



# Floating microplastic debris in a rural river in Germany: Distribution, types and potential sources and sinks

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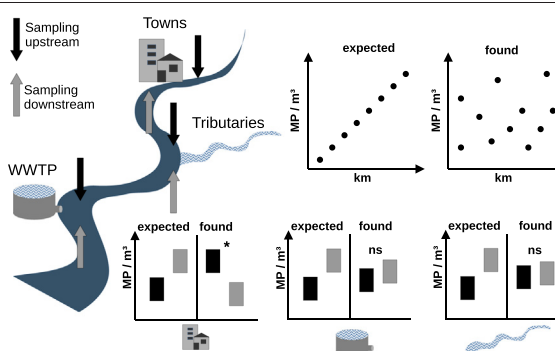
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## HIGHLIGHTS

- Microplastic pollution of a rural river was studied with high spatial resolution.
- Results showed considerable microplastic loads despite an agricultural catchment.
- Shapes of microplastics differed from those detected in larger streams.
- The contamination pattern did not increase along the river course.
- Weirs in towns served as potential sinks for floating microplastics.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Microplastic debris affects marine as well as freshwater ecosystems and an increasing number of studies have documented the contamination in aquatic environments worldwide. However, while the research focuses on oceans and larger rivers, little is known about the situation in smaller rivers within rural catchments. Since microplastics pose various risks to ecosystems, wildlife and human health, it is important to identify potential sources, sinks and transport patterns, which are probably different for small rivers. In this study, we investigate the contamination with microplastic debris of the river Ems, representing a smaller river in Northwest Germany with an agricultural catchment. We hypothesised that with increasing river length the plastic concentration increases, especially downstream of towns, waste water treatment plant (WWTP) effluents and major tributaries as they may be important point sources of microplastics. We collected 36 surface water samples at 18 sampling sites within the first 70 km using manual driftnets. We sampled every 7 km and upstream and downstream of three larger towns, four major tributaries and four WWTP effluents. Overall, we found  $1.54 \pm 1.54$  items  $m^{-3}$ , which corresponds to the plastic concentrations in larger streams. However, the shape of the detected items differed as we did not find potential primary microplastic. Furthermore, the pattern contradicts our assumption, that the contamination increased with distance to the river's source. Downstream of towns, we found significantly less floating microplastic indicating possible sinks due to sedimentation at sites with slowing flow velocity caused by weirs in towns. Hence, the non-linear distribution pattern of microplastics indicates potential sinks of microplastics due to flow alterations on the river course.

This should be considered in future studies modelling microplastic distribution and transport. Furthermore, studies especially in smaller rivers are urgently needed to identify and quantify sources and sinks and to find applicable solutions to reduce microplastic loads.

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## 1. Introduction

Imagining today's life without plastic products is hardly possible, especially when considering their diverse social and economic benefits. On the one hand, plastic properties such as lightweight, longevity, robustness and low costs enabled the development of innumerable and important inventions (Barnes et al., 2009), on the other hand, these same qualities constitute disposal problems and immense environmental threats (Zbyszewski and Corcoran, 2011; Thompson et al., 2009). Numerous studies documented the contamination of aquatic environments with plastic debris and reported several risks that especially microplastics (particle size < 5 mm) pose to ecosystems, fauna as well as human health (Browne et al., 2007; Wright et al., 2013; Syberg et al., 2015; Yu et al., 2020; Hale et al., 2020).

Besides marine environments – all oceans (e.g. Cole et al., 2011; Ivar do Sul and Costa, 2014; Lusher et al., 2014; Shim and Thomposon, 2015; Isobe et al., 2017) and several shorelines (Browne et al., 2011; Wessel et al., 2016) contain alarming amounts –, microplastics also affect freshwater ecosystems such as lakes (e.g. Eriksen et al., 2013; Wang et al., 2018; Egessa et al., 2020), rivers (e.g. Frei et al., 2019; Tibbetts et al., 2018; Wong et al., 2020) and estuaries (e.g. Zhao et al., 2015; Gray et al., 2018; Zhang et al., 2019). In addition to acting as additional transport pathways for terrestrial (micro)plastic debris into marine environments (Jambeck et al., 2015; Van Emmerik and Schwarz, 2020) this has implications for the respective freshwater ecosystem itself.

