



# Methodology to address potential impacts of plastic emissions in life cycle assessment

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## Abstract

**Purpose** Products made of plastic often appear to have lower environmental impacts than alternatives. However, present life cycle assessments (LCA) do not consider possible risks caused by the emission of plastics into the environment. Following the precautionary principle, we propose characterization factors (CFs) for plastic emissions allowing to calculate impacts of plastic pollution measured in plastic pollution equivalents, based on plastics' residence time in the environment.

**Methods and materials** The method addresses the definition and quantification of plastic emissions in LCA and estimates their fate in the environment based on their persistence. According to our approach, the fate is mainly influenced by the environmental compartment the plastic is initially emitted to, its redistribution to other compartments, and its degradation speed. The latter depends on the polymer type's specific surface degradation rate (SSDR), the emission's shape, and its characteristic length. The SSDRs are derived from an extensive literature review. Since the data quality of the SSDR and redistribution rates varies, an uncertainty assessment is carried out based on the pedigree matrix approach. To quantify the fate factor (FF), we calculate the area below the degradation curve of an emission and call it residence time  $\tau_R$ .

**Results and discussion** The results of our research include degradation measurements (SSDRs) retrieved from literature, a surface-driven degradation model, redistribution patterns, FFs based on the residence time, and an uncertainty analysis of the suggested FFs. Depending on the applied time horizon, the values of the FFs range from near zero to values greater than 1000 for different polymer types, size classes, shapes, and initial compartments. Based on the comparison of the compartment-specific FFs with the total compartment-weighted FFs, the polymer types can be grouped into six clusters. The proposed FFs can be used as CFs which can be further developed by integrating the probability of the exposure of humans or organisms to the plastic emission (exposure factor) and for the impacts of plastics on species (effect factor).

**Conclusions** The proposed methodology is intended to support (plastic) product designers, for example, regarding materials' choice, and can serve as a first proxy to estimate potential risks caused by plastic emissions. Besides, the FFs can be used to develop new CFs, which can be linked to one or more existing impact categories, such as human toxicity or ecotoxicity, or new impact categories addressing, for example, potential risks caused by entanglement.

**Keywords** Degradation · Plastics · LCIA · SSDR · Fate · Impact · Plastic emissions

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## 1 Background and aim

Life cycle assessment (LCA) is a methodology used to estimate potential environmental impacts of products and processes, such as global warming caused by greenhouse gas emissions. LCA studies often estimate environmental impacts of plastic products to be lower than those of alternatives, e.g., due to light-weight design or a lower resource input (Amienyo et al. 2013; Humbert et al. 2009; Saleh 2016). One reason that the LCA studies come to this conclusion might be that plastic emissions caused by loss

of plastics, e.g., by abrasion, aging, fragmentation, or littering (Sonnemann and Valdivia 2017), are currently not well considered by an appropriate impact category.

Plastics can be found in nearly all ecosystems worldwide (Li et al. 2020). Even though the impacts are not fully understood, there is a clear consensus between politicians, industry, and consumers that plastic should not be released to the environment (Nielsen et al. 2020). It is assumed that plastics in the environment can threaten biodiversity and ecosystem services, such as fish reproduction, which negatively influence the economy, such as the fishery or tourism industry (Burns and Boxall 2018). There is also initial evidence that microplastics might cause inflammatory bowel diseases in humans (Yan et al. 2022). In order to address these effects, LCA should take into account emissions of plastics into the environment and their potential impacts (Schwarz et al. 2019; Sonnemann and Valdivia 2017; Woods et al. 2016).

In order to assess the potential risks caused by plastic emissions, two steps are needed: first, the definition and quantification of plastic emissions, and second, the estimation of their fate in the environment. Current research focuses on the leakage of plastics, which covers both micro- and macroplastics, e.g., Siegfried et al. (2017), Civancik-Uslu et al. (2019), Unice et al. (2019), Peano et al. (2020), Chitaka and von Blottnitz (2021), Stefanini et al. (2021), and Amadei et al. (2022). However, this knowledge has only partially been linked to life cycle inventory (LCI) flows, and corresponding characterization factors (CFs) for plastic emissions are lacking.

Besides information regarding leakage amounts, suitable elementary flows and corresponding CFs need to be defined to incorporate plastic emissions into LCA. A CF describes the extent to which the elementary flow contributes to an impact category, e.g., global warming or a prospective new impact category. In this paper, we develop the basis for a new impact category “plastic pollution” which addresses potential hazards caused by plastic emissions. According to the framework proposed by Udo de Haes et al. (2002), CFs are defined as follows:

$$\text{Characterization factor} = \text{Fate factor} \times \text{Exposure factor} \times \text{Effect factor} (\times \text{Severity Factor}) \quad (1)$$

- The fate factor (FF) refers to the transmission and distribution of a plastic item between the environmental compartments and its degradation within the environmental compartments. Plastics initially released into one compartment (e.g., a river) might be transmitted to another compartment (e.g., the ocean), where they degrade. Since degradation rates differ between the compartments, it is necessary to know the compartment where most of the degradation takes place.

- The exposure factor addresses the probability of exposure of humans or animals to the plastic emission, e.g., by ingestion, inhalation, and entanglement.
- The effect factor represents the sensitivity of a species to the pollutant.
- The severity factor varies with the expected severity of the damage. A severity factor is to be specified when calculating an endpoint indicator. Therefore, the severity factor is given in parenthesis in the formula for calculating the CF.

A first attempt to calculate FFs was made in the conceptual framework SecuPlast of Croxatto Vega et al. (2021), in which the formation of secondary microplastic due to photooxidation in LCA is tackled. The exposure of organisms to microplastics has been investigated for selected species (e.g., Cole et al. 2011). Although exposure and effects caused by the intake and accumulation of microplastics such as starvation, disturbances of the reproduction and in energy metabolism, or changes in liver physiology (Anbumani and Kakkar 2018), as well as toxic reactions to specific polymers (e.g., Gagné 2017) or additives (Gallo et al. 2018) have been described, the consideration of the effects of different polymers and shapes at the normal concentration found in the environment remains challenging. Woods et al. (2016) structurally investigated research needs to quantify marine ecological impacts. Within the MariLCA project, this work is currently expanded to cover other environmental compartments such as soil, freshwater, or air (Boulay et al. 2021; Woods et al. 2021). A first effect factor was proposed by Woods et al. (2019) and extended by Høiberg et al. (2022), which conveys the risk of entanglement of organisms with a critical size compared to the size of the plastic item. Even less knowledge is available concerning the severity of the effects caused by plastic emissions. In another recent study, Lavoie et al. (2021) developed an effect factor for micro- and nano-sized plastics regarding different aquatic species. They found effect differences between polystyrene and other polymers when analyzing many aquatic species

(Lavoie et al. 2021). Saling et al. (2020) have proposed a life cycle impact assessment (LCIA) approach to address the potential ecological impacts of microplastics in oceans. Although this approach links dose-dependent experiments, in particular effect concentrations (EC50) and lethal concentrations (LD50), to different polymers and shapes of microplastics, there is still limited knowledge concerning adverse impacts in the environment and the consequences of increasing bioaccumulation of hydrophobic organic

compounds into organisms (ECHA 2019; Koelmans 2015). Another attempt to cover the impacts of direct microplastic emissions to freshwater was conducted by Salieri et al. (2021), who couple degradation models with the USEtox framework to calculate CFs for a limited number of polymer types. In this article, we focus on plastic emissions (the loss of plastic items into the ecosphere) and their fate in the environment, particularly their residence time, dependent on the emission's degradation in the environment. Usually the risk potential of substances is addressed by well-established classes of substances such as persistent, bioaccumulative and toxic (PBT) or very persistent and very bioaccumulative (vPvB) substances. Persistent substances that are not bioaccumulative or toxic have not been characterized as risky. However, the potential risk of plastics seems to be mainly driven by their extreme persistence (ECHA 2019). Hence, following the precautionary principle, we propose a CF that focuses on plastics' residence time in the environment and does not consider the exposure and effect of plastic emissions. Accordingly, at this stage, we assume that the exposure and the effect factor in Eq. (1) both equal 1 and the CF is solely dependent on the FF. However, in the future, this FF can be supplemented by exposure and effect factors to develop CFs linked to one or more existing impact categories such as human toxicity or ecotoxicity or as a starting point for new impact categories focusing, for example, on physical impacts on biota, invasive species, or potential impacts on cultural heritage.

The emission's persistence in the environment is dependent on its degradation. In literature, the term degradation is used for changes in chemical or physical properties, depolymerization, mass loss, or mineralization (Chamas et al. 2020). In general, degradation refers to the reduction of mass through mineralization, measurable by the production of CO<sub>2</sub> (or CH<sub>4</sub> under anoxic conditions), or the consumption of O<sub>2</sub>. However, many environmental degradation studies typically measure the loss of mass, since direct measurements of gas production is not possible. In order to consider these studies, we also consider mass loss as measure for degradation although this might lead to overestimated degradation rates because mass loss may include fragmentation. The term degradation addresses both, biodegradation as characterized by the three processes biodegradation, biofragmentation, and assimilation (Lucas et al. 2008) as well as other forms of degradation such as photodegradation which is usually mainly driven by oxidation and hydrolysis. Saling et al. (2020) highlighted that weight loss is associated with (bio)degradation and fragmentation. The process of fragmentation is driven by UV radiation, thermal oxidation, mechanical weathering and occurs along with chemical degradation.

## 2 Methods and materials

As a prerequisite to integrate the plastic-related elementary flows into LCIA, a convention for defining plastic emission types is proposed (cf. "Sect. 2.1"). Then, FFs are calculated per plastic emission type based on the following elements:

- Redistribution patterns and the final compartment share of plastic emissions (cf. "Sect. 2.2");
- Degradation rates for each plastic emission type and environmental compartment (cf. "Sect. 2.3").

The FF of a plastic emission (characterized by a particular polymer type, size, and shape and initially released into a particular environmental compartment) is determined by the final compartment share after a possible redistribution and the expected degradation times in those compartments compared to a reference degradation time, which is set to 1 year in our methodology. We chose this reference time since plastics degrade at different rates in different environmental compartments. Moreover, by choosing a reference time we avoid uncertainties related to the measurement of the degradation of a certain plastic. The resulting FF is expressed as kg plastic pollution equivalent (PPE) per kg plastic emitted. The FF allows for a comparison of various plastic emissions and enables assessing emissions to different compartments by considering the different degradation rates.

Since degradation rates vary in different compartments, ideally, various environmental compartments should be considered, such as the marine environment (eulitoral, pelagic, benthic), freshwater systems, marine and river sediment, soils, and air. However, since the knowledge about the transfer rates and degradation rates of plastic items in different compartments is currently very limited, we propose to consider plastic emissions exclusively into the initial compartments fresh or marine water, soil, and air, and redistribution only to the final compartments marine water, marine and river sediment, and soil, as a first step.

The requirements regarding the definition of elementary flows and initial compartments and the materials these requirements are based upon are outlined in "Sect. 2.1." "Sect. 2.2" explains the patterns underlying plastic redistribution in the ecosphere based on polymer-specific and generic research regarding transport processes in the environment. The calculation of the residence time is dependent on polymer and compartment specific degradation rates extracted from a comprehensive literature review and equations developed by the authors to describe the persistence of plastic emissions in the environment. Details

regarding the materials used and the formulas derived can be found in “Sect. 2.3.” “Sect. 2.4” explains how specific surface degradation rates are calculated based on the degradation model. Finally, “Sect. 2.5” illustrates the data quality assessment applied to the literature-based data and its implications for further use.

