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3D printing to enable the reuse of marine plastic waste with reduced environmental impacts

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Abstract

Over the years, our oceans have witnessed an enormous accumulation of marine plastic waste resulting from ocean-related economic activities. As plastic pollution adversely affects marine wildlife and habitat, our society requires urgent solutions to address this increasingly alarming dilemma. Here, we turn our attention to circular economy principles to reduce the amount of nonbiodegradable petroleum-based marine litter. We consider a production process based on 3D printing to fabricate products for the marine industry, which uses marine plastic waste as a source material. Additionally, the suitability of virgin bio-based polyamide (bio-PA), polylactic acid (PLA), and polyhydroxybutyrate (PHB) is explored. PHB is selected due to its extraordinary rapid biodegradation in aquatic environments. To quantify the environmental impacts of the proposed processes, a cradle-to-grave life cycle assessment (LCA) is applied according to ISO 14040:2006 and ISO 14044:2006 standards. Different endof-life alternatives are proposed, including landfill deposition, thermal degradation, and composting. LCA results reveal that the use of marine plastic waste is environmentally preferred in comparison with bio-PA, PLA, and PHB. Specifically, the global warming indicator, considered a prime driver toward sustainability, shows a 3.7-fold decrease in comparison with bio-PA. Importantly, the environmental impacts of PHB production through crude glycerol fermentation are quantified for the first time. Regarding the end-of-life options with a composting scenario, PLA and PHB are preferred as they yield biogenic carbon dioxide (CO₂), which can be used as a renewable energy source.

KEYWORDS

3D printing, bioplastics, circular economy, industrial ecology, life cycle assessment, marine plastic waste

1 | INTRODUCTION

With a worldwide plastic production of 370 million tons in 2019 (PlasticsEurope, 2020), it is estimated that 12.7 million tons of plastics enter the ocean annually. This amount is to be added to the already 150 million metric tons that currently circulate our marine environments (Jambeck et al., 2015). The dumping of plastics into the sea mainly originates from land-based sources (landfills) and aquatic human activities such as fishing

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(Galimany et al., 2019). The marine pollution produced by plastic materials is considered one of the greatest dangers of the 21st century as it seriously threatens plant and animal species (Calabrò & Grosso, 2018), and several economic sectors such as tourism, shipping, and fishing (Barboza et al., 2018). Marine plastic pollution also threatens human health. The accumulation of plastic debris is transferred along the food chain through bioaccumulation and biomagnification processes, finally harming human health (Markic et al., 2020; Rochman et al., 2014; Wang et al., 2016). As the plastic debris accumulate along the entire water column, both on the surface of the sea and on the seabed (Barnes et al., 2009), plastic pollution is widespread throughout all the world's oceans (Jambeck et al., 2015). As a matter of fact, reports on plastic debris in the marine environment date back to the 1970s (Law, 2017).

Plastic contamination has reached a turning point where academia, industry, and society must provide urgent and viable solutions. In this sense, mainly three types of solutions are proposed: reducing the use of plastic, promoting the reuse of plastic waste (Adam et al., 2020; Schnurr et al., 2018), and replacement of traditional petroleum-based polymers by biodegradable polymers based on renewable raw materials are being sought (Luckachan & Pillai, 2011).

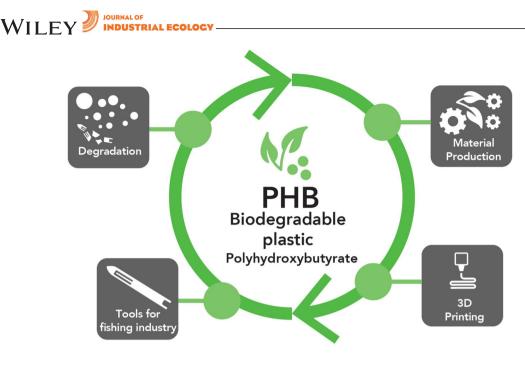
This work is focused on the last two proposals: Reusing marine plastic waste and using biodegradable plastics. However, plastic waste revalorization or the use of renewable and biodegradable materials does not involve per se a direct reduction on the environmental footprints as the impacts associated with energy use or transport could be increased. Therefore, to get a holistic vision on how the environmental impacts could be reduced by incorporating marine plastic waste into the production process, the environmental impacts of the process from the life cycle's point of view should be quantified (Baumers et al., 2011). In this framework, life cycle assessment (LCA) methodology is considered a reference for the study of the environmental impacts associated with products, processes, and services (Crenna et al., 2019). By identifying and quantifying the emissions and consumption of resources at every stage of the life cycle (Sambito & Freni, 2017), it is possible to successfully evaluate the environmental performance of products/processes as varied as batteries (Iturrondobeitia et al., 2021), lignin valorization (Kulas et al., 2021), aluminum industry (Zhu & Jin, 2021), or mineral recovery (Huang et al., 2021). ISO 14040:2006 and ISO 14044:2006 standards set the principles, framework, requirements, and guidelines for conducting an LCA study so the production patterns and end-of-life (EoL) scenarios can be carefully analyzed to provide cues for the reduction of the global environmental pressures (Crenna et al., 2019). In this context, Zabaniotou and Kassidi reported in 2003, one of the first studies applied to the determination of the environmental impacts of plastics, where the use of polystyrene and recycled paper in food packaging was compared (Zabaniotou & Kassidi, 2003). Henceforth, LCA has been gaining further relevance, for example, by evaluating the environmental sustainability of biodegradable polymeric films (Vidal et al., 2007).

