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Full length article

Microplastic aquatic impacts included in Life Cycle Assessment

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ABSTRACT

Although Life Cycle Impact Assessment (LCIA) methods assess a wide range of environmental impacts, ecological impacts of plastic pollution are not commonly included. Here, characterization factors of Polypropylene (PP), Low density polyethylene (LDPE) and Polyethylene Terephthalate (PET) microplastics were assessed. Fate was assessed through the multimedia fate model Simplebox4Plastics. Ecological effects were based on species sensitivity distributions. Macroplastic impacts were included though a conversion fraction. The characterization factors were included in ReCipe2016 method and applied to two consumer packaging films to show the relevance of including plastic pollution in LCAs. Plastic losses were assessed using material flow analysis. The freshwater and marine ecotoxicity midpoint indicators were dominated by plastic pollution impacts, whilst these impacts were limited on ecosystem quality as endpoint. Extending this methodology to additional polymers and additional methodological developments will help to obtain a more complete picture of plastic pollution in LCA and to identify effective mitigation options.

1. Introduction

Environmental plastic pollution is abundant all over the planet (Alvarez-Zeferino et al., 2020; Lechner et al., 2014; Qadeer et al., 2021; Ryan et al., 2009; van Sebille et al., 2015). While designed for durability, plastic products can fragment during its life cycle and form smaller particles of the same material (Andrady and Neal, 2009; Chamas et al., 2020). These fragments and derivates are lost to the environment (Kawecki and Nowack, 2019; Ryberg et al., 2018; Schwarz et al., 2023). The sizes of plastics are defined as macroplastic (MaP) (>5 mm) and microplastic (MiP) (<5 mm - > 1 μ m) (Andrady, 2011; Quik et al., 2023). Plastic pollution can negatively affect ecosystems and human health through various impact pathways. Plastics can have external physical effects when species become entangled or smothered by MaP (Gall and Thompson, 2015; van Bijsterveldt et al., 2021; Woods et al.,

2021). Internal physical effects can occur when ingested by species, where it can block the digestive system, lead to lower energy availability and can even be lethal (Gall and Thompson, 2015). Ingested MiP can move up in the trophic system leading to bioaccumulation in higher predators (Miller et al., 2020). MaP and MiP also pose negative effects in terrestrial environments, affecting soil health and plant growth (Gao et al., 2022; Huerta Lwanga et al., 2016).

To quantify the environmental impacts of a product over the full life cycle, Life Cycle Assessment (LCA) is used. LCAs do not commonly include the impacts from plastic pollution which may results in a potential underestimation of impacts of plastic products in an LCA. For inclusion, data about mass loss quantities to the environment are required for inclusion. In several LCA studies, plastic mass loss estimates to the environment have been included per functional unit (Galafton et al., 2023; Loubet et al., 2022). These mass loss flows can be defined

Abbreviation: MiP, Microplastic; MaP, Macroplastic; LC(i)A, Life cycle (impact) assessment; FF, Fate Factor, in days; EF, Effect Factor, in m^3/kg ; CF, Characterization factor; ETP, Environmental toxicity potential; PDF, Potentially disappearing fraction of species; PAF, Potentially affected fraction of species; DCB-eq, Dichlorobenzene equivalents; M, Mass, in kg; HC50, Hazardous concentration where 50% of species is affected; LC50, Lethal concentration for half the test subject; EC50, Half maximal effective concentration; k_{frag} , Fragmentation rate constant (S⁻¹); k_{min} , Mineralization rate constant (S⁻¹); k_{deg} , Degradation rate constant (S⁻¹), sum of $k_{frag} + k_{min}$; LDPE, Low density polyethylene; PP, Polypropylene; PET, Polyethylene terephthalate; I, Individualist; H, Hierarchist; E, Egalitarian.

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using Material flow analysis (Kawecki and Nowack, 2019; Schwarz et al., 2023). Furthermore, cause-effect pathways in life cycle impact assessment (LCIA) methods are required for plastics to summarize the ecological impact per unit. This is done using a substance characterization factor (CF), addressing both environmental fate and ecological effects on species of the substance (Huijbregts et al., 2017). For MiPs, environmental fate has been assessed by including sedimentation and degradation (Corella-Puertas et al., 2023; Maga et al., 2022; Malli et al., 2022). The MiPs ecological effects are typically based on species sensitivity distributions, using acute or chronic physical toxicity data from laboratory tests on single species (Lavoie et al., 2022; Li et al., 2023; Loubet et al., 2022). Combining fate and effects result in CFs of plastic pollution, as reported by a number of studies (Corella-Puertas et al., 2023; Croxatto Vega et al., 2021; Maga et al., 2022; Salieri et al., 2021; Saling et al., 2020; Zhao and You, 2022). Unfortunately, these CFs are so far not included LCIA methods, such as ReCiPe2016. Hence, the importance of plastic pollution compared to other stressors, such as chemical pollution or climate change, is missing from literature. Moreover, the environmental fate assessment of MiP is to be further improved by including other relevant fate processes for MiPs, such as heteroagglomeration and intermedia transport from land and freshwater to marine environments.

In this study, CFs were obtained for MiP and MaP for three polymers (LDPE, PP, PET), aligned with impact categories marine and freshwater ecotoxicity in the LCIA methodology ReCipe2016. To apply and assess the extended LCIA methodology, a comparative LCA is performed for two case studies of multilayer packaging films for consumer products. The results from this study may support structural and comprehensive inclusion of plastic pollution impacts in LCAs, as well as an increase in awareness and understanding of impacts of plastic products through pollution.

