Experimental method for quantifying macroplastic fragmentation in rivers

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Abstract: Direct field measurements of macroplastic fragmentation during its transport in rivers are unavailable, and there is no method to perform such measurements. Recent theoretical works have hypothesised that river channels may be hotspots of macroplastic fragmentation. Here, we propose a methodology for quantifying riverine macroplastic fragmentation by conducting repeated measurements of tagged macroplastic items' mass before and after their transport in the river. A 52-65-day experimental test of the proposed methodology allowed us to provide the first quantification of fragmentation of 1-liter PET bottles during their transport in a mountain river channel. We calculated the mass loss of tracked bottles (*n*=43), ranging from 0.025 grams/year (0.07%/year) to 1.0 gram/year (3%/year), with a median of 0.26±0.04 grams/year (0.78%/year), and the rate of bottle surface degradation, ranging from -0.29 μm/year to -11.88 μm/year (median = 3.77±0.43 μm/year). These results suggest their complete fragmentation under conditions represented by our experiment (low to medium flow) within a timescale ranging from 33.63 years to 332.81 years (median = 128.92±31.07 years). Our methodology can be flexibly adapted to quantify macroplastic fragmentation in various types of rivers and other surface environments.

Key words: field experiment, secondary microplastic, plastic breakdown, plastic fragments, mountain river

Introduction

Tracking rates of macroplastic fragmentation in various environmental compartments is fundamentally important for evaluating the risk of plastic pollution, because it provides direct insights into the amount of secondary microplastics released within these compartments[1]. Field-based information on the rates of macroplastic fragmentation in different environments is, however, very limited[1,2,3] especially for rivers[4,5]. Recent works have, however, hypothesised that river channels can operate as hot-spots of macroplastic fragmentation because of constant movement of water and sediments in this zone which can favour mechanical interactions of macroplastic with water, sediments, and riverbeds[5,6]. 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[7,8]. 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 understanding the extent to which they can be transported downstream to lowland rivers and oceans[5,8]. Information on this process can be particularly useful for assessment of future risk of secondary microplastic release (during macroplastic fragmentation) for mountain river biodiversity[9,10] and quality of resources they provide for human populations (e.g., water resources[11]).

Here, we propose a simple field-experiment based methodology for quantifying macroplastic fragmentation rates during its transport in river channels. Our methodology implements mass loss quantification of macroplastic objects, previously utilized in laboratory experiments[12], to tagged macroplastic objects transported in river channel (Fig. 1). Using this methodology, we have quantified, for the first time, 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.

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[12], with macroplastic tracking techniques previously used to quantify the travel distance of tagged macroplastic objects transported in river channel[13,14]. The proposed workflow consists of four steps: (1) measurement of the masses of virgin macroplastic objects, (2) transport of tagged items in the river, (3) repeated measurements of macroplastic object masses, and (4) calculation of object mass loss over time resulting from their transport. The primary advantage of using mass loss as a proxy for macroplastic fragmentation in rivers, compared to other laboratory techniques previously used for quantifying macroplastic degradation and fragmentation[2], is its low cost and minimal need for laboratory analysis. Below, we describe how we applied this four-step procedure to quantify the fragmentation rate of 1-litre PET bottles transported in the Skawa River in the Polish Carpathians (Fig. 2).

