



Short communication

First attempt to measure macroplastic fragmentation in rivers

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ABSTRACT

Direct field measurements of macroplastic fragmentation during its transport in rivers are currently unavailable, and there is no established method to perform them. Previous studies have showed that macroplastic fragmentation results in the production of harmful microplastics, and river channels can be hotspots for this process. Therefore, obtaining information about this process is crucial for quantifying the production of secondary microplastics in rivers and assessing the related risks for riverine biota and human health. Here, we propose a simple low-cost methodology for quantifying riverine macroplastic fragmentation by conducting repeated measurements of the mass of tagged macroplastic items before and after their transport in the river. As a proof-of-concept for this method, we conducted a 52–65 day experiment that allowed us to measure a median fragmentation rate of 0.044 ± 0.012 g for 1-liter PET bottles during their transport at low to medium flow in the middle mountain Skawa River in the Polish Carpathians. Using the obtained data ($n = 42$), we extrapolated that during low to medium flows, the median yearly mass loss of PET bottles in the study section is 0.26 ± 0.012 g/year (0.78 ± 0.036 % of bottle mass), and the median rate of bottle surface degradation is 3.13 ± 0.14 $\mu\text{m}/\text{year}$. These estimates suggest a relatively high fragmentation rate for a PET bottle in a mountain river even under low to medium flow conditions without high-energy transport. We discuss how our simple and relatively low-cost methodology can be flexibly adapted and future optimized to quantify macroplastic fragmentation in various types of rivers and their compartments, informing future mitigation efforts about the rates of formation and dispersion of secondary microplastics.

1. Introduction

Tracking rates of macroplastic fragmentation in various environmental compartments is fundamentally important for evaluating the risk of plastic pollution for biota and human health. This provides direct insights into the amount of secondary microplastics (Maga et al., 2022) as well as their harmful additives and contaminants that can be released within these compartments, ultimately harming living organisms and, through the food chain, affecting human health (Hahladakis et al., 2018; Karlsson et al., 2021; Chen et al., 2022; Behnisch et al., 2023; Wagner et al., 2024). Field-based information on the rates of macroplastic fragmentation in different environments is, however, very limited (Chamas et al., 2020; Hurley et al., 2020; Maga et al., 2022) especially for rivers (Liro et al., 2020, 2023a,b; Delorme et al., 2021; Honorato-Zimmer et al., 2021). Recent works have, however, hypothesized that river channels can operate as hot-spots of macroplastic fragmentation because of constant movement of water and sediments in this zone which can favor mechanical interactions of macroplastic with water,

sediments, and riverbeds (Liro et al., 2023a,b). The intensity of these interactions can be particularly high in the case of mountain river channels, where high-energy water and sediment transport coincide with the presence of numerous physical obstacles such as boulders, bedrock, and large wood within the river channel (Liro et al., 2023a). Field experiments exploring this process have not yet been conducted. However, obtaining direct information about the rate of macroplastic fragmentation in mountain rivers is crucial for quantifying the production of secondary microplastics in these environments and evaluation of related risks to their biodiversity (see e.g., Wohl, 2010; Hauer et al., 2016), quality of resources they provide for human populations (e.g., water resource (Viviroli et al., 2020), human health (Hahladakis et al., 2018; Karlsson et al., 2021; Chen et al., 2022; Behnisch et al., 2023; Wagner et al., 2024) and understanding the extent to which they can be transported downstream to lowland rivers and oceans (Liro et al., 2023a, b).

Here, we propose a simple field-experiment based methodology for quantifying macroplastic fragmentation rates during its transport in

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river channels. Our methodology implements mass loss quantification of macroplastic objects, previously utilized in laboratory experiment (Gerritse et al., 2020) to tagged macroplastic objects transported in river channel (Fig. 1). Using this methodology, we have quantified the mass loss of 1-litre PET bottles occurring during their short-term transport (52–65 days) over distances ranging from 0.37 km to 16.27 km in a mountain river channel in the Polish Carpathians, under low- to medium-flow conditions (Fig. 2). The objective of this work is to present this methodology and report the first insights into macroplastic fragmentation in mountain rivers obtained through its application.