2. Methodology

2.1. ReCiPe2016 methodology

The ReCiPe2016 methodology addresses impacts through Characterization factors (CFs) on two levels: midpoints and endpoint. Midpoint CFs focus on single environmental problems, such as climate change, whereas endpoint CFs show the environmental impacts on a higher aggregation level, such as effects on biodiversity. In total, ReCiPe2016 includes 17 midpoint and 3 endpoint categories (Huijbregts et al., 2017). Additionally, ReCiPe2016 includes three cultural perspectives based on the cultural theory of Thompson 1990 (Huijbregts et al., 2017; Thompson et al., 1990). The choice for cultural perspectives includes assumptions on multiple aspects of data and impacts, such as time horizon for exposure, area of impact and required level of certainty for a specific impact. Integration of plastic pollution in the ecotoxicity CF is chosen, as the impacts of plastic pollution can be derived with the same type of models and lab experiments as the ecotoxicological impacts of chemicals. Ecotoxicity is covered in 3 different midpoint categories reflecting different ecosystems, i.e., freshwater, terrestrial and marine.

2.2. Microplastic characterization factors

The Ecotoxicity Potential (ETP) for a MiP of polymer (x) is the compartment specific midpoint characterization factor (CF) and is derived using the standardized equation in ReCiPe2016 for ecotoxicity (Eq. (1)) (Huijbregts et al., 2017). The FF is the fate factor (in years) for the receiving compartment j of polymer x emitted to compartment i. The EF is the effect factor in the receiving compartment (j), with unit Potentially Disappearing Fraction of Species (PDF) m³ kg⁻¹. The FF and EF of polymer x are divided by the reference flow for dichlorobenzene (1.4-DCB) to obtain the required units for ReCiPe2016 (SI 1.7). This is done for all cultural perspectives (c). The cultural perspective affects the time horizon of exposure, where 'Individual' (I) covers 20 years, the

'Hierarchist' (H) 100 years and 'Egalitarian' (E) on an infinite time horizon.

$$ETP_{MiPx,i,j,c} = \sum_{g} \left(\frac{FF_{x,i,j,c} \times EF_{x,j,c}}{FF_{DCBj,c} \times EF_{DCBj,c}} \right)$$
(1)

It was assumed all aquatic MiP are bioavailable, often expressed as an exposure factor (XF). Here, the XF is omitted as it is assumed to be equal to 1, like other LCA studies (Corella-Puertas et al., 2023). The endpoint CFs refer to ecosystem quality and hence require a translation from midpoint to endpoint. For that, the same midpoint-to-endpoint conversion factors for ecotoxicity are applied as reported in the Recipe2016 framework (Huijbregts et al., 2017).

2.3. Microplastic fate factors

Multimedia fate models are used in LCA to determine the fate factor (FF) of a chemicals released to the environment. The resulting FFs refer to the net residence time of a substance in the compartments which are assessed. Recipe2016 used the multimedia fate model USES-LCA 2.0 to quantify FF of chemicals which uses an adaptation of the Simplebox3.0 model (Hollander, 2004). Similarly, UseTOX can be used to assess fate factors for chemicals in LCA. Both Simplebox and UseTOX are developed for chemicals and not directly applicable to particle fate assessment.

The recently published simplebox4plastics (Quik et al., 2023), an adapted variation on the Simplebox3.0 model, was used to calculate FFs for MiP following the USES-LCA 2.0 modifications (Van Zelm et al., 2009). The initial emission compartments required (natural soil, agricultural soil, ocean, freshwater river & freshwater lake) were selected on the continental scale. Receiving compartments are freshwater and marine water at the continental and global scale. The regional scale was excluded by setting the Water Flow-continental Sea water and airflow continental-regional to regional sea water to zero. This adaptation aligns with USES-LCA 2.0, where emissions are summed up per pollutant regardless of their geographical place of occurrence, as happens in most LCAs (Huijbregts et al., 2005). Further modelling details are described in SI 1.3. The steady state version (i.e. a state of equilibrium) of Simplebox4plastics was used to derive the FFs through dividing the mass over the total volume of the compartment. These FF are a result for an infinite time horizon, which corresponds to the E-perspective. The FFs for 20 years, and 100 years (corresponding to the I-perspective and H-perspective, respectively) were obtained with the dynamic Simplebox4plastics Rshell, which was set at a time interval between 1 and 100 years. The concentrations after respectively 20 and 100 years of continuous emissions were taken from the assessment and used for the I and H horizon, following the flux-pulse solution proposed by Heijungs

Within Simplebox4plastics a standard particle properties dataset is available (Quik et al., 2023). For PP, LDPE and PET in this study, polymer density, particle size, fragmentation rate constant (kfrag) and mineralization rate constants (kmin) were collected for different environmental compartments. To describe the removal processes accordingly, the term degradation is used to illustrate environmental removal processes of MiP particles, which includes both fragmentation and mineralization. Mineralization is defined as the disappearance of MiP polymers into hydrocarbons that cannot be considered plastic. The term fragmentation refers to the size reduction of MiP to sizes $< 1 \mu m$ (nanoplastics). Studies that report on fragmentation of plastics primarily refer to MaP fragmentation, and exclude MiP fragmentation to smaller particles. Due to this data limitation, the fragmentation rate of MiP was assumed to be similar to the fragmentation rate of MaP. This assumption follows other LCA and modelling studies (Corella-Puertas et al., 2022; Koelmans et al., 2017; Maga et al., 2022). Further details on data collection and Simplebox4plastics are described in SI 1.1-1.3.