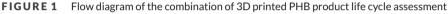
Regarding the reuse of marine plastic waste, this work proposes the use of 3D printing technology to manufacture needles to fix broken nets using marine plastic waste (fishing nets composed of petroleum-based polyamide) as a feed material (Garrido et al., 2020). The fishing nets are collected at different seaports along the Atlantic coast. This work focuses its attention on the nets collected at the seaport of Ondarroa (year 2020), one of the most important seaports in the Bay of Biscay (Atlantic Ocean) according to fishing volume. Many studies are focused on the recycling and reusing of marine plastic waste, for instance, polyamide 66 fishing nets (Mondragon et al., 2020; Srimahachota et al., 2020); besides this study is complemented by the 3D processing technique as well as LCA.

3D printing is gaining increasing prominence to fabricate products with complex geometries (Scott et al., 2020) or to implement new approaches to reuse and recycle plastics (Sanchez-Rexach et al., 2020). 3D printing is generally defined as the production of components by successive deposition of layers of materials; fused filament fabrication, in particular, uses thermoplastic filaments as feedstock material. These printable filaments can be composed from a wide variety of materials provided they show thermoplastic properties, opening the path to different valorization possibilities of plastic waste to obtaining brand-new consumable products (Mikula et al., 2021). It could be thus expected that the use of waste as a feed material in a 3D printing process could yield environmental benefits as the contribution of the raw material extraction will be reduced.

The alternative of using biopolymers has drawn enormous attention as a viable alternative to conventional plastics and has seen increasing relevance in the fields of sustainable packaging (Shogren et al., 2019), energy storage (Lizundia & Kundu, 2021), biomedicine (DeStefano et al., 2020), and textiles (Jahandideh et al., 2021). In 2019, with 3.8 million tonnes, the global production capacity of biodegradable plastic solely represented 1% of all plastic production (Bioplastics Magazine, 2020). Polylactic acid (PLA) is considered as the most prominent bioplastic due to its low price and physico-chemical properties. Unfortunately, its slow biodegradation kinetics makes this material nondesirable in applications where the end-of-life (EoL) scenario is critical. In this sense, polyhydroxyalkanoates (PHAs) are polyesters that can be degraded under composting and marine environments with no harmful byproducts, a rather uncommon feature in the family of biopolymers. Polyhydroxybutyrate (PHB), the most widely used polymer within the PHA family, has similar thermomechanical properties than the petro-based polypropylene and is synthesized via a fermentation process by microorganisms (Chen et al., 2020).

With no doubt, waste- and bio-based plastics are appealing candidates to supplant petroleum-derived materials in many applications so that future demands on the sustainability in the life cycle of plastics can be fulfilled (Sanchez-Rexach et al., 2020). In this hypothesis, we propose to lessen the environmental pressures caused by marine waste accumulation by reusing polyamide fishing nets as a feed material for a 3D printing process. The environmental impacts of the 3D printing process using different feed materials including a virgin conventional petroleum-based polyamide, a bio-derived polyamide, and two biodegradable polymers (PLA and PHB) are also compared. The literature shows scarcity of studies that combine 3D printing technology, PHB bio-based polymer, and LCA. A cradle-to-grave LCA covering the process of the recovery of maritime plastic waste, the production of raw materials, the processing through 3D printing, and the EoL is performed. Importantly, to the best of our knowledge, no works have





evaluated the environmental hotspots arising from the production/collection and subsequent reuse of plastic waste through LCA. Combined with 3D printing, this work has the potential to serve as a roadmap to guide follow-on works in the field of plastic waste reuse and LCA. Interestingly, the use of waste as a feed material for brand new products also reduces the need of raw materials, one of the cornerstones of circular economy (Ingrao, Arcidiacono, et al., 2021; Ingrao, Nikkhah, et al., 2021).

2 | METHODS

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2.1 | Goal and scope

As summarized in Figure 1, this study aims to compare the environmental impacts during the whole life cycle of a product used in the marine industry obtained by 3D printing (example: a needle for mending nets). The main target is to reduce the environmental impacts caused by the conventional plastic industry through the reuse of marine plastic waste. To that end, two approaches have been studied:

- 1. Plastic waste (mainly nets, which after being analyzed, the main material was found to be polyamide 66) collected from the sea is processed through 3D printing into a new product, and the environmental impacts of the process are quantified using the LCA methodology.
- The use of biodegradable polymers to produce the same product by 3D printing and the environmental impacts of the process are quantified using the LCA methodology.

As reference, raw virgin polymer (polyamide 66) has been used for the same processes and product and environmental impacts have also been quantified using the LCA methodology. The first alternative is carried out in the lab at semi-industrial scale. The plastic waste was collected at the port (during the year 2020), so waste from the open sea is not included. From all the plastic waste collected, only fishing nets are used. For the second alternative, we explore the use of biodegradable plastics from renewable origin, PLA and PHB as a plausible alternative to lessen the environmental impacts of the process (Figure 1). For this step, the same processing techniques used for the first step are considered. For both alternatives different EoL cycle scenarios are estimated: landfill, thermal degradation, and composting. Therefore, LCA is applied to five different scenarios (Figure 2), where 3D printing processes are used for the fabrication of a product for the marine industry using different feed materials and EoL. The originated environmental impacts are compared so a recommendation to lessen overall environmental impacts can be made. To perform the study, 1 kg of 3D printed material is used as the functional unit (FU). The following scenarios are considered:

 <u>Scenario 1 (PA-Petrol)</u>: Petroleum-based polyamide (PA-66) raw material. Here, two different EoL scenarios are chosen: landfill deposition and thermal degradation.