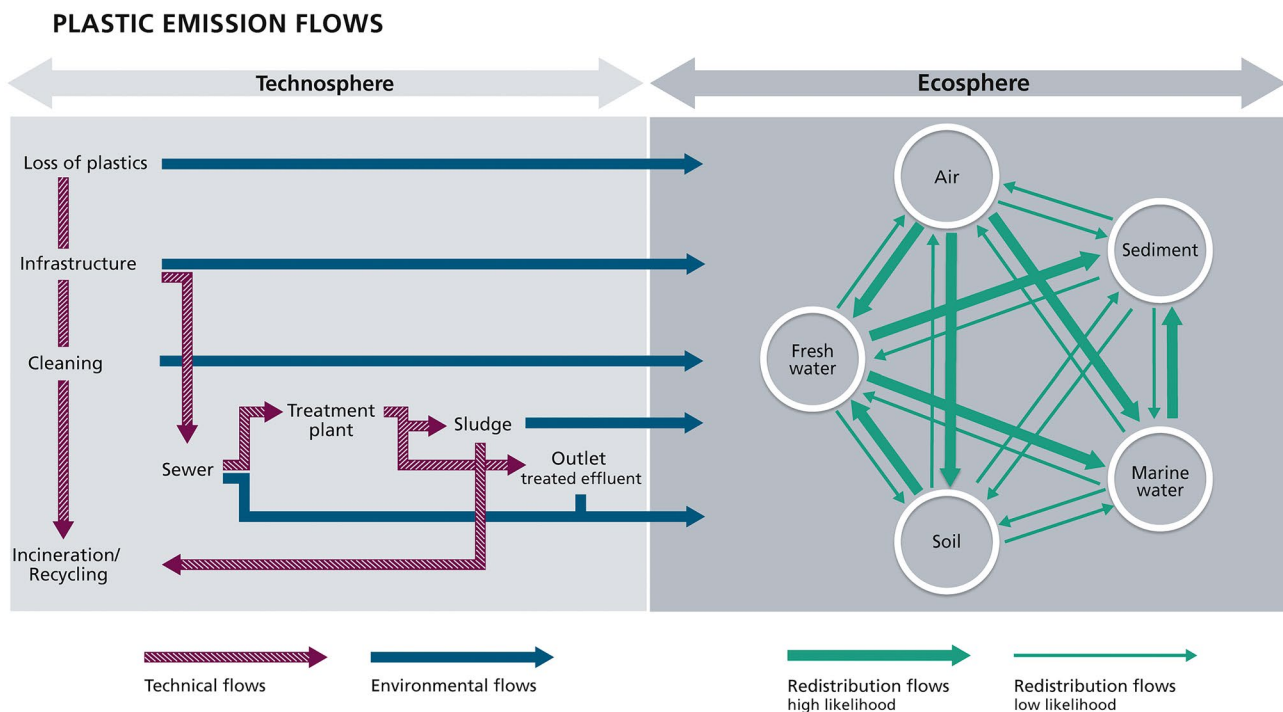
## 2.1 Defining elementary flows and initial compartments

Elementary flows are used to describe a plastic flow from the technosphere into an environmental compartment of the ecosphere. For each elementary flow, it is necessary to quantify the initial release rate of the corresponding plastic. Following the definition of Edelen et al. (2017), the initial release is the quantity of plastic emissions that leaves the technosphere and enters the ecosphere. Examples of plastic losses that directly enter the ecosphere are lost fishing nets in the sea, the burying of mulch films, or other plastic applications in an open environment, such as geotextiles which are not removed. However, the challenge is that the boundaries between the technosphere (e.g., streets or sewage systems) and the ecosphere (e.g., soil or freshwater) are often unclear (Maga et al. 2020). In this paper, we differentiate between technical flows like microbeads in wastewater, environmental flows addressing the initial release, such as microplastics directly emitted to agricultural soil, and redistribution flows which occur between different environmental compartments.

Figure 1 presents the distinction between these three flow types and the boundaries between the technosphere and the ecosphere made in this paper. It is visualized with a simplified model that shows the possible pathways of plastic emission from the point of loss to sinks. Redistribution flows that are assumed to have a higher probability are displayed in bold, those with a smaller likelihood are thinner.

The initial compartment is the environmental compartment where a plastic item is first emitted from the technosphere. For example, during a picnic in the park, plastic cutlery might be emitted onto urban soil. Another example is the emission of plastic microbeads, which can be found in some cosmetics. They most likely reach a wastewater treatment plant as part of the wastewater, where they are either retained and later partially emitted onto agricultural soil as part of sewage sludge or pass the treatment process and are emitted to freshwater. Since degradation speed and transport processes between environmental compartments are country-specific, e.g., dependent on soil and water temperatures or the ratio of water to land, region-specific elementary flows should be defined and characterized, where possible.

Besides the initial compartment, the polymer type, shape, and size of plastic emissions influence transport characteristics and degradation speed. Therefore, these attributes need to be included in the definition of elementary flows. The size, shape, and material type are crucial parameters concerning potential effects (de Ruijter et al. 2020). Regarding the shape, the emission can be



**Fig. 1** Simplified model to address plastic flows between technosphere and ecosphere

characterized as a film, a fiber, or a nearly spherical pellet or particle. To simplify, larger plastic items such as bags or cutlery are considered formed film in this publication. The characteristic length refers to the emission's diameter (fiber, particle) or thickness (film). As shown in "Sect. 2.3," the shape and characteristic length of an emission and the environmental compartment to which the emission is finally redistributed strongly influence its degradation time. Therefore, these attributes should be part of the definition of the elementary flow. Ogonowski et al. (2016) have shown for *Daphnia magna* that plastic items' shape is relevant for exposure and potential impacts. Likewise, Ziajahromi et al. (2017) found differences in effects on the water flea *Ceriodaphnia dubia* between beads and fibers. Although the knowledge about the influence of the shape of plastic emissions on adverse impacts in organisms is limited to date, the shape might also be relevant to address the potential effects of plastic emissions more in detail in the future.

The naming of the elementary flow, therefore, should include first the region, second the material type (including specification, e.g., rigid vs. foam), third the shape of the plastic emission (film, fiber, or particle), fourth the characteristic length of the emitted plastic item. Fifth, according to Edelen et al. (2018), the initial environmental compartment into which the plastic is emitted should be part of the definition.

In order to cover the majority of possible plastic emissions, we propose to differentiate between the following three ranges of characteristic lengths as a proxy: <0.1 mm, 0.1–1 mm, and > 1 mm. These size classes address the range of the characteristic length of films, fibers, and particles which are typically released to the environment either as microplastic or as macroplastic emissions. As a conservative assumption, to not overestimate the degradation rate, each residence time (cf. "Sect. 3") is calculated using the maximum characteristic length of the respective class. For the class of the largest emissions, a characteristic length of 10 mm is used. When calculating a specific well-known plastic emission's FF, the exact characteristic length should be used instead (cf. "Sect. 2.3").

Some frequently used LCA processes, such as a transport process via truck, result in various plastic emissions such as tire wear and abrasion of road markings. These plastic emissions should be treated as different flows. Besides, if a plastic emission consists of various polymers on a single-particle level (e.g., tire wear consists of natural rubber and synthetic rubber), it should be treated as one flow. In this case, degradation data should be used that reflects the degradation behavior of this specific emission type. If degradation rates are unavailable for this specific emission type, the degradation rate of the slowest degrading polymer type of the complex should be used as a conservative estimation.

Various approaches to estimate the initial release of macro- and microplastics exist (Maga et al. 2020), e.g., Kawecki and Nowack (2019) for Switzerland and Peano et al. (2020) and Boucher et al. (2020) for other countries.

## 2.2 Estimation of redistribution between environmental compartments

In order to estimate the redistribution of an emission between different environmental compartments, as presented in Fig. 1, several research papers addressing the fate of plastics were analyzed. For some plastic emissions, specific data could be extracted. For instance, according to the research conducted by Unice et al. (2019) about the Seine watershed (France), tire wear particles initially accumulate on the road and are washed off by rain equally into the road runoff (water) and onto the surrounding soil. Considering the sewage system and partial re-emittance of tire wear particles as part of the sludge, only 24% of the particles are accurately managed. The rest is emitted to and redistributed in the environment with a final compartment share of 56% on soil, 13–16% in river sediment, 2–5% in the ocean, and 2% in air. However, we assume that the 2% emitted to air do not remain in air but are redistributed to soil and water, as presented in Table 1.

For other plastic emissions, data concerning redistribution and compartment shares are unavailable. In these cases, assumptions are made based on parameters suggested in the literature, which affect the behavior of plastic emissions in the environment, such as:

- Environmental compartment initially emitted to (e.g., Kawecki and Nowack 2019);
- Density of the plastic (e.g., Kowalski et al. 2016; Nizzetto et al. 2016; Horton and Dixon 2018);
- The emission's size and shapes (e.g., Chubarenko et al. 2016; Fazey and Ryan 2016; Kowalski et al. 2016).

Regarding the environmental compartment the plastic is initially emitted to, there is a chance that plastic items (mostly macroplastics) emitted to terrestrial environments are transported to water on the surface, e.g., by wind and rain-based erosion (Jambeck et al. 2015). The amount of plastic transferred from land into water bodies, which in most cases finally ends up in the ocean, highly depends on local conditions such as the local waste management system, climate conditions, and the proximity of the plastic waste emission to water bodies. According to the model of Jambeck et al. (2015), macroplastics initially emitted onto soil as mismanaged waste are partly redistributed to the ocean via inland waterways, wastewater outflows, wind, or tides if the plastic is emitted within 50 km of a coast. Accordingly, macroplastics emitted further away from the coast do not end up in the ocean. Although this

**Table 1** Redistribution of plastic emissions to final compartments after initial release into the ecosphere

Emission	Initial compartment	Final compartment share after redistribution (cf. $T_{ij}$ in Eq. (12))			
		Soil	Marine water	River sediment	Marine sediment
NR/SBR	Soil	100%	0%	0%	0%
	Fresh water	0%	0%	89%	11%
	Marine water	0%	0%	0%	100%
	Air	94.7%	0%	4.7%	0.6%
Polymers with a density $\geq 1 \text{ g/cm}^3$ (PA, PBAT, PBS, PBSA, PBSe, PBSeT, PC, PCL, PET, PHB, PHBV, PLA, PS, PU, PVC, starch-blend)	Soil	97%	0%	2.7%	0.3%
	Fresh water	0%	0%	89%	11%
	Marine water	0%	0%	0%	100%
	Air	94.7%	0%	4.7%	0.6%
Polymers with a density $< 1 \text{ g/cm}^3$ (HDPE, LDPE, PE, PEA, PES, PP)	Soil	97%	3%	0%	0%
	Fresh water	0%	100%	0%	0%
	Marine water	0%	100%	0%	0%
	Air	94.7%	5.3%	0%	0%

rough estimation was only made for macroplastics, we assume the same for microplastics. No indication for other redistribution mechanisms (e.g., subsurface redistribution) from soil to other compartments could be found. Hurley and Nizzetto (2018) concluded that soil systems could store microplastics. Likewise, Fauser et al. (1999) found little downward movement for tire wear particles initially emitted onto soil, as most particles were found in the upper 1 cm of the soil, and 30 times less, only 2 cm further into the ground. This suggests that there is a very small probability of microplastics reaching groundwater through vertical movement. Bioturbation, the physical displacement of solutes and solids in soils caused by the activities of organisms, particularly by burrowing activities of earthworms, can technically lead to a downward transport in soil (Huerta Lwanga et al. 2016). However, since no quantifiable and reliable data are available for transfer rates from soil to groundwater, we neglect these mechanisms and only assume an average redistribution rate from soil to marine water of 27.5% (Jambeck et al. 2015 suggested a range of 15–40%), applied to the respective coastal population share of the analyzed region. Furthermore, we assume that once plastic items reach the ocean, the same redistribution patterns apply to plastics directly emitted into the sea.