The FFs were obtained for all emission compartments to obtain an emission matrix with receiving and emitting compartment (freshwater,

ocean water, industrial / natural soil and agricultural soil). To assess the sensitivity of the steady state FF to the selected input data, the lower and higher bound input values for particle size, and degradation rates in water and soil were used to calculate the deviation from the mean FF. Due to the limited input data points for these parameters, we were not able to perform a full uncertainty analysis.

2.4. Microplastics effect factor

To determine the effect factor of MiPs, effect data from MiP particle exposure in lab experiments data of individual species is used, including invertebrates, bacteria and algae species (Corella-Puertas et al., 2023, 2022; Everaert et al., 2020; Lavoie et al., 2022; Salieri et al., 2021). The EF is quantified in ReCiPe2016 as follows:

$$EF_{x} = \frac{\Delta PDF}{\Delta C} = \frac{\Delta PAF_{L(E)C50}}{\Delta C} = \frac{0.5}{HC50_{x}}$$
 (2)

The HC50 is the Hazardous Concentration of MiP exposure affecting 50 % of the species, based on L(E)C50 data (in kg/m³). The use of L(E)C50 data is in line with effect factor calculations of chemicals in ReCiPe2016. Here, L(E)C50 data is used as L(E)C50 data of chemicals are shown to correlate with the observed disappeared fraction of species in the field (Posthuma and De Zwart, 2006). Corella-Puertas et al. (2023) has summarized concentration-response studies of plastic pollution for aquatic species. From this study, the studies reporting the Effect concentration (EC50) and Lethal concentration (LC50) have been selected for ten individual species from five different phyla (SI 1.6).

2.5. Macroplastic conversion fraction

Most MiPs in the environment are fragmentation products from MaP released in the environment (Schwarz et al., 2023). A simplified MaP fragmentation is assumed, where MaP fully and exclusively fragments to MiP with a particle size similar to what was collected for the MiP FF modelling (SI 1.1–1.3) which was based on a model proposed by (Boersma et al., 2023). Hence, MaP disappears at similar rate as MiP formation from MaP. To include the fragmentation rate of MaP to MiP for polymer (x) and the impacts resulting from that, the polymer-specific midpoint CF of a MaP emission is derived through the following simplified approach:

$$ETP_{MaPx} = Fr_{MaP \to MiPx} \times ETP_{MiPx}$$
 (3)

Where $Fr_{Map->MiP}$ is the polymer-specific conversion fraction (dimensionless) from MaP to MiP in the environment. The conversion depends on the polymer-specific k_{frag} and the time horizon selected for each cultural perspective. The fragmentation rate of MaP for PP, LDPE and PET is derived for soil and water, assuming UV is present, as this is considered as one of the main factors influencing of MaP fragmentation (Chamas et al., 2020). The potential effect of macroplastic shape on the conversion fraction was not quantified in the fragmentation model due to lack of data. Data collection of the k_{frag} and further explanation of the MaP to MiP conversion fraction calculation and assumptions are elaborated in SI 1.1 and 1.2.

2.6. LCA case study

2.6.1. Goal and scope

A comparative LCA for two types of consumer multilayer packaging films is completed to illustrate the potential relevance of MaP and MiP emissions on the results of an LCA. Multilayers are complex materials which can be composed of at least two layers and make up a large quantity of the packaging sector (Horodytska et al., 2018). The packaging sector is the largest plastic consuming sector globally, with 172 Mt produced in 2021 (Plastics Europe, 2022). Additionally, consumer films make up a significant portion of identifiable littered waste in the

environment (Morales-Caselles et al., 2021).

The functional unit of the LCA is defined as 'one film packaging of 1 m² used to conserve food requiring average oxygen barrier properties'. The system boundaries are set from Cradle-to-Grave, and include the production, use and end-of-life processes including landfilling, incineration and mechanical recycling (SI 1.8). Both emissions and hence impacts from MiP and MaP are included. Processes and chemicals required for printing are excluded from the scope. The first case study is a multilayer of biaxially oriented polypropylene (BOPP) laminated to a layer of metalized PP (PPmetBOPP) with a thickness of 20 μm and a total mass of 36.75 gr. It is assumed that the different orientation of the PP and the metalized layer does not influence fragmentation, mineralisation and with that, degradation rates. The second case study is a multilayer of PET laminated to LDPE, the most common type of multilayer film in food packaging, with a thickness of 70 µm and a total mass of 81.84 gr. For the multilayer of PET-LDPE, it was also assumed that this would not affect the polymer degradation for MiP and fragmentation patterns for MaP. The geographical scope is set to Europe to limit complexity of the LCA. Full inventory details are available in SI 1.8, the production data of the use cases was provided by Leygatech and Eversia. Impacts from particle sizes below the size range of MiP and additive release are omitted from the study. Background inventory data from the industry 2.0 database and ecoinvent 3.9 are used. The ReCiPe2016 V1.1 midpoint and endpoint results for all three cultural perspectives are assessed. The LCA is executed in Brightway 2.5 (Mutel, 2017) and activity browser (Steubing et al., 2020).

2.6.2. Loss estimates of MaP and MiP

Per functional unit, quantities of loss need to be defined for both MiP and MaP during the products life cycle. These mass losses are quantified using material flow analysis (MFA) of MaP and MiP losses to the environment (Schwarz et al., 2023). This MFA approach is chosen as it provides detailed loss data over the full plastic life cycle from multiple plastic pollution sources (Kawecki and Nowack, 2019; Kaza et al., 2018; Peano, 2020). Furthermore, the model distinguishes different product groups, MaPs and (direct and primary) MiPs, countries, and environmental compartments emitted to. To obtain the losses for the film product in the case study, the littering profiles from consumer films for 27 EU countries have been extracted from an MFA study (Schwarz et al., 2023). The model data input was transformed to resemble 1 kg of plastic consumer film, which is then scaled to the functional unit . The environmental compartments of the MFA do not fully align directly with the emission compartments of LCA, hence some environmental compartments from the MFA are combined to align with the LCA compartment (SI 1.8, Table S11&S12). Data calculations, environmental compartment alignment and output are presented in SI 1.8.