2. Materials and methods

2.1. Proposed methodology for quantify riverine macroplastic fragmentation

Our methodology combines mass loss quantification of macroplastic objects, previously utilized in laboratory experiments for determining macroplastic fragmentation (Gerritse et al., 2020), with macroplastic tracking techniques previously used to quantify the travel distance of tagged macroplastic objects transported in river channels (see e.g., Duncan et al., 2022). The proposed workflow consists of six steps: (1) measurement of the masses of virgin macroplastic objects, (2) transport of tagged items in the river, (3) cleaning and biofilm removal, (4) repeated measurements of macroplastic object masses, (5) correction of measured mass losses by accounting for cleaning error, and (6) calculation of macroplastic fragmentation metrics from the obtained mass loss information (see Fig. 1). The primary advantage of using mass loss as a proxy for macroplastic fragmentation in rivers, compared to other techniques previously used for quantifying macroplastic degradation and fragmentation (cf. Chamas et al., 2020), is its low cost and minimal need for laboratory analysis. Below, we describe how we applied this six-step procedure to quantify the fragmentation rate of 1-liter PET

bottles transported in the Skawa River in the Polish Carpathians (Fig. 2).

2.2. Measurement of the masses of virgin macroplastic objects (step 1)

Measurement of macroplastic mass loss as a proxy for its degradation and fragmentation has primarily been employed in laboratory experiments aimed at determining the effects of UV radiation, water movement, and biofilm formation on these processes (Gerritse et al., 2020). In our experiment, we utilized 177 ($n = 177$) virgin 1-liter bottles made from polyethylene terephthalate (PET). Initially, the mass of each bottle was determined (as the mean of triplicate measurements) (Table S2) using a precise laboratory balance with an accuracy of 0.001 g. Subsequently, the bottles were tagged with numbers drawn on the bottle caps and on a foil tag placed inside them (Fig. 3A).

2.3. Transport of tagged items in the river (step 2)

The field experiment was performed in the Skawa River (Polish Carpathians), a right-bank tributary of the Vistula River (the largest river in Poland) (Fig. 2A). The Skawa River has a total length of 96 km and originates at 700 m a.s.l. Its channel width ranges from 5 to 40 m within the study section. The river has a mountainous hydrological regime with little hydrological inertia, resulting in considerable flow variability and sudden but short-lasting floods. The total catchment area is 1160 km² and the average annual flow is 11 m³/s. The riverbed is predominantly composed of gravel and cobbles, with some sections of bedrock present in the middle course of the study section. All bottles were sealed with caps (Fig. 3A) and deployed into the river channel at three locations along the Skawa River in the Polish Carpathians on July 11th, 2022 (Fig. 1A-B). These locations were chosen along the 20 km-long study reach of the river, spanning from Osielec Village (location 1) to the Świnna Poręba Dam Reservoir (as depicted in Fig. 2B). After 52 days (September 1st), 57 days (September 6th), and 65 days (September

