112</sup><sup>e</sup>Average price of ethanol between 2016–2018.<sup>113</sup><sup>f</sup>Average price from sodium acetate reported in e-commerce webpage.<sup>114</sup><sup>g</sup>115.<sup>h</sup>Average price from ammonium acetate reported in e-commerce webpage.<sup>116</sup><sup>i</sup>117.<sup>j</sup>Average price from sugar beet pulp for the Netherlands.<sup>118</sup>

scenarios with the exception of the Netherlands SB case. When energy allocation is used for lignocellulosic biomass conversion routes, the burden of GHG emissions is spread only between lactide, ethanol, and furfural. The higher energy content of ethanol dictates that a larger share of GHG emissions is allocated to ethanol compared with other allocation methods. The relatively larger share of hemicellulose in BG entails that larger quantities of ethanol are produced in comparison to other conversion routes. This results in higher GHG emissions for ethanol in this conversion route as a result of the higher energy content of ethanol compared to lactide. A smaller share of emissions

is allocated to lactide when energy allocation is applied compared to the base cases. Nevertheless, most of the GHG emissions from chemicals inputs at the conversion step and heat demand derive from lactide production. Thus, energy allocation over all outputs of the biorefinery without an intermediate allocation step leads to a considerable burden of GHG emissions from lactide production being allocated to ethanol. Still, RED II has no clear suggestions and solutions of how to avoid such inaccuracy in burden share for such multi-output systems and the results of such exercise with energy allocation are a shortcoming that can lead to inadequate conclusions.

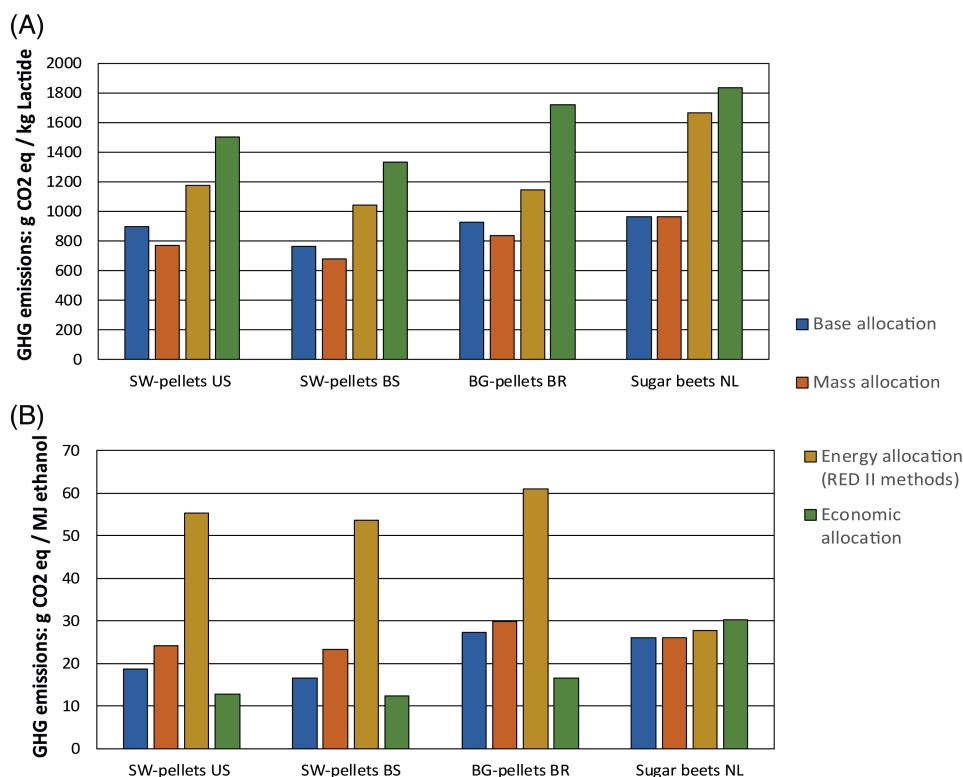


Figure 9. Sensitivity analysis results for (A) lactide and (B) ethanol between different allocation procedures.