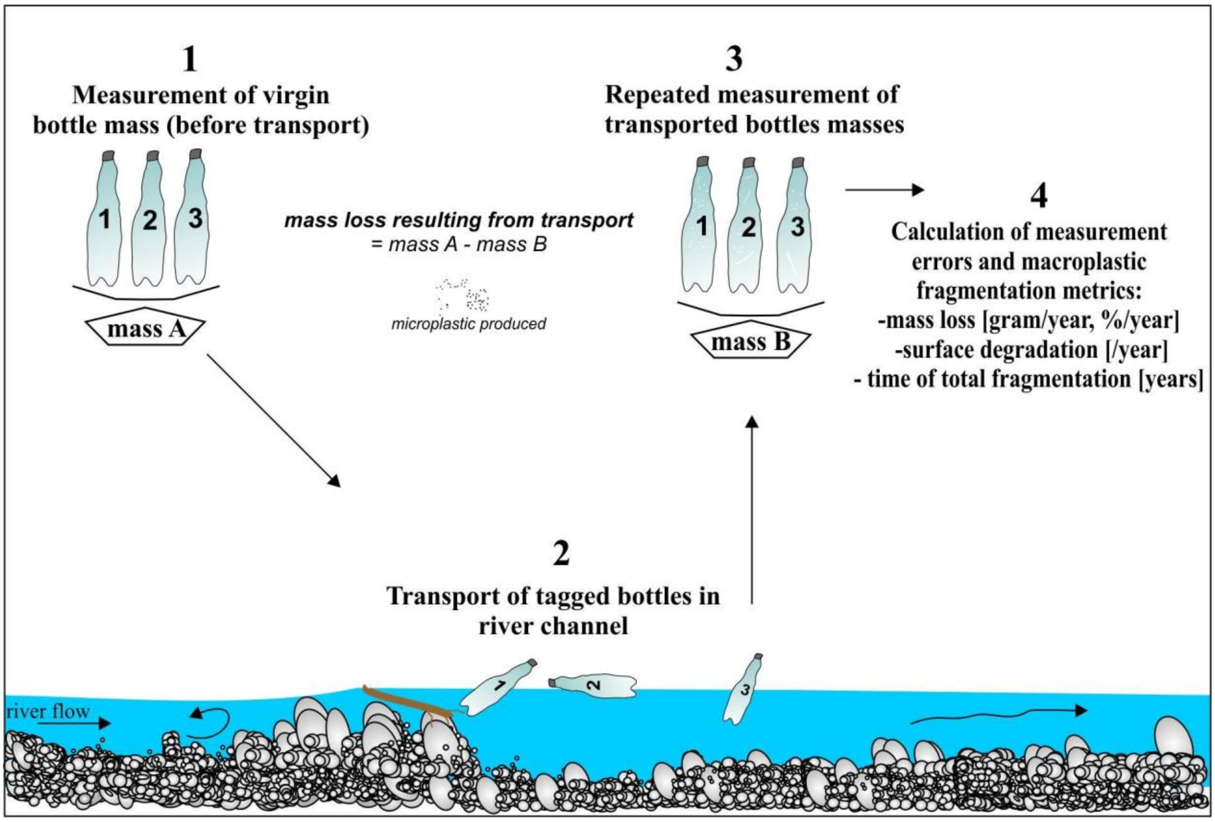

Figure 1. Proposed experimental workflow.

Measurement of the masses of virgin macroplastic objects

Measurement of macroplastic mass loss as a proxy of its degradation and fragmentation have primarily been employed in laboratory experiments aimed at determining effects of UV radiation, water movement, and biofilm formation on these processes[12]. In our experiment, we utilised 177 (n=177) virgin 1-litre bottles made from polyethylene terephthalate (PET). Initially, the mass of each bottle was determined (as the mean of three replicate measurements) using a precise laboratory balance with an accuracy of 0.001g. Subsequently, the bottles were tagged with numbers drawn on the bottle caps and on the foil tag placed inside them (Fig. 3A). Depending on the planned experiment budget, the size of the rivers, the planned duration of the experiment, and the hypotheses being tested, various tracking techniques can also be considered for future works, including GPS, RFID, radio transmitters, and printed items.)[13,14].

Transport of tagged items in the river

Field experiment was performed in Skawa river, right-bank tributary of Vistula (largest river in Poland). It flows at 700 m a.s.l. and its length is over 96 km. The width of the river in the study section ranges from 5 to 40 metres. 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. 2A) 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 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 were transported to the laboratory for cleaning and to measure their mass loss resulting from mechanical fragmentation during their transport in the river channel.

