

# Article

# A Methodology for Measuring Microplastic Transport in Large or Medium Rivers

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Abstract: Plastic waste as a persistent contaminant of our environment is a matter of increasing concern due to the largely unknown long-term effects on biota. Although freshwater systems are known to be the transport paths of plastic debris to the ocean, most research has been focused on marine environments. In recent years, freshwater studies have advanced rapidly, but they rarely address the spatial distribution of plastic debris in the water column. A methodology for measuring microplastic transport at various depths that is applicable to medium and large rivers is needed. We present a new methodology offering the possibility of measuring microplastic transport at different depths of verticals that are distributed within a profile. The net-based device is robust and can be applied at high flow velocities and discharges. Nets with different sizes (41 µm, 250 µm, and 500 µm) are exposed in three different depths of the water column. The methodology was tested in the Austrian Danube River, showing a high heterogeneity of microplastic concentrations within one cross section. Due to turbulent mixing, the different densities of the polymers, aggregation, and the growth of biofilms, plastic transport cannot be limited to the surface layer of a river, and must be examined within the whole water column as for suspended sediments. These results imply that multipoint measurements are required for obtaining the spatial distribution of plastic concentration and are therefore a prerequisite for calculating the passing transport. The analysis of filtration efficiency and side-by-side measurements with different mesh sizes showed that 500 µm nets led to optimal results.

Keywords: microplastics; plastic pollution; freshwater; microplastic sampling

### 1. Introduction

Due to their excellent material properties, plastics started off their triumphant advance to wide popularity in the 1950s. Worldwide plastic production is still increasing rapidly, from 230 million tons in 2005 to 322 million tons in 2015 [1], which equals a growth rate of 29% in the last ten years. Hence, roughly 40 kg of plastics are produced for each human being every year. Moreover, the useful lifetime of plastics, especially for single-use packaging material which accounts for around 40% of the plastic material demand in Europe [1], is often relatively short. Their primary advantages of being waterproof and resistant also make plastics highly persistent in our environment. The longevity of plastic is

estimated to be hundreds to thousands of years, meaning that, apart from incineration, the vast majority of all plastic ever made is still present in the environment [2,3]. Although recycling rates are increasing in recent years (at a rate of 64% from 2006 to 2014 in the EU28+2) [1], some fraction escapes the controlled waste streams and ends up in our environment. Plastic debris is classified by size, ranging from megadebris (>100 mm), macrodebris (>20 mm), mesodebris (20–5 mm) to microdebris (<5 mm) [2]. There is no defined size for a particle categorized as "microplastic", but an upper limit of 5 mm is generally agreed upon in the literature [4,5]. Particles are present in terrestrial, freshwater, and marine environments (for example, [5]) but have also been observed in the remotest areas of the world [6]. Studies proved plastic particle appearance in deep-sea sediments [7] as well as trapped in Arctic Sea ice [8]. When exposed to sunlight, wind, and the chemical environment, the material alters and breaks down to debris of a few micrometres in size, which can easily be dispersed. Little is known about the biological effects, such as organism interaction with and ingestion of microplastics [9], but particle consumption has been widely observed in marine species [6], and particles may be retained in the gut or even cross the gut wall to be translocated within the body [10].