Although the shape and characteristic length of the emission might play a role in the redistribution from soil to other compartments, there is no universal information about their influence. Therefore, soil redistribution rates to different compartments are assumed to be independent of the shape, characteristic length, emission material type, and density.

On the other hand, in water bodies, the plastic emission's density plays a more prominent role, especially concerning the vertical movement of the emission and its proneness to sediment (Chubarenko et al. 2016; Kowalski et al. 2016).

Depending on the density of the plastic  $\rho_p$  compared to the density of the water  $\rho_w$ , the plastic item might float ( $\rho_p < \rho_w$ ), stay in the water column ( $\rho_p \approx \rho_w$ ), or sink to the sediment ( $\rho_p > \rho_w$ ). When Sanchez-Vidal et al. (2018) analyzed 29 sediment samples collected from southern European seas, most fibers found were polymers with higher densities, such as polyester, acrylic, and polyamide. We assume that plastic emissions with a density greater than or equal to that of water ultimately sink and become part of the respective water body's sediment (river or marine sediment). Emissions with a density smaller than that of water float and therefore remain in the water. We assume that initial plastic emissions to freshwater bodies, such as rivers, with a density below that of freshwater, are transported on the water surface and ultimately reach the sea. Therefore, we do not consider freshwater as a final compartment. Although it might be possible for plastic items with a density below that of freshwater, which are emitted into a water body without connection to a flowing water body, to remain in that freshwater body, we assume this neglectable when determining generic distribution rates.

Although, polymers with lower densities than water can also sink and, ultimately, sediment, for example, through turbulence, (bio-)fouling, or heteroaggregation with suspended solids (Besseling et al. 2017), we neglect these mechanisms, because defouling might occur, which might resuspend the emission, creating a loop of sinking and suspending over time (Ye and Andradý 1991).

According to Fazey and Ryan (2016) and Chubarenko et al. (2016), smaller plastic emissions and those with a high surface area tend to sink faster as they are more susceptible to biofouling due to their surface area-to-mass ratio. On the other hand, water turbulence, e.g., by wind or currents, increases the vertical movement, especially of microplastics

in the water (Kooi et al. 2016; Lebreton et al. 2018). Since clear assignments are impossible, the redistribution rates from fresh and marine water to the river or marine sediment do not consider the emissions’ size and shape.

Like Kawecki and Nowack (2019) and Peano et al. (2020), we assume that all plastics emitted to air are deposited onto soil or water, with compartment shares dependent on the water to land surface ratio. They are further redistributed in the same way as plastics directly emitted into these compartments.

Since we assume that transport velocities between the compartments are relatively high compared to the degradation times (especially transport by air and in running waters), any degradation occurring during the redistribution is not considered. For example, water within the river Rhine only takes a few weeks to travel from its source at Tomasee (Switzerland) to its delta at Hoek van Holland in

the Netherlands. Due to a lack of available data, a possible recollection from environmental compartments into the technosphere, e.g., by beach cleanups, is not considered. Most assumptions presented in “Sect. 2.3” are generic and can be applied in the same way globally and to specific countries. The redistribution of items emitted to soil and air, however, is country-specific because the water to land surface ratio differs per country. The FF presented in SM3 are given for Germany as an example. They may be adapted to suit other regions.

### 2.3 Calculation of degradation rates, total lifetimes, and residence times

In order to determine the degradation rates of different polymers, a comprehensive literature review was conducted. As presented in Fig. 2, 146 research papers were identified from

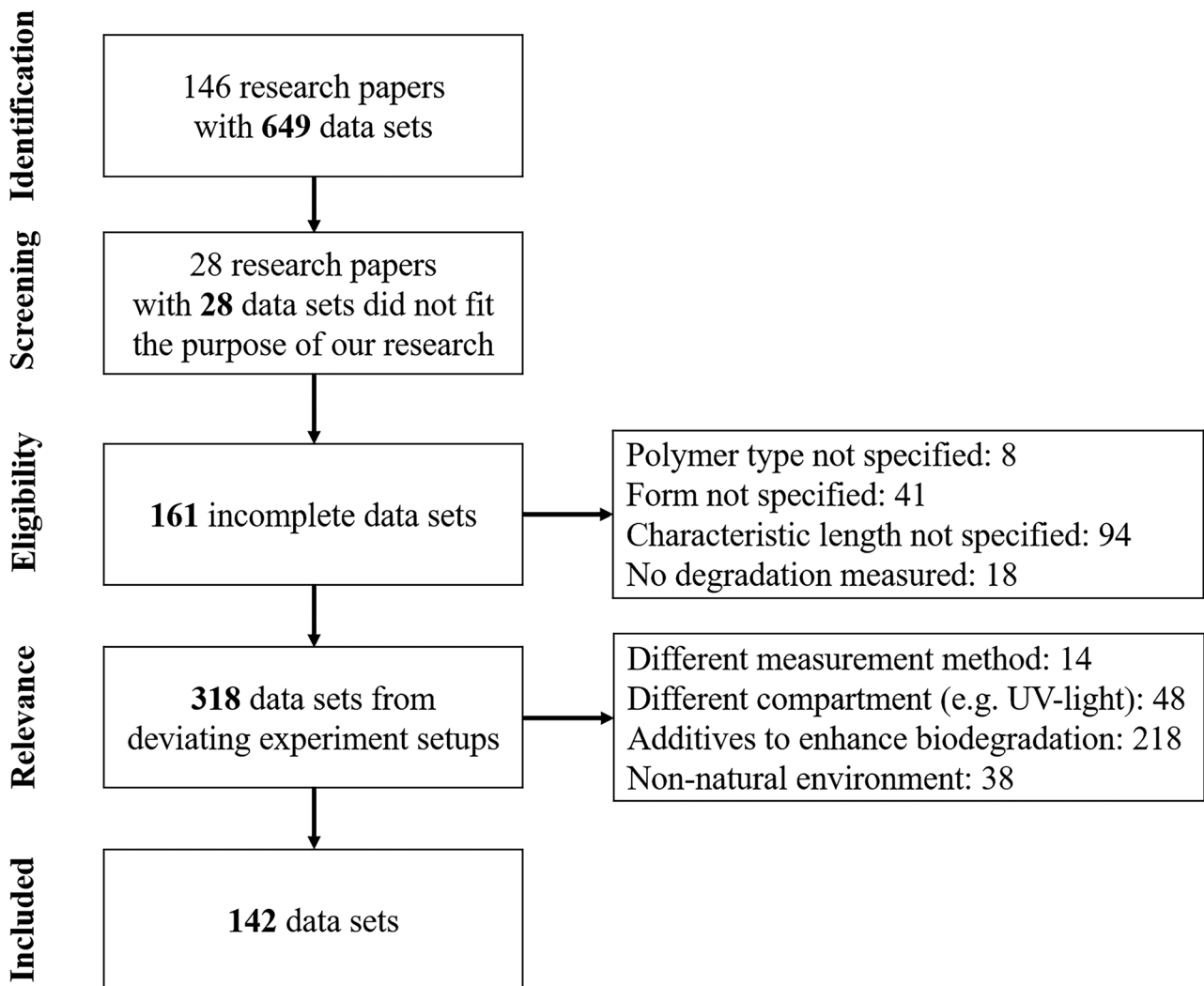


Fig. 2 Methodological approach of the literature review

peer-reviewed journals accessible via the search engines Web of Science, ScienceDirect, and Google Scholar based on keywords such as “plastic,” “fate,” “degradation,” “depolymerization,” “mineralization,” “mass loss,” and “impact.” Following a snowball sampling approach, research papers quoted in the identified publications were also considered. For polymers for which no sufficient data could be obtained via the described method, research papers were searched applying the same approach, adding into the search term that specific polymer.

Research papers that did not disclose all necessary information, e.g., regarding the shape and characteristic length of the investigated plastic item, were excluded. Besides, only studies were taken into account where degradation measurements were based on either weight loss, biochemical oxygen demand, or the amount of CO<sub>2</sub> formed during the degradation. Studies examining material property changes, such as tensile strength or crystallinity, were left out as there is no available correlation to material loss. Only for polymers for which no data are available concerning the described measurement methods, studies were taken into account that measured viscosity at higher temperatures and relied on Arrhenius projection to deduct degradation speed at temperatures found in nature. The few degradation studies focusing on other environmental compartments and pure laboratory studies under artificial conditions were ruled out.

The data found pertain to many different polymer types, including both fossil-based and biobased polymers. Some fossil-based polymer types are commonly assumed not to be biodegradable (ASTM D7611 standard codes 01–06) and are referred to in this study as conventional fossil-based polymers. Nevertheless, as explained further below, we do assume a slow degradation of these polymers. We refer to polymer types of ASTM D7611 code 07 (other) as either biodegradable fossil-based polymers or biobased polymers, depending on their source of material.

For some polymers and compartments, several data sets from one or more publications were available. If any of these data sets did not indicate any degradation, but others for the same polymer and compartment did, those without degradation measurements were excluded, which was considered a measurement error or an insufficient accuracy of the measurement device. For the same reason, data sets that did not indicate any degradation, but were the only data sets available for the respective polymer and compartment, were set to an SSDR of 0.001  $\mu\text{m}$  per year, as further calculations would be impossible with an SSDR of 0  $\mu\text{m}$  per year. The chosen value is slightly lower than the lowest SSDR measured that was unequal to zero to not overestimate degradation for those polymers and compartments.

Since data availability for various environmental compartments is limited, we only differentiate between degradation in soil, river and marine sediment, as well as marine

water at this stage of work. As investigated by Lott et al. (2020, 2021) for polyhydroxyalkanoate copolymer (PHA) in eulitoral, pelagic, and benthic habitats of the Mediterranean Sea and Southeast Asia (Pulau, Bangkam Sulawesi), there are relevant differences in degradation rates. Degradation was observed to be faster in the benthic zone compared to the pelagic zone. The region, however, had the greatest influence on the degradation time. Degradation rates of PHA films in SE Asia were observed to be higher than in the Mediterranean Sea. While degradation rates for different regions are generally not available yet, we distinguish between the marine water body and marine and river sediment.

The extracted data sets contain information regarding the investigated plastics (polymers, additives), the shape and characteristic length of the research subject, the degradation test compartment, whether the degradation conditions could be considered natural, measured degradation, and the time span of the experiment. Data extracted from research papers that did not indicate having examined plastics with additional additives to enhance biodegradation were marked as “no enhancing additives.” Nevertheless, it can be assumed that these plastics include typical additives such as plasticizers, antistatic agents, flame-retardants, or UV stabilizers that are indifferent to or even reduce the degradability. However, the knowledge about types and quantities of additives might be relevant for future integration of more realistic degradation rates and additional ecotoxicity impact assessments.

