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A predictive model to assess the accumulation of microplastics in the natural environment

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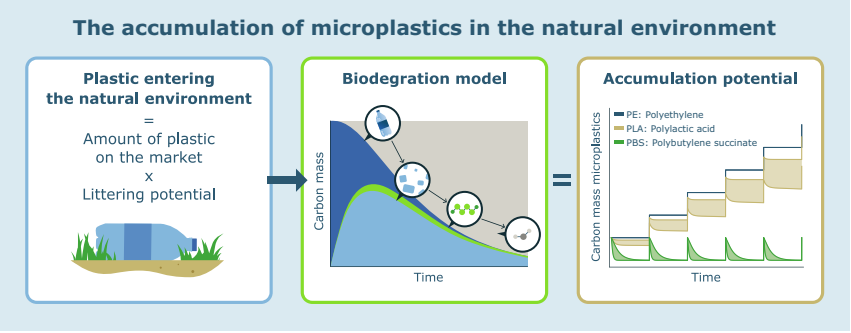
HIGHLIGHTS

- Integrated biodegradation model enables the assessment of microplastic accumulation
- Accumulation potential defined as time-integrated concentration of microplastics
- Biodegradability leads to significantly lower polymer accumulation potential
- High biodegradation rates prevent microplastic accumulation in soil

GRAPHICAL ABSTRACT



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ABSTRACT

The use of plastics inevitably leads to (micro-)plastics entering and accumulating in the natural environment, affecting biodiversity, food security and human health. Currently, a comprehensive and universally applicable methodology to quantify microplastic accumulation in the natural environment is lacking. This study proposes an integrated biodegradation model that provides the possibility to examine and compare the microplastic formation and accumulation of different polymer types in diverse natural environments. The proposed model derives carbon mass flow streams from experimental mineralisation curves (CO_2 evolution) of polymers and predicts the concentrations and residence times of the different plastic states during their biodegradation processes. The model allows for the description of the accumulation potential of polymers, as the time-integrated concentration of microplastics present in the natural environment during a timeframe of 100 years after a polymer enters the natural environment. The model is applied to estimate the accumulation potential of three polymers with different biodegradation profiles in soil: polybutylene succinate (PBS), polylactic acid (PLA) and polyethylene (PE). It is demonstrated that the dimensionless accumulation potential of PBS in soil is near zero (between $3.0{\cdot}10^{-4}$ and 0.002) which corresponds to a potentially very low level of accumulation. On the other hand PE shows a near maximum value of 1 which corresponds to the almost completely non-biodegradable character of this polymer in soil. PLA exhibits a wide range of values in between that of PBS and PE which reflects its reported relatively slow biodegradation in soil. The proposed model can be used to guide material

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selection in product design by quantifying the microplastic accumulation of these different polymer types. To demonstrate its use, plastic candy wrappers and agricultural mulch films were selected as case studies. Both case studies show that high biodegradation rates can limit or prevent microplastic accumulation in soil.

1. Introduction

Plastics are an important part of modern daily life, due to their unparalleled combination of material properties such as excellent mechanical and barrier properties, low weight, durability, cost-effectiveness and versatility in tuneable characteristics. Consequently, the global production of plastics is still increasing at an annual rate of approximately 2.5 %. However, suboptimal collection and recycling systems result in a substantial portion of used plastics that end up in the natural environment. The leakage of these plastics to the environment can be reduced by the implementation of well-designed waste management structures, but cannot be prevented completely (OECD, 2022). Furthermore, due to their persistence, even minute leakage levels can lead to the accumulation of plastics in the natural environment. This plastic pollution manifests in various forms, ranging from visible litter in terrestrial and aquatic ecosystems to less visible micro/nano-plastics and associated chemicals, resulting from polymer degradation and wear. The environmental effects of these diverse types of plastic pollution have been subject of many studies in recent years, raising concerns regarding its negative effects on biodiversity, food security and human health (OECD, 2022; Gasperi et al., 2018; Kosuth et al., 2018; Mintenig et al., 2019; Rainieri and Barranco, 2019; Cverenkárová et al., 2021; Saeedi, 2024; Mamun et al., 2023).

Biodegradable polymers emerge as a potential solution to mitigate plastic pollution, offering stable performance during use while exhibiting shorter residence times in natural environments. A clear barrier for the application of these polymers is that biodegradation rates are often difficult to predict and can vary substantially per environment. This is a consequence of the fact that the biodegradation process is influenced by both material composition and environmental conditions. Standardised laboratory tests are used to assess biodegradation by measuring the disintegration or the formation of the ultimate biodegradation product, carbon dioxide (CO_2) (Van der Zee, 2020; *SAPEA: Science Advice for Policy by European Academies, Biodegradability of Plastics in the Open Environment*, 2020). However, as environmental conditions vary in the natural environment, degradation rates in standardised lab tests are not always the same as in real-life environments. To comply with certification standards for soil biodegradability, polymers need to show complete biodegradation (i.e. >90 % mineralisation to CO2) in specified lab tests within a 2-year time frame (TÜV AUSTRIA, 2012; DIN CERTCO, Certification Scheme: Biodegradable in Soil, in according to DIN EN 17033 and/or ISO 23517, 2023). Polymers that biodegrade in the timeframe of decades, such as polylactic acid (PLA), are classified as non-biodegradable in soil and therefore fall into the same category as conventional polymers like polyethylene (PE) that are known to persist for over centuries. Although both are non-biodegradable polymers by definition, their environmental impact could be of a completely different magnitude. Another complication for biodegradation assessment is that during the biodegradation process microplastics and biochemicals are formed as transient products (Degli-Innocenti et al., 2022), which adds to the difficulty of assessing and predicting the environmental effects of plastics in specific environments.

Life cycle assessments are an insightful and widely accepted approach to assess the sustainability of materials and (consumer) products and therewith guide material selection in product development. Although the environmental effects linked to plastic pollution are gaining recognition, they are often not included in standard life cycle assessments (LCA) (Boone et al., 2023). Attempts to incorporate these environmental effects show that among others, knowledge on the location, the (plastic) state (e.g. macroplastic, microplastic, intermediate product, etc.), concentration and residence time of (micro-)plastics in the natural environment is needed for successful implementation (Woods et al., 2021; Saling et al., 2020; Maga et al., 2022; Colwell et al., 2023; Ward and Reddy, 2020). Also in plastic systems assessments, such as material flow analysis, the leakage of plastics into the natural environment is hardly incorporated. In those studies in which the leakage is addressed, the processes that occur once these materials enter natural ecosystems are not included (Schwarz et al., 2023; Winterstetter et al., 2023; Lobelle et al., 2023). Hence, there is a need for a comprehensive and universally applicable methodology to quantify microplastic formation and accumulation. The incorporation of such a methodology into environmental and system assessments will enable better inclusion of the impact of plastics accumulation in the natural environment.

