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The global warming potential and the material utility of PET and bio-based PEF bottles over multiple recycling trips

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ABSTRACT

Biomass use and recycling are among the few options to reduce the greenhouse gas (GHG) emissions of the growing plastics sector. The bio-based plastic polyethylene furanoate (PEF) is a promising alternative to polyethylene terephthalate (PET), in particular for small bottle applications. For the first time, we assessed the life cycle global warming potential (GWP) for 250 mL PET and PEF bottles over multiple mechanical (MR) and chemical (CR) recycling trips in the Netherlands. We found that bio-based PEF would offer 50–74% lower life cycle GHG emission after one recycling trip compared to PET, depending on the waste management case. Our results also show that deposit-based recycling systems significantly reduce the cumulative cradle-to-grave net GHG emissions for both bottle types, especially when multiple recycling trips are applied. We propose complementary material utility (MU) indicators to reveal synergies and trade-offs between circularity and GWP: While deposit-based CR shows the best performance in terms of MU, it falls behind deposit-based MR when it comes to net GHG emissions due to the energy intensity of CR. Hence, combining mechanical and chemical recycling could contribute to achieving the goals of the circular economy and climate change mitigation alike.

1. Introduction

The production volume of plastics has grown faster than any other bulk material since 1971 (IEA, 2018) and is expected to double until 2050 compared to today's levels (Stegmann et al., 2022). The plastics sector was estimated to be responsible for 4.5% of the global GHG emissions in 2015 (Cabernard et al., 2021). With a contribution of almost 45%, packaging poses the largest demand for plastic polymer resins (Geyer et al., 2017). Among them, polyethylene terephthalate (PET) covers 22.5% of the global plastic packaging market, making it the second most used polymer resin in plastic packaging after low-density polyethylene (LDPE) (Geyer et al., 2017). Moreover, PET is the most recycled polymer in Europe (EPBP, 2017a; Eunomia, 2022).

Biomass use and recycling are among the few options to lower the plastic sector's growing greenhouse gas emissions (GHG) emissions and reduce dependence on virgin fossil feedstocks (IEA, 2018; Meys et al., 2021; Zheng and Suh, 2019). Together, biomass use and recycling are an integral part of a circular bioeconomy; a concept increasingly brought forward within the European Union (Stegmann et al., 2020).

A potential renewable alternative to PET is 100% bio-based polyethylene furanoate (PEF). PEF is formed by polymerising sugar-based furandicarboxylic acid (FDCA) with bio-based mono-ethylene glycol (MEG). PEF was developed by the Dutch company Avantium and the world's first commercial FDCA facility is expected to be completed by 2024 (Avantium, 2022a). PEF has superior gas barrier properties compared to PET, especially for O_2 (~10x) and CO_2 (~15x), thus requiring less material to achieve the same shelf life as conventional PET (Burgess et al., 2014 a-c; de Jong et al., 2022). Moreover, PEF has a higher modulus than PET, which allows for producing containers of equivalent mechanical strength with less material (de Jong et al., 2022).

This makes PEF particularly suited for food packaging applications that require a long shelf life while keeping the packaging lightweight.

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PEF can be used as monolayer bottles for soft drinks, beer, and juices, replacing glass bottles, aluminium cans, and multilayer bottles. The applicability of PEF is especially attractive in small packaging applications as these have a relatively high material footprint per unit of packaged product volume. Hence, one of Avantium's initial focus areas for the use of PEF are small bottles for carbonated or oxygen-sensitive products. PEF can be recycled using the same technologies as for recycling PET (de Jong et al., 2022). While not being biodegradable under industrial composting conditions as described in the European standard EN 13432, initial tests showed that PEF degrades substantially faster than PET: Under industrial conditions, 90% of PEF biodegraded within 240 and 385 days, in weathered and unweathered state respectively (de Jong et al., 2022).

A recent Life Cycle Assessment (LCA) conducted by the nova-Institut showed a GHG emission reduction potential for a clear, 250 mL monolayer PEF bottle of 33% when compared to an equivalent PET bottle over their life cycle (Puente and Stratmann, 2022). An assessment by Eerhart et al. (2012) estimated cradle-to-grave GHG emissions savings in the range of 45–55% when comparing PEF and PET polymers, but disregarding any application. Regarding the end-of-life (EoL), Eerhart et al. (2012) only assessed incineration.

Also for other bio-based plastics, the EoL phase has so far received limited or no attention in scientific literature. Only for polylactid acid (PLA) there are eleven LCA studies also addressing end-of-life options, followed by two studies on thermoplastic starch (TPS) and a few individual ones for other plastic types (Spierling et al., 2020). While Puente and Stratmann (2022) included a simplified end-of-life scenario for PEF bottles based on incineration and open-loop mechanical recycling, the study did not analyse alternative scenarios or the impact over multiple recycling trips. The effect of multiple recycling trips on GHG emissions has so far not been assessed for bio-based plastics from a LCA perspective and only twice for PET (Komly et al., 2012; Shen et al., 2011). Analysing multiple recycling trips would allow for calculating the overall GHG emissions of the polymers over their entire life cycle, including the emissions occurring after the first EoL phase. Multiple recycling trips would contribute to circular economy goals by increasing the product's utility (Ellen MacArthur Foundation, 2015; Konietzko et al., 2020). The Ellen MacArthur Foundation (2015) defines a product's utility as a combination of the length of a product's use phase and the intensity of its use.

Next to mechanical recycling (MR), chemical recycling (CR) is increasingly considered an alternative solution for treating plastic waste (Simon and Martin, 2019). MR refers to recovering plastic waste via mechanical processes, like shredding, washing and re-granulating, while CR breaks down the polymer structures of plastics. For PET, depolymerisation via glycolysis is seen as one of the most promising CR options (Raheem et al., 2019) and has already been implemented in industrial pilots in the Netherlands and Italy (Simon and Martin, 2019). Also for PEF, CR via glycolysis has already been demonstrated to be feasible (Gabirondo et al., 2021). Avantium is investigating technologies to chemically recycle PEF, amongst them glycolysis (de Jong et al., 2022; Sipos and Olson, 2013). There are initial LCAs for the glycolysis of PET (Lindgreen and Bergsma, 2018; Shen et al., 2010), but these do not assess multiple recycling trips even though such an analysis could highlight the advantages of CR technologies in terms of higher recycling yields and better quality of recyclates.

A lack of understanding of the impact of EoL options could hamper the transition to a circular (bio)economy in plastics value chains and lead to incomplete (life cycle) assessments of the overall climate benefit of bio-based compared to fossil plastics. While a bio-based plastic might have a lower global warming potential (GWP) than a fossil competitor in production, this advantage might be (partly) counterbalanced by worse performance in the EoL. Technical barriers or contaminations caused by bio-based plastics could hamper their integration into existing recycling systems (Alaerts et al., 2018). Simultaneously, a separate collection and treatment of bio-based plastics is economically challenging due to their current small market shares (Carus and Dammer, 2018). These issues could prevent the recycling of bio-based plastics or allow fewer or lower quality recycling trips compared to their fossil competitors.

We want to address these challenges and identify the cradle-to-grave climate impact of different waste management cases in the Netherlands for a small (250 mL) plastic bottle made from bio-based PEF compared to fossil-based PET, including the effects of multiple recycling trips. By complementing this with an analysis of the material utility, we want to identify and discuss potential trade-offs between circular economy and climate change mitigation goals.

With this work we provide the first comprehensive end-of-life assessment for bio-based PEF and, to our knowledge, the first LCA that considers multiple recycling trips for bio-based plastics. Moreover, we propose complementary indicators for LCAs that allow for analysing trade-offs and synergies between material utility or circularity and conventional LCA impact categories such as GWP.

We focus on the Netherlands as this is one of the potential initial target markets of Avantium's PEF bottles, after signing bottle offtake agreements with Refresco, a bottling company located in the Netherlands, and Resilux, a Belgian preform and bottle producer (Avantium, 2021). Furthermore, waste management data is well available for this country. The Netherlands recently introduced a deposit system for the more than 900 million small plastic bottles sold every year (Rijksoverheid, 2020), which we compare to the previous collection systems. This study is complementary to an LCA conducted by Puente and Stratmann (2022), which provides a detailed assessment of PEF bottles compared to PET bottles, including only one simplified EoL scenario. This study adds a more thorough analysis of the EoL by analyzing the impact of different Dutch waste management cases over multiple recycling trips.