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FIGURE 2 Schematical representation summarizing the followed LCA scope and boundaries

- <u>Scenario 2 (PA-NETS)</u>: The material used for 3D printing originates from marine plastic waste composed by PA-66. Before printing, the marine plastic waste is treated through extrusion, watertight bathing, and pelletization. Similarly, landfill deposition and incineration are considered as EoL choices.
- Scenario 3 (PA-Bio): Bio-based from castor oil polyamide (PA-66) raw material. Landfill deposition is considered as EoL scenario.
- <u>Scenario 4 (PLA)</u>: PLA is used as a material for 3D printing. This thermoplastic aliphatic polyester is biocompatible and biodegradable and can be produced from renewable resources (Naser et al., 2021; Vroman & Tighzert, 2009). Its relatively low melting temperature of 150–180°C allows reduced energy consumption during printing in comparison to other thermoplastics (Przekop et al., 2020). Due to its degradability, landfill deposition and composting are considered as EoL.
- <u>Scenario 5 (PHB)</u>: PHB is used for 3D printing. Apart from being based on renewable resources, PHB can be readily degraded in marine environments (Naser et al., 2021). For the first time, to the best of our knowledge, we computed the impacts arising from the synthesis of PHB. Composting is considered as the EoL scenario.

2.2 | Inventory analysis

During the inventory analysis, mass and energy balances are identified and the inputs (materials, energy, and resources) and outputs (wastes and emissions) for the system are defined (Hauschild et al., 2018). Primary data has been used to define the inventory as the information was directly collected from main sources at Leartiker Company (production plant is Leartiker Company in Markina-Xemein, Biscay, Basque Country, see Table 1 and Figure 3). All the inventories are disclosed in Supporting Information as Tables S1-S6.

All the processes and amounts of inputs and outputs were measured in the laboratory. Then, these were the references to define the processes for the rest of the scenarios. The EoL scenarios were defined based in the used databases.

Plastic waste from fishing nets is used to manufacture the final product. After cleaning and grinding down the nets, a drying (DESTA H 300), extrusion (requiring calcium carbonate), bathing, and pelletization process is applied (Labtech LTE 26–40/22kw) to prepare the filaments for

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TABLE 1 Inventory and modeling for the life cycle materials and energy flows for 1 kg of material processed by 3D printing using polyamide from marine nets waste origin "PA-nets" and its EoL. (a) waste pretreatment; (b)optimization; and (c) 3D printing and EoL

		Flows	Incut	Outer	A	Unite	Source/provider (Ecoinvent 3.7 + Gabi
(a) Process	Character	Flows	Input	Output	Amount	Units	bioplastics 2019)
Waste pretreatment	Cleaning	Nets Water	x		2 3	kg L	x Tap water production, conventional treatment tap water Cutoff, U-Europe without Switzerland
		Nets clean		х	1.6	kg	х
		Waste nets		x	0.4	kg	Landfill of plastic waste, at landfill site, landfill including landfill gas utilization and leachate treatment
	Shredding	Nets clean	х		1.6	kg	Х
		Energy	x		5	kWh	Electricity voltage transformation from high to medium voltage electricity, medium voltage Cutoff, U—ES
		Shredded nets		х	1.4	kg	Х
		Waste		x	0.2	kg	Landfill of plastic waste, at landfill site, landfill including landfill gas utilization and leachate treatment
(b) Process		Flows	Input	Ouput	Amount	Units	Source/provider (Ecoinvent 3.7 + Gabi bioplastics 2019)
Compounding	Dry	Shredded nets	х		1.4	kg	Х
		Energy	x		5	kWh	Electricity voltage transformation from high to medium voltage electricity, medium voltage Cutoff, U - ES
		Shredded + dried nets		х	1.4	kg	x
	Extrusion + Bath	Additive (Calcium Carbonate)	x		0.11	kg	Calcium carbonate production, precipitated calcium carbonate, precipitated Cutoff, U—RER
		Energy	x		12.7	kWh	Electricity voltage transformation from high to medium voltage electricity, medium voltage Cutoff, U—ES
		Optimized plastic		x	1.5	kg	X
		Waste (polypropy- lene [PP])		x	0.5	kg	Landfill of plastic waste, at landfill site, landfill including landfill gas utilization and leachate treatment

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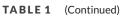
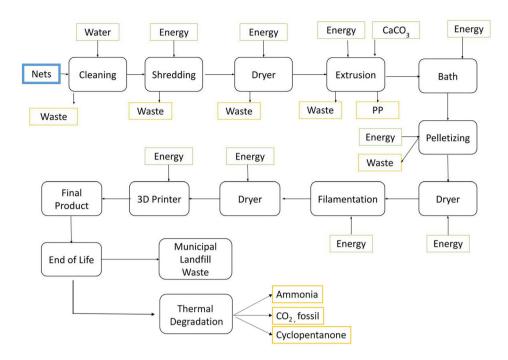
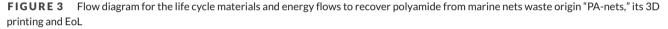




