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An environmental and economic assessment of bioplastic from urban biowaste. The example of polyhydroxyalkanoate

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HIGHLIGHTS

- Polyhydroxyalkanoates production from food waste and sewage sludge was investigated.
- Life cycle assessment (LCA) and societal life cycle costing (LCC) were applied.
- Polyhydroxyalkanoates from urban biowaste outperform polyurethane.
- Possible improvements in the production process chain are identified.
- Local conditions are critical for the polyhydroxyalkanoate performance.

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ABSTRACT

Bio-based and biodegradable plastics promise considerable reductions in our dependency on fossil fuels and in the environmental impacts of plastic waste. This study quantifies the environmental and economic consequences of diverting municipal food waste and wastewater sewage sludge from traditional management to the biorefinery-based production of polyhydroxyalkanoates (PHA) in five geographical regions. The results show that PHA can outperform fossil polyurethane and PHA from first-generation biomass (sugarcane and maize) with respect to both environmental impacts and societal costs (four times lower impacts and eight times lower costs than polyurethane). To outperform other fossil polymers like low-density polyethylene (LDPE), biorefinery performance should be improved further by more efficient utilization of sodium hypochlorite during PHA extraction, minimization of methane leakage in biogas facilities, upgrading of biogas to biomethane, and more effective handling of the liquid fraction from digestate dewatering.

1. Introduction

Plastic is a versatile, light, and cost-efficient material. Over recent years, several negative aspects of this widespread plastic use have been highlighted (e.g. littering, marine plastics, and fossil fuel consumption), pushing consumers and manufacturers to look for alternative options, in particular bioplastics. In the public perception, bioplastics are produced from plants or other organic materials (solving the problem of fossil fuel dependency), are biodegradable in any type of environment (solving terrestrial and marine pollution), and simultaneously decouple environmental impacts and economic growth. Yet, the majority of biodegradable bioplastics are manufactured from fossil fuels and only biodegrade under specific and favorable conditions (e.g. industrial composting) (Alaerts et al., 2018), and the majority of bio-based plastics are not biodegradable. Only a few options exist that are both bio-based and biodegradable, namely, polylactic acid, polyhydroxyalkanoates (PHA), and starch blends (European Bioplastics, 2019a). Among them, PHA is expected to see one of the largest growth rates in the coming years (Aeschelmann and Carus, 2017; European Bioplastics, 2019b). PHAs are a broad family of bio-based, biodegradable polyesters that are accumulated by specific bacteria in conditions of excessive organic carbon and scarce inorganic nutrients (Nikodinovic-Runic et al., 2013). PHAs are conventionally produced from first-generation biomass (Tsang et al., 2019), can be compounded and utilized in several applications depending on the chain length of the volatile fatty acids (Laycock et al., 2014), and are mainly used in flexible and rigid packaging (European

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Bioplastics, 2019c). The main challenge in PHA manufacturing is production costs, which are significantly higher than for other biopolymers, especially due to the raw materials used as carbon substrates and the chemicals used during PHA extraction (marketsandmarkets.com, 2019). To reduce some of these costs, secondary residual feedstocks can be utilized (Yadav et al., 2020) and this would in turn mitigate the competition with food for land. Examples of PHA from biogas, wastewater, and sewage sludge have been demonstrated (Bengtsson et al., 2017a, 2017b; Fernández-Dacosta et al., 2015; Morgan-Sagastume et al., 2015).

Life cycle assessment (LCA) and life cycle costing (LCC) are common tools employed to evaluate the environmental and economic performance of alternative systems. Published LCAs have provided rather conflicting results when comparing bioplastics to their fossil counterparts. Generally, bio-based bioplastics are reported to have lower environmental impacts with respect to climate change and fossil fuel dependency, but they exhibit higher impacts in other categories such as eutrophication and toxicity (Chen et al., 2016; Tabone et al., 2010; Yates and Barlow, 2013). Nonetheless, even climate benefits may be outbalanced when including climate effects from land-use changes caused by first-generation biomass cultivation (Piemonte and Gironi, 2010). Almost all existing LCAs addressing PHA have involved first-generation biomass as feedstock (Harding et al., 2007), with contradictory results because of varying methodologies and case-specific assumptions (Heimersson et al., 2014). The few environmental assessments involving second-generation biomass feedstock mainly focused on municipal (Heimersson et al., 2014) and industrial (Fernández-Dacosta et al., 2015) wastewater. Previous LCA and LCC studies on bioplastics and PHA production have been inconclusive, primarily because: i) The methodologies were inconsistent, due to inappropriate inventory data (Leong et al., 2017; Tabone et al., 2010), ii) several approaches for the accounting of multi-functionality were mixed in the same study (e.g. Chen et al., 2016; van der Harst et al., 2014) or not clearly stated (Akiyama et al., 2003; Harding et al., 2007), and iii) geographic and regional framework conditions were not systematically evaluated. None of the aforementioned studies considered the effects of diverting biowaste resources from current management (hereafter called 'counter-factual') induced by the production of bioplastics.

The aim of this study is to quantify the environmental and economic consequences of diverting municipal food waste and wastewater sewage sludge from their traditional management (i.e. the counter-factual) to a biorefinery producing PHA. Identified research gaps are addressed by three specific objectives: i) Provide a full life cycle inventory and mass balance of PHA production in five different geographical regions in Europe; ii) compare the production of PHA from residual bioresources with competing fossil plastic alternatives and PHA from first-generation biomass, and iii) identify environmental hotspots in the system.

2. Material and methods

2.1. Goal, scope, and system boundaries

The functional unit was the production of 1 kg of polymer for use in film blowing and the treatment of the waste generated from such consumption in 5 geographical clusters (the metropolitan areas of Barcelona, Copenhagen, Lisbon, South Wales, and the province of Trento). Fig. 1 shows the system boundaries for the three systems fulfilling the functional unit: a) PHA from urban biowaste; b) fossil low-density polyethylene (LDPE) and polyurethane (PUR); c) PHA from first-generation biomass. The choice of fossil polymers was based on the potential applications of PHA from urban biowaste (Fantinel, 2019) and their inventory was retrieved from Ecoinvent 3.6 - consequential. Three PHAs from first-generation biomass were considered, two from sucrose in sugarcane (Harding et al., 2007; Kookos et al., 2019) and one from glucose in maize (Gerngross, 1999). Indirect land-use changes from producing sugarcane and maize were added following the method described in Tonini et al. (2016). The system boundaries of the PHA from urban biowaste were divided into five parts (color-coded in Fig. 1, a): *i*) the biorefinery plant (in blue), including the pre-treatment of the food waste, fermentation, PHA accumulation, and extraction, *ii*) the management of biorefinery residues (in orange), *iii*) the waste management of the PHA (in red), *iv*) the food waste counter-factual (yellow-dotted), *v*) the sewage sludge counter-factual (green-dotted). Inputs into the biorefinery (food waste and sewage sludge) carried the impacts of the avoided counter-factual management: the counter-factuals (*iv* and *v* in Fig. 1) were subtracted from the direct impacts (*i*, *ii* and *iii* in Fig. 1) and all the processes that were a net burden in the counter-factuals (e.g. methane emissions from landfilling) became an avoided burden in the new system, whilst all the processes that were a net saving (e.g. energy recovered in an incinerator) became an avoided saving. Finally, the biorefinery residue management (*ii*) was always equal to the sewage sludge counter-factual (*v*), due to the stricter legislation for sewage sludge management.