2. Materials & methods

2.1. LCA goal & scope definition

2.1.1. Goal

We assessed the GWP of PEF and PET systems, following the LCA methodology laid out in the ISO standards 14040 and 14044 (ISO, 2006b, 2006a), using the LCA software SimaPro (version 9.1.0.11) and the Ecoinvent database version 3.7 for background data.

We aim to quantify the potential global warming impacts of 250 mL fossil-based PET and bio-based PEF bottles including four different waste management cases for the Netherlands (see also Fig. 1), being:

A. the waste management system for small plastic bottles in the Netherlands until 2021, based on post-separation, source-separation, MR and incineration with energy recovery (ER).
B. a waste collection predominantly based on a deposit system combined with MR and ER.
C. a waste collection predominantly based on a deposit system combined with CR and ER.
D. a non-circular scenario, assuming the complete incineration of the bottles with energy recovery.

The GWP over the life cycle of PET and PEF bottles including the above mentioned waste cases were assessed using the impact assessment method 'IPCC 2013 GWP100a'.

2.1.2. Functional unit

The functional unit of this study is *a* 250 mL *monolayer plastic bottle designed for single-use, providing minimum shelf life of at least 12 weeks for carbonated soft drinks*. The monolayer PET bottle fulfilling this function should weigh 24 g, and the monolayer PEF bottle weighs 13 g, according to calculations of Avantium, and substantiated by literature review, and feedback of industry experts (Puente and Stratmann, 2021, 2022). The weights were calculated based on the gas permeability values and

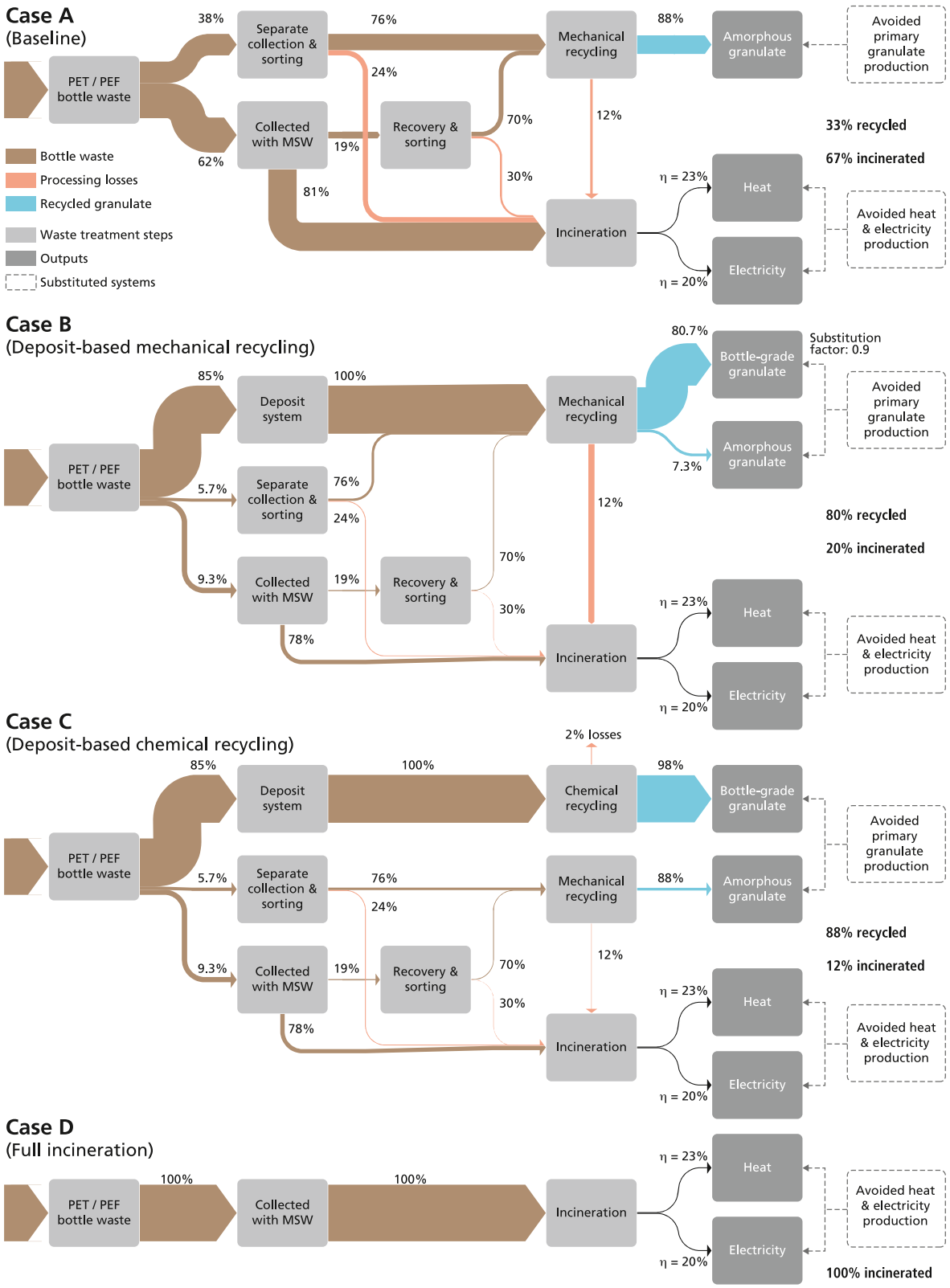

Fig. 1. The four analysed waste management cases for small PET & PEF bottles.
Notes: prepared with esankey 4.

material strength of the polymers, assuming no barrier-enhancing additives were used, and that ideal stretch ratios were gained during bottle blowing. The sensitivity of the results to the bottle weights was assessed in Appendix B. Due to superior barrier properties, the shelf life of the PEF bottle extends to more than 20 weeks (Puente and Stratmann, 2021, 2022). This functional unit is in line with the LCA of Puente and Stratmann (2022) and represents one of the potential initial target markets of Avantium's PEF bottles.

2.1.3. Product systems

The LCA has a scope from cradle-to-grave with a strong focus on EoL, following the goal of the study. The waste treatment cases are in the foreground analysis. The impacts of bottle production (cradle-to-gate), assessed in an LCA of nova-institute (Puente and Stratmann, 2021, 2022), were taken as the background system in our analysis. Due to our focus on the material flows of PEF and PET, we exclude the bottle's caps, neck rings, and labels since we assume they can be identical in PET and PEF bottles.

We cover the production of PET bottles from petrochemical feedstock and PEF bottles from bio-based feedstocks from cradle-to-gate, using results of existing assessments (Puente and Stratmann, 2022, 2021; CPME, 2017). Figs. 2 and 3 provide an overview of the bottle production. Potential emissions from the use phase are excluded because the impacts are considered negligible and comparable between PET and PEF bottles. However, the shelf-life difference between both bottles is addressed when discussing the material utility of both bottle types for multiple recycling trips.