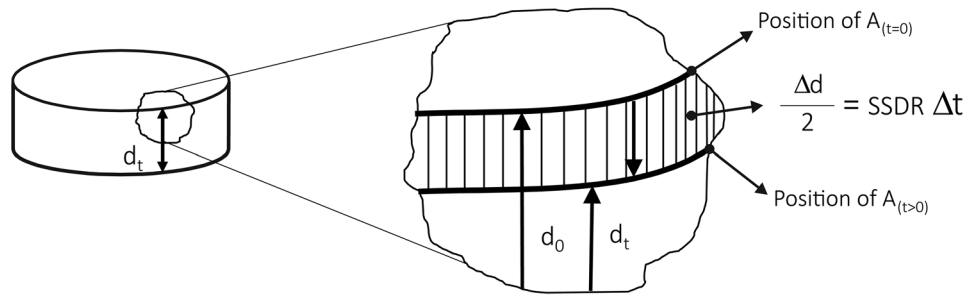
As a result, 38 studies concerning 172 data sets for various polymer types and the environmental compartments marine water, marine sediment, river sediment, and soil under natural or near-natural conditions and without additional additives to influence degradation were used for calculating degradation rates. Data for degradation in soil include values measured during experiments with compost under near-natural conditions. Many studies, especially concerning conventional fossil-based polymers, did not last long enough to reach a significant degradation. In these cases, the value at the last measuring point is taken. In cases where experiments lasted long enough to reach a degradation of more than 50%, the value at the measuring point closest to 50% degradation is taken.

As mentioned before, the residence times depend on (1) the polymer type, (2) the shape of the plastic item, (3) the initial size of the investigated item, and (4) the environmental compartment where the degradation takes place. Following Chamas et al. (2020), three assumptions are made:

- (a) The degradation mainly happens in the top layer at the surface of the emitted item (Fig. 3) (cf. Ohtake et al. 1998).
- (b) A specific surface degradation rate (SSDR)  $v_d$  can be defined that depends on the type of plastic and the environmental compartment the degradation takes place.



**Fig. 3** Surface degradation



(c)  $v_d$  is assumed to be constant during the entire time of degradation.

Naturally, these assumptions are simplifications: some polymers will show degradation in the bulk material or might be eroded in part by mechanical influences. However, also following Chamas et al. (2020), we assume that surface degradation is the factor that ultimately determines the amount of time needed for a plastic emission to vanish.

The initial plastic item is eroded from the overall outer surface. Therefore, the characteristic length  $d_t$  reduces over time  $t$  by twice the degradation rate  $v_d$ , irrespective of the item's shape (Fig. 3). Only for hollow items (e.g., closed bottles), this might not be true initially. However, it can be assumed that these items fracture quickly.

Films are considered to degrade at the main surfaces  $A$  (from both sides) only, fibers are considered to be cylinders whose length  $L$  does not reduce significantly over time, and particles are regarded as spheres. As illustrated in Fig. 4 and Eq. (2), the characteristic length  $d_t$  at a point in time  $t$  refers to the film's thickness or the fiber or particle's diameter, respectively.

$$d_t = d_0 - 2v_d t \tag{2}$$

From this equation, the total lifetime  $\tau_L$  of the emission until the polymer is completely degraded can directly be calculated by setting  $d_t=0$ :

$$\tau_L = \frac{d_0}{2v_d} \tag{3}$$

The total lifetime only depends on the initial size and the specific surface degradation rate and is independent of the shape of the emission due to our degradation model chosen. Nevertheless, the persistence of a specific plastic emission depends not only on this total lifetime but also on the temporal degradation behavior. This temporal degradation behavior depends on the shape because even with a constant SDR, the velocity of mass degradation varies over time due to a change of the surface-to-volume ratio. Hence, we introduce the residence time  $\tau_R$  as measure for the persistence of plastic emissions in the environment (see below). The degrading volume  $V_t$  of a particular plastic emission at time  $t$  is calculated depending on its shape:

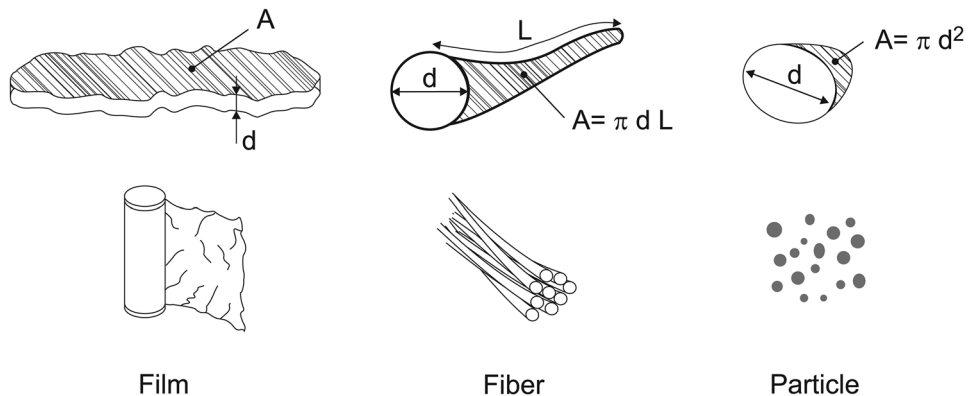
$$\begin{aligned} V_{t\text{film}} &= A d_t^1 \\ V_{t\text{fiber}} &= \frac{\pi L}{4} d_t^2 \\ V_{t\text{particle}} &= \frac{\pi}{6} d_t^3 \end{aligned} \tag{4}$$

Here  $A$  is the surface area of the foil and  $L$  the length of the fiber. In general, the volume can be calculated by a constant and the characteristic length to the power of  $a$ :

$$V_t = \text{const.} d_t^a \tag{5}$$

where  $a$  equals 1 for infinite films, 2 for fibers, and 3 for particles, respectively. Particles are idealized as spheres. However, assuming particles as cubes would lead to the same

**Fig. 4** Schematic illustration of surfaces and characteristic length  $d$  of different shapes (film, fiber, and particle)



result. Nevertheless, irregular shapes of three-dimensional items may be approximately reflected by using a different constant or power in Eq. (5). As long as the constant and power  $a$  in Eq. (5) stay approximately constant during the item’s degradation, they cancel out in the following calculation. Combining Eqs. (2) and (5) leads to the emission’s remaining volume  $V_t$  at a particular time concerning the initial volume  $V_0$ .

$$\frac{V_t}{V_0} = \left(\frac{d_t}{d_0}\right)^a = \left(1 - \frac{2v_d t}{d_0}\right)^a = \left(1 - \frac{t}{\tau_L}\right)^a \quad (6)$$

Assuming a constant density, the same relation holds for the masses of the emission (cf. Fig. 5):

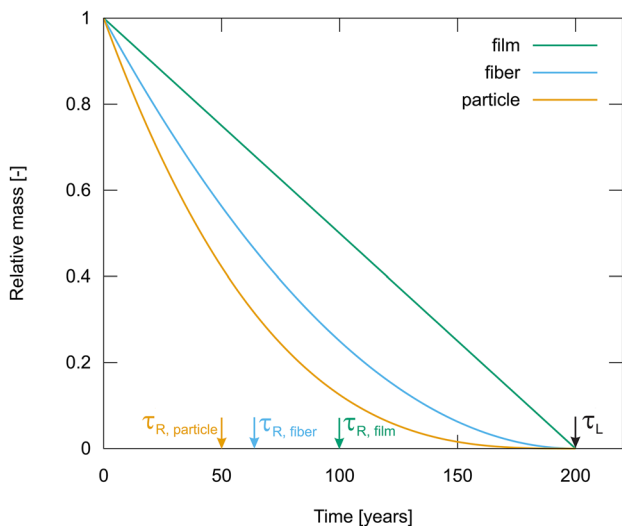
$$\frac{m_t}{m_0} = \left(\frac{d_t}{d_0}\right)^a = \left(1 - \frac{2v_d t}{d_0}\right)^a = \left(1 - \frac{t}{\tau_L}\right)^a \quad (7)$$

The environmental impact according to our approach can be calculated by integrating the curve function of the actual dimensionless fraction of the remaining mass: the dimension of this quantity is time and it will be called residence time  $\tau_R$  (explanation see below):

$$\tau_R = \int_0^{\tau_L} \left(1 - \frac{t}{\tau_L}\right)^a dt = \frac{1}{a+1} \tau_L \quad (8)$$

Hence, the residence time  $\tau_R$  of a film, fiber, or particle is:

$$\begin{aligned} \tau_R &= \frac{1}{2} \tau_L, & \text{for film} \\ \tau_R &= \frac{1}{3} \tau_L, & \text{for fiber} \\ \tau_R &= \frac{1}{4} \tau_L, & \text{for particle} \end{aligned} \quad (9)$$

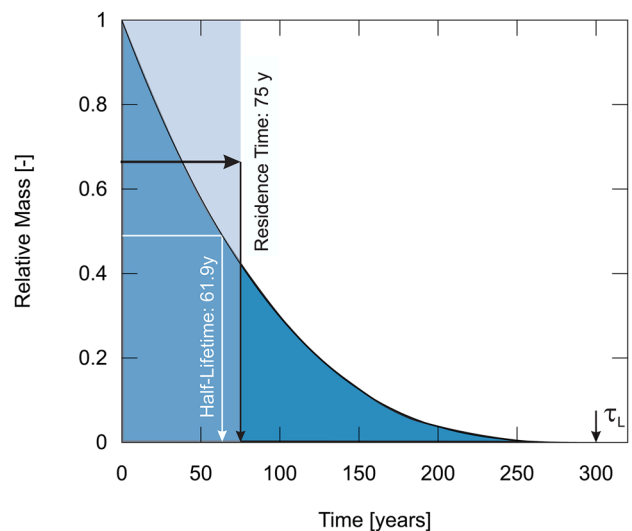


**Fig. 5** Degradation of emission with same total lifetime ( $\tau_L=200$  years) but different shape and different degradation curves (functions see Eq. (10))

As shown in Fig. 5, the velocity of mass degradation of films is constant, resulting in a linear reduction of mass, while for fibers and particles, the degradation is faster in the beginning and slower at the end. This results in a higher residence time  $\tau_R$  for films than for particles at similar life time Eq. (9).

Figure 6 shows the residence time  $\tau_R$  of a particle. In this case, the residence time is 75 years. In contrast, the point in time at which 50% of the mass is degraded (half-lifetime) of the particle would be slightly lower with 61.9 years. The name residence time was chosen, because during the total degeneration process mass will continuously vanish and the average age of this leaving mass is equal to the residence time introduced. Furthermore, the residence time can be interpreted as that time a non-degradable emission would have to stay in the environment to give the same impact: in Fig. 6, the gray box has the same area as the integral below the degradation curve.