The biorefinery design (*i* in Fig. 1) was based on the Treviso pilot plant (Moretto et al., 2020), receiving sewage sludge and source-separated food waste. Consumption of energy and ancillary material was upscaled by sketching an industrial-scale facility and deriving consumptions/costs from process-engineering calculations. Before entering the biorefinery, food waste undergoes a pre-treatment similar to wet anaerobic digestion, i.e. impurities are removed, waste is shredded and water is added to reach the desired total solids (TS) content. To have a steady process, the volatile solids (VS) from sewage sludge must be a maximum of 25% of the total VS input (Moretto et al., 2020). The mixed substrate enters acid fermentation, where volatile fatty acids are accumulated, and after passing through a centrifuge and a membrane, the liquid part is sent to two aerobic steps, where PHA is accumulated in the cells of microorganisms with a fast-famine regime. After a second centrifuge, the PHA is extracted from the biomass with the use of sodium hypochlorite, sodium hydroxide, and other inorganic oxidizing agents, dried and readied for manufacturing into new products. Residues from the centrifuges and membrane represent the biorefinery residues.

2.2. Impact assessment

The production of 1 kg of PHA from urban biowaste in the 5 geographical clusters was modeled for 6 alternatives describing different food waste and sewage sludge counter-factuals (described in section 2.3) and 8 framework scenarios (described in section 2.4), totalizing 240 scenarios (Fig. 2). Mass balance, environmental and economic impacts were calculated for each of the 240 scenarios, for the competitive fossil plastic, and for PHA from first-generation biomass, and their results are described in sections 3.1, 3.2 and 3.3, respectively.

First, urban waste generation, current biowaste treatment, and mass balances were defined for the 5 individual clusters, allowing for estimating potential PHA production according to specific local conditions.

The environmental assessment was performed with a consequential LCA (Ekvall and Weidema, 2004; Weidema, 2003), due to the interest in quantifying the effect of diverting urban biowaste to the new biorefinery. Marginal technologies were calculated for processes marginally affected (Ekvall and Weidema, 2004) (e.g. electricity consumption), and multi-activity processes were solved with system expansion (e.g. electricity produced from an incinerator avoided marginal electricity; nutrients provided from the use-on-land of compost or digestate avoided the production and use of marginal mineral fertilizers). The life cycle impact assessment method included the 16 impact categories recommended in the environmental footprint EF3.0 (JRC-EC, 2020), thereby providing normalization and weighting factors to calculate one single weighted indicator. Economic sustainability was quantified with a societal LCC (Martinez-Sanchez et al., 2015; Swarr et al., 2011), defined as the sum of shadow prices and externalities. Shadow prices were obtained by multiplying the budget costs (i.e. financial costs without profit

