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A framework for the assessment of marine litter impacts in life cycle impact assessment

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ABSTRACT

Purpose: Marine litter, mostly plastics, is a growing environmental problem. Environmental decision makers are beginning to take actions and implement regulations that aim to reduce plastic use and waste mismanagement. Nevertheless, life cycle assessment (LCA), a tool commonly used to assist environmental decision making, does not yet allow for considering the consequences of plastic waste leaked into the environment. This limits the application of LCA as a tool for highlighting potential tradeoffs between impact categories and the relative significance of their contribution on a specific Areas of Protection (AoP). A coordinated research effort to cover various parts of the marine litter impact pathway is required to ultimately produce characterisation factors that can cover this research gap. Here, we design a consistent and comprehensive framework for modelling plastic litter impact pathways in LCIA models. This framework is to support such coordinated research progress towards the development of harmonized pathways to account for impacts of plastic litter, specifically to the marine environment. The framework includes an overview of life cycle inventory requirements (leakage to the environment; a focus of other research efforts), and a detailed description of possible marine litter impact pathways, modelling approaches and data(-type) requirements. We focus on marine plastic litter and consider the potential contribution of different impact pathways to overall damage in the main operational AoPs, as well as recently proposed ones.

Results and conclusions: The proposed framework links inventory data in terms of kg plastic leaked to a specified environmental compartment (air, terrestrial, freshwater, marine) to six AoPs: ecosystem quality, human health, socio-economic assets, ecosystem services, natural heritage and cultural heritage. The fate modelling step, which includes transportation, fragmentation and degradation processes, is common to all included impact pathways. Exposure and effect modelling steps differentiate between at least six exposure pathways, e.g. inhalation, ingestion, entanglement, invasive species rafting, accumulation, and smothering, that potentially compromise sensitive receptors, such as ecosystems, humans, and manmade structures. The framework includes both existing, e.g. human toxicity and ecotoxicity, and proposed new impact categories, e.g. physical effect on biota, and can be used as a basis for coordinating harmonized research efforts.

1. Introduction

1.1. Background

Anthropogenic litter is accumulating in the marine environment and is associated with impacts on marine biodiversity (Li et al., 2016). Between 1997 and 2015 the number of species that are known to have

become entangled in or ingested marine anthropogenic litter doubled from 267 to 557 species (Kühn et al., 2015). In addition, such overview estimates likely underrepresent the prevalence of entanglement and ingestion due to underreporting in the scientific literature, e.g. Parton et al. (2019). Whilst entanglement and ingestion impact mechanisms operate at the level of individual organisms, effects may scale up to the level of species populations and ecosystems (Browne et al., 2015).

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Consequently, the provision of marine ecosystem services may be compromised (Beaumont et al., 2019). Furthermore, even in the absence of biotic damage, ecosystem services, particularly cultural services, can be affected due to the unsightliness of anthropogenic litter that distracts from natural beauty (Rangel-Buitrago et al., 2018). For example, Jang et al. (2014) observed reduced ecotourism associated with increased coastal litter pollution. Additionally, the potential for marine litter, specifically plastics, to end up in the human food supply may have consequences for human health (Wright and Kelly, 2017).

Globally, approximately three quarters of the marine anthropogenic litter composition (and more than 98% of micro-litter) is plastic (Tekman et al., 2019). Plastic originates from the technosphere, i.e. the human modification of the environment, and its production continues to increase at a rate of approximately 4% annually (PlasticsEurope, 2017), with plastics being used widely throughout a large range of product value chains (Andrady and Neal, 2009). Such an increase in plastic use is, in part, due to its versatility as a material, e.g. lightweight, mouldability and chemical- and light-resistance (Andrady and Neal, 2009), and the importance of such material properties for some applications e. g. in the health sector. Efforts to seek substitutive materials to fossilbased plastic have been recent and, in most cases, still lack competitiveness at a market level (Lettner et al., 2017). Moreover, many innovative biobased polymers do not necessarily show significant environmental improvements as compared to fossil-based polymers in many planetary boundaries (Escobar et al., 2018; Ita-Nagy et al., 2020). The current plastic *lock-in* for many economic sectors, coupled with the potential for inappropriate disposal of plastic waste, which can occur both before or after use, ultimately leads to plastic litter in the environment and environmental damage (Jambeck et al., 2015, 2018).

Attempts to reduce the generation of potentially environmentally damaging marine plastic litter have started recently, including in the EU, which has new rules on single-use plastics by 2021 (European Commission, 2019), and Canada (Environment and Climate Change Canada, 2019). The EU rules on single-use plastics have already started to be implemented by, for example, Portugal (DRE, 2020). The rules exemplify an application of both upstream interventions that reduce supply, e.g. a ban on single-use products made of plastic for which alternatives exist on the market, and downstream interventions, such as a 90% collection target for plastic bottles by 2029, and extended producer responsibility e.g. covering costs of clean-up (European Commission, 2019).

Sustainable development goal 14 "Conserve and sustainably use the oceans, seas and marine resources for sustainable development" further motivates the requirement for actions to stem the marine litter problem. For example, goal 14.1 targets a prevention and significant reduction in marine pollution of all kinds by 2025, including marine debris and nutrient pollution arising from land-based activities (e.g. van Puijenbroek et al. (2019)). For further discussion of SDGs in relation to plastic pollution, see, for example, Plastic Soup Foundation (2018).

Whilst it is becoming clear that marine plastic litter is a mainstream environmental problem, current strategies to reduce marine plastic litter may only reduce the rate of plastic litter accumulation in the ocean (Borrelle et al., 2020; Lau et al., 2020). Additionally, such strategies potentially result in trade-offs with other environmental problems and the magnitude of the impact of marine plastic litter on humans and ecosystems needs to be compared to other environmental impacts. Life Cycle Assessment (LCA) is a tool for supporting environmental decision making that is specifically designed to allow for the identification of these potential trade-offs between product alternatives e.g. a single use plastic cup versus a paper one, looking at a broad set of impacts on human health and ecosystems such as climate change, human and ecotoxicity, eutrophication and acidification. However, whilst LCA is widely used in support of decision making, approaches for considering a number of existing marine impacts in LCA methods remain generally lacking (Woods et al., 2016; Ziegler et al., 2016). This absence of assessment methods has become particularly apparent for marine plastic

litter, e.g. Schweitzer et al. (2018), and is recognised by the LCA research community as an area in need of urgent developments (Sonnemann and Valdivia, 2017). As such, whilst trade-offs between different plastic reduction options can already be assessed using LCA, an impact assessment approach for indicating the potential impact of residual plastic litter is still lacking.

However, research on the sources, fate and consequences of marine litter is still maturing. This presents both challenges and opportunities for extending life cycle impact assessment methods to account for marine littering. For this reason, a new international working group was formed in late 2018 (Boulay et al., 2019), supported by the Life Cycle Initiative hosted by UN Environment, as well as the Forum for Sustainability through Life Cycle Innovation (FSLCI), with the overall goal to "Foster the harmonized development of environmental impact pathways and characterization factors for marine impact assessment in LCA, in priority associated with marine litter, especially plastic" (marilea.org). The group, named MARILCA (MARine Impacts in LCA), aims first at proposing a harmonized framework for the developments of marine litter impacts in LCA, with a first focus on plastic. This framing paper is the result of this work.

1.2. Objectives

The main objective of this study is to design a consistent and comprehensive framework for modelling plastic litter impact pathways in LCIA models to ensure that the development of characterization factor models is harmonized and compatible with each other, which allows for a more holistic picture on plastic impacts in life cycle impact assessment (LCIA) methods. The aim is to draw a framework where we pinpoint the parts and aspects that need, in our current opinion, investigating. We hope that this can contribute to mutual discussions and collaborations among LCIA developers for the greater goal of a comprehensive approach for characterizing plastic impacts. This framework paper has emerged from an extensive review of scientific publications and data, put in relation with the LCIA impact pathway framework.

In the LCIA stage of an LCA of a product or service, an inventory of environmental interventions, i.e. a list of resources used and emissions produced in association with the life cycle of the product or service, is converted into indicators of environmental impact (Verones et al., 2017). This conversion is termed impact characterisation and is conducted using characterisation factors (CFs). CFs are resource- or emission- substance-specific and link to specific environmental problems (impact categories). CFs are constructed through impact pathway modelling that link e.g. a mass of emission (kg) to a metric of damage to an Area of Protection (AoP), and as such include consideration of e.g. the dispersion and lifetime of a substance in the environment and the potential for exposure and effects on sensitive receptors e.g. species that may suffer deleterious effects.