The use of economic allocation shifts the burden to lactide and away from ethanol compared to the other allocation methods; with the exception of ethanol from the Netherlands conversion route. The higher price from lactide determines that a large share of the GHG emissions burden from the lactide / ethanol production are allocated to lactide. However, economic allocation is bound to the market volatility that leads to instability and price fluctuations. To illustrate, the market value for ethanol used in this study is a two-year average from 2016 to 2018 (0.66 €/kg); in this same period of time ethanol prices fluctuated between 0.83 €/kg and of 0.55 €/kg. Still, economic allocation reflects a higher degree of relatedness in terms of outputs and the system's primary function.

## Inventory data limitations

No foreground data for the conversion from calcium lactide to lactide, electricity and heat demand were available. A stoichiometry and mass balance equation, as suggested by Vink and Davies,<sup>87</sup> was applied to calculate the amount of  $\text{Ca}(\text{OH})_2$  and  $\text{H}_2\text{SO}_4$  required for the conversion process. Heat and electricity demand from the lactic acid to lactide conversion was approximated with a back-end calculation

following the results from Vink *et al.*<sup>100</sup> and Vink and Davies.<sup>87</sup> The biorefinery for this study was assumed to produce 272 kt<sub>lactide</sub>/year. However, current PLA/lactide facilities have a much lower capacity. The NatureWorks corn-based facility in Blair, Nebraska, has a capacity of 150 kt<sub>PLA</sub>/year. Meanwhile, Corbion recently finalized their sugarcane lactide plant expansion (100 kt<sub>lactide</sub>/year) and a 75 kt<sub>PLA</sub>/year plant in Thailand.<sup>101</sup>

## Improving hot-spots in GHG emissions performance

The assessment of the conversion routes is affected by the choice of input parameters such as transport modes and distances, choice of heat supply, yields and other assumptions about the life-cycle inventory. These parameters can strongly steer the magnitude of the results and the direction of the analysis. To understand the impact of these parameters, a sensitivity analysis was carried out with the base-case scenarios displayed in Fig. 2. These conversion routes were selected for illustrative purposes. In addition, the variation of assumptions for the sensitivity analysis base-case scenarios has an equivalent impact if applied to the other conversion routes (e.g. alternative forest feedstock). The relevant parameters for the sensitivity analysis were selected based on

their influence on the results (GHG hotspots in the supply chains). As a result, three processes along the conversion routes were selected for this analysis: (1) the conversion step; (2) transport of feedstock; and (3) cultivation (higher yields). The variations in inputs are applied with a view to superior GHG emissions performance for the selected processes and are based on plausible scenarios. The sensitivity conversion routes are defined as best cases.

For the conversion step (1), the majority of the GHG emissions result from the use of chemicals in the process. However, no distinct improvement potential could be identified with the limited information and data available. Improvements were therefore limited to alternative sources of heat supply. Different transport modes and shorter feedstock transportation distances were assumed for the feedstock transportation process (2), and higher yields were considered mainly for the cultivation process in the SB conversion route (3). Table 9 includes a summary of the parameters and description adopted for the sensitivity analysis base cases.

Figure 10 displays the comparison between the base supply chains with the best performing alternatives assessed in the sensitivity analysis. As shown in Fig. 10(A), the best performance conversion routes have a reduction of

15 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (SW pellets from the US), 52 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (SW pellets from Baltic states), 276 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (BG pellets from Brazil) and 156 g CO<sub>2eq</sub>/kg<sub>lactide</sub> (SG from the Netherlands) when compared with the base scenarios. There is a tradeoff between GHG emissions from logistics and conversion to lactide for the woody lignocellulosic biomass scenarios. The use of biomass to cover system heat demand as a natural gas replacement results in a reduction of GHG emissions at the conversion stage. However, the mobilization of additional biomass to supply heat demand generates an increase in logistics GHG emissions (Fig. 10(B)). To illustrate, the reduction of 89 g CO<sub>2eq</sub>/kg<sub>lactide</sub> in conversion to lactide GHG emissions from biomass heat supply for the SW pellets from the USA is offset by 74 g CO<sub>2eq</sub>/kg<sub>lactide</sub> due to the increase of GHG emissions in logistics; the same GHG emissions tradeoff occurs for the SW pellets from the Baltic states.