Although terrestrial environments and freshwaters are recognized as the origins and transport paths of plastics, the majority of research to date focuses on the marine environment [6]. However, studies in freshwater environments have been rapidly advancing over recent years. Horton et al. [6] gave a detailed overview of freshwater studies ranging from lakes (for example, [11,12]) to rivers [13–19] and river sediments (for example, [20,21]). A number of studies have been performed that addressed the transport of microplastics in riverine systems. Measurements were undertaken in recent years in tributaries of the Great Lakes [13], the Seine River [14,22], various rivers in Switzerland [15], the Rhine River [17], various river sites near Chicago [18], and the Danube River [16]. The researchers all used benthic nets [16] or surface trawls as first used by Carpenter et al. [23], described by Brown and Cheng [24], and proposed as a standard methodology for surface waters by Lippiat et al. [25]. For this methodology, a manta net is towed horizontally at the surface. The net is 60–100 cm in width, designed to capture material to a depth of 15–35 cm, and is deployed from the back or the side of a vessel for approximately 0.5 nautical miles at a speed of 1–3 knots, a duration of approximately 15 min [25]. A flowmeter is usually attached in the centre of the net orifice to allow the calculation of concentration (items\*m<sup>-3</sup>) based on the filtered water volume. Due to this sampling strategy, all of these studies only address the particles floating in the near-surface layer (upper 10-30 cm of the water column). Moore et al. [26] tried to address multiple depths by using different devices including a modified large Helley Smith sampler in concrete-lined creeks near Los Angeles. Also, Dris et al. [22] addressed different depths in one point in the center of the Seine River by coupling a plankton net (addressing fibres) with a propeller-type current meter to sample down to a depth of 2 m. However, so far, no applicable methodology has existed to standardly sample the entire cross section with a multipoint method to address microplastic transport in medium and large rivers.

Plastics consist of different polymers that can be buoyant, neutral, or sink, depending upon the composition, density, and shape of the plastic [4,5]. They can also become higher density polymers when mineral fillers are added during production [27]. Plastic particles change in size and density by aggregation or by the growth of biofilms [28–30], and flow turbulence moves them within rivers [31]. They are sometimes even concentrated near the river bed and interact with the sediment. In the oceans, plastic debris has been encountered in all layers, from the surface down to the sediment [6–8,22,32]. If the density of plastics is less than the density of water, they can float or will be close to the water surface. However, given their different densities, turbulence in rivers, biofilms, and so forth, they can be rather compared to the behaviour of suspended sediment particles and are encountered within the whole water column. When looking for an appropriate methodology to address microplastic transport, it should then follow the research experience available for suspended sediment sampling.

For suspended sediment transport, usually a combination of direct and indirect methods is applied to collect measurements at the required spatial and temporal resolution [33]. To address the temporal variability, optical sensors are installed, which continuously record the turbidity at one point

in the channel cross section. Additionally, the distribution of the suspended sediment concentration in the cross section (spatial variability) has to be considered [34]. To establish the cross-sectional mean concentration (Cm), the multipoint method described in ISO 4363 [35] and by Edwards and Glysson [36] is recommended. Using this method, the suspended sediment concentration and flow velocity are measured in various vertical profiles and at different depths [33]. To account for the temporal resolution, various measurements are performed over the entire year and discharge spectrum or indirect devices are used. Adapting to measuring microplastic transport, it is also of high importance to determine the spatial and temporal variability.

The second important challenge, next to the spatial distribution, for collecting measurements in medium- and large-sized natural streams is keeping the device stable at the required points in the water column. High flow velocities and turbulence provide a demanding environment, especially when handling large-sized nets. Similar problems occur for bedload transport measurements and are solved by using heavy, hydraulically optimized devices. Liedermann et al. [37] adapted a basket sampler (BfG sampler) currently used by the Federal Institute of Hydrology in Koblenz, Germany, which is based on the bedload transport meter of Delft Hydraulics [38], and applied it for bedload transport measurements in the Austrian Danube River up to a 200-year flood event (10,738 m<sup>3</sup> s<sup>-1</sup>).

Mesh sizes vary throughout the different studies (between 80  $\mu$ m and 800  $\mu$ m), depending on the focus of the work. In most studies in riverine environments, a mesh size of 300  $\mu$ m was chosen to focus on primary (manufactured in the micrometre size) and secondary microplastics (secondary fragments). When addressing fibres, an even smaller diameter should be used (Dris et al. [22] had a 250-times higher probability of sampling fibres when using an 80  $\mu$ m mesh compared to a 330  $\mu$ m mesh).