Large rivers have been particularly studied regarding their microplastic pollution in recent years (e.g. Lechner et al., 2014; Mani et al., 2015). Large rivers differ from small rivers in the criterion of navigability. Due to their often large catchment areas in urban and industrial settings as well as inland navigation, considerable amounts of primary and secondary microplastic particles have been documented. Primary microplastic particles are produced in sizes of less than 5 mm mostly consisting of spheres or pellets e.g. for plastic industry or cleaning products (Browne et al., 2007; Ryan et al., 2009; Thompson et al., 2009). They often correlate with the presence of large industrial plants near the river (Mani et al., 2015). Secondary particles originate from larger plastic products, fragmenting in the environment under physical or chemical conditions such as mechanical abrasion or photodegradation (Browne et al., 2007; Ryan et al., 2009). Both, primary and secondary microplastic particles often enter rivers through wastewater treatment plants (WWTP; Frei et al., 2019), cities (Browne et al., 2007; Faure et al., 2012), tyre abrasion (Kole et al., 2017; Knight et al., 2020) or landfills (Su et al., 2019; Golwala et al., 2021). Studies in some great European streams such as the Seine River (Gasperi et al., 2014), the Danube River (Lechner et al., 2014) and the Rhine River (Mani et al., 2015; Mani and Burkhardt-Holm, 2020) investigated the distribution pattern of microplastic debris, which corresponded to the densely populated and industrial areas along their riversides.

While recent studies provided valuable insights into the situation of large freshwater bodies, information for smaller rivers with a mainly agricultural dominated catchment area is scarce. However, microplastic loads and types, entry paths and distribution patterns in small rivers are likely to differ from those identified in greater streams. Smaller rivers in rural catchments are usually less affected by industrial discharges and thus presumably less exposed to primary microplastic sources. In addition, these rivers are rarely affected by inland navigation or fisheries, which can be additional sources of microplastics in larger fluvial systems (Mani et al., 2019) or lakes (Wang et al., 2018). Furthermore, studies investigating microplastic pollution in larger rivers also work on a larger scale, e.g. Mani et al. (2015) examined 11 sampling sites along a river range of about 700 km. Focusing on a larger scale can certainly help to elucidate general patterns or to estimate the total contribution of a river to microplastic pollution of marine ecosystems. However, on this larger scale it is more difficult to identify particular sources and potential sinks. Hence, we worked on a much smaller scale (18 sampling sites along a river range of 70 km) to identify local

sources and potential sinks more in detail and to test whether the microplastic contamination results in a more or a less homogenous distribution pattern on the smaller scale. Therefore, this survey intends to document the extent of microplastic contamination within the river Ems in Germany, representing a small river basically influenced by agricultural land use. With the focus on floating microplastic particles, we expect the microplastic contamination of the river Ems (1) to increase with the distance to the rivers' spring and (2) to show high local peaks directly downstream wastewater treatment plants (WWTP), towns and tributaries hence indicating those as sources for microplastic debris in small rivers.

