



## Life cycle impact assessment of bio-based plastics from sugarcane ethanol



I. Tsiropoulos <sup>a,\*</sup>, A.P.C. Faaij <sup>b</sup>, L. Lundquist <sup>c,1</sup>, U. Schenker <sup>c,2</sup>, J.F. Briois <sup>d,3</sup>, M.K. Patel <sup>e,4</sup>

<sup>a</sup> Copernicus Institute of Sustainable Development, Section Energy and Resources, Utrecht University, Heidelberglaan 2, 3584 CS Utrecht, The Netherlands

<sup>b</sup> Energy and Sustainability Research Institute, University of Groningen, Blauwborgje 6, 9700 AE Groningen, The Netherlands

<sup>c</sup> Nestlé Research Center, Nestlé LTC, Vers-chez-les-Blanc, 1000 Lausanne 26, Switzerland

<sup>d</sup> Nestlé Water Management and Technology, PTC Water, B.P. 101, 88804 Vittel Cedex, France

<sup>e</sup> Energy Group, Institute for Environmental Sciences and Forel Institute, University of Geneva, 1227 Carouge, Geneva, Switzerland

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### ABSTRACT

The increasing production of bio-based plastics calls for thorough environmental assessments. Using life cycle assessment, this study compares European supply of fully bio-based high-density polyethylene and partially bio-based polyethylene terephthalate from Brazilian and Indian sugarcane ethanol with production of their petrochemical counterparts in Europe. Bio-based polyethylene results in greenhouse gas emissions of around  $-0.75 \text{ kg CO}_2\text{eq/kg}_{\text{polyethylene}}$ , i.e. 140% lower than petrochemical polyethylene; savings on non-renewable energy use are approximately 65%. Greenhouse gas emissions of partially bio-based polyethylene terephthalate are similar to petrochemical production ( $\pm 10\%$ ) and non-renewable energy use is lower by up to 10%, partly due to the low bio-based content of the polymer. Assuming that process energy is provided by combined heat and power reduces the greenhouse gas emissions of partially bio-based polyethylene terephthalate production to a range from  $-4\%$  (higher) to  $15\%$  (lower) compared to petrochemical polyethylene terephthalate depending on the methodological choices made. Production from Brazilian ethanol leads to slightly higher impacts than production from Indian ethanol due to dampening effects of allocation on Indian ethanol produced from sugarcane molasses, different sugarcane pre-harvesting practices and inter-continental transport of Brazilian ethanol to India. Internal technical improvements such as fuel switch, new plants and best available technology offer savings up to 30% in greenhouse gas emissions compared to current production of petrochemical polyethylene terephthalate. The combination of some of these measures and the use of biomass for the supply of process steam can reduce the greenhouse gas emissions even further. In human health and ecosystem quality, the impact of the bio-based polymers is up to 2 orders of magnitude higher, primarily due to pesticide use, pre-harvesting burning practices in Brazil and land occupation. When improvements are assumed across the supply chain, such as pesticide control and elimination of burning practices, the impact of the bio-based polymers can be significantly reduced. Realising such improvements will minimise the greenhouse gas and other emissions and resource use associated with bio-based polyethylene terephthalate and will allow to alleviate further pressure on fragile ecosystems.

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**Abbreviations:** BAT, best available technology; CHP, combined heat and power; EQ, ecosystem quality; FIT, feed in tariff; GHG, greenhouse gas; HDPE, high-density polyethylene; HH, human health; LCA, life cycle assessment; LDPE, low-density polyethylene; LLDPE, linear low-density polyethylene; MEG, monoethylene glycol; NREU, non-renewable energy use; Pchem, petrochemical; PE, polyethylene; PET, polyethylene terephthalate; PTA, purified terephthalic acid.

\* Corresponding author. Tel.: +31 30 253 5535.

**E-mail addresses:** [i.tsiropoulos@uu.nl](mailto:i.tsiropoulos@uu.nl) (I. Tsiropoulos), [a.p.c.faaij@rug.nl](mailto:a.p.c.faaij@rug.nl) (A.P.C. Faaij), [lars.lundquist@rdls.nestle.com](mailto:lars.lundquist@rdls.nestle.com) (L. Lundquist), [urswalter.schenker@rdls.nestle.com](mailto:urswalter.schenker@rdls.nestle.com) (U. Schenker), [jean-francois.briois@waters.nestle.com](mailto:jean-francois.briois@waters.nestle.com) (J.F. Briois), [martin.patel@unige.ch](mailto:martin.patel@unige.ch) (M.K. Patel).

<sup>1</sup> Tel.: +41 21 785 8071.

<sup>2</sup> Tel.: +41 21 785 9512.

<sup>3</sup> Tel.: +33 3 29 08 70 77.

<sup>4</sup> Tel.: +41 22 379 0658.

## 1. Introduction

Since 1980, petrochemical plastics production increased by an average compound annual growth rate of about 5%, resulting in a global production volume of 288 million tonnes in 2012 (PlasticsEurope, 2013). This production accounts for 5% of the global total primary energy supply (BP, 2013; PlasticsEurope, 2013).<sup>5</sup> In Europe, low-density, linear low-density and high-density polyethylene (LDPE, LLDPE and HDPE, respectively) and polyethylene terephthalate (PET) together represent 36% of plastics demand (PlasticsEurope, 2013).

It is known that the use of renewable resources for applications other than fuels, such as chemicals, oleochemicals, paper and textiles, generally offers higher value added (Nova Institut, 2010). Recently, the use of bio-based plastics for packaging has received a lot of attention due to emerging technological options (Shen et al., 2010). Polylactic acid, bio-based polyethylene (bio-PE), and partially bio-based PET (bio-PET) are notable examples. In 2011, bio-PE and bio-PET represented 56% of the global bioplastics' production capacity reaching 650 ktonnes (European Bioplastics, 2012). The capacity is expected to further increase since several producers have commissioned new production plants (JBF, 2012; TTS, 2011). Daioglou et al. (2014) estimate that the global feedstock energy demand for chemicals and refinery products is expected to increase from 30 EJ today to over 100 EJ by 2100. Biomass can supply over 40% of the total primary energy required for non-energy purposes and thus reduce greenhouse gas emissions by 20% in 2100 (Daioglou et al., 2014). Bio-based products and plastics could hence become an important strategy in the transition process towards sustainable bio-based economies (EC, 2009, 2011; EU, 2011). To ensure that adequate decisions are made, it is essential to assess the potential environmental impacts of the entire process chain taking into consideration local production practices and boundary conditions.

The purpose of this study is to assess the environmental impacts of bio-PE and bio-PET from sugarcane ethanol. The selected products represent a large share of current bio-based plastics production capacity and will continue to do so in the short and medium term (Shen et al., 2010). While numerous studies have been published on biofuel production from various feedstocks (e.g. Börjesson and Tufvesson, 2011; von Blottnitz and Curran, 2007), to our knowledge, there is only one peer-reviewed article that assesses the environmental impacts of bio-LDPE (Liptow and Tillman, 2012). However, Brazilian sugarcane ethanol data and data on ethanol conversion to bio-ethylene need to be updated. Polymer producers also publish environmental profiles of their bio-based products without, however, disclosing detailed background information (Hunter et al., 2008). Other studies, in which ethylene is a precursor, do not report environmental impacts of bio-ethylene, but aggregated results for the final polymer (bio-PVC; Alvarenga et al., 2013). Chen and Patel (2012) used literature data to prepare a rough estimate of non-renewable energy use and greenhouse gas emissions for bio-PET from sugarcane and maize. However, process data on ethanol dehydration need to be revisited and, for a comprehensive analysis, it is important to assess additional environmental impacts on ecosystem quality, human health, water-use and land use.

In the following, we describe the production of bio-PE and bio-PET from sugarcane ethanol. We then present the methodology used to assess their environmental performance, and compare the

results with the production of their petrochemical counterparts in Europe.

## 2. Process description

Both bio-PE and bio-PET are currently produced from first generation ethanol, i.e. ethanol derived from food crops such as sugarcane. Ethanol is subsequently catalytically dehydrated to ethylene and a) is polymerised to polyethylene or b) is oxidised to ethylene oxide and then hydrolysed to bio-based mono-ethylene glycol (bio-MEG), the bio-based component of bio-PET. Regardless whether the feedstock is bio-based or petrochemical, further conversion of ethylene to these polymers remains the same. The comparability of bio-PE and bio-PET with their petrochemical counterparts is ensured since they are identical polymers. Although ethanol is produced from various food crops such as sugarcane, maize and wheat, we concentrate on production from sugarcane since it is currently the only feedstock used to produce bio-PE and bio-PET. Also, we focus on Brazilian and Indian production because they are the world's largest sugarcane and sugarcane ethanol producers and today's production of bio-PE and bio-PET is established in Brazil and India, respectively (de Jong et al., 2012).

### 2.1. Sugarcane ethanol production in Brazil and India

The production chain of ethanol in Brazil and India is described in detail in Tsiropoulos et al. (2014). This section focuses on main differences between ethanol production in south-central Brazil and Uttar Pradesh, India. Brazilian sugarcane cultivation offers high yields (around 85 t<sub>cane</sub>/ha) and is highly mechanised; pre-harvesting burning practices are partly applied but they are gradually being phased out. In India, agricultural practices rely mainly on human and animal labour, yields are significantly lower (around 55 t<sub>cane</sub>/ha) and irrigation is required. In Brazil, fresh sugarcane juice is directly fermented and distilled to ethanol whereas in India only sugarcane molasses are used.

In both countries, ethanol production yields co-products, which are used internally and reduce inputs (e.g. fertilisers), make the process less dependent on external energy sources and provide surplus electricity and biomass. During sugarcane juice extraction, juice is separated from the fibrous stalks and the obtained shredded bagasse is used in co-generation facilities to produce steam and electricity to meet process energy requirements. An increasing number of mills both in Brazil and in India generate surplus electricity, which they sell to the national grid. The remaining bagasse is typically sold as a solid biofuel or as feedstock for the paper industry (ISMA, 2011a,b; Seabra et al., 2011). Residues of juice filtration, typically referred to as filtercake or mud, are mixed with ashes from boilers and are returned to sugarcane fields as fertilisers. The distillation generates a significant amount of wastewater (stillage). In Brazil, after cooling in open ponds, stillage is distributed onto the fields and valuable nutrients are recycled (Lisboa et al., 2011). In India, stillage is typically treated in anaerobic digesters to generate biogas; the biogas is used in co-generation facilities and contributes to on-site energy supply (Tewari et al., 2007). Depending on filtercake availability a number of distilleries use part of the stillage to produce bio-compost, which is either sold or offered to farmers for free (ISMA, 2012).