For the best BG pellets from the Brazil scenario, the increase in emissions due to additional supplied biomass is counterbalanced by the use of trains for pellets transport. Transporting pellets by trains is more efficient than trucks and generates lower GHG emissions. To ensure this advantage, the location of the cargo terminal is of paramount

**Table 9. Sensitivity analysis parameters for best case scenarios.**

Base cases	Parameter	Value	Unit	Description/assumption
US pellets – SW – NL C	Heat supply	1.07 <sup>a</sup>	kg <sub>dry pellets</sub> /kg <sub>dry pellets</sub>	Heat demand supplied by burning additional (wood or bagasse) pellets at the CHP instead of natural gas
BS pellets – SW – NL C		1.07 <sup>a</sup>	kg <sub>dry pellets</sub> /kg <sub>dry pellets</sub>	
BR pellets – BG – NL C		1.20 <sup>a</sup>	kg <sub>dry pellets</sub> /kg <sub>dry pellets</sub>	
BR pellets – BG – NL C	Transport	100 km by truck and 264 km by train	km	Large-scale bagasse pellet production could use the railway system in São Paulo state to transport pellets to port (COSAN, 2018). Sugarcane bagasse pellets are transported from pellet facilities by road to a storage / load terminal (2) from the terminal, pellets are loaded in freight trains and transported to the port of Santos.
SB NL C	Heat supply	3.58 <sup>b</sup>	MJ <sub>steam</sub> /kg <sub>lactide</sub>	It is possible that sugar beet conversion facilities / refineries include a biogas plant to process waste slop and use the heat from this procedure (biogas) for the conversion process to lactide and ethanol
	Transport	50	km	Sugar beet cultivation can increase in the proximity of the plant driven by bio-refinery biomass demand. Therefore, a shorter transportation distance from the biomass supply area to the biorefinery can be considered.
	Yield	120	t <sub>sugar beets</sub> /ha	The suitability method applied in this paper to calculate spatially explicit sugar beet yields can achieve values as high as 120 t/ha (ideal conditions). Hence, this yield was considered for the cultivations stage.