Recent scientific efforts to relate the persistence of a product to the biodegradation behaviour of polymers in the natural environment either miss important details about the chemical conversions that are taking place during the degradation process or are not shown to be suitable for non- or slow degrading polymers. On the one hand, there are existing approaches that predominantly rely on surface degradation rates, conceptualizing the degradation process as occurring solely at the surface of the polymer object or particle where the polymer breaks down into smaller fragments (Saling et al., 2020; Chamas et al., 2020; Harrison et al., 2022). In these models, biodegradation is modelled as a mass loss of the original polymer product. This assumption implies ignoring that worn off particles can still prevail as micro- or nano-plastics. One can only be certain that this material has fully biodegraded, and is not present in the environment as micro- or nanoparticles, when a material is fully converted into gaseous products and minerals. On the other hand, there are studies that propose modelling approaches describing the biodegradation behaviour of specific biodegradable polymers (Degli-Innocenti et al., 2022; Nelson et al., 2022; Ghimire et al., 2020). These modelling approaches do include the complete mineralisation of these polymers. However, these models do not describe the biodegradation of slow or non-biodegradable polymers. Hence, an all-encompassing methodology to assess and compare the biodegradation behaviour and therewith the accumulation of different types of polymers in the natural environment is not available at this moment.

This study proposes an integrated and universally applicable biodegradation model that enables the assessment and comparison of microplastics formation and accumulation of different polymers in a specific natural environment. The model derives carbon mass flows from experimental mineralisation curves (CO_2 evolution) obtained with standard lab tests, resulting in the concentration and residence times of the various plastic states during the biodegradation processes. In doing so, the model allows for the description of the accumulation potential of polymers, as the time-integrated relative concentration of microplastics in the natural environment during a timeframe of 100 years after a polymer enters the natural environment. Subsequently, it is shown how this model can be used to guide material selection in product design by quantifying the microplastic formation and accumulation of these different polymer types in specific applications. Plastic candy wrappers and agricultural mulch films were selected as case studies. Application of the model on these cases shows how the microplastic accumulation depends on both the polymer that is used and on the product-system in which it is applied.

2. Methodology

2.1. Conceptual basis

The conceptual basis of the model presented in this study is based on two principles. The first is that the residence times of the substances formed during the biodegradation process can be determined by a carbon mass balance as a function over time (Fig. 1). This mass balance follows the biodegradation cycle of a plastic product (macroplastic) that is first converted into microplastics, then into monomers and/or biochemicals and ultimately into CO_2 and biomass. The model assumes full aerobic biodegradation of a polymer, in which all carbon in the polymer is converted to carbon dioxide, water and sequestered carbon (process described in Eq. 1). Although the chemical structure changes over time, the total mass of carbon remains the same at all stages during this process. Hence, for each polymer the substances formed during biodegradation in a specific environment and under specific conditions can be described over time. The second model principle is that this carbon mass balance can be modelled for all materials, as long as the mineralisation of a material is measured and documented. For biodegradable polymers, measuring the mineralisation curves is a standard analysis method in the certification procedure for biodegradation in a specific environment. However, these curves can be obtained for other materials as well, including polymers that are non-biodegradable, such as PE (Albertsson, 1980). Hence, our model states that all polymers will eventually biodegrade but the timespan in which this process takes places varies greatly between polymers.

$$C_{polymer} + O_2 = CO_2 + C_{biomass} \quad (1)$$

The model generates a polymer-environment specific mass balance which predicts the concentration and residence times of the substances formed during the biodegradation process. The area under the curves of the formed substances can be used to determine the accumulation potential of polymers in a specific environment which is defined as the normalized time-integrated concentration of these substances in the environment (eq. 2). This polymer accumulation potential (dimensionless) indicates the mass and residence time of the chemical structures that are present in the natural environment in the form of microplastics (and macroplastics), relative to the total amount that was discarded. In this study, the accumulation potential is calculated for 1 mass unit of material over a timeframe 100 years. This period of 100 years was chosen to balance the short-term and long-term presence of microplastics. Furthermore, it is it expected that microplastics have the most impact on species, ecosystems and human health in the first 100 years after release, as it is likely that after 100 years these particles have settled in areas with lower exposure rates (e.g. the deep ocean or deeper soil layers) (Woods et al., 2021). Other carbon phases, like small intermediate products (SIPs), were excluded from the polymer accumulation potential. Although the presence of the SIPs in the environment might also have a (probably different) impact on the environment, the SIPs are expected to have a relatively short residence time compared to microplastics. The accumulation potentials of different polymers can be compared, when they are determined under similar environmental conditions.

$$Accumulation\ potential = \frac{1}{100y} \times \int_0^{100y} Microplastic\ concentration \bullet dt \quad (2)$$

2.2. Data collection

The proposed model describes the carbon mass balance during the biodegradation process by fitting experimental biodegradation data. Mineralisation curves (CO_2 evolution over time) are used as input for the model as it is considered the most appropriate data to assess the polymer mass balance over time. These tests directly represent the mineralisation of the polymer over time and are a direct measure of the end product of this mineralisation process. Generally, the powdered test substances used in these tests have a particle size <5 mm (*EN-ISO 17556, Plastics - Determination of the Ultimate Aerobic Biodegradability of Plastic Materials in Soil by Measuring the Oxygen Demand in a Respirometer or the Amount of Carbon Dioxide Evolved*, 2019) which corresponds to the threshold for the definition of microplastics (Woods et al., 2021; Frias and Nash, 2019; da Costa et al., 2017). Other biodegradation characterization methods (enzyme assays, respiration tests, laboratory simulated biodegradation test and field trials) cannot be used as input for the model as they do not generate suitable data for the calculations. For instance, in laboratory simulated biodegradation test or field trials it is impossible to capture the intermediate products and assess their weight. Hence, we chose the

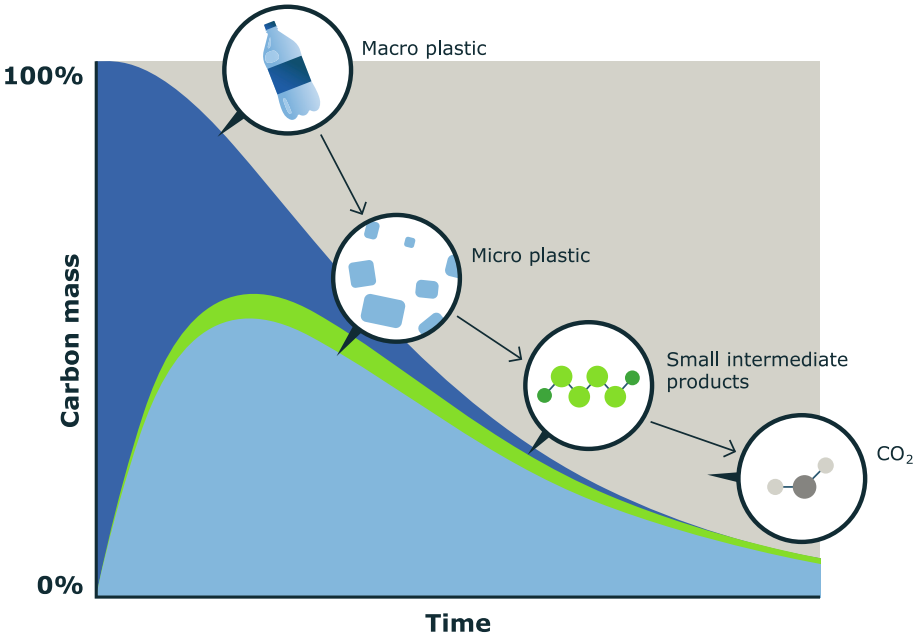

Fig. 1. Mass balance of carbon as a function of time (conceptual idea).

mineralisation curves of polymers under specific conditions as input for the model that fits the time constants of a biodegradation process. In this study, we applied the model by assessing three polymers (polyethylene (PE), polylactic acid (PLA) and polybutylene succinate (PBS)) in a soil environment at natural temperatures. The data sources and a brief description of the biodegradation conditions for the mineralisation curves for all polymers included in this study are given in the supplementary material S1.