There is a knowledge gap in monitoring the presence of microplastics in freshwater systems [8], and it is of the utmost importance to standardize methods for the collection, processing, and analysis of environmental samples [6]. Hence, we aimed to develop a sampling strategy that is applicable to medium- and large-sized rivers, addressing not only the water surface, but the entire transport of plastic particles in the water column. The new methodology presented here should be robust and broadly applicable for measuring microplastic transport in riverine systems.

#### 2. Materials and Methods

#### Development of a Device for Measuring Microplastic Transport

Numerous tests were conducted to develop a sampling strategy and optimise the device. The first tests within the free-flowing section of the Austrian Danube clearly showed the necessity of a strong and stable equipment carrier; therefore the modified BfG basket sampler was used to form the basis of the device. The aim was to use the largest possible net dimensions in order to get the highest possible discharge through the nets. Hence, the net openings were fixed to  $600 \times 600$  mm, with two nets attached at each depth (Figure 1a). The nets are positioned at the surface, in the middle of the water column, and at the bottom of the river. As mesh sizes varied throughout the previous studies, different sizes were used to also address the differences between larger and smaller mesh types. On one side, a 250 µm net was used, and on the other, a 500 µm net was used, at each depth. A 41 µm net was also tested and successfully gathered data at lower discharges, but failed at higher discharges as high suspended sediment and organic loads stressed the net material. At flow velocities of around 2–3 m/s for the mean flow condition, the application of meshes that are too fine is not feasible. However, as an option, a small version can be attached to the centre frame. Furthermore, fibres and microbeads were not the intended focus of this study, but primary and secondary microplastics.

The nets have an actual porosity of 38%, hence a length of about 2.5 m was necessary for reaching the required "open area ratio" of three [39]. The uppermost net assemblage was equipped with a buoyant body to ensure that these nets are skimming the water surface (Figure 1c). The net frame was equipped with a fin on each side to assure streamwise alignment (Figure 1a), but when tests showed that this was not sufficient, another 1.6 m-long fin was added in the middle of the frame

(Figure 1d). The centre nets can be adjusted in height according to the prevailing water depth by using a displaceable stopper (Figure 1b, insert). The lowermost net assemblage was destroyed due to material fatigue at a Danube discharge of 3000 m<sup>3</sup>/s, hence the system was changed by mounting a single centred net to the sampler near the bed instead of the basket (Figure 1e). In the first attempts, the frames were fixed to the steel cable with shackles (Figure 1b), but to accommodate the inclination of the cable, the upper shackle of each net was replaced by an inclination rack (Figure 1d), which allowed the nets to have an upright position when deployed (Figure 1f). A sampling container was constructed at the end of the nets to reduce the emptying time (Figure 1g). After sweeping the content to the container by using a high-pressure sprayer (Figure 1h), the catch can easily be emptied within comparatively short operational times (30–40 min for all nets). A mechanical flow meter was attached (Figure 1i) to measure the discharge through the nets, which is required for calculating plastic concentration.



**Figure 1.** Pictures depicting the development of the device: (a) first prototype with small fins and  $600 \times 600$  mm frames at each level; (b) steel rope and shackles, and adjustable stopper to keep the net assemblage at the right position; (c) buoyant body to ensure near-surface skimming; (d) long fin and inclination rack for good positioning; (e) centred single net near the bed; (f) upright position when inside the water; (g) sampling container to reduce the emptying time; (h) high-pressure cleaning of the nets; (i) mechanical flow meter to measure the discharge through the nets.

## 3. Results

## 3.1. Final Configuration of the Device

After numerous tests and adaptations, the final assembly of the device was reached and can be seen in Figure 2. The uppermost frame carrying the buoyant bodies is only fixed by a stopper; to avoid dropping too low during handling, the stopper of the middle net is adjusted to the correct depth after measuring the water depth in the field. Both are equipped with inclination racks and fins to achieve good positioning in the stream.