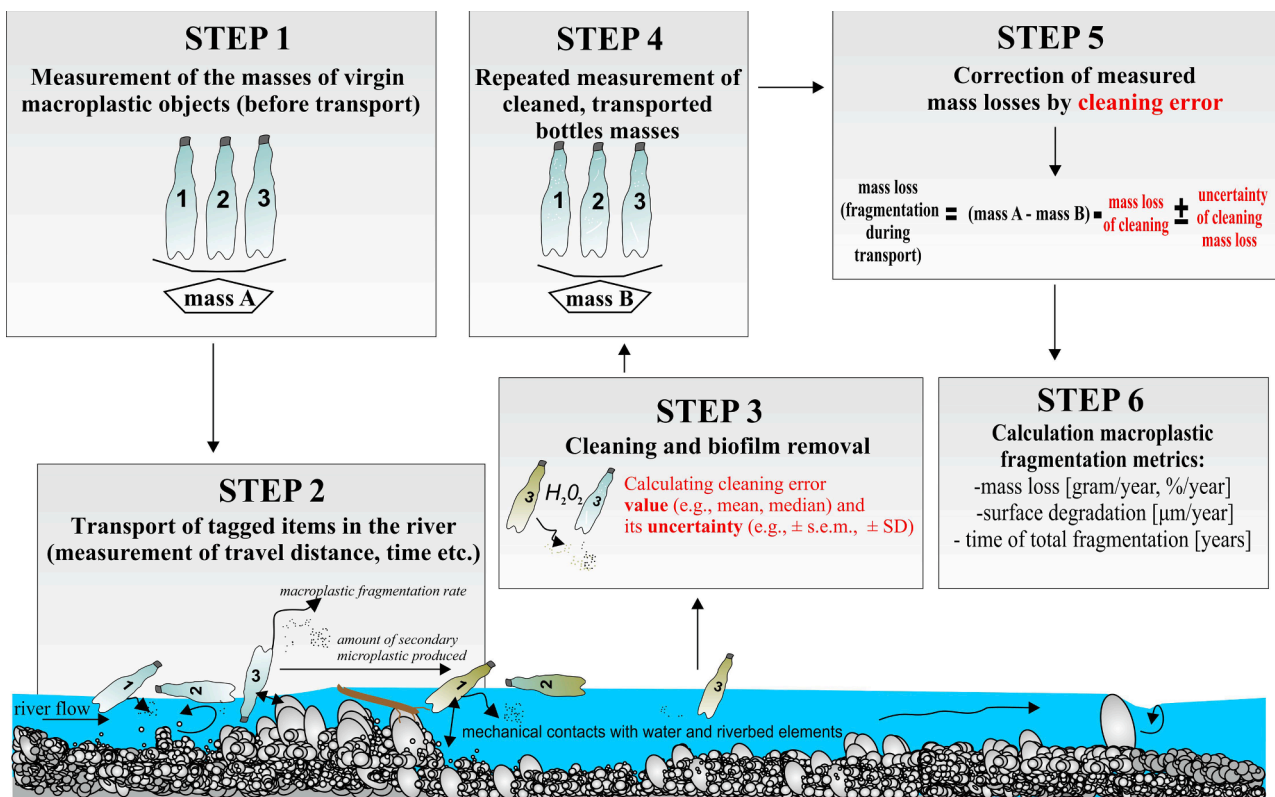


Fig. 1. The workflow of the proposed methodology for the quantification of riverine macroplastic fragmentation. Detailed explanations for the described steps are presented in the text.