3. Results

3.1. Fate and effect factors

Table 1 summarizes the MiP fate factors (FFs) for the three polymers, three cultural perspectives and the freshwater and marine compartments. PP MiP shows the highest FFs in the marine compartment, followed by LDPE. This includes the marine deep-sea where PP and LDPE are found to accumulate in the Simplebox4plastics model. FF for LDPE and PP are relatively similar for the freshwater compartment, where contributions from emissions to the soil compartments have a slightly lower FF for LDPE compared to PP (136–146 days and 158 days, respectively). The variation in FF per emission compartment is higher for LDPE compared to PP for both marine and freshwater receiving compartment. This is a result of the faster degradation rate of LDPE in soil compartments. PET has a low FF in both the marine and freshwater compartment (< 1 day). This is caused by the density of PET which is higher than water, in contrast to PP and LDPE with a density lower than water. PET accumulates in sediment compartments with FF values over

Table 1The Fate Factors (FF) of MiP emissions assessed through Simplebox4plastics, in days.

| Substance | Receiving compartment | Freshwater | | | Marine | | | |
|-----------|--------------------------|---------------|-------------|-------------|---------------|-------------|-------------|--|
| | Emission compartment | Individualist | Hierarchist | Egalitarian | Individualist | Hierarchist | Egalitarian | |
| PP | Freshwater lake | 121 | 154 | 154 | 3,300 | 22,205 | 45,431 | |
| PP | Freshwater | 159 | 159 | 159 | 4,135 | 25,399 | 47,073 | |
| PP | Ocean water | 0 | 0 | 0 | 6,925 | 25,503 | 47,129 | |
| PP | Natural/ industrial soil | 158 | 158 | 158 | 6,663 | 25,185 | 46,747 | |
| PP | Agricultural soil | 158 | 158 | 158 | 6,578 | 25,134 | 46,735 | |
| LDPE | Freshwater lake | 99 | 113 | 113 | 1,174 | 1,551 | 1,551 | |
| LDPE | Freshwater | 157 | 157 | 157 | 2,119 | 2,163 | 2,163 | |
| LDPE | Ocean water | 0 | 0 | 0 | 2,151 | 2,191 | 2,191 | |
| LDPE | Natural/ industrial soil | 146 | 146 | 146 | 1,971 | 2,013 | 2013 | |
| LDPE | Agricultural soil | 136 | 136 | 136 | 1,830 | 1,872 | 1,872 | |
| PET | Freshwater lake | 9.61E-06 | 1.01E-05 | 1.01E-05 | 2.23E-07 | 3.44E-07 | 3.46E-07 | |
| PET | Freshwater | 9.49E-02 | 9.95E-02 | 9.95E-02 | 2.21E-03 | 3.40E-03 | 3.41E-03 | |
| PET | Ocean water | 0 | 0 | 0 | 4.24 | 5.44 | 5.46 | |
| PET | Natural/ industrial soil | 9.36E-02 | 9.84E-02 | 9.84E-02 | 2.16E-03 | 3.36E-03 | 3.37E-03 | |
| PET | Agricultural soil | 9.30E-02 | 9.79E-02 | 9.79E-02 | 2.13E-03 | 3.34E-03 | 3.35E-03 | |

2000 days (SI 2.1, Table S14). Cultural perspective choice, indicating the exposure time of impacts, is especially important for the marine FF. The I-perspective (20 years' time horizon) has significantly lower FF for all polymers, and in lesser amount this is also observed for the H-perspective (100 years).

The sensitivity of the FF towards aquatic degradation rates and

particle size is depicted in Fig. 1. The variation in marine FF of PP and LDPE is particularly influenced by changes in the degradation rate. The PP freshwater FF is not affected by degradation rate, whilst the FF variation is limited for LDPE. In contrast, particle size particularly influences the FF for PET. Hence, smaller particle size affects the retention time of high-density polymers in aquatic environments, whilst

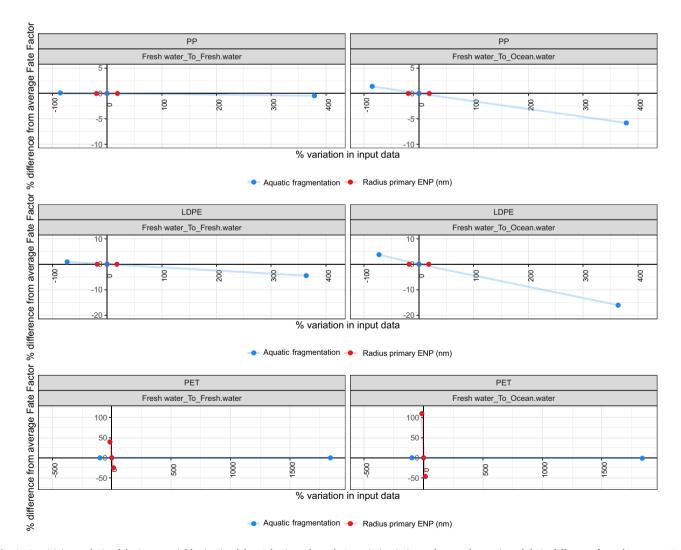


Fig. 1. Sensitivity analysis of the input variables in Simplebox4plastics, where the% variation in input data on the x axis and the% difference from the average Fate Factor on the Y axis. Emission compartment is set to fresh water, where other emission compartments are available in the Supplementary information.