The consideration of time horizons  $\tau_H$  is a common practice in LCIA (Hauschild and Huijbregts 2015). When calculating FFs (“Sect. 3.3” and supplementary material), time horizons of 100, 500, and 1000 years are applied. Both a 100- and 500-year time horizon are commonly used for other environmental impact assessments, e.g., global warming potential. The longer time horizons allow for greater differentiation between hardly degradable polymers. By selecting a short time horizon, e.g., 100 years, FFs will only differ when emissions degrade faster than 100 years while polymers with life times of, e.g., 500 years, 10,000 years, or 50,000 years will all obtain an residence time close to 100 years. This might encourage politicians and the industry to focus on fast degradable polymers if



**Fig. 6** Residence time of a particle: the area of the box and the area below the degradation curve are equal

resulting emissions into the environment are hardly avoidable. When applying a time horizon, the integration is performed until the time horizon is reached and Eq. (8) becomes:

$$\tau_{R,\tau_H} = \int_0^{\tau_H} \left(1 - \frac{t}{\tau_L}\right)^a dt = \frac{\tau_L}{a+1} \left(1 - \left(1 - \frac{\tau_H}{\tau_L}\right)^{a+1}\right) \text{ for } \tau_L > \tau_H \quad (10)$$

This means, as illustrated in Fig. 7, by applying a time horizon, only the area inside the time horizon is considered (blue area), while everything after the time horizon is omitted (gray area).

The ratio of  $\tau_R$  and  $\tau_H$  ranges between 0 and 1 and measures the “occupation” of the time horizon. However, the results need to be interpreted cautiously: the residence time in a time horizon is always smaller than the residence time without a time horizon. Especially durable polymers like PVC with residence times greater than 1000 years without considering a time horizon might appear more favorable than they are when interpreting the residence time with a short time horizon (e.g., 100 years). Consequently, the residence time must always be interpreted relative to the time horizon considered. If the value of the residence time is close to the value of the time horizon, very little degradation occurs during this time horizon. The equations obtained (8 and 10) hold for the degradation models chosen. However, the approach can be easily adapted to other degradation mechanisms (e.g., Junker et al. 2016) as long as the degradation curves are known.

Another derivation of the residence time and the FF as measure of the environmental impact is shown in Fig. 8. Assuming a constant yearly flow  $\dot{m}$  there is only a partly

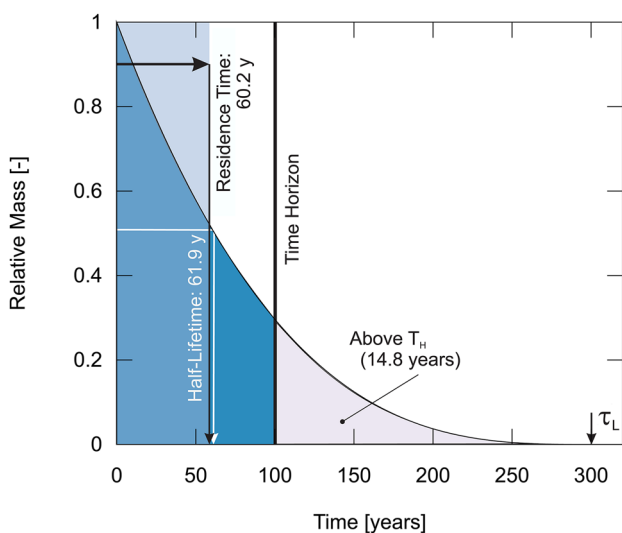


Fig. 7 Calculation of the residence time of a particle for a time horizon given. The area of the light blue box and the blue area are equal

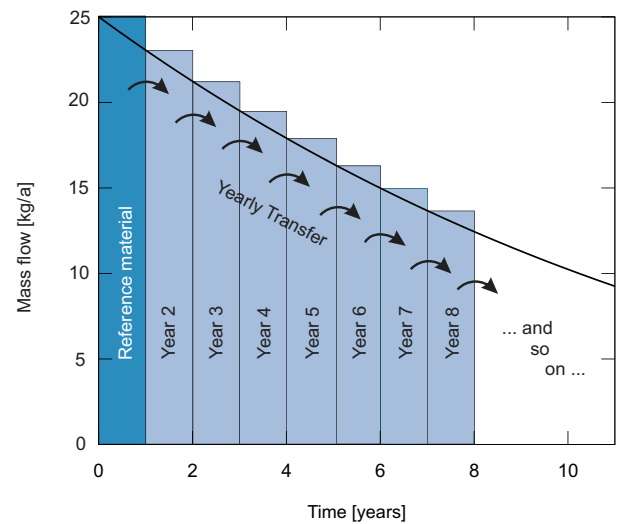


Fig. 8 Visualization of the residence time approach and the reference material

degeneration in the first year. After 1 year, the remaining amount is transferred to the second year and a fresh inflow will appear (and so on). The curve will have the same shape as in the calculations before. When summing up all yearly amounts by integrating the curve Eq. (9), the hold-up  $M$ , i.e., the total mass accumulated in the environment due to this emission, is calculated. The degeneration of this hold-up balances the emission flow in the steady state case. Dividing this hold-up by the emission flow a residence time is calculated, which is equal to the residence time before.

$$\tau_R = \frac{M}{\dot{m}} \quad (11)$$

This shows vividly the equivalence of mass-flow and residence time as stated before. One unit of an emission A with a specific residence time results in the same hold-up as an emission of two units of an equally sized emission B with half the residence time of emission A. This holds for diluted, widely spread emissions. It is not suitable to calculate effects of local or temporal concentration hot spots.

With the residence time approach, it is straightforward to calculate the fate of an emission that is divided and transferred to different final environmental compartments. The mass of the initial release is distributed according to the transfer factors  $T_{ij}$  and for each environmental compartment, residence times are calculated separately. The transfer factors are the fractions of an emission  $i$  transferred to compartments  $j$ . The overall residence time is calculated as a weighted sum of the individual ones.

$$FF_{i,tot} = \sum_{j=1}^n \left[ T_{ij} \frac{\tau_{R,i,j}}{\tau_{R,ref}} \right] = \sum_{j=1}^n [T_{ij} FF_{i,j}] \text{ with } \sum_{j=1}^n (T_{i,j}) = 1 \quad (12)$$

In Fig. 9, an emission is divided into two compartments (30/70%) with total lifetimes of 200 and 100 years, respectively. The residence time of the entire emission is the weighted sum of the individual ones (50 and 25 years, respectively). From the residence time (32.5 years), a total lifetime of 130 years of the emission in a hypothetical compartment can be calculated by Eq. (9).

## 2.4 Calculation of specific surface degradation rates

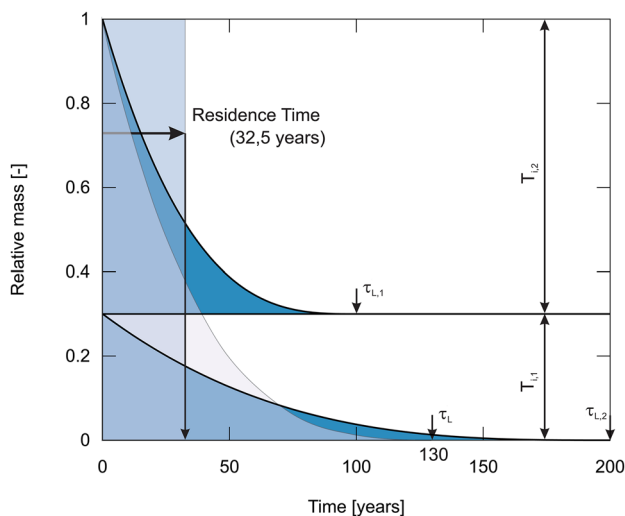
The degradation model described is used to evaluate experimental results in the literature.

Experimental studies on degradation usually report the loss of mass  $\Delta m$  relative to the initial mass  $m_0$  during a period. In order to calculate the specific surface degradation rate  $v_d$  (SSDR) of a plastic emission, Eq. (7) can be rearranged:

$$v_d = \frac{1}{2} \frac{d_0}{t} \left( 1 - \sqrt[a]{1 - \frac{\Delta m}{m_0}} \right) \quad (13)$$

This yields the SSDR of the experimental mass loss  $\Delta m/m_0$  during a period  $t$  and the given initial characteristic length  $d_0$  and shape (to determine the power  $a$ ). SSDRs are calculated for each polymer type and each compartment. The SSDR values displayed in the supplementing material are based on experimental data extracted from the research papers analyzed, as explained at the beginning of this section. For polymers for which data are insufficient to calculate SSDRs, values are estimated according to the following assumptions, which have to be confirmed or improved in the future:

- Where data for only one of the environmental compartments are available, degradation is assumed to be com-



**Fig. 9** Residence time for a plastic emission (particle) that is redistributed to two environmental compartments

parable in the other three, and the same value is utilized for all four compartments.

- Where data are available for river or marine sediment but not the other type of sediment, the same value is utilized for both types.
- Where data are available for marine water and soil, but not river and marine sediment, the lower degradation rate of the two compartments, marine water and soil, is used for both sediment types as a conservative estimate not to overestimate the sediment's degradation.
- For polystyrene (PS) and polyvinyl chloride (PVC), no data are available to calculate SSDR in any compartment. SSDR of PS and PVC are estimated to aim towards 0 (cf. Chamas et al. 2020 based on information published by Otake et al. 1995, obtained by a measurement method otherwise irrelevant to our research: through observation by phase contrast microscope and scanning electron microscopy or mere estimation). For further calculations, the SSDR for these polymers is set to 0.001  $\mu\text{m}$  per year.

## 2.5 Data quality and uncertainty analysis

The procedure to select data for calculating FFs is based on a data quality assessment. For example, for some polymer types and environmental compartments, several studies with different degradation rates were available. The assessment of the data quality enabled the decision for the most accurate degradation rate to be used. In order to indicate the quality of the data provided in this paper, we adapted the pedigree matrix approach, which was first introduced by Funtowicz and Ravetz (1990) and was adapted by Weidema and Wesnæs (1996) for life cycle inventory data, and applied it to our input data. The pedigree matrix approach allows to assess data quality and translation to uncertainty values although a small sample size for most of the SSDRs is given. If the number of SSDRs increases or uncertainty values for SSDR measurements are provided, these values should be used instead. Since no pedigree matrix exists for degradation rates and transfer coefficients between environmental compartments, we altered the initial categories to suit the research purpose. Like Laner et al. (2016), we distinguish between experimental data and expert judgments. Scores indicate good data quality (data quality indicator score (DQIS) = 1) up to low data quality (DQIS = 4).

The quality of experimental data is affected by its reliability, completeness, temporal and geographical correlation as well as the measurement method used. For the geographical correlation, Germany is set as a reference, to match the country of the redistribution patterns, as described in "Sect. 2.3." The quality of expert judgments depends on the foundation of the judgment, for example, on an (empirical) database, the experts' qualification, and the transparency of the procedure by which the judgment was obtained (Laner et al. 2016).

Based on these DQIS, uncertainty scores are calculated for each input data set: transfer coefficients and degradation rates as explained in SM1. The modeler sets the size and shape of a plastic emission for each elementary flow; therefore, these parameters are assumed not to induce additional uncertainty. However, this could be the case in terms of measurement uncertainties. Nonetheless, the uncertainty induced by size and shape is assumed to be small compared to the uncertainty induced by transfer coefficients and degradation rates. For transfer coefficients, expert judgment is used to apply relevant information from existing publications to our research case and Germany's environmental conditions. For degradation rates, where more than one data set per polymer and compartment were found, the data set with the lower uncertainty score is used for further calculations. The geometric average of SSDRs is used when more than one data set had the same lower uncertainty score. The uncertainty calculation of the FFs is done using Monte Carlo simulation based on the geometric standard deviation (GSD) and median, taking log-normal distributions for the estimates of SSDR and redistribution (Limpert et al. 2001).