## 2. Methods

### 2.1. Study site

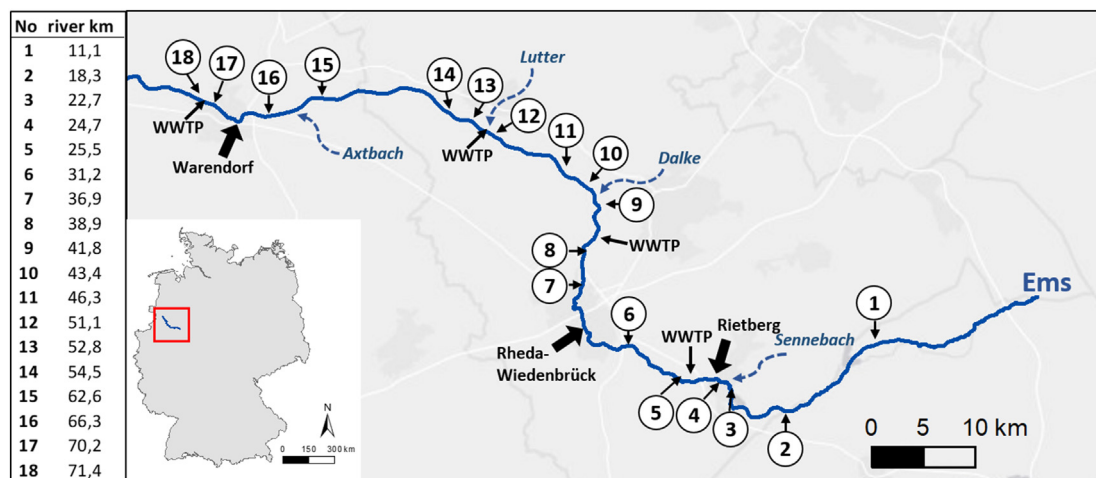
The study was conducted in the river Ems, a 371 km long lowland river, rising in the Westphalia Bay in North-West Germany and flowing into the North Sea (FGG Ems, 2015). Samples were taken within the first 70 km downstream the source. The catchment of the river Ems in this region is dominated by agriculture (41.3%), grassland (18.5%) and forests (20.3%), whereas settlements and commercial areas (together 16.8%) are of less importance (MULNV NRW, 2020). About 300,000 inhabitants live in this area (MULNV NRW, 2020). At our first sampling site, the river Ems showed a width of 3 m, at the last sampling site of 15 m. Mean annual river discharge is 79.9 m<sup>3</sup>/s (measured at Versen; FGG Ems, 2015).

### 2.2. Sampling

Samples were taken every 7 km for the first 70 km of the river downstream the rivers' spring (Fig. 1). Additional samples were taken upstream and downstream of three towns (Rietberg, Rheda-Wiedenbrück, Warendorf), four effluents of wastewater treatment plants and four smaller tributaries. We took two samples within the surface water of the river Ems at each sampling site (resulting in 36 samples at 18 different sampling locations) and then averaged the microplastic concentrations per site. Samples at the same site were collected in pairs at the same day. Each sample was taken by a driftnet (rectangular opening 10 cm × 50 cm, mesh size 250 µm), which floated for 5 min filtering the upmost 10 cm of the water column leading to a mean sampling volume of 7.78 m<sup>3</sup> (±5.06 m<sup>3</sup>) for each sample (n = 18). For further details, see Hübner et al. (2020). We calculated the amount of filtered water according to the respective flow velocity and determined the abundance of microplastic debris (items m<sup>-3</sup>). Water samples were stored in glass jars and preserved with 60% ethanol. All samples were taken in March and April 2014 under dry and similar weather conditions.

### 2.3. Sample processing

Each sample was filtered over cellulose filters (pore size < 10 µm, Melitta) and examined under a microscope (magnification between 8- and 35-fold) to identify plastic particles. Detected particles were counted out visually taking into account the physical properties of the item, such as its colour, structure, texture or flexibility (Norén, 2007). According to their shape, the items identified as potential microplastic particles were subsequently categorized as different groups: flakes (=particle fragments), fibres, spherules/spheres, pellets and plastic films following the classification scheme proposed by Hidalgo-Ruz et al. (2012). After visually identifying a particle as a potential microplastic item, we touched it with a hot needle (hot needle test; e.g. De Witte et al., 2014) to check if the material melted. To validate this preliminary identification and to analyse the plastic type selected plastic particles were sorted according to colour, size and shape. Items larger than 1 mm (n = 123 items out of 236 items) were subsequently



**Fig. 1.** Map of the study area (Ems) with the 18 sampling sites, three main towns (Rietberg, Rheda-Wiedenbrück and Warendorf; downstream 4, 6 and 16, respectively) and sites of the four waste water treatment plants (WWTP; downstream 4, 8, 12, 17) and four small tributaries (blue dashed arrows; downstream 3, 9, 12, 15).

analysed by a Fourier transform infrared spectroscopy (FTIR, Agilent Cary 670 FTIR) with a spectrum from 500 to 4000 nm. For this analysis, particles were mixed with potassium bromide in a ratio of 1:100 and were pressed into small pellets. The spectrum that was analysed by a FTIR was consequently compared with spectra from the literature, using the SDBS database (Saito and Kinugasa, 2011) to compare the FTIR spectra of our samples against validated spectra of the database.

