#### Short communication

First attempt to measure macroplastic fragmentation in rivers

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1	First Attempt to Measure Macroplastic Fragmentation in Rivers
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Abstract: Direct field measurements of macroplastic fragmentation during its transport in 11 rivers are currently unavailable, and there is no established method to perform such 12 measurements. Previous studies have suggested that macroplastic fragmentation results in the 13 production of harmful microplastics, and river channels can be hotspots for this process. 14 Therefore, obtaining information about this process is crucial for quantifying the production 15 of secondary microplastics in rivers and assessing the related risks for riverine biota and 16 human health. Here, we propose a simple low-cost methodology for quantifying riverine 17 macroplastic fragmentation by conducting repeated measurements of the mass of tagged 18 macroplastic items before and after their transport in the river. As a proof-of-concept for this 19 method, we conducted a 52-65 day experiment that allowed us to measure a median 20 fragmentation rate of  $0.044 \pm 0.012$ g for 1-liter PET bottles during their transport at low to 21 medium flow in a 20-km-long section of the middle mountain Skawa River in the Polish 22 Carpathians. Using the obtained data (n=42), we extrapolated that during low to medium 23 flows, the median yearly mass loss of PET bottles in the study section is  $0.26 \pm 0.012$  g/year 24  $(0.78 \pm 0.036 \%$  of bottle mass), and the median rate of bottle surface degradation is  $3.13 \pm$ 25 26 0.14 µm/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. 27 We discuss how our simple and relatively low-cost methodology can be flexibly adapted and 28 future optimized to quantify macroplastic fragmentation in various types of rivers and their 29 30 compartments, informing future mitigation efforts about the formation and dispersion of secondary microplastics. 31

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

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#### 36 Introduction

37 Tracking rates of macroplastic fragmentation in various environmental compartments is fundamentally important for evaluating the risk of plastic pollution for biota and human 38 health. This provides direct insights into the amount of secondary microplastics (Maga et al., 39 2022) as well as their harmful additives and contaminants that can be released within these 40 compartments, ultimately harming living organisms and, through the food chain, affecting 41 human health (Hahladakis et al., 2018; Karlsson et al., 2021; Chen et al., 2022; Behnisch et 42 al., 2023; Wagner et al., 2024). Field-based information on the rates of macroplastic 43 fragmentation in different environments is, however, very limited (Chamas et al., 2020; 44 Hurley et al., 2020; Maga et al., 2022) especially for rivers (Liro et al., 2020, 2023a,b; 45 Delorme et al., 2021; Honorato-Zimmer et al., 2021). Recent works have, however, 46 hypothesized that river channels can operate as hot-spots of macroplastic fragmentation 47 because of constant movement of water and sediments in this zone which can favor 48 mechanical interactions of macroplastic with water, sediments, and riverbeds (Liro et al., 49 2023a,b). The intensity of these interactions can be particularly high in the case of mountain 50 river channels, where high-energy water and sediment transport coincide with the presence of 51 numerous physical obstacles such as boulders, bedrock, and large wood within the river 52 53 channel (Liro et al., 2023a). Field experiments exploring this process have not yet been conducted. However, obtaining direct information about the rate of macroplastic 54 fragmentation in mountain rivers is crucial for quantifying the production of secondary 55 microplastics in these environments and evaluation of related risks to their biodiversity 56 57 (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 58 59 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., 60 61 2023a,b).

Here, we propose a simple field-experiment based methodology for quantifying 62 63 macroplastic fragmentation rates during its transport in river channels. Our methodology 64 implements mass loss quantification of macroplastic objects, previously utilized in laboratory experiment (Gerritse et al., 2020) to tagged macroplastic objects transported in river channel 65 66 (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 67 to 16.27 km in a mountain river channel in the Polish Carpathians, under low- to medium-68 flow conditions (Fig. 2). The objective of this work is to present this methodology and report 69 the first insights into macroplastic fragmentation in mountain rivers obtained through its 70 application. 71

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### 73 Materials and methods