In order to calculate the confidence interval of 68%, the FFs need to be multiplied by the GSD for the upper limit and divided by it for the lower limit. To calculate the confidence interval of 98%, multiply or divide the given FFs by  $GSD^2$ . The Python script for the FFs' uncertainty calculation is given in SM4 for reproducibility and traceability reasons.

### 3 Results

While one of the key contributions of this paper is the development of a new methodology to include plastic-related elementary flows in LCA, the following chapter presents the data found in literature regarding redistribution patterns and degradation rates, which were used to calculate FFs for a vast number of elementary flows. The list of FFs provided in SM3 is non-exhaustive. Following our approach, LCA modelers are able to calculate FFs for their specific plastic emission regarding polymer type, size, shape, and initial compartment. Besides, the methodology can be applied to different areas or countries. Because certain elements, e.g., redistribution and the data quality assessment of the degradation rates, require a regional focus, the results are given for Germany as an exemplary region. The modeler might choose a different regional focus and therefore adjust the FFs based on the equations presented in this paper.

#### 3.1 Compartment shares and redistribution

Table 1 provides an overview of the final distribution of plastic emissions between environmental compartments due to their redistribution. For natural and synthetic rubber,

data on initial release and redistribution are based on Unice et al. (2019), who analyzed the fate of tire wear particles in the Seine watershed (France). Redistribution rates are suggested for the other polymers based on the initial environmental compartment and the polymer's density, as described in "Sect. 2.2." Redistribution rates from fresh and marine water to the other compartments are generic. As explained in "Sect. 2.1," redistribution rates from soil and air are given for Germany. The shares of coastal populations of other countries can, e.g., be based on the CIA World Factbook and the geographic information system (GIS) data provided by Jambeck et al. (2015).

Based on a redistribution rate from soil to marine water of 27.5% and a coastal population share of 11% in Germany, 97% of plastics emitted to soil will remain in soil; the rest might sink to the river sediment (2.7%) or be redistributed to the ocean and remain in the marine water (3%) or sink to the marine sediment (0.3%) (Unice et al. 2019), dependent on the polymer's density.

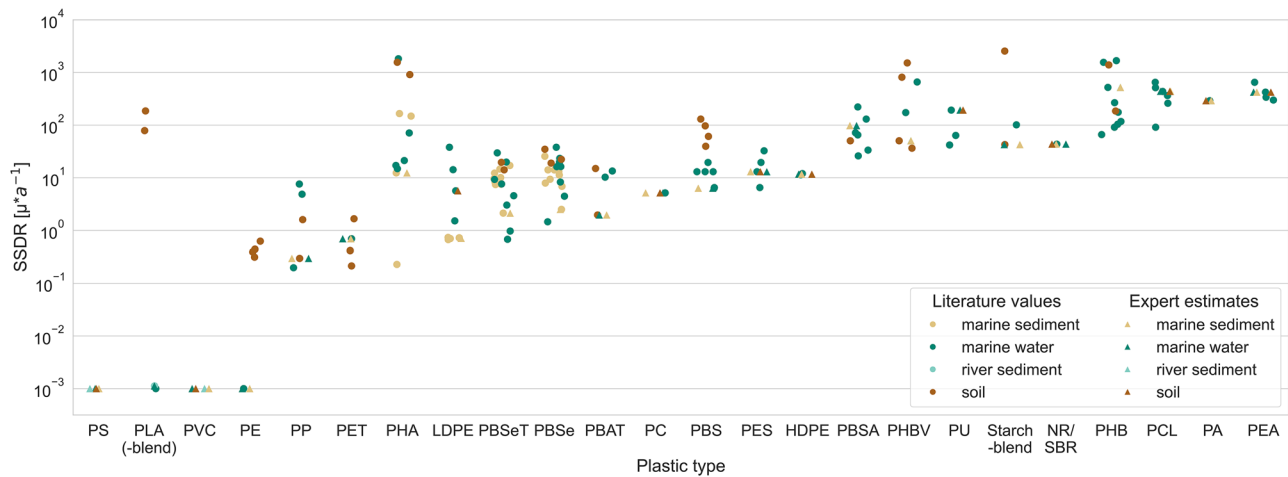
After initially being emitted into the air, 2.4% of the plastics will be deposited onto fresh water and 97.6% onto soil, based on a surface share of 2.4% water and 97.6% infrastructure or vegetation (both considered soil) in Germany (Statistisches Bundesamt 2017).

#### 3.2 Specific surface degradation rates

The SSDR of fossil-based plastics, natural and synthetic rubber, and biobased plastics in the four compartments, river sediment, marine water and sediment, and soil, are displayed in Fig. 10. Beige dots and triangles display degradation rates in marine sediment, dark green dots and triangles degradation rates in marine water, light blue dots and triangles represent degradation rates in marine sediment, and brown dots and triangles degradation rates in soil (including compost). Dots indicate values found in literature and triangles represent expert estimates.

Figure 10 displays all degradation rates found during the literature review, independent of the data quality. As can be seen in Fig. 10, very few data are currently available for conventional fossil-based plastics. Only Yabannavar and Bartha (1994) and Boyandin et al. (2013) conducted experiments in soil with polyolefins (PE and PP), and no data are available to calculate degradation rates of conventional fossil-based plastics in marine and river sediment. Most studies investigating conventional fossil-based plastics were conducted in marine water.

For some kinds of PE, degradation rates vary very little. For example, for HDPE, Sudhakar et al. (2007) and Artham et al. (2009) conducted experiments in marine water at the Bay of Bengal. The corresponding degradation rates are 11.4–12.0  $\mu\text{m}$  per year. Likewise, for PE, the variation among the data is very small (0.0–0.6  $\mu\text{m}$  per year in studies



**Fig. 10** Specific surface degradation rates of different polymers in different environmental compartments in  $\mu\text{m}$  per year on a logarithmic scale (data can be found in SM2)

conducted by three different research groups: Yabannavar and Bartha (1994); Rutkowska et al. (2002a); Boyandin et al. (2013)). On the other hand, Sudhakar et al. (2007) and Artham et al. (2009) conducted the same experiments as for HDPE also for LDPE, but their results on LDPE differ substantially (14.4–38.0  $\mu\text{m}$  per year). The higher SSDR of LDPE compared to HDPE can be explained due to more amorphous zones. Besides, highly crystalline PE is highly stable against degradation. This tendency, however, is not reflected by all data sets. Similarly, Eich et al. (2020), who also investigated the degradation of LDPE, found no degradation in the marine water of the Mediterranean Sea. In summary, it can be stated that there exist huge differences for different kinds of PE and even the degradation speed of the same kind of PE (LDPE) varies clearly between different studies.

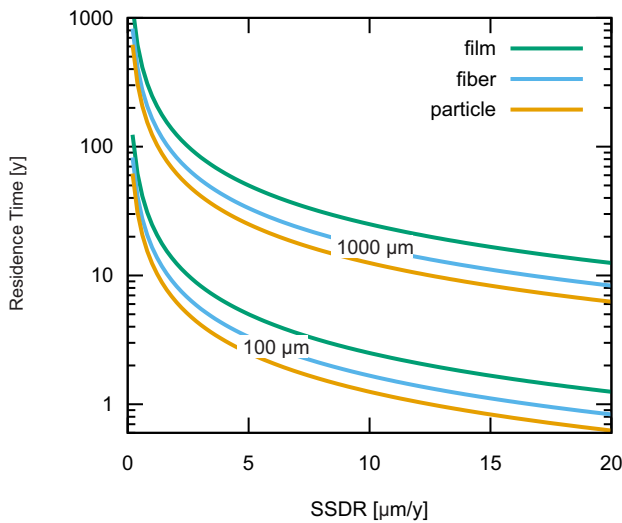
In the case of PP, degradation rates range from 0.2  $\mu\text{m}$  per year (Resmeriță et al. 2018) to 7.6  $\mu\text{m}$  per year (Sudhakar et al. 2007) and for PU from 0 to 193  $\mu\text{m}$  per year (both Rutkowska et al. 2002b). The reported low degradation rates might be related to the limited bioavailability of the plastic's molecules to microorganisms. For example, according to Gilan et al. (2004), the low degradation rates observed for PE might be since most bacterial surfaces are hydrophilic and PE is hydrophobic. In practice, other nutrition/energy sources might be more easily available to microorganisms than those contained in plastics. Besides, the amount of species of microorganisms that can degrade plastics is limited. For example, according to Kumar Sen and Raut (2015), only 19 genera of bacteria and 12 fungal genera are known to degrade LDPE.

Compared to the conventional fossil-based polymers, more data are available for biodegradable fossil-based

and biobased polymers. Similar to conventional fossil-based polymers, more degradation data are available for marine water than for soil. Nevertheless, soil degradation rates are available for PBAT, PBS, PHA, PHB, PHBV, PLA(-blends), and starch-blends. Besides, some data are available concerning degradation in freshwater (for PBS, PBSe, PHBV, PU, Starch-blends, NR/SBR, PHB, PCL, PES, PEA, PHB, and PLA-blends), in river sediment (for PLA-blends), and in marine sediment (for PBSe and PBSeT). Degradation rates are available for most biobased polymers (except PBAT) for at least two different compartments. As might be expected, degradation seems to be faster for polymers that are commonly characterized as biodegradable or compostable. Nevertheless, in total, there is a smooth gradient from high to low degradability.

Concerning conventional fossil-based polymer types, data are insufficient to either support or contradict the conclusion of Andrady (2011) that degradation is faster on land than in aquatic systems, e.g., due to greater exposure to degradative forces such as UV light. For biodegradable fossil-based and biobased polymer types, the extracted data from the literature supports that hypothesis for all cases where data are sufficient to calculate both rates.

Besides differences in degradation rates related to the polymer type, the residence time  $\tau_R$  of plastic degradation depends on the shape (film, fiber, particle) and size (characteristic length  $< 0.1$  mm,  $0.1$ – $1$  mm, and  $> 1$  mm) as expressed in Eq. (10) and visualized in Fig. 11. Although only moderate changes of the input variables degradation rate, characteristic length, and shape are considered, the average lifetimes span several orders of magnitude: particles always degrade faster than fibers and films of the same characteristic length,



**Fig. 11** Residence times  $\tau_R$  depending on surface degradation rates (SSDR), the shape, and characteristic length of plastic emissions

although the initial characteristic length’s influence is more dominant than the shape of the emission. Especially for low degradation rates, the average lifetime exponentially increases,

which leads to less reliable experimental results for such polymers in reasonable experiment times.

### 3.3 Calculation of fate factors

According to the methodology described in “Sects. 2.3 and 2.4,” FFs were calculated for Germany for different time horizons: 100, 500, and 1000 years. Examples of typical plastic emissions are presented in Table 2. The complete list of FFs is given in SM3. SM3 also provides the FFs without applying any time horizon and the corresponding GSD.

In general, the FF represents the extent of the persistency of a plastic emission compared to a reference emission with a residence time of 1 year. Conventional fossil-based plastics with a characteristic length of 1 mm and more result in very long residence times of more than 200 years up to 1000 years. This is the case, for example, for yogurt cups (PS), picnic cutlery (PS), plastic caps (PE), microbeads from cosmetics (PE), PET bottles but also for pellet losses (e.g., PVC). No significant degradation occurs when the FF for the different time horizons is close to the respective time horizon. This applies to yogurt cups (PS), picnic cutlery (PS), microbeads (PE), and pellet losses (PVC).