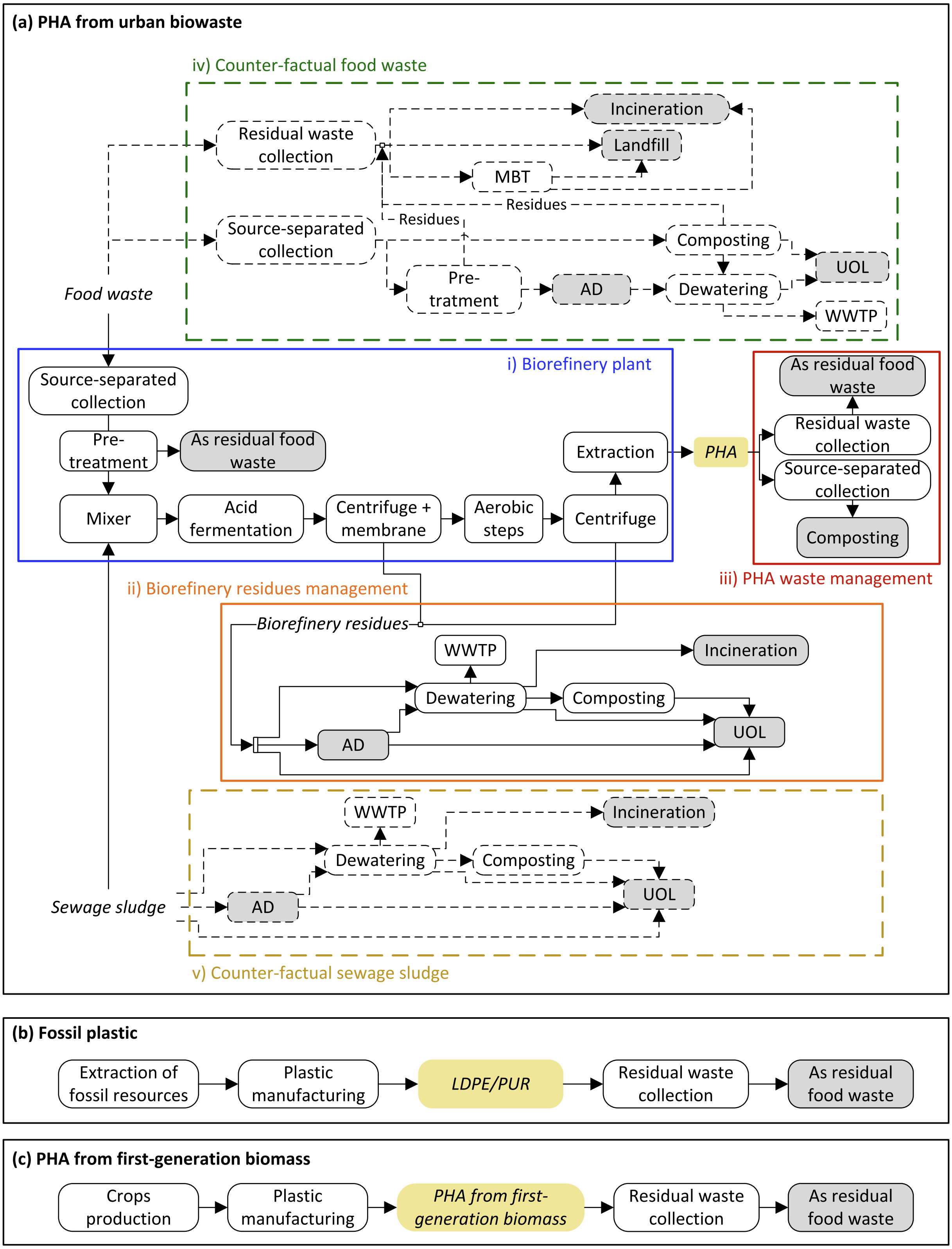

Fig. 1. System boundaries of polyhydroxyalkanoates (PHA) produced from urban biowaste (a), compared to competitive fossil plastics (b), and PHA from first-generation biomass (c). The PHA from urban biowaste (a) was divided into the plant itself (*i* in blue), the treatment of the biorefinery residues (*ii* in orange), and the incineration/landfilling of the PHA (*iii* in red). The dotted lines indicate the food waste counter-factual (*iv* in green) and sewage sludge counter-factual (*v* in yellow), which were modeled by subtracting them from the PHA production process. The treatment of the biorefinery residues (*ii*) was always equal to the sewage sludge counter-factual (*iv*). Filled processes indicate the presence of by-products (energy or fertilizers) that avoid the production of marginal energy or marginal mineral fertilizers. Transport and capital goods are not explicit in the figure but were included in the modeling. LDPE: low-density polyethylene; PUR: polyurethane; MBT: mechanical biological treatment; AD: anaerobic digestion plant; UOL: use-on-land. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Functional unit: "1 kg of polymer for use in film blowing and the treatment of the waste generated from such consumption in 5 geographical clusters"

a) PHA from urban biowaste

5 geographical clusters:
- Barcelona,
- Copenhagen
- Lisbon
- South Wales
- Trento

6 alternatives

Name	Food waste counter-factual	Sewage sludge counter-factual and biorefinery residues management
I) "FW_sep."	Current management of source-separated food waste*	Current management of sewage sludge*
II) "FW_residual"	Current management of residual waste*	as in "FW_sep."
III) "FW_AD"	AD followed by use-on-land**	as in "FW_sep."
IV) "FW_Inc"	Incineration	as in "FW_sep."
V) "SS_AD_Inc"	as in "FW_sep."	AD followed by dewatering and incineration
VI) "SS_AD_UOL"	as in "FW_sep."	AD followed by use-on-land**

8 framework scenarios:
- Marginal electricity modelled as average electricity in 2020
- Marginal space heat modelled as average space heat in 2020
- Biogas upgrading to biomethane (both in the biorefinery residues management and in the counter-factual)
- Biogas upgrading to biomethane (only in the biorefinery residues management)
- Reduction of the CH_4 leakage in the AD*** (both in the biorefinery residues management and in the counter-factual)
- Reduction of the CH_4 leakage in the AD*** (only in the biorefinery residues)
- Avoidance (or complete recirculation) of sodium hypochlorite in the PHA extraction
- PHA composting instead of treatment with the residual waste

b) Fossil plastic

- Low-density polyurethane (LDPE)
- Polyurethane (PUR)

c) PHA from first-generation biomass

- PHA from sucrose in sugarcane based on Harding et al. (2007)
- PHA from sucrose in sugarcane based on Kookos et al. (2019)
- PHA from glucose in maize based on Gerngross (1999)

Fig. 2. Summary of the systems fulfilling the same functional unit compared in the study. *management depends on the cluster. **the presence of dewatering and composting depends on the cluster. ***AD plant includes the AD digester, digestion tank, and biogas utilization units. FW: municipal food waste; AD: anaerobic digestion.

share, taxes, subsidies, and fees) by 1.325 according to the Ministry of Danish Finance (2017) and Martinez-Sanchez et al. (2015). Externalities represented the external costs society has to bear, and we monetized emissions into the environment based on CE Delft's report (De Bruyn et al., 2018). All costs were corrected for inflation to EUR2019 (herein called simply 'EUR'), using the Harmonized Indices of Consumer Prices (Eurostat, 2018).

The LCA and the LCC shared the same functional unit and system boundaries (illustrated in Fig. 1), meaning that, also in the LCC, the costs of the counter-factual were subtracted from the costs of the biorefinery. Both LCA and LCC were performed using the EASETECH software package (Clavreul et al., 2014; DTU Environment, 2020).

2.3. Alternatives

The 6 modeled alternatives represent different systems producing PHA from urban biowaste in the 5 clusters (see Fig. 2). All alternatives had in common the same biorefinery plant (*i* in Fig. 1) but differed in the biorefinery residues management (*ii* in Fig. 1), in the treatment of the waste PHA (*iii* in Fig. 1), and in the counter-factual of food waste and sewage sludge (*iv* and *v* in Fig. 1).

- Alternative I ("FW_sep."): The biorefinery residues and the sewage sludge counter-factual were modeled as the treatment of sewage sludge in 2018 in each cluster: In Barcelona, Lisbon, South Wales, and Trento, the sewage sludge was dewatered (with or without anaerobic digestion, and with or without composting) and used on land. In Copenhagen, 100% of the sewage sludge was anaerobically digested and incinerated. The food waste counter-factual involved the treatment of the source-separated food waste in 2018 in each cluster: Anaerobic digestion followed by direct use-on-land in Copenhagen; composting followed by use-on-land in South Wales; anaerobic digestion followed by composting and use-on-land in Barcelona, Lisbon, and Trento (in Trento, anaerobic digestion represented the treatment of 75% of the source-separated food waste, while the remainder was sent to composting). The waste PHA was treated as the residual waste in each cluster.