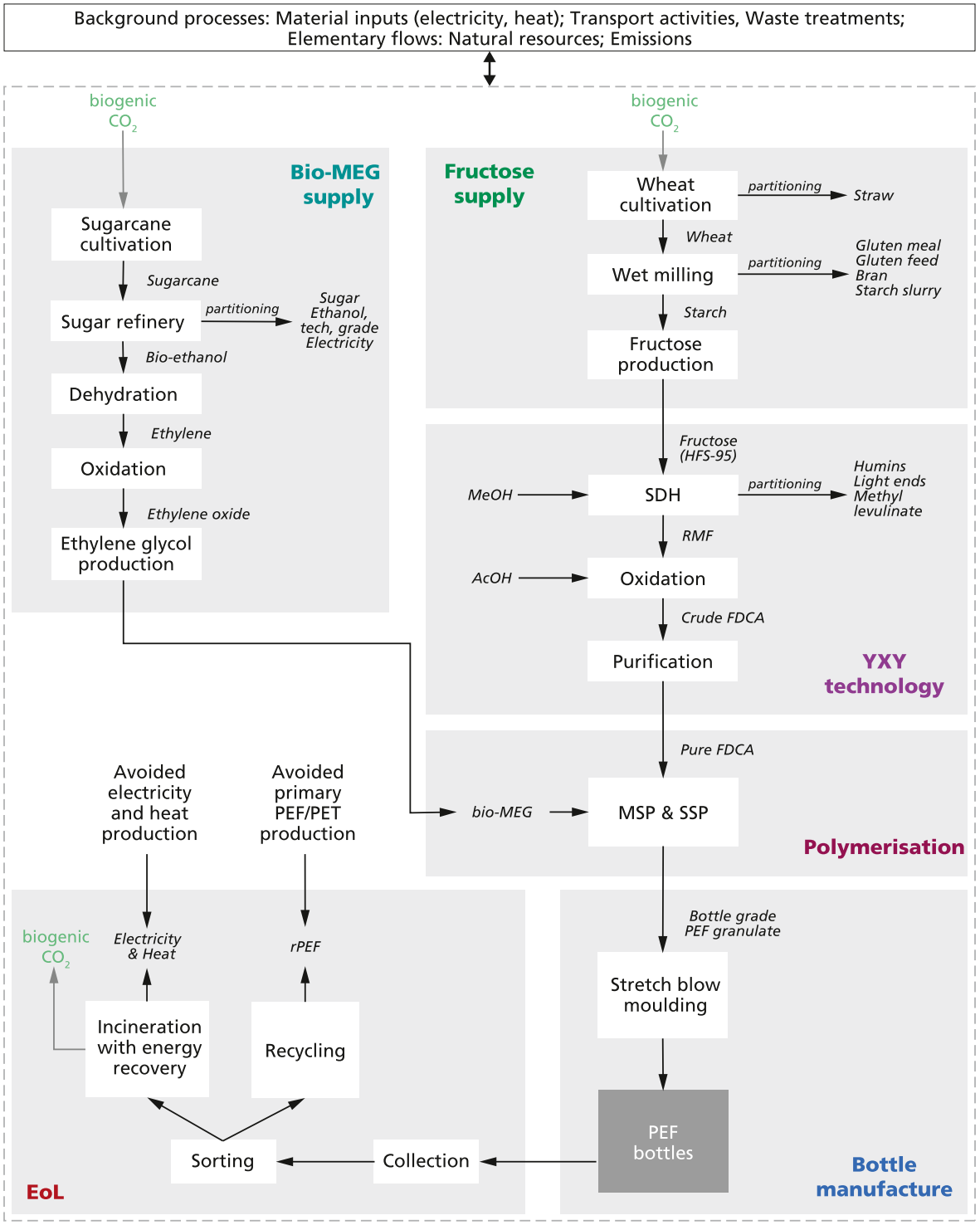

Fig. 2. Product system of PEF bottles.
Notes: adapted from Puente and Stratmann (2021); for simplicity, links to background processes and co-production were removed from the flowchart.

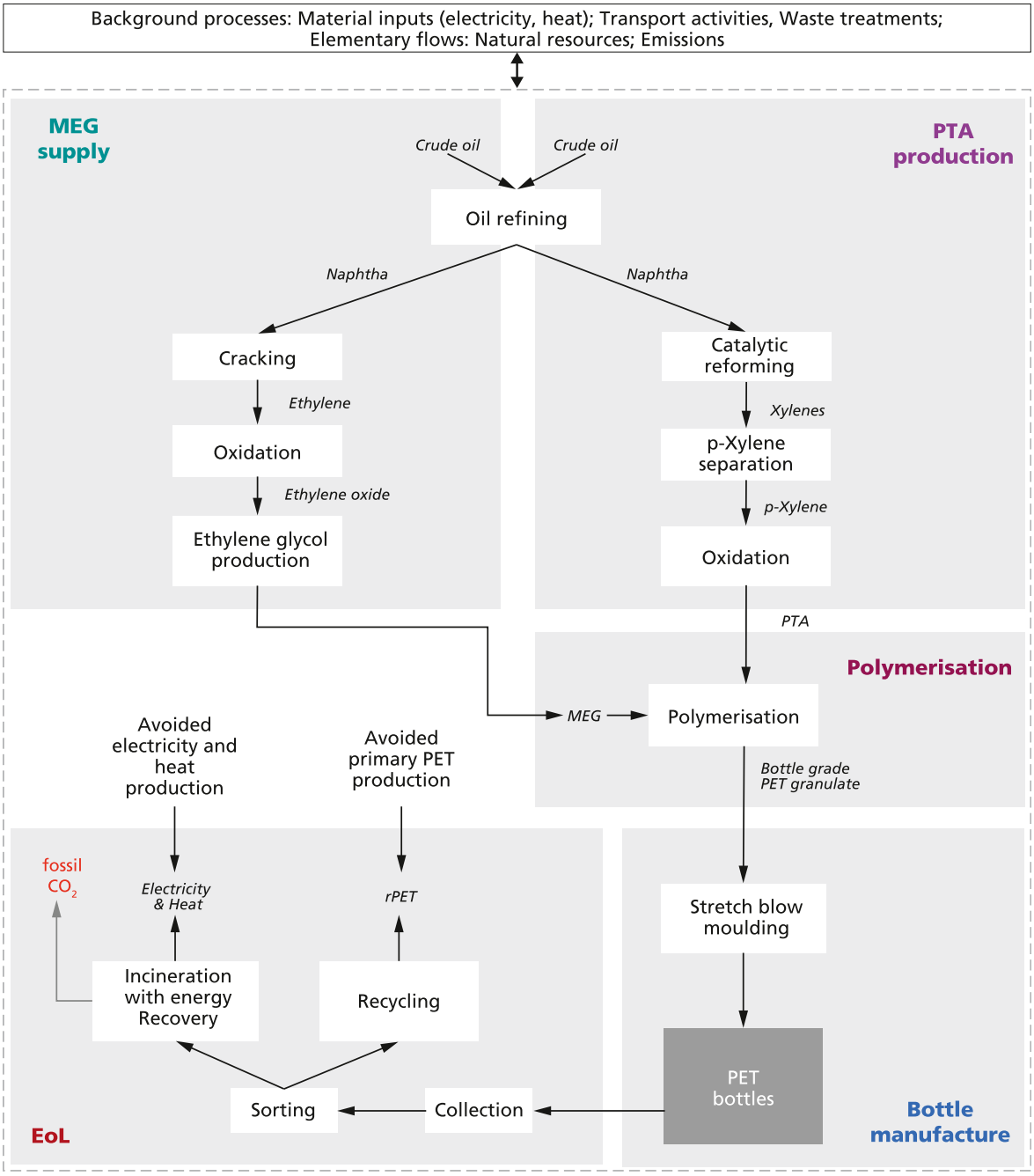

Fig. 3. Product system of PET bottles.
Notes: adapted from Puente and Stratmann (2021); for simplicity, links to background processes and co-production were removed from the flowchart.

We cover the EoL of the plastic bottles, consisting of collection, sorting, and waste treatment, including the transportation within and between the EoL stages. We distinguish between four waste management cases (see Fig. 1) for PEF and PET bottles. These cases differ in collection & sorting methods (post-consumer separation from municipal solid waste (MSW), source separation, and deposit system), recycling technologies (MR and CR), and the corresponding differences in the amount and quality of the recycled material. We assume that all bottles are eventually collected and ignore the impacts of littering plastic bottles. In the Netherlands, post-consumer plastic packaging waste collection differs by municipality. An assessment by Brouwer et al. (2019) estimates that in 2017 38% of Dutch post-consumer plastic packaging waste was collected separately at the source, and 62% ended up in MSW. 19% of the MSW fraction is sent to material recovery facilities for sorting, with the rest being sent to incineration plants (M. Brouwer et al., 2019). We assume the same collection rates for the small PET and PEF bottles. The mass flows of all processes are displayed in Fig. 1 and described in Section 2.2.3.

