Macroplastic fragmentation in rivers

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Abstract. The process of macroplastic (>0.5 cm) fragmentation results in the production of smaller plastic particles (micro- and nanoplastics), which threaten biota and human health and are difficult to remove from the environment. The global coverage and long retention times of macroplastic waste in fluvial systems (ranging from years to centuries) create long-lasting and widespread potential for its fragmentation and the production of secondary micro- and nanoplastics. However, the pathways and rates of this process are unknown, which constitutes a fundamental knowledge gap in our understanding of macroplastic fate in rivers and the transfer of produced microparticles throughout the environment. To set the stage for future research aiming to fill this gap, we present a conceptual framework which identifies two types of riverine macroplastic fragmentation controls: intrinsic (resulting from plastic item properties) and extrinsic (resulting from river characteristics and climate) processes. First, based on the existing literature, we identify the intrinsic properties of macroplastic items that make them particularly prone to fragmentation (e.g., the film shape, low polymer resistance, previous weathering). Second, we propose a conceptual model showing how extrinsic controls can modulate the intensity of macroplastic fragmentation in perennial and intermittent rivers. Using this model, we hypothesize that the inundated parts of perennial river channels—as specific zones exposed to the constant transfer of water and sediments—provide particular conditions that accelerate the physical fragmentation of macroplastics resulting from their mechanical interactions with water, sediments, and riverbeds. The non-inundated parts of perennial river channels provide conditions for biochemical fragmentation via photo-oxidation. In intermittent rivers, the whole channel zone is hypothesized to favor both the physical and biochemical fragmentation of riverine macroplastics, with the dominance of the mechanical type during the wet season. Our conceptualization aims to support future experimental works quantifying macroplastic fragmentation rates in different types of rivers.

Key words**:** *plastic abrasion, plastic breakdown, mechanical weathering, mechanical degradation, river hydrodynamics*

Highlights

- Rivers as unexplored environments of macroplastic fragmentation.
- A proposed conceptual model for riverine macroplastic fragmentation.
- Physical characteristics of river channel as controls of mechanical fragmentation.
- Climate as a control of biochemical fragmentation.

1. Introduction

Rivers are practically unexplored as macroplastic (plastic particles larger than > 0.5 cm) fragmentation environments, and information on its control and rates here is mostly unknown (Delorme et al., 2021; Williams and Simons, 1996). However, in most climates, river channels are characterized by a continuous flow of water and sediments and exposure to sunlight, which can favor the fragmentation of macroplastic waste through both physical and mechanical forces. Moreover, recent works have indicated that rivers are polluted with macroplastic globally, and the timescales of its retention in fluvial system can range from years to centuries (Liro et al., 2020; van Emmerik et al., 2022). This long-lasting presence and widespread occurrence of macroplastics in rivers pose risks of the production and dispersal of secondary micro- and nanoplastics in and beyond rivers.

Existing works have demonstrated that macroplastic can be fragmented into smaller micro- (<1-5 mm) and nanoplastic particles (<0.1-1 μm) through biochemical and physical forces (Andrady, 2015; Dimassi et al., 2022; Geewert et., al., 2015; Zhang et al., 2021) (Fig. 1). The produced microparticles (called secondary micro- and nano-plastics) are difficult to track and remove from the environment (Napper and Thomson, 2019; van Wijnen et al., 2019) and are known to pose numerous risks for biota and human health (Leslie et al., 2022). The process of macroplastic fragmentation, defined as the breaking of a macroplastic object into smaller particles, in the environment is very poorly understood, and up to now, it has mostly been studied in oceans (Andrady 2015; Andrady et al., 2022; Dimasi et al., 2022; Zhang et al., 2021) or by laboratory experiments (Boersma et al., 2023; Chubarenko et al., 2020; Gerritse et al., 2020; Kalogerakis et al., 2017; Lambert and Wagner, 2016). The macroplastic fragmentation rate is quantified by (i) the loss of weight of the macroplastic object (mass loss/time) (e.g., Maga et al., 2022), (ii) changes in its surface characteristics (e.g., surface roughness change (Reineccius et al., 2023)) or specific surface degradation rate (e.g., SSDR in Chamas et al., 2020), or (iii) the number or mass of microparticles produced (e.g., Lambert and Wagner, 2016).