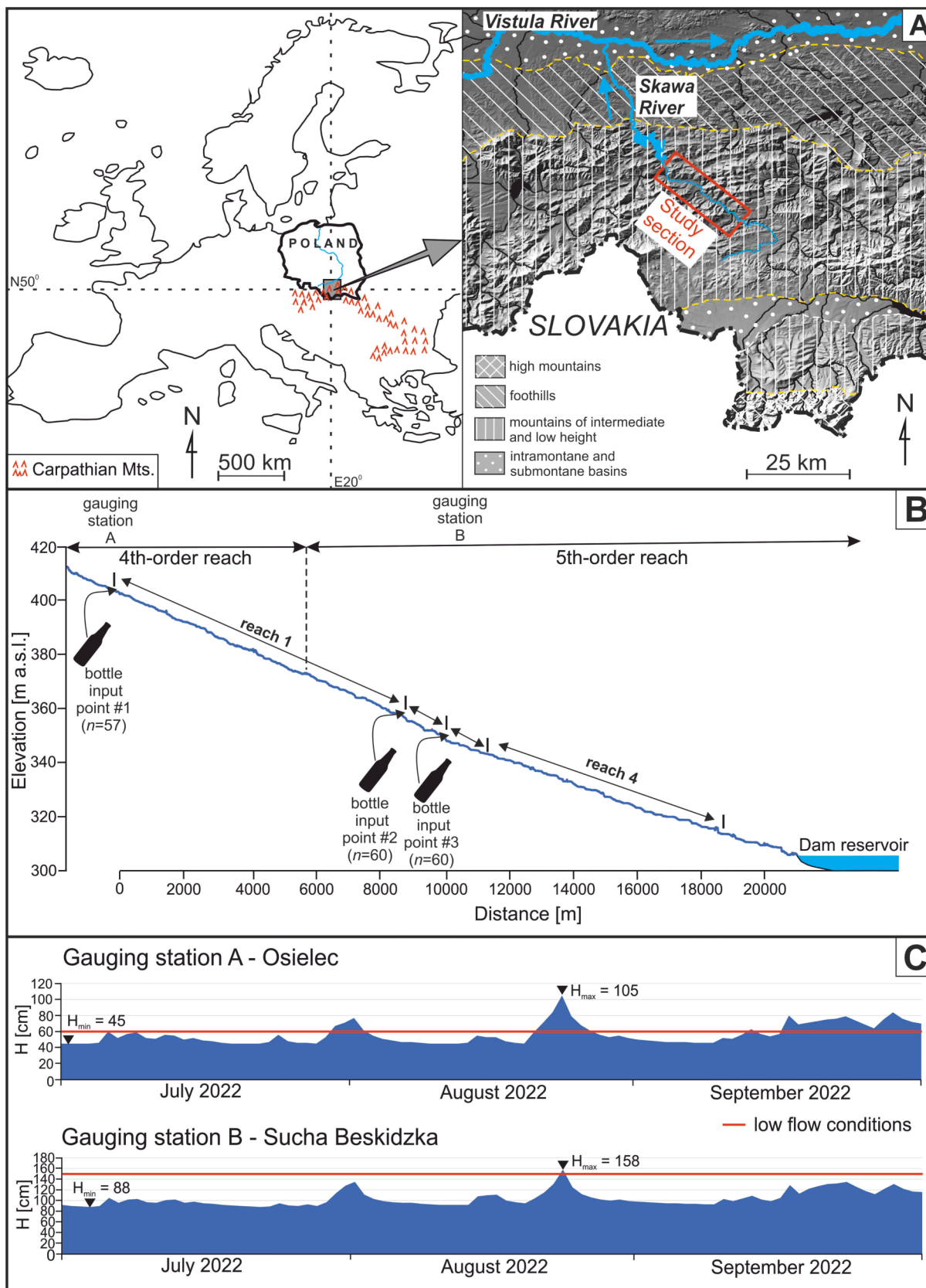


Fig. 2. A – Location of the study area; B – Longitudinal profile of the surveyed river section with bottle delivery points marked; C – Hydrograph of water levels for the gauge stations in the Osielec village and Sucha Beskidzka city occurring during the experiment.



Fig. 3. Tagged 1-litre PET bottles used in the experiment. Tagged bottles before (A), during (B, C) and after experiment (D). Last photo (D) indicates small cracks formed on the bottle surface during 52 days of transport in the river channel.

14th), the study reaches were surveyed by four persons (two on each river bank), enabling them to collect 43 of the previously deployed tagged bottles (as shown in Fig. 2A-C). The travel distances for each bottle were calculated as the thalweg distance between the point of bottle deployment and the location where the bottle was collected along the study reach (measured using an RTK GPS receiver). Subsequently, the collected bottles (Fig. 3B-C) were transported to the laboratory for cleaning and measurement of their mass loss resulting from mechanical fragmentation during their transport in the river channel.

2.4. Bottle cleaning and biofilm removal (step 3)

Before measuring the bottles' mass after their transport in the river, we employed a cleaning procedure similar to that used by Gerritse et al. (2020) (Fig. 1). Initially, the bottles were cleaned with tap water and detergent using a nylon brush, followed by a 12-hour incubation period in 30 % H₂O₂ to eliminate biofilms and other organic matter from their surfaces (Gerritse et al., 2020). Then, the bottles were rinsed in distilled

water and dried at 45 °C for six hours. Before drying, the bottles were opened, and the tagging numbers placed inside them before the experiment were removed. After cleaning, biofilm removal, and drying, each bottle was weighed, and the mass loss for each was determined in grams (Gerritse et al., 2020).