<sup>a</sup>Calculation based on values from.<sup>80</sup>

<sup>b</sup>Calculation based on values from.<sup>69</sup>

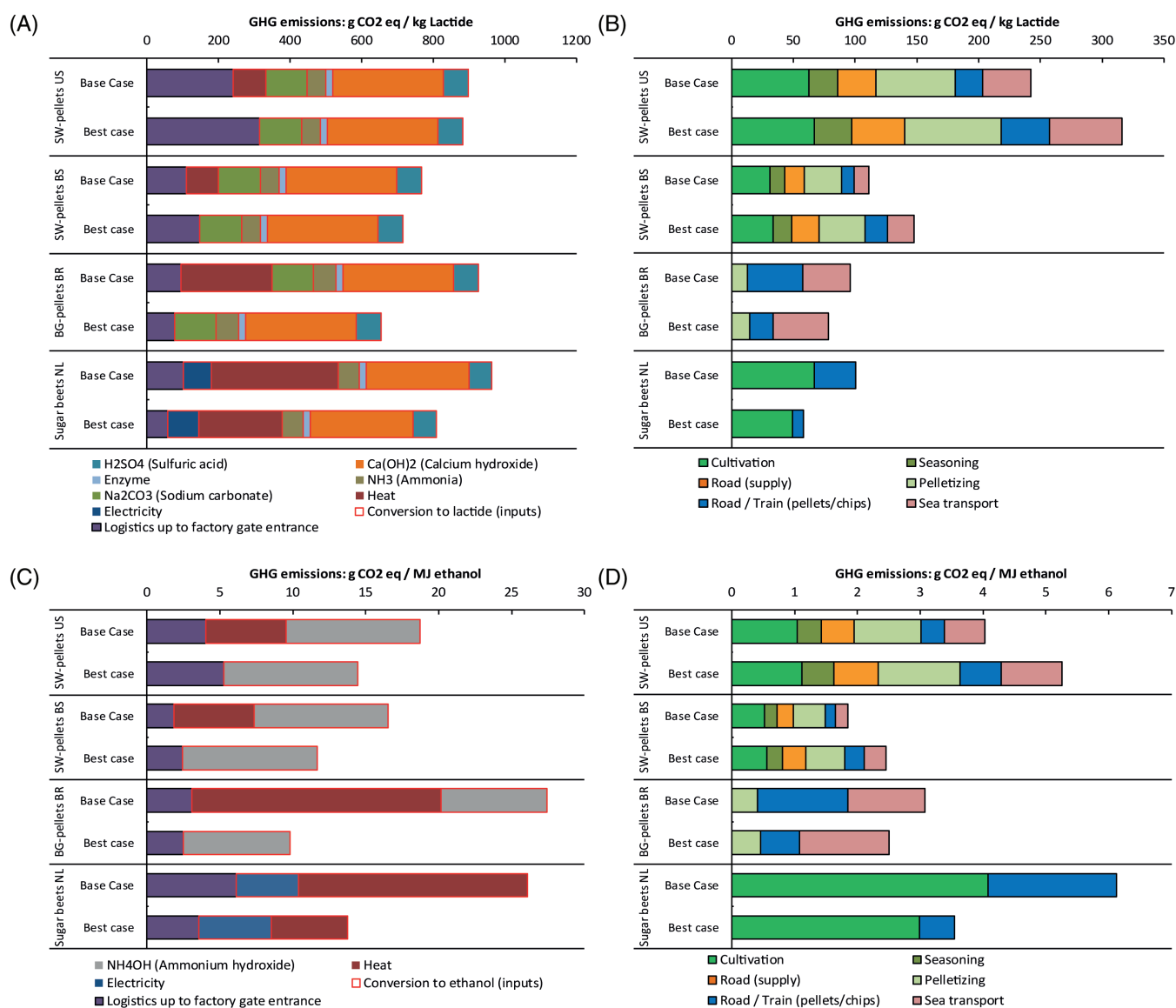


Figure 10. Sensitivity analysis results for lactide with (A) and without conversion stage (B), and for ethanol with (C) and without (D) conversion stage. Best case assumptions include higher feedstock yields, more efficient transport chains and replacement of natural gas with biomass.

importance for the structured transport of pellets from inner São Paulo state to the port of Santos. This requires important infrastructure developments. For the best SB from the Netherlands scenario, higher SB yields and shorter supply distance results in a reduction of 42 g CO<sub>2</sub>eq/kg<sub>lactide</sub> in logistics up to the factory-gate entrance. To obtain a higher yield, besides adequate climate and soil conditions, an increase in agricultural inputs can be expected. An increase in such inputs could result in higher GHG emissions from SB production. The heat supply from a biogas plant can only cover to an extent the heat demand from the conversion to the lactide stage. However, the heat supply from the biogas

plant reduces by 122 g CO<sub>2</sub>eq/kg<sub>lactide</sub> the impact at this stage. The majority of GHG savings from the best case scenarios are related to the use of additional biomass for heat supply at the conversion stage with the highest improvement on the BG pellet from the Brazil conversion route.