TABLE 1	(Continued)						
(b) Process		Flows	Input	Ouput	Amount	Units	Source/provider (Ecoinvent 3.7 + Gabi bioplastics 2019)
		Shredded + dried nets	х		1.5	kg	Х
	Pelletizing	Pellets		х	1.5	kg	Х
		Optimized plastic	х		1.5		Х
		Energy	x		5	kWh	Electricity voltage transformation from high to medium voltage electricity, medium voltage Cutoff, U—ES
(c) Process		Flows	Input	Ouput	Amount	Units	Source/provider (Ecoinvent 3.7 + Gabi bioplastics 2019)
	Filament + Dry	Waste		x	0.5	kg	Landfill of plastic waste, at landfill site, landfill including landfill gas utilization and leachate treatment
		Filamented optimized plastic		x	1	kg	х
		Pellets	х		1	kg	Х
		Energy	x		18.8	kWh	Electricity voltage transformation from high to medium voltage electricity, medium voltage Cutoff, U–ES
Transport	Transport	Transport	x		38.18	TKm	Market for transport, freight, lorry 16–32 metric ton, EURO5 transport, freight, lorry 16–32 metric ton, EURO5 Cutoff, U–RoW
3D printing	Dry	Filamented optimized plastic dried		x	1	kg	x
		Filamented optimized plastic	x		1	kg	x
		Energy	x		10	kWh	Electricity voltage transformation from high to medium voltage electricity, medium voltage Cutoff, U–ES
	3D Impression	Waste		Х	0.5	%	Landfill of plastic waste, at landfill site, landfill including landfill gas utilization and leachate treatment
		Final Product		х	0.95	kg	Х
EoL	Landfill/thermal Degradation	Processes from	Ecoinvent 3	s	Landfill of plastic waste, at landfill site, landfill including landfill gas utilization and leachate treatment		





printing (3devo Filament extruder). Before 3D printing (Ultimaker 2 and extended printer), a final drying step is also applied (DESTA H 300). Because of the nonbiodegradable character of polyamide, landfill is selected as the most likely EoL option. The flow diagrams for the rest of the scenarios are available in the Supporting Information 1 and 2.

2.3 | Life cycle interpretation

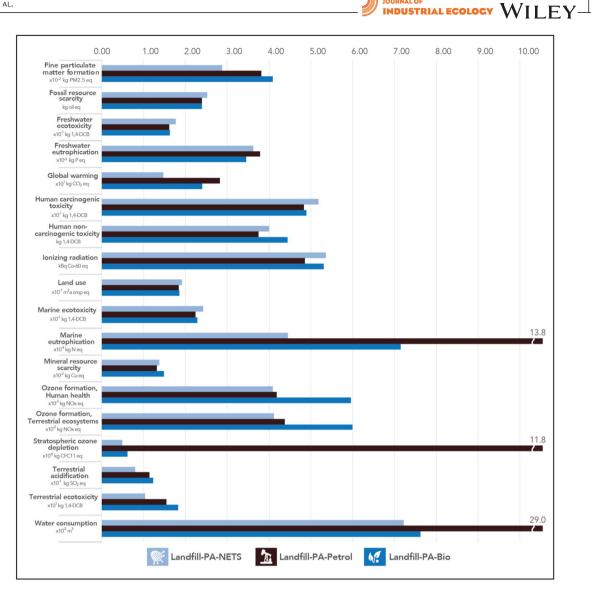
LCA studies are performed with OpenLCA 1.10.3 software using Ecoinvent 3.7 and Gabi bioplastics 2019 databases. Environmental impacts are categorized into midpoint indicators according to the ReCiPe 2016 Midpoint method. The use of ReCiPe 2016 over other methods such as CMLbaseline provides additional impact categories that can certainly help provide more meaningful information for future follow-on works. Precisely, several categories worthy of analysis in this study are not covered by other methods such as the CML-IA (land use, fossil resource scarcity, mineral resource scarcity, or water consumption). Additionally, midpoint indicators were selected over endpoint ones because the former focus on single environmental problems, while the latter show the environmental impact on three higher aggregation levels (human health, biodiversity, and resource scarcity) that increase data uncertainty. A cradle-to-grave perspective was followed to understand the whole life cycle, considering the material procurement, production, use, and EoL.

Sensitivity analysis is also carried out varying the source of the "Energy" used in the processes of life cycle of 1 kg of 3D printed material with PA-NETS: *Electricity voltage transformation from high to medium voltage* | *electricity, medium voltage* | *Cutoff, U*—ES and *electricity production, photovoltaic, 3kWp slanted-roof installation, multi-Si, panel, mounted, label-certified* | *electricity, low voltage, label-certified* | *Cutoff, U*—GLO. In the selected life cycle, the electricity plays an essential role, being the cause of the main impacts. The results of the analysis are shown in Supporting Information S11 and they reveal that the selection of the origin of the inputs, for instance, the "energy" required for each process under study is critical and that the results vary 50% in absolute values.