- Alternative II ("FW_residual"): The biorefinery residue management, the sewage sludge counter-factual, and the PHA waste treatment were the same as in alternative I. The food waste counter-factual was modeled as the current treatment of the residual waste: Mechanical biological treatment (before incineration or landfilling) and incineration in Barcelona; only incineration in Copenhagen and South Wales; mechanical biological treatment (before landfilling) and direct landfilling in Lisbon and Trento.
- Alternative III ("FW_AD"): The biorefinery residues management, the sewage sludge counter-factual, and the PHA waste treatment were the same as in alternative I. The food waste counter-factual was 100% anaerobic digestion, whereby the biogas was combusted in a combined heat and power engine that produced electricity to be sold to the grid, and heat to be used internally. The digestate was either spread directly on land (in Copenhagen and South Wales) or was previously dewatered and composted (in Barcelona, Lisbon, and Trento).
- Alternative IV ("FW_Inc"): The biorefinery residues management and the sewage sludge counter-factual were the same as in alternative I. The food waste counter-factual and the PHA waste treatment was incineration with energy recovery.
- Alternative V ("SS_AD_Inc"): The biorefinery residues and the sewage sludge counter-factual were sent to anaerobic digestion followed by dewatering and incineration. The food waste counter-factual and the PHA waste treatment were the same as in alternative I.
- Alternative VI ("SS_AD_UOL"): The biorefinery residues and the sewage sludge counter-factual were sent to anaerobic digestion followed by use-on-land. After anaerobic digestion, digestate was dewatered and composted in Barcelona, Lisbon, and Trento, while it was directly spread on land in Copenhagen and South Wales. The food waste counter-factual and the PHA waste treatment were the same as in alternative I.

2.4. Uncertainty and scenario analysis

All 440 LCA parameters and 70 LCC parameters in the model were parametrized with an uncertainty distribution based on an extensive literature review. Uncertainty distributions were propagated with a Monte Carlo analysis having 1,000 iterations; the average and 95% confidence intervals of the obtained distributions are shown in the results section. Parametrization of the models helped identify the parameters that contributed the most to overall uncertainty, by using a global sensitivity analysis (Bisinella et al., 2016).

Six framework scenarios were modeled to test the robustness of the results in different conditions (see Fig. 2): a) consumed and avoided electricity was modeled with the national average composition in 2020 instead of the marginal value; b) avoided space heating was modeled with the national average composition in 2020 instead of the marginal value; c) all biogas was upgraded to a purity high enough to be fed into the natural gas grid, thus avoiding the production and combustion of natural gas (in both the biorefinery residues management and the counter-factuals); d) the biogas was upgraded only in the biorefinery residues management; e) methane leakages in the anaerobic digesters, the digestion tank, and the biogas combustion engine were minimized from 6% of the generated CH_4 to 2% (in both the biorefinery residues management and the counter-factuals); g) methane leakage was minimized only in the biorefinery residues management. Furthermore, we tested the impact of avoiding (or completely recirculating) sodium hypochlorite in the extraction step (g) and composting PHA instead of treating it in the residual waste (h).

3. Results and discussion

3.1. Mass balance

The mass balance of the analyzed system (summarized in Table 1) provides important information for interpreting the following LCA and LCC results. First, since the efficiency of the biorefinery was defined as kg PHA/ kg VS entering the aerobic steps, the quantities of food waste and sewage sludge needed to produce 1 kg of PHA depended on the content of VS in the two fractions. Generally, the production of 1 kg of PHA in the proposed biorefinery requires the source-separation of 41 to 50 kg of food waste (Trento and Barcelona, respectively) and between 15 and 78 kg TS of sewage sludge (Trento and Barcelona, respectively).

Table 1
kg of municipal food waste and sewage sludge needed to produce 1 kg of polyhydroxyalkanoates (PHA) in each cluster; total food waste and sewage sludge generated in each cluster; and maximum amount of PHA calculated assuming 100% food waste source-separation. All the values are expressed in wet weight, apart for sewage sludge, which is expressed in total solid (TS).

	For 1 kg PHA		Tot generated			
	Food waste kg	Sewage sludge kg TS	Food waste Mt	Sewage sludge Mt TS	Max PHA t	kg/ inhabit
Barcelona	50	4.8	467	78	8,061	3.0
Copenhagen	44	4.6	208	41	4,194	3.9
Lisbon	44	4.5	562	58	11,257	4.8
South Wales	50	4.4	162	43	2,914	1.8
Trento	41	4.1	52	15	1,067	2.4

Second, the technical limitations of the studied pilot plant (i.e. a maximum of 25% of the input VS could come from sewage sludge) made the food waste the limiting factor. If 100% of the food waste was source-separated and sent to the studied biorefinery, 58% (Barcelona), 52% (Copenhagen), 98% (Lisbon), 33% (South Wales), and 36% (Trento) of the generated sewage sludge could be used for PHA production. This suggests that while the biorefinery could be a relevant technology for food waste treatment, a large share of the sewage sludge would still need to be treated elsewhere, except for the metropolitan area of Lisbon.

Third, the maximum amount of PHA that can be produced in each cluster was calculated for 100% source-separated food waste and corresponded to: 3 kg PHA/inhabitant in Barcelona (total 9,673 t), 3.9 kg PHA/inhabitant in Copenhagen (total 5,033 t), 4.8 kg PHA/inhabitant in Lisbon (total 13,508 t), 1.8 kg PHA/inhabitant in South Wales (total 3,496 t), and 2.4 kg PHA/inhabitant in Trento (total 1,281 t). This variability demonstrates how important it is to quantify correctly available resources, chemical characteristics, and potential limiting factors when assessing the potential benefits induced by a novel material.

Fourth, rates for food waste source-separation in 2018 estimate the magnitude of the potential diversion of food waste from residual waste, namely, 33% in Barcelona, 20% in Copenhagen, 5% in Lisbon (where only a very small quantity of food waste from restaurants is collected), 43% in South Wales, and 84% in Trento. This indicates that there is little potential for increasing collection rates in Trento, while larger improvements are possible (and foreseeable) for the other clusters, especially as a result of the new Directive 2018/851 mandating that at least 55% of municipal solid waste should be prepared for reuse and recycling by 2025 (EC, 2018). However, to date, no information on a plan to implement the source-separation in Lisbon has been found.

3.2. Environmental impacts

Environmental impacts were calculated by normalizing and weighting the midpoint impact categories recommended by the EF3.0 (JRC-EC, 2020). However, two sets of weighting factors are provided, including or excluding the three impact categories describing human and water toxicity, namely, *Freshwater eco-toxicity* and *Human toxicity, carcinogenic*, and *Human toxicity, non-carcinogenic*. While showing, in absolute values, the largest potential impacts, *Eco-toxicity* and *Human toxicity, non-carcinogenic* were not included in the calculated single indicator, as these categories are associated with significant uncertainty and are not seen as adequately robust "to be included in external communications or in a weighted result" (Sala et al., 2018). Furthermore, the current characterization factors for these impact categories do not consider background concentrations or complex speciation of heavy metals that naturally occur in ecosystems (Saouter et al., 2020), and both background concentration and metal speciation may significantly affect impacts derived from use-on-land of any organic matter including heavy metals in the form of digestate (Boldrin, 2009). For this reason, we discussed these two impact categories separately in section 3.2.4, but we excluded them from the single weighted indicator presented in section 3.2.5. When excluding toxicity, *Depletion of fossil* and *abiotic resources*, *Climate change*, *Freshwater,* and *Marine eutrophication* are the most relevant impact categories in the single weighted indicator, and for this reason, they are specifically discussed in the following sections.