### 74 Proposed methodology for quantify riverine macroplastic fragmentation

Our methodology combines mass loss quantification of macroplastic objects, previously 75 utilized in laboratory experiments for determining macroplastic fragmentation (Gerritse et al., 76 77 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., 2020). 78 The proposed workflow consists of six steps: (1) measurement of the masses of virgin 79 80 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 81 measured mass losses by accounting for cleaning error, and (6) calculation of macroplastic 82

- 83 fragmentation metrics from the obtained mass loss information (see Fig. 1). The primary
- 84 advantage of using mass loss as a proxy for macroplastic fragmentation in rivers, compared to
- 85 other techniques previously used for quantifying macroplastic degradation and fragmentation
- 86 (cf. Chamas et al., 2020), is its low cost and minimal need for laboratory analysis. Below, we
- 87 describe how we applied this six-step procedure to quantify the fragmentation rate of 1-liter
- 88 PET bottles transported in the Skawa River in the Polish Carpathians (Fig. 2).



Figure 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.

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# 94 *Measurement of the masses of virgin macroplastic objects (step 1)*

95 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 96 UV radiation, water movement, and biofilm formation on these processes (Gerritse et al., 97 2020). In our experiment, we utilized 177 (n=177) virgin 1-liter bottles made from 98 polyethylene terephthalate (PET). Initially, the mass of each bottle was determined (as the 99 mean of triplicate measurements) (Table S2) using a precise laboratory balance with an 100 accuracy of 0.001g. Subsequently, the bottles were tagged with numbers drawn on the bottle 101 caps and on a foil tag placed inside them (Fig. 3A). 102

# 103 Transport of tagged items in the river (step 2)

The field experiment was performed in the Skawa River (Polish Carpathians), a rightbank 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 meters 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 km<sup>2</sup> and the average annual flow is 11 m<sup>3</sup>/s. The 109 riverbed is predominantly composed of gravel and cobbles, with some sections of bedrock 110 present in the middle course of the study section. All bottles were sealed with caps (Fig. 3A) 111 and deployed into the river channel at three locations along the Skawa River in the Polish 112 Carpathians on July 11th, 2022 (Fig. 1A-B). These locations were chosen along the 20 km-113 long study reach of the river, spanning from Osielec Village (location 1) to the Świnna Poreba 114 Dam Reservoir (as depicted in Fig. 2B). After 52 days (September 1st), 57 days (September 115 6th), and 65 days (September 14th), the study reaches were surveyed by four persons (two on 116 each river bank), enabling them to collect 43 of the previously deployed tagged bottles (as 117 shown in Fig. 2A-C). The travel distances for each bottle were calculated as the thalweg 118 distance between the point of bottle deployment and the location where the bottle was 119 collected along the study reach (measured using an RTK GPS receiver). Subsequently, the 120 121 collected bottles (Fig. 3B-C) were transported to the laboratory for cleaning and measurement 122 of their mass loss resulting from mechanical fragmentation during their transport in the river 123 channel.



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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 the Osielec village and Sucha Beskidzka city occurring during the experiment.

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# 129 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 139 140 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 141 overestimation of the final results. To quantify this error and correct the final values of mass 142 loss, we conducted a test cleaning on 24 reference bottles, measuring their masses before and 143 after the cleaning procedure. The values of bottle mass loss during cleaning, determined from 144 the mass loss of the 24 reference bottles (one bottle was excluded due to contamination during 145 cleaning), were normally distributed, and their mean was found to be 0.022 g (Table S1). The 146 uncertainty of this value estimation was quantified as the standard deviation of the mean 147 (±0.012 g). 148

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150 *Repeated measurements of macroplastic object masses (step 4) and cleaning error* 151 *correction (step 5)* 

The mass loss of macroplastic objects resulting from their transport in rivers was 152 determined by conducting repeated measurements of the dry macroplastic masses before and 153 after their transport. The mass loss values determined for the bottles transported in rivers 154 (n=43) were not normally distributed. These values were then corrected using the mean value 155 of bottle mass loss occurring during the cleaning procedure (0.022 g) and presented with the 156 uncertainty of the cleaning error estimation ( $\pm 0.012$  g) (Table S1-2). One of the corrected 157 measurements of mass loss, which was lower (0.004 g) than the calculated cleaning error 158 uncertainty ( $\pm 0.012$  g), was removed from further analysis, and 42 measurements (n=42) were 159 further analyzed. 160

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162 *macroplastic mass loss<sub>transport</sub> = (mass<sub>before transport</sub>-mass<sub>after transport</sub>)-mass loss<sub>cleaning procedure</sub>*±uncertainty of cleaning error estimation (1)

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164 *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 (Figs. 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<sup>3</sup>) and assumed that bottle fragmentation occurs evenly across their entire external surface (~610 cm<sup>2</sup>) (Fig. 5C).