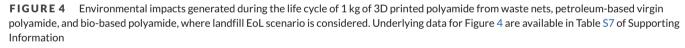
3 | RESULTS

3.1 Environmental impacts of polyamide-based product with EoL landfill: Scenarios 1–2–3

We firstly considered three scenarios involving the use of polyamide from different resources as a feed material for the 3D printing process. Firstly, Figure 3 summarizes the process in which plastic waste from fishing nets is used to manufacture the final product. Because of the nonbiodegradable



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character of polyamide, landfill is selected as the most likely EoL option. On the other hand, Figure S1a depicts the process that uses petroleumbased virgin polyamide as a feed material. In this scenario, the raw material is transported to the production plant. After a drying step, filament formation, and an additional drying, the material is ready to be printed. Landfill is also chosen as the EoL option. Finally, Figure S1b shows the process using bio-based polyamide.

Figure 4 shows the environmental impacts arising from these three scenarios, which are categorized into 18 indicators according to ReCiPe 2016 Midpoint (see Table S7 for further specific quantitative values). It can be seen that the process using polyamide waste nets as a feed material involves a reduction in the global warming category by 48–62%. In fact, a contribution of 1.47 kg·CO₂ eq. is achieved for this process, in comparison with the 2.82 and 3.83 kg·CO₂ eq. estimated for the processes using petroleum-based virgin polyamide and bio-based polyamide, respectively. However, increased impacts are observed in the categories of water depletion, urban land occupation, ionizing radiation, and ozone depletion. When polyamide of petrochemical origin is used, large impacts in urban land occupation, water depletion, marine eutrophication, and ionizing radiation are achieved. This is probably due to the process of the extraction of petroleum and its processing, including the large equipment required for those purposes in land and oceans. The use of bio-based polyamide largely contributes to metal depletion, global warming, ozone depletion, terrestrial acidification, and human and terrestrial ecotoxicity categories. Bio-based materials do not necessarily have small environmental impacts. High-energy demand is particularly due to renewable energy from sun, which is required to grow the castor oil plant. Corn field also requires energy from the sun, but the impact is much lower. Altogether, these results indicate that the use of polyamide based on marine plastic waste is environmentally preferred over the other two options as it shows the lower impacts in 11 of the 18 studied categories.

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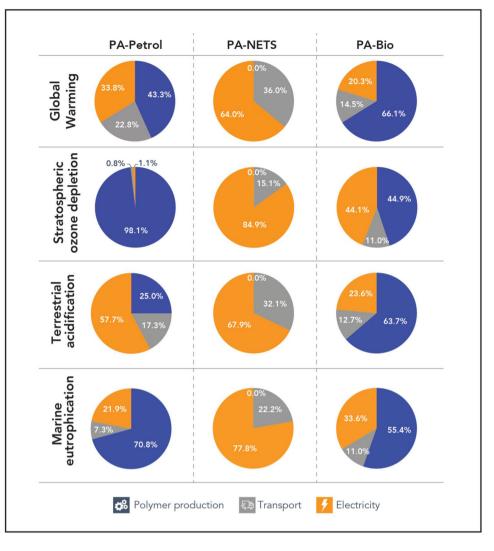


FIGURE 5 The contribution of different life cycle stages to global warming; terrestrial acidification; marine eutrophication, and stratospheric ozone depletion. Underlying data for Figure 5 are available in Table S12 of Supporting Information

To get further insights into how the different scenarios could contribute toward environmental sustainability, four impact categories are further analyzed. The global warming category is selected given its relevance as impact indicator to smooth our transition to a low-carbon economy and thus meeting the %55 emission reduction by 2030 (Paris Agreement). Terrestrial acidification, marine eutrophication, and ozone depletion are also considered given the scope of the materials studied here, which end up in oceans, landfills or under composting conditions, or originate from agricultural resources using extensive amounts of fertilizers or pesticides. Figure 5 shows the contribution of different activities and supplies: polymer production, electricity, and transport. For PA-petrol and PA-bio, the electricity is referred to the supply required for the 3D printing, whereas for PA-NET is the energy required to recycle the waste as well as to process it by 3D printing. The transport describes mainly the route from the production of the polymer to the processing. The global warming potential, which is defined as the warming of the climate system due to human activities, where greenhouse gases (GHGs) such as CO_2 , CH_4 , or nitrous oxides are the main contributors (Levasseur, 2015). The GWP generated by petroleum-based polyamide production is 2.82 kg·CO₂ eq., material production and the electricity being the main drivers with a relative contribution of 43.0% and 34.0%, respectively. On the contrary, in the case of bio-based polyamide (total impact of 3.83 kg·CO₂ eq.), the production stage reaches a 66.0% of the whole impact. Finally, the process using plastic waste-based polyamide generates 1.47 kg of CO₂ eq., the use of electricity being the most polluting phase (64.0 %). Those results can be explained by the fact that even though fossil resources are not essentially required to produce bio-based polyamide, current technology generates large CO2 emissions (Kamau-Devers & Miller, 2020). In this sense, Cheroennet et al. (2017) argued that the electricity demand in the production of bio-based plastic can greatly exceed the one required by petrochemical-based polymers. Figure 5 also presents the distribution of terrestrial acidification throughout the life cycle of the three scenarios. Terrestrial acidification is analyzed because it causes the devastation of forests and many animals with shells, being mainly caused by acidifying pollutants such as sulfur dioxide (SO₂), hydrochloric acid (HCl), ammonia (NH₃), and nitrogen oxides (NO_x) (Kim & Chae, 2016). Petroleum-based polyamide generates an impact