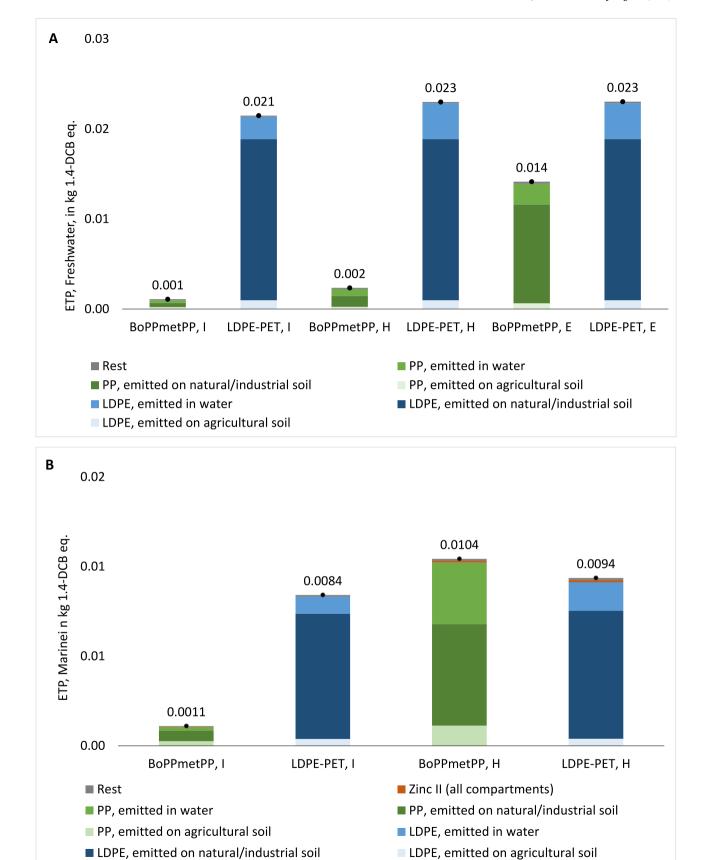


Fig. 2. LCA results on the midpoint indicators (Environmental Toxicity Potential, ETP, in kg 1.4-DCB eq.) for Freshwater (A) and Marine (B, C) ecotoxicity for the two case studies and all ReCiPe2016 cultural perspectives. Emission compartment contributions are referred to separately.

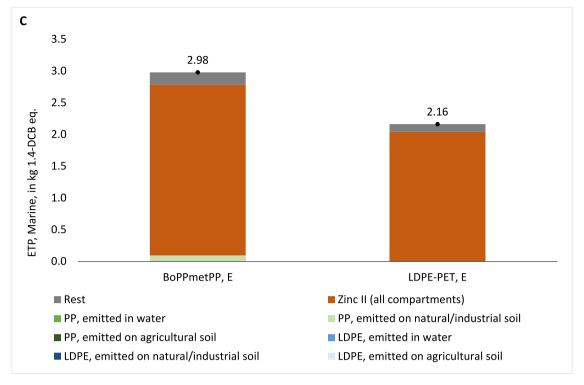


Fig. 2. (continued).

degradation rates affect lower density polymers. Spider diagrams for all emission compartments are available in SI 2.2.

The Effect Factor (EF) equals 29.1 PDF $\rm m^3~kg^{-1}$, (90 % confidence interval 5.3–159.6 PDF $\rm m^3~kg^{-1}$). As studies were limited (n=10, SI 1.6), no selection on type of polymer was made, hence all sizes and polymer types were included in assessment of the EF. Therefore, with this EF it is assumed all MiPs have similar effect of species, not considering size, shape and polymer type.

Unique MaP conversion fractions were obtained for three polymers, three time horizons and two compartments. The conversion fraction for all polymers in the E perspective equal 1 due to the infinite time horizon assumed. For the I and H perspective, PP has the lowest conversion fraction (0.01–0.28), followed by PET (0.4–1) and LDPE (0.6–1). PP All conversion fractions are presented in Table S5.

3.3. Case study

3.3.1. Midpoint results

The MiP midpoint ETPs, or CF, (kg 1–4 DCB eq.) for the three polymers in ReCipe2016 are presented in SI 2.3. PP and LDPE have a comparable ETP, where PP has the highest freshwater and marine ecotoxicity. PET has a low ETP, a result from a low FF for freshwater and marine compartments. The differences in CFs are reflected in the case study LCA, where the PP MiP emissions from the BOPPmetPP film contribute 88–98 % to the total freshwater ecotoxicity indicator for all cultural perspectives. The LDPE MiP from the PET-LDPE film contributes more than >99 % to the total freshwater ecotoxicity indicator (Fig. 2a). The impact score for freshwater ecotoxicity is about a factor 10 higher for the LDPE-PET film compared to the BOPPmetPP film, even though the LDPE-PET film is only 2x heavier (Fig. 2a).

For marine ecotoxicity, the contribution of LDPE MiP from the LDPE-PET film is also dominant in the I- and H-perspective (97–99 %) (Fig. 2b). The PP MiP from the BOPPmetPP film contributes 94–98 % to marine ecotoxicity. BOPPmetPP only scores lower for marine ecotoxicity in the I-perspective, mainly a result of low MaP fragmentation for PP

(Fig 2b). It is clear that cultural perspective choice affects the outcomes, where contributions of MiP are relatively small in the E perspective for marine ecotoxicity. Here, MiP impacts contribute 3 % and 1 % to the total midpoint score for marine ecotoxicity, for BOPPmetPP and LDPE-PET respectively. Zinc II emissions that occur during the life cycle, increase the marine ecotoxicity ETP with a factor 100 in the E perspective (Fig. 2c). The contribution of PET MiP is negligible for both marine and freshwater ecotoxicity in LDPE-PET, for all cultural perspectives.