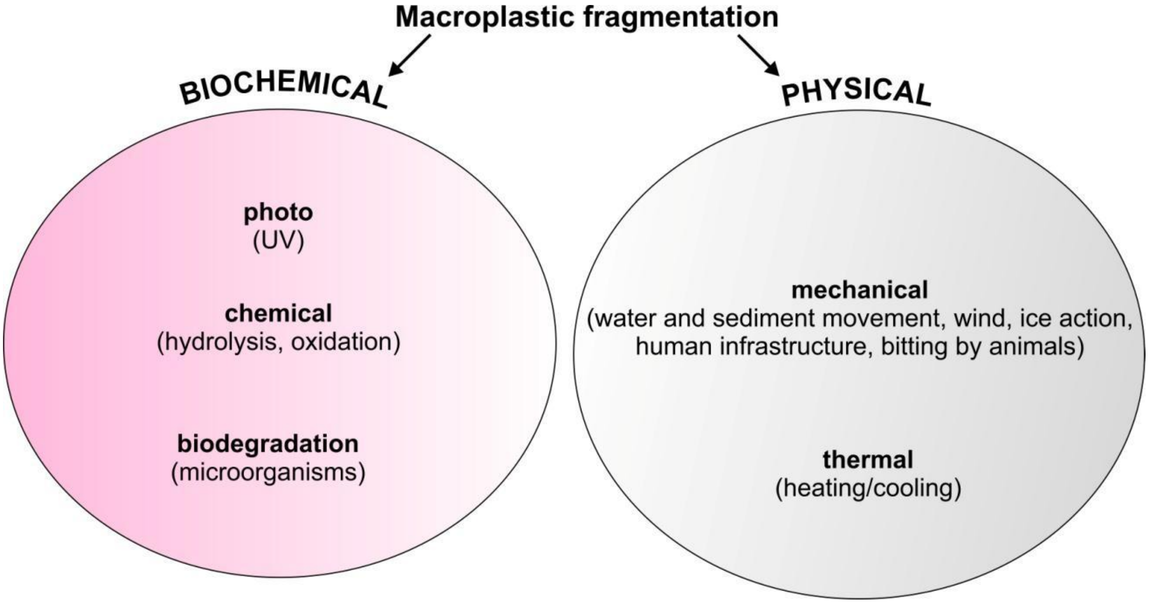

Figure 1. Outline of the types of forces which can result in macroplastic fragmentation. The proposed division into biochemical and physical degradation and fragmentation processes was developed based on previous works (Andrady, 2015; Dimmisi et al., 2022; Geewert et., al., 2015; Zhang, 2021) (for details, see Section 2).

Among the physical forces able to fragment macroplastic, mechanical force, such as wave action on a beach, is known to provide an opportunity for the collision and abrasion of macroplastic, resulting in its breakdown (Corcoran et al., 2009). Recent experiments showed that macroplastic fragmentation induced by mechanical forces (or the interaction of mechanical and biochemical forces) can proceed a few orders of magnitude faster than that resulting from solely biochemical forces (Sun et al., 2022). Moreover, microparticles produced through mechanical fragmentation are smaller (nanometer scale) than those produced through biochemical fragmentation (see Sun et al., 2022). Mechanical fragmentation accelerates biochemical fragmentation, because fragmented macroplastic items have increased surface-area-to-mass ratios (providing a larger surface area available for biochemical forces) (Chamas et al., 2020) and surface textural changes, which initiate biochemical fragmentation (Copper and Corcoran, 2010; Corcoran et al., 2009; Song et al., 2017; Zbyszewski and Corcoran, 2011) (Fig. 2). This relationship also indicates the importance of mechanical fragmentation, which can be seen not only as a product of macroplastic degradation but also as an environment-dependent process (Hurley et al., 2020) which, itself, can accelerate the rate of macroplastic fragmentation (Sun et al., 2022) (Fig. 2) and thus control the future presence of macroplastics in the environment.