2.3. State-space description of the biodegradation process

In this model, we describe the biodegradation process as a set of time-invariant first-order differential equations expressed as a state space model. This allows for simulating the biodegradation process using commonly used tools (e.g. Python, R, Matlab). The input $u(t)$ is the amount of polymer entering the environment per unit of time and is normalized to 1 to fit the data of the mineralisation curves. This normalised input is also used for determining the accumulation potential. The state vector $x(t)$ represents the amount of material in each of the states of degradation (see eq. 3).

$$x(t) = \begin{bmatrix} \textit{mass in state } 1 \ (\textit{entry into the environment}) \\ \textit{mass in state } 2 \\ \vdots \\ \textit{mass in state } n \ (CO_2) \end{bmatrix} \tag{3}$$

Different carbon mass flow schemes with varying number of states were evaluated (see Supplementary Materials S4). The time derivative of $x(t)$, $\dot{x}(t)$, can be written as:

$$\dot{x}(t) = \underbrace{\begin{bmatrix} -\left({}^{1}/_{\tau_1} + {}^{1}/_{\tau_4}\right) & 0 & 0 & 0 \\ {}^{1}/_{\tau_1} & -{}^{1}/_{\tau_2} & 0 & 0 \\ 0 & {}^{1}/_{\tau_2} & -{}^{1}/_{\tau_3} & 0 \\ 0 & 0 & {}^{\alpha}/_{\tau_3} & 0 \end{bmatrix}}_{A} x(t) + \underbrace{\begin{bmatrix} 1 \\ 0 \\ 0 \\ 0 \end{bmatrix}}_{B} u(t) \tag{4}$$

where $\tau_1 \ldots \tau_4$ are the time constants of the mass flows between the states and α is the fraction of the carbon mass that is converted into CO2 as opposed to the carbon mass that is converted into sequestered carbon. The mineralisation curves of fully biodegradable polymers often level off at a CO2 conversion of around 80 %–90 % of the theoretical maximum. This is mainly because part of the polymers' carbon is sequestered in biomass (the micro-organisms use the polymer as an energy source and for growth) (Sander et al., 2024). A more in-depth description of the derivation of the state-space representation is given in the supplementary material S2.

2.4. Fitting of mineralisation curves and data processing

For each polymer, the experimental CO_2 evolution curves were used to fit the state-space model described in eq. 4. Model fitting was performed by non-linear least-squares optimisation using the scipy.optimize.curvefit function in Python. As non-linear optimisation is susceptible to finding local minima dependent on the initial parameter estimates, each CO_2 evolution curve was fitted with 500 different sets of pseudo-randomly generated initial guesses for fit parameters $\tau_1 \ldots \tau_4$. To account for the wide variation of time constants among the different polymers, two sets of fit conditions were defined: one set for fast-degrading polymers and one set for slow degrading polymers. Table 1 shows the parameter bounds and the range of the initial guesses for all parameters for both polymer types. For fast degrading polymers (e.g. polybutylene succinate (PBS)) mineralisation curves describe the full biodegradation of the polymer. Therefore, the initial guess range of the parameters $\tau_1 \ldots \tau_4$ was set at 0.005–5 years and the initial guess estimations of the parameter α was set at 0.8. For slowly degrading polymers (polyethylene (PE) and polylactic acid (PLA)), only the first years (or even less) of the degradation process were described by the mineralisation curves. Therefore, the initial guess estimation range of the parameters $\tau_1 \ldots \tau_4$ for these polymers was set to the broad range of 5 to 5000 years. The parameter α was fixed for these polymers at 0.8, i.e. it is assumed that all polymers are fully degraded at 80 % CO2 conversion. It should be noted that the relative outcomes of the model are not affected by the value of α as along as α is kept constant for all slowly degrading polymers.

Table 1
Fit conditions.

Degradation speed	Polymer	Parameters $\tau_1 \ldots \tau_4$ [y]		Parameter α	
		Bounds	Initial guess range	Bounds	Initial guess
Fast	PBS	0–10,000	0.005–5	0–1	0.8
Slow	PE, PLA	0–10,000	5–5000	Not fitted; fixed at 0.8	

For all fit results from the different initial guesses, the residual sum squared (RSS), Akaike information criterium (AIC) number, coefficient of determination (R^2), covariance matrix and condition number of the covariance matrix (κ) were calculated, as described in supplementary material S3. All fits where at least one of the found time constants was <0.001 y (~1/3 day) were directly omitted as this is not considered to be physically realistic. Subsequently, the resulting fits were analysed and the most reliable fits were selected based on R^2 and κ. First, all fits with $1\text{-}R^2 > 2 \cdot (1\text{-}R^2_{max})$ were omitted, i.e. all fits where the deviation between R^2 and 1 (a perfect fit) is more than twice that deviation for the best R^2. Next, all fits that fall outside the 25th percentile of the remaining unique fits sorted by lowest κ were omitted. Taking the 25th percentile provided a good balance between the number of fits found and the reliability of the fit.

These calculations result in a variety of probable sets of parameters that can describe the biodegradation process of the different polymers. With these sets parameters, the residence times and concentration of the substances formed during the biodegradation process can be determined, which can then be used to calculate their accumulation potential (following eq. 2).

2.5. Simulation of scenarios with continuous and pulsed material input (case studies)

The proposed biodegradation model can be used to predict the substance concentrations in the environment during the degradation process, which is exemplified by two case studies (section 3.3). The input mass $u(t)$ is determined for a single product (for instance to compare two different materials), or a complete product system (for instance the amount of products littered in a country). Depending on the case, the input material enters the environment in either a continuous or a pulsed manner (e.g. annual release of material into the environment).

The scenario with a continuous material input rate is simulated by computation of the step response of the state space system. The step response is the evolution of the system state from $x(0) = 0$, for input $u(t) = 1$ for $t > 0$ (Lathi, 2005). The step response represents the amount of material in each of the states given a constant input of 1 mass per time unit into the environment. This step response is then multiplied by the amount of material that enters the natural environment.

We use a convolution approach for simulating pulsed scenarios. First the impulse response of the state space system is computed. For this, the

state is instantaneously updated at $t = 0$ and $u(t) = 0$ for $t > 0$ (no further input into the environment, only the degradation). This thus represents the input of a single release into the environment. As a state space system is linear and time-invariant, the resulting output for a single release may be shifted to subsequent periods representing subsequent releases and these results may be added to obtain the cumulative result.

3. Results

3.1. Model of polymer biodegradation in the natural environment

The carbon mass flow of the biodegradation process is best described using a system with four carbon states placed in series with a parallel path from the first state to the third state, as this captures all characteristics of the experimental curves. In Fig. 2 the input data (measured CO_2 evolution curves) and the best modelled fit results are shown. These plots show that the chosen model describes the mineralisation process well for both slow and fast degrading polymers.