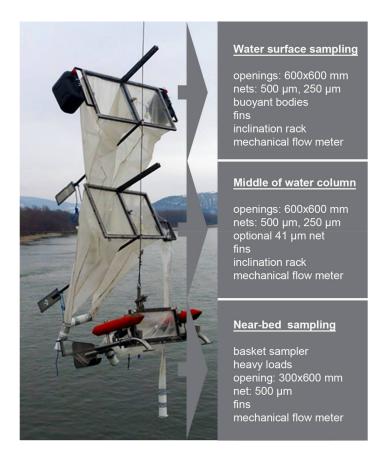


Figure 2. Final device configuration used for collecting measurements in the Austrian Danube River.

At each river cross section, 6–7 vertical profiles were distributed over the entire wetted area; hence a maximum of 35 points were sampled to comprise one multipoint measurement. We aimed to deploy the nets as long as possible because a maximum volume of water should be filtered during the measurements. Nevertheless, one multipoint measurement should be finished in one day, as constant discharge conditions are a prerequisite for further yield calculations. Hence, due to limited daylight time, the sampling duration was set to 30 min for each vertical profile. This had to be slightly reduced on some occasions due to time restrictions (longer cleaning times), or strongly reduced during flood-event measurements due to high loads of suspended sediments and organic matter.

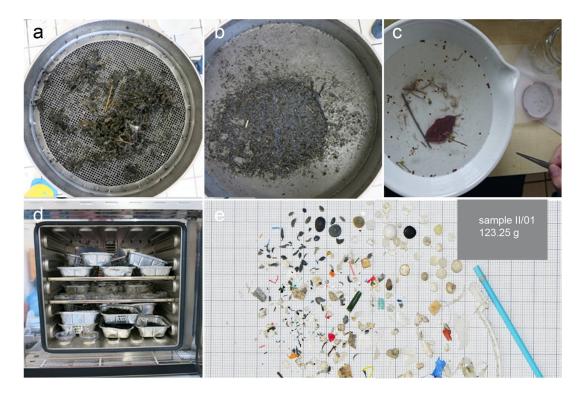
In addition to the water flow rate measured by the mechanical flow meter, acoustic Doppler current profiler (ADCP) measurements were performed to add flow distribution and total discharge to the data collected.

#### 3.2. First Measurements and Sample Processing

With the new device, numerous multipoint measurements were performed at two different cross sections of the Austrian Danube River (at River-km 2160.00 near Aschach and at River-km 1886.24

near Hainburg), covering different discharge conditions. The regulated low discharge (RNQ, 94% exceedance duration) equals 980 m<sup>3</sup> s<sup>-1</sup>, the annual mean discharge (MQ) is 1930 m<sup>3</sup> s<sup>-1</sup>, and the highest navigable and bankfull discharge (1% exceedance duration) is 5130 m<sup>3</sup> s<sup>-1</sup> at the Hainburg study site. As an example of the gathered data and as proof of concept, one dataset is presented in the following.

After washing the load into the sampling container with a high-pressure sprayer, the samples were washed into a labelled sampling container. In the lab, the samples were washed through a cascade of sieves to divide the samples and simplify the further procedures (Figure 3a,b). For the visual sorting, water and NaCl were added to the samples to increase the density of the liquid and allow the plastic particles to float (Figure 3c). For samples with a high quantity of organic matter, H<sub>2</sub>O<sub>2</sub> was added, but as disintegration did not totally break down larger organic items (e.g., leaves), this was not satisfactory for most of the samples. After the first wet sorting, the samples were dried at 50 °C and afterwards dry sorted (Figure 3d). The dry organic matter could then easily be ground, allowing more plastic items to become visible. The plastic particles were separated into fractions <5 mm and then weighed. An example of some plastic particles of one sample can be seen in Figure 3e. For particle identification, attenuated total reflection infrared spectroscopy (ATR-IR) was used.