To set a starting point for the future exploration of this knowledge gap, here, we developed a conceptual framework for the study of riverine macroplastic fragmentation. First, we divided controls of riverine macroplastic fragmentation into intrinsic (resulting from plastic item properties) (Section 2.1; Fig. 3) and extrinsic (resulting from river characteristics and climate) factors (Section 2.2; Fig. 4). Then, we conceptualized how extrinsic controls can modulate the fragmentation of macroplastics transported and stored in the inundated and non-inundated parts of the river channel. Our conceptualization can help planning of future experimental works aimed at the direct quantification of macroplastic fragmentation rates in rivers

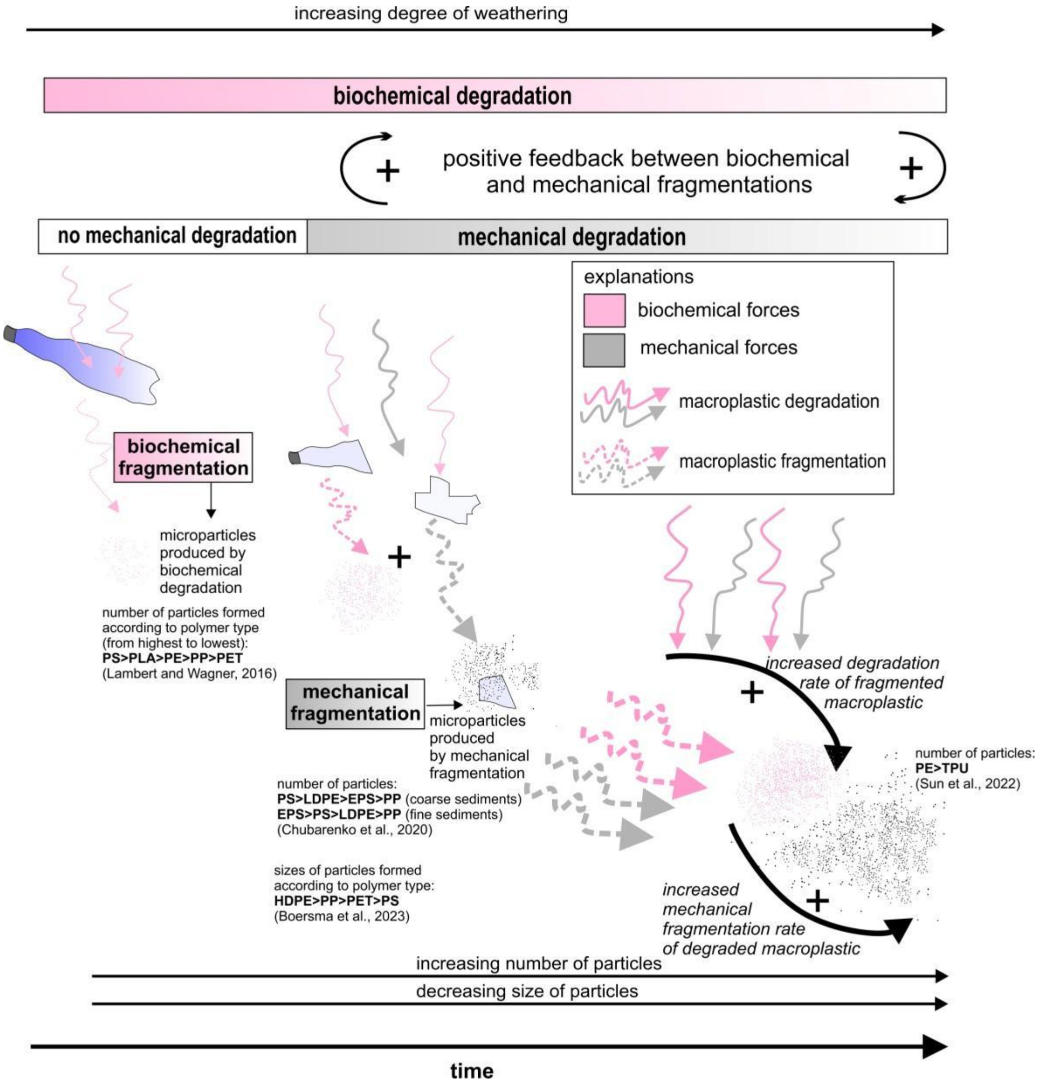

Figure 2. Visualization of microparticles formed through biochemical and mechanical fragmentations and the positive feedback processes between them.