The model is a good representation of the physical and chemical processes that occur during the biodegradation process, as the states of the model can be coupled to the phases of the biodegradation process (Fig. 3). After the plastic enters the environment, micro-organisms start colonizing the surface of the product and the metabolism of the microbes adapt to the plastic at hand. The microbes start to release enzymes causing depolymerisation, which contributes to the fragmentation of the polymer. This process results in a larger surface area for the enzymes to reach the material (τ1). The extracellular (enzymatic) depolymerisation continues (τ2) resulting in small intermediate products (SIP), such as alcohols or acids that can be taken up by the microbes for their metabolism. These SIP are subsequently metabolised by enzymes inside the microbial cells and either converted further into CO_2 (τ3) or into sequestered carbon, in case the carbon is incorporated in biomass. It is also possible that SIPs are already formed without prior fragmentation, e.g. when the extracellular enzymes directly cut off small molecules from polymer chain ends (τ4). For some polymers (e.g. PLA and PGA), the fragmentation processes (τ1, τ2 and τ4) can also occur without the catalytic action of enzymes, for instance due to chemical hydrolysis and/or oxidation. By connecting the different carbon states of the model to a particular phase in the biodegradation process the model is able to predict the carbon mass over time for all the intermediate substances in the biodegradation process.

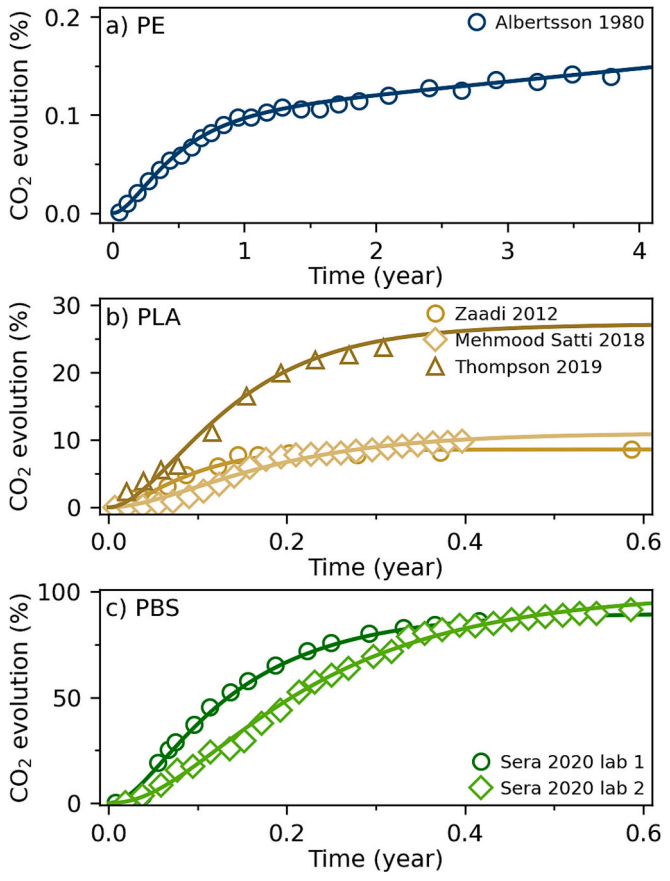

Fig. 2. The input data and modelled fit results for the CO_2 evolution of all datasets that were analysed in this study.

3.2. Substance carbon mass and corresponding time constants

Fig. 4 shows examples of the carbon mass of the different substances over a one-year timespan for the biodegradation process of three polymers (PE, PLA and PBS) that possess distinctly different biodegradation rates in soil. The detailed modelling results over a timeframe of 100 years are given in the supplementary material S5 (for all evaluated data sets). The corresponding time constants are provided in Table 2. The model predicts that for all polymers the carbon mass passes the micro-I phase relatively quickly (0.5–2 year), which is reflected in the small value of time constant τ1 of <0.5 year (Table 2), and the short lag phase in the input data for all polymers (Fig. 2).

The predicted carbon mass present in the micro-II phase is significantly different for the different polymers, which is reflected by the large variation of the value of time constant τ2. This suggests that the step from micro-II to SIP is the rate limiting process. For PBS, the carbon has passed the micro-II phase within <0.5–2 years after starting the biodegradation process. In contrast, for PLA and PE the amount of carbon that has passed the micro-II phase is almost negligible after 2 years. Extension of the biodegradation time to 100 years (Fig. S7–9) shows that for PLA there is some carbon mass passing the micro-II phase, but this is very slow. Logically, the time constants τ2 vary significantly between PBS, PLA and PE with values <<1 year for PBS, in the order of hundreds to thousands of years for PLA and multiple thousands of years for PE.

For all polymers, the conversion from the SIP state to CO_2 is quick (τ3 < < 1 year), which matches the known biodegradation rates of these substances (Batiste et al., 2022; Baba et al., 2017). The fast degradation kinetics of PBS results in a fast formation and decay of SIPs, with 99 % carbon mass passing this phase in <0.6 year. For PLA, an initial peak in the carbon mass of the SIP is observed with a subsequent steep increase in CO_2. This might be surprising given the slow rate of carbon mass transfer from micro-II to SIP state. It can be explained mathematically by the relatively low value of τ4 (<1 year) that describes the mass transfer from the micro-I state directly to the SIP stage. However, the carbon mass from the micro-I state is converted relatively quickly into the micro-II state. Consequently, the mass flow from micro-I to SIP decays as the micro-I state depletes. The formation of SIP thus becomes solely dependent on the path from micro-II, which is a very slow process. As the dominant degradation process for PLA is hydrolysis (Lott et al., n.d.; Lunt, 1998), it is expected that in reality the value of τ4 is much closer to that of τ2. Nevertheless, based on Fig. 2, it can be concluded that the model is able to describe the biodegradation process of all polymers well.

3.3. Polymer accumulation potential

The polymer accumulation potential describes the mass fraction and residence time of microplastics (sum of phase micro-I and micro-II) that are present in the natural environment over a period of 100 years. Table 2 shows the accumulation potential and the corresponding time constants, for the different polymers that were evaluated in this work. The time-integrated mass of the individual substances is provided in the supplementary material S6. In all cases a best (lowest accumulation