During laboratory processing of samples blank controls were run in parallel. Filters were rinsed with the same amount of tap water as samples and filters were treated the same way and for the same time as the sample filters. After finishing the samples, the blank filters were analysed and potentially detected fibres (e.g. originating from the air) were subtracted from the samples. Particles or films were not detected in the blanks. Apart from the drift net, we tried to avoid plastic instruments for treating the samples, i.e. we used glass vials with metal lids for sample transportation, glass funnels and glass petri dishes, cellulose filter, steel tweezers and glass vials in the laboratory.

#### 2.4. Statistical analysis

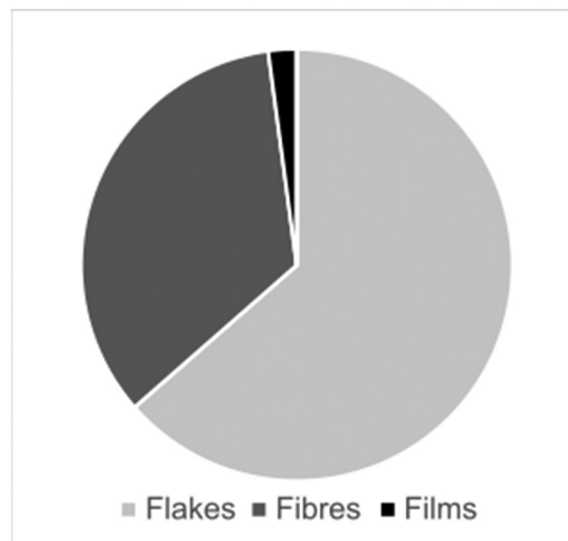
All of the following analyses were performed in R version 4.0.2 (R Core Team, 2020). The cut-off level for determining significance was  $p < 0.05$  for all analyses. Data exploration was applied on the data following the protocol suggested by Zuur et al. (2010), explanatory variables (distance to the spring of the river, site specificity, flow velocity) were tested for collinearity using the R package *corrplot* (Wei and Simko, 2021). Since flow velocity was included in the calculation of the dependent variable (items per  $m^3$ ) and thus showed an autocorrelation, flow velocity was not included as an explanatory variable in the following analyses. We performed two linear models to test the amount of microplastic items per  $m^3$  with the distance to the spring of the river. The first model (items per  $m^3$  ~ distance to the spring of the river) included the distance to the spring of the river (in km) as fixed covariate (continuous variable). In a subsequent linear model (items per  $m^3$  ~ river km + site specificity) we incorporated the fixed covariate site specificity (categorical variable with two levels; "downstream", if sampling site was directly located downstream a town, WWTP or tributary, or "others" for all other sampling sites) to account for potential effects of site-specificity. No interaction terms were included into the two models. To test for differences between specific groups, we subsequently classified the sampling locations into six different groups according to their location along the river: upstream ( $n = 4$ ) and downstream ( $n = 4$ ) tributaries, upstream ( $n = 3$ ) and downstream

( $n = 3$ ) towns, upstream ( $n = 4$ ) and downstream ( $n = 4$ ) wastewater treatment plants (WWTP). We used a pairwise Student *t*-test with a Bonferroni correction for multiple testing to test for differences between upstream and downstream wastewater treatment plants, towns, and tributaries, respectively.