Contribution of fragmented (converted) MaP to the total impacts is significant but strongly depends on mainly ratio of MaP vs MiP emitted per emission compartment, polymer type and cultural perspective. For the E perspective, MaP contributions vary between 97 % for natural/industrial soils to 68 % for agricultural soils. Contribution of fragmented MaP is the lowest for PP in the I perspective, only contributing 4- 41 % to the total impact.

3.3.2. Endpoint results

The ReCipe2016 endpoint values (species year) for the three polymers are presented in SI 2.3. Freshwater and marine ecotoxicity impacts on the midpoint level aggregate on the ecosystem endpoint. For the total endpoint impact score, the climate change impact contribute most, varying between 26 and 86 % per perspective and case study (Fig. 3). The contribution of MiP to the aggregated endpoint indicator is relatively low for both case studies. The total MiP impacts are highest for the LDPE-PET film, with highest contribution in the I-perspective (3.3 %). For both films, the freshwater ecotoxicity of MiP has a higher contribution compared to marine ecotoxicity of MiP to the total endpoint impact score for ecosystem quality. The endpoint impact score is higher for the BoPPmetPP film than the LDPE-PET film, even though the BOPPmetPP is a lighter product with less polymer included. This result can be explained by the impacts from other materials in the BOPPmetPP film (aluminium).

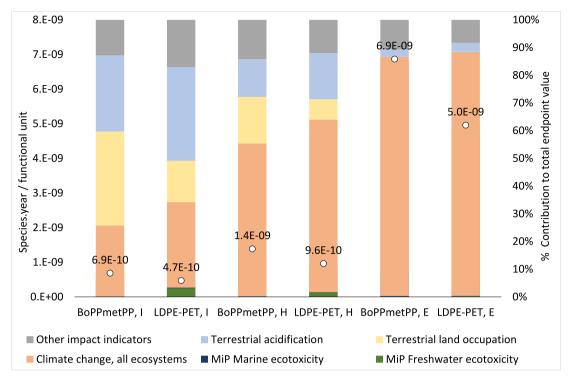


Fig. 3. LCA impact contributions (in% for the fill of the columns) and absolute endpoint impact scores for ecosystem quality, in species year per functional unit. for the two case studies and all ReCiPe2016 cultural perspectives.

4. Discussion

4.1. Fate factor

Simplebox4plastics is the first multimedia fate model used to quantify MiP fate factors (FF). Multimedia fate models are valuable tools for risk assessment and LCA methodologies but are not without uncertainties, partially due to dependency on input variables. The marine FFs assessed in the current study are observed to be sensitive for aquatic degradation rate. This is in line with findings in Quik et al., 2023. Unfortunately, data for fragmentation rates of MiP is limited, and MaP fragmentation data to MiP is used. Degradation processes occurs on the surface of plastics, hence the fragmentation rate will increase when particles size decreases (Chamas et al., 2020). Therefore, it is likely that the fragmentation rate of MiPs to nanoplastics is higher than assumed in the current study. It is key that more accurate data for MiP fragmentation is needed for different polymer types in various environments.

The sensitivity analysis indicated that FF of low density polymers was not affected by particle size. For high density polymer PET, larger particle size increases sedimentation rates and decreases the FF respectively. When addressing the uncertainty of particle size occurring with model estimates (Boersma et al., 2023), validation through independent measurement data is required, with high density polymers as a priority. The low density polymers (LDPE, PP) have particularly high FF in oceans and especially deeper ocean layers. Deep ocean layers are found to be a potential sink for MiP in literature, identified as marine snow, where it is bioavailable for benthic species (Porter et al., 2018). Within Simplebox4plastics assessments of the current study, accumulation of low-density polymers in sediments did not occur. This is in contrast to environmental observations where low density polymers are observed in sediments (Schwarz et al., 2019) as well as other oceanic modelling studies (Kaandorp et al., 2020). Additional vertical and horizontal transport mechanisms can play a role, which are not all included in Simplebox4plastics (SI 1.2 and SI 1.4). Additionally, the retention effect of freshwater systems can affect FF for freshwater and soil emissions. Modelling studies have found retention of smaller plastics,

especially in freshwater systems (Besseling et al., 2017). We expect that the FF reported in the current study are likely at the high end for low density polymers, as these removal and retention processes discussed above are not included. Inclusion of additional transport processes and environmental observations can improve FF modelling within multimedia fate models such as Simplebox4plastics, as well as additional MiP properties such as shape (Askham et al., 2023).