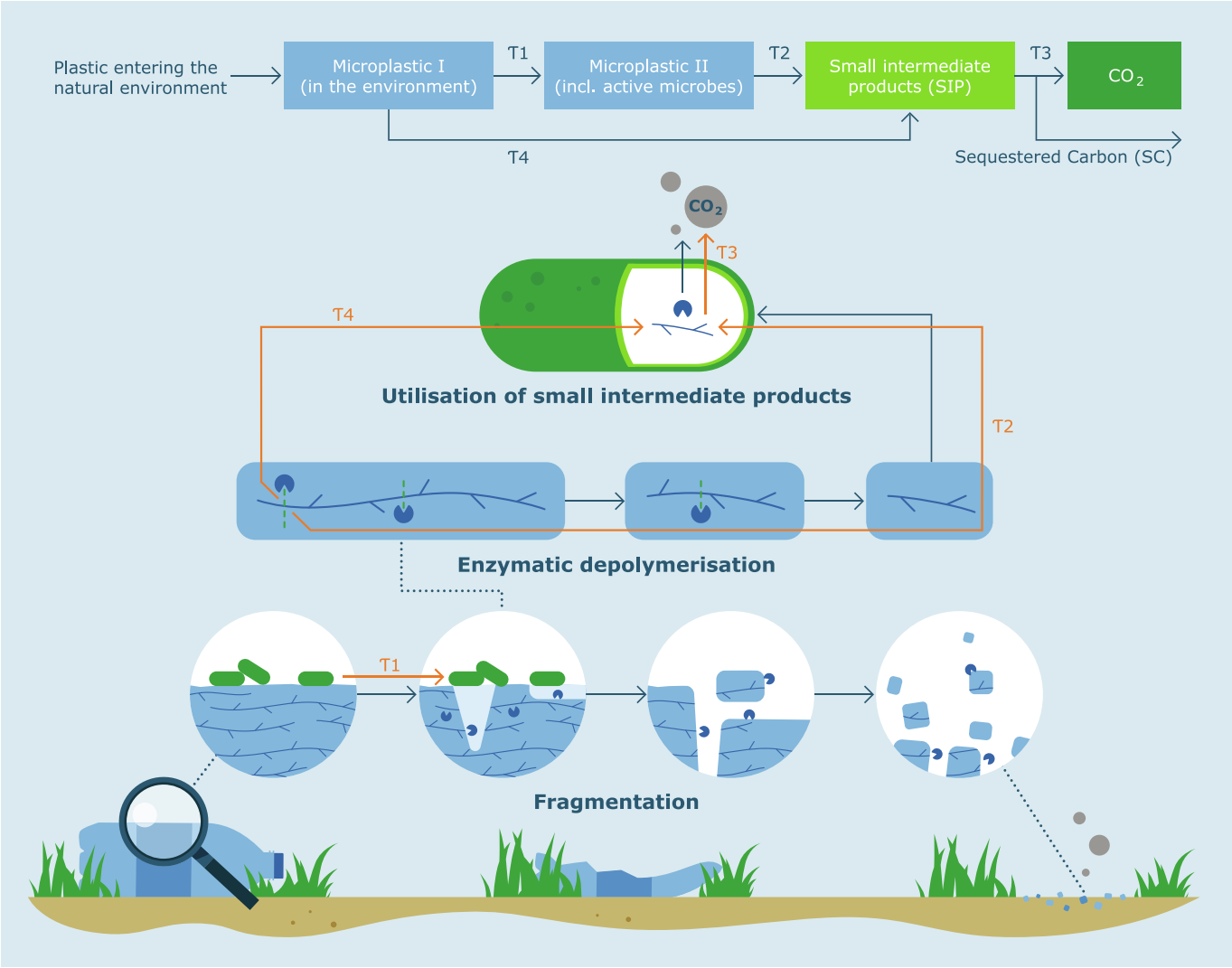

Fig. 3. Schematic representation of the model and the degradation processes and phases that are simulated, τ1…τ4 indicate the time constants of the processes.

potential) and worst case (highest accumulation potential) is given based on the variations in the model output.

The accumulation potential has a maximum value of 1 when all carbon mass stays in the micro-I or micro-II phase, i.e. all carbon mass remains in the environment as microplastics. The accumulation potential of PE is very close to this maximum value (0.99), which indicates that PE is a very persistent material. In contrast, the maximum accumulation potential of PBS is found to be 0.002, which reflects its fast biodegradation and low persistency in soil. The modelled accumulation potential values of PLA lie in between those of PBS and PE in a range of 0.30–0.88.

The obtained differences in accumulation potential can be related to the known biodegradation characteristics and chemical compositions of the polymers. PE is an addition polymer with a carbon-carbon (C—C) backbone. This long C—C backbone structure is not found in natural organic matter and not susceptible to hydrolysis, which is considered a crucial step in biodegradation in many environments (Lott et al., n.d.; European Commission and Directorate-General for Research and Innovation, Biodegradability of Plastics in the Open Environment, 2021). As a result, PE tends to be non-susceptible to biodegradation in virtually any environment, although specific examples of micro-organisms that break down PE do exist (Zhang et al., 2022). PBS and PLA, on the other hand, are polyesters, a class of polymer that is often found in natural polymers and can be hydrolysed under the right conditions. As a result, many polyesters are susceptible to biodegradation, although the environmental conditions govern the biodegradation kinetics and certain polyesters are more susceptible to biodegradation than others in common biological environments. Asides from the environmental conditions, the chemical properties (e.g. molecular weight, tacticity and hydrophilicity) and physical properties (e.g. crystallinity and glassiness) of the polymer also determine whether a polymer will degrade and at which speed this will occur (Wang et al., 2024). However, as the exact interplay between polymer structure and the environmental conditions is very complex, it is not yet fully understood why PBS degrades much faster in soil than PLA.

3.4. Case studies

To exemplify how this model can be used to guide material selection in product design, two case studies were performed: the littering of plastic candy wrappers and the seasonal use of agricultural mulch film. For each case, multiple scenarios were evaluated. The results of these case studies can be used directly to assess and compare the accumulation of different polymers, or could be used as input for sustainability studies such as a life cycle analysis (LCA).