2. Conceptualizing riverine macroplastic fragmentation

We categorized the types of macroplastic fragmentation, in relation to their main control factors, into *biochemical fragmentation* and *physical fragmentation,* similarly as was previously used for plastic degradation processes (Andrady, 2015; Dimmisi et al., 2022; Geewert et., al., 2015; Zhang, 2021). Biochemical fragmentation is defined as the breaking of macroplastic objects into smaller particles as a resulting of photodegradation (via UV irradiance), oxidation, hydrolysis, and biodegradation by microorganisms. Physical fragmentation is defined as the breaking of macroplastic objects into smaller particles as a result of mechanical and thermal factors. In the riverine environment, mechanical fragmentation describes the breaking of macroplastic objects as a result of interactions with the riverbed, water, and sediments (mineral, organic, synthetic), as well as plants (e.g., root action), ice (collision, freezing/thawing), animals (e.g., biting), and anthropogenic infrastructure (e.g., ships, weirs). Thermal fragmentation refers to the disaggregation of macroplastic objects resulting from heating/cooling (e.g., sunlight, wildfire). The degradation and fragmentation of plastics through thermal forces alone require high temperatures (Dimassi et al., 2022), which rarely occur in the ambient riverine environment; thus, in this paper, thermal forces are treated as accelerators for other types of degradation (see, e.g., thermal photo-oxidation in Dimmassi et al., 2022). The above-defined macroplastic fragmentation process can be seen as part of a more general plastic degradation process, understood as changes in the chemical and physical properties of plastics (see Andrady et al., 2015) (Fig. 2).

2.1. Intrinsic controls of macroplastic fragmentation

Information on intrinsic controls was extracted from the existing literature on macro- and microplastic fragmentation (Boersma et al., 2023; Chamas et al., 2020; Chubarenko et al., 2020; Song et al., 2017; Sorasen et al., 2022; Sun et al., 2022) and is summarized in Figure 3. The existing literature indicates that the potential for macroplastic fragmentation resulting from the intrinsic properties of macroplastics is a function of the plastic polymer composition, degree of previous weathering (see the x-axis in Fig. 3), and mass-to-surface-area ratio of the macroplastic item (Chamas et al., 2020) (see the y-axis in Fig. 3).

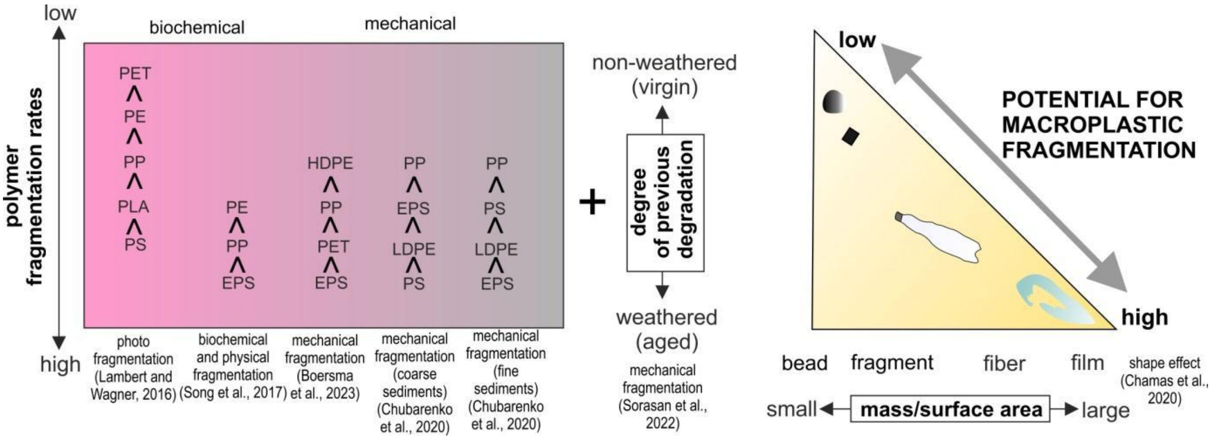

Figure 3. The synthesis of intrinsic plastic properties effects on macroplastic fragmentation. Intrinsic properties of macroplastic fragmentation are synthesized as a function of polymer resistance to biochemical and mechanical fragmentation (based on experiments conducted by Boersma et al., 2023; Chubarenko et al., 2020; Lambert and Wagner, 2016; Song et al., 2017), the degree of previous weathering of macroplastic item (Sorasan et al., 2022) (x-axis), and its mass-to-surface-area ratio (Chamas et al., 2020) (y-axis).