### 2.2. Bio-PE production

Historically, bio-based ethylene was derived from ethanol dehydration. However, after the mid-1940s, with the rise of the petrochemical industry, steam cracking of petroleum liquids and heavier fractions of natural gas became the dominant processes for

<sup>5</sup> Based on total global primary energy supply of 522 EJ (87% is fossil-based; BP, 2013). The contribution of petrochemical plastics (288 Mtonnes, 2012) is calculated based on the weighted average specific energy consumption of plastics (76.7 GJ/tonne), of which approximately 46% is process energy requirements.

ethylene production (Kochar et al., 1981). As the industry has renewed its attention to produce ethylene from ethanol, the process has been further optimised and new, improved catalysts have been developed (Chematur, 2011).

Ethanol is catalytically dehydrated in a vapour phase reaction to remove one water molecule per molecule of ethanol, thereby yielding ethylene. The process is endothermic and based on the theoretical reaction enthalpy it requires 1.63 MJ/kg<sub>ethylene</sub>. Diethyl ether is formed as intermediate product at temperatures between 150 °C and 300 °C, while ethylene formation is predominant between 300 °C and 500 °C (Morschbacker, 2009). Due to their high selectivity, productivity and resistance to deactivation, alumina or silica-alumina catalysts are used in fixed-bed or fluidised-bed reactors. Minor quantities of ethane, propylene, butylenes, acetaldehyde and negligible amounts of methane, carbon monoxide and dioxide, ethyl ethers and hydrogen are formed from side-reactions. The effluent stream consists mainly of water containing acetaldehyde, diethyl ether and non-reacted ethanol. It is treated by stripping, reaching a chemical oxygen demand level lower than 100 ppm. Commonly, light organic by-products are flared and heavy organics are collected for fuel use, which reduces the net energy requirement of the process (Chematur, 2011).

Depending on the desired application ethylene is converted to LDPE in high-pressure tubular or autoclave reactors (PlasticsEurope, 2008a). HDPE is mainly produced in low-pressure reactors via suspension or gas phase polymerisation (PlasticsEurope, 2008b). LLDPE is produced at relatively low pressures and temperatures by solution or gas phase polymerisation (PlasticsEurope, 2008c). Among these polymers, LDPE has the highest primary energy demand due to high electricity requirement. The primary energy demand of HDPE and LLDPE is lower despite high steam demand, because steam is typically produced with higher conversion efficiencies than electricity (EC, 2007).<sup>6</sup>

### 2.3. Bio-PET production

Ethylene is mixed with oxygen, CO<sub>2</sub>, argon and methane or nitrogen and the dilute gas mixture is fed to a tubular catalytic reactor. The reaction to ethylene oxide (EO) is highly exothermic; the reaction temperature is controlled by producing steam and by controlling the pressure in the steam drum. The EO is scrubbed with water. By-product CO<sub>2</sub> is removed and returned to the reactor loop, and the EO is steam-stripped and recovered as concentrated aqueous solution. The EO stream is sent to the glycol reactor in which ethylene glycols are produced by reaction with water. After the reactor a multi-effect evaporator system is used for water removal. Glycols are dried, cooled and sent to a distillation train for separation and purification, where MEG is separated from the heavier diethylene glycol (DEG) and triethylene glycol (TEG) (HCP, 2010).

Bio-MEG represents 27.7% of the inputs in mass terms required for bio-PET production.<sup>7</sup> The other monomer is purified terephthalic acid (PTA), produced from paraxylene. So far no commercial bio-based route to paraxylene exists. Xylenes are produced mainly by solvent extraction and fractionated distillation of aromatic rich streams in refineries. Paraxylene and acetic acid are used to produce PTA. The monomer is polymerised with MEG to amorphous

PET in a direct esterification or melt polymerisation process. For bottle applications a second polymerisation in solid state is required (PlasticsEurope, 2011).

### 3. Methodology

To evaluate the environmental impacts of products and services, life cycle assessment (LCA) is used (ISO, 2006a,b) which is widely applied in environmental assessments of bio-based materials (Uihlein et al., 2008; Weiss et al., 2012; Álvarez-Chávez et al., 2012), and can also be used for comparative assertions between products that deliver equivalent functions (ISO, 2006a).

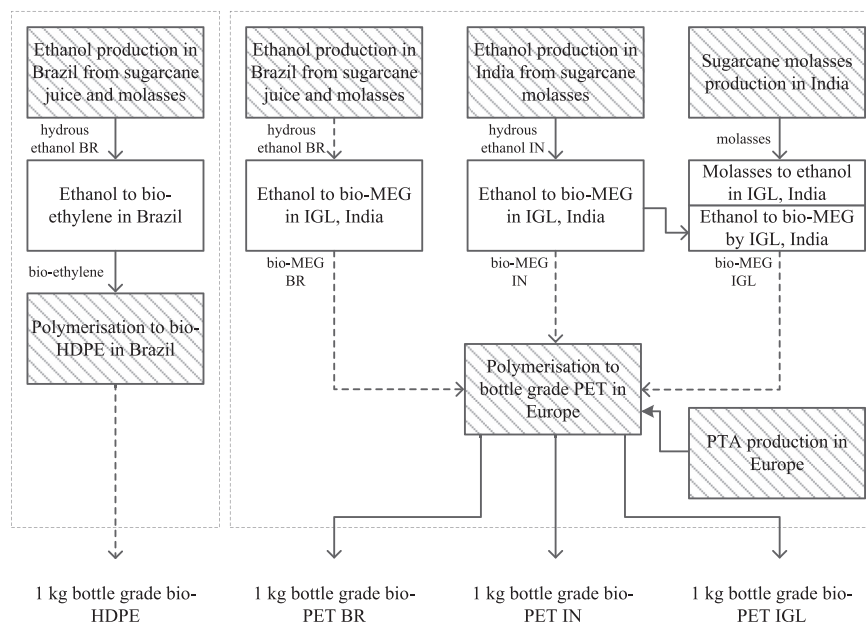
#### 3.1. System boundaries, functional units and data

The systems are assessed from cradle to gate. Both Brazilian and Indian sugarcane ethanol are used for bio-PET, while only Brazilian ethanol is used for bio-HDPE. For bio-HDPE we consider bio-ethylene production and polymerisation to bio-HDPE in Brazil, and transport to Europe. For bio-PET we consider bio-MEG production in India. We analyse three cases, where bio-MEG is produced at India Glycols (IGL) from: a) Indian sugarcane ethanol production in Uttar Pradesh (bio-MEG<sub>IN</sub>), b) ethanol of the distillery attached to the MEG facility in India Glycols (bio-MEG<sub>IGL</sub>), c) Brazilian sugarcane ethanol (bio-MEG<sub>BR</sub>). These routes currently supply bio-MEG to the market. We include transport of bio-MEG to Europe, PTA production and polymerisation to bio-PET in Europe. Final transport is included because we compare with European production of fossil-based HDPE and PET. The functional units are 1 kg of bio-based HDPE and 1 kg partially bio-based PET, produced in three product-systems: bio-PET<sub>IN</sub>, bio-PET<sub>IGL</sub>, and bio-PET<sub>BR</sub> (Fig. 1).

For production of sugarcane ethanol we use cradle-to-gate results from our previous study, which focused on the comparison of Brazilian and Indian ethanol (Tsiropoulos et al., 2014). For conversion of ethanol to bio-ethylene in Brazil, data from technology licensors are used. We assume that process electricity is sourced from the national grid or produced by natural gas depending on the allocation approach. For steam production we assume the use of natural gas and oil, on a 76:24 primary energy ratio, based on Ecoinvent v2.2. Other heat requirements are supplemented by natural gas. Biomass is also suitable for industrial heat production, however, this option is not assessed in this study (Saygin et al., submitted for publication). For ethylene polymerisation to bio-HDPE we estimate gate-to-gate impact assessment results, by deducting the cradle-to-gate data for the monomer from cradle-to-gate data for the polymer (Ecoinvent v2.2 data; Hirschier, 2007). These data are representative for European production sites; however, due to lack of more accurate information, we assume that they are representative also for Brazil. For bio-MEG production in India, we use data from the producer. We also present results based on data provided by technology licensors (Section 4). Similar to bio-HDPE, process electricity is sourced from the national grid or from the local grid depending on the allocation. Steam is produced from coal (bio-PET<sub>BR</sub>, bio-PET<sub>IN</sub>) or from coal and biogas (bio-PET<sub>IGL</sub>). Emissions for biogas combustion are based on Stucki et al. (2011). Other heat requirements are supplemented by natural gas. We also present results assuming that heat and electricity is supplied by an on-site coal-based combined heat and power (CHP) plant (Section 4). For PTA production and polymerisation, we use cradle-to-gate results calculated from the latest environmental profiles published by the European Plastics association (Mersiowsky, 2011; PlasticsEurope, 2011) and Ecoinvent v2.2 (Supporting information). We adapt the electricity mix of electricity-intensive processes, namely oxygen, nitrogen and sodium hydroxide

<sup>6</sup> Primary energy requirement is 6.4, 8.2, 4.8 MJ/kg HDPE, LDPE and LLDPE, respectively. Calculated from steam demand with 90% efficiency (Saygin et al., 2011) and from electricity demand with 42% efficiency (HCP, 2010).

<sup>7</sup> The share of ethylene glycol based on the carbon content of the reactants (ethylene glycol and terephthalic acid) is 20%. The share of the molecular weight of MEG relative to the repeating unit of PET is 31%.



**Fig. 1.** Main process steps in bio-HDPE (left) and bio-PET (right) production for further use in Europe. Data for processes marked with highlighted boxes are based on literature sources, databases or industry averages. Data for processes marked with clear boxes are based on primary producers or technology licensors. Dashed arrows indicate international transport.

production, to the regional electricity mix. In addition, for oxygen production we adapt process electricity requirements based on information provided by the bio-MEG producer. As background database for secondary inputs and processes we use Ecoinvent v2.2 (Ecoinvent, 2010). Table 1 gives an overview of the data used in this study. The impact assessment results we used for the process steps ethylene polymerisation to bio-HDPE, PTA production and polymerisation to bio-PET are presented in the Supporting information.