Similarly to previous mesocosm experiments utilizing mass loss as a proxy for macroplastic fragmentation (Gerritse et al., 2020), we accounted for the possibility that the cleaning procedure itself could cause a small-scale mass loss, potentially leading to an overestimation of the final results. To quantify this error and correct the final values of mass loss, we conducted a test cleaning on 24 reference bottles, measuring their masses before and after the cleaning procedure. The values of bottle mass loss during cleaning, determined from the mass loss of the 24 reference bottles (one bottle was excluded due to contamination during cleaning), were normally distributed, and their mean was found to be 0.022 g (Table S1). The uncertainty of this value estimation was quantified as the standard deviation of the mean (± 0.012 g).

2.5. Repeated measurements of macroplastic object masses (step 4) and cleaning error correction (step 5)

The mass loss of macroplastic objects resulting from their transport in rivers was determined by conducting repeated measurements of the dry macroplastic masses before and after their transport. The mass loss values determined for the bottles transported in rivers ($n = 43$) were not normally distributed. These values were then corrected using the mean value of bottle mass loss occurring during the cleaning procedure (0.022 g) and presented with the uncertainty of the cleaning error estimation (± 0.012 g) (Table S1-2). One of the corrected measurements of mass loss, which was lower (0.004 g) than the calculated cleaning error uncertainty (± 0.012 g), was removed from further analysis, and 42 measurements ($n = 42$) were further analyzed.

$$\text{macroplastic mass loss}_{\text{transport}} = (\text{mass}_{\text{before transport}} - \text{mass}_{\text{after transport}}) - \text{mass loss}_{\text{cleaning procedure}} \pm \text{uncertainty of cleaning error estimation} \quad (1)$$

2.6. Calculation of macroplastic fragmentation metrics from mass loss data obtained (step 6)

Utilizing the corrected mass loss values for the 42 bottles obtained during the 52–65 day experiment (Fig. 4), we calculated the yearly mass loss expressed in grams and as a percentage of the initial bottle mass (Fig. 5A-B). Additionally, we determined the rate of bottle surface degradation resulting from the calculated mass losses (Fig. 5C). For this calculation, we used the density of PET plastic (1.38 g/cm^3) and assumed that bottle fragmentation occurs evenly across their entire external surface ($\sim 610 \text{ cm}^2$) (Fig. 5C).

3. Results

The mass loss of the tracked 1-litre PET bottles ($n = 42$) during the 52–65-day transport in the river channel ranged from 0.015 g to 0.152 g (Table S2), with a median value of 0.045 and uncertainty of ± 0.012 g (Fig. 4).

Using the obtained data on bottle mass losses, we extrapolated the yearly mass loss to range from 0.098 g to 1 g (Fig. S2) with a median of

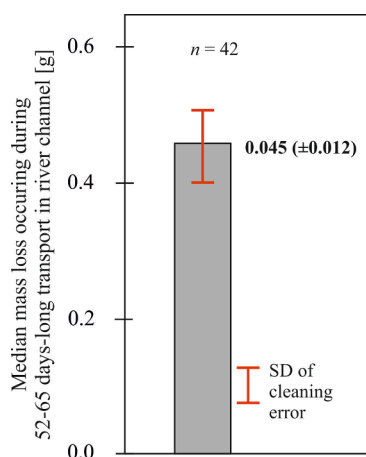


Fig. 4. Mass loss of 1-litre PET bottles occurring during 52–65 days of low to medium flow conditions.

0.26 ± 0.012 g (Fig. 5A), which constitutes 0.29 % to 2.97 % (median = 0.78 ± 0.036 %) of their initial masses (Fig. 5B) and surface degradation rates from 1.19 to 11.82 $\mu\text{m}/\text{year}$ (median = $3.13 \pm 0.144 \mu\text{m}/\text{year}$) (Fig. 5C). Based on these values, we estimated that the complete fragmentation of a used 1-liter PET bottle under the conditions represented by our experiment (low to medium flows) would take between 33.63 and 332.81 years, with a median estimate of 127.15 ± 0.046 years (Fig. 6).