Marine eutrophication consists of an excess of nutrients generated by chemical fertilizers or discharged sewage (generally containing phosphates and nitrates) that cause undesired algal growth (Kim & Chae, 2016). With a relative contribution of 71% and 55% in the marine eutrophication category (Figure 5), material production is the most polluting phase for both virgin polyamide (total impact of 1.38×10^{-3} N kg eq.) and bio-based polyamide (total impact of 4.45×10^{-4} N kg eq.), respectively. Contrarily, when polyamide is extracted from marine waste (total impact of 9.2×10^{-4} N kg eq.), the use of electricity is the main polluting activity with a weight of 78%.

Finally, Figure 5 accounts for stratospheric ozone depletion, which is a relevant category as the ozone layer absorbs the most energetic ultraviolet radiation that can interact with biological tissues, causing possible tumors and mutations in aquatic and terrestrial ecosystems (Gaur et al., 2018). Chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) are the main contributors here (Burkholder et al., 2015). With a contribution of 96% for material production process and 84% for the use of electricity, they are the aspects that most largely influence the ozone depletion in the case of virgin petroleum-based polyamide and waste-derived polyamide, respectively (total contribution of 1.18×10^{-4} kg CFC-11 eq. and 4.93×10^{-6} kg CFC-11 eq.). As with the other impact categories, material production has a large share in the process relying on bio-based polyamide, a 45%, as well as the electricity, 44% (total impact of 1.12×10^{-5} kg CFC-11 eq.). In this context, Tamburini et al. (2021) also found that bioplastics display a larger impact in the category of ozone depletion. Indeed, it is accepted that conventional/industrial farming practices of biomass (which use fertilizers generating nitrous oxide (N₂O) emissions, the single most important driver of ozone depletion globally) are the key contributors to the large acidification, eutrophication, and ozone depletion potentials of bio-based materials (Weiss et al., 2012). Altogether, those results reveal that the scenario involving polyamide based on marine plastic waste is the most environmentally friendly option among the three studied alternatives. It does not only exhibit the lowest impacts in most of the categories but also generates a markedly reduced global warming potential (3.7-fold decrease in comparison with bio-based polyamide), which is considered a prime driver toward sustainability.

To put these results into context, we focus our attention on current efforts to lessen the environmental pressures arising from plastic production and consumption. Novel alternatives such as the collection and reuse of waste from marine and terrestrial environmental are highly recommended (2030 Climate Target Plan, 2022). In fact, considering the large share of the electricity required to process marine plastic waste, the environmental impacts can be even lowered by changing the source of the electricity used throughout the process. We have modeled an "electricity medium high voltage," which uses nonrenewable energy sources. Therefore, we expect that the impacts could be further reduced upon the use of renewable energy sources. Furthermore, in this scenario, plastic waste is removed from the sea, limiting the amount of human-created waste that has been released into the ocean and contributing to the Sustainable Development Goal 14, "Life below water" (United Nations Sustainable Development, 2022). Unfortunately, in the current scenario, bio-based materials generate significantly larger impacts in the categories of terrestrial acidification and eutrophication. Those results are in line with the recent conclusions drawn by Kamau-Devers and Miller (2020), who found that bio-based plastics present higher impacts than any petrochemical plastic as a result of the extensive use of agricultural resources (land, water, fertilizers or pesticides) during industrial biomass cultivation.

3.2 Environmental impacts of polyamide-based product with EoL thermal degradation and PLA with composting EoL: Scenarios 1–2–4

The environmental impacts arising from the use of materials having a different EoL scenario are analyzed here. Specifically, Figure S1 and S2 depict the flow diagrams for the processes involving use of polyamide plastic waste with a thermal degradation EoL, petroleum-based polyamide with a thermal degradation EoL, PLA with landfill EoL, and PLA with a composting EoL. Generally, all these processes include the transport of the material to the processing plant (of the cleaning and grinding of the nets in the scenario where polyamide nets are used), and a conditioning process involving drying, filament formation, additional drying, and 3D printing. The thermal degradation of the polyamide yields low molecular gases such as NH₃, CO₂ of fossil origin, water (H₂O), and carbon oxide (CO) as well as cyclopentanone byproducts (Pagacz et al., 2015). These cyclopentanone byproducts are considered flammable and irritant to the skin and eyes (GHS Hazard Statements of H226, H315, and H319). It also rates a 2 in health, 3 in flammability, and 0 in instability according to NFPA 704 (Standard System for the Identification of the Hazards of Materials for Emergency Response). On the contrary, the EoL considered for PLA results in the formation of nonhazardous methane (CH₄), biogenic CO₂, and H₂O (Kliem et al., 2020; Obarzanek-Fojt et al., 2014).

Figure 6 summarizes the environmental impacts grouped into 18 categories arising for the scenarios considered earlier (with different EoL). Further information could be found in Table S8. It is observed that PLA with a composting EoL displays a reduced impact on freshwater eutrophication, while for petroleum-based polyamide larger impacts are obtained in the categories of stratospheric ozone depletion, water consumption, and global warming. For the sake of comparison GWP, terrestrial acidification, and marine eutrophication categories are selected. Regarding the EoL with landfill scenario, polyamide based on marine plastic waste can be considered as the best option, with a total impact of 14.7 kg·CO₂ eq. In this scenario, bio-based polymers are found to generate less impacts than fossil-based polymers in this impact category (Walker & Rothman, 2020).