We compare the results with cradle-to-gate environmental profiles of petrochemical PET (pchem-PET) and HDPE (pchem-HDPE) production in Europe. Ethylene polymerisation to HDPE, PTA production and polymerisation to bottle-grade PET are the same for the bio-based and the petrochemical route.<sup>8</sup> For computation of impact assessment results we use the software Simapro v 7.3 (Pré Consultants, 2011).

### 3.2. Multifunctionality

For multifunctional processes a suitable method must be applied to assign the environmental interventions to the multiple outputs. Based on the International Organization for Standardization, one should first subdivide the multifunctional system into distinct sub-processes and associate process-related impacts with individual products or co-products. This is not always possible due to non-separable processes or data availability. The second approach, termed system expansion, enlarges the system boundaries to include the additional functions of the co-products.<sup>9</sup> The third and fourth options allocate environmental interventions

between co-products based on physical or other relationships (e.g. economic value), respectively (ISO, 2006b).

In this study there are several process steps which lead to multiple outputs. The sugarcane ethanol product-system involves co-production of sugarcane trash, filtercake, surplus electricity and bagasse from sugar mills and distilleries. In the Indian ethanol product-system we are faced with the multifunctionality problem due to co-production of sugar and molasses. To account for these outputs we apply four different allocation approaches, which are explained in Tsiropoulos et al. (2014). An overview is presented in Table 2. Furthermore, in bio-MEG production, DEG, TEG and heavier glycols (HEG) are also produced. These are all valuable products with applications in the automotive and packaging industry (IGL, 2011). We allocate their impacts based on mass.

### 3.3. Impact assessment methodology

We present results for greenhouse gas (GHG) emissions over a 100 year timeframe (IPCC, 2007).<sup>10</sup> GHG emissions are the most commonly used metric to assess the sustainability impacts of products. However, as Laurent et al. (2012) indicate, GHG emissions are correlated with other environmental impacts only when these predominantly originate from fossil fuels. When toxicity to humans or ecosystems and land-use are of concern, then GHG emissions alone are a weak indicator. For bio-based products, the use of agrochemicals during biomass cultivation is expected to contribute to toxicity-related impacts, as opposed to GHG emissions and non-renewable energy use (NREU), for which bio-based products generally show savings when compared to petrochemical products (Weiss et al., 2012). Therefore, we extend the analysis to NREU, land-use, freshwater eutrophication, water-use at the midpoint level and to the endpoints human health (HH) and ecosystem

<sup>8</sup> PlasticsEurope released a new version of impact assessment results for petrochemical ethylene and MEG (PlasticsEurope, 2012). It is expected that results for pchem-HDPE will be updated accordingly. While preparing this study, these were not available. Therefore, the comparison is based on Ecoinvent v2.2. We discuss findings of the latest PlasticsEurope report in Section 4. For pchem-PET we use the latest available data published by PlasticsEurope (Table 1).

<sup>9</sup> Although not explicitly stated in ISO (2006a,b) this approach is typically considered equivalent to subtracting the additional functions of the system, i.e. by deducting credits (Heijungs, 2014). This “credit approach” is applied in this study.

<sup>10</sup> Fossil and biogenic methane characterisation factors adapted to 27.75 kg CO<sub>2eq</sub>/kg CH<sub>4</sub> and 25 kg CO<sub>2eq</sub>/kg CH<sub>4</sub>, respectively (Muñoz et al., 2013).



**Table 1**  
Representativeness, regional and temporal information on data used in this study.

Process step	Region	Period	Representativeness	Source
Sugarcane ethanol	South-central Brazil	2008–2009	Regional	Tsiropoulos et al. (2014)
	Uttar Pradesh, India	2009–2010	Regional	
Ethanol to bio-ethylene	Brazil	2011	Technology licensor <sup>a</sup>	Personal communication
Bio-ethylene to bio-HDPE	Brazil	2006	European industry average	Hischier (2007)
Ethanol to bio-MEG	India	2011	Producer, <sup>a</sup> technology licensor <sup>a</sup>	Personal communication
PTA	Europe	2009/2000 <sup>b</sup>	European industry average	PlasticsEurope (2011)
Polymerisation to bio-PET	Europe	2009/2000 <sup>b</sup>	European industry average	
Transoceanic transport <sup>c</sup>	International	2003	LCA database	Spielmann et al. (2007)
Petrochemical PET	Europe		European industry average	PlasticsEurope, (2011)
Petrochemical HDPE	Europe		European industry average	Hischier (2007)

<sup>a</sup> Conversion of ethanol to bio-ethylene and bio-MEG is based on proprietary data. For this reason it is not possible to provide an extensive life cycle inventory.

<sup>b</sup> For PTA production and for polymerisation we use cradle-to-gate impact assessment results for greenhouse gas emissions and non-renewable energy use calculated from the latest PlasticsEurope report (PlasticsEurope, 2011). These data are representative for 2009. For other impact categories we use data from Ecoinvent v2.2 (Hischier, 2007), which are representative for 2000.

<sup>c</sup> GHG emissions of sea transport are estimated at 8.8 g CO<sub>2eq</sub>/tkm. They are in line with the emissions reported by the European Environment Agency (8 g CO<sub>2</sub>/tkm) for bulk dry sea transport (EEA, 2009).

**Table 2**  
Allocation approaches chosen for multifunctional processes in sugarcane processing in Brazil and India (Tsiropoulos et al., 2014).

Approach <sup>a</sup>	Products	Description	Credits/allocation factors [per kg <sub>ethanol</sub> ]
System expansion, conservative (SE-C)	- Energy outputs: electricity (Brazil, India) - Material outputs: ethanol and bagasse (Brazil), sugar, molasses and bagasse (India)	- Displacement of <i>low</i> CO <sub>2</sub> emission intensity grid power - Economic allocation between material outputs	Brazil: 0.16 kWh (0.22 kg CO <sub>2eq</sub> /kWh) India: 0.5 kWh (0.55 kg CO <sub>2eq</sub> /kWh), sugar 91.5%, molasses 8%, bagasse 0.5%
System expansion, optimistic (SE-O)	- Energy outputs: electricity, heat from bagasse (Brazil, India) - Material outputs: ethanol (Brazil), sugar, molasses and bagasse (India)	- Brazil: Displacement of <i>high</i> CO <sub>2</sub> emission intensity power from natural gas and oil-based heat India: Displacement of <i>high</i> CO <sub>2</sub> emission intensity grid power and coal-based heat - Economic allocation between material outputs	Brazil: 0.16 kWh (0.65 kg CO <sub>2eq</sub> /kWh), 0.9 MJ <sub>heat</sub> (0.09 kg CO <sub>2eq</sub> /MJ <sub>heat</sub> ) India: 0.5 kWh (1.1 kg CO <sub>2eq</sub> /kWh), 0.38 MJ <sub>heat</sub> (0.13 kg CO <sub>2eq</sub> /MJ <sub>heat</sub> ), sugar 92%, molasses 8%
Economic allocation, conservative (EA-C) and optimistic (EA-O)	- Material outputs: ethanol, electricity and bagasse (Brazil). Sugar, molasses, electricity and bagasse (India)	- Economic allocation between material outputs	Brazil: ethanol 97.5%, electricity 2%, bagasse 0.5% India: sugar 85%, molasses 7.5%, bagasse 0.5%

<sup>a</sup> In the system expansion approaches process electricity input in the foreground system and energy intensive material inputs has the same emission intensity with surplus electricity. Tsiropoulos et al. (2014) present results for one economic allocation approach in which process electricity is supplied from the national grid. In this study we extend the analysis by assuming a) process electricity for bio-ethylene from the national grid and for bio-MEG from the regional grid in Uttar Pradesh (EA-C) and b) process electricity for bio-ethylene from natural gas and for bio-MEG from the national Indian grid (EA-O).

quality (EQ). All categories are analysed using Impact 2002+ (Joliet et al., 2003).<sup>11</sup> Water-use is reported at the inventory phase and at the endpoint using factors in Pfister et al. (2009). The impact of infrastructure is excluded from the analysis.

## 4. Results and discussion

### 4.1. Bio-based carbon

We include the bio-based carbon content of the final polymer as carbon storage (negative bar section in Figs. 2 and 3, amounting to 3.2 kg CO<sub>2</sub>/kg<sub>bio-HDPE</sub> and 0.45 kg CO<sub>2</sub>/kg<sub>bio-PET</sub>). It is important to account for the bio-based carbon in cradle-to-gate systems as it may affect the final ranking of alternative options when compared to ranking based on cradle-to-grave assessments (Pawelzik et al., 2013). In this case, net emissions of the bio-based polymers (symbols in Figs. 2 and 3) should be compared with gross emissions of

the petrochemical counterpart (shaded stacked bars, excluding end-of-life). Next to the base case ('Net') the graph shows high and low performance cases for mills and distilleries with high surplus electricity versus no surplus electricity and dependency on the grid based on Tsiropoulos et al. (2014). Figs. 2 and 3 also include results at the end-of-life, assuming incineration without energy recovery. In this case, gross results for the bio-based polymers (without carbon storage) should be compared with the petrochemical polymers at the end-of-life. Apart from process emissions from polymer production (shaded stacked bars in Figs. 2 and 3) also feedstock carbon emissions are accounted for (dotted bars in Figs. 2 and 3). Bio-based carbon emissions are neutral while fossil carbon emissions contribute to global warming. Bio-HDPE is fully bio-based hence there are no fossil end-of-life emissions. In bio-PET emissions from fossil carbon in PTA are included. For petrochemical polymers all carbon content is of fossil origin and is accounted for its contribution to global warming.