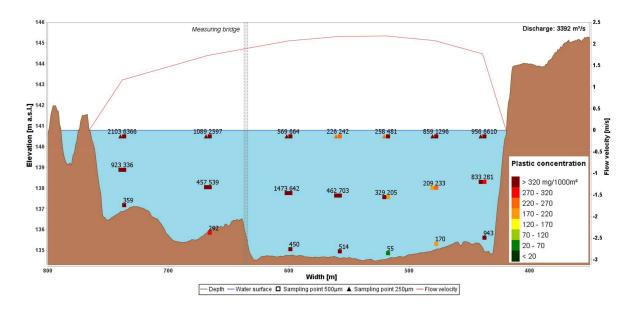


**Figure 3.** Pictures showing the sample processing: (**a**) and (**b**) examples of the sieving cascade for splitting the material, (**c**) manual sorting using water and NaCl, (**d**) samples in the drying stove, and (**e**) example showing plastic particles of one sample.

Plastic concentration  $C_{i,j}$  (g m<sup>-3</sup>) for each vertical profile (i) and each net (j) is calculated using the measured plastic mass  $m_{i,j}$  (g) and the filtered water volume  $V_{i,j}$  (m<sup>-3</sup>) as follows:

$$C_{i,j} = m_{i,j} / V_{i,j} \tag{1}$$

The calculated plastic concentration can be seen in Figure 4. The measurements clearly showed that plastic is not only found in the uppermost layer of the river, but is distributed over the full water column (Figure 4). Some measurements showed an accumulation effect near the river banks (Figure 4) or on one side of the river, but for others, the highest yields were encountered in the middle of the river.

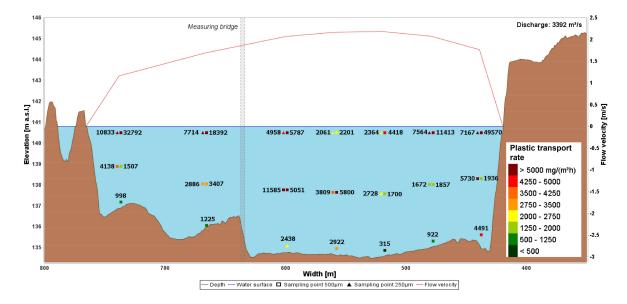


**Figure 4.** Example of the data gathered for one multipoint measurement performed in the Danube River near Hainburg. The sampling was conducted on 13 January 2015, at a Danube discharge of  $3.392 \text{ m}^3 \text{ s}^{-1}$ . The plastic concentration (mg/1000 m<sup>3</sup>) is displayed for each net. a.s.l: above sea level.

A transport rate  $q_{i,j}$  (g m<sup>-2</sup> s<sup>-1</sup>) is then calculated as a product of plastic concentration  $C_{i,j}$  (g m<sup>-3</sup>) and the measured flow velocity  $v_{i,j}$  (m s<sup>-1</sup>):

$$q_{i,j} = C_{i,j} v_{i,j} \tag{2}$$

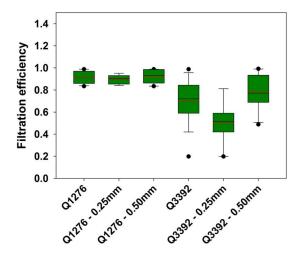
The plastic transport rate for the measurements on 13 January 2015, at River-km 1886.24 near Hainburg are depicted in Figure 5.



**Figure 5.** Example of the data gathered for one multipoint measurement performed in the Danube River near Hainburg. The sampling was conducted on 13 January 2015, at a Danube discharge of  $3.392 \text{ m}^3 \text{ s}^{-1}$ . The plastic transport rate (mg m<sup>-2</sup> h<sup>-1</sup>) is displayed for each net.

A mean plastic transport value for the cross section and an estimation of yearly yields based on a number of measurements at different discharge conditions can then be determined, comparable to the analysis of suspended sediments in rivers, for example, as in [33].