Overall, baseline case A has a high share of incineration with energy recovery (67%) and a mechanical recycling rate of 33%. The term recycling rate is used differently in literature, often referring to the plastics sent to recycling. We define the recycling rate as the net weight of recycled material divided by the net weight of collected material. Our recycling rate is thus the product of the sorting efficiency and the efficiency of the recycling process.

In July 2021, the Netherlands introduced a deposit system for plastic bottles smaller than 0.5 L (Rijksoverheid, 2020). We assume that 85% of the small PET and PEF bottles will be collected via a deposit system in

waste management cases B and C, based on a prognosis for the Netherlands (Schalkwijk and Mulder, 2011) and a study of the relationship between the collection rate and the deposit amount (Hogg et al., 2015). The remaining 15% are assumed to be collected in the same ratio as in case A. Case B continues with the MR of the plastic bottles, now achieving a higher recycling rate and higher plastic quality compared to Case A due to the introduced deposit system. In contrast, Case C uses CR for the plastic bottles collected via the deposit system. The remaining bottles in the CR case are mechanically recycled or incinerated in the same ratio as in case A. Case D assumes that all plastic bottles are collected along with the mixed municipal solid waste and then directly sent to ER. The mass and energy balances for the waste management cases in Fig. 1 are described in the inventory, chapter 2.2.

2.1.4. Assessing the waste treatment of PEF

Given the novelty of PEF plastic, there is no data available for PEF waste treatment. Due to similarities between PET and PEF, Avantium claims that PEF can be recycled using existing PET mechanical recycling assets like dryers, extruders, crystallizers and SSP equipment (de Jong et al., 2022). Moreover, PEF could be sorted by commercial near-infrared sorting equipment (EPBP, 2017b; de Jong et al., 2022). Furthermore, tests showed that small shares of PEF would not have a negative impact on recycled PET if mixed (Avantium, 2022b; EPBP, 2017b). An assessment by Avantium even claims that a 5% fraction of PEF in the PET stream would improve the quality of recycled PET and lead to a better crystallinity and a longer shelf life (Visser, 2020). Hence, the European PET Bottle Platform (EPBP, 2017b) provided an interim approval for up to 2% market penetration of PEF. In the absence of PEF specific data, we used the same waste treatment data for PEF as PET in this study after adjusting for differences in heating value and carbon content. With small market shares in the short term, Avantium expects that PEF will be integrated into PET recycling processes (open loop), but closed-loop PEF to PEF recycling systems are preferred, certainly at higher market shares (de Jong et al., 2022). For the purpose of this article we refer to open-loop when PEF is recycled together with PET and closed-loop for PEF-to-PEF or PET-to-PET recycling.

2.1.5. Geographical, temporal, and technological scope

The feedstock supply for Avantium's PEF production is based on starch from wheat cultivated in France used for fructose production and on ethanol-based bio-MEG produced from sugarcane in India. Avantium's FDCA plant will be located in Delfzijl, Netherlands, and the polymerisation of FDCA will mainly happen in European facilities. PET production data represents average European production. We assume that polymerisation, bottle manufacturing and waste treatment occur in the Netherlands.

Data for Avantium's YXY technology (see Fig. 2) was taken from Avantium's 5 kt/a flagship plant design (de Jong et al., 2022), scaled up to 100 kt/a, to represent the first commercialisation phase of PEF (Puente and Stratmann, 2022). The datasets are based on Aspen Plus process simulations and experimentally-derived data from Avantium's pilot plant in Geleen. Background energy inputs are based on the energy mix as presented in Ecoinvent (IEA data from 2017, extrapolated to 2020). Regarding the EoL, we combine state-of-the-art data for sorting, mechanical recycling and incineration (from Ecoinvent) with small-scale production data for chemical recycling (Shen et al., 2010). All EoL data was adapted to the Netherlands.

2.2. Life cycle inventory analysis

Table A.1 in appendix A summarizes all data used and assumptions made for the inventory of the LCA.

2.2.1. Allocation

In PEF production, partitioning the environmental burdens is required for by-products in wheat cultivation, wet milling, Avantium's YXY technology and the sugarcane refinery, see Fig. 2 (Puente and Stratmann, 2021). We use the cradle-to-gate PEF and PET polymer production results from Puente and Stratmann (2021, 2022) as input to our study, who applied economic allocation to allocate the environmental burdens. This is in line with the PAS 2050 recommendation on allocation choices when assessing bio-based products (BSI, 2012).

For modelling the end-of-life, the avoided burden approach was applied, following the recommendation of the ISO standards (ISO, 2006b, 2006a). This approach enables us to capture the environmental consequences of the analysed waste management cases. Some cases achieve better quality in recycled output (e.g., higher viscosity, clean streams suitable for bottle-grade applications), and PET achieves a higher energy recovery in incineration, which we want the results to reflect. Accordingly, we provide credits to our production systems for the recycled material and the generated energy, see section 2.2.3 on substituted product systems.

2.2.2. PET and PEF bottle production

PEF production consists of wheat cultivation, wet milling, and fructose production, followed by FDCA production via Avantium's YXY technology. The FDCA is then copolymerized with bio-based MEG to PEF granulate, see Fig. 2. Data on bottle-grade PEF granulate production was taken from the LCA conducted by nova-Institute (Puente and Stratmann, 2021, 2022).

PET production consists of oil refining, MEG and PTA production and their copolymerisation into PET. Puente and Stratmann (2021, 2022) used Ecoinvent data to model PET granulate production (CPME, 2017). Their results for PEF and PET production were calculated using the Ecoinvent 3.6 database, while our assessment uses the Ecoinvent 3.7 database for background data. To have consistent results in the granulate production, we used the results for both PEF and PET granulate production from Puente and Stratmann (2021, 2022) as input to our study.

Bottle-grade polymers are stretch blown into bottles. For stretch-blow moulding, we used Ecoinvent data (see Table A1 in appendix A). The downstream processing steps (polymerisation and bottle production) are adjusted for the Dutch energy mix as provided by Ecoinvent (IEA data of 2017, extrapolated to 2020).

2.2.3. Post-consumer waste management of PET and PEF bottles

We gathered the data for the waste management systems for PET and PEF bottles via literature review and interviews. As waste treatment is assumed to occur in the Netherlands, all waste management processes described below are adjusted with the Dutch heat and electricity resources. All processes are assumed to be the same for PEF unless a difference is explicitly mentioned.

Table A.1 in appendix A summarizes all data and assumptions related to the waste management of small plastic bottles used for this study. Table A.2 in appendix A details the datasets and transport distances used for all transportation steps in the waste management steps.

2.2.3.1. Collection & transportation. We chose Swiss Ecoinvent data to assess plastic waste collection with a 21-ton lorry, see Appendix A, table A.2. The differences in transportation distances between the collection systems are considered, using the average transportation distances of waste collection systems in the Netherlands (Bergsma et al., 2011). For the deposit system, we did not consider the transport of the consumers to the bottle collection points, assuming that it is part of regular consumer movements for groceries. We also used Ecoinvent data to model the plastic waste transportation emissions between the different waste treatment facilities for each analysed waste management case, adjusted for average Dutch transport distances, see Appendix A, Table A.2).

2.2.3.2. Sorting. We assume no sorting losses for the bottles collected by the *deposit system*. They are directly sent to PET and PEF recyclers.

The plastics collected by *source separation* are sorted into fractions based on plastic type. A standard sorting process consists of the removal of impurities like metals, followed by sorting into different materials and colours, using sink-float separation, electronic sorting via laser- and near-infrared sensors, and finally manual sorting (Dinkel et al., 2018; Kägi and Dinkel, 2022). In the Netherlands, sorted fractions are distinguished based on the DKR (Der Gruener Punkt) standards adopted from Germany, with DKR 328-1 containing PET bottles (M. T. Brouwer, 2018). Brouwer et al. (2019) report that 76% of small PET bottles (≤0.5 L) are sorted for recycling. The remaining fraction is assumed to be incinerated with energy recovery (Brouwer et al., 2019). We used Swiss data on sorting PET waste into sorted PET bales (Haupt et al., 2018), and adjusted it for the Dutch energy mix, see Table A1 in appendix A.