#### 4.1.1. Greenhouse gas emissions and non-renewable energy use

GHG emissions are presented for bio-HDPE and bio-PET in Figs. 2 and 3, respectively. Bio-HDPE results are broken down to ethanol production, bio-ethylene production, polymerisation to bio-HDPE and final transport of the polymer from Brazil to Europe. Bio-PET results are broken down to ethanol production, bio-MEG

<sup>11</sup> Including the ozone depleting potential of nitrous oxides (N<sub>2</sub>O), i.e. 0.017 kg CFC-11eq/kg N<sub>2</sub>O (Ravishankara et al., 2009). Aquatic acidification and freshwater eutrophication are associated with ecosystem quality based on 8.82 · 10<sup>-3</sup> PDF m<sup>2</sup> y/kg SO<sub>2eq</sub> and 1.4 PDF m<sup>2</sup> y/kg PO<sub>4</sub><sup>3-</sup>eq, respectively (Humbert et al., 2012).

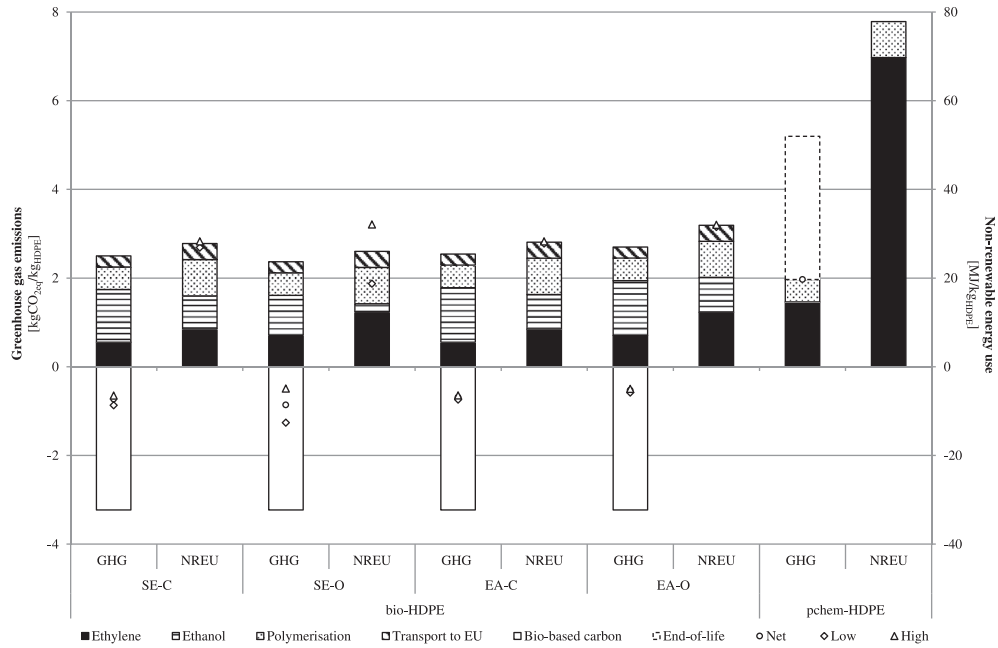


Fig. 2. Greenhouse gas emissions and non-renewable energy use of bio-based HDPE production from Brazilian sugarcane ethanol.

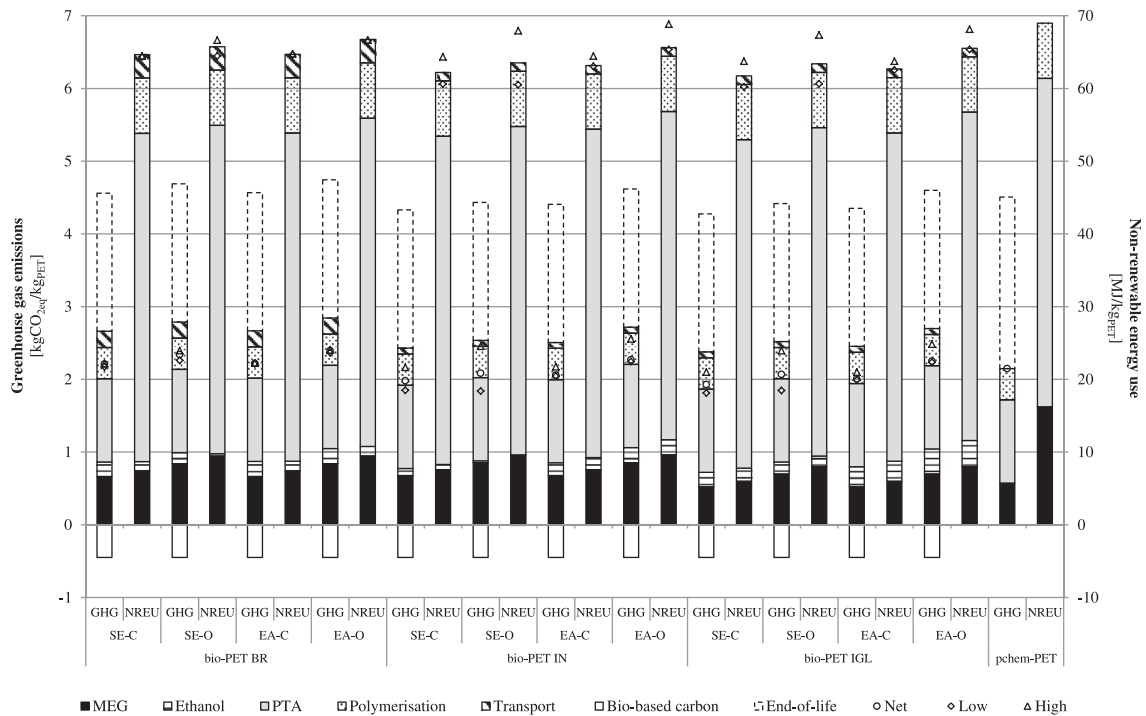


Fig. 3. Greenhouse gas emissions and non-renewable energy use of partially bio-based PET production from Brazilian and Indian sugarcane ethanol.

production, transport of bio-MEG from India to Europe,<sup>12</sup> PTA production and polymerisation to bio-PET in Europe.

Production of bio-HDPE results in average net CO<sub>2</sub> storage in the range of 0.75 kg CO<sub>2</sub>eq/kg<sub>bio-HDPE</sub> (Fig. 2). When compared to pchem-HDPE it leads to approximately 140% savings. The result is

similar across the four allocation approaches (SE-C, SE-O, EA-C, EA-O) but the absolute contribution of each step differs slightly. In SE-O, ethanol production contributes less than in SE-C, EA-C and EA-O, while bio-ethylene production is somewhat higher. In SE-O, ethanol receives large credits for the co-products surplus electricity and surplus bagasse (Table 2), thus the cradle-to-gate impact of ethanol is lowest. However, due to the high CO<sub>2</sub> emission intensity of the electricity in ethanol dehydration the contribution of bio-ethylene step increases when compared to SE-C, EA-C and EA-

<sup>12</sup> Transport results for bio-PET<sub>BR</sub> also include transport of ethanol from Brazil to India.

O. While this tradeoff does not influence the base case it has an impact when applied in the two extreme cases (high, low) under SE-O. The overall net GHG emissions for bio-HDPE range approximately  $\pm 50\%$  in the high and low performance case.

Bio-PET production leads to comparable GHG emissions as pchem-PET, even after deducting the bio-based carbon of bio-MEG (Fig. 3). These cases do not consider improvement potentials by integration of a CHP, process integration and other options which will be discussed in Section 4.1.4. Depending on the source of ethanol (Brazil, India) and the allocation approach (SE-C, SE-O, EA-C, EA-O) the difference to pchem-PET (2.15 kg CO<sub>2eq</sub>/kg<sub>PET</sub>) ranges by approximately  $\pm 10\%$ . Bio-PET<sub>BR</sub> results in slightly higher GHG emissions than bio-PET<sub>IN,IGL</sub>, partly due to the higher GHG emissions of Brazilian ethanol (Tsiropoulos et al., 2014) and partly due to the contribution of transport of Brazilian ethanol to India. PTA production, polymerisation and transport of bio-MEG to Europe account approximately for two-thirds of the GHG emissions. The former two steps are common in the bio-based and petrochemical route. The remainder of the contribution is due to bio-MEG production and originates primarily from process energy requirements, which are met by coal-based steam and grid electricity.

The system expansion approaches (SE) are associated with credits assigned to the product-systems of ethanol production in Brazil, and molasses and ethanol production in India. SE-C assumes lower credits than SE-O, which ultimately affect the ethanol cradle-to-gate results. More specifically, ethanol has higher impacts under SE-C when compared to SE-O. It may be expected that results for bio-PET would follow a similar pattern, however, the reverse is noticed. Bio-PET has lowest cradle-to-gate emissions under SE-C, followed by EA-C, SE-O, and finally EA-O shows most conservative estimates. This is due to electricity consumption at the bio-MEG production step, the source of which (national, highly coal-based or local, highly hydro-based) depends on the allocation approach. Per kilogram of bio-MEG the total electricity consumed by the system is larger than the surplus provided at the ethanol step. Therefore the influence of the credits is overcompensated by the electricity input for bio-MEG production, which is the final determinant for ranking the allocation approaches. Economic allocation (EA) is not related with system credits; however, it assumes regional grid (low CO<sub>2</sub> emission intensity in EA-C) and national grid electricity input for bio-MEG production (high CO<sub>2</sub> emission intensity in EA-O). The difference between the two EA approaches is approximately 10%. The different electricity source also explains the varying contribution of the bio-MEG step across the four allocation approaches.

For bio-PET<sub>IGL</sub> the contribution of bio-MEG production is lowest, because steam is not only produced from coal but also from biogas available from the attached distillery. However, the latter has an impact on the contribution of ethanol production, which is higher than in bio-PET<sub>IN</sub>. This is because, the integrated bio-MEG and ethanol production (bio-MEG<sub>IGL</sub>) has the same steam source, while heat requirements of ethanol production in Uttar Pradesh, India are covered by bagasse and biogas. In other words, the benefit of using biogas is fully assigned to ethanol in bio-PET<sub>IN</sub>, while it is shared among ethanol and MEG production in bio-PET<sub>IGL</sub>.<sup>13</sup> The SE-O approach leads to high spread between the high and the low performance case for bio-PET<sub>IN,IGL</sub>, when compared to the base case (approximately  $\pm 15\%$  from the average 2 kg CO<sub>2eq</sub>/kg<sub>bio-PET,IN</sub>). The

smallest variation is noticed for bio-PET<sub>BR</sub> under EA-C (approximately 1% from the average 2.3 kg CO<sub>2eq</sub>/kg<sub>bio-PET,BR</sub>).