Here, we describe marine plastic litter impact pathways leading to damage to different Areas of Protection (AoPs), and, more specifically, identify the predominant impact pathways from plastic leakage to environmental damage. We first discuss key considerations related to plastic litter impact pathways before structuring these within a framework for the modelling of relevant pathways within LCIA. Our approach to identifying the main impact pathways is rooted in, primarily, a qualitative assessment of which impact pathways contribute most to 'leaked' plastic waste. Where possible, we discuss the potential contribution of different impact pathways to overall damage on each of the AoPs recommended by the UNEP-SETAC life cycle initiative (Verones et al., 2017): ecosystem quality, human health, natural heritage, cultural heritage, socio-economic assets and ecosystem services. Consideration of potential damage on the natural resources AoP is mostly covered by the assessment of upstream plastic manufacturing processes (Schulze et al., 2020) and is beyond the scope of the present framework focusing on marine litter.

Within the framework we comment on the strength of scientific

evidence supporting different stages of plastic litter impact pathways. In addition, we identify initial data requirements for the proposed modelling steps in the framework and relevant required levels of spatial differentiation and of litter / waste categorization in terms of e.g. fragment size and material type.

2. Key considerations surrounding marine plastic litter generation and impacts

In analogy to other life cycle impact categories such as human and ecotoxicity e.g. Rosenbaum et al. (2008), characterizing impacts of marine litter requires to define and understand the *elementary inventory flows* of plastic waste release to environmental compartments, the *fate* of plastic litter in the environmental (sub)compartments including transport, fragmentation, degradation and sinks, the *exposure* of sensitive receptors, and the *effects* of these exposures on species and ecosystems, human health or socio-economic assets. In the subsections below, we review and define the processes and pathways relevant to characterizing each step of the marine litter impact pathway.

2.1. Terminology

The first reports of floating plastic debris on the sea surface go back to the early 1970 s (Carpenter and Smith, 1972). Since then, the analysis of this environmental hazard has increased exponentially, emphasizing the growing need to categorize plastic debris in size classes, type of plastic or other parameters. Nevertheless, although marine plastic litter is commonly classified according to fragment size, a consensus nomenclature for size range of plastic objects is yet to be defined. Many authors adopt a stratified classification system comprising 5 size classes (UN Environment, 2018), i.e. mega-, macro-, meso-, micro- and nanoplastics. Simplistically, other studies consider two size classes, microplastics and macroplastics, with microplastics encompassing fragments less than 5 mm in size and macroplastics encompassing fragments greater than 5 mm. Moreover, in some cases, a distinction is made between small microplastics, below 1 mm, and large microplastics, from 1 to 5 mm. Regardless of the different classifications, there is a relatively wide consensus that the upper threshold for microplastics is 5 mm (Arthur et al., 2009; Lee et al., 2013; Eriksen et al., 2014; Pinto da Costa et al., 2016). As described by GESAMP (2015), this threshold allows assuming that most of the wide range of small particles below 5 mm can be ingested by a majority of marine biota, whereas increasing values above this threshold would only affect megafauna. In contrast, particles larger than 5 mm can also be a threat through other mechanisms such as entanglement (GESAMP, 2015), potentially leading to starvation (Lambert et al., 2014; Senko et al., 2020), increased energetic costs, or reduced mobility that may increase risk of predation e.g. (van der Hoop et al., 2014; Nunes et al., 2018).

In addition, it is important in an LCA context to distinguish between waste and litter. Waste is a broad term and is defined in the Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and Their Disposal as "substances or objects which are disposed of or are intended to be disposed of or are required to be disposed of by the provisions of national law" (Secretariat of the Basel Convention, 2017). Waste is therefore a product of disposal, which may remain within or be reused from an anthropogenic waste management system. Conversely, litter is within the environmental realm, outside anthropogenic control. For example, marine litter is "any persistent, manufactured or processed solid material discarded, disposed of, or abandoned in the marine and coastal environment" (UN Environment, 2019). In LCA terms, waste corresponds to a technosphere (intermediate) flow between a unit process and a waste management process, and litter corresponds to an elementary flow, i.e. release to the environment either directly from a unit process or via a waste management process.

2.2. Release of plastic waste into environmental compartments

At the life cycle inventory stage of LCA the aim is to quantify exchanges between the technological system and the environment for the functional unit (product or process) being assessed. For marine plastic litter, the transfer from plastic waste to plastic litter, or direct generation of plastic litter, constitutes the exchange between the technological system and the environment. This exchange occurs due to the mismanagement or inappropriate disposal of plastic waste, or directly from use-phase emissions e.g. the release of microplastic fragments from tire abrasion.

Plastic waste is released into the environment from both land- and marine-based sources, both directly and via waste management systems. Whilst land-based sources are considered the dominant source of marine plastic debris (GESAMP, 2016), the contribution from marine-based is still significant and underestimated e.g. (Xue et al., 2020). The United Nations Environment Programme (2018) estimated that 11.4% of marine macroplastic litter originates from marine-based sources (principally fisheries-related), with this figure becoming 7.2% with the inclusion of microplastics.

For waste that enters formal or informal waste management systems such as open dumps or landfills and wastewater treatment facilities (Axelsson and van Sebille, 2017), the release rate to the environment depends on both how closed/effective the waste management system is and the influence of environmental conditions. This results in spatially heterogeneous plastic release rates from plastic waste. For example, open dumps are now seldom used in Europe and North America, but in the case of Latin America and the Caribbean they represent 33% of total waste disposal (Hoornweg and Bhada-Tata, 2012; Margallo et al., 2019). The prevalence of open dumps is as high as 100% for Haiti and Suriname, 85% in Belize or 70% in Guatemala (Tello et al., 2010). Previous studies, some of which include a life cycle perspective, have demonstrated higher groundwater pollution levels close to open dumpsters as compared to sanitary landfills (Aiman et al., 2016; Han et al., 2016; Ziegler-Rodriguez et al., 2019). Nevertheless, recent studies also confirm the existence of microplastics in landfill leachates (He et al., 2019; Kazour et al., 2019). Direct littering, from either land- or marine-based sources, constitutes up to a 100% transfer from waste to litter. In urban environments the waste to litter transfer rate may be offset by technological measures such as street cleaning. Other sources with a 100% transfer from waste to litter include several sources of primary microplastics that enter the environment from e.g. tire abrasion or via untreated wastewater flows.

Jambeck et al. (2015) estimated the amount of plastic waste "available" to be released to the environment, e.g. plastic waste in open dumps or littered, using economic development as a proxy for capturing spatial differentiation in waste management standards. More specifically, they considered littering to be the sole source of litter in highincome countries, with waste mismanagement becoming the main issue in medium- and low-income countries. Nevertheless, these country-specific estimates of land-based plastic waste "available" to enter the ocean retain uncertainties and assumptions, particularly with respect to the involvement of environmental conditions in plastic release rates. For example, natural disasters and heavy rainfall events typically increase the rate of transfer from "available" plastic waste to environmental litter (Gabrielli et al., 2018; Moy et al., 2018). These can be seasonal and include those related to El Niño-Southern Oscillation (ENSO) and monsoons, that mostly occur in low and medium income nations with informal waste management, e.g. Li and Zeng (2003).

2.3. Fate: Transport, degradation, fragmentation and sinks of plastic litter in the environment

Once plastic is released into an environmental compartment, environmental fate processes determine the spatial distribution, residence time, and properties of plastic litter such as fragment size. Transport,

degradation and fragmentation processes are described below, including additional comments on the sinks from and accumulation in the "marine plastic litter system" specifically.

2.3.1. Transport

Plastic litter is transported within and between environmental compartments. Transport processes are therefore a key determinant of the residence time of plastic litter within an environmental compartment, and thus determining hotspots of plastic litter accumulation. Prominent transport vectors coupling the terrestrial compartment with the marine compartment, include wind, surface run off, and river systems (Schwarz et al., 2019). River systems ultimately deliver a large quantity of plastic to the oceans (Lebreton et al., 2017), especially after flooding events (Ryan et al., 2009). In the marine compartment plastic litter is further distributed by ocean hydrodynamics (Auta et al., 2017).