In a *post-separation* system, ca. 19% of packaging plastics collected with MSW are sent to material recovery & sorting facilities (MRF), where 70% of the small clear PET bottles (≤0.5 L) entering these facilities are recovered (M. Brouwer et al., 2019). In a pre-treatment step, hard plastics are recovered from the remaining MSW before being sorted into specific plastic fractions. According to industrial data obtained from Dutch MRF's (Bergsma et al., 2011), the energy requirements for this pre-treatment step are approximately four times higher than those for sorting into plastic fractions. We adapted the Ecoinvent processes for the recovery and sorting from MSW with energy use including pre-treatment (see Table A.1 in appendix A). The processing losses from the post-separation system are assumed to be sent to incineration plants, along with the remaining MSW.

2.2.3.3. Mechanical recycling (MR). For MR, the sorted PET bales are opened, purified (removing labels, caps and contaminants), and shredded, before the purified PET flakes are washed and dried (Haupt et al., 2018). Recycled PET and PEF directed at bottle-grade applications also goes through a solid-state polymerisation process (SSP) to improve crystallinity.

The technology data of MR is the same in all three recycling cases A-C. We modelled the production of recycled PET bottle-grade granulate (rPET) by adapting the Ecoinvent process *"Polyethylene terephthalate, granulate, bottle-grade, recycled {CH}"* (Haupt et al., 2018), with the Dutch energy mix as provided by Ecoinvent. We derived the production of recycled amorphous PET from the same dataset by subtracting the requirements for SSP. Following our system boundaries, we excluded the waste treatment of bottle accessories (caps, labels). We assume a recycling efficiency from sorted, baled PET bottles to recycled PET pellets of 88% based on assessments of the Dutch and Danish waste management systems for PET plastic packaging (Brouwer et al., 2019; Faraca et al., 2019).

We assume the same data for PEF bottles. Some processes in PEF recycling are expected to require less and others more energy than PET recycling. Therefore, in the absence of sufficient data, we assume that the overall energy requirement of mechanical recycling would be similar between PET and PEF.

2.2.3.4. Chemical recycling (CR). Our waste management case C focuses on CR based on depolymerisation via glycolysis. This CR technology is one of the most advanced for PET (Damayanti and Wu, 2021; Raheem et al., 2019) and has also been proven to work with PEF (Gabirondo et al., 2021).

We assume that only the deposit fraction is directed to glycolysis since so far the EU regulations on the guaranteed 95% food packaging origins also apply to chemically recycled material (EFSA, 2011; KIDV, 2018). Data on glycolysis is hard to come by in publicly available literature. Shen et al. (2010) present data from a Taiwanese small-scale production plant, covering the glycolysis of PET waste to the oligomer bis-hydroxyl ethylene terephthalate (BHET), which is then filtered and repolymerised to PET. We adapted the data of Shen et al. (2010) to our system boundaries, see Table A.1 in appendix A. In the absence of data on CR of PEF, we assumed the same process requirements as for CR of PET.

2.2.3.5. Incineration with energy recovery (ER). Sorting and recycling losses from waste management cases A-C are assumed to be incinerated with energy recovery. Case D assumes full incineration of PET and PEF waste with energy recovery. In the Netherlands, incineration plants' average electricity and heat generation efficiencies are 20% and 23%, respectively (RvO, 2020). The Swiss Ecoinvent process of waste incineration was adjusted accordingly (see Table A.1 in appendix A).

The CO_2 emissions from incineration are calculated based on the carbon content of PET and PEF, which are 62.2% and 52.7%, respectively (calculated based on the molar masses). Also, the generated energy through incineration differs between PET and PEF, as PET has a lower calorific value of 22.1 MJ/kg (Ecoinvent) whereas the lower caloric value of PEF is 16.7 MJ/kg (based on experimental calorimetric calculations by Avantium).

2.2.3.6. Substituted product systems. We follow the "avoided burden approach" and provide credits to our production systems for the provision of recycled material and recovery of energy in the end-of-life (see chapter 2.2.1).

Sorted PET bottle fractions from the source separation and post-separation of MSW have a 17–24% share of non-food flasks in the Netherlands (van Thoden van Velzen et al., 2016), while 5% is the legal limit set by the European Food Safety Authority (EFSA) for food-grade recycling (EFSA, 2011). Hence, PET and PEF bottles collected in MSW and via source separation can legally not be recycled into new bottle applications used for drinks. Therefore, we assume that all recycled PET and PEF bottles from these collection methods will be "downcycled". As a consequence, the recycled polymers are assumed to substitute virgin *amorphous PET and PEF*.

We assume that only bottles collected via a deposit system are recycled for bottle-grade applications, as those conform with the EFSA regulation (van Thoden van Velzen et al., 2016). However, due to quality losses in MR (e.g., decrease in crystallinity), we apply a substitution factor of 0.9 to account for the quality loss for recycled bottle-grade PET and PEF when substituting virgin *bottle-grade PET and PEF*. This factor reflects the decrease of intrinsic viscosity when mechanically recycling blue post-consumer PET bottles compared to virgin PET (Elamri et al., 2015) and is also recommended by the *"Product Environmental Footprint"* guide (Annex C) of the European Commission (Manfredi et al., 2012). Moreover, Brouwer et al. (2020) showed that recycled PET from mono-collection systems could meet industry standards of bottles at a recycled content ratio of around 90%.

The differences in intrinsic viscosity are also present in the recycled amorphous granulate. However, we do not apply a substitution factor to amorphous granulate, assuming that a decreasing viscosity does not affect their use for amorphous applications (e.g., fibres). CR does not require a substitution factor since it achieves the same polymer quality as primary production.

For recycled PEF, we analyse the substitution of both PET and PEF. We do so as initially the PEF bottles are planned to be recycled together with PET, hence, replacing primary PET production. Once larger amounts of PEF are on the market, closed-loop recycling of PEF substituting primary PEF bottles is assumed. For open-loop recycling of PEF, we assume that 1 g of PEF will substitute 1 g of PET. In theory, PEF could substitute a higher weight of PET, due to its better material properties (see functional unit). However, in practice, we do not expect this to influence the weight of PET bottles blended with PEF as long as the PEF market shares stay as low as the 5% limit set by the EPBP (2017).

We also credit the energy generated from incinerating the bottle waste, assuming a substitution of the average Dutch electricity and heat production mix according to Ecoinvent (IEA data of 2017, extrapolated to 2020).

2.3. Biogenic emissions

Our GWP results include the uptake of CO_2 during the growth of the bio-based feedstock and the emissions of biogenic CO_2 when PEF is incinerated. We do not credit the biogenic CO_2 uptake for the substituted product system (PEF recyclates substituting primary PEF production).

We do not apply any credits for the delayed emission of carbon because of the relatively short carbon cycles of the bottle application. However, we specify the overall storage time of carbon per waste management case as complementary information (see section 2.5 and Fig. 7).

2.4. Assessing multiple recycling trips

PET has a higher recyclability than other packaging plastics, as it absorbs fewer post-consumer contaminations than, e.g., polyolefins (Pinter et al., 2021). Nevertheless, there is limited information on how often PET could be mechanical recycled without losing its critical properties, like its intrinsic viscosity, colouring and the presence of contaminants. Pinter et al. (2021) assessed eleven recycling trips for PET in a closed-loop system, showing that the quality of the mechanically recycled bottles was not negatively affected when mixed with 25% virgin PET. Brouwer et al. (2020) assessed the accumulation of contaminants over ten recycling trips, showing that recycled bottles from mono-collection systems could meet acceptable standards when mixed with around 10% of virgin PET. Lab-scale assessments by Avantium showed that recycled PEF resins could keep their mechanical properties over 12 loops at a 70% and 90% recycled content ratio (Personal communication of Roy Visser from Avantium, 11.5.2022).

We assess the cumulative net GHG emissions and material utility achieved by the waste management cases A and B for PET and PEF bottles over 10 recycling trips. After that, we assume that the remaining material will be incinerated. CR does not cause material quality deterioration and could therefore achieve more recycling trips. We chose to assess 15 recycling trips for case C to make this advantage of CR visible.