Similar to GHG emissions, bio-HDPE shows significant savings in NREU when compared to pchem-HDPE (around 65%, Fig. 2). Apart from fossil energy use to meet process energy requirements, pchem-HDPE is produced from petrochemical feedstocks which is reflected in the large NREU.<sup>14</sup> For bio-HDPE, fossil energy requirements are primarily due to process energy requirements, production of material inputs, transport, and NREU due to ethanol production (diesel use in sugarcane production) while the feedstock is exclusively bio-based. The range across the allocation approaches does not show significant variation. When the higher and the lower performance cases are assumed, then the results range only for SE-O approximately by  $\pm 25\%$ , compared to the base case result.

Bio-PET offers NREU savings from 3 to 11% compared to pchem-PET (Fig. 3). While the NREU for PTA and polymerisation is identical for both routes, savings occur from producing bio-MEG from renewable feedstocks. However, the contribution of transport decreases the overall difference. Similar to GHG emissions, bio-PET<sub>BR</sub> causes higher NREU than bio-PET<sub>IN,IGL</sub>, due to higher NREU of Brazilian ethanol and its transport to India. The higher and lower performance cases lead to the widest variation for SE-O, which however, does not influence significantly the relative savings.

#### 4.1.2. Emissions from land use change

Due to the large influence that land use change (LUC) may have in GHG emissions of bio-based plastics (Piemonte and Gironi, 2011) and the significance they receive in the policy agenda (EC, 2012), we attempt to account for both direct and indirect LUC emissions as described in Tsiropoulos et al. (2014). For bio-HDPE, additional GHG emissions due to LUC, range from 0.16 to 2.38 kg CO<sub>2eq</sub>/kg<sub>bio-HDPE</sub>, for low (3 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub>) and high (46 g CO<sub>2eq</sub>/MJ<sub>ethanol</sub>) LUC emission factors, respectively (Wicke et al., 2012). Adding this value to the results presented in Fig. 2, the net GHG emissions range from -0.7 to 1.8 kg CO<sub>2eq</sub>/kg<sub>bio-HDPE</sub>, which are approximately 130% and 20% lower than the GHG emissions of today's pchem-HDPE, for low and high LUC emission factor, respectively. For bio-PET<sub>BR</sub> the LUC GHG emissions range from 0.03 to 0.4 kg CO<sub>2eq</sub>/kg<sub>bio-PET,BR</sub>. The highest LUC emission factor essentially cancels the bio-based carbon storage credit of bio-PET<sub>BR</sub>. This entails that bio-PET<sub>BR</sub> production may even lead to an increase in CO<sub>2</sub> emissions compared to pchem-PET, by 4–13% and 22–30%, for low and high LUC emission factor, respectively. Due to the wide range in LUC emission factors, these results should be interpreted with caution.

Taking into account only direct LUC, emissions of bio-HDPE range from -0.55 to -0.88 kg CO<sub>2eq</sub>/kg<sub>bio-HDPE</sub> depending on the allocation approach (2–3% lower compared with bio-HDPE GHG emissions without direct LUC, Fig. 2). For bio-PET<sub>BR</sub> there is very limited influence of direct LUC emissions (less than 0.2% of emissions shown in Fig. 3). For bio-PET production from Indian sugarcane molasses, we do not account for direct LUC and indirect LUC because to our knowledge, there is a lack of publicly available reliable data that could support calculations on the effects of land use change for Indian sugarcane ethanol production. However, if ethanol production in India increases displacement effects are likely to take place and LUC emissions should be accounted for (Tsiropoulos et al., 2014).

#### 4.1.3. Stillage treatment

The assumption that stillage is treated via anaerobic digestion is critical for estimating CO<sub>2</sub> emissions. In particular, if stillage is

<sup>13</sup> In bio-PET<sub>IGL</sub> the ethanol is partly from the integrated distillery and partly from ethanol procured from distilleries in Uttar Pradesh (31% and 69%, respectively; share estimated based on plant capacities of 80,000 kl ethanol/year and 200,000 tMEG/year).

<sup>14</sup> The higher and lower heating value of polyethylene is 42.8 and 42.5 MJ/kg, respectively.

disposed without prior treatment, and anaerobic conditions prevail, then Indian ethanol emissions may range from 2.6 to 3.1 kg CO<sub>2eq</sub>/kg<sub>ethanolIN</sub>, as opposed to 0.09 to 0.64 kg CO<sub>2eq</sub>/kg<sub>ethanolIN</sub> which are the emissions of the reference system (Tsiropoulos et al., 2014). This increase is primarily due to the high global warming potential of methane but also due to the lower biogas recovery of the system – thus lower credits for surplus power provided to the grid. The emissions of bio-PET<sub>IN</sub> increase significantly to 2.8–3.1 kg CO<sub>2eq</sub>/kg<sub>bio-PET,IN</sub>, as opposed to 2–2.3 kg CO<sub>2eq</sub>/kg<sub>bio-PET,IN</sub> in the base case.

#### 4.1.4. CHP plant and other improvement options

In the production premises of the MEG producer's ethanol distillery a CHP plant is operated (UNFCCC, 2006). In Fig. 4 we present results for the GHG emissions of bio-PET<sub>BR,IN</sub> which assume that the CHP plant covers all steam demand of MEG production.<sup>15</sup> For co-production of steam and electricity we apply the four allocation approaches as described in this study (Table 2, Table 3). In one case, we assume that a feed-in tariff (FIT) for CHP electricity is in place, which makes it profitable for the producer to sell CHP electricity and purchase grid electricity for process requirements (CHP FIT). In this case, only CHP steam is used in the MEG process and electricity is supplied from the grid. To assess the effect of the FIT we also present results assuming that no FIT is in place (CHP no FIT). In this case both CHP steam and electricity are used in the MEG process, and only the remaining (net) process electricity requirement is supplied from the grid. Incorporating the CHP plant to the study reduces the GHG emissions of bio-PET by 3–10% when compared to the base case results of Fig. 3 (Section 4.1.1). The difference is smallest (i.e. showing a limited effect of CHP on the GHG emissions of bio-PET) under the SE-C approach due to the low credits assigned for displacement of grid electricity by the CHP. The difference is largest (i.e. showing a large effect of CHP on the GHG emissions of bio-PET) under economic allocation because impacts from CO<sub>2</sub> intensive coal-based steam generation are allocated due to co-generation of electricity. The emissions of bio-PET estimated with a FIT are by 3–8% lower compared to a no FIT case in SE-C, EA-C and EA-O. However, under SE-O the FIT assumption leads to higher emissions than assuming no FIT because emissions from process electricity from the grid (CHP FIT) are higher than the emissions from process electricity from the CHP (CHP no FIT).

In addition to CHP, we assume four further improvement scenarios. These include: a) integration of the bio-MEG plant with a distillery that fully meets the ethanol demand for bio-MEG, b) fuel switch from coal to natural gas as primary fuel for steam production c) advanced MEG production technology based on technology licensor instead of producer information and d) implementation of best available technology for bio-MEG production. Other possible improvements, e.g. process heat integration or the fuel switch from coal or natural gas to biomass at the CHP plant, are not assessed in this paper. To ensure comparability with the base case bio-PET production, we assume that all improvements are applied in India.

- a) Integration: The integrated production of bio-MEG and ethanol makes it possible to utilise biogas for process steam production, thus reducing net coal input required to meet the steam demand for bio-MEG and ethanol production. In the

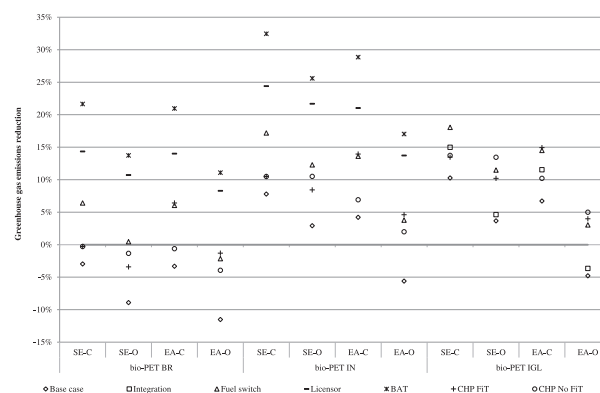


Fig. 4. Cradle-to-gate greenhouse gas emission reduction potentials of partially bio-based PET compared to today's petrochemical PET (0% line).

case studied above (bio-PET<sub>IGL</sub>), the distillery's capacity is not sufficient to meet the bio-MEG plant's maximum capacity and the ethanol demand is supplemented from other distilleries. This constrains the availability and use of biogas at the bio-MEG production site. Assuming that bio-MEG production is integrated with a distillery that fully meets its ethanol demand, the biogas availability per kilogram of bio-MEG increases; hence the net coal input decreases. The latter has an impact on GHG emission reduction in the range of 15%, 5%, 12% and –4%, when compared to pchem-PET for SE-C, SE-O, EA-C and EA-O, respectively ('Integration', Fig. 4). To place this in perspective, the GHG emission reduction of a non-integrated case (bio-PET<sub>IN</sub>) is 10%, 4%, 7% and –5% for SE-C, SE-O, EA-C and EA-O, respectively ('Base case', Fig. 4). This scenario is applied only in bio-PET<sub>IGL</sub>.

- b) Fuel switch: India has abundant coal supplies, which are used as a primary fossil fuel for several energy applications. The high emission factor of coal combustion along with other environmental impacts compromises the environmental benefits of the systems studied. A fuel switch from coal to natural gas for steam generation in bio-MEG production would result in further reduction of approximately 10% of the GHG emissions of bio-PET production when compared to the base cases ('Fuel switch', Fig. 4). This scenario is applied in bio-PET<sub>BR</sub>, bio-PET<sub>IN</sub> and bio-PET<sub>IGL</sub>.
- c) Licensor: For this scenario we use data provided by technology licensors. The results could apply to a newly commissioned bio-MEG plant ('Licensor', Fig. 4). To compare this scenario with the base case we assume steam production from coal. Compared to the base case bio-PET, this would lead to additional GHG emission reduction in the range of 20%. This scenario is applied in bio-PET<sub>BR</sub> and bio-PET<sub>IN</sub>.
- d) BAT: This scenario assumes for bio-PET similar process energy requirements for ethylene dehydration as assumed for bio-ethylene production (i.e. the technology assumed in bio-HDPE, which was found to have lowest energy requirements per kilogram of ethylene). Furthermore, for the steps ethylene oxidation and hydrolysis-to-ethylene glycol we assume the same process energy requirements as in petrochemical MEG production. In addition, we assume that natural gas is used for steam generation. By aggregating process energy requirements of each step we conclude that significant GHG emission reduction can be achieved ('BAT', Fig. 4). These savings range from 10% to 20% and 15% to 30%, compared to today's pchem-PET, for bio-PET<sub>BR</sub> and bio-PET<sub>IN</sub>, respectively.