The filtration efficiency was calculated by comparing the measured flow velocities using an ADCP device to the ones obtained using the mechanical flow meter. The filtration efficiency is calculated as the ratio between the volume of water filtered and the volume of water encountered [40]. The efficiency in our case corresponds to the sustained filtration efficiency, as it was assessed after a complete sample cycle in one vertical, including already the potential clogging of the nets. Following the recommendations of Smith et al. [40], a sustained filtration efficiency of at least 0.85 should be obtained. The lowermost nets were excluded for this analysis as ADCP data is missing in the bed-near areas due to side-lobe interference. This analysis was made for two different measurements: the one from 13 January 2015, at a higher Danube discharge  $(3392 \text{ m}^3 \text{ s}^{-1})$  and another at a discharge near low flow conditions (1276  $\text{m}^3 \text{s}^{-1}$ ). The filtration efficiency at the low discharge for a sampling duration of about 25 min is high for both net types (Figure 6). The minimum efficiency was found at 0.83 (Table 1); the mean value could be determined at 0.91. The difference between the 250  $\mu$ m and the 500  $\mu$ m net is considerably low (mean value 0.90 for 250 µm, 0.92 for the 500 µm net). Despite the reduced sampling time (approximately 5 min), the efficiency is lower for the higher discharge, especially for the smaller mesh size (250 µm). With an average efficiency of 0.52, the 250 µm net performed poorly compared to the 0.78 of the 500 µm net, indicating a faster clogging of the fine mesh at these conditions (higher abundance of fine material, primarily suspended sediments). Regarding the 500 µm net, two sample points had a filtration efficiency of around 0.50. As the material caught was comparable to samples in other nets, it might be possible that blockage of the mechanical flow meter with leaves led to an underestimation of the filtered water volume, hence the efficiency.



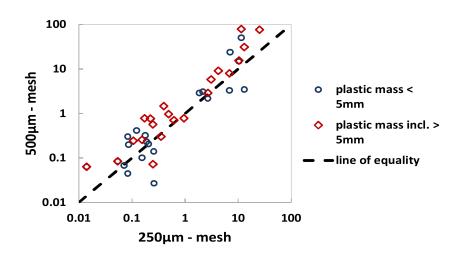
**Figure 6.** Filtration efficiency calculated by comparing acoustic Doppler current profiler (ADCP) measurements to the values of the flow meter mounted in the center of the nets. The analysis was made for two different discharge stages in Hainburg (with Q1276 and Q3392 denoting the discharges of 1276 m<sup>3</sup> s<sup>-1</sup> and 3392 m<sup>3</sup> s<sup>-1</sup>) and both net types (250  $\mu$ m and 500  $\mu$ m).

As different mesh sizes were used, the sampled plastic mass was compared for both net types. Spatial variability is high and can also be observed when using the same mesh sizes side by side (e.g., Figure 4, center point at the vertical to the right of the bridge pier). However, there is a tendency that even more material is sampled with nets of the 500 µm mesh size, as most values are found above the line of equality (Figure 7).

Especially the comparison of the plastic mass of particles exceeding 5 mm in size shows that more material is sampled with 500  $\mu$ m nets.

**Table 1.** Calculated values for the filtration efficiency during two measurements at two different discharge stages in Hainburg (1276 m<sup>3</sup> s<sup>-1</sup> and 3392 m<sup>3</sup> s<sup>-1</sup>). The values are calculated for all measured points and for the 250  $\mu$ m nets and 500  $\mu$ m nets separately.

		Min	Max	Mean
lowest mesh excluded	Q1276	0.83	0.99	0.91
	Q1276—250 μm	0.84	0.95	0.90
	Q1276—500 μm	0.83	0.99	0.92
	Q3392	0.20	0.99	0.70
	Q3392—250 μm	0.20	0.81	0.51
	Q3392—500 µm	0.49	0.99	0.78



**Figure 7.** Comparison of plastic mass sampled with both net types (250  $\mu$ m and 500  $\mu$ m side by side) for three different measurements (discharges 1276 m<sup>3</sup> s<sup>-1</sup>, 3179 m<sup>3</sup> s<sup>-1</sup>, and 3392 m<sup>3</sup> s<sup>-1</sup>). Material smaller than 5 mm is depicted in blue; the mass of particles larger than 5 mm is plotted in red.