3.4.1. Case 1: Littering of plastic candy bar wrapper(s)

In this case study we analyse the effect of polymer selection (PE, PLA

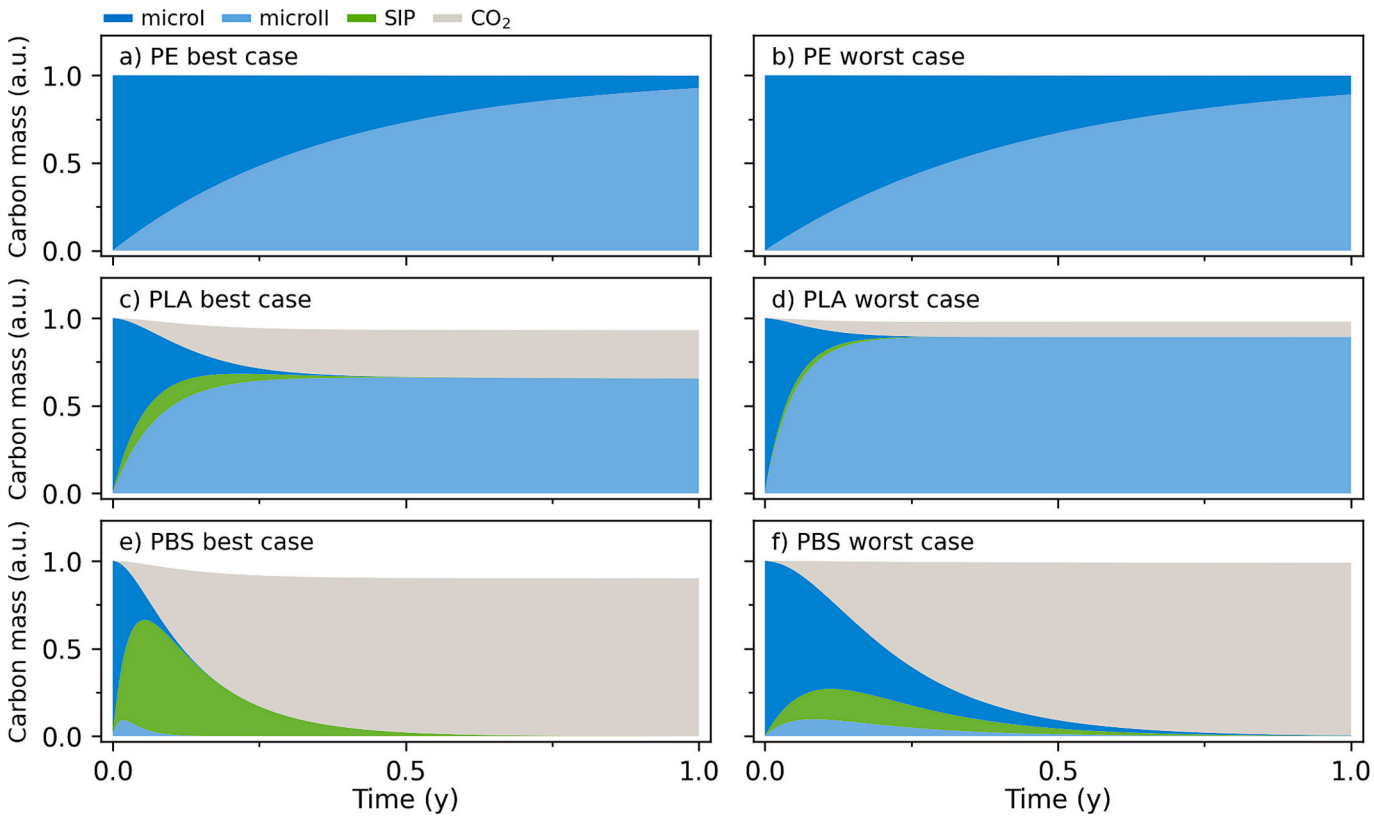

Fig. 4. Prediction of the carbon mass over a one-year timespan for all four phases of the biodegradation process for a) PE, b) PLA and c) PBS.

Table 2
Modelled accumulation potential for different polymers and the corresponding time constants. The accumulation potential is defined as the time-integrated concentration of microI+microII for 1 unit of carbon mass over a timeframe of 100 years.

Polymer		Accumulation potential [−]	τ1 (y)[a]	τ2 (y)[a]	τ3 (y)[a]	τ4 (y)[a]	α
PE							
Albertsson, 1980	Best	0.990	0.38	6000	0.14	300	0.80
	Worst	0.991	0.45	6300	0.095	360	0.80
PLA							
Satti et al., 2018	Best	0.723	0.11	270	0.096	0.70	0.80
	Worst	0.858	0.11	5200	0.095	0.71	0.80
Thompson et al., 2019	Best	0.301	0.11	53	0.073	0.22	0.80
	Worst	0.659	0.11	1800	0.073	0.23	0.80
Saadi et al., 2012	Best	0.874	0.054	2400	0.048	0.45	0.80
	Worst	0.883	0.054	4600	0.048	0.45	0.80
PBS							
Šerá et al., 2020 lab 1	Best	$3.0{\cdot}10^{-4}$	0.070	0.012	0.12	0.048	0.90
	Worst	0.002	0.31	0.63	0.064	0.077	1.00
Šerá et al., 2020 lab 2	Best	$7.0{\cdot}10^{-4}$	0.061	0.041	0.18	0.16	1.00
	Worst	0.002	0.30	0.048	0.058	0.37	0.99

[a] In case multiple fit results have the lowest/highest accumulation potential, the time constants are given for the fit with the lowest condition number.

or PBS) on the microplastics accumulation of a candy bar wrapper that enters the natural environment in three different littering scenarios. In all scenarios the dimensions of the candy wrapper are independent of the polymer type. In scenario 1 A, the model is used to predict the microplastics mass that is present in the natural environment over time after littering one candy bar wrapper. In scenario 1B, continuous littering of candy bars is assessed. The functional unit of this assessment is 45 million candy bars, which is roughly the amount candy bar wrappers for chocolate products on the Dutch market in 2019 (Seters and Brunt, n. d.). The littering potential of the candy bar wrappers is assumed to be equal for the different polymer types with a value of 0.2 %. As littering can be considered to be a constant process, the effect on carbon mass of the microplastics is determined by calculating the step response of the model. A more detailed calculation of the amount of polymer that is entering the environment in the different scenarios and a justification of the chosen littering potentials is given in the supplementary material S.7. Based on the best and worst case scenarios of the accumulation potential and their corresponding time constants (Table 2), the total carbon mass of the microplastic stages is modelled (Fig. 5). The bandwidths in the figure are caused by the variation between the best and worst case scenario which in some case originate from different data sources. The time-integrated mass of microplastics in the environment

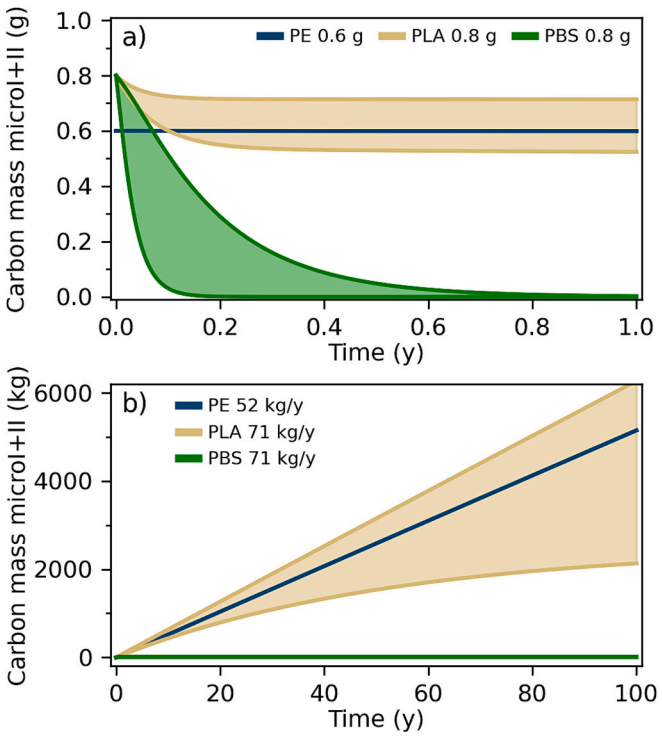

Fig. 5. The carbon mass of microplastics (micro-I + micro-II) over time for different polymers modelled as a continuous inflow in the system given the best and worst scenario of the accumulation potential (Table 2). a) The amount of microplastics as function of time (up to 1 year) for the littering of one candy bar wrapper (scenario 1 A), b) The amount of microplastics over 100 years assuming a constant littering potential of 0.2 % per year (scenario 1B).

over a timeframe of 100 years and the amount of microplastics in the environment after 100 years is provided in Table S7 and the modelled CO_2 production is given in de Fig. S12.