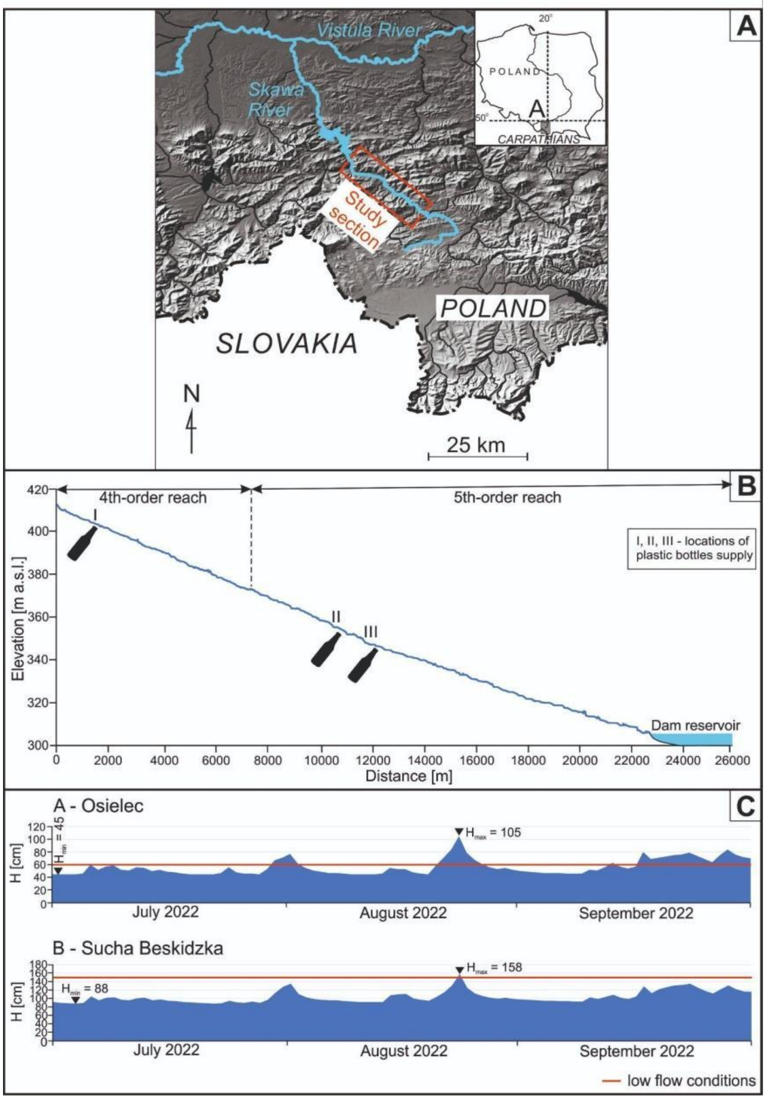

Figure 2. A - Location of the study area; B - Longitudinal profile of the surveyed river section with bottle delivery points marked; C - Hydrograph of water levels for the gauge stations in Osielec village and Sucha Beskidzka city occuring during the experiment.

Repeated measurements of macroplastic object masses

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. Before measuring the bottle's mass after their transport in the river, we employed a cleaning procedure similar to that used by Gerritse et al.[12](Fig. 3B). Initially, the bottles were cleaned with tap water and detergent, followed by a 12-hour incubation period in 30% H2O2 to eliminate biofilms and other organic matter from their surfaces. Then, 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 of them was determined in grams.

We accounted for the possibility of the cleaning procedure itself causing a small-scale mass loss, which could potentially overestimate the final results. To assess this error, we conducted a test cleaning on 24 reference bottles, measuring their masses before and after the procedure. The mean value 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), was found to be 0.021 g.