The relative importance of different transport processes in determining the spatio-temporal distribution of released plastic varies depending on the characteristics of the released plastic and the prevailing environmental conditions within the region where the plastic is released. For example, surface runoff is dependent on weather conditions, such as the frequency of high rainfall events, wind, and regional topography and vegetation (Jambeck et al., 2015). Furthermore, properties of plastic litter fragments, such as size, shape, and density, influence their transportability. Low density fragments are more buoyant than fragments of high-density polymers and therefore more susceptible to longer distance transport in ocean surface currents. In contrast, higher density particles will tend to sink and will accumulate in marine, estuarine and mangrove sediments (Alomar et al., 2016). Nevertheless, the original properties of plastic can be altered, such as biofouling causing previously buoyant plastic fragments to sink (Kaiser et al., 2017). An overview of transport processes within and between environmental compartments is provided in Table S1 (supporting information) and further discussed in Section 3.

Within the interconnected plastic distribution in the environment, various accumulation points have been observed. These include: sediment under freshwater lakes and soils (mainly fluvisols and agricultural land), subtropical ocean gyres (Eriksen et al., 2014), coastlines (Critchell and Lambrechts, 2016) and the seabed (Woodall et al., 2014). The presence of artificial barriers such as weirs and dams act as particle sinks in river systems (Mani et al., 2015). For marine litter impact assessment, accumulation of plastic in non-marine compartments effectively prevents plastic litter from reaching the 'marine-litter system'. Nevertheless, plastic fragments remaining in non-marine compartments potentially cause environmental damage that should be considered in future LCIA modelling efforts.

Transport processes begin once plastic fragments are released from the technosphere into the natural environment. To model plastic fragment transport processes it is therefore critical to understand where the limits are with respect to the technosphere, and apply this limit consistently across models. For example, to draw analogy with the case of pesticide emissions to the environment, the PestLCI model simulates the technosphere as a 50 m air column and a 1 m soil column for agricultural land, as well as the field borders. Beyond those limits, emitted pesticides are assumed to be entering the environment (Dijkman et al., 2012). Similarly, this could be considered a reasonable boundary for plastic litter from agricultural activities that are entering the neighbouring environmental compartments. In the LCA context, the priority is to achieve consistency in defining the boundary between the technosphere and the natural environment such that LCI modelling efforts are complementary and compatible with LCIA modelling efforts (see section 3).

Furthermore, plastic fragments of different sizes can be recovered from mismanaged waste through different types of technologies and infrastructure. Municipal street sweeping, beach cleaning or stormwater catchment devices are some examples of these technologies (Jambeck et al., 2015), and novel equipment is currently being implemented to

further the capture of plastic fragments from the environment, especially macroplastics directly from the ocean surface, e.g. The Ocean Cleanup (2020). While some of these technologies are clearly within the technosphere (e.g., street sweeping which prevents the waste to reach the natural environment), others, such as cleaning of marine litter in the intertidal zone of beaches, could be considered a direct intervention in the natural environment and, consequently, a sink of plastic from a plastic litter transport model.

2.3.2. Degradation and fragmentation

Over time, plastic pieces are affected by their environment and breakdown in smaller pieces. Whilst fragmentation occurs on land, in freshwater and at sea, e.g. Ryan et al. (2009), the rate of fragmentation is dependent on environmental conditions (Chamas et al., 2020). For instance, compromised structural integrity is a precursor for plastic fragmentation. UV radiation from sunlight triggers oxidation of the polymer matrix (Cole et al., 2011), which weakens chemical bonds, making plastic fragments more brittle and susceptible to fragmentation (Andrady, 2011). In addition, UV radiation, and associated photo-oxidation of the polymer matrix, may result in the leaching out of plastic additives from the plastics (Talsness et al., 2009), such that plastic fragments may contain lower concentrations of plastic additives over time.

Compromised structural integrity of plastics leads to faster fragmentation due to physical processes such as abrasion, wave-action and turbulence. Cold haline conditions (as found in large parts of the ocean), however, may prohibit this photo-oxidation (Cole et al., 2011). The fragmentation process is particularly prevalent in coastal zones due to direct exposure to sunlight, high oxygen availability and the presence of physical processes (abrasion, wave-action, turbulence) that enhance the fragmentation rate. Plastic fragmentation rates are therefore spatially heterogeneous.

Fragmentation, which results in increasingly small plastic fragments, can act as precursors to further biodegradation, since the surface area available for microorganisms is augmented. Still, fragmentation is usually triggered by abiotic processes and only later complemented with biotic pathways (Andrady, 2015; Chamas et al., 2020). The process of agglomeration can, in contrast, lead to an increasing fragment size, although the prominence of the agglomeration process relative to fragmentation is unknown. The process of agglomeration occurs as a result of biofilm formation on the surface of plastic fragments (Summers et al., 2018). The biofilm then acts as an adhesive, binding plastic fragments together and resulting in the (re)formation of larger plastic fragments (Summers et al., 2018).

2.4. Exposure of sensitive receptors

Plastic in the environment potentially interacts with sensitive receptors via exposure pathways. These exposure pathways determine the extent of occurrence of incidences, such as an entanglement event, that may contribute to compromising AoPs. Here we discuss the marine exposure pathways for the sensitive receptors: species and ecosystems, humans, and structures with instrumental or cultural value to humans.

2.4.1. Species and ecosystem exposure

Species and ecosystems can be exposed to marine environmental plastic litter via a variety of exposure pathways including ingestion, entanglement, smothering, and the rafting and introduction of invasive species. Exposure through *ingestion* is species-specific and demonstrated in a range of marine organisms (Kühn et al., 2015; Egbeocha et al., 2018; Karbalaei et al., 2019; Zhu et al., 2019). Different organisms are exposed via potentially ingesting plastic fragments within different size ranges. Filter feeders, deposit feeders and detritivores are known to ingest microplastic fragments (Thompson et al., 2004). Ingested microplastic fragments (0.08 mm to 0.67 mm) and fibres (0.21 mm to 4.90 mm) have been found in the deep sea invertebrates, *Ophiomusium*

lymani, a brittle star, and *Hymenaster pellucidus*, a star fish, at the sampling site Rockall Trough at a depth of 2200 m (Courtene-Jones et al., 2019). Over a four-decade study period the incidences of individuals containing ingested microplastics was on average 51% for *O. lymani* and 22% for *H. pellucidus* (Courtene-Jones et al., 2019). Larger marine species are also observed with ingested plastic particles larger than 5 mm (Bråte et al., 2016).

Furthermore, in addition to the ingestion rate being influenced by the environmental concentration and size characteristics of the environmental plastic fragments, the ingestion rate can be influenced by organism behaviour, e.g. feeding strategy. For example, whilst indiscriminate feeders, particularly lower-trophic level organisms, don't discriminate between plastic fragments and food, discriminate feeders select prey items and may mistake plastic fragments as prey items. In such instances, characteristics of plastic fragments, e.g. size, shape and colour, can also play a role in determining species ingestion rates. For example, it is suspected that turtles ingest transparent particles (resembling jellyfish) more frequently than e.g. blue particles (Schuyler et al., 2014), even though other studies contrastingly found more white particles being ingested (Eastman et al., 2020). Boerger et al. (2010) also reported preferences for white particles in planktivorous fish in the North Pacific. The position of biota in the water column and the colour of the debris can also influence its uptake (Santos et al., 2016).

Another important factor is the rate at which organisms release ingested plastic back to the environment. Combining knowledge of both the uptake and release rates of plastic fragments for a specific organism, the organism exposure at a given environmental concentration of plastic can be determined i.e. at a given environmental concentration of plastic, how much (internal) plastic are individuals of different species exposed to

Ecosystem exposure can be considered as exposure at the species-assemblage level. Wilcox et al. (2015) provide an example of this by developing a model to predict seabird exposure to marine debris. Wilcox et al. (2015) trained their model on species- and year-specific ingestion rates (from empirical research studies), species' geographic distributions and estimated marine debris for the period 1960–2010. They estimate, at a 1 decimal degree resolution, the number of bird species potentially exposed to ingested plastic. This therefore describes the incidence of ingestion at the seabird species-assemblage level, but not the ecological consequence of such an incidence of ingestion.

Coupled to the ingestion of plastic fragments is the potential for exposure to plastic additives and toxic substances transferred from seawater (Teuten et al., 2009) into organisms (Cole et al., 2011). Plastic additives are chemicals added to plastic during production and that may leach out into the environment and exposed organisms over time (Tanaka et al., 2020). The transference of chemicals from the surrounding environment to exposed organisms occurs via adsorption of chemicals onto plastic fragments that are subsequently ingested (Fred-Ahmadu et al., 2020). This may be particularly the case for microplastic uptake as microplastics have a high surface area to volume ratio that may allow for a high transfer of pollutants, such as persistent organic pollutants (Mattsson et al., 2015a, 2015b). With respect to the uptake of plastic additives by organisms, Garcia-Garin et al. (2020) detected the presence of seven different organophosphate flame retardants (OPFRs; chemical additives added to plastics) in the fin whale Balaenoptera physalus, a filter-feeding cetacean, and five OPFRs in samples of their main prey species, krill Meganyctiphanes norvegica.