<sup>15</sup> It depends on the size of the CHP plant and the needs of the entire site whether or not the CHP should be included for the conversion of Brazilian and Indian ethanol to MEG (as required for bio-PET<sub>BR,IN</sub>). Such decisions are often difficult to take for a concrete industrial setting if the provided data are ambiguous.



**Table 3**  
Approaches chosen for the multifunctionality problem at the CHP plant.

	SE-C	SE-O	EA-C	EA-O
<b>Electricity</b>				
FIT	Displacement of power grid electricity: Low CO <sub>2</sub> emission intensity (0.55 kg CO <sub>2eq</sub> /kWh) Process uses electricity from local grid	High CO <sub>2</sub> emission intensity (1.1 kg CO <sub>2eq</sub> /kWh) Process uses electricity from national grid	Allocation factor: 40% Process uses electricity from local grid	40% Process uses electricity from national grid
No FIT	No displacement of power grid electricity: CHP electricity used in process Remainder electricity supplied from local grid	CHP electricity used in process Remainder electricity supplied from national grid	Allocation factor: 40% Remainder electricity supplied from local grid	40% Remainder electricity supplied from national grid
<b>Steam</b>				
FIT/no FIT	Used in process	Used in process	60% Used in process	60% Used in process

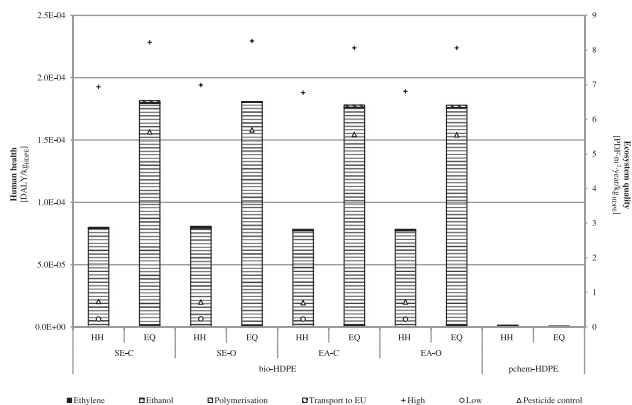
**Table 4**  
Net cradle-to-gate greenhouse gas emissions of bio-PET produced from Brazilian and Indian ethanol under different improvement options in kg CO<sub>2eq</sub>/kg<sub>PET</sub>.

	bio-PET <sub>BR</sub>				bio-PET <sub>IN</sub>				bio-PET <sub>IGL</sub>				pchem-PET
	SE-C	SE-O	EA-C	EA-O	SE-C	SE-O	EA-C	EA-O	SE-C	SE-O	EA-C	EA-O	
Base case	2.21	2.34	2.22	2.39	1.98	2.08	2.06	2.27	1.93	2.07	2.00	2.25	2.15
CHP FIT	2.15	2.22	2.01	2.17	1.92	1.97	1.85	2.05					
CHP no FIT	2.15	2.18	2.16	2.23	1.92	1.92	2.00	2.10					
Integration									1.83	2.05	1.90	2.23	
Fuel switch	2.01	2.14	2.02	2.19	1.78	1.88	1.85	2.07	1.76	1.90	1.84	2.08	
Licensor	1.84	1.92	1.85	1.97	1.62	1.68	1.70	1.85					
BAT	1.68	1.85	1.70	1.91	1.45	1.60	1.53	1.78					

Fig. 4 and Table 4 show that 'BAT' offers the highest savings on greenhouse gas emissions compared to the base case and pchem-PET ('BAT' is displayed on the upper side on the improvement options shown in Fig. 4). Other options also offer improvements; in order of decreasing contribution these are 'Licensor', 'Fuel switch' and 'CHP'. The combination of some of these measures (e.g., 'Fuel switch' and 'CHP') and the use of biomass for the supply of process steam can potentially reduce the greenhouse gas emissions even further, depending on the implemented technological option and the chosen assessment methodology.

#### 4.2. Human health and ecosystem quality

Potential impacts of bio-HDPE and bio-PET on HH and EQ are significantly higher than those of their petrochemical counterparts. The impact of bio-HDPE on HH is 50 times higher and on EQ is 2 orders of magnitude higher than pchem-HDPE (Fig. 5). The results



**Fig. 5.** Potential impacts of bio-based HDPE production from Brazilian sugarcane ethanol on human health and ecosystem quality.

are similar across all four allocation approaches. Ethanol production dominates (98%) in both impact categories, with agriculture being the main contributing factor. The remaining impacts originate from ethanol dehydration, polymerisation and final transport to Europe. The contribution analysis of Brazilian sugarcane ethanol production on HH shows that carcinogenic and non-carcinogenic emissions due to pesticide application are responsible for around 80% of the impact. The remainder is primarily due to pre-harvesting burning practices but also to some extent due to bagasse combustion in boilers in Brazil (Tsiropoulos et al., 2014).

Due to uncertainties in active ingredients of pesticides applied in sugarcane cultivation we analyse two cases: a) we exclude impacts of a highly toxic substance (daconate) from unspecified pesticides ('Pesticide control', Fig. 5) b) we assume that all unspecified pesticides are the pesticide daconate ('High', Fig. 5). In addition, we consider a third case c) where we assume that no pre-harvesting burning practices are applied ('Low', Fig. 5). The total impact of bio-HDPE production on HH declines by 75% if daconate is eliminated and an additional 70% reduction is achieved if pre-harvesting burning practices are phased out. The remaining impact is four times higher than pchem-HDPE, of which one-fourth is due to international transport of bio-HDPE to Europe, polymerisation and ethanol dehydration.

For EQ, land occupation contributes approximately 80% to the total impact (4.3 m<sup>2</sup> org.arable/kg<sub>bio-HDPE</sub>). Excluding land occupation, 90% of the remainder impact is associated with terrestrial ecotoxicity, and 9% with terrestrial acidification and nitrification. Eliminating daconate reduces the EQ impact of bio-HDPE by approximately 50% (if land occupation is included reduction is 13%; 'Low', Fig. 5). However, the comparison with pchem-HDPE still indicates a factor 40 higher impact of bio-HDPE on EQ. This is primarily related with heavy metals in sugarcane production. 10% of the impact is due to bio-ethylene production and transport to Europe. For terrestrial acidification and nitrification the two processes that contribute are pre-harvesting burning and bagasse

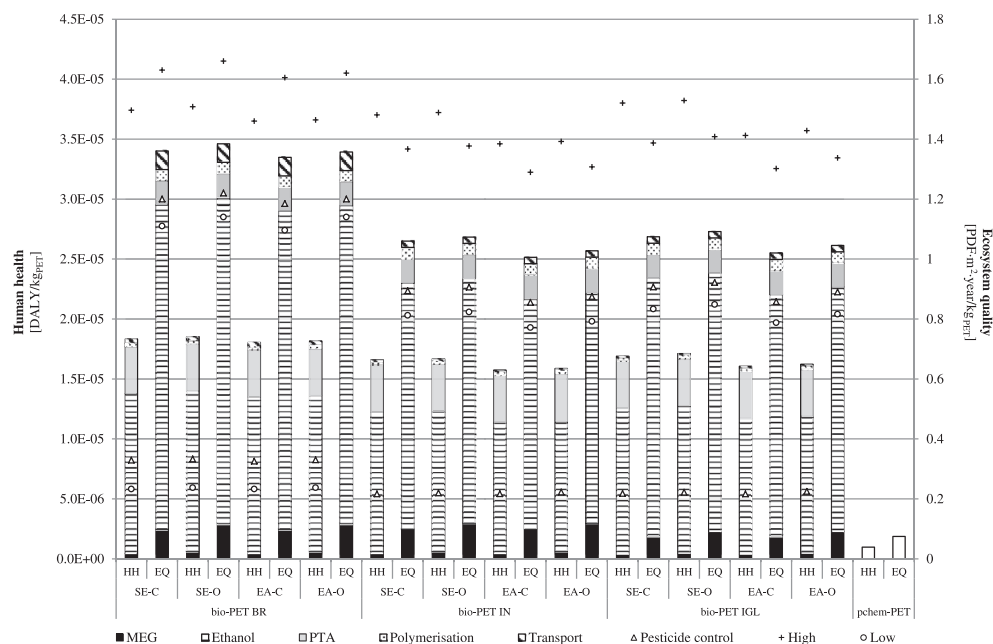


Fig. 6. Potential impacts of bio-based PET production from Brazilian and Indian sugarcane ethanol on human health and ecosystem quality.

burning in co-generation facilities. Eliminating the former reduces further the impact by 3%. On the contrary, if the unspecified fraction of pesticides is assumed to be daconate, the potential impact of bio-HDPE on HH and EQ increases by 140% and 25%, respectively ('High', Fig. 5).

Next we normalise the results based on factors of Impact 2002+ (Jolliet et al., 2003). The impacts on HH appear to be significantly higher than those on EQ, climate change or resources. The impact categories that contribute most are non-carcinogens, carcinogens and respiratory inorganics. Eliminating daconate and pre-harvesting burning practices slightly changes the pattern and respiratory inorganics become the dominating impact category, while carcinogens and non-carcinogens are significantly reduced. Even though HH is still the most important area of protection, the relative difference to EQ, climate change and resources is reduced from more than 1 order of magnitude to a factor 2, 4 and 5, respectively (Supporting information). Despite benefits that bagasse co-generation exhibits (eliminates the need for external fuels and generates surplus electricity, assigned as credit to the ethanol product-system) it contributes to impacts of bio-HDPE production on HH through particulate matters emissions. These emissions need to be reduced by appropriate combustion technologies. It should be noted that spatial differentiation and local conditions are critical for appropriately assessing the fate of and exposure to these pollutants.