2.4.1. Cumulative net GHG-emissions

We calculate the cumulative, cradle-to-grave net GHG emissions *'CE'* for each waste management case *'i'* and bottle type *'k'* by adding the cumulative net end-of-life emissions over all end-of-life trips *'t'* to the cradle-to-gate bottle production emissions *'PE'* as follows: $CE_{k,i} = PE_k + \sum_{t=1}^{n} m_{k,i}(t-1) * b_{k,i}$. The maximum number of end-of-life trips *'n'* differs per waste management case and is ten for cases A and B, 15 for C, and one for D. The net GHG emissions are the sum of direct GHG emissions, the GHG credit received for the substituted virgin plastic granulate and the substituted energy, and the biogenic carbon uptake. The cumulative end-of-life net GHG emission is calculated based on the mass *'m'* of the PET and PEF material entering the waste treatment at each recycling trip and the net GHG emissions *'b'* of one recycling trip (in g CO_2 eq./g polymer waste). All equation variables are further explained in appendix A.

2.4.2. Material utility

A product's utility is defined by the length and the intensity of the product's use (Ellen MacArthur Foundation, 2015). We propose the concept of material utility, inspired by the product utility defined by Ellen MacArthur Foundation (2015). The *material utility* consists of the *material use intensity* and the *length of the material's use*, which we assess separately.

2.4.3. Material use intensity

This study defines the cumulative material use intensity *'MI'* as the percentage of additional material use achieved out of the initial virgin material, as follows: $MI_{i,k} = (\sum_{t=1}^{n} m_{k,i}(t-1) * r_i) / m_k(t=0)$. We calculate the weight 'm' of the cumulatively recycled polymers for a maximum of $n = 15$ recycling trips *'t'*, using the overall recycling rate *'r'* (sorting yield times recycling yield), for each waste management case *'i'*. To calculate the material intensity, we then divide this by the weight of the virgin PET or PEF bottle type *'k'*. All equation variables are further explained in appendix A.

2.5. Length of material use expressed in carbon sequestration time

The duration of a material's use is considered part of the material's utility. By assessing the overall amount and duration of carbon sequestration of the initial virgin bottle material, we combine an assessment of the use time of the PET and PEF material, expressed in bottle shelf life, with an evaluation of the total embedded CO_2 emissions. The effect of delayed emissions is not part of our LCA results but merely presented as complementary information.

We use the initial bottle weight *'m'* per bottle type *'k'* (24 g for PET, and 13 g for PEF bottle) and the carbon content *'CC'* (62.5% for PET, 52.7% for PEF) as input. To calculate the amount of sequestered carbon 'C' remaining after each recycling trip *'t'*, we multiply the overall recycling rate *'r'* of each waste management case *'i'* with the remaining carbon from the previous recycling-trip (t-1), as follows:

$C_{k,i}(t) = CC_k * m_{k,i}(t-1) * r_i$. The molecular weight ratio of carbon dioxide to carbon (44/12) is used to report the embedded CO_2 emissions.

We put the sequestered carbon over multiple recycling trips in relation to the use time of the bottle material (see Fig. 7). As a proxy for the length of use, we chose the bottle's shelf life, which is 12 weeks for a PET bottle and 20 weeks for the PEF bottle in our product system, due to the superior barrier properties of PEF.

3. Results & discussion

3.1. GWP of the bottles assuming one recycling trip

3.1.1. Comparing the waste management cases

For both PEF and PET bottles, the order of waste management cases in terms of GWP is the same, see Fig. 4: Case B performs the best, followed by Case C and Case A. The complete incineration with energy recovery (Case D) shows the largest emissions.

Incineration of bottle waste is the major contributor to the EoL emissions. Also, the benefit of substituting primary plastic production has a crucial impact on the results (see Figure B.1 in appendix B). Hence, assuming a PEF-to-PEF recycling system (green line) shows lower net GHG emissions compared to a PEF-to-PET system (red line). This is

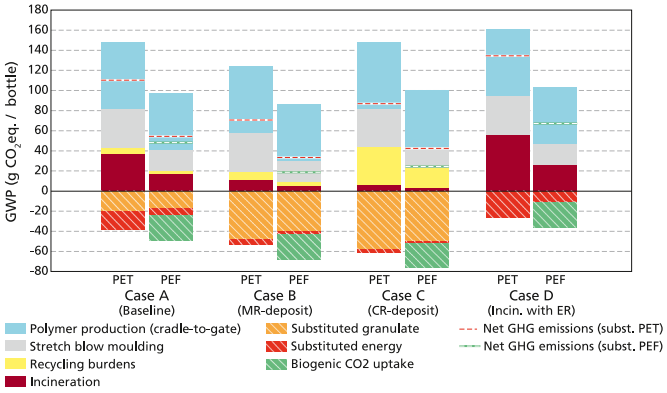

Fig. 4. The cradle-to-grave net GHG emissions for one bottle after one recycling trip.
Notes: Recycling burdens include collection, sorting, and transportation; The columns display closed-loop recycling (PEF subst. PEF, PET subst. PET); For PEF, the net GHG emissions differentiate between substituting PEF (closed-loop) and PET (open-loop).

because we assume PEF substitutes PET one to one, despite its advantages in material properties (see methods), and because 1 g of PEF granulate is more emission-intensive than 1 g of PET if we ignore the biogenic carbon uptake.

The overall recycling rate is a key driver of the emissions advantages achieved by the waste management cases B and C because it defines how many bottles are recycled to substitute primary plastics and how many are incinerated. Hence, the deposit system greatly influences the overall net emissions since it avoids sorting losses: Case C for PET bottles has 21% lower net GHG emissions than the baseline (case A) and case B even 36%. For PEF-to-PEF recycling, these savings compared to Case A could reach up to 49% and 61% respectively. The net GHG emission savings of case B compared to case D could even reach 72% for PEF when substituting PEF. Also the substitution rate plays a significant role (see discussion of sensitivities in Appendix B).

However, despite the fact that CR achieves the highest recycling rate and substitution factor, case C performs worse than case B. This is because the high energy requirements of the CR process undermine its advantages regarding the amount and the quality of recycled plastics produced. To perform better than MR after one recycling trip, CR would need to reduce its process emissions by 44% for PET bottle recycling and by 28% for PEF-to-PEF bottle recycling.

3.1.2. Comparing PET & PEF bottles

Overall, in terms of cradle-to-grave net GHG emissions, a PEF bottle performs better than a PET bottle if we assume the same waste management case and one recycling trip, see Fig. 4. However, it makes a difference if an open-loop recycling system (PEF substituting PET) or a closed-loop system (PEF substituting PEF) is in place. If PEF is assumed to substitute PEF, it receives more credits for recycling, as the impact of the displaced primary PEF granulate production is higher when ignoring the biogenic carbon uptake. For such a PEF-to-PEF system, the cradle-to-grave net GHG emissions of a PEF bottle could be 56%–74% lower than for the PET bottle after one recycling trip (depending on the waste management case). When substituting PET, the relative GHG emission savings only range from 51 to 53%, as the substituted primary PET granulate is less emission-intensive when substituted one to one. Hence, recycling PEF becomes even more beneficial in terms of net GHG savings when PEF is recycled separately in a closed-loop system. The relative emission savings of a PEF bottle are 50% when comparing the full incineration of bottles (Case D). The net GHG emission advantage of PEF bottles diminishes if PEF would be recycled less than PET or even incinerated, e.g., comparing case B for PET with case A or D for PEF. Hence, it is important to properly integrate PEF into the recycling system to maintain its advantage in net GHG emissions.

When ignoring the biogenic carbon uptake during biomass cultivation, the cradle-to-gate GHG emissions of the PEF bottle production are 26% smaller than those of the PET bottle. A key driver for this is the superior barrier qualities of PEF, which could reduce the polymer use in bottle production by almost 46%. This difference in the cradle-to-gate GHG emissions increases to 50% when we also account for the sequestration of biogenic carbon (25 g/bottle), which is taken up during biomass cultivation. Further impact categories were assessed in a separate LCA (Puente and Stratmann, 2021, 2022).