**Table 2** FFs for selected plastic emissions expressed as kg plastic pollution equivalent (PPE) per kg plastic emitted

Example	Elementary flow	Example of related process	Fate factor (FF) $\left[ \frac{\text{kg}_{\text{PPE}}}{\text{kg}_{\text{emission}}} \right]$		
			Time horizon = 100 years	Time horizon = 500 years	Time horizon = 1000 years
1	DE: LDPE film <0.1 mm (emission to soil)	Littered plastic bag	4	4	4
2	DE: NR/SBR film <0.1 mm (emission to air)	Rubber balloons	<1	<1	<1
3	DE: NR/SBR fiber 0.1–1 mm (emission to soil)	Tire wear from transport processes	3	3	3
4	DE: PA particles <0.1 (emission to soil)	Abrasion of string from lawn trimmer	<1	<1	<1
5	DE: PE film 0.1–1 mm (emission to soil)	Littered plastic caps	93	348	419
6	DE: PE particles 0.1–1 mm (emission to soil)	Microbeads from cosmetics	83	205	224
7	DE: PE particles 0.1–1 mm (emission to freshwater)	Microbeads from cosmetics; abrasion of wear of street markings	100	499	997
8	DE: PET fibers 0.1–1 mm (emission to freshwater)	Abrasion of textile fibers from washing	87	233	239
9	DE: PET film 0.1–1 mm (emission to freshwater)	Littered PET bottle	93	326	358
10	DE: PLA film <0.1 mm (emission to soil)	Littered plastic bag	3	15	30
11	DE: PP particles <0.1 (emission to freshwater)	Abrasion from drinking and sewage pipes	2	2	2
12	DE: PS film 0.1–1 mm (emission to soil)	Littered yoghurt cup	100	500	999
13	DE: PS film > 1 mm (emission to soil)	Picnic cutlery	100	500	999
14	DE: PVC particle > 1 mm (emission to soil)	Pellet loss	100	500	1000

In products with a characteristic length less than 1 mm, such as plastic bags (LDPE, PLA), shorter residence times are estimated. Surprisingly, considering a time horizon of 500 and 1000 years, the residence time of LDPE bags emitted to soil was calculated to be shorter than for PLA bags also emitted to soil. In contrast, with a time horizon of 100 years, the residence time of PLA bags (3 years) is slightly shorter than that of LDPE bags (4 years). This is because 97% of the PLA bag degrades very fast in soil and only 3% is redistributed to river and marine sediment where PLA shows no significant degradation. That means plastic emissions that degrade significantly faster in one compartment receive a comparably higher FF with an increasing time horizon than plastic emissions with a similar degradation speed in all compartments. Therefore, to allow for a deeper interpretation of the results, compartment-specific residence times should be analyzed.

Very short residence times of less than 4 years ( $FF < 4$ ) were calculated for tire wear, balloons (NR/SBR), abrasion from drinking and sewage pipes (PP), and very fine plastic particles such as those released by a lawn trimmer (PA) string abrasion. With a time horizon of 100 years, hardly degradable plastic emissions receive an FF close to 100. Compared to a time horizon of 500 years, conventional fossil-based plastics with a characteristic length of 1 mm or more are characterized similarly, all with the highest FF. One exception is the abrasion of PET fibers from washing with a residence time of 233 years with a time horizon of 500 years. When changing the time horizon to 1000 years, littered plastic caps (PE) are assigned an FF of 419 years and the littered PET bottle of 358 years. Accordingly, it can be concluded that a larger time horizon allows for more differentiation.

## 4 Discussion

Applying the methodology results in elementary flows and corresponding FFs that can be integrated into LCA. Depending on certain modeling choices (e.g., the time horizon) and the share of compartment-specific FFs compared to the total FFs, differences among polymer types become more or less apparent (cf. “Sect. 4.1”). While this paper is the first to present FFs for plastic-related elementary flows, there are still some challenges regarding the application to LCA (cf. “Sect. 4.2”).

### 4.1 Underlying patterns of plastic emissions’ fate in the environment

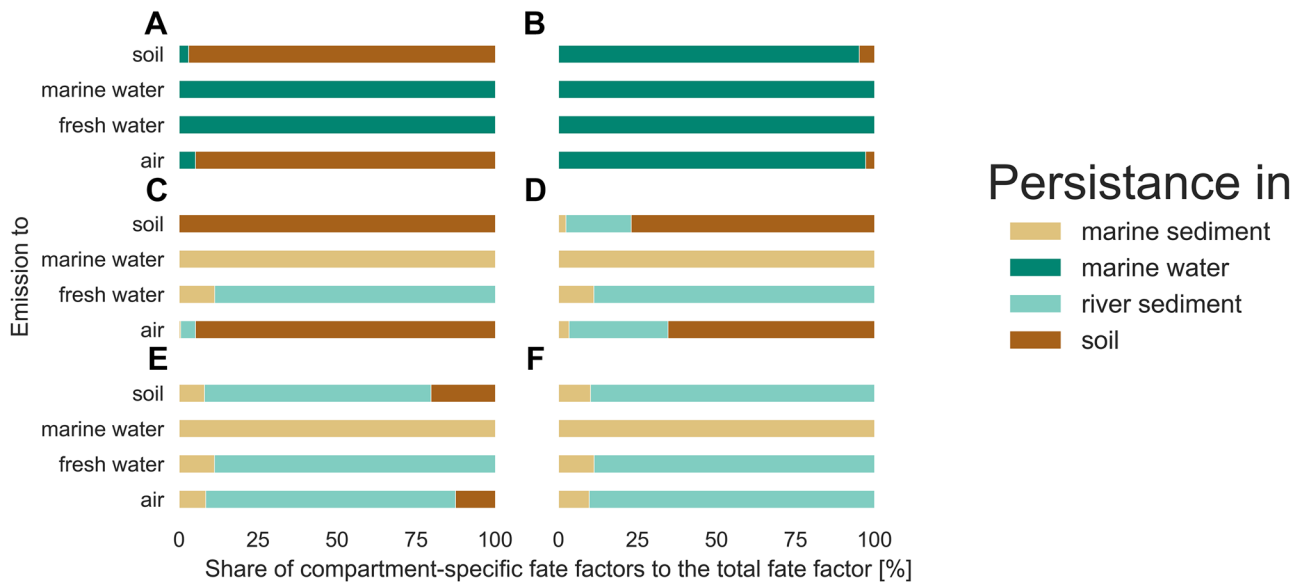
The FFs are more than pure degradation data of a polymer type, because size, shape, initial compartment, and redistribution are considered, too. When not applying a time horizon and combining Eqs. (4) and (9), it can be

concluded that the characteristic length influences the fate linearly while the SSDR enters the equation inversely proportionally (Fig. 11). At all times, the remaining mass of the emission is smaller for particles and fibers than for films due to a faster degradation in the beginning (Fig. 5). Some polymers show very different degradation rates in different compartments (e.g., PLA). Consequently, under some circumstances conventional fossil-based polymers degrade faster than biodegradable fossil-based polymers (cf. Table 2, examples 1 and 10).

When applying a time horizon, the patterns are even more complex Eq. (11): if there is no appreciable degradation of an emission within a time horizon, i.e., the residence time is close to the time horizon, the time horizon is “saturated” and changes of the degradation parameters (SSDR and initial size) do not alter the value of  $\tau_A$  significantly (e.g., Table 2, examples 12 and 13). Looking at the example of the plastic bags again (Table 2, examples 1 and 10), it can be noted that the PLA bag degrades faster than the one made of LDPE when applying a time horizon of 100 years, while it degrades much slower when applying a 500 or 1000 year time horizon, due to a very fast degradation of the major fraction in soil and a very slow degradation of the minor fraction distributed to marine and river sediment.

In this publication, we were able to develop FFs for Germany for 24 polymers. In order to find similarities and differences in polymers’ behavior in the environment, we clustered them. For the clustering, we compare the compartment-specific FFs with the total FF (see SM3). The polymer types are grouped into six clusters (types A to F in Fig. 12), independent of the shape and size of the emission. In Fig. 12, it is shown in which compartment degradation happens when a polymer is emitted to a specific compartment. For instance, when emitting a type A polymer to soil, degradation mainly occurs in soil and partly in marine water, but no degradation is observed in the sediment compartments. Types A and B are polymers with a density lower than water. When emitted to soil or air, most of their mass is found in soil after the redistribution. When emitted to fresh or marine water, types A and B emissions float on the water surface and are ultimately transported to marine water. Type A polymers (HDPE, LDPE, PEA, PES, PP) degrade equally fast in all final compartments. Therefore, the contribution of the FFs for the different environmental compartments is equal to the final compartment share of the emitted plastic mass. Type B polymers (PE) differ from type A polymers in higher SSDRs in soil than in all other compartments, resulting in a lower degradation time in soil. For emissions to fresh and marine water, the fate is similar to type A polymers. For emissions to soil and air, however, the overall fate of type B polymers is dominated by the high persistency in





**Fig. 12** Clustering of the different degradation behavior of plastic emissions in different compartments

marine water, despite the bigger compartment share of the mass in soil. The polymers of types C to F have a higher density than water and thus sink through the water column and ultimately reach either the river or the marine sediment when emitted to water. This results in a degradation in marine and river sediment, as well as soil, but no degradation in marine water for polymers of types C to F. SDRs of type C polymers (NR/SBR, PA, PBAT, PBSA, PC, PCL, PET, PHB, PHBV, PS, PU, PVC, starch-blend) are in the same order of magnitude in all final compartments and the redistribution drives the degradation pattern. The share of compartment-specific FFs to the total FF for types D to F polymers is explained by the ratio of the SDR in soil to the SDRs in the sediments. The greater the ratio, the less soil degradation plays a role. PBS, PBSe, and PBSeT are type D polymers (SSDR in soil is approximately ten times higher than the SDR in the other compartments), PHA is the type E polymer (SSDR in soil is approximately 100 times higher than the SDR in the other compartments), and PLA(-blend) for a type F polymer (SSDR in soil is 70,000 times higher than the SDR in the other compartments). In the future, it might be possible that a different clustering is necessary. For instance, new classes could be introduced when considering other polymers. In addition, other polymers could be assigned to the already defined classes for which no FF has yet been determined due to lack of data.

## 4.2 Application of the fate factors to LCA

The proposed methodology links FFs to the residence time of plastics in the environment and addresses how many years

it takes for the plastic emission to degrade in the final compartments. We assume that the risk potential of plastic in the environment is correlated to its persistence in nature.

The prerequisite for applying the proposed methodology to life cycle analysis is properly modeling technical flows, combined with accurate initial release rates. As mentioned before, the most precise option is to measure or calculate specific initial release rates for the investigated products in their entire life cycle. If the precise modeling of the product-specific initial release is not possible, less precise data for estimating initial release rates for other countries can be taken, e.g., from Peano et al. (2020). When determining the initial release of plastics emitted by a product, the corresponding elementary flows should be named according to the convention described in “Sect. 2.2,” including the precise characteristic length instead of the size classes, if available. If the characteristic length is not available, the proposed elementary flows can be used as proxies. However, it needs to be taken into account that we used the upper end of the range of the characteristic length for the calculations, leading to relatively long residence times.