For bio-PET, impacts on HH and EQ are shown in Fig. 6. Similar to bio-HDPE the allocation approach does not influence the final results. Both HH and EQ impacts related to bio-PET<sub>IN,IGL,BR</sub> are between a factor 14 and 19 higher than for pchem-PET. PTA production contributes approximately 20% to the HH impacts and approximately 6–8% to the EQ impacts of bio-PET. To estimate the impacts of PTA production and polymerisation on HH and EQ we use data from Ecoinvent (Hischer, 2007), while the comparison with pchem-PET is based on new eco-profile data (PlasticsEurope, 2011). This entails that the comparison for HH and EQ is not fully aligned for PTA production and polymerisation. These steps should have the same absolute contribution to both bio-based and petrochemical PET. However, even when excluding the contribution of

these steps from bio-PET impacts, it still has higher impact than pchem-PET, which is a plausible outcome (by approximately a factor of 15 for bio-PET<sub>BR</sub> and 12 for bio-PET<sub>IN</sub> on HH and EQ). Similar to bio-HDPE, most of the impacts originate from ethanol production. Ethanol contributes approximately 70% on HH and 80% on EQ, for bio-PET<sub>BR</sub> and bio-PET<sub>IN</sub>, respectively. In addition, 4% of the impact on HH comes from transport, and MEG production (each contribute from 1 to 3%). Bio-MEG production contributes 7–11% depending on the ethanol source and allocation, while transport contributes between 2 and 5% on the EQ impacts, for bio-PET<sub>BR</sub> and bio-PET<sub>IN</sub>.

The absolute impact of bio-PET<sub>BR</sub> is slightly higher because in this process chain all impacts of sugarcane production are allocated to ethanol. In the process chain of bio-PET<sub>IN,IGL</sub>, impacts of sugarcane production are allocated among sugar and molasses. The (allocated) sugarcane required for bio-PET from Brazilian ethanol is 5 kg<sub>cane</sub>/kg<sub>bio-PET,BR</sub>, while it is only 2.6 kg<sub>cane</sub>/kg<sub>bio-PET,IN</sub> from Indian ethanol. While the Brazilian sugarcane input is twice the Indian sugarcane input, the relative difference on HH and EQ impacts is significantly lower, by 10% and 20%, respectively. This is a consequence of the higher HH and EQ impacts of Indian sugarcane as compared to Brazilian sugarcane production (Tsiropoulos et al., 2014).

Eliminating the pesticide daconate reduces the impact on HH by approximately 55% in bio-PET<sub>BR</sub> and 65% in bio-PET<sub>IN</sub>. Phasing out pre-harvesting burning practices in Brazil reduces the impact on HH further by 30%. Thereafter bio-PET is a factor 6 higher than pchem-PET (pre-harvesting burning is not applied in Uttar Pradesh) ('Low' for bio-PET<sub>BR</sub> and 'Pesticide control' for bio-PET<sub>IN</sub>, Fig. 6). Assuming all unspecified pesticides as daconate practically doubles the impact on HH for both process chains ('Pesticide control' and 'High', Fig. 6). For EQ the results are similar, i.e. excluding daconate reduces the impact by approximately 10–15% ('Pesticide control', Fig. 6). Assuming that all unspecified pesticides have the impact of daconate increases the EQ impact by 20% and 30% for bio-PET<sub>BR</sub> and bio-PET<sub>IN</sub>, respectively ('High', Fig. 6). A possible fuel switch from natural gas further reduces the impacts of bio-PET on HH by 5% and on EQ by 5 and 10% for bio-PET<sub>BR</sub> and bio-PET<sub>IN</sub>

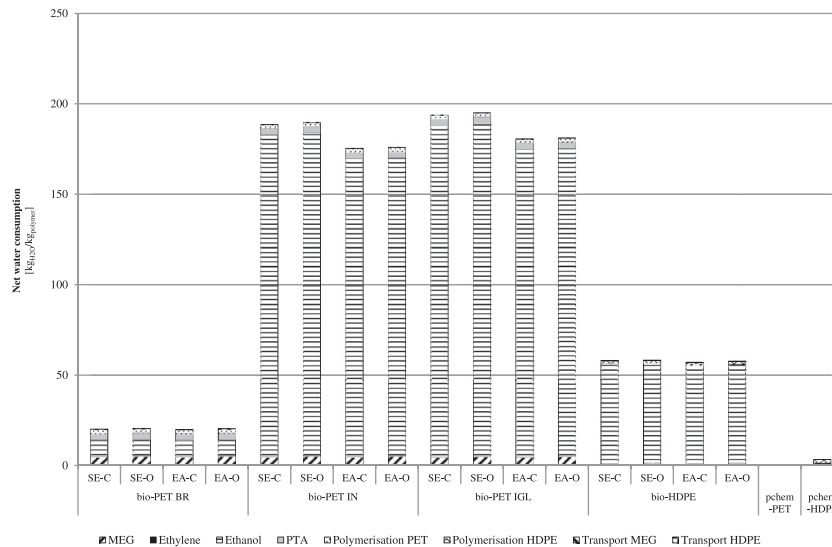


Fig. 7. Net water consumption of bio-based and petrochemical HDPE and PET production.

(‘Low’, Fig. 6). When excluding the contribution of land occupation on EQ ( $0.7 \text{ m}^2 \text{ org.arable/kg}_{\text{bio-PET, BR}}$ ,  $0.5 \text{ m}^2 \text{ org.arable/kg}_{\text{bio-PET, IN}}$ ), the lowest estimate is 4 times higher for bio-PET<sub>BR</sub> and approximately 3 times higher for bio-PET<sub>IN</sub>, when compared to pchem-PET. PTA production, polymerisation and transport account for approximately 50% of the remaining impact. Terrestrial ecotoxicity and acidification/nutritification of ethanol production are contributing most to the remaining impacts, due to heavy metals, other pesticides,  $\text{NH}_3$  and  $\text{NO}_x$  emissions due to fertilisers and bagasse burning.

Normalising the results of bio-PET<sub>IN</sub> shows that impacts on HH are significantly higher than impacts on EQ, namely by a factor 30. However, when compared to climate change and resources they are larger only by a factor 10 and 6, respectively. This indicates that both impacts on HH and on climate change and resources have high importance. When daconate is excluded from Indian sugarcane production, then the impacts on HH are still dominating but the difference with climate change and resources is reduced to a factor 3 and 2, respectively (Supporting information). The normalised results of bio-PET<sub>BR</sub> show a similar pattern, with the exception of normalised respiratory inorganics, which are comparable with GHG emissions and NREU.

#### 4.2.1. Net water consumption and its contribution to human health and ecosystem quality

Due to large use of water resources in agriculture and consequently in the production chain of bio-based products, we present results for bio-HDPE and bio-PET, and assess their impact on HH and EQ based on cause–effect relationships explained in Pfister et al. (2009). Net water consumption is calculated based on freshwater use for irrigation and water consumed for the processing steps that lead to the final polymers. Water outputs such as stillage recycling by ferti-irrigation in Brazilian sugarcane fields or other effluents returned to nature are deducted to calculate net water input. Results are shown in Fig. 7.

In south-central Brazil, sugarcane crops are practically not irrigated and only rainwater is used, which is not included in the above results. On the contrary, in Uttar Pradesh, India sugarcane plantations are irrigated by freshwater, which is included in the results. This explains why water consumption for bio-PET<sub>BR</sub> is by a factor 10 lower from bio-PET<sub>IN</sub>. Assuming water efficiency improvements in sugarcane irrigation for existing crops in Uttar Pradesh (Tsiropoulos et al., 2014) would reduce water consumption to  $120 \text{ l/kg}_{\text{bio-PET, IN, IGL}}$ , which

is still a factor 6 higher than bio-PET<sub>BR</sub>. Given the local water stress in the region, such a difference can be considered significant as additional demand for ethanol or Indian bio-based materials may put more pressure in the depleting groundwater resources. Taking into account regional water stress indices and damage factors (Pfister et al., 2009) the impact of bio-PET<sub>IN</sub> increases by 3% on HH and 9% on EQ, respectively (Supporting information).

#### 4.2.2. Eutrophication

Increasing nutrient loads in water bodies due mineral fertiliser use in agricultural production is a major concern for water quality. We estimate that freshwater eutrophication related to the production of the bio-based polymers is  $0.45 \text{ g PO}_4/\text{kg}_{\text{bio-HDPE}}$  and bio-PET<sub>BR</sub> while it is  $0.55 \text{ g PO}_4/\text{kg}_{\text{bio-PET, IN, IGL}}$ . If we assume higher P-surface runoff factor (10% P of P-fertilisers, representative for Brazilian soils (Ometto et al., 2009)) then the emissions increase to  $1.3 \text{ g PO}_4/\text{kg}_{\text{bio-HDPE}}$ ,  $0.6 \text{ g PO}_4/\text{kg}_{\text{bio-PET, BR}}$  and  $1 \text{ g PO}_4/\text{kg}_{\text{bio-PET, IN}}$ . We notice a factor 3 increase in bio-HDPE because high quantities of sugarcane are required for its production. For bio-PET the increase is 30% and 50% from Brazilian and Indian ethanol due to lower sugarcane requirement. For bio-PET<sub>IN</sub> the increase is higher compared to bio-PET<sub>BR</sub> due to significantly higher P-fertiliser input in sugarcane production (Tsiropoulos et al., 2014). The contribution of freshwater eutrophication to EQ is very low (<1%). When compared to petrochemical counterparts, freshwater eutrophication of bio-HDPE is approximately 60 times higher. Note that latest eco-profile data for petrochemical ethylene suggest that eutrophication is  $1.1 \text{ g PO}_4/\text{kg}_{\text{ethylene}}$  (3 times higher when compared to petrochemical ethylene profiles used in this study (PlasticsEurope, 2012)). Assuming a similar increase for pchem-HDPE then eutrophication of bio-HDPE is a factor 20 higher.<sup>16</sup> For bio-PET<sub>IN</sub>, if stillage

<sup>16</sup> PlasticsEurope indicates that the eutrophication potential of pchem-PET is  $0.81 \text{ g PO}_4/\text{kg PET}$  (PlasticsEurope, 2011). It is calculated based on CML impact assessment, which includes characterisation factors for N-emissions, relevant for near shore eutrophication. In our analysis we calculate freshwater eutrophication potential limited to P-emissions and chemical oxygen demand. To compare freshwater eutrophication potential of pchem-PET with bio-PET we calculate the impacts of the former with Impact 2002+. The calculated emissions are by a factor 2 higher for bio-PET<sub>BR</sub> and by a factor 3 higher for bio-PET<sub>IN</sub> than pchem-PET. Similarly, accounting for the eutrophication potential of nitrogen emissions in bio-HDPE production results in a factor 10 higher estimate compared to current pchem-HDPE.