The *entanglement exposure* pathway can be described similarly to the ingestion pathway in that species are exposed to incidences of entanglement at different rates due to the environmental plastic concentration and the behavioural response (or absence of response) to the presence of plastic fragments, i.e. 'indiscriminate' entanglements that occur randomly and 'discriminate' entanglements that occur, in part, due to organism behaviour. For example, juvenile Australian fur seals *Arctocephalus pusillus doriferus* appear to be exposed to a higher incidence of entanglement in plastic fragments than adult Australian fur

seals, perhaps due to higher curiosity in plastic fragments among younger individuals (Lawson et al., 2015). Alternatively, Lawson et al. (2015) also acknowledge that the more frequent entanglement of juvenile Australian fur seals could also be a function of body size coupled with mesh size. Entanglement rates are therefore likely to be a function of plastic shape and size, as well as organism behaviour and body size, which can change over time with organism development. Impacts from entanglements can range from mild pain to death and can also be caused in the long-term through blood loss and inflammations (Gall and Thompson, 2015; Dehnhard et al., 2019).

An additional exposure pathway on species is via *smothering*, whereby plastic fragments, particularly sheets, cover the seafloor and associated biota (Gregory, 2009). Whilst plastic debris settles on the seafloor at all depths, i.e. from intertidal to abyssal environments (Gregory, 2009), biota on the seafloor in deeper and still waters is particularly exposed to the potential anoxia and hypoxia induced by plastic smothering inhibiting gas exchange between water in sediments below the plastic and the sea water above (Gregory, 2009). This is because in deep and still waters plastic will smother the seabed for a long time, thus exposing species to the potentially anoxic or hypoxic conditions for a long time.

Additionally, ecosystems can be exposed to invasive species introduced through rafting on plastic litter fragments (Gregory, 2009; Rech et al., 2018). Some species, including bryozoans, barnacles and mollusks (Geburzi and McCarthy, 2018), attach to floating debris (Gregory, 2009), including plastic debris. This phenomenon is most observed in the case of larger fragments of plastic (Geburzi and McCarthy, 2018). Transport of floating plastic litter (see section 2.3.1) with organisms attached may accelerate the natural, but slow, process of trans-oceanic species dispersal on drifting natural flotsam (Gregory, 2009; Geburzi and McCarthy, 2018).

2.4.2. Human exposure

Humans ingest between 39,000 and 52,000 plastic particles per year (Cox et al., 2019). Some of these fragments are consumed via the marine environment. As mentioned in section 2.3.1 marine species can ingest plastic, either directly from the seawater or via the consumption of an organism already exposed to ingested plastic. Human exposure can then occur through the consumption of plastic-contaminated seafood. Further research efforts are required to better understand the trophic transfer of plastic fragments within the food web, including the potential for biomagnification or trophic dilution of plastic fragments and associated toxins (Akhbarizadeh et al., 2019), and the variety of human exposure routes e.g. (Revel et al., 2018). It should be noted that, whilst beyond the scope of impacts from marine plastic litter, humans can also be exposed to microplastics (and nanoplastics) through inhalation (Wright and Kelly, 2017).

2.4.3. Exposure of structures with instrumental or cultural value to humans Sites with natural beauty that confer economic or cultural value, such as beaches and coral reefs that support ecotourism, and other sites with cultural value, can be exposed to plastic litter through visual exposure. The presence and accumulation of plastic litter in the ecosystem can distract from the value-giving aesthetic properties of a species or landscape (Gregory, 2009). For example, Jang et al. (2014) associated a reduction in tourist numbers in 2011 at Geoje Island, South Korea, to increased marine plastic litter density around the island's coastline.

Additionally, man-made structures such as boats and fishing nets can be exposed to marine plastic litter through physical interactions. For example, Nash (1992) report entanglement of marine plastic litter in boat propellers and the fouling of operational fishing nets with plastic litter.

2.5. Effect or damage to Areas of Protection

Following the exposure of sensitive receptors there is potential for effects on or damage to AoPs. The consequences are determined, in part, by the effect/damage that stems from the exposure to plastic. These effect pathways include both physical and toxic effects on ecosystems and humans, and additionally the consequence of visual pollution on some ecosystem services.

2.5.1. Physical effects on species and ecosystems

Entanglement in, ingestion of, or smothering by plastic debris has potential consequences for individual organisms via physical effect pathways, e.g. Gregory (2009). For example, ingestion can impair an organism's feeding capacity by blockage of the digestive tract and/or false satiation. Impaired feeding can subsequently cause starvation and general debilitation, often reducing organism fitness, e.g. growth, reproduction and predator avoidance, or cause death (Gregory, 2009). In addition, ingestion of plastic fragments can cause internal injuries increasing the risk of infection (Seif et al., 2018). Similarly, the physical consequences of entanglement include death from starvation or debilitation, physical injury (internally or externally), increased risk of infection, compromised mobility, behavioural change, reduced growth and reproduction (Gregory, 2009). The effects of smothering by plastic litter are typically associated with sunken debris that covers the seafloor (Gregory, 2009). Such seafloor smothering may inhibit gas exchange between water in seafloor sediments and the water column, and thereby create anoxic or hypoxic conditions in benthic ecosystems and subsequent changes in ecosystem function and composition (Goldberg, 1997).

Whilst several hundred species are known to have been exposed to plastic via ingestion and entanglement (Kühn et al., 2015), the severity of the consequences is likely to be plastic-type- and dose-dependent, as well as species-specific and vary between individuals (Gall and Thompson, 2015). For microplastics, physical effects of microplastic ingestion are being explored in lab tests. However, ecotoxicological tests of microplastic impacts require procedural standardization (Connors et al., 2017). It is often difficult to distinguish in ecotoxicological studies the exact mechanism inside the organism which is responsible for death or altered functions.

Nevertheless, whilst the potential for sublethal to lethal effects of ingestion and entanglement is there, considerable uncertainty remains (Gall and Thompson, 2015). Moreover, Gall and Thompson (2015) further point out that whilst there will be a negative consequence at the level of individuals, defining the nature of sub-lethal effects, and the potential contribution of these impacts to individual mortality is complex.

Additional effect pathways are triggered because of how the plastic fragments alter environmental conditions. For example, smothering can expose species to hypoxic and anoxic conditions, although there is still limited bibliography on how species respond to these environmental changes. Similarly, biofouling processes in surface layers can generate important alterations in solar radiation in the photic zone (VishnuRadhan et al., 2019), which lead to changes in the warming and cooling patterns of the water column, and ultimately to a variety of effects on biota. Another example is that plastic debris change properties of sediment – increased permeability and decreased heat absorbance – such that sediment would reach a lower maximal temperature than in absence of plastic particles. Such temperature changes could have consequences for marine biota e.g. sex-determination in turtle eggs, and greater permeability leads to increased risk of desiccation in sediment-dwelling organisms (Cole et al., 2011).

Gall and Thompson (2015) conclude that "whilst it is inevitable that the biological and ecological performance of some individuals will be compromised, at present there is no clear evidence of population level consequences of encounters between plastic and marine life". Due to the difficulty associating changes in natural populations to single causative agents, a lack of evidence does not necessarily imply a lack of causation. As linking

effects to a single pressure is a prerequisite for a life cycle impact assessment model, at the current stage of knowledge on the physical effects of plastic on biota it may, therefore, be appropriate to adopt an approach similar to Browne et al. (2015), i.e. using basic principles to estimate how individual-level effects could scale up to ecosystem level effects.

2.5.2. Toxic effects on species and ecosystems

Toxic substances, from the leaching of additives from plastic fragments, and the transfer of extraneous pollutants into organisms (Cole et al., 2011), can have effects on organism development and survival (Hermabessiere et al., 2017). Organisms can also exhibit sub-lethal effects in terms of behavioral change. Seuront et al. (2021), for example, observed species-specific trait strengthening in four species of intertidal mussels, which could affect mussel bed characteristics and their suitability for supporting biodiversity. There is also the possibility of bioaccumulation and biomagnification of toxic substances such as POPs across trophic levels, although the efficiency of such transfers is not yet known (Andrady, 2011).