### 4. Discussion

The first measurements with the device, addressing multiple depths within the water column and various vertical profiles distributed over the cross section, showed the spatial heterogeneity of the plastic transport distribution. Due to different densities of the polymers, aggregation, or the growth of biofilms, and because of flow turbulence, particles are not constrained to only floating on the surface [6–8,22,32]. Particles may accumulate in areas near the bank; a slight tendency of higher near-bank concentration respectively to transport can be seen in the presented results (Figures 4 and 5). Hence, studies addressing only the surface layer (by using a manta trawl) or sampling only at parts of the cross section (groyne fields or near-bank areas) may strongly over- or underestimate plastic transport in riverine systems. For the example presented, plastic transport varies by up to a factor of 30 when only sampling the near-surface layer (as would occur using a manta trawl), depending on which section is chosen.

The chosen net types (250  $\mu$ m and 500  $\mu$ m) were analyzed regarding their sampling efficiency. For the lower discharges, efficiency is above 90% for both mesh sizes, but decreases substantially for the 250  $\mu$ m mesh at higher discharges. Suspended sediment concentration and grain size as well as organic matter increases with the discharge, leading to a more rapid clogging of the nets. Hence, the sampling duration was adapted for further measurements and should be well attended in future studies. The comparison between plastic mass sampled side by side with the two net types showed that at higher discharges, even more material is sampled with the 500  $\mu$ m net. Especially bigger particle sizes seem to be sensitive to back pressure. Hence, the usage of the 500  $\mu$ m nets is recommended for rivers during stages with higher turbidity, whose grain sizes already are in the range of the 250  $\mu$ m mesh.

# 5. Conclusions

A new device was introduced that is the first to address microplastic transport at various depths and multiple vertical profiles in medium and large streams, for the purpose of addressing the knowledge gap in the monitoring of freshwater systems. Conditions in rivers are often inhospitable and harsh due to high flow velocities, turbulence, and a high transport of suspended loads and organic matter. Therefore, it is highly demanding to deploy floating nets in accurate positions within the system. The new device is robust and applicable also at high flow velocities and discharges, and was also operative at a one-year flood event (discharge:  $5.700 \text{ m}^3 \text{ s}^{-1}$ ; flow velocities around  $3 \text{ m s}^{-1}$ ). First measurements proved that multipoint measurements are necessary for obtaining the spatial distribution of plastic concentration and are a prerequisite for calculating the transport that is occurring. The 500 µm mesh sizes are recommended for a partly turbid stream.

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**Author Contributions:** Marcel Liedermann, Philipp Gmeiner, Sebastian Pessenlehner, Marlene Haimann, and Helmut Habersack designed and built the monitoring device. Marcel Liedermann, Philipp Gmeiner, Sebastian Pessenlehner, and Marlene Haimann performed the field measurements. The draft text was prepared by Marcel Liedermann; all other co-authors provided completions in their fields.

Conflicts of Interest: The authors declare no conflicts of interest.

# Abbreviations

The following abbreviations are used in this manuscript:

- EU28+2 European Union member states and candidate countries
- ISO International Organization for Standardization
- BfG Bundesanstalt für Gewässerkunde (Federal Institute of Hydrology), Koblenz, Germany
- ATR-IR Attenuated total reflection infrared spectroscopy
- RNQ The regulated low discharge at Hainburg (94% exceedance duration; time series 1981–2010) equals 980 m<sup>3</sup> s<sup>-1</sup>
- MQ The annual mean discharge at Hainburg (time series 1981–2010) equals 1930  $m^3 s^{-1}$

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