The differences in microplastics accumulation of candy wrappers made of different polymers are determined by a combination of the littering potential and the polymer accumulation potential. The microplastics that are formed during PBS biodegradation are only present in the natural environment for a short period of time (see Fig. 5a). Even when PBS is littered continuously, the microplastic build-up in the environment is limited (Fig. 5b). In contrast, due to the higher accumulation potential of PE and PLA, the majority of the carbon in these polymers remains in the microplastics phase in the first year after littering a single candy wrapper (Fig. 5a). In case these plastics are continuously littered, the microplastics accumulate in the natural environment (Fig. 5b). Even when the littering rate of biodegradable packaging (like PBS) is increased to 3.5 % the amount of microplastics is still negligible compared to that of slow biodegrading polymers with a low littering potential (Fig. S13). Hence, although biodegradability should not provide a 'license to litter', high polymer biodegradation rates can prevent microplastic accumulation in the natural environment caused by littering.

3.4.2. Case 2: Seasonal input of agricultural mulch film

As opposed to plastics that continuously enter the natural environment, there are also plastic products that are applied in a very specific timeframe and consequently enter the environment in a pulsed manner. A clear example is the use of agricultural mulch films, that are typically placed on the fields once a year. After harvest of the crop, these films are (partially) removed, or left on the field to biodegrade. In this case study we compare and analyse the effects of agricultural films made from different polymers (PE, PLA and PBS) in two scenarios.

In scenario 2 A all agricultural films are used on the land and are retrieved after harvest with an equal recovery ratio for all polymers. The recovery ratio is determined by a multitude of factors such as the fragmentation of the films due to weathering, handling losses, parts of the film that are accidentally ripped off, parts that are accidentally left on the field and parts that are taken by animals. In scenario 2B, the PBS film is purposely left on the field to biodegrade and is compared to PE and PLA films that are retrieved after use. In both case studies, the amount of residual plastic after harvest is calculated and is used as input for the model to predict the amount of microplastics over time given a yearly inflow of this amount (Fig. 6). As in the previous case study, the best and worst case scenarios of the accumulation potential and their corresponding time constants are used and the provide bandwidths shown in the fig. A more detailed calculation of the amount of polymer that is entering the environment in the different scenarios and a justification is given in the supplementary material S.8. Furthermore, the time-integrated mass of microplastics in the environment over a timeframe of 100 years and the amount of microplastics in the environment after 100 years is provided in Table S10 and the modelled CO_2 production is given in de Fig. S14.

This case study shows the effect of seasonal emission of plastics into agricultural soil (Fig. 6). Both scenarios show that biodegradable PBS forms spikes in microplastic concentration that diminish quickly. These spikes are higher or lower depending on the scenario: whether or not the PBS is recovered from the land and the recovery rate of the plastic after use. In contrast, the other polymers (PE and PLA) form persistent microplastics in the environment that accumulate over time by their continued use. In case the PBS film is retrieved from the land with the same recovery rate as the other polymers, the microplastic concentration of PE and PLA exceed the temporally formed PBS microplastics directly after the first year. In case the PBS film is left on the land, at first the PBS microplastic concentration will be higher than the amount of microplastics resulting from PE and PLA. However, these spikes of high microplastic concentration are only present at a certain season in the year and will diminish quickly. Furthermore, with continued use of these plastic films at the same location, the accumulating amount of microplastics from PE and PLA will eventually exceed the PBS microplastic concentration for any of the assessed scenarios. Hence, agricultural films with high biodegradation rates can prevent microplastic accumulation in soil compared to the consistent use of non-biodegradable films.

4. Discussion

4.1. Model limitation and justification

The presented model can be used to assess and compare the accumulation of microplastics of different polymers in the natural environment. As with all predictive models, there are limitations of the specificity and accuracy of this model.

The input data for the model are the experimentally determined mineralisation curves (CO_2 evolution). The fact that this is one of the few standardised data sources that can be obtained from a biodegradation process, makes the model practically applicable. Nevertheless, the accuracy of the model would greatly benefit from the availability of quantitative data sources on the mass of the intermediate states (microplastics and SIPs) during biodegradation. Unfortunately, as this data is difficult to gather it is currently not available. Another factor that impacts the model accuracy in this study is the timeframe over which the CO_2 evolution was measured for the polymers that are compared. For both PE and PLA, the CO_2 evolution that was used as input for the model was terminated well before full biodegradation was obtained (3.8 and 0.6 years respectively). Although this makes sense from an experimental time and cost perspective it is anticipated that the model performance greatly improves with the availability of prolonged biodegradation measurements. Still, with the available data, the model already demonstrates the ability to predict clear differences in the accumulation

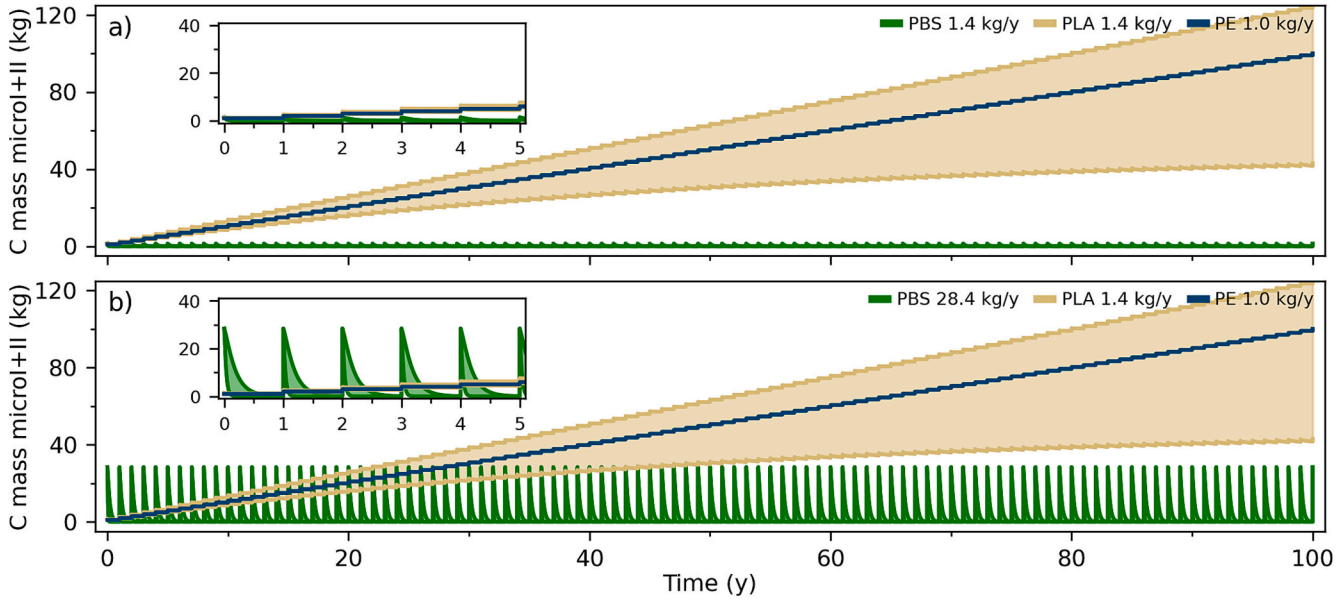

Fig. 6. The carbon mass of all microplastics (micro-I+micro-II) over time for different polymers assuming a pulsed inflow in the system over a period of 100 years (inset: period of 5 years). a.) with a recovery ratio of 95 % for all polymers (scenario 2 A), b.) PBS is left on the land to decompose (recovery ratio of 0 %) and PE and PLA are retrieved with a recovery ratio of 95 % (scenario 2B).

potential for different types of polymers. When more accurate data becomes available, more accurate predictions can be made.