For ethanol (Fig. 10(C)), the GHG emissions from the best case scenarios are reduced between 6 g CO<sub>2</sub>eq/MJ<sub>ethanol</sub> (SW pellets from the USA) and 21 g CO<sub>2</sub>eq/MJ<sub>ethanol</sub> (BG pellets from Brazil). As with the lactide results, the replacement of natural gas with biomass to generate process heat induces the largest influence for GHG emissions reduction for the best case scenarios. It is worth noting that the worst performing

base-case scenario (BG pellet from Brazil) becomes the best performing conversion route in terms of GHG emissions. Delivering heat with additional biomass for the ethanol conversion process for the Brazil supply chains results in a reduction of  $17 \text{ g CO}_{2\text{eq}}/\text{MJ}_{\text{ethanol}}$ . Likewise, the heat supply from a biogas plant reduces by  $10 \text{ g CO}_{2\text{eq}}/\text{MJ}_{\text{ethanol}}$  the impact generated at the conversion to ethanol process from the SB supply chain. Overall, several of the considered improvements need to be assessed more extensively due to the possible cascade effects that they could induce. For instance, an increase of SB cultivation in the vicinity of the biorefinery can induce the displacements of other land-use types.

### Impact of the choice of biorefinery location

The total GHG emissions ‘from cradle to factory gate’ can differ between conversion routes depending on the biorefinery location. To understand the impacts of the

biorefinery location choice, alternative scenarios for the location of the biorefinery in the country of biomass supply were assessed with the base-case scenarios displayed in Fig. 2 (only results from lactide are displayed for illustrative purposes). The biorefinery is considered to be located at the terminal from which biomass is exported in other supply chains. Lactide is transported by containers ships to Rotterdam and ethanol stays in biomass supply country. Figure 11 displays the comparison between the base-case conversion routes and routes when the biorefinery is located in the country of biomass supply. In comparison with lactide conversion in the Netherlands, the ‘cradle-to-factory-gate’ emissions for 1 kg of lactide production in the USA are  $75 \text{ g CO}_{2\text{eq}}$  less. Greenhouse emissions from chemicals supply chains are relative similar with the exception of sulfuric acid and sodium carbonate. Mining of sulfur and production of sulfuric acid and sodium carbonate in the USA are less energy intensive than in Europe;<sup>102</sup> this results in lower GHG emissions from the conversion to lactide step in the