2.5.3. Invasive species effects on species and ecosystems

Non-native introduced species have the potential to become invasive and cause ecological damage (Pyšek and Richardson, 2010). Whilst the potential for non-native species introductions associated with the oceanic dispersal of plastic litter is increasingly recognized (Geburzi and McCarthy, 2018), the overall contribution of floating marine plastic litter to the regional spread of alien invasive species remains to be quantified (Rech et al., 2016). Nevertheless, to date, this species introduction pathway is considered unlikely to be a major introduction pathway, i.e. relative to other pathways, notably ballast water and hull fouling, but may contribute in some regions (Gregory, 2009; Gall and Thompson, 2015).

2.5.4. Physical and toxic effects on human health

Whilst exposure pathways have been documented, predominantly ingestion (Galloway, 2015), followed by inhalation and dermal contact (Rahman et al., 2021), the human health effects of microplastic exposure are unknown (Wright and Kelly, 2017). Potential effect mechanisms include both particle toxicity, which may enhance or induce an immune response, and chemical toxicity, i.e. effects due to additives, adsorbed environmental pollutants or the polymer itself (Wright and Kelly, 2017). More specifically, in humans, microplastic exposure may be linked with toxicity effects via oxidative stress, inflammatory lesions and increased uptake or translocation across membranes (Prata et al., 2020).

Though the effects of plastic exposure on humans are expected to be dose-dependent, a robust evidence base of exposure levels and effect mechanisms is currently lacking (Wright and Kelly, 2017). In addition, while knowledge on the toxicity of microplastic is still limited, an even bigger challenge is associated with the fact that microplastics may release their constituents with their own toxic effect (Groh et al., 2019).

2.5.5. Damage to socio-economic assets and ecosystem services

Damage to socio-economic assets and ecosystem services, i.e. the respective instrumental value of both manmade and natural systems to humans economically and socially, can be affected by both the presence of, and damaging interactions with, plastic litter. Examples of effects of marine plastic litter on socio-economic assets and ecosystem services are wide-ranging and include e.g. physical impairment and the loss of revenue caused by impaired fishing practices (Gregory, 2009), and the reduced economic value of sites due to visual pollution. For instance, Jang et al. (2014) estimated a US\$29 – 37 million tourism revenue loss in 2011 at Geoje Island, South Korea, following increased marine plastic litter around the island, and Rodríguez et al. (2020) estimated a cost equivalent to 0.02% of the Azores' GDP linked to direct impacts of marine litter on the nation's economic activities. In fact, fisheries productivity and tourism (including heritage and recreation) have been

recurrently highlighted as the two sectors that would suffer the most in terms of social and economic value loss (Fadeeva and Van Berkel, 2021). Nevertheless, Beaumont et al. (2019) have identified negative impacts on the provision of most marine ecosystem services, quantifying a reduced natural capital of between 3300 and 33,000 USD per additional tonne of marine plastic in terms of the 2011 ecosystem services values.

3. Overall framework, priorities and knowledge gaps

This section sets out a proposed LCIA modeling framework for plastic litter impacts, with a focus on marine litter (Fig. 1). This includes identifying to which new or existing impact categories in LCA plastic litter impacts would be associated. Plastic litter inventory data contributes via a variety of impact pathways to multiple impact categories potentially affecting six different Areas of Protection. Macro-, microand nano- plastic litter may be released to multiple environmental (sub-) compartments, with the marine compartment differentiated into subcompartments such as the surface, pelagic, demersal and seabed for coastal zones and the deep ocean. The fate describes the fragmentation and transportation of plastic litter within and between subcompartments. The exposure describes marine litter inhalation, ingestion, entanglement, rafting or accumulation and how litter can bioaccumulate via the food web or facilitate the transport of invasive species. Exposure-response characterizes the effects on species, ecosystems, humans and structures via physical, chemical, indirect (invasive species) or visual effects, whereas severity of each of these responses might enable to characterize and compare their respective damage on the relevant Areas of Protection.

The following sub-sections provide additional modelling considerations framing the possible approaches and data available for each of these steps, including the influence of environmental plastic on other impact categories, i.e. influence of plastic litter on the marine ecotoxicity and marine invasive species impact categories, this latter category needing to be further developed for LCIA. In addition, we provide a modified version of Fig. 1 in the supporting information (Fig. S1) to exemplify the framework using potential units, e.g. the time-integrated concentration of plastic in an environmental compartment (kg-yr/m³) resulting from fate processes and an inhalation rate (m³/yr) leading to human exposure.

3.1. Inventory modelling and data

Whilst inventory modelling is beyond the scope of this impact assessment framework, it is important to acknowledge the need for compatibility between impact assessment modelling and data derived from inventory modelling efforts. The Plastic Leak Project (PLP), a project with aims to compile LCI data for plastic litter, offers interesting insights by modeling two transfer processes: plastic loss and plastic release (Peano et al., 2020). Plastic loss refers to plastic leaving a product system, e.g. into a waste water treatment plant, but not necessarily the environment, and is akin to the generation of plastic waste. Plastic release refers to the transfer of lost plastic into the environment and is akin to generation of plastic litter, i.e. plastic released into the environment, and thus potentially leading to environmental consequences. It is the combination of loss and release, termed plastic leakage by the PLP (Peano et al., 2020), that is relevant for determining the life cycle inventory and subsequent impact assessment.

The PLP has generated initial LCI data for plastic litter, i.e. plastic flows from product processes into the environment, for a variety of process including tire use, textile manufacturing and use, and packaging production (Peano et al., 2020). This LCI data includes the initial release to three different environmental compartments, marine, freshwater and terrestrial, attributes information on polymer type, and uses regional data, where available, to assess loss and release rates within different regions (Peano et al., 2020).

Our modelling framework for marine plastic litter impacts is designed to start from inventory data expressed in kilograms (kg) of plastic leaked to a specific environmental compartment e.g. air, terrestrial, freshwater, marine. The exact definition of the technosphereenvironment boundary can be ambiguous, e.g. the limits of the soil and air columns in an agricultural system (see section 2.3.1). Nevertheless, it is of importance that the boundary defined is consistent in both LCI and LCIA modelling such that inventory data match characterisation factors without duplicated or absent modelling steps. In addition, inventory data should be attributed with details of location, receiving environmental compartment, material type and fragment size (i.e. inventory attributes; Table 1) to enable appropriate modelling of fate, exposure and effect processes, and align with the data capabilities in inventory modelling. Furthermore, it could be beneficial to distinguish between non-pyroplastics and pyroplastics, which are derived from the burning of plastic (Turner et al., 2019) and may have physicochemical properties different to the original polymer(s) that in turn

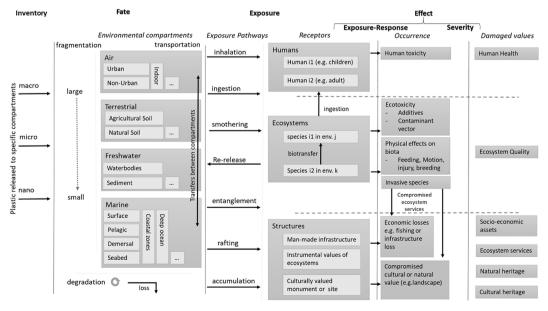


Fig. 1. An LCIA modelling framework for marine plastic litter impacts (explanations and details, see text).

Table 1

An overview of key properties influencing the potential environmental consequences of plastic litter and potential inventory attributes to capture the influence of these properties in LCIA modelling steps. Additional properties can also be relevant.

Property	Inventory attribute	LCIA modelling relevance	Classification	Why?
Location	Receiving environmental compartment and location	Fate	Spatial units for environmental compartments: air, terrestrial, freshwater, marine	Influences starting location of fate processes and ultimately the spatio-temporal distribution of a plastic litter flow
Primary fragment size	Primary fragment size	Fate, Exposure and Effect	Macro, micro, nano	Influences transport processes and impact mechanism possibilities
Density	Material type	Fate processes (transport)	Material type groups	Spatial distribution in the environment
Shape	Material type	Fate and Effect	Linked to industrial process origin	Influences object density (and therefore transport processes). Influences fragmentation rates (ter Halle et al., 2016). Influences effect mechanisms
Fragmentability	Material type	Fate	Material type groups	Longevity of plastic litter within each size class: associated with transportability and possible impact mechanisms
Degradability	Material type	Fate	Material type groups	Longevity of plastic
Toxicity	Material type	Exposure, Effect	Material type groups	Use of plastic additives, which potentially have different toxicities, differ between materials

alter fate and subsequent effects (De-la-Torre et al., 2021).