The uncertainty resulting from the applied cleaning procedure constitutes 26.9 % of the mass loss measured during the 52–65 day experiment, 4.6 % of the extrapolated yearly mass loss, and 0.036 % of the extrapolated time for total fragmentation of the bottle.

4. Discussion and future outlook

We have implemented the measurement of macroplastic mass loss, previously utilized in laboratory experiments on macroplastic fragmentation (Gerritse et al., 2020), to quantify the rate of this process occurring during short-term transport of macroplastic in a middle mountain river channel. This method allowed us to quantify the fragmentation rates of 1-liter PET bottles (expressed as their mass loss) with an uncertainty of ± 0.012 g (SD of cleaning procedure error). The uncertainty resulting from the applied cleaning procedure constitutes 26.9 % of the median mass loss measured during the 52–65 day experiment. This relatively high proportion of measurement uncertainty (± 0.012 g) to the recorded mass loss change during the experiment (median = 0.045 g) can be explained by the short duration of the experiment, the absence of higher flows during its course, and the simple technique used for bottle cleaning. The uncertainty in the measurable value of mass loss using the proposed method could potentially be reduced in future experiments by applying our method to longer-term studies (e.g., one year). This would account for occurrences of higher mass losses even during low-energy conditions in rivers or by conducting experiments during higher-energy conditions (e.g., floods) or in rivers with higher-energy hydromorphologies (e.g., high-mountain streams). Future studies should also optimize cleaning procedures to reduce the value (0.022 g) and uncertainty (± 0.012 g) of cleaning errors. This will allow for the measurement of smaller magnitude mass losses during shorter-term experiments. Despite the aforementioned need for future improvements, the proposed method using mass loss as a proxy for macroplastic fragmentation offers a promising, cost-effective tool for collecting baseline information on the rates of macroplastic fragmentation in rivers and other environments.

In our experiment, we used a simple manual tagging method (numbers on a foil tag inserted into the bottle), which reduced the cost of the experiment but increased the time required to collect bottles in the field and reduced the bottle recovery rate. For future, longer-term experiments, it is essential to use appropriate tracking techniques (e.g., GPS, RFID, radio transmitters) (see e.g., Duncan et al., 2022), which facilitate easier retrieval of the objects from the field, thus improving the overall efficiency of the experiment and ensuring a higher recovery rate of the tracked macroplastics.

The bottles in our experiment were filled with air and sealed, causing them to float, which decreased their potential for mechanical fragmentation resulting from interactions with riverbed elements (Liro et al., 2023b). Transport in flotation can also increase their exposure to UV

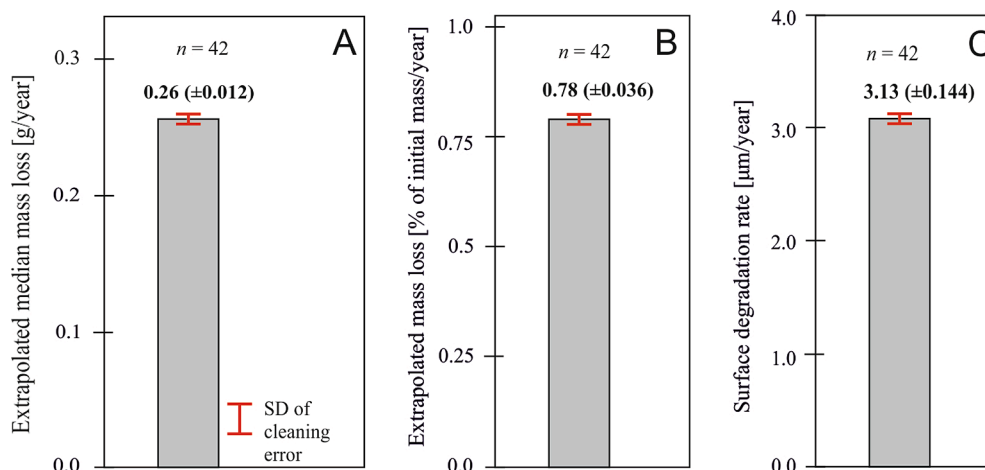


Fig. 5. Yearly mass loss (gram/year) (A) and surface degradation rate ($\mu\text{m}/\text{year}$) (B) of 1-litre PET bottles estimated based on the experiment results.