- 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.
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171

- 176 **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  $\pm 0.012$ g (Fig. 4).



Figure 4. Mass loss of 1-litre PET bottles occurring during 52-65 days of low to mediumflow conditions.

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Using the obtained data on bottle mass losses, we extrapolated the yearly mass loss to 184 range from 0.098 g to 1 g (Fig. S2) with a median of  $0.26 \pm 0.012$  g (Fig. 5A), which 185 constitutes 0.29% to 2.97% (median =  $0.78 \pm 0.036$ %) of their initial masses (Fig. 5B) and 186 surface degradation rates from 1.19 to 11.82  $\mu$ m/year (median = 3.13  $\pm$  0.144  $\mu$ m/year) (Fig. 187 5C). Based on these values, we estimated that the complete fragmentation of a used 1-liter 188 PET bottle under the conditions represented by our experiment (low to medium flows) would 189 take between 33.63 and 332.81 years, with a median estimate of  $127.15 \pm 0.046$  years (Fig. 190 191 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.







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

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### 203 Discussion and future outlook

We have implemented the measurement of macroplastic mass loss, previously utilized in 204 laboratory experiments on macroplastic fragmentation (Gerritse et al., 2020), to quantify the 205 rate of this process occurring during short-term transport of macroplastic in a middle 206 mountain river channel. This method allowed us to quantify the fragmentation rates of 1-liter 207 208 PET bottles (expressed as their mass loss) with an uncertainty of  $\pm 0.012$  g (SD of cleaning procedure error). The uncertainty resulting from the applied cleaning procedure constitutes 209 26.9% of the median mass loss measured during the 52-65 day experiment. This relatively 210 high proportion of measurement uncertainty  $(\pm 0.012 \text{ g})$  to the recorded mass loss change 211 during the experiment (median = 0.045 g) can be explained by the short duration of the 212 experiment, the absence of higher flows during its course, and the simple technique used for 213 bottle cleaning. The uncertainty in the measurable value of mass loss using the proposed 214 method could potentially be reduced in future experiments by applying our method to longer-215 term studies (e.g., one year). This would account for occurrences of higher mass losses even 216 during low-energy conditions in rivers or by conducting experiments during higher-energy 217 conditions (e.g., floods) or in rivers with higher-energy hydromorphologies (e.g., high-218 mountain streams). Future studies should also optimize cleaning procedures to reduce the 219 value (0.022g) and uncertainty ( $\pm 0.012$  g) of cleaning errors. This will allow for the 220 measurement of smaller magnitude mass losses during shorter-term experiments. Despite the 221 aforementioned need for future improvements, the proposed method using mass loss as a 222 proxy for macroplastic fragmentation offers a promising, cost-effective tool for collecting 223 baseline information on the rates of macroplastic fragmentation in rivers and other 224 environments. 225

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, longerterm experiments, it is essential to use appropriate tracking techniques (e.g., GPS, RFID, radio transmitters) (see e.g., Duncan et al., 2020), 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, 233 234 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 235 to UV irradiation, a crucial factor in polymer degradation (Chamas et al., 2020; Andrady et 236 al., 2022). Previous studies have shown that UV exposure can accelerate fragmentation by 237 breaking down polymer chains, making the plastic more brittle and prone to future 238 mechanical fragmentation (Chamas et al., 2020; Andrady et al., 2022; Liro et al., 2023b). To 239 240 quantify the importance of UV-induced degradation in macroplastic fragmentation, our methodology could be extended to include exposing control bottles solely to UV and air, and 241 comparing their mass loss with bottles subjected to both UV exposure and river transport. 242 This approach will be especially important for future long-term experiments, allowing for 243 measurable mass loss due to UV-induced fragmentation. 244