Polymer type

The polymer composition (Boersma et al., 2023) and degree of its previous weathering (Sorasan et al., 2022) control the physical and chemical characteristics of plastics. The additives added to commercial polymers are also changing their fragmentation potential (Dimmisi et al., 2022), which may cause different rates of biochemical fragmentation of the same polymer type in the same environments (Maga et al., 2022). Existing laboratory experiments showed that the rate of plastic fragmentation (understood as the number of produced particles or macroplastic item mass loss) was the highest for rigid (PS) or expanded polystyrene (EPS) and lower for low-density polypropylene (LDPE), polypropylene (PP) polyethylene (PE), and polyethylene terephthalate (PET) (Fig. 3). The fragmentation of PS and EPS polymer types was identified as the highest in experiments on mechanical (Boersma et al., 2023; Chubarenko et al., 2020; Song et al., 2017) and biochemical fragmentation (Lambert and Wagner, 2016; Song et al., 2017). Some experiments documented, for example, that rate of mechanical fragmentation (measured as the item mass loss) is 1000-10000 higher for PS and EPS than it is for PP polymer (Chubarenko et al., 2020). It was also indicated that mechanical fragmentation resulted from macroplastic interactions with coarser sediments (pebbles), producing 5 to 145 times more microparticles than fine sediments (sand), with the value of these difference being highly dependent on the polymer type (143x PS, 145x LDPE, 5x EPS, and 25x PP (Chubarenko et al., 2020). These observations provide an interesting background for the further quantification of riverine macroplastic fragmentation in rivers with coarse- and fine-grained bed (see Liro et al., 2022). It was also experimentally documented that microparticles formed via EPS and PS polymer fragmentation have smaller sizes than those released from PP and PET, with the largest formed from HDPE plastic (Boersma et al., 2023). Numerous riverine macroplastic waste items (e.g., cups, meat trays, plastic cutlery and straws, building insulation) are built from these fragmentation prone polymers (EPS, PS) (Plastic Europe 2021; Gonzalez-Fernandez et al., 2021; Tasseron et al., 2023). Gaining field-based information on their fragmentation rates seems to be especially important for our further understanding of the risks resulting from riverine macroplastic pollution. To describe consequences of fragmentation of different types of riverine macroplastics waste, we introduce the term *plastic footprint* which reflects the number and size of plastic particles released to the environment during its presence in the river.

Degree of previous weathering

As one of the intrinsic properties of macroplastic important for its fragmentation, we also considered the degree of plastic weathering resulting from its previous degradation (Sorasan et al., 2022) (Fig. 3). Previous experiments showed that aged plastics have a fragmentation rate three orders of magnitude higher than the virgin one (Sorasan et al., 2022). Moreover, the fragmentation of aged plastic releases more nanoparticles (Sorasan et al., 2022), which are particularly harmful for biota and human health (Leslie et al., 2022). Thus, the inclusion of the degree of riverine macroplastic weathering as one of the controls of fragmentation seems to be important, because rivers are frequently eroding floodplains, thus remobilizing sediments containing aged macroplastic waste dumped in these locations in the past. Future efforts should focus on detecting and managing such zones to avoid the input and accelerated fragmentation of aged macroplastics in rivers. Taking into account the fact that weathering makes macroplastic more brittle (Boersma et al., 2023), special attention should be paid to the avoidance of its input into inundated channel zones where it can be mechanically fragmented (see Fig. 4).

Mass-to-surface-area ratio

Biochemical and physical processes able to fragment macroplastic items take place mostly on the surfaces of macroplastic objects (Chamas et al., 2020). The potential for fragmentation of macroplastic objects was reported to be the highest for thin-film- and fiber-shaped objects, which have the highest surface-area-to-mass ratios, giving them larger surface areas which can actively interact with biochemical and physical factors of plastic degradation (Chamas et al., 2020) (Fig. 3). For example, the degradation of film-shaped macroplastic objects was reported to be 260 to 1100 times higher than that of fiber- and bead-shaped objects of the same volume and polymer type (see Chamas et al., 2020). There are numerous examples of riverine macroplastic waste with film shapes, which enter rivers as packaging, buildings, and agricultural litters. Such items are frequently trapped in obstacles present in the inundated parts of river channels (e.g., boulders, wood jams) (Liro et al., 2022, 2023) or riparian vegetation (Cesarini and Scalici, 2022), providing them with ideal exposure to both mechanical (via water movement, wind, rain) and biochemical factors of fragmentation (via photo-oxidation) (Fig. 4).