These inventory attributes would allow for capturing a range of plastic litter properties that influence LCIA modelling steps. Location could be given as a country of origin or other supranational areas or watersheds, among other options. Properties including density, fragmentability and toxicity could be captured by grouping polymers into a classification system for material types. Material type groups would effectively link specific polymer types to characterisation factors calculated with parameter values that correspond to a set of polymer properties. As such, different polymer types would need to be classified into material type groups before the application of characterisation factors. Material type groups can be defined based on the importance of different material properties, e.g. resistance to fragmentation, degradability, and buoyancy (Andrady, 2015; Ryan, 2015), over the different LCIA stages (fate, exposure and effect). These properties influence both residence time in a compartment and longevity within a fragment size class.

Fragment size at the time of leakage to the environment is the primary size, e.g. primary microplastics are less than 5 mm in diameter at the time of release to the environment. Primary microplastics therefore include, for example, those used in facial-cleansers and cosmetics (Zitko and Hanlon, 1991), virgin plastic production pellets (Andrady, 2011), and e.g. plastic fragments released into the environment from tire abrasion. For the case of life cycle methodologies, as shown in Fig. 2, we recommend adopting the threshold of 5 mm to distinguish macro- from microplastics. This aligns with existing plastic litter classification systems and will therefore simplify inventory data collection from existing datasets. In addition, GESAMP (2015) suggest that particles sized smaller than 5 mm diameter are readily ingestible by a range a biota, whereas particles larger than 5 mm are predominantly a threat via other mechanisms such as entanglement. In addition, a 5 mm lower threshold for macroplastics would include plastic fragments potentially causing visual pollution. Whilst at-sea visual surveys of plastic litter typically include a minimum fragment size of 25 mm (Lippiatt et al., 2013),

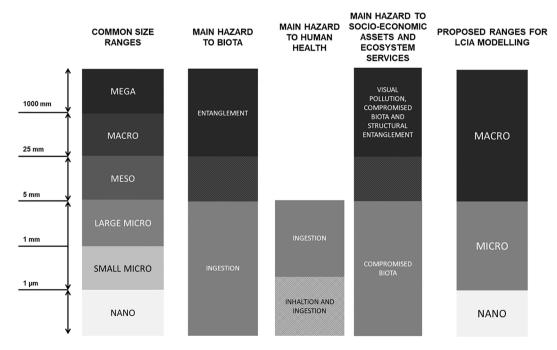


Fig. 2. Size class categories for application in LCI and LCIA, and approximate correspondence to common size class ranges, and impact pathways leading to damage on biota, human health, and socio-economic assets and ecosystem services.

fragments smaller than 25 mm could still contribute to visual pollution, particularly in high enough concentrations. To divide microplastics from nanoplastics we recommend a threshold at 1 μ m. This threshold is linked to the fact that nanoplastics may be ingested by organisms that are at the base of the food web (Pinto da Costa et al., 2016). In addition, 1 μm approximately aligns with the finding that inhaled particles smaller than 1–3 μm can potentially diffuse deep into lung tissue (Thomas, 2013). Nevertheless, fragment size in relation to human health impacts via inhalation may need to be further distinguished such that treatment of potential impact mechanisms aligns with, for example, characterizing impacts of inhaled particulate matter in the size-range 10-2.5 μm separately from particles smaller than 2.5 μm (Humbert et al., 2011). As depicted in Fig. 2, we recommend for LCI data to consolidate the six common size class ranges (see section 2.1) into a three class system of macro, micro and nano plastic litter, also adopted by PLP (Peano et al., 2020), which also approximately aligns with size-relevant impact mechanisms for consideration in LCIA modelling of plastic litter impacts. Whilst there would be a variety of sizes within each size-class, which would introduce uncertainty, this size-class simplification can sufficiently depict the main cause-effect chains of plastics in the marine

3.2. Fate

An LCIA fate model for plastic litter needs to characterize the environmental distribution and longevity of plastic litter within spatially-defined environmental compartments and sub-compartments i.e. the time-integrated concentration of plastic litter within environmental (sub)compartments. A fate model should include a representation of fragmentation, degradation and redistribution between (sub)compartments (Fig. 1).

3.2.1. Transport

Transport modelling is based on transfer rates between environmental compartments. During a transfer the plastic type and fragment size would stay the same. Transfer rates are specific to different combinations of environmental compartments, plastic types and litter size classes, and whilst the transfer process occurs over time, for an individual litter fragment the transfer, e.g. from air to water, could be assumed to take place in a moment brief enough for no degradation or

fragmentation processes to act.

The overall fate model needs to be subdivided into different main environmental compartments (i.e. air, terrestrial, freshwater, marine), such that relevant advective processes such as wind patterns, river flows, and ocean hydrodynamics, can be captured appropriately. Dispersal of plastic litter fragments within compartments, and across subcompartments (e.g. surface, pelagic, demersal and seabed in coastal and deep ocean areas of the marine compartment), should be captured and transfers between compartments (and sub-compartments) taken as outputs from each, contributing as inputs in the others (see Fig. 3). For example, consider a flow of macroplastic litter released into the terrestrial compartment. During a heavy rain event this macroplastic litter could be washed into a nearby freshwater river and subsequently taken out to the marine compartment. Over time, through fragmentation processes (see section 3.2.2), this macroplastic could be transferred into micro- and then nanoplastics. This example fate pathway is depicted on Fig. S2.

Considering the marine plastic litter problem specifically, fate model development should first focus on predominant (by mass flow) and best understood transport pathways. From our literature review (see section 2.3.1), these pathways are linked mainly to freshwater flows into the ocean, such as rivers and wastewater discharges, which account for considerable volumes of macro- and microplastics entering the ocean from multiple sources (e.g., cosmetics, road runoff, mismanaged waste). The air compartment is also an important pathway for microplastics entering the ocean, such as tire abrasion or city dust. Finally, direct ocean discharges due to fishing, cruise ship or marine freighting activities constitute an important pathway for both macro- and microplastics (Boucher and Friot, 2017, Schmidt et al., 2017). It should be noted, however, that these pathways identified as critical in terms of current mass flows may vary in importance across different regional or national contexts.

Nevertheless, the challenge remains for fate model developers to figure out how to model some flows identified in Fig. 3. In addition, Table S1 (Supporting Information) includes other transport pathways that may be relevant to include in future iterations of fate model development, some of which are not covered by research/evidence yet.

3.2.2. Fragmentation

Over time, fragmentation processes result in macroplastic fragments

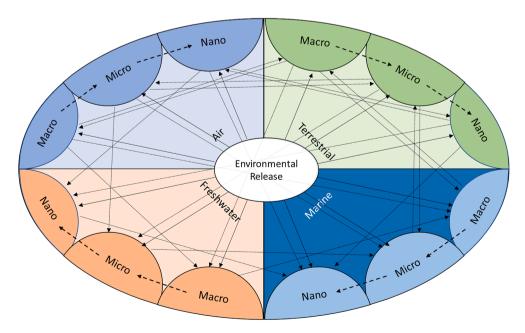


Fig. 3. Transport (dotted arrow) and fragmentation (dashed arrow) linkages between environmental compartments and plastic litter size classes of plastic litter released into the environment (solid arrow).

being broken down into microplastic fragments and, for some polymer types, microplastic can fragment further into nanoplastic fragments (as depicted on Fig. 3). Differences in fragmentation rate, influenced by material (polymer) type and environmental conditions (see section 2.3.2), should be captured by the combination of material type category (an attribute of the inventory flow) and environmental conditions, which can be attributed to the different environmental compartments and sub-compartments considered in the fate model.

Whilst agglomeration of smaller fragments into larger fragments can occur, e.g. (Summers et al., 2018), and the dominance of fragmentation processes to overall changes in plastic fragment sizes over time, inclusion of the fragmentation process in fate modelling is a more important first step, since it will be common to all the polymers entering the environment.

3.2.3. Degradation

Due to degradation processes, the total mass of a single release of plastic litter decreases overtime. Compartment properties, i.e. environmental conditions, influence the rate of plastic degradation (Gewert et al., 2015; Chamas et al., 2020). An appropriate classification of material types and environmental compartments needs to be determined to allow for distinguishing differences between types of plastic litter flow. For example, over a given time horizon, due to the faster degradation of biodegradable plastics than non-biodegradable plastics, the time-integrated concentration of biodegradable plastics in the environment would be less than for a non-biodegradable plastic.