Subsequently, the mass loss values determined for the bottles transported in rivers (n=43) were corrected using the mean value of bottle mass loss occurring during the cleaning procedure (0.021 g) *(1)*.

$$\textbf{\textit{macroplastic mass loss}}_{\textbf{\textit{transport}}} = (mass_{before\ transport} - mass_{after\ transport}) - mass\ loss_{cleaning\ procedure} \quad (1)$$

Utilising the corrected mass loss values for the 43 bottles obtained during the 52-65-day experiment, we calculated the yearly mass loss expressed in grams and as a percentage of the initial bottle mass. Additionally, we determined the rate of bottle surface degradation resulting from the calculated mass losses. For this calculation, we used the density of PET plastic (1.38 g/cm^3), and we assumed that bottle fragmentation occurs evenly across their entire external surface (~610 cm^2).

Figure 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.

Results

The mass loss of the tracked 1-litre PET bottles (*n*=43) during the 52-65-day transport in the river channel ranged from 0.004 g to 0.153 g, with a median value of 0.044 ± 0.006 g (Fig. 4).

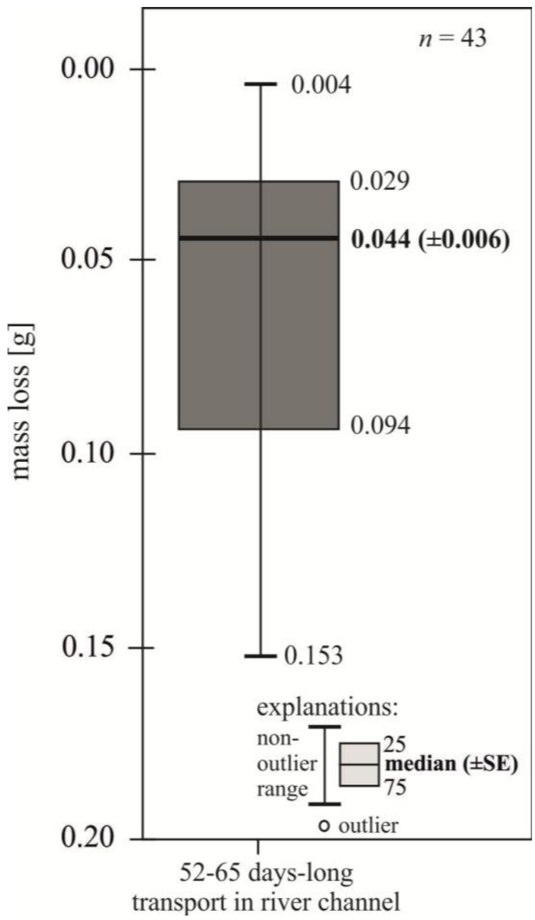

Figure 4. Mass loss of 1-litre PET bottles occurring during 50-65 days of field experiment.

Using obtained data on bottle mass losses, we extrapolated yearly mass loss from 0.025 g to 1g with median 0.26±0.04 g, which constitute respectively from 0.7% to 3% (median = 0.78±0.11 %) of their initial masses (Fig. 4) and surface degradation rates from 1.95 to 11.88 μm/year (median = 3.08±0.43 ±0.11 μm/year) (Fig. 5). Based on these values, we calculated that complete fragmentation of a used 1-litre 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 128.92 ± 31.07 years. (Fig. 6).