Other studies that have addressed FFs for LCA applied a different approach (Table 2). FF modelling for freshwater systems has been preliminary addressed using USEtox, where the physicochemical properties inputs were adapted to simulate MiP particles, such as a low partitioning coefficient (Salieri et al., 2021). This resulted in an FF for Polyester (PET) between 120 and 143 days for freshwater, similar to PP/LDPE FF in the current study. Interestingly, the high density of PET was not reflected in the assessed FF, and neither was degradation rate a significant factor (Salieri et al., 2021). The absence in Salieri et al. (2021) of particle characteristics, such as density and particle size, or aquatic transportation processes can explain the different outcomes to the current study. Maga et al. (2022) proposed a simplified method to quantify the FF for MiP using transfer coefficients from global plastic studies, combined with degradation rates. In their study, degradation, time horizons, polymers, shape and size were distinguished. Unfortunately, a freshwater assessment was missing in the study. The shape depended on degradation assessment from Maga et al. (2022) is an interesting approach that could be combined with Simplebox4plastics modelling, although shape was identified to have limited effect by itself. Next, Corella-Puertas et al. (2023) derived FFs for different sizes, shapes and polymers using sedimentation and degradation as removal processes in marine and freshwater systems. Corella-Puertas et al. (2023) observed a high sensitivity to the degradation rate and less so for the particle size. These findings are similar to the current study. Separate scenarios were build depending on a select number of degradation speeds. Slow degradation resulted in an FF of 40,000- 50,000 days for low density polymers in marine and freshwater compartments respectively (Table 2). The FF assessed in the current study decrease the reported ranges of FF significantly compared to Corella-Puertas, et al. 2023. The

Table 2 Comparison of Fate Factor (FF) in other LCA studies for Microplastics (MiP). The final columns include qualitatively whether a differentiation in MiP aspects was made (Y=Yes, N=No).

| Study | Impacts | Polymer | Freshwater FF (days, min - max) | Marine FF (days, min - max) | FF: multimedia modelling approach | FF: emission compartments | FF: shape | FF: size | FF: time horizons |
|--------------------------------|----------------------|---------------------------------------|------------------------------------|-----------------------------------|---|---------------------------|--------------|-------------|----------------------|
| Polypropylene (PP) | | | | | | | | | |
| Current study | MiP physical impacts | PP (default shape, mean size) | 158 - 159 | 4,135 - 47,130 | Y | Y | N | Y | Y |
| Corella-Puertas et al. 2023 | MiP physical impacts | PP (spheres, default) | 4.5 - 37,751 | 6 - 50,335 | N | N | Y | Y | N |
| Maga et al. 2022 | undefined | PP (all shapes, sizes) | n.a. | 0.06-412 | N | Y | Y | Y | Y |
| Low density polyeth | ylene (LDPE) | | | | | | | | |
| Current study | MiP physical impacts | LDPE (default shape, mean size) | 136 - 157 | 1,830- 2,191 | Y | Y | N | Y | Y |
| Corella-Puertas et al. 2023 | MiP physical impacts | LDPE (spheres, default) | 1 - 39,485 | 1 - 52,646 | N | N | Y | Y | N |
| Maga et al. 2022 | undefined | LDPE (all shapes, sizes) | n.a. | 0.07 - 443 | N | Y | Y | Y | Y |
| Polyethylene tereph | thalate (PET) | | | | | | | | |
| Current study | MiP physical impacts | PET (default shape, mean size) | 0.09 - 0.1 | 0.002 - 5.5 | Y | Y | N | Y | Y |
| Corella-Puertas et al. 2023 | MiP physical impacts | PET (spheres, default) | 0.04 - 4 | 0.4 - 40 | N | N | Y | Y | N |
| Maga et al. 2022 | undefined | PET (all shapes, sizes) | n.a. | 0 - 0 | N | Y | Y | Y | Y |
| Salieri et al. 2021 | undefined | PET | 120-143 | n.a. | Y | N | N | N | N |

maximum freshwater FF for low density polymers deviates strongly from the current study, mainly as additional fate removal processes were included in the current study such as the hydraulic transfer from freshwater to marine water (Hollander et al., 2016; Quik et al., 2023). For PET, FFs are relatively similar, as sedimentation is the dominant process in both studies. The current study demonstrates that using multimedia fate models can limit FF range estimates, keeping in mind the input data sensitivities and model assumptions. A uniform modelling approach to FF modelling of particles is required for LCA to limit the range of FF and simplify the process for LCA practitioners.

4.2. Macroplastics conversion fraction

The MaP conversion fraction proposed in the current study is a first simplified assessment to include MaP plastic pollution in LCA, through its fragmentation to MiP. However, there is significant room for improvement of this model. Firstly, it was assumed that all MaP fragment to form MiP. This linear fragmentation model excludes mineralisation and nanoplastic formation directly from MaP. Although mineralisation rates are significantly smaller than fragmentation rates (see Table S1), this can affect the volume of MiP produced and hence the total volume of MiP that results in damage to the environment. Furthermore, the collected fragmentation rates include high variation and uncertainty within the datapoints and due to the variation in product shapes (see SI 1.1, 1.3, 1.5), both affecting the volume of MiP in the LCA for the I and H perspective. Furthermore, the transport from emission compartment to different receiving compartments was not included in the conversion fraction. This will not only affect the MaP conversion directly, but also indirectly as the fragmentation rates may be different in different environmental compartments. Several studies have identified coastal zones and freshwater systems as MaP hotspots, or plastic sinks, including shores, vegetation, estuaries, mangroves and sediments (Ivar do Sul et al., 2018; Morales-Caselles et al., 2021; Onink et al., 2021; van Emmerik et al., 2022). In short, a full fate assessment for MaP requires a fragmentation model, environmental effects through an EF and its fate through an FF. This will improve the MiP formation estimates from MaP as well. Potentially, multimedia fate models could be adapted to both include MaP particles, as well as additional receiving compartments which are key for MaP fate due to accumulation.