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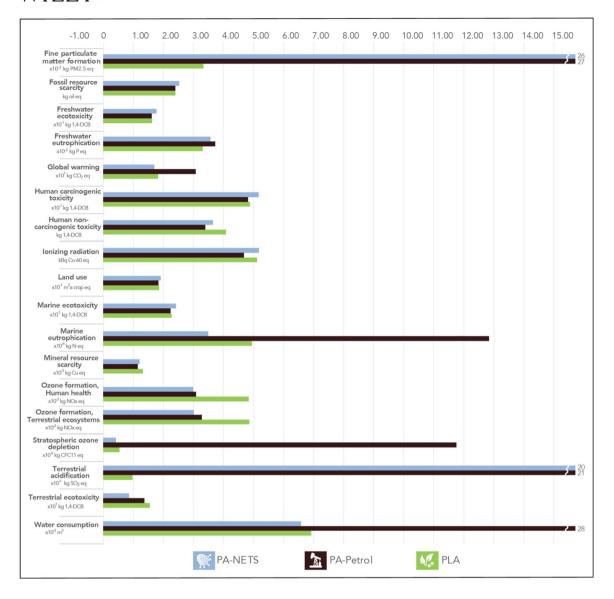
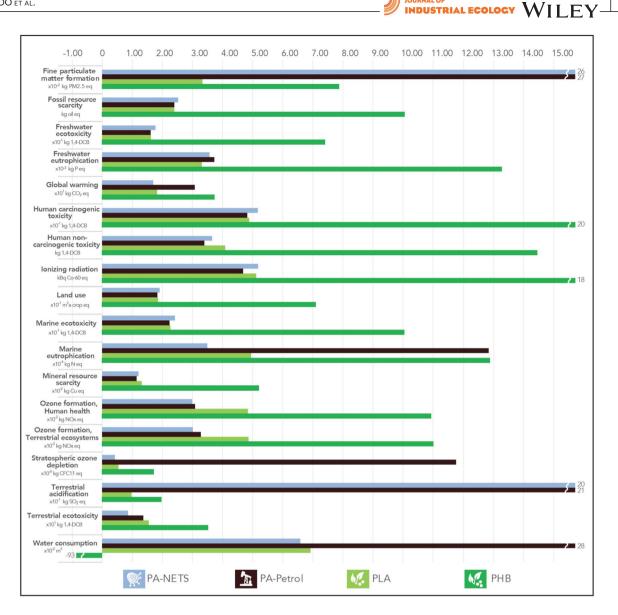


FIGURE 6 Environmental impacts regarding the life cycle of 1 kg of 3D printed depending on three scenarios: petroleum-based polyamide and PA originating from marine waste with thermal degradation EoL and PLA with composting EoL. Underlying data for Figure 6 are available in Table S8 of Supporting Information

These results can be explained by the amount of CO_2 absorbed by the biomass during its growth. For the second EoL alternative, the plastic wastebased polyamide material continues to show the lowest impacts (17.7 kg·CO₂ eq. for global warming). Moreover, the impact generated by PLA is similar to that of plastic waste-based polyamide (Kamau-Devers & Miller, 2020), and the highest is that of petroleum-based polyamide, contributing to 30.87 kg·CO₂ eq. Based on those results, we analyzed the potential of composting to lower the impacts of PLA (Teixeira et al., 2021). The main difference lies in the fact that while incineration of polyamide results in fossil CO₂ emissions that are not considered a renewable energy source (Mohn et al., 2012), the biogenic character of the CO₂ produced during PLA composting can be considered a renewable energy source. Moreover, terrestrial acidification and fine particulate matter formation impacts are reduced by using PLA.

As for terrestrial acidification and marine eutrophication, PLA generates the smaller impacts, with 9.96×10^{-2} kg SO₂ eq. and 4.96×10^{-4} kg N eq., respectively. For marine eutrophication, on the other hand, the material with the highest impact is petroleum-based polyamide with a contribution of 1.28×10^{-3} kg N eq. Overall, bio-based polyamides behave similarly to PLA as they are advantageous regarding the impact categories, acidification and eutrophication. On the contrary, global warming is intensified when using PLA and bio-based polyamide, mainly due to the cultivation process that is carried out to produce the materials (Kamau-Devers & Miller, 2020).

Finally, fine particulate matter formation is analyzed as it is a significant contributor to death and disease globally, as it makes reference to the intake fraction of fine particles (particulate matter, PM2.5, particles with diameters below $2.5 \,\mu$ m) (Morão & de Bie, 2019). Here, PLA gives remarkably interesting results, as the impact in this category is lower than in the landfill EoL. Land use is an important environmental impact category



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FIGURE 7 Environmental impacts regarding the life cycle of 1 kg of 3D printed depending on four scenarios: petroleum-based polyamide and PA originating from marine waste with thermal degradation EoL and PLA and PHB with composting EoL. Underlying data for Figure 7 are available in Table S10 of Supporting Information

regarding bioplastics produced from plant-based feed stocks. Generally, soil carbon stocks on land used for agriculture are generally lower than those in forests, so when conversion occurs on cropland, soil carbon stocks will decrease (Poeplau & Don, 2013).