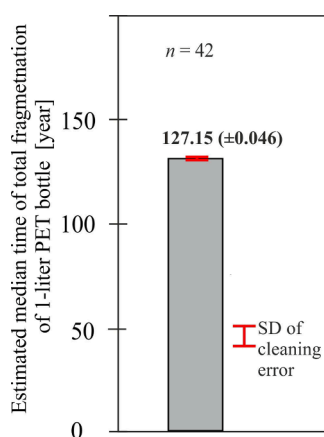


Fig. 6. Time of total fragmentation of 1-liter PET bottle estimated from the extrapolation of data obtained during experiment.

irradiation, a crucial factor in polymer degradation (Chamas et al., 2020; Andrady et al., 2022). Previous studies have shown that UV exposure can accelerate fragmentation by breaking down polymer chains, making the plastic more brittle and prone to future mechanical fragmentation (Chamas et al., 2020; Andrady et al., 2022; Liro et al., 2023b). To quantify the importance of UV-induced degradation in macroplastic fragmentation in river channel, our methodology could be extended to include exposing control bottles solely to UV and air, and comparing their mass loss with bottles subjected to river transport. This approach will be especially important for future long-term experiments, allowing for measurable mass loss due to UV-induced fragmentation.

Regardless of the tracking method used, short-term experiments may still be useful for quantifying the mechanical fragmentation of macroplastics occurring during floods, which have previously been suggested as key factors enhancing macroplastic fragmentation in rivers (Liro et al., 2023a, b). Such experiments can also allow for comparing the rates of mechanical fragmentation of macroplastics along river reaches with different hydromorphological characteristics (e.g., channelized vs. unregulated) or between different river types (e.g., lowland vs. mountain rivers). Previous works (Liro et al., 2023a,b) suggest that mountain streams, with high-energy water and sediment transport (see e.g., Wohl, 2010; Hauer et al., 2016), differ substantially from lower-energy lowland rivers and lakes. In mountain rivers, mechanical interactions should be the key factor in the fragmentation of macroplastic (Liro et al., 2023a), while only macroplastics transported on the water surface through flotation may also undergo fragmentation resulting from exposure to UV irradiation (Liro et al., 2023b).

It seems that the proposed methodology can be useful not only for recording differences in macroplastic fragmentation between different types of rivers and their smaller-scale compartments (Liro et al., 2023b), but also among other environments on Earth where macroplastic transport and its mechanical interactions with water and sediments occur, such as seas or beaches (Corcoran et al., 2009). Considering the general lack of direct field measurements of macroplastic fragmentation resulting from its transport in different environments (Hurley et al., 2020; Maga et al., 2022), the proposed low-cost experimental design, which utilizes a simple comparison of macroplastic masses that is easy to repeat in various environments, can be viewed as a promising tool to provide standardized baseline information on this process globally.

Despite our experiment being conducted during low and medium flow conditions, which are generally suggested to be less effective for the mechanical fragmentation of macroplastic compared to high flows (Liro et al., 2023a), the results indicate that macroplastic is effectively fragmented in the studied mountain river channels, with a median fragmentation time for 1-liter PET bottles estimated at 127.15 ± 0.046 years. However, this estimate is substantially underestimated due to the simplified assumption that the fragmentation rate remains constant throughout the entire lifespan of the bottle. Previous works have suggested that over time, the increasing degree of macroplastic degradation and the increasing surface area of fragmented macroplastic will accelerate the rate of fragmentation (see Fig. 2 in Liro et al., 2023b). Future longer-term experiments, including observations during flood events, are necessary to further elucidate our findings. However, even considering the potential underestimation, this value exceeds those commonly estimated for PET bottles (~ 500 years) in other environments (Chamas et al., 2020). This supports our previous hypotheses that mountain river channels can serve as hotspots for the mechanical fragmentation of macroplastics being transported through them (see Liro et al., 2023a, b).