Each polymer type will likely have a unique degradation rate in specific conditions (see Table 1 in Chamas et al. (2020)). The LCIA fate modeller needs to determine the appropriate material type resolution to capture differences in degradation rates between polymer types, whilst maintaining realistic data demands for life cycle inventory modelling. The material type resolution needed here will contribute to determining an appropriate material type resolution for the inventory. In the PLP, which includes an initial plastic litter fate step which overlaps in part with the fate step of our proposed LCIA framework for plastic litter, degradation rates for eight specific polymer types are specified without proposing a grouping of material types with similar properties (Peano et al., 2020). Ultimately, the fate model will include a best available estimate/average degradation rate to each material group.

3.2.4. Environmental compartments resolution

The sub-compartments in Fig. 1 are illustrative of the relevance of considering vertical compartments (i.e. surface, pelagic, demersal, and seabed) and the distinction between ocean hydrodynamics in coastal zones and the deep ocean. Nevertheless, the sub-compartment resolution of a fate model would be determined during model development. Additional (or different) sub-compartments may prove beneficial. An example could be the marine intertidal zone, which lies at the boundary of the terrestrial and marine compartments. Here, the flow of litter between transport models for the terrestrial and marine compartments could be aided by considering inputs and outputs to a shared compartment: the marine intertidal zone. In addition, a simplification of the fine line between the marine and terrestrial compartments along the coast may hide the impacts of plastic litter in beach, intertidal, estuarine and other coastal environments, areas with high concentrations of plastics which may trigger increased exposure for the resident biota.

3.2.5. Recapture of plastic litter

Within fate sub-models, i.e. covering terrestrial, freshwater, air and the marine environment, there is potential for technological solutions to remove plastic litter from the environment. For example, storm water drains can recapture plastic litter from the freshwater system. Beach cleaning or street sweeping in urban environments may also be considered as basic technologies for recovering plastics and other potential litter. Moreover, recent efforts have been developed to remove plastic fragments directly from the ocean through cleanup campaigns

and adapted technology for this purpose (Oliveira et al., 2019), although reducing the plastic stock in the ocean by 15% in 2030 would imply an expenditure equivalent to 1% of global GDP (Cordier and Uehara, 2019).

The rate of these 'losses or sinks' from the environment needs to be assessed and potentially included in plastic litter fate modelling. An important distiction, however, must be made between upstream preventive solutions to avoid plastic from reaching the environment and downstream palliative measures to remove plastic litter from the environment (Cordier and Uehara, 2019). For the former, which includes recycling, incineration and the improvement of waste disposal infrastructure, it can be considered that the plastic never crosses the system boundary of the technosphere and are, therefore, not considered releases to an environmental compartment. For the latter, plastic residues enter the environment and must be accounted for as such. Hence, to account for those plastic fragments that have reentered the technosphere after being released as plastic litter into the environment, we suggest to assign a negative impact flow to capture such techological processes, in a similar way to how carbon sequestration is accounted for in global warming potential metrics.

3.2.6. Distribution and mobility of plastic fragments over a variable time

For the quantification of time-integrated concentrations of plastic litter, a time horizon is required. Given the strength of the bonds within polymers, as well as the high molecular weight, plastics tend to be highly resistant to degradation, leading to high persistence when released to the environment (Welden and Cowie, 2017). Despite their persistence, these materials are subject to embrittlement and loss of mechanical properties, which ultimately leads to further fragmentation (Massey, 2006; Urbanek et al., 2018).

For instance, Ioakeimidis et al. (2016) analysed the behaviour of polyethylene terephthalate (PET) bottles over differing periods of time in the marine environment. Their results showed that PET appears to remain essentially unaltered in the marine environment for the first 15 years. Thereafter, there is a gradual decrease in the mechanical properties of PET. Acknowledging that not all polymers will show this temporal behaviour in the marine environment, and that interactions between plastics, marine biota and the rest of the marine environment are not fully understood at the present time (Kedzierski et al., 2018), it appears imperative for life-cycle metrics to differentiate between time horizons and analyse at which time the effects that plastic exposure will generate on marine biota and humans will occur.

Rather than arbitrarily calculating impact factors for a given time horizon, it might be more attractive, based on the chain of exposure and effects that the plastic particles would create, to differentiate between impacts that take place at different time scales (e.g. before 100 years and after 100 years) as proposed by Bulle et al. (2019) and implemented by Verones et al. (2016). Initially, damage caused by primary macro (and micro) plastics, close to the source of plastic leakage, will dominate the impact score. In contrast, when considering impacts occurring on a longer-term (e.g. after 100 years), the immediate consequences of plastic release into the environment are minimized, and longer-term impacts may be associated with areas of low exposure rates or in which data is difficult to obtain (e.g., the deep ocean). In contrast to GHG emissions, however, the mobility and fragmentation/degradation rate of the polymers will most likely alter the array of effects and damage that they may generate. That is because over time the distribution, mass and size-class profile of the plastic litter, as well as spatially-specific sensitive receptors, changes.

3.2.7. Additional fate considerations for invasive species and ecotoxicity

Fate processes for invasive species and ecotoxicity impacts are also based on transport processes of plastic fragments in the ocean. For invasive species, a fate model would additionally need to consider the attachment and release of organisms onto and from plastic fragments being transported in the marine compartment. For ecotoxicity, additive

leaching and the adsorption of toxic substances from the environment are additional considerations.

3.3. Exposure, effects and damage to Areas of Protection

Sensitive receptors are potentially exposed to plastic litter from the environment via a variety of exposure pathways. LCIA exposure modelling characterizes the extent to which sensitive receptors are exposed to a given environmental litter concentration via the relevant exposure pathways. It is this exposure to environmental litter that leads to the potential occurrence of effects that damage Areas of Protection (Fig. 1), rather than simply the presence of environmental plastic litter. In our LCIA framework for plastic litter we consider humans, organisms within ecosystems, and natural and manmade structures as potentially sensitive receptors. The LCIA framework for modelling the exposure of each of these sensitive receptors, together with the occurrence of potential effects and damage to Areas of Protection, is described in the subsections below.

3.3.1. Species (organism) exposure, effects and damage to Areas of Protection

3.3.1.1. Physical effects on biota. Ingestion and entanglement impact pathways both contribute to effects within this new proposed impact category (physical effects on biota; Fig. 1) to capture the physical impacts (external and internal) of plastic litter. As described in section 2.3.1, organism exposure via ingestion is a function of both uptake and expulsion of plastic fragments, processes that are organism-specific and influenced by the concentration of plastic litter in the environment. The potential for trophic transfers of plastic fragments up a food web is an additional potential consideration. Together, these processes determine the internal dose of plastic fragments an organism is exposed to at a given environmental concentration of plastic litter. Organism effects can then occur if this dose surpasses an effect threshold.

The entanglement exposure pathway describes the rate of entanglement events within a species population at a given concentration of plastic litter in the surrounding environment. The entanglement exposure rate at a given plastic litter concentration is species-specific and a function of organism size and behavior coupled with attributes of the plastic litter such as size and shape. Furthermore, the effects of entanglement occurrences (at a given rate in a population) are not equally detrimental to the entangled organism. Effects at the organism level depend on the consequences of entanglement in terms of e.g. reduced mobility, survivorship and reproductive rate. In LCIA, a species could be considered potentially affected at the environmental plastic litter concentration that results in a defined rate of entanglement events with specific detrimental organismal consequences occurring within an exposed population.

Both the ingestion and entanglement exposure and effect pathways could be characterized by life cycle impact assessment modelling approaches based on species sensitivity distributions (SSD; Posthuma and De Zwart (2014)). In other words, first determine the environmental plastic litter concentration at which species become affected. Then, after ranking these species in this sample by their sensitivity to environmental plastic litter, a cumulative distribution function can be calculated. A cumulative distribution function can be used to estimate the proportion of species potentially affected at a given concentration of environmental plastic litter.

Such an approach would link the environmental plastic litter concentration directly with a species-assemblage response (i.e. multiple species affected), thus combining exposure and effect modelling steps. Explicit modelling of exposure and effects would demand data and knowledge beyond that which is currently available. Nevertheless, species-sensitivity-based exposure and effect modelling for ingestion and entanglement currently faces data and knowledge challenges. For

example, current levels of knowledge on the consequences of ingestion or entanglement is often incomplete. The incidence of ingestion or entanglement in species has potential to cause effects, but the severity of these effects is often unknown. It has to be further studied whether such an effect factor of an environmental pressure is more easily defined in terms of a potentially affected fraction of species or in terms of a potentially disappeared fraction of species as for ecotoxicity (Owsianiak et al., 2019).