3.3 | Environmental impacts of polyamide-based product with EoL thermal degradation and PLA and PHB with composting EoL: Scenarios 1-2-4-5

Once the environmental impacts of PHB are computed (see Supporting Information 3) for the evaluation of the production of PHB and the comparison with the production of other polymers, we proceed with the additional stages before 3D printing, which involve material transportation, conditioning though drying, filament formation, and a final drying step (Figure S1.2b). A composting scenario is considered as the most likely option, where methane, biogenic CO_2 , and water are formed as byproducts (Kliem et al., 2020; Naser et al., 2021). To gain a better overall perspective, obtained environmental impacts are compared with 3D printing processes using PLA, petroleum-based polyamide, and marine-waste polyamide (see Table S1.10). Figure 7 summarizes the environmental impact results obtained. Overall, the use of PHB generates larger environmental impacts in comparison with the other materials. However, smaller burdens are observed in the categories of water consumption, fine particulate matter formation, and terrestrial acidification. Analyzing global warming potential, the use of bioplastics generates the lower (PLA) and the highest (PHB)

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impact values, 18.2 kg·CO₂ eq. for PLA (similar to 17 kg·CO₂ eq. produced by the use of waste-plastic-based polyamide) and 71.2 kg·CO₂ eq. for PHB. The use of PHB might also be a handicap for marine ecotoxicity, human noncarcinogenic toxicity, and ionizing radiation. Besides, it is noticed that the use of polyamide obtained from recycled marine nets is the optimum choice to reduce marine eutrophication, stratospheric ozone depletion, and terrestrial ecotoxicity. No LCA results have been found in the literature considering a composting EoL scenario for PHB. However, the incineration with energy recovery has been assessed, with average reported values of 4.0 kg·CO₂ eq. for PHB and 3.5 kg·CO₂ eq. for PLA (Hottle et al., 2013; Kookos et al., 2019). For the production of 1 kg of material, global warming potential (GWP100) (kg CO₂ eq.) is reported in literature, showing the following results: for PHB: 2.3 kg·CO₂ eq., for PLA: 0.6 kg·CO₂ eq., and for bio-based PA 6:.4 kg·CO₂ eq. (Spierling et al., 2018). The results obtained from this study are much larger than those found in literature, which is understandable due to the complexity of the cycle studied in this work (production, transport, process, and EoL).

These a priori surprising results are explained by the technology readiness of PHB production, which results in small batches in comparison with the other materials considered as commodities. However, we estimate that PHB has a bright future to lower the environmental impacts of plastic products given its rapid biodegradability under both aerobic and anaerobic conditions and the formation of no harmful byproducts (in sharp contrast with materials from fossil origin) (Narodoslawsky et al., 2015; Rajan et al., 2017). In this sense, we expect that the environmental impacts of PHB production will be reduced as PHB technology evolves (Rajan et al., 2017).

To lessen the overall environmental impacts, new waste reutilization approaches should be explored in the near future. These include, for example, the use of polymers able to undergo a selective depolymerization process back to their initial constituent's feedstock (Lizundia et al., 2017; Shi et al., 2021). This way, the energy and time requirements for filament production (grinding the nets, drying, extrusion, bathing, and pelletization) could be reduced.

4 | CONCLUSIONS

Based on a circular economy perspective, here, we propose a solution to reduce the pollution caused by nonbiodegradable petroleum-based polymers floating on the ocean. To that end, waste originating from polyamide fishing nets is collected and processed through 3D printing into a new product. Cradle-to-grave LCA results reveal that the use of marine plastic waste as a feed material for a 3D printing process is environmentally preferred over virgin bioplastics derived from renewable resources such as bio-based polyamide (bio-PA), PLA, or PHB. By doing so, a 3.7- and 1.8-fold reduction in the global warming potential could be achieved when compared with bio-based and virgin petroleum-based polyamide respectively, markedly reducing the global warming potential of the plastic industry and representing a step forward on track to meet the 55% emissions reduction target for 2030. Besides the CO₂ eq. emission reduction, the use of marine plastic waste to produce fishing goods also encompasses lower impacts in 11 of the 18 categories analyzed. For the first time, we quantified the environmental impacts of PHB production through crude glycerol fermentation, demonstrating that widening the boundaries of the LCA, from the production process until the end of life, the environmental impacts such as fine particulate matter formation, terrestrial acidification, and water consumption can be reduced by using PHB as raw material. We estimate bio-based polymers represent an interesting alternative to lower CO2 emissions of produced plastic goods provided their implementation is optimized (agricultural resource optimization, use of renewable energy). As regard the EoL scenario, PLA and PHB are preferred as their composting results in biogenic CO₂, which can be then used in additional processes, for example, as a source of renewable energy. Precisely, its readily biodegradable character makes PHB a potential material for a zero plastic-waste society, although its production process needs to be optimized to avoid undesired environmental impacts. The results from LCA provide cues for further improvement, the use of renewable energy being highly recommended in the scenario where marine plastic waste is used as a feed. This work anticipates the potential use of polymeric waste as a feed material to manufacture brand-new products with reduced environmental impacts. (Figure S3, Figure S4, Table S11)

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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