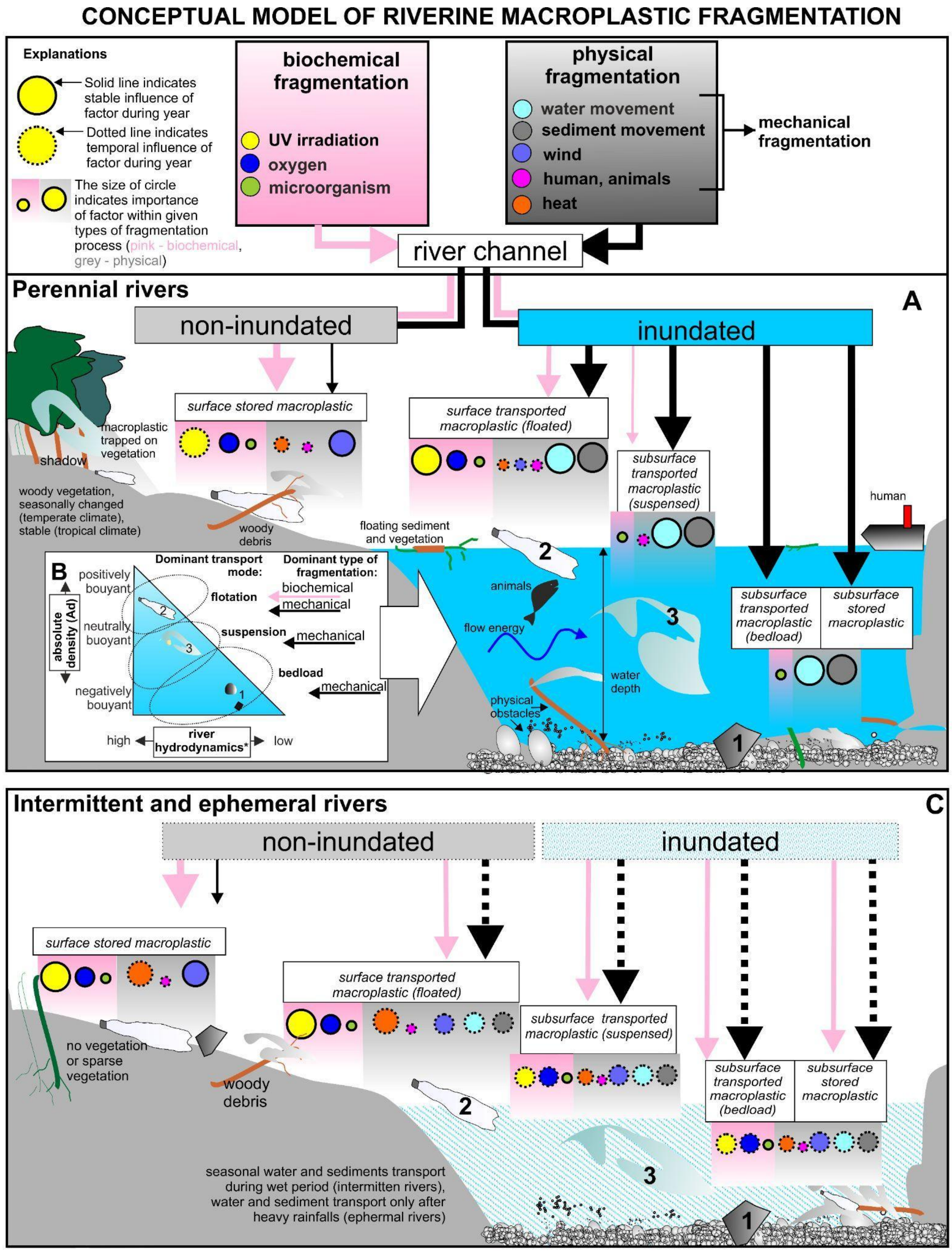

Figure 4. Conceptual model illustrating the intensity of biochemical and physical forces governing processes of macroplastic fragmentation in rivers. The model structures differences between these two

types of fragmentation in the inundated and non-inundated parts of the river channel for perennial (A) and intermittent rivers (C). The relationship between the absolute density of macroplastic objects and river hydrodynamics as controls of the dominant transport mode of macroplastic (B) is based on Russell et al., 2023. The numbers 1 (flotation), 2 (suspension), and 3 (bed-load) visualize different transport modes of macroplastic debris in flowing water. The intensity of biochemical and physical fragmentation is indicated by the arrows' widths. The importance of given factors of fragmentation is visualized by circle sizes.