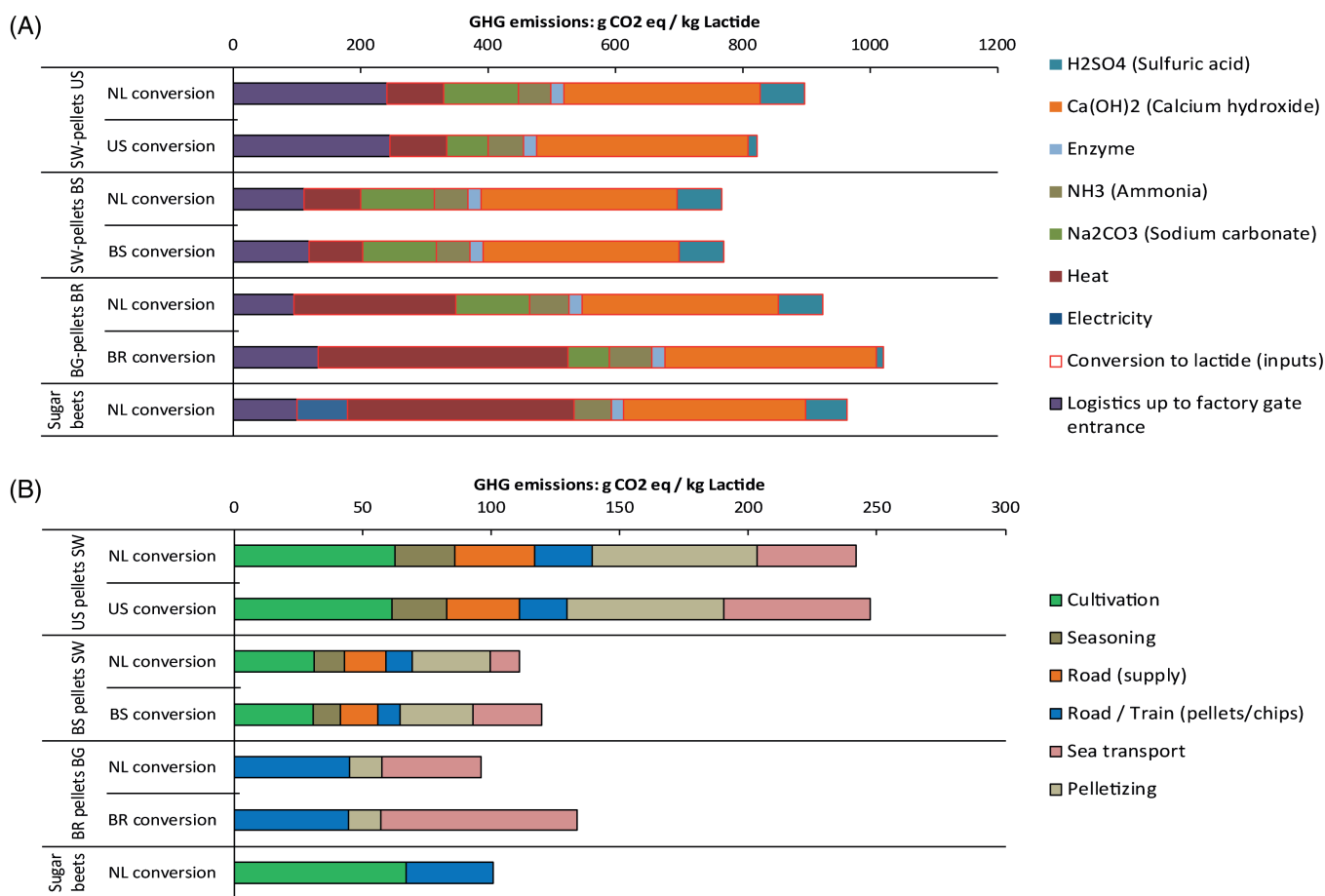


Figure 11. Comparison between GHG emissions when locating the biorefinery and producing lactide at the country of biomass source to the Netherlands with (A) and without (B) conversion stage.

USA. There are small variations in GHG emissions from the use of natural gas to provide heat demand and upstream emissions from other used chemicals. When the biorefinery is located in the Baltic states (Latvia), there is almost no difference between GHG performance. When the biorefinery is located in Brazil and BG is used as feedstock, higher GHG emissions are generated from the use of natural gas for heat supply (the extraction of natural gas in Brazil is more energy intensive). Overall, the 'cradle-to-factory-gate' emissions for 1 kg of lactide increase by 95 g CO<sub>2eq</sub> between locating the biorefinery in Brazil or the Netherlands.

For upstream emissions (Fig. 11(B)), the main difference between locating the biorefinery in the country of biomass source or in the Netherlands is the mode of sea transport. Sea transport of pellets has a better GHG performance as a result of bulk transportation mode than lactide transportation in containers. Transporting lactide from biomass supply countries results in an increase of 18 g CO<sub>2eq</sub>/kg<sub>lactide</sub> for the US conversion route, 15 g CO<sub>2eq</sub>/kg<sub>lactide</sub> for the Baltic states conversion route and 38 g CO<sub>2eq</sub>/kg<sub>lactide</sub> for the Brazil conversion route. However, on average, differences in sea

transportation mode are small and have little impact when compared to the conversion step. Overall, it is concluded that the location of the biorefinery in either the country of biomass source or the Netherlands has little impact on overall GHG emissions performance when considering the total 'cradle-to-factory-gate' GHG emissions from the conversion routes.

## System comparison

The differences in scope, system design, and functional unit limit the scope for comparison with other multi-output biorefinery studies such as Mandegari *et al.*<sup>103</sup> and Farzad *et al.*<sup>104</sup> Nevertheless, the GHG footprint of the individual outputs of the biorefinery to lactide and ethanol are compared with single-output processes as shown in Table 10. For comparison purposes, the system boundaries were extended with the conversion of lactide to PLA by adding the emissions from the ring opening polymerization of lactide for (PLA) production as reported in Vink and Davies.<sup>87</sup> In the literature, different results are generally reported for lactide and similar ones for ethanol (see supporting information S3 in File S1).