At the current state of knowledge, a lack of consistency between studies, and often specific spatial scale of studies, makes it difficult to apply available data in LCIA effect models and draw generic conclusions regarding entanglement and ingestion across species assemblages and regions. For example, individual studies each have their own specific aim, and therefore report different information. Gall and Thompson (2015) reported, for example, that not all reports of ingestion provide detail of the size or number of ingested items, the prevalence of ingestion within the population or the amount of harm caused by this ingestion.

The most studied of these impact pathways to date is perhaps the ingestion of microplastics, which have been the subject of several laboratory studies for a range of organisms. Also here, there is a lack of consistency in the data (Connors et al., 2017). Laboratory experiments typically use virgin plastic, i.e. without additives, to isolate the physical effects of ingested plastic (though perhaps a small fraction of the effect would be caused by "toxicity of the polymer itself"). These laboratory tests can observe the impairment of biological function, such as reduced motility and fitness, but most studies so far do not identify the specific pathway causing the effect, and effects occur via several effect mechanisms, several of them physical. Nevertheless, these data are detailed enough for informing the generation of an SSD, which includes exposure and effect pathways implicitly. To enable the generation of the SSDs, the test statistics for each species included need to be appropriately comparable - this is something for the LCIA model developer to determine and the requirements for inclusion may become stricter as more and better data become available, e.g. through Connors et al. (2017).

A further challenge pertains to species representation and spatial coverage. Reports of entanglement and ingestion are "most commonly made for sea turtles, marine mammals and sea birds" (Gall and Thompson, 2015). Often, studies and reviews focus on certain spatial areas and concentrate on single or few species groups, such as pinnipeds (Jepsen and de Bruyn, 2019), turtles (Duncan et al., 2017), birds (Ryan, 2018), sharks and rays (Parton et al., 2019). However, there is a "lack of reports considering low trophic level organisms" (Gall and Thompson, 2015). Furthermore, there is a "geographical reporting bias" (Gall and Thompson, 2015). The first species-sensitivity based effect factors (incorporating both exposure and effect) are therefore likely to be modelled on patterns and observations from organisms using coastal shelf regions and surface waters. The response of these species will enable representation of plastic litter effects in LCA, but with the caveat that potential damage to deep sea ecosystems is still neglected.

3.3.1.2. Other impact categories: Ecotoxicity and invasive species. For completeness, our framework for including plastic litter impacts in LCIA includes the contribution of plastic litter to two other impact categories linked to impacts on ecosystems: ecotoxicity and invasive species. The contribution of plastic litter to the ecotoxicity impact category is associated with plastic litter ingestion. Further development of marine ecotoxicity impact models would be required to incorporate both new substances, i.e. plastic additives, and potentially enhanced exposure due to the adsorption of toxic substances on the surface of plastic fragments taken in by organisms. Such model developments could be included in the marine ecotoxicity modelling as either new substances or a parameter influencing the toxicity exposure pathway. With respect to the influence of the presence of plastic litter on marine toxic exposure, this could be achieved in the first instance by incorporating a background concentration of plastic litter in the ecotoxicity exposure model.

However, in such a case, toxicity impacts would still be attributed to elementary flows of toxic substances rather than characterizing the marginal change in toxicity impact due to a change in the background concentration of plastic litter.

With respect to invasive species, the redistribution of plastic may inadvertently expose ecosystems to novel species introduced via the 'rafting' species introduction pathway. In the event a species is introduced to a 'new' ecosystem, this could expose the receiving ecosystem to threats associated with invasive species. It remains unclear how ecosystem impacts of invasive species could be quantified/modelled in LCIA. To date, the only LCIA model for invasive species impacts pertains to invasive species impacts associated with shipping in the freshwater Rhine-Danube waterway (Hanafiah et al., 2013).

3.3.1.3. From species effects to damage to Areas of Protection. Effects on species contribute to damage to several Areas of Protection. Ecosystem quality is directly damaged by a potential reduction in species diversity. Indirect impacts could occur on ecosystem services (e.g. reduced provisioning services), natural and cultural heritage (e.g. loss of a culturally important species) and human health (e.g. via subsequent human consumption). These indirect connections are indicated by arrows on the overall framework figure (Fig. 1).

3.3.2. Human exposure, effects and damage

Although it has been shown that humans are exposed to plastic via food consumption, as well as freshwater consumption and inhalation, human toxicity effects are not yet demonstrated, but should be considered and further investigated. Research needs to mature further before the first LCIA modelling approaches can be developed. Incidence of diseases associated with plastic exposure first need to be detected, and their severity subsequently assessed in terms of Disability Adjusted Life Years, analogous to other human health impacts categories (Fantke et al., 2019). For human toxicity effects of plastic ingredients, additional knowledge is needed to either directly determine exposure—response for different types of plastic or assess the fraction of plastic ingredient released in humans and their bioavailability.

3.3.3. Natural and man-made structures: exposure, effects and damage to emerging Areas of Protection

In some cases of natural and man-made structures, the accumulation of plastic within a defined site/area defines the exposure process. This could be a one-to-one translation from the output of the fate model i.e. the time-integrated plastic litter concentration e.g. in (visible) surface waters of a culturally or economically valued site, specifically for its aesthetics. In other cases, particularly with manmade structures, the exposure pathway becomes more about e.g. physical entanglement such as the cases of plastic litter becoming entangled in fishing boat apparatus or underwater tidal turbines.

Effects on these sensitive receptors are diverse and include economic losses from, for example, fisheries, infrastructure loss, and reduced tourism. These effect pathways ultimately lead to damage to socioeconomic assets.

In addition, the presence of plastic litter can compromise a natural value e.g. the intrinsic beauty of a landscape and lead to damage to the natural heritage AoP. In such a case it would be the visible plastic that is potentially damaging the natural heritage or cultural value. The final damage to the cultural or natural heritage would depend on how much this location was valued by humans for its nature or culture and how much the (visible) plastic detracts from this.

3.3.4. Natural resource depletion

It should be noted that in the present study natural resource depletion, which is an Area of Protection typically assessed in LCA studies, was not included. For plastic, impacts on abiotic resources are mostly covered by the assessment of upstream plastic manufacturing processes

and focus on the removal of natural resources. Such a modelling pathway is independent from the plastic litter impact pathways we included in our framework. Nevertheless, there is a potential overlap with respect to recycling of potentially recaptured marine litter that could be considered in the future as substituting raw natural resources.

4. Conclusions

The framework we present in this manuscript provides an overview of modelling steps and considerations for the development of characterisation factors for plastic litter impacts. More specifically, the framework includes impact pathways from plastic leakage to indicators of damage on the AoPs: ecosystem quality, human health, natural heritage, cultural heritage and socio-economic assets and ecosystem services. Our qualitative assessment of the literature indicates that elements of fate modelling, particularly transport processes within the marine compartment, together with effect modelling related to microplastic ingestion and to entanglement in macroplastic, have to date received most research attention. As such, it is currently more feasible to model those impact pathways that link plastic litter with damage on ecosystem quality. In addition to the increased knowledge and evidence developed for these impact pathways to ecosystems damage, this AoP is well established in LCIA, which is an advantage in terms of harmonized model development across research efforts in the near term. Human health is also a well-established Area of Protection in LCIA. However, knowledge of the potential for, and importance of, deleterious consequences of plastic exposure on human health is yet to be established. Further primary research is required before the necessity for LCIA modelling for human health impacts can be assessed and then, if required, developed.

The AoPs socio-economic assets, ecosystem services, natural heritage and cultural heritage are not yet operational in the LCIA framework (Verones et al., 2017). In addition to understanding the underlying exposure and effect mechanisms leading to damage on these AoPs, a concurrent discussion is required within the broader LCIA research community to fully establish endpoint metrics for these. Consideration of plastic litter exposure and effects, i.e. novel suggestions for endpoint metrics, for these AoPs offers an opportunity to contribute to this process.

Overall, diverse expertise and data are required to fully cover the modelling steps described. As such, and in light of the urgency for expanding the LCIA toolbox to include methods for quantifying indicators of damage associated with plastic litter, the framework presented can be used as a basis for coordinating harmonized research efforts between research organizations, a task that the MARILCA group, hosted by the UN Environment Life Cycle Initiative and FSLCI, is undertaking (https://marilca.org/).

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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Appendix A. Supplementary data

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