4.3. Effect factor

The EF in the current study is based on a selection of L(E)C50 values from 10 species from 5 phyla, summarized by Corella-Puertas et al. (2023). The proposed EF in Corella-Puertas et al. (2023) is higher compared to the current study (1067.5 PAF m³ kg⁻¹ compared to 29.1 PDF.m³.kg⁻¹). The main difference is the starting point effect concentrations for which 50 % of the population was affected after exposure in the current study. Corella-Puertas et al. (2023) used chronic EC10-values, i.e. effect concentrations for which 10 % of the population is affected after chronic exposure. L(E)C50 values were selected for the current study to align with the EF calculation for toxicants in ReCiPe2016, as elaborated in SI 1.6. This difference also explains the higher value reported by Corellas-Puertas et al. (2023) compared to the current study. This is also reflected in the different EF units, where the current study used the PDF (Potentially Disappearing Fraction of Species) whilst Corellas-Puertas et al., 2023 used PAF (Potentially Affected Fraction of Species). Furthermore, with ten species and five phyla represented in the calculated EF, only a limited view on species impacts is given. This value requires an update when new exposure data for species from other phyla (e.g. Chordata) is available. Finally, note that an EF is not yet available for MaP, to include impacts of smothering, ingestion impacts and entanglement impacts (Gall and Thompson, 2015; van Bijsterveldt et al., 2021; Woods et al., 2021). This limitation results in an underestimation of the impacts of plastic pollution in LCA case studies.

4.4. LCA case study

The case study of a consumer film highlights the relevance to include plastic pollution in the midpoint evaluation of ecotoxicity impacts. Choice in polymer for a product is important to consider, highlighted by the differences between the case study results. PET has limited impacts to the measured aquatic impact categories, while PP and LDPE are dominant for the results on the midpoint level. The marine ecotoxicity

values are much higher for the E perspective, where the impacts of zinc and other metals are more dominant than the MiP impacts. However, these impacts of metals in the E perspective in oceanic environments have been criticised due to extreme long residence times of metals in the marine environment (Huijbregts et al., 2001; Ligthart et al., 2004). The ecotoxicity impacts contributions are small in the endpoint assessment of ReCiPe2016, where other impact categories, including climate change, are dominant in the final ecosystem impacts. Now that plastic pollution impacts can be included in LCA, product choices can be made to reduce impacts from MiP, including polymer type and plastic quantities used. In the inventory, the average mass loss of a consumer film in Europe was used, which is estimated to be 2.6 %. However, on a global scale, these loss values can increase when proper waste management is missing, increasing or decreasing the contribution of MiP ecotoxicity when the geographical scale is changed.

The plastic pollution impacts assessed in the current study are far from complete and should be continued. First, Zhu et al., and Royer et al., observed CO2 and CH4 formations in oceans through MiP mineralization, which are not yet included as emission (Royer et al., 2018; Zhu et al., 2020). Ocean acidification has been identified due to the mineralization products of plastics as well (Romera-Castillo et al., 2023). Human toxicity is out of scope for the current study, although highlighted in literature (Leslie et al., 2022; Ragusa et al., 2022). This is especially relevant for smaller plastic particles ($< 1 \mu m$, or nanoplastics), as these can be damaging when passing blood barriers in humans (Leslie et al., 2022). This also highlights that bioavailability with size and potentially shape might vary and should be further explored. Also terrestrial ecotoxicity is omitted from the study, although most impact might occur in terrestrial environments due to the high MaP and MiP emission rates (Kawecki and Nowack, 2019; Schwarz et al., 2023). Recent studies are focusing on exposure routes and impact methodology development for terrestrial environments (Li et al., 2023). The potential toxicity of additives which are leached from the polymer matrix is not included, as well as other hydrophobic chemicals that can adsorb to the MiP which can increase toxicity in aquatic environments (Chen et al., 2018). In short, MiP pollution physical impacts do not cover all impacts of plastic pollution. Regular updates and improvement on the proposed methodology are therefore necessary. At last, physical impacts of MiPs were quantified through the current ecotoxicity midpoint categories of ReCiPe2016. However, physical impacts are different from chemical impacts. Hence, the terminology of the ecotoxicity impact categories may require adaptation. This can be solved by reformulating the ecotoxicity category to micropollution, which would then refer to both impacts of chemicals and microplastics.

4.5. Concluding remarks

Microplastic (MiP) impacts were included the LCIA-method ReCiPe2016. Characterisation factors for LDPE, PP and PET were derived for three different time horizons. Environmental fate of plastic pollution was comprehensively assessed using the multimedia fate model Simplebox4plastics, while the effect factor was based on a species sensitivity distributions, based on L(E)C50 toxicity data. We applied the updated ReCiPe2016 method to an LCA case study of two consumer packaging films, including plastic pollution losses. The contribution of low density MiP dominated the midpoint impacts for freshwater and marine ecotoxicity with contributions ranging between 88 and 100 % for the Individualist and Hierarchist perspective. In the Egalitarian perspective, MiP contribution was minimal for marine ecotoxicity, highlighting the importance of cultural perspective choices for a LCA. Overall impacts on ecosystem quality of the two packaging films were, however, dominated by climate change impacts. The updated ReCiPe2016 method is expected to be particularly relevant for assessing life cycle ecotoxicological impacts of plastic products and for services, such as cleanup assessments. By including plastic pollution in LCA, choices to minimize impacts of plastic pollution can be evaluated and

supported.

CRediT authorship contribution statement

A.E. Schwarz: Conceptualization, Data curation, Formal analysis, Methodology, Visualization, Writing – original draft. S. Herlaar: Software, Data curation, Writing – review & editing. Q.M. Cohen: Validation, Writing – review & editing. J.T.K. Quik: Validation, Writing – review & editing. M. Golkaram: Methodology, Writing – review & editing. J.H. Urbanus: Writing – review & editing. T.H.M. van Emmerik: Writing – review & editing. M.A.J. Huijbregts: Methodology, Supervision, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Supplementary materials

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