Another consequence of the usage of experimental CO_2 evolution curves is that each model prediction is linked to a specific plastic material in a specific environment. Differences in polymer grade (e.g. crystallinity, additives, *co*-polymer content or blends), environments (soil type, compost, fresh or salt water) and environmental conditions (e.g. temperature, UV radiation, oxygen availability, humidity or chemical circumstances) yield different biodegradation data which subsequently results in different predictions for the accumulation potential. On the one hand, these anticipated predictions correspond to the differences in the observed biodegradation for specific polymer-environment combinations which is a unique aspect of the proposed model and offers the possibility to not only compare between polymers but also compare the biodegradation behaviour of one polymer in different environments or under different environmental conditions. On the other hand, for specific accumulation descriptions, accurate experimental input data is essential. The model proposed in this study has been applied to polymers in a soil environment and should be further validated for the application in other circumstances (e.g. aquatic environments). As biodegradation is measured under various environmental conditions (incl. Aquatic, marine, home or industrial composting, etc.) using similar methods with comparable output (i.e. CO*2* evolution curves) it is anticipated that the model can be applied for other environments and conditions as well. Although the presented model is fully targeted at aerobic biodegradation, it would also be possible to include anaerobic digestion processes. In that case the formation of methane (CH_4) needs to be included in the mass balance. Based on the experimentally acquired CH_4 and CO_2 evolution curves the accumulation of a specific polymer in an anaerobic environment can then be determined using the same methodology.

The final consideration that must be accounted for when using experimental CO_2 evolution data is that these measurements are typically performed on powdered materials. Therefore, the model describes the process from microplastic to CO_2 and sequestered carbon. However, plastic materials generally enter the environment in other states. In the case of product wear and disintegration (e.g. tire abrasion, textile microfibre shedding) the polymer may enter the environment as microparticles, but in the case of littering or outdoor product usage without recovery, the initial fragmentation of the plastic object needs to be added to the model. This could be achieved by either measuring the mineralisation curves of polymer objects or by including the fragmentation step as another time constant to the model. For the latter option, object-specific data on the fragmentation rate would be required for an accurate prediction of the accumulation potential of a plastic product. Nevertheless, the relative differences in accumulation potential observed for PE, PLA and PE are already considerable and are not expected to alter significantly upon inclusion of this factor in the model.

4.2. *Model implementation*

This study provides an integrated and widely applicable approach to determine the accumulation potential of microplastics originating from plastic product use. The output of this model can be used directly to compare the polluting effects of different polymers. In addition, the model output can be used in environmental assessments such as LCA studies. By implementing this model in future LCA studies, different polymer types can be adequately compared using the same model for all polymers.

In contrast to other proposed methodologies, our model describes biodegradation of polymers as a full conversion to CO_2 and minerals and is also suitable for non- and slow degrading polymers such as PE and PLA. The model only predicts the quantity of microplastics and SIPs over time and does not differentiate between the effects that the precise morphology and chemical nature of these products can have on species, ecosystems or human health. These effects of microplastics are currently not fully understood and the most important impact seems to be related to the concentrations and residence times of the microplastics in the natural environment rather than their chemical nature (Lavoie et al., 2022; Corella-Puertas et al., 2023), which makes the output of the model already a good approximation of the related environmental impacts. However, clear differences between microplastics of different chemical nature could be used as characterisation factors in an LCA. Additionally, this study does not differentiate between the exact shapes and sizes of the plastic fragments in the micro I and micro II phase and therefore all carbon mass in this phase is treated as microplastics. In practice, parts of the carbon in these phases will probably be sufficiently small to formally categorize as nano-plastics which are reported to have different toxicological effects (Yin et al., 2021). This study does not allow for

distinction between different plastic fragment morphologies as quantitative data on the mass and size of the carbon in these intermediate states is currently not available. Furthermore, this study does not include the accumulation and impact of plastic additives that are omnipresent in plastic formulations and are suspected of having a relatively high impact on the environment in which they accumulate (Pinaeva and Noskov, 2024). New insights and data that are expected to be generated by future studies on these topics can be used in LCA studies in combination with the model presented in this study.

The results presented in this study show that the total concentration of microplastics in the environment depends on both the amount and speed at which plastic are being emitted to the environment and the biodegradation rate of the polymers of which they are composed. When the average emission, either continuous or pulsed, outweighs the biodegradation rate, the system moves towards a near steady state (provided constant input). This phenomenon is also described by Pecchiari, et al. (Pecchiari et al., 2024) and was experimentally demonstrated by Ghimire, S., et al. (Ghimire et al., 2020). This implies that even when only rapidly biodegradable polymers are used in certain system, a consistent dose of microplastics will still be present. However, for non-degradable or slowly degrading polymers the near steady-state would only be reached after thousands of years. In a system where the overall biodegradation rate outweighs the input of new plastic material, the near steady state will be net zero which implies that no accumulation of microplastics is observed. Upon striving for a system without persistence of microplastics, the anticipated system input (e.g. littering potential) can be used to determine a threshold value for the accumulation potential of a polymer to aid material selection in product design.

5. Conclusions

This study proposes an integrated and universally applicable biodegradation model that enables the assessment and comparison of microplastics formation and accumulation of different polymers in a specific natural environment. Based on experimentally determined mineralisation curves (CO_2 evolution), carbon mass flow streams are calculated that predict the concentration and residence time of the different plastic states (e.g. microplastics, small intermediate products, CO_2) during the biodegradation processes. In doing so, the model effectively converts the biological biodegradation processes of polymers in a wide range of environments to input data that can be utilized by LCA and other environmental (system) analysis to describe the impact of polymer accumulation. In the current study, the model was applied to three polymers (PE, PLA and PBS) in a soil environment at natural temperatures.

The use of the model is demonstrated with two case studies: the littering of plastic candy wrappers and use of agricultural mulch films. For plastic candy wrappers, a continuous influx of littered products yields only a marginal increase in microplastic accumulation over time when it is made of a plastic having a sufficiently high biodegradation rate. For mulch films, which are placed on the land periodically, the use of biodegradable plastics results in periodical spikes of the microplastic concentration in soil. However, provided the biodegradation rate is sufficiently high, a net zero steady state will be observed that outweighs the input of new plastic material and subsequent does not lead to plastic accumulation.

Ultimately, the proposed methodology can be used to determine the required biodegradation behaviour to prevent the microplastic accumulation of specific plastic products in the natural environment. This will facilitate the transition to a system in which products are designed based on their intended functionality and anticipated end-of-life scenario.

CRediT authorship contribution statement

Marieke T. Brouwer: Writing – review & editing, Writing – original draft, Visualization, Methodology, Formal analysis, Conceptualization. **Wouter Post:** Writing – review & editing, Conceptualization. **Maarten van der Zee:** Writing – review & editing, Validation. **Rob Reilink:** Writing – review & editing, Methodology. **Remko Boom:** Writing – review & editing. **Evelien Maaskant:** Writing – review & editing, Methodology, Formal analysis, Conceptualization.

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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.177503.

Data availability

The data sources used in this study are specified in de Supplementary Materials

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