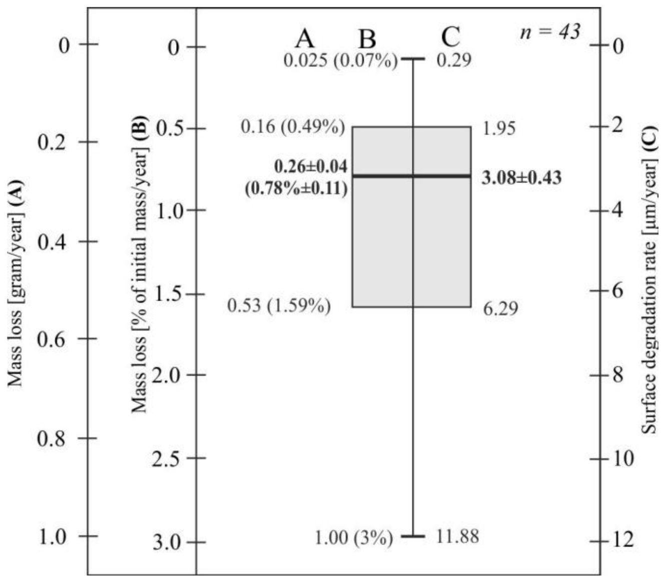

Figure 5. Yearly mass loss (gram/year) (A) and surface degradation rate (µm/year) (B) of 1-litre PET bottles estimated based on the experiment results.

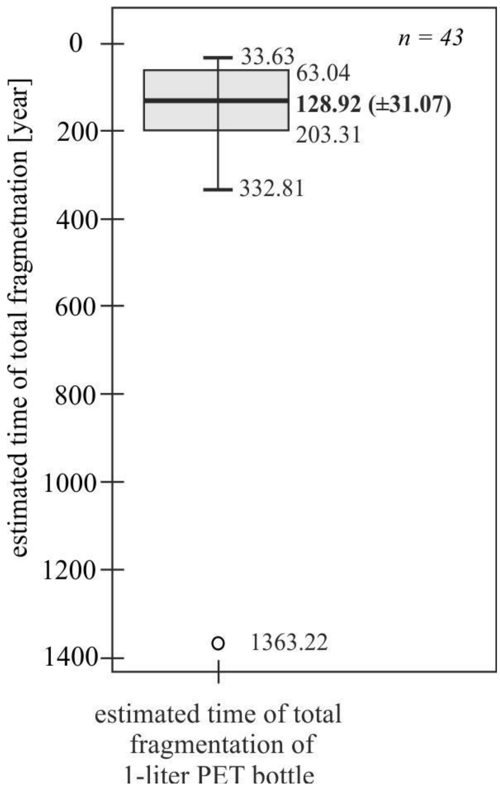

Figure 6. Time of total fragmentation of 1-litre PET bottle estimated from the extrapolation of data obtained during experiment.

Discussion and future outlook

We have implemented the measurements of macroplastic mass loss, previously utilised in laboratory experiments on macroplastic fragmentation[12], to quantify the rate of this process occurring during short-term transport of macroplastic in a mountain river channel. This method allowed us to quantify the fragmentation rates of 1-litre PET bottles (expressed as their mass loss) higher than 0.021g (error introduced by the cleaning procedure). Despite the relatively short duration of the experiment and the absence of higher flows, this relatively small error enabled us to measure the amount of fragmentation effectively. This suggests that the proposed method can provide baseline information on the rates of mechanical fragmentation of macroplastics even with a relatively short experimental period. However, we highlight that longer-term experiments are necessary to verify and detail these values, as they may be substantially underestimated due to the short duration of our experiment (52-65 days) and the absence of floods during its duration.

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)[13,14] 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 (especially during longer-term experiments).

Regardless of the tracking method used, short-term experiments may be still useful for quantifying macroplastic fragmentation occurring during floods, which are previously suggested as factors enhancing mechanical fragmentation of macroplastic in rivers[5,8]. It seems that the proposed methodology can be useful not only for recording macroplastic fragmentation in other river types[8] but also in other surface environments on Earth where macroplastic transport and its mechanical interactions with water and sediments occur, such as seas or beaches[15]. Considering the general lack of direct field measurements of macroplastic fragmentation in different environments[1,3], the proposed experimental design (repeated in various environments using the same macroplastic object) can be viewed as a promising tool to provide standardised 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 fragmentation compared to high flows[5], the results indicate that macroplastic is effectively fragmented in mountain river channels (with a median time of 1-litre PET bottle fragmentation estimated at 128.92 ± 31.07 years). 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 typically estimated for PET bottles (~500 years) in other environments[2]. This provides support for our previous hypotheses that mountain river channels can serve as hotspots for mechanical fragmentation of macroplastics being transported through them[5,8].