Despite the mass losses observed in our experimental bottles (Fig. 4), macroscopic features observed on their surfaces after the experiment indicate intensive mechanical interactions with objects in the river channel (Fig. 3D). For future research, a more detailed analysis of such surface cracks formed during macroplastic transport could be valuable (for methods, see e.g., Delorme et al., 2021). Our results suggest a lack of correlation between travel distance (ranging from 0.37 km to 16.27 km) and mass loss during bottle transport ($R^2 = 0.004$; $p = 0.56$) (Fig. S1), indicating that the amount of mechanical interaction experienced by a given bottle cannot be solely explained by its travel distance. This likely reflects the high diversity of mountain river hydromorphology and the resulting complexity of transport patches within which a given bottle can be transported along the same reach of the mountain river channel.

Field observations conducted during the initiation of the experiment revealed, for example, that some bottles were intensely rotating in the

same place of the river channel due to hydraulic jumps formed behind physical obstacles such as boulders. The occurrence of such phenomena, especially in the shallower parts of the river channel where rotating bottles can interact with riverbed elements, can explain why some bottles may become relatively highly fragmented without undergoing distant transport, even under the low-energy conditions occurring during the experiment (low and medium flow) (Fig. 2C).

Our experiment did not utilize trackers capable of measuring the details of macroplastic transport. However, future experiments employing GPS trackers integrated with accelerometers could explore this phenomenon further by applying our methodology to correlate the mass loss of riverine macroplastics not only with their travel distance but also with other characteristics of the transport process (e.g., time, residence time in a given hydromorphological unit, number of bottle rotations).

Based on the obtained data, we estimated a yearly mass loss of plastic bottles ranging from 0.07 % to 3 %, with a median of 0.78 % (± 0.036 %) (Fig. 5B), which is lower than those measured previously in a mesocosm experiment (4.9 % mass loss/year for PET bottles), which reported a moderate rate of fragmentation for PET bottles compared to other plastic litter made from PS, PP, and PE (<1%) and compostable polymers like PLA (7–27 %) (Gerritse et al., 2020). However, the plastic bottles in their experiment were not sealed, as they were in ours, which can increase the availability of the bottle surface area for fragmentation. Our estimation of yearly fragmentation may also be substantially underestimated because the data were collected during low-flow conditions with no high-energy flows (which favor mechanical fragmentation). It is also important for future works to directly quantify the rate of riverine macroplastic fragmentations for litter made from PE, PP, and PS polymers, which were reported by Schwarz et al. (2019) to constitute 92.2 % to 95.8 % of the total plastic litter in freshwater environments.

Considering the general lack of direct experimental data on fragmentation rates of different polymer types in rivers, and the limited relevance of existing laboratory experiments to ambient river conditions (for review, see Liro et al., 2023b), future studies could use our methodology to compare the fragmentation rates of similarly shaped objects made from different polymers commonly found in rivers under similar conditions (e.g., within one river reach). This approach would provide a baseline understanding of fragmentation rates for common macroplastic waste items in rivers, informing waste management policies and plastic product designers on how to reduce the risk of secondary microplastic production in rivers.

Finally, the information obtained through the proposed experimental methodology in different types of rivers could be valuable for calibrating future flume, mesocosms, and numerical models aimed at simulating riverine macroplastic fragmentation.

5. Conclusion

We report the initial test of a low-cost methodology that uses a simple comparison of macroplastic object mass loss to quantify macroplastic fragmentation during its transport in river channels. We applied this method to 1-liter PET bottles transported in a mountain river, detecting measurable fragmentation rates during a short-term experiment (52–65 days) under low- to medium-flow conditions. These results suggest that tracking the mass loss of macroplastic objects during their transport in rivers can serve as a low-cost and easy-to-implement approach for providing valuable information on macroplastic fragmentation in various types of rivers.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgments

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Author contributions

ML conceptualisation, methodology, planning of field experiment design, fieldwork and laboratory analysis, writing the original draft, and creating original figures with the input from AZ and PM; AZ literature review, fieldwork, data analysis and manuscript writing; PM fieldwork, manuscript writing and figures preparation.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2024.108935>.

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