When the net results (black dots) of the Figs. 3 and 4 are above the

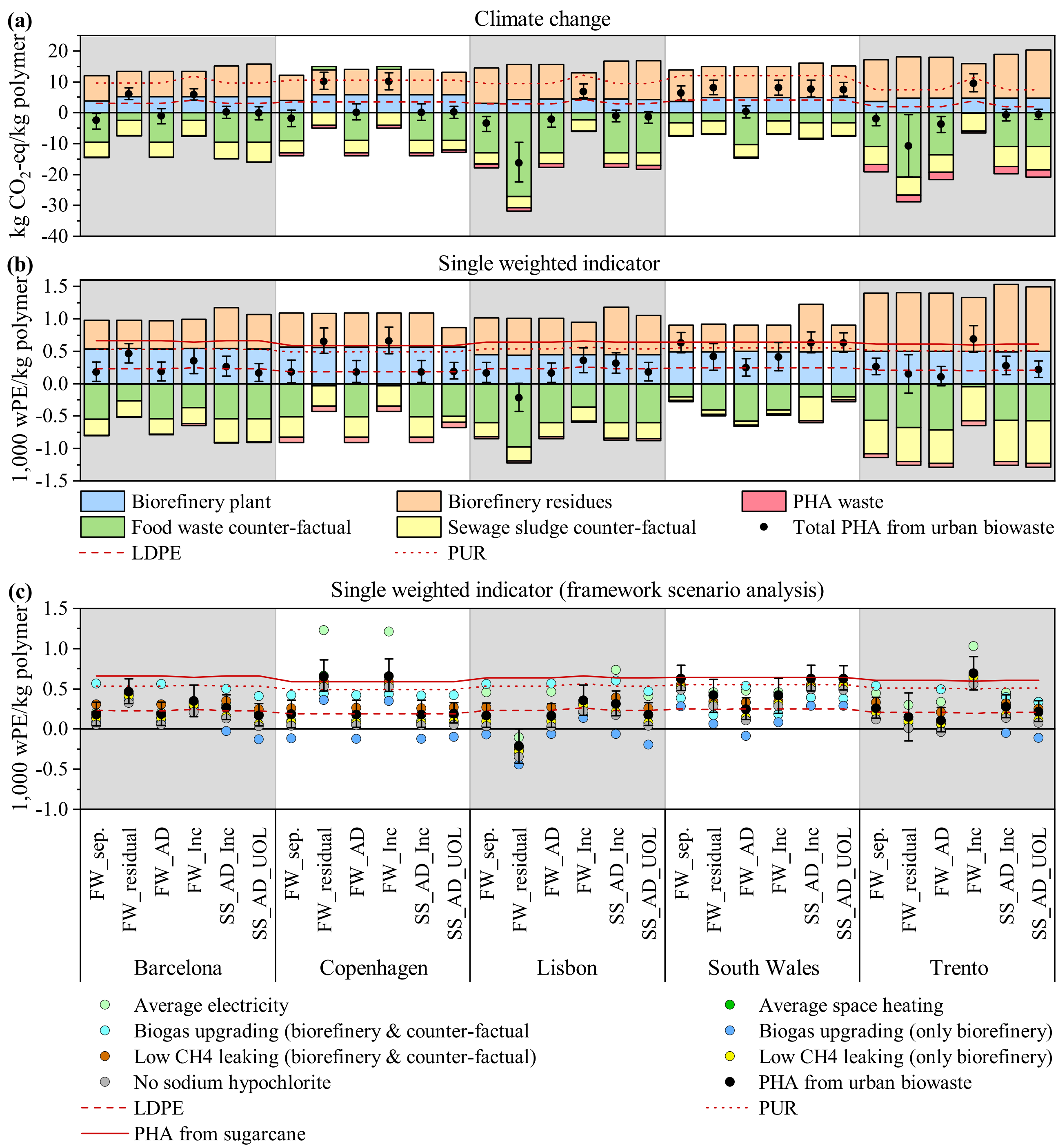

Fig. 3. *Climate change* (a), single weighted indicator (calculated by normalizing and weighting 13 midpoint categories, excluding toxicities) (b), and results of the framework scenario analysis of the single weighted indicator (c) to produce 1 kg of polyhydroxyalkanoates (PHA) from urban biowaste in the 6 alternatives for the 5 clusters. The PHA from urban biowaste is divided in the five parts of the system described in Fig. 1, and is compared to 1 kg of low-density polyethylene (LDPE), polyurethane (PUR), and PHA from sucrose (based on Harding et al. (2007)) represented by the three red lines. The uncertainty bars represent the 95% confidence interval. FW: municipal food waste; AD: anaerobic digestion, Inc: incineration, SS: sewage sludge, UOL: use-on-land, WWTP: wastewater treatment sludge. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

line associated with the competitive plastics, it indicates that the PHA has a worse environmental impact, but when the net results are below, PHA has a lower environmental impact. Finally, being the net results a sum between the processes happening in the newly built biorefinery and the avoided processes in the counter-factuals, the larger the counter-factuals the lower the net result; the lower the counter-factuals (indicating an already efficient system) the higher the net results.

3.2.1. Depletion of fossil resources

Depletion of fossil resources, quantified in MJ-eq, assesses the anthropogenic depletion of energy carriers (coal, oil, gas, peat, and uranium) compared to the ultimate reserve of the total amount of resources in the earth's crust. It is strongly dependent on the energy balance of the system, as well as on the energy avoided/consumed in each process.

The results for PHA from urban biowaste range from 40 MJ-eq in South Wales to 80 MJ-eq in Trento, in both cases when the food waste counter-factual is incineration. The low result in South Wales is due to the lack of dewatering that is connected to the consumption of iron (III) chloride for flocculation in the wastewater treatment plant. The flow with the greatest impact is the biorefinery plant, due to the consumption of sodium hydroxide during PHA extraction, while PHA incineration, landfilling, and composting have almost no impact. The analyzed PHA always performs better than PUR (80–90 MJ-eq) and can be either higher or lower than LDPE (60–70 MJ-eq), depending on the alternative. The PHA results from first-generation biomass ranged instead between 41 and 51 MJ-eq, due to the natural gas consumption in Kookos et al. (2019), to 145–155 MJ-eq, due to glucose production in Harding et al. (2007).