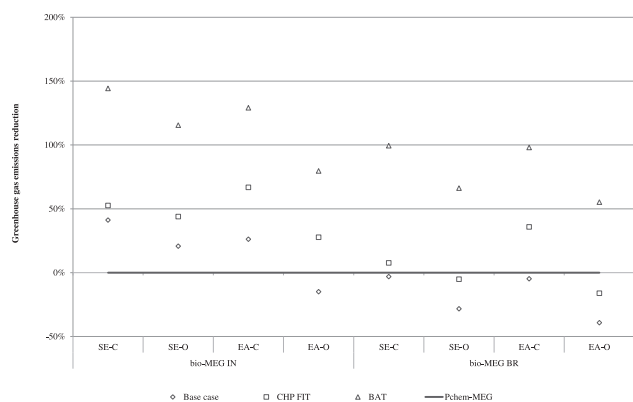


Fig. 8. Comparison of petrochemical MEG production (0% line) with bio-based MEG production from Indian and Brazilian ethanol.

is not treated but instead is released to the soil, the eutrophication increases by a factor 30 and the impact on EQ increases by 18%. The impact is even higher if stillage is disposed to water bodies (factor 50 increase in eutrophication by approximately and 28% on EQ).

#### 4.3. Comparison with other studies

The GHG emissions of bio-HDPE in this study differ with those reported in literature. The results of our EA-C are higher than those of the attributional approach of Liptow and Tillman (2012) by roughly 1.2 kg CO<sub>2eq</sub>/kg<sub>PE</sub>. More than 50% difference is associated with sugarcane production, for which Liptow and Tillman (2012) in their attributional approach did not include mechanised harvest, fossil CO<sub>2</sub> emissions from lime and urea, N<sub>2</sub>O and CH<sub>4</sub> emissions from unburned trash. The remaining difference is associated with ethanol production, dehydration and polymerisation. Ethanol production in Liptow and Tillman (2012) is not associated with GHG emissions. The small difference in ethanol dehydration to ethylene (0.15 kg CO<sub>2eq</sub>/kg<sub>PE</sub>) can be related with different process energy requirements based on different data sources. For polymerisation we assumed European production, while Liptow and Tillman (2012) assumed Brazilian production. The difference between our findings and those of Hunter et al. (2008) is in a similar range (1.3 kg CO<sub>2eq</sub>/kg<sub>PE</sub>). However, our study differs in scope and the product-systems studied. In the Supporting information we present a set of assumptions made to limit the difference between the two studies (e.g. excluding transport, process energy for dehydration met by bagasse), which reduce the difference on GHG emissions to 20%. High difference remains in eutrophication and acidification potential, which is associated with N-fertilisers and returned N-residues to soil and bagasse use in boilers.

The comparison of the base case results for bio-MEG production (excluding transport to Europe) with pchem-MEG (PlasticsEurope, 2012) reveals that bio-MEG<sub>IN</sub> has better performance on GHG emissions than pchem-MEG by 25–50% under SE-C, SE-O and EA-C (difference of 0.33–0.65 kg CO<sub>2eq</sub>/kg<sub>MEG</sub>). Only under EA-O bio-based MEG<sub>IN</sub> results in 15% more GHG emissions than pchem-MEG (0.23 kg CO<sub>2eq</sub>/kg<sub>MEG</sub>). Bio-MEG<sub>BR</sub> shows worse performance on GHG emissions by 3–40% under the different allocation approaches. Chen and Patel (2012) estimate that sugarcane-based bio-PET emits 1.0 kg CO<sub>2eq</sub>/kg<sub>PET,BR</sub>, which is lower by 50% from the results of this study. Main differences are associated with ethanol requirement for ethylene where the authors use stoichiometric yields, energy requirement for dehydration which the authors based on the theoretical heat of formation, and cradle-to-gate emissions of ethanol production which are based on older Brazilian sugarcane ethanol data. In addition, the authors do not take into account that production of

bio-MEG takes place in India in order to account coal-based process related emissions and do not include emissions from transoceanic transport.

The results become clearly more favourable when improvement potentials are considered (Section 4.1.4): If a CHP plant (with FIT) is implemented then the emission reduction of bio-MEG<sub>IN,BR</sub> increases by 10–40%. In this case emissions from bio-MEG<sub>BR</sub> under the SE approaches are comparable to pchem-MEG. Under EA-C bio-MEG<sub>BR</sub> shows lower emissions by 35% (0.57 kg CO<sub>2eq</sub>/kg<sub>MEG</sub>) and under EA-O it shows higher emissions by 15% (0.25 kg CO<sub>2eq</sub>/kg<sub>MEG</sub>). When assuming implementation of BAT bio-based MEG<sub>BR,IN</sub> has lower emission profile by 50–150% across all allocation approaches. Other improvement options discussed in Section 4.1.4 also bring benefits on GHG emissions, as well as their potential combinations (e.g. combination of ‘Licensor’ or ‘BAT’ with ‘CHP’). It should be noted that bio-based carbon from bio-MEG is deducted (1.45 kg CO<sub>2</sub>/kg<sub>MEG</sub>) (Fig. 8). In the Supporting information, comparison between other impact categories is presented.

## 5. Conclusions and recommendations

The purpose of this study was to assess the potential environmental impacts of bio-based HDPE and partially bio-based PET production across their production chain, to highlight where environmental pressures may be caused and indicate potentials for improvements. At the same time, results were compared with petrochemical counterparts of the bio-based plastics to demonstrate savings and tradeoffs between impact categories. This study confirms the findings of a review study on bio-based materials which suggests that they may lead to savings in NREU and GHG emissions relative to conventional materials but may on the other hand increase impacts associated with application of fertilisers (e.g. eutrophication) and pesticides during biomass cultivation (Weiss et al., 2012).

Compared to pchem-HDPE, the bio-based route offers significant savings in GHG emissions and NREU. These savings are reduced when iLUC emissions are considered, which does not change the general conclusion that bio-HDPE production is beneficial with regard to these impact categories.

Bio-PET production as assumed in the base case (without CHP) is in a similar range as pchem-PET production, with respect to GHG emissions and NREU (±10%). For the base case different allocation approaches were found to influence the results significantly. The difference between the highest and the lowest estimation on GHG emissions of different allocations is 9, 14 and 15 percent points in bio-PET<sub>BR,IN</sub> and bio-PET<sub>IGL</sub>, respectively. When comparing with GHG emissions of pchem-PET, the difference due to allocation in bio-PET systems may lead to different conclusions. The comparison with other studies revealed that previous estimates on GHG emissions of bio-PET were more optimistic with regard to potential savings taking the base case production into account. In view of comparable profiles with pchem-PET for the base case, uncertainties in the supply chain, but also taking a conservative and precautionary approach by accounting for iLUC emissions, it becomes evident that the production of bio-PET needs further improvements with regard to its environmental performance.

If the on-site CHP plant is assumed within the system boundaries then bio-PET has up to +15% lower GHG emissions than pchem-PET; again, the additional emission reduction by CHP varies depending on the case (BR, IN, IGL) and the allocation method. Bio-PET offers limited savings of GHG emissions when compared with fully bio-based plastics (bio-HDPE) but this is expected since the biogenic carbon content of bio-PET is much lower (approximately 70% of the polymer's weight is petrochemical PTA, which is the same in bio- and pchem-PET; hence comparative savings are limited). A first step could be to



switch the primary energy source of steam production in the bio-MEG plant from coal to natural gas. With regard to Indian ethanol production, it is important that the industry moves towards providing surplus electricity to the grid (for those 55% of mills that currently do not yet do so) and that stillage is treated prior to any disposal to soils or water bodies. Strong efforts should be made to implement the latest technology developed for the petrochemical MEG production, which would offer significant improvement potentials for bio-PET (which in some cases were shown to reach approximately 30% compared to pchem-PET). Such performance can be expected by new bio-MEG plants. Other potential improvement options, which however are not assessed by this study, include a combination of the above measures (e.g. 'BAT' with 'CHP') and replacement of coal or natural gas by biomass. Nevertheless as explained above, the environmental profile is expected to improve in the future.

With respect to HH and EQ, bio-HDPE and bio-PET were found to have a higher impact by factors or even orders of magnitude compared to pchem-HDPE and pchem-PET. The largest contribution comes from sugarcane production. However, this result is subject to large uncertainty concerning specific pesticides used. It is therefore advised to establish a chain of custody which ensures that certain substances such as daconate are not used. Also for bio-based plastics produced from Brazilian ethanol it is important to ensure supply from areas where no pre-harvesting burning practices are applied or to promote termination of burning practices by other means. Apart from carcinogenic and non-carcinogenic impacts on human health, the normalisation analysis showed that for both bio-HDPE and bio-PET from Brazilian ethanol, respiratory inorganics caused by bagasse use in co-generation facilities and pre-harvesting burning practices play an important role. It is therefore advised to monitor closely and on a local level the possible pathways of those pollutants and the impacts that they may have to the population and sugarcane field workers surrounding fields and distilleries. Also proper technologies may be installed to reduce particle emissions to the atmosphere. Further work is needed on inventories of sugarcane pesticides but also on impact assessment methodologies related with toxicity. Moreover, further analysis is required for estimating PTA profiles and land use change emissions of Indian sugarcane production. For bio-PET from Indian ethanol, water consumption is very high; given the water stress of the region, this not only reduces available water resources but it is also related with an increase of the impact on HH and EQ. Last, in India the eutrophication can increase significantly if higher emission factors are assumed due to the high P-fertiliser use.

It becomes evident that there is untapped potential in the production chains of bio-HDPE and partially bio-based PET, which if fully exploited can contribute to further improvements in environmental and human health impacts compared to petrochemical plastics.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.jclepro.2014.11.071>.

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