**Table 10. Literature comparison.**

Feedstock	Supply country	End product	GHG performance	Unit	Source
Wood biomass	US	Lactide	−1096 and −828 (+200) <sup>a</sup>	g CO <sub>2eq</sub> /kg <sub>lactide</sub>	This study
Wood biomass	Baltic states	Lactide	−1138 and −1063 (+200) <sup>a</sup>	g CO <sub>2eq</sub> /kg <sub>lactide</sub>	This study
Sugarcane bagasse	Brazil	Lactide	−904 (+200) <sup>a</sup>	g CO <sub>2eq</sub> /kg <sub>lactide</sub>	This study
Sugar beets	Netherlands	Lactide	−866 (+200) <sup>a</sup>	g CO <sub>2eq</sub> /kg <sub>lactide</sub>	This study
Corn	USA	PLA	2023	g CO <sub>2eq</sub> /kg <sub>PLA</sub>	100
Corn	USA	PLA	620	g CO <sub>2eq</sub> /kg <sub>PLA</sub>	87
Corn	USA	PLA	1303	g CO <sub>2eq</sub> /kg <sub>PLA</sub>	119
Sugarcane	Thailand	PLA	500–800	g CO <sub>2eq</sub> /kg <sub>PLA</sub>	120
Sugarcane/corn	Thailand/USA	PLA	590 <sup>b</sup>	g CO <sub>2eq</sub> /kg <sub>PLA</sub>	98
Wood biomass	USA	Ethanol	16–20.4	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	This study
Wood biomass	Baltic states	Ethanol	15.3–16.5	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	This study
Sugarcane bagasse	Brazil	Ethanol	27.4	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	This study
Sugar beets	Netherlands	Ethanol	26.1	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	This study
Eucalyptus	Spain	Ethanol	208.9 <sup>c</sup>	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	105
Black locust	Spain	Ethanol	155.51 <sup>c</sup>	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	105
Poplar	Italy	Ethanol	182.76 <sup>c</sup>	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	105
Sugar beet	-	Ethanol	30.8	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	69
Sugarcane	-	Ethanol	28.1	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	69
Eucalyptus	-	Ethanol	37.31	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	69
Forest residue	-	Ethanol	26.62	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	69
Straw	-	Ethanol	13.7	g CO <sub>2eq</sub> /MJ <sub>ethanol</sub>	69

<sup>a</sup>200g CO<sub>2eq</sub>/kg<sub>PLA</sub> for ring opening polymerization.

<sup>b</sup>Average from corn and sugarcane.

<sup>c</sup>'Cradle to gate' values. Converted from kg CO<sub>2eq</sub>/kg<sub>ethanol</sub>. These values omit for comparison purposes the CO<sub>2</sub> binding in the dry matter while feedstock is grown which is deducted.

The multi-output / parallel production characteristics of biorefineries result in considerably lower GHG emissions than stand-alone conversion systems of biomass to lactide (or similar) and stand-alone conversion systems to ethanol. Biomass utilization is optimized for the production of different marketable products with a lower carbon footprint for each product in comparison with values reported in literature. The multi-output design determines that the burden of emissions is divided according to feedstock biochemical composition (mainly between lactide and ethanol); however, for biorefineries with one main product output the whole burden of GHG emissions is assigned to one product, as in ethanol production from wood biomass as reported in González-García *et al.*<sup>105</sup> or PLA production reported in Vink and Davies.<sup>87</sup> Nevertheless, it is suggested that to obtain such advantages from multi-output biorefineries, the use of lignocellulosic biomass feedstocks with high lignin content is crucial as this is typically used to cover internal heat demand.