Without the biogenic carbon uptake and the credits from the avoided impacts, the contribution of EoL waste treatment in total cradle-to-grave gross GHG emissions of the PEF bottle is ranging from 11% (Case B) to 25% (Case D) (see Figure B.2 in appendix B). For the PET bottles, this range is 15–35%. The EoL phase of a PEF bottle causes fewer GHG emissions than the EoL of a PET bottle, as a PEF bottle has a lower weight and carbon content.

3.2. Cumulative net GHG emissions for multiple recycling trips

When looking at the development of the cumulative net GHG emissions over multiple recycling trips, the relative performance of the waste management cases changes significantly, see Fig. 5. While the results for case A are barely affected by assuming multiple recycling trips, the net GHG emission saving benefits of both deposit-based cases (B and C) increase with each recycling trip compared to the baseline case A and the non-recycling case D. For example, for PET bottles, the cumulative net GHG emission reductions compared to case D increase from 64 g CO_2 eq. (case B) and 48 g CO_2 eq. (case C) after one recycling trip to, respectively, 165 and 130 g CO_2 eq. after 10 (case B) and 15 recycling trips (case C).

Case B benefits less from increasing the number of recycling trips than case C. After five trips, case B already achieved 85% of total cumulative net GHG emission reductions through recycling, while case C achieves around 72% of its total cumulative reductions (in 15 trips) until the fifth recycling trip.

Moreover, the difference between cases B and C reduces after the 10th recycling trip, as we assume that the polymers cannot be mechanically recycled more than ten times, while CR enables additional recycling trips as its recyclates are equivalent to primary plastics. Case C performs better for PEF bottles, as their lower weight significantly reduces the high energy use of chemical recycling. For PEF-to-PEF recycling, case C even becomes the best option starting with the 8th recycling trip. CR retains more bottle material due to the higher recycling and substitution rate and thus benefits most from the higher carbon benefit achieved by displacing primary PEF production. However, case B stays the cumulatively best performing option for the entire number of analysed recycling trips for both systems substituting PET. Moreover, also for PEF-to-PEF recycling it stays superior until the 7th recycling trip.

3.3. Material utility

Fig. 6 expresses the material use intensity as the % of additional material use achieved from the initial, virgin plastic material over 10–15 recycling trips. The baseline case A only achieves 48% of additional material use and only for lower-grade amorphous applications. The deposit-based cases achieve a significantly higher material intensity with over 300% (case B) and almost 500% (case C). Moreover, their outputs are to a large extent useable in new bottle applications. The cumulatively produced and recycled bottle-grade material would equal a total of 3.8 and 5.7 bottles respectively (incl. the initial virgin bottle).

CR reaches the highest material use intensity, due to its higher recycling efficiency and because it allows for more recycling trips than MR, which reaches its recyclability limit earlier due to quality losses. This advantage in the material use intensity of CR would even be higher if we would have analysed more than 15 recycling trips.

Next to the material use intensity, the overall length of the material use is another part of a material's utility. By showing the carbon sequestration over time per bottle and waste management case, Fig. 7 combines an assessment of the overall use time of the bottle material with the corresponding embedded carbon emissions. Also here CR (case C) clearly outperforms the other analysed waste management cases as it sequesters more of the material (see material use intensity) over a longer time.

The PEF bottle could theoretically achieve a longer carbon sequestration time over multiple recycling trips than a PET bottle (a maximum of 320 weeks compared to 192 weeks for a PET bottle for case C over 15 recycling trips), due to the longer shelf-life a PEF bottle provides. Moreover, the PEF bottle acts as a short-term carbon sink due to its biogenic carbon content. However, this effect is negligible because of the short lifetime of bottles. Only in long-term applications such as in the building & construction sector, the effect of bio-based carbon sequestration could be considerable (de Oliveira et al., 2020).

Overall, CR (case C) is superior to the other waste management cases in terms of material utility. However, the high energy use of CR largely offsets its advantages in material utility when assessing the GWP.

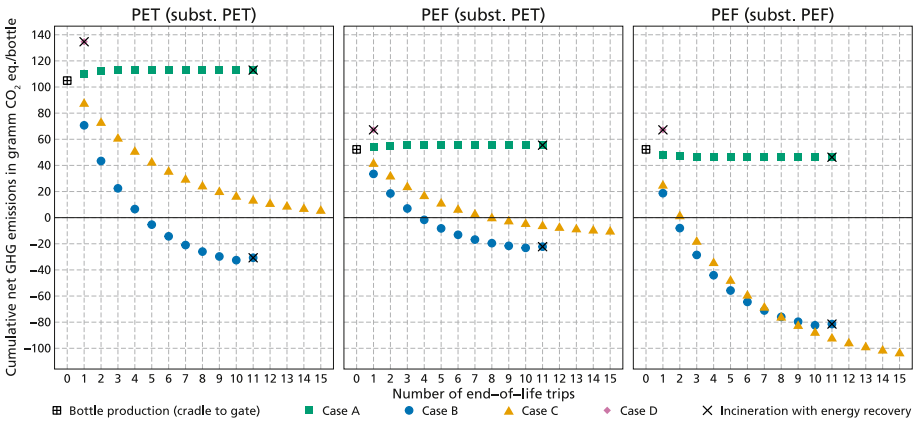

Fig. 5. Cumulative cradle to grave net GHG emissions over 10 recycling trips for mechanical recycling (for cases A and B) and 15 trips for chemical recycling (case C).
Notes: The 11th EoL trip for case A and B represents the incineration of the remaining material with energy recovery; The negative emissions achieved in Cases B and C are the result of allocating the future benefits of the recycled bottle material only to the initial virgin bottle. This representation excludes additional virgin material needed to produce the subsequent bottle after each recycling trip. This display was chosen to illustrate the potential cumulative benefit of keeping the original material in use.

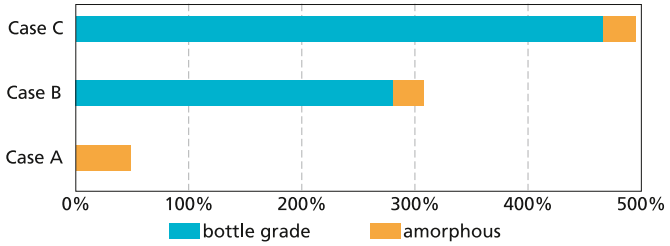

Fig. 6. Material use intensity achieved by the three recycling cases over 10 (MR) and 15 (CR) recycling trips.
Notes: expressed as the percentage of additional material use achieved out of the initial virgin material over multiple recycling trips, differentiating between bottle and amorphous applications; It is the same for PET and PEF bottles since we assume the same waste management cases.

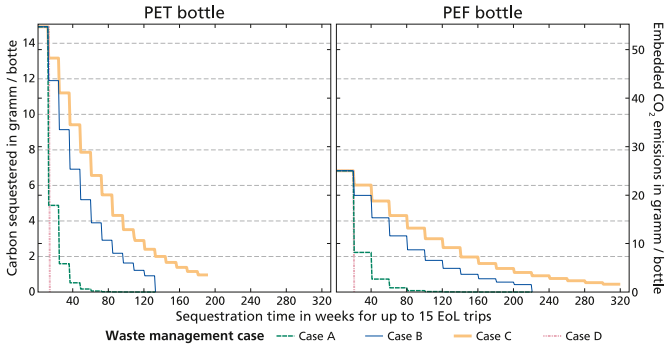

Fig. 7. Carbon sequestration over time per bottle type and waste management case, showing how long the carbon of the initial bottle is sequestered and how much of it (in new bottles or other applications).

3.4. Discussion & limitations

3.4.1. Comparing PEF and PET bottles

Assuming the same waste management cases for PEF and PET bottles, the cradle-to-grave net GHG emissions of a 250 mL PEF bottle are 50–74% lower than the ones of an equivalent PET bottle after one end-of-life trip, depending on the waste management case. The combined effect of biomass use and material savings (46%) due to enhanced barrier properties is the key reason behind the 50% lower cradle-to-gate GHG emissions of the 250 mL bio-based PEF bottle compared to its PET equivalent. The lower weight and carbon content of the PEF bottle reduces its emissions at the EoL, as less process energy is required and less carbon emitted during incineration. Moreover, a PEF bottle offers a longer shelf-life, which could increase its material utility.