As we illustrated in Figure 3, intrinsic properties can bidirectionally interact with each other, enhancing or reducing potential for macroplastic fragmentation. For example, due to its shape, foil, having a high surface-area-to-mass ratio, can compensate for the low fragmentation potential of such an object as a result of its high resistance (e.g., HDPE), and vice versa, the fragmentation of an object built with a polymer having high degradation potential (e.g., EPS or PS) can be reduced due to its compact shape. Taking into account a very wide range of reported differences in plastic fragmentation rates resulting from different polymers (e.g., up three to four orders of magnitude, Chubarenko et al., 2020) and shapes (e.g., up to three orders of magnitude, Chamas et al., 2020), the degree of such compensation can be potentially very high.

2.2 Effects of extrinsic controls on riverine macroplastic fragmentation

Our conceptualization illustrates different intensities of biochemical and physical fragmentation between the inundated and non-inundated zones of perennial and intermittent rivers channels. These differences result from the changing influence of factors controlling these processes. The most important difference between the inundated and non-inundated parts of river channels is presence of water and sediment transport, which favors mechanical fragmentation and limits potential for biochemical fragmentation (by photo-oxidation) (Fig. 4). We also assume that potential for biochemical fragmentation may be regulated by the macroplastic transport mode because of different levels of exposure of differently transported and stored macroplastics to UV radiation and oxygen (Fig. 4B).

River hydromorphology

In the inundated parts of perennial rivers (transporting water throughout the whole year), mechanical fragmentation is hypothesized to be dominant because of the chronic potential for mechanical collision of transported and stored macroplastics with water and sediments (Fig. 4A). Perennial rivers in populated areas are also frequently utilized by humans for navigation, recreation, and other purposes (e.g., dams, water extraction), which can favor the mechanical interaction of macroplastics with human infrastructure (e.g., with vessels, electric dam turbines, weirs). The potential for biochemical fragmentation in inundated parts of perennial river channels is generally reduced by the presence of water, which limits UV irradiation and oxygen exposure, representing the main drivers of biochemical fragmentation (Andrady, 2015). Only macroplastic transported in flotation can be influenced by UV irradiation and oxygen in similar ways like those stored in non-inundated part of the river channel (Fig. 4A). Water penetration by UV irradiance is highly dependent on location and water characteristics (e.g., dissolved and suspended sediment concentration) and is typically substantially reduced in water deeper than a few meters (Dunne and Brown, 1996). Thus, macroplastics transported in suspension and as a bedload in most perennial rivers can be fragmented mostly through mechanical forces (Fig. 4A and B). Only in the very shallow river waters transporting clear water a potential for UV irradiation and related fragmentation exists. The mode of transport of each sediment transported in rivers (including plastics) is ultimately controlled by river hydrodynamics and sediment particle characteristics (see Kuizenga et al., 2022; Valero et al., 2022; Russell et al., 2023). The dominant transport mode of a macroplastic can be precisely evaluated using absolute density (see Russell et al., 2023) (Fig. 4B). For example, air-filled plastic bottles, independent of the density of the polymer types they are composed of, will be transported in river channels in flotation because of their low net density. The same type of bottle filled with water or sediments will be transported in suspension or as a bedload (see Fig. 13A in Russell et al., 2023). It is important to highlight here that different transport modes of macroplastics in flowing water also modulate the intensity of macroplastic interactions with water, sediments, and riverbeds. For example, the same macroplastic object can be transported in suspension and as a bedload, respectively, in fast-flowing rivers with steep channel slopes and in slow-flowing rivers with gentle channel slopes. The modes of transport can also vary within the same river reach for different flow conditions, with high potential for transport during floods and lower potential in normal conditions. It is important to note that the mode of transport of the same macroplastic object can also dramatically change locally, for example, between different bed morphologies and channel patterns. The example can be a river with a steep pool sequence, as is common in mountain rivers (Wohl, 2010). Such a bed morphology has hydrodynamics which are highly variable locally with fluctuations between shallow, fast-flowing step zones and deeper, slower-flowing pool zones (Wohl, 2010). Thus, river hydrodynamics, together with the buoyancy of macroplastic waste objects, are important and unexplored factors determining the intensity of macroplastic interactions with river water, sediments, and beds. Taking into account the fact that river hydrodynamics are highly variable in both time and space, there is a need for future field experiments to directly quantify the intensity of macroplastic fragmentation for macroplastic items with different fragmentation potential (Fig. 3) in different flow conditions and for different hydromorphological types of rivers (see, e.g., Liro et al., 2022). Artificial modifications of river hydromorphology (e.g., through channel regulation, bed and bank construction), can also be investigated in the context of riverine macroplastic fragmentation in future works.