The framework scenario analysis does not affect the comparison with PUR. However, PHA always becomes better than LDPE (in any alternative and for all clusters) if the biogas is upgraded only in biorefinery residues management, or if no sodium hydroxide is used in the extraction process, leading to a reduction in overall impacts of 50%-250% and 30%-60%, respectively (the ranges indicate the differences in the clusters). It is noteworthy that if anaerobic digestion were improved in both the biorefinery and in the counter-factuals, the net results would be two-fold larger. This is due to the larger effect of these measures in the counter-factuals, where more biogas is produced compared to the biorefinery option, owing to the lower quantity of VS being consumed in the biorefinery.

Finally, the uncertainty bars are similar in all alternatives and clusters, mainly due to the variations in the sodium hydrochloride utilization (60%) and diesel consumption during food waste collection.

3.2.2. Climate change

Climate change (Fig. 3, a) measures the integrated infrared forcing increase of greenhouse gases in kg CO_2-eq. In this study, *Climate Change* is strongly influenced by the direct emissions (or avoided emissions) of fossil CO_2 and CH_4 into the atmosphere. The net impacts range between −16 and – 11 kg CO_2-eq when diverting food waste from landfilling in Lisbon and Trento ("FW_residual" in Fig. 3) to 8 and 10 kg CO_2-eq in Copenhagen and Trento when avoiding food waste incineration ("FW_Inc" in Fig. 3). This compares with 2 to 4 kg CO_2-eq per kg of LDPE and 5 to 7 kg CO_2-eq per kg of PUR. The PHA from first-generation biomass ranges between 3 and 5 kg CO_2-eq, mainly due to first-generation biomass production. Impact ranges for fossil plastics (LDPE and PUR) depend on how the plastic is disposed of (landfilling or incineration), and in the case of incineration, the impacts are almost doubled, due to direct fossil CO_2 emissions. As seen for *Depletion of fossil resources*, the waste management of the produced PHA (i.e. incineration, landfilling, or composting, shown in the red bars in Fig. 3, a) has a negligible effect compared to the rest of the system.

The net result of *Climate change* is lower when the counter-factuals are associated with significant impacts, such as food waste landfilling, anaerobic digestion without the strict control of CH_4, leakage, and without biogas upgrading. Conversely, the reduction is smaller when counter-factuals have a low impact profile (e.g. incineration). The results indirectly show that food waste anaerobic digestion is better than incineration, but only when the biogas is upgraded, and that national legislations and good practices should focus on minimizing leakages of CH_4 from anaerobic digestion plants (both digester, tank, and biogas utilization) and N_2O emissions from treating the liquid fraction in the dewatering step. When methane leakages from the digester, open tank, and combustion engine are minimized and the biogas is upgraded to natural gas, then the impact of the biorefinery decreases by approximately 60%. If these conditions were modeled in both the biorefinery and the counter-factuals, and the counter-factuals included anaerobic digestion, the net results would increase, because larger amounts of biogas would be produced in the counter-factuals without the biorefinery, as also observed in *Depletion of fossil resources* (section 3.2.1). However, if these conditions were implemented only in the biorefinery, the net impact of PHA from biowaste would always be lower than for fossil plastics, with the case of Copenhagen as an exception. Also for this cluster, assumptions mostly affecting the results are related to the use of sodium hypochlorite (heavily decreasing the net results) and marginal electricity.

Uncertainty in *Climate Change* is largely associated with variations in N_2O emissions from the wastewater treatment plant treating the liquid fraction derived from digestate dewatering (when present), sodium hypochlorite utilization, CH_4 leakage in the anaerobic digestion plant, and diesel consumption during food waste collection.

3.2.3. Depletion of abiotic resources

The *Depletion of abiotic resources* quantifies the extraction of minerals and metals compared to the ultimate reserve, and it is quantified in kg Sb-eq. This impact category is strongly influenced by the consumption of chemicals (e.g. chemicals in PHA extraction, iron (III) chloride in the wastewater treatment plants) and by the energy balance. Energy consumed or avoided is especially important when marginal electricity includes high shares of photovoltaics (as in Barcelona and Trento). The waste management of both PHA and fossil plastic does not have a strong impact on the net results.

In most alternatives or framework scenarios, the impacts of PHA from urban biowaste are higher than PUR and LDPE. The only exception is South Wales in "FW_sep.," "FW_residual," "FW_Inc," and "SS_AD_UOL" in Fig. 3, due to the lower impact of biorefinery residues management when the digestate is not dewatered but spread directly onto soil (meaning no iron (III) chloride is consumed).

Compared to first-generation biomass, PHA from urban biowaste is always better than PHA from glucose, while it performs better than PHA from sucrose only when food waste is diverted from AD as in "FW_AD" in Fig. 3 (excluding the cases of Copenhagen and Trento) and when the biorefinery residues are spread on land instead of being incinerated ("SS_AD_UOL" in Fig. 3), thanks to the avoided use of mineral fertilizers. The uncertainty in the results is mostly due to the variance in sodium hypochlorite utilization, to the low heating value of food waste (influencing the quantity of energy avoided during incineration), and to the capacity of the dewatering step in diverting phosphorus between the liquid and the solid fraction (influencing the quantity of avoided mineral fertilizers when organic matter is spread on land).

3.2.4. Freshwater toxicity and Human toxicity, non-carcinogenic

Freshwater eco-toxicity is mainly due to the manufacturing of sodium hypochlorite during PHA extraction, and in particular to the direct emission of 0.48 kg of chloride into surface water per kg of sodium hypochlorite produced. *Human toxicity, non-carcinogenic* is primarily caused by the use of organic fertilizers on agricultural soil, owing to the quantity of heavy metals (especially mercury, zinc, and lead) directly added to soil that is not offset by the avoided heavy metals in mineral fertilizers with the assumed fertilizer substitution factors. However, heavy metals in the digestate/compost proved to be lower than the maximum limit set by national legislations.

3.2.5. Single weighted indicators

Fig. 3, b and c, illustrate the results of the single weighted indicator without considering the toxicity impact categories. The net results (black dots) show a lower indicator than PUR (4.9E-04 to 5.6E-04 wPE) and than the three PHAs from first-generation biomass (between 5.9 and 8.0E-04 wPE), while the comparison with LDPE (1.9E-04 to 2.6E-04 wPE) depends on several conditions. This highlights the importance of first targeting the niche market of polyurethane sealants and adhesives before entering the mass market of commodity films (as also confirmed in *Depletion of fossil resources* and *Climate change*).