In order to apply the FFs to a specific region, the redistribution rates (cf. Table 1) can be adapted to region-specific conditions. Following these steps, regionalized and emission-specific FFs can be determined.

The proposed methodology is intended to support (plastic) product designers, for example, to support materials’ choice. In particular, products with higher littering rates or those where abrasion leads to the release of microplastics should be assessed by conventional LCIA categories and analyzed regarding plastic emissions. However, the proposed FFs can also be used to evaluate products ex-ante, such as

giving additional advice to producers on which packaging solution performs better from a plastic emission point of view.

### 4.3 Limitations of the methodology

The concept presented in this article is limited on several accounts due to the varying certainty of the applied parameters. Data on polymers' degradability are scarce for some polymers, particularly for conventional fossil-based polymers such as PET or PP, as presented in Fig. 10. One reason for the limited data availability is the need for long experiment durations for slowly degrading polymers and the associated high costs for degradability tests that are close to reality. Besides, there is a lack of reliable field test methods and standards for assessing and certifying biodegradation (Lott et al. 2020). Test methods differ, for example, in trial duration, the method to measure degradation (e.g., weight loss vs. CO<sub>2</sub> emission), climatic and environmental conditions. Norms addressing degradation measurements were mainly developed exclusively for biobased plastics focusing on compostability in industrial compost (e.g., EN 13,432, ISO 17088, EN14995, ISO 18606, ASTM D6400, AS 4736) or garden compost (AS 5810, NF T 51–800). Only the DIN EN norm 17,033 was developed to measure the biodegradability of mulch films in agricultural soils and horticulture (EN 17033:2018 2018).

Following the proposed methodology to calculate residence times of plastic emissions, we probably estimate relatively low residence times since it is scientifically challenging to differentiate between (bio)degradation and fragmentation processes as primary drivers for the observed mass losses of plastics in the environment. Additionally, slowly degrading polymers tend to have too short calculated

residence times compared to polymers with faster degradation speed since initial losses of better degradable monomers, small molecules, or additives can lead to underestimated degradation rates. Most studies on the degradation of conventional fossil-based plastics only lasted long enough to reach a minimal degradation. Long-term experiments are needed to measure the degradation to substantial values (> 10%, ideally to  $\geq 50\%$ ) in different environmental compartments. Besides, because microplastics found in the environment and those used in laboratory experiments differ (Phuong et al. 2016), laboratory results are not transferable to the field and reduce the amount of usable data. If no data were available for the degradation of a polymer in a particular environmental compartment, degradation data from another environmental compartment are used as approximate values, leading to higher data uncertainty.

In order to analyze the uncertainty introduced by SSDR and redistribution, GSDs of the FFs calculated for Germany are presented as boxplots in Fig. 13. In the boxplots, the median represents the middle GSD of polymers' FFs. Uncertainty represented by the median GSD varies between 1.2 and 3.2. Low uncertainty (with a median GSD lower than 2) is given for most plastic types (22 out of 24). Low uncertainty is due to representative values for SSDR. The uncertainty introduced due to redistribution is the same for all plastic types (GSD = 1.3). FFs of PVC and PS emissions have higher GSDs (higher than 3) as SSDRs for these plastic types rely on expert estimates. Expert estimates go along with the greatest possible uncertainty as defined here. Variability in FFs is represented by the GSD distribution spread and can be quantified by the interquartile range (IQR). The IQR represents the difference between the 25th and 75th percentiles of a distribution. For PET, the IQR is 1.3, which represents the highest variability among all FFs GSDs. The GSDs of PET's SSDRs are the reason for the variability of

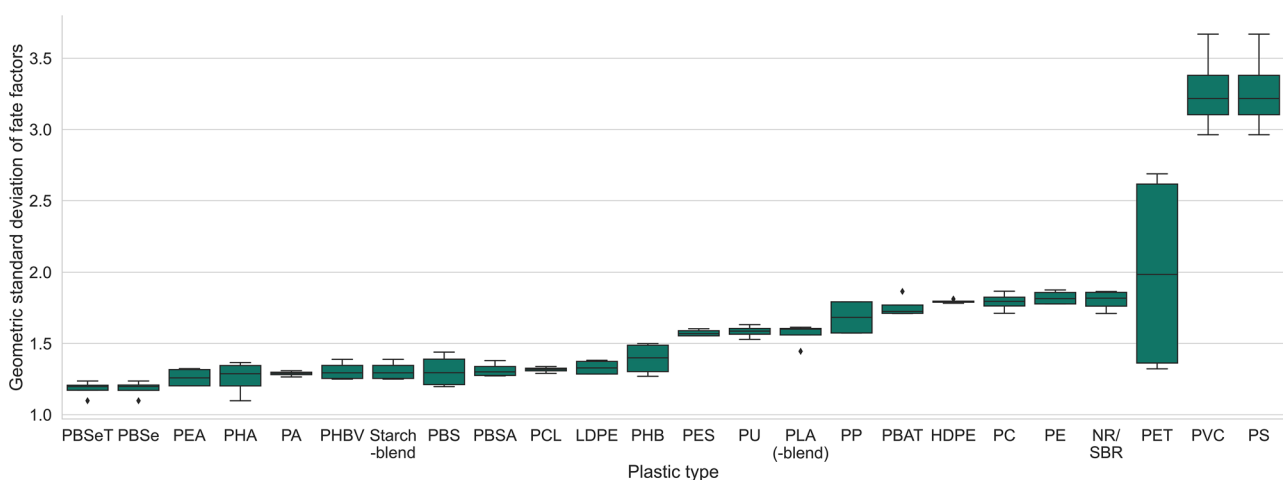


Fig. 13 Geometric standard deviation of FFs for each plastic type

the GSD values for the FFs of PET. The GSD of the SDDR in marine and river sediment is 1.37, in marine water 1.25, and in soil 2.87. For the remaining polymers, FFs GSDs are lower (IQR ranges between 0.01 and 0.3).

We assume that degradation happens exclusively at the surface of the emitted item. However, some polymers might degrade more volume-driven (bulk degradation) than surface-driven, such as PET and polyamides (Lyu et al. 2005; Pickett and Coyle 2013). Although the total surface area of the plastic emission (e.g., macro vs. microplastic) might play a role in the redistribution of the emission, our methodology only considers its characteristic length. All other differences like shape and size are neglected, although they might have an impact on the redistribution (e.g., airborne particles, migration in soil).

Another limiting factor is that specific additives were not taken into account in this publication. For example, the general composition of car tire is approximately 40–60 wt% NR/SBR (Wagner et al. 2018; Wik and Dave 2009); the remains are a mixture of fillers, softeners, vulcanization agents, and additives (Baensch-Baltruschat et al. 2020; Wagner et al. 2018; Wik and Dave 2009). Similarly, Ioakeimidis et al. (2016) based their degradation measurements on littered PET bottles found in the Aegean Sea, for which no information is available concerning additives used in production. For similar cases, degradation of the emissions is different from pure polymers or a mixture of polymers.

We limit the environmental compartments to freshwater, marine water, soil, river sediment, and marine sediment in the described methodology. However, in the future, when more data become available for redistribution and degradation, additional environmental compartments should be considered. For example, we did not differentiate between different marine compartments other than water and sediment, although the degradation rate can differ in various marine compartments (eulitoral, pelagic, benthic) and climate zones (Lott et al. 2020). Differentiation in climate zones is still an issue regarding regionalized FFs. Regionalization should also be applied to the above-mentioned redistribution rates. For example, road runoff treatment is central in determining the number of plastic particles ending up in surface water. Water management differs between countries or regions, resulting in large differences in the amount released to freshwaters. Even within one country, different runoff water treatment systems can co-exist.

## 5 Conclusions and perspectives

The proposed methodology is a crucial step to consider plastic emissions to the environment in LCA. We proposed FFs and respectively CFs for plastic emissions allowing to

calculate impacts of plastic pollution measured in plastic pollution equivalents, based on plastics' residence time in the environment. We also provided a basis for developing a future impact category addressing potential impacts caused by plastic emissions in LCA. Regarding some aspects of the calculations, assumptions or estimations were made, which call for quality assurance by increasing available information. The methodology consists of several independent elements which can be replaced or improved independently:

- Degradation measurements (SSDRs) retrieved from literature,
- The degradation model (in our case surface-driven degradation),
- Redistribution patterns,
- An FF based on the residence time,
- Estimation of the data quality by a pedigree matrix and uncertainty analysis.

As pointed out before, the SSDRs derived from literature entail uncertainty due to, e.g., measurement inaccuracies or additives. The retrieved values can be replaced by more accurate or certain ones, once available, especially concerning degradation data the redistribution between different environmental compartments. In the future, these data should focus on those from experiments where degradation measurements are obtained according to the test setup suggested by Lott et al. (2020). That means, after proving a polymer's biodegradability in the laboratory, an as good as possible standardized real-life experiment should be conducted, leading to more realistic and comparable data and reducing the impact of artificial conditions on the one hand and exceeded weight loss rates caused by fragmentation on the other hand.

Other more sophisticated approaches can replace the degradation model (surface-driven degradation), e.g., Junker et al. (2016). Even different approaches for different polymers can be used in one model. The residence time approach shows several advantages when treating time horizons and combining different compartments compared to the more familiar half-lifetime approach. However, the latter one could be used instead, if preferred.

Concluding, the proposed FFs can serve as the first indication of plastic emissions' fate in the environment. They may be used as a proxy where more detailed information is unavailable to evaluate micro- and macroplastics' potential aquatic and terrestrial impacts. The proposed methodology is flexible and can be adapted to available data concerning a specific product or process, e.g., the characteristic length or shape of the emission. In order to fully characterize the impact of plastic emissions on ecosystems, biodiversity, and humans, our proposed FFs need to be combined with factors for the probability of the exposure of humans or

organisms to the plastic emission (exposure factor) and for the impacts of plastics on species (effect factor). The effect factor should also take into consideration the expected severity of the damage. Coupling our approach with other work on exposure factors, effect factors, and marine litter impact models is possible but might need adjustments (Lavoie et al. 2021; Saling et al. 2020; Woods et al. 2019, 2021). Besides, each methodological approach only addresses a particular type of impact, for example, physical impacts such as entanglement or chemical impacts such as toxicity. Combined, they are not exhaustive and leave out specific impacts caused by alien species or pathogens transported on plastic emissions.

In addition, the process of releasing chemicals and metabolites during degradation is still not investigated but could be crucial for assessing the effect of plastic emissions (Lambert et al. 2014). For example, additional hazards caused by the release of additives processed in the plastics can later be incorporated into this model in a way similar to the USEtox methodology (Rosenbaum et al. 2008). In order to address possible toxicological risks caused by the release of additives, fillers, or reinforcement substances and to integrate the suggested model with ecotoxicological data, their types and degree should be included in the elementary flows' definition if known. Therefore, additional research is needed to yield comprehensive effect factors and develop complete CFs for micro- and macroplastics in marine and terrestrial environments. Finally, although not yet in the public focus, soluble polymers such as used, for example, in detergents, might be harmful, too. Further methodological development is needed for their consideration since they probably behave differently in the environment and parameters such as the characteristic length are difficult to assess.

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**Data availability** All data generated or analyzed during this study are included in this published article and its supplementary information files.

## Declarations

**Conflict of interest** The authors declare no competing interests.

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