River channel hydromorphology, as a control of the physical fragmentation of macroplastics, seems to be less important in the case of intermittent and ephemeral rivers which transfer water and sediments only periodically (Fig. 4C). Such flow events in intermittent or ephemeral rivers are frequently triggered by heavy rainfall and have high energy capable of transporting coarse sediments (Martin-Vide et al., 2019). As discussed in the previous section, such a phenomenon can effectively fragment macroplastic present in a river channel (see Figs 3). Intermittent rivers can also transport products previously subjected to biochemical fragmentation during such events.

Climate

In non-inundated parts of perennial rivers and within the whole channel zone of intermittent rivers, biochemical fragmentation is hypothesized to be dominated because of exposure to UV irradiation and oxygen, and physical fragmentation can occur mostly due to wind action (Fig. 4). Depending on the local setting (e.g., the surrounding vegetation types, human infrastructure) and climate, the intensity and importance of a given fragmentation type will change. It may be interesting to experimentally compare biochemical fragmentation rates of the same macroplastic wastes in the non-inundated zones of rivers in different climatic zones in future works (see e.g., Delorme et al., 2021).

Future works can also explore effects of macroplastic colonization by biota as factor modulating biochemical and mechanical fragmentation rates of riverine macroplastic. Recent evidences suggest a positive feedback mechanism between colonization and degree of macroplastic degradation in perennial rivers (Galitelli et al., 2023). However, the pathway of this process in perennial and intermittent rivers in different climates is unknown.

Future outlook

Our theoretical framework aims to provide a guide for the future exploration of riverine macroplastic fragmentation. We identified (i) macroplastic item properties that impact its fragmentation, (ii) the relevant biochemical and mechanical fragmentation processes, and (iii) their relevant importance in different riverine environments. Our conceptualization can support planning of future experimental work aimed at the direct quantification of macroplastic fragmentation rates in rivers. Such information is currently mostly unavailable for rivers (Delorme et al., 2021), but is of crucial importance for understanding secondary micro- and nano-plastic production in rivers, the transfer of these harmful particles throughout the natural environment, and the imposed risks. We hypothesize that inundated part of the perennial river channels provide conditions that particularly accelerate mechanical fragmentation of macroplastic resulting from its mechanical interaction with water, sediments and riverbed. We highlights a need for (i) future experimental quantification of the rates of mechanical fragmentation of common macroplastic items between lower-energy lowland rivers and higher-energy mountain rivers, (ii) the evaluation of the importance of artificial modifications (e.g., channelization, dams) of river physical characteristics on the spatial and temporal trajectories of mechanical fragmentation of macroplastics, (iii) the detection of compartments of fluvial systems (e.g., geomorphological forms, reach types, catchments, zones of fluvial systems) operating as generators and sinks of secondary micro- and nanoplastics (see, e.g., Liro et al., 2022), and (iv) the evaluation of the downstream dispersal of secondary microplastics from such hotspots. Such data will allow for an evaluation of the amount of secondary microparticles formed during the macroplastic's journey throughout the river. Information on this *plastic footprint* for a given type of macroplastic waste is a first step in the planning of its future mitigation.

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

Author Contributions

ML conceptualized paper idea, wrote the original draft, and created original figures; **AZ** reviewed the literature and contributed to writing the original draft and the figures' preparation; **TvE** took part in writing and revising the manuscript and figures.

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