Chemical consumption during PHA extraction makes a significant contribution in most impact categories and is hence critical for the sustainability of the biorefinery. Such a result, however, is rarely reported in the literature, with only three previous studies (Chen et al., 2001; Leong et al., 2017; Pérez et al., 2020) including detailed inventories in this regard. If the use of sodium hypochlorite in extraction were avoided, the biorefinery would become greener compared to LDPE in all alternatives, the only exception being food waste diverted from incineration ("FW_Inc" and some of "FW_residual" in Fig. 3). However, if extraction modification were coupled with an improvement in the anaerobic digesters treating the biorefinery residues (i.e. lower CH_4 leaking and biogas upgrading), compared to the anaerobic plants in the counter-factuals, the produced PHA results would always be better than LDPE. This is due to the generic lower environmental impact of food waste incineration compared to anaerobic digestion (and a lower counter-factual causes a higher net factor) with current anaerobic digestion conditions.

The single indicator was mainly influenced by biogas upgrading and the use of sodium hypochlorite (since they affected many impact categories), while CH_4 leakage affected only *Climate change.* Another relevant process is the treatment of the reject water from digestate dewatering in the wastewater treatment plant, due to N_2O emissions (in the case of *Climate change*), the consumption of iron (III) chloride (in the case of *Depletion of abiotic resources*), and nitrogen emitted into water bodies (for either *Marine eutrophication* or *Freshwater eutrophication*, depending on the cluster). Furthermore, waste management of the produced PHA was almost negligible in the results, whilst for the fossil polymer, it only affected 10% of the single weighted indicator for fossil plastic. In many of the impact categories (and in the single weighted indicator), the Copenhagen cluster was where it is more difficult to achieve environmental benefits by diverting food waste from incineration, due to the very high energy efficiency of the incineration plants modeled for this cluster.

Finally, assumptions behind the composition of electricity and space heating, and different waste management regimes for PHA (composting versus treatment with the residual waste), did not affect the ranking of the alternatives.

3.3. Economic impacts

Fig. 4 shows the budget costs (a), the externalities (b), and the final societal LCC (c). Budget costs (Fig. 4, a) range between −0.6 (Trento, "SS_AD_UOL" in Fig. 4) and 2.0 EUR/kg PHA (Lisbon, "SS_AD_UOL"). This is generally lower than PUR (2.6 EUR/kg) and PHA from first-generation biomass (3.1 EUR/kg), while the comparison with LDPE (1.5 EUR/kg) is less clear. The budget costs of PUR, LDPE, and PHA from first-generation biomass were calculated from their corresponding market value, subtracting the profit share and adding the cost of waste treatment. The lowest budget costs for all clusters were found in the alternative "SS_AD_UOL" in Fig. 4, where both biorefinery residues and sewage sludge in the counter-factual were anaerobically digested and spread onto agricultural land (between −0.6 in Trento and 0.5 EUR/kg PHA in Lisbon), because the costs in the biorefinery and in the counter-factuals balanced out very similarly.

The most relevant contribution to budget costs (and also to the societal LCC) is the cost associated with source-separated food waste collection in the biorefinery (76% of the budget cost of the biorefinery plant) or in the counter-factuals (65%-75% of the budget costs of the food waste counter-factual). For this reason, the blue and green bars are very important in Fig. 4, c compared to biorefinery residues management (orange bars) and to the sewage sludge counter-factual (brown bars). However, collection costs do not appear too prominent when overlapping the alternative scenarios to the counter-factuals (violet bars in Fig. 4, a and b), since they are either equal in both parts of the system or slightly different when food waste is collected with the residual waste in the counter-factuals (146 EUR/t on average versus 92 EUR/t). Two other important contributions in terms of budget costs are the incineration plants (grey bars in Fig. 4, a and b), which are not offset by the generated energy, especially due to the high capital expenditure, and the significant cost of spreading digestate when not composted, owing to the high water content in digestate. All of the results involving food waste incineration in the counter-factual are lower than the others, due to the economic loss occurring when food waste is incinerated, compared to when it is composted or digested.

Collection and incineration of food waste affect uncertainty the most in "FW_Inc" in Fig. 4: Residual waste collection costs, source-separation costs, and incineration capital expenditure contribute to 73% (75%), 12% (11%), and 9% (8%), respectively, of uncertainty in the net results for the budget costs (and for societal costs).

Externalities (Fig. 4, b) are mainly the result of the emissions of a few substances into the air: Methane (both fossil and non-fossil), fossil carbon dioxide, nitrogen oxides, particulates smaller than 2.5 μm, sulfur dioxide, dinitrogen monoxide, lead, and ammonia. Externalities show a similar pattern to *Climate change* (section 3.2.2).

Between 60% and 80% of the societal costs are generally due to budget costs (transformed in shadow prices), while externalities contribute around 20%-40%. Societal costs range between −0.6 (in Trento, "SS_AD_UOL") and 4 (in Copenhagen, "FW_residual" or "FW_Inc") EUR/kg PHA, compared to 1.9 to 2.3 kg LDPE, 4 to 4.3 EUR/kg PUR, 4.8 to 6.5 kg PHA from first-generation biomass. Societal costs are lower than LDPE in all clusters apart from South Wales in "FW_sep.", "FW_AD", and "SS_AD_UOL" in Fig. 4. However, similarly to the environmental results, the analyzed PHA shows consistently lower societal costs, only if the biorefinery is combined with lower CH_4 emissions, biogas upgrading, and the avoidance of sodium hypochlorite.

Finally, while the management of 1 kg of PHA waste is negligible in terms of budget costs, externalities, and societal costs (red bars in Fig. 4, c), it increases all costs by 8% in the case of PUR, and by 15% (for budget costs and societal LCC) and 27% (for externalities) for LDPE.

3.4. Recommendations to policy-makers

When seeking to lower the environmental impact of plastic consumption by substituting fossil plastic with bioplastics, this study identifies some recommendations to policy-makers.

First of all, LCA and societal LCC are indispensable tools to scientifically compare the impacts of different polymers and biopolymers providing the same functionality.

Second, when designing economic incentives for bioplastics and biorefineries, the environmental hotspots in the entire management chain have to be considered, including the biorefinery residue management. Producing bioplastics from waste instead than from first-generation biomass, as in the studied PHA, can lower the environmental and economic impacts of the bioplastics due to the avoided production of crops. In this case, the treatments that the waste would have without the construction of the biorefinery need to be included in the analysis (here called "counter-factuals") and these counter-factuals could result in either environmental savings or burdens.