Regardless of the tracking method used, short-term experiments may still be useful for 245 quantifying the mechanical fragmentation of macroplastics occurring during floods, which 246 have previously been suggested as factors enhancing macroplastic fragmentation in rivers 247 (Liro et al., 2023a, b). Such experiments can also allow for comparing the rates of mechanical 248 fragmentation of macroplastics along river reaches with different hydromorphological 249 250 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 251 streams, with high-energy water and sediment transport (Wohl, 2010; Hauer et al., 2016), 252 differ substantially from lower-energy lowland rivers and lakes. In mountain rivers, 253 mechanical interactions should be the key factor in the fragmentation of macroplastic (Liro et 254 al., 2023a), whereas macroplastics transported on the water surface through flotation may also 255 undergo fragmentation resulting from exposure to UV irradiation (cf. Andrady et al., 2022). 256

It seems that the proposed methodology can be useful not only for recording differences 257 in macroplastic fragmentation between different types of rivers and their smaller-scale 258 compartments (Liro et al., 2023b), but also among other environments on Earth where 259 macroplastic transport and its mechanical interactions with water and sediments occur, such 260 as seas or beaches (Corcoran et al., 2009). Considering the general lack of direct field 261 measurements of macroplastic fragmentation resulting from its transport in different 262 environments (Hurley et al., 2020; Maga et al., 2022), the proposed low-cost experimental 263 design, which utilizes a simple comparison of macroplastic masses that is easy to repeat in 264 various environments, can be viewed as a promising tool to provide standardized baseline 265 information on this process globally. 266

Despite our experiment being conducted during low and medium flow conditions, which 267 are generally suggested to be less effective for the mechanical fragmentation of macroplastic 268 compared to high flows (Liro et al., 2023a), the results indicate that macroplastic is 269 effectively fragmented in mountain river channels, with a median fragmentation time for 1-270 liter PET bottles estimated at  $127.15 \pm 0.046$  years. However, this estimate is substantially 271 underestimated due to the simplified assumption that the fragmentation rate remains constant 272 throughout the entire lifespan of the bottle. Previous works have suggested that over time, the 273 increasing degree of macroplastic degradation and the increasing surface area of fragmented 274 275 macroplastic will accelerate the rate of fragmentation (see Figure 2 in Liro et al., 2023b). 276 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 282 features observed on their surfaces after the experiment indicate intensive mechanical 283 interactions with objects in the river channel (Fig. 3D). For future research, a more detailed 284 analysis of such surface cracks formed during macroplastic transport could be valuable (for 285 methods, see e.g., Delorme et al., 2021). Our results suggest a lack of correlation between 286 287 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 288 289 experienced by a given bottle cannot be solely explained by its travel distance. This likely 290 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 291 mountain river channel. 292

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 306 from 0.07% to 3%, with a median of 0.78% ( $\pm 0.036\%$ ) (Fig. 5B), which is lower than those 307 measured previously in a mesocosm experiment (4.9% mass loss/year for PET bottles), which 308 reported a moderate rate of fragmentation for PET bottles compared to other plastic litter 309 made from PS, PP, and PE (<1%) and compostable polymers like PLA (7–27%) (Gerritse et 310 al., 2020). However, the plastic bottles in their experiment were not sealed, as they were in 311 ours, which can increase the availability of the bottle surface area for fragmentation. Our 312 estimation of yearly fragmentation may also be substantially underestimated because the data 313 were collected during low-flow conditions with no high-energy flows (which favor 314 mechanical fragmentation). It is also important for future works to directly quantify the rate of 315 riverine macroplastic fragmentations for litter made from PE, PP, and PS polymers, which 316 were reported by Schwarz et al. (2019) to constitute 92.2% to 95.8% of the total plastic litter 317 in freshwater environments. 318

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.

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# 331 Conclusion

We report the initial test of a low-cost methodology that uses a simple comparison of 332 macroplastic object mass loss to quantify macroplastic fragmentation during its transport in 333 river channels. We applied this method to 1-liter PET bottles transported in a mountain river, 334 335 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 336 macroplastic objects during their transport in rivers can serve as a low-cost and easy-to-337 338 implement approach for providing valuable information on macroplastic fragmentation in various types of rivers. 339

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344

# 345 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.

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