However, if PEF would be recycled less than PET or even incinerated, the cumulative net GHG emission advantage of PEF compared to PET diminishes or could even turn in favour of PET after multiple recycling trips (see Figs. 4 and 5). Only if we achieve the same recycling performance for PEF as for PET do we maintain the full advantage of PEF compared to PET in terms of GHG emissions.

3.4.2. The importance of the bottles' end-of-life and the accounting method

Our results showed the importance of the EoL in the overall cradle-to-grave gross emissions of PET and PEF bottles. Depending on the case, the EoL has a share of 15–35% in gross cradle-to-grave emissions of PET bottles and a share of 11–25% for PEF bottles. For the recycling cases (A-C), these shares increase further the more recycling trips we assume. However, also the benefits of recycling could increase the more recycling trips are achieved. This could result in cumulative net negative GHG emissions after 2–8 recycling trips for some cases when accounting for the replaced primary polymer production (see Fig. 5). These negative values should not be interpreted as net physical carbon removals, as they only illustrate the climate impact of avoiding primary material production. Moreover, these net negative emissions are the result of allocating all future benefits of the recycled bottle material to the initial, virgin bottle. This representation excludes additional virgin material needed to produce the subsequent bottle after each recycling trip. This display was chosen to illustrate the potential cumulative benefit of keeping the original material in use.

All analysed recycling cases are clearly improving the net GHG emissions when compared to the full incineration of bottle waste. Recycled PEF is expected to replace primary PET production in the initial years. With higher PEF market shares, PEF-to-PEF recycling could be established, further increasing the net GHG benefit of PEF recycling.

Changing the analysis from one recycling trip to multiple trips changes the relative performance of the waste management cases, see Fig. 5. The cumulative net emissions savings of the deposit-based cases B and C increase with each recycling trip compared to case D (incineration with energy recovery), while case A (baseline) barely changes. Especially CR benefits from increasing the number of recycling trips, while MR already achieves 85% of its cumulative net GHG emissions savings after 5 trips (case B) or even close to 100% for case A. Furthermore, CR is catching up with deposit-based MR in terms of cumulative net GHG emissions after ten recycling trips. However, for PET recycling and PEF recycling to substitute PET, deposit-based MR (case B) remains the option with lowest net GHG emissions. Only for PEF-to-PEF recycling do we see a preference for CR (case C) compared to MR (case B), but only after 8 recycling trips. Increasing the number of recycling trips also clearly improves the material utility achieved by the deposit-based waste management cases B and C, with CR (case C) providing the best result.

3.4.3. Potential trade-offs between climate change mitigation and circular economy goals

Our results reveal a trade-off between circular economy goals (expressed in material utility), which favour CR (case C), and climate change mitigation, which favours MR (case B). The high recycling yield and substitution factor of CR (case C) make it the best option in terms of material utility, by keeping more of the plastics in the technosphere for a longer period of time. However, the high energy requirements of CR hinder its performance when looking at the GWP impact. Deposit-based MR (case B) shows the lowest net GHG emissions. Combining MR and CR could be a promising synergy between the material utility and the GWP. CR could upgrade polymers that have been degraded through MR. This would allow for further recycling trips and thus contribute to keeping the polymers longer in use and avoiding primary plastic production.

However, increasing the material utility or circularity of a product should not be a goal by itself but rather a mean to reduce negative environmental impacts (Geyer et al., 2016). Moreover, a circular economy can provide an additional value by contributing to the sovereignty of countries, i.e., making them less dependent on new resources, through keeping materials in use longer and recycling them. This became increasingly important in the light of recent resource supply issues due to conflicts and the COVID-19 pandemic.

3.4.4. Key limitations & recommendations for further research

While our work provides valuable insights into the net GHG emissions and material utility of different waste management cases for small PET and PEF bottles over multiple recycling trips, our results have to be used with caution, due to data limitations, modelling choices (e.g., allocation), and the specificity of the analysed product systems. A key limitation is the mismatch of technology data, leading to flawed comparisons. While there is industrial data for the highly optimised PET production and MR, this is still missing for PEF production and CR. Updated assessments would be worthwhile, once commercial data on PEF production and CR in the Netherlands is available, the impact of the recently introduced deposit systems in the Netherlands is known, and once there is more evidence on the behaviour of PEF in mechanical and chemical recycling systems. Moreover, the quality implications of MR need to be better understood. We integrated the impact on polymer quality via a substituion factor (see methods). Higher quality losses in MR would reduce the benefit of MR and the number of potential recycling trips. The sensitivities of these and further limitations are discussed in appendix B.

We focused on a comparison of GHG emissions and material utility and would recommend that also future LCAs consider complementary circularity indicators. However, our work excluded other environmental impact categories, which were considered for the bottle production in a previous assessment (Puente and Stratmann, 2021, 2022). Although, also for the waste management additional environmental impacts should be considered to identify potential further trade-offs and synergies.

The impact of future energy supply on the analysed systems should be better understood by conducting prospective environmental impact assessments, i.e., linking the LCA to long-term scenario inputs such as from integrated assessment models (Sacchi et al., 2022). Such prospective assessments should also consider the use of second generation lignocellulosic feedstocks for PEF production. Moreover, we would recommend the analysis of additional applications of PEF and the respective waste management cases, as the benefits of PEF, e.g., high barrier properties, are less relevant in other applications.

4. Conclusions & recommendations

The combined results of Puente and Stratmann (2021, 2022) and this article showed that switching from PET to PEF is a robust strategy to reduce the GHG emissions of small plastic bottles, and that LCAs are a useful tool to guide an informed transition from fossil to renewable resources. The LCA of nova institute comparing PEF and PET bottle types already showed that application matters when comparing different plastics: PEF performs best in applications that require high barrier properties and light weight. This highlights the importance of considering the application-specific benefits in the use phase when comparing novel with conventional plastics.

Our analysis concludes that also EoL and the amount of recycling trips matter. Only a *circular* bioeconomy, i.e., integrating PEF bottles into the recycling systems, ensures that the GWP advantage of PEF bottles can be sustained. It is important that new plastic materials maintain their quality also over multiple recycling trips; otherwise, their overall cradle-to-grave performance compared to existing materials worsens. Hence, it is important to consider the EoL already during product design, and to conduct full cradle-to-grave assessments over multiple recycling trips when assessing a product.

We analysed four waste management scenarios for PET and PEF bottles over 10–15 recycling trips, which clearly showed the superiority of deposit-based recycling systems over the baseline based on the 2017 Dutch mix of post-separation and source separation. Therefore, policy makers should extend the use of deposit systems.

Lastly, policy goals matter: When aiming for material utility alone, e.g., to limit finite resource depletion or to improve the sovereignty, i.e., resource independence, of a country, CR prove to be the superior option in our study. If the goal is climate change mitigation, our results favour deposit-based MR in most cases. These trade-offs between MR and CR could be overcome by combining the two waste treatment options to achieve the goals of both the circular economy and climate change mitigation alike. Moreover, the further development of CR technologies should be fostered to increase their technology readiness level, and reduce their costs, energy consumption, and climate impact.

CRediT authorship contribution statement

Paul Stegmann: Writing – original draft, Methodology, Software, Investigation, Visualization, Validation, Conceptualization. **Ties Gerritse:** Methodology, Software, Investigation, Writing – original draft. **Li Shen:** Methodology, Writing – review & editing, Supervision, Conceptualization. **Marc Londo:** Writing – review & editing, Supervision, Conceptualization, Funding acquisition. **Ángel Puente:** Writing – review & editing, Investigation, Resources. **Martin Junginger:** Writing – review & editing, Supervision, Conceptualization, Funding acquisition, Project administration.

Declaration of competing interest

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

Data availability

All data sources are included in the manuscript (Method section and Appendix A).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.136426.

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