## Conclusion

This study compared the GHG footprint of different supply chains design for a biorefinery that uses either imported lignocellulosic biomass or SB from the domestic supply for the production of bio-based chemicals (lactide) and biofuels (ethanol) in the Netherlands. The highest performance of the pathways considered corresponds with SR pellets from the Baltic states with 692 g CO<sub>2eq</sub>/kg<sub>lactide</sub> when not considering the embedded carbon up to the factory gate and net GHG emissions of −1138 g CO<sub>2eq</sub>/kg<sub>lactide</sub> and 15 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub>. In terms of feedstock GHG emissions, SR and sugarcane BG outperform other feedstock types. Other feedstock types are characterized by higher GHG emissions from agricultural inputs and fuel-use for agricultural machinery. Pelletization is preferable over long-distance overseas transport of wood chips due to the increased bulk density and improved handling characteristics further downstream in the supply chain. In terms of supply regions, it has been shown that in the Baltic states (Latvia), adequate biomass supply, the relatively short transport distance between biomass sources and pelletization facilities, and the shorter overseas transport distance between the port of Riga and Rotterdam result in a lower GHG footprint compared with the intercontinental supply chains (Brazil, US southeast) and the national (Netherlands) supply chain. This output challenges the general assumption that local-sourced biomass attains higher GHG emissions by default; it has been shown that the explicit GHG performance depends largely on specific feedstock characteristics and supply-chain configurations. For the conversion stage to the lactide and ethanol stage, process heat requirements and upstream emissions from the production of chemicals used in the

conversion process contribute substantially to the overall GHG footprint. Lignin is used to generate process heat that is otherwise generated from natural gas. The implication of this supply chain design assumption is that wood supply chains, with a higher lignin content compared to bagasse, have a better GHG emissions performance at the conversion stage; this is valid for the analyzed supply chains designs. Processes from the upstream conversion stage have been demonstrated to have a relatively small impact share on the total GHG emissions conversion routes.

All conversion routes, with the exception of the SB Netherlands, are unable to comply with the strict GHG emissions criteria for biofuels in RED II. However, the GHG emissions calculations in line with RED II do not consider the challenges of burden allocation in multi-output systems. This shortcoming largely affects ethanol GHG emissions outcomes when calculated with RED II criteria. Overlooking the challenges of burden allocation can discourage the production of renewable forms of energy in multi-output systems. In addition, it can constrain the incorporation of highly efficient systems and conversion routes that can support to maximize energy security and reduce GHG emissions. For lactide, all conversion routes report high savings when compared with relevant fossil-based counterparts. The high savings are mainly attributed to negative emissions from the imbedded carbon in the materials. The end-of-life phase from bio-based and fossil-based materials or the embedded carbon in bio-based materials needs to be accounted for an adequate comparison. The end-of-life phase from the products subjected to comparison between bio-based and fossil-based can have major repercussions for the total GHG emissions performance and possible policy outcomes. Therefore future research should focus on exploring the complexity from this stage related to plausible end-of-life scenarios.

The results have been tested against alternative supply-chain configurations with the biorefinery located in the country of biomass supply, improvements along the supply chains' GHG hotspots, and different allocation choices. Locating the biorefinery in the USA results in a better GHG performance for the conversion stage than in Brazil or the Baltic States due to the upstream emissions from chemicals supply. However, the net advantage over the total life cycle ('cradle to factory gate') is small. It can therefore be concluded that the development of an integrated biorefinery in the Netherlands, instead of importing final commodities such as ethanol or chemicals, is a viable strategy. The GHG emissions performance from the supply chains can be further improved mainly by utilizing biomass as fuel to supply heat demand from conversion to the lactide / ethanol process instead of natural gas.

Multifunctional systems are highly sensitive to allocation procedures. Next to variations from actual supply chain

design variations, the chosen method to allocate GHG emissions over the multiple outputs of the biorefinery results in large variations in the total GHG footprint. For multi-output systems we recommend using a multi-level allocation method as a base and apply different allocation methods to explicate the impact of methodological choices on the results.

Future research should focus on technologies / processes that allow for a reduction in the consumption of process chemicals in the production of lactide. Given the level of maturity, biorefinery systems and bio-based chemicals still have large development potential in terms of environmental performance by technological development. Allocation guidelines that consider the challenges in allocating the burden of impacts for multifunctional systems should be developed.

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