Despite the mass losses observed in the 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.,[4]). 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), 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, wherein 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 low-energy conditions occurring during the experiment (low and medium flow) (Fig. 2C).

Our experiment did not utilise 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).

Finally, the information obtained through the proposed experimental methodology in different types of rivers could be valuable for calibrating future physical (e.g., flumes[cf.16], mesocosms[12]) and numerical[17] models aimed at simulating riverine macroplastic fragmentation.

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Author contributions
ML conceptualisation, methodology, planning of field experiment design, fieldworks 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.

References
1. Maga, D. *et al*. Methodology to address potential impacts of plastic emissions in life cycle assessment. *Int. J. Life Cycle Assess*. **27**, 469–491 (2022).
2. Chamas, A. *et al*. Degradation rates of plastics in the environment. ACS Sustain. *Chem. Eng.* **8**(9), 3494–3511 (2020).
3. Hurley, R., Horton, A., Lusher, A. & Nizzetto, L. Plastic waste in the terrestrial environment in *Plastic Waste and Recycling* (ed. Letcher, T. M.) 163–193 (Academic Press, 2020).
4. Delorme, A. E. *et al*. The life of a plastic butter tub in riverine environments. *Environ. Pollut*. **287**, 117656 (2021).
5. Liro M. *et al.* The unknown fate of macroplastic in mountain rivers. *Sci. Total. Environ*. **865**, 161224. (2023).
6. Liro, M., van Emmerik, T. H. M., Wyżga, B., Liro, J. & Mikuś, P. Macroplastic storage and remobilization in rivers. *Water* **12**, 2055 (2020).
7. Honorato-Zimmer, D. *et al.* Mountain streams flushing litter to the sea – Andean rivers as conduits for plastic pollution. *Environ. Pollut*. **291**, 118166 (2021).
8. Liro, M., Zielonka, A. & van Emmerik, T. H. M. Macroplastic fragmentation in rivers. *Environ. Int*. **180**, 108186 (2023).
9. Wohl, E. (ed). Mountain Rivers Revisited. *Water Resour. Monogr*. (American Geophysical Union, 2010).
10. Hauer, F. R. *et al.* Gravel-bed river floodplains are the ecological nexus of glaciated mountain landscapes. *Sci. Adv*. **2**, e1600026 (2016).
11. Viviroli, D., Kummu, M., Meybeck, M., Kallio, M. & Wada, Y. Increasing dependence of lowland populations on mountain water resources. *Nat. Sustain*. **3**, 917–928 (2020).
12. Gerritse, J., Leslie, H. A., de Tender, C. A., Devriese, L. I. & Vethaak, A. D. Fragmentation of plastic objects in a laboratory seawater microcosm. *Sci. Rep*. **10**, 10945 (2020).
13. Duncan, E. M., *et al.* Message in a bottle: open source technology to track the movement of plastic pollution. *PLoS ONE* **15**, e0242459 (2020).
14. Newbould, R. A., Powell, D. M. & Whelan, M. J. Macroplastic debris transfer in rivers: a travel distance approach. *Front. Water* **3**, 724596 (2021).

15. Corcoran, P. L., Biesinger, M. C. & Grifi, M. Plastics and beaches: a degrading relationship. *Mar. Pollut. Bull*. **58**, 80–84 (2009).
16. Valero, D., Belay, B.S., Moreno-Rodenas, A., Kramer, M. & Franca, M.J. The key role of surface tension in the transport and quantification of plastic pollution in rivers. *Water Res*. **226**, 119078 (2022)
17. Wyżga, B., Mikuś, P., Zawiejska, J., Ruiz-Villanueva, V., Kaczka, R.J., Czech, W.. Log transport and deposition in incised, channelized, and multithread reaches of a wide mountain river: tracking experiment during a 20-year flood. *Geomorphology* **279,** 98-111 (2017)