Third, it is important to identify the fossil material that is competing with the produced bio-plastics since different fossil polymers can have

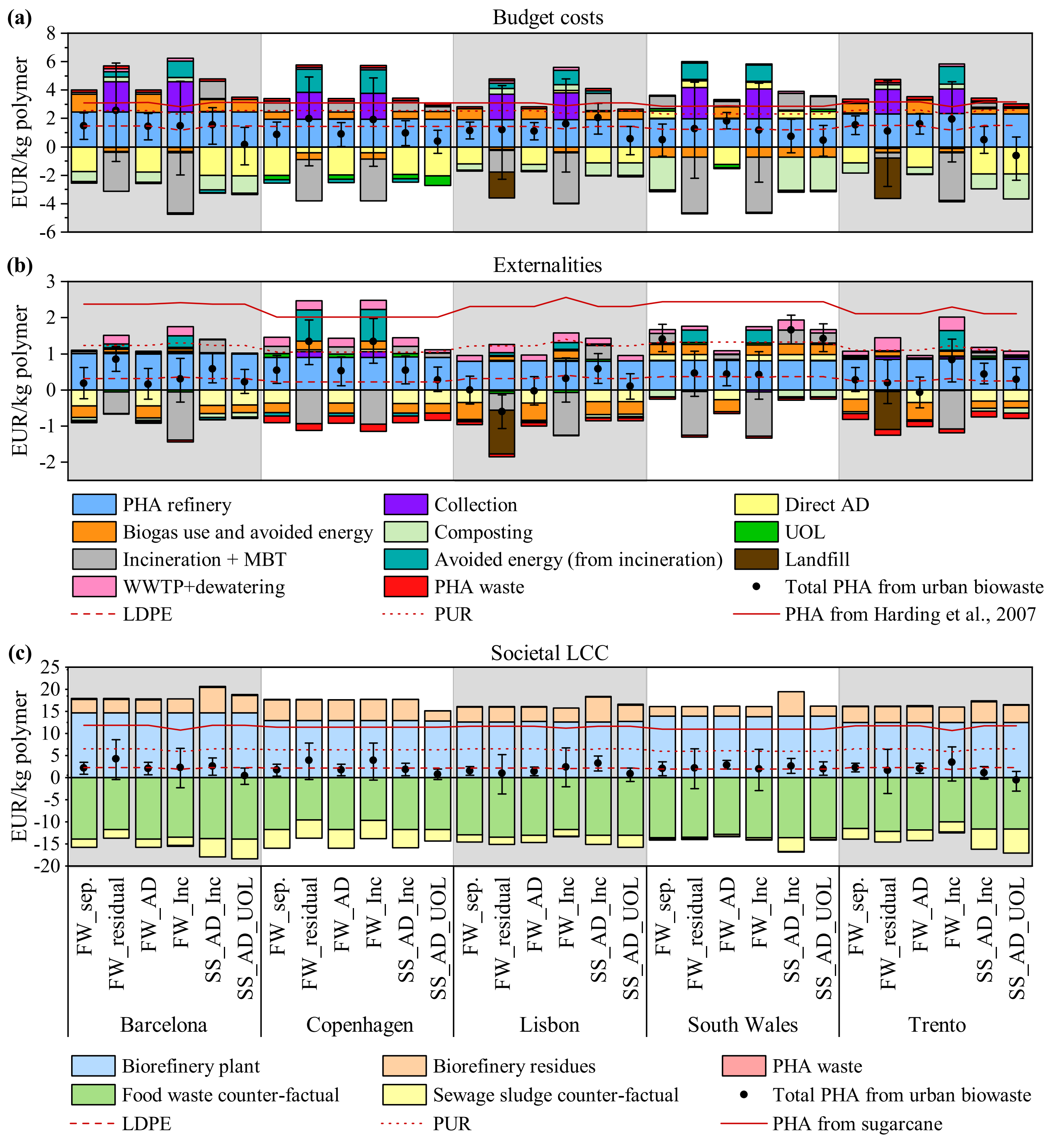

Fig. 4. Budget costs (a), externalities (b), and societal LCC (c) required to produce 1 kg of polyhydroxyalkanoates (PHA) from urban biowaste, compared to 1 kg of low-density polyethylene (LDPE), polyurethane (PUR), and PHA from sucrose (based on Harding et al. (2007)) represented by the three red lines . (a) and (b) are divided per process, where each process represents the difference between the same process in the biorefinery and in the counter-factuals; (c) is divided in the five parts of the system described in Fig. 1. Uncertainty bars represent the 95% confidence interval. MBT: mechanical–biological treatment; FW: municipal food waste; AD: anaerobic digestion, Inc: incineration, SS: sewage sludge, UOL: use-on-land, WWTP: wastewater treatment sludge. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

quite different environmental performance and market value. The studied biodegradable and bio-based PHA from urban biowaste resulted in lower environmental and economic impacts compared to the niche market of polyurethane, representing a small market with high environmental impacts, market values and a non-existent recycling chain. However, the results were not as clear for the much larger market represented by LDPE. These conclusions should incentivize the future biorefineries producing PHA to ensure that the technical properties of the biopolymer could be used to compete with traditional sealants and adhesives made of polyurethane. At the same time, it indicates that specific regulative actions should be considered to support these changes in the plastic market.

4. Conclusions

Generally, PHA performed better environmentally and economically than polyurethane and PHA from first-generation biomass; consequently, products such as sealants and adhesives should be future targets for PHA. However, the results strongly depended on the alternative management of urban biowaste, the management of biorefinery residues, and local framework conditions. Performance-wise, biorefineries should minimize the use of sodium hypochlorite in PHA extraction, reduce CH_4 leakage from anaerobic digestion, ensure biogas is upgraded to biomethane, and improve treatment of the liquid fraction from digestate dewatering. The potential for PHA production from local biowaste was strongly affected by local resource availability and (bio) chemical characteristics.

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Credit authorship contribution statement

Susanna Andreasi Bassi: Conceptualization, Formal analysis, Methodology, Investigation, Software, Writing - original draft. **Alessio Boldrin:** Conceptualization, Funding acquisition, Methodology, Supervision, Validation, Writing - review & editing. **Giammarco Frenna:** Investigation, Formal analysis, Writing - review & editing. **Thomas F. Astrup:** Conceptualization, Methodology, Supervision, Funding acquisition, Writing - review & editing.

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.

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Data availability

Datasets (system boundaries, life cycle inventory, LCA and LCC results) related to this article can be found at https://doi.org/10.11583/DTU.13636436, hosted at DTU Data (Andreasi Bassi, 2021).

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