### 3. Results

#### 3.1. Microplastic content

Within the river samples ( $n = 36$ ) the abundance of microplastics ranged from no detected items  $m^{-3}$  (min.) to 5.28 items  $m^{-3}$  (max.) with an average of  $1.54 \pm 1.54$  items  $m^{-3}$ . Items characterised as flakes occurred in most samples in various colours and sizes and were the prevailing item type (Fig. 2). We furthermore found a high amount of microplastic fibres (Fig. 2). Plastic films showed various colours (Fig. S1, Appendix). Based on the spectrum of the FTIR analysis analysed flakes and films were identified as polyethylene (Figs. S2, S3, Appendix).



**Fig. 2.** The pie chart shows the percentages of the identified microplastic particle types. Of a total of 236 particles detected in the river Ems, 150 were classified as flakes, 81 as fibres and five particles were categorized as films. Spherules or pellets were not detected in the study.

### 3.2. Spatial distribution

In contrast to the hypotheses the concentration of floating microplastic did not increase with distance to the river source. Within the first 20 km the microplastic concentration already reaches amounts comparable with more distant sites. Concurrently, there occur sampling points with a great distance to the river source which indicate lower or almost no contamination (Fig. 3). This demonstrates a very low relation between distance from source and amount of microplastic concentration (linear model including the fixed covariate “distance to the spring of the river”,  $R^2 = 0.004$ ,  $p = 0.810$ ). The linear model including also the fixed covariate “site specificity” did not indicate effects of the specific location (directly downstream a town, WWTP or tributary) of the sampling sites ( $R^2 = 0.014$ ,  $p = 0.903$ ).

### 3.3. Effects of potential point sources of microplastics

Contrasting our hypotheses, neither WWTP, towns nor major tributaries were correlated with elevated concentration in the river water column (Fig. 4). In contrast, microplastic concentrations were significantly lower downstream of towns ( $p = 0.012$ , Fig. 4). Microplastic concentrations downstream of WWTP tended to be higher than upstream of these effluents, but this trend was not significant.

## 4. Discussion

In contrast to our hypotheses the river Ems does not show a significant increasing pollution with microplastics with distance to the source, but an alternating concentration pattern similar to the larger scale investigations in larger rivers (e.g. Mani et al., 2015). Having a closer look on the hypothesised sources of microplastic it becomes evident that in our case towns turned out to not be dominant sources of microplastic (as postulated by e.g. Eriksen et al., 2013; Free et al., 2014; Lechner et al., 2014) but potential sinks of microplastics as microplastic concentrations in the water were significantly lower downstream of the towns than upstream. In all the examined towns there are weirs for flood protection and to surpass large differences in water levels (e.g. 3.6 m in the most downstream town; engineering

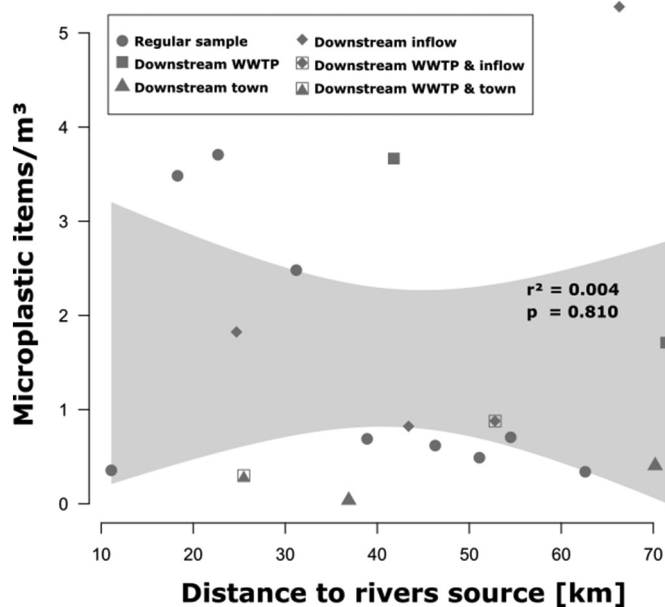


Fig. 3. Linear model of the microplastic particles (items  $m^{-3}$ ) against the distance to the spring of the river Ems. No significant correlation was found; the grey shadow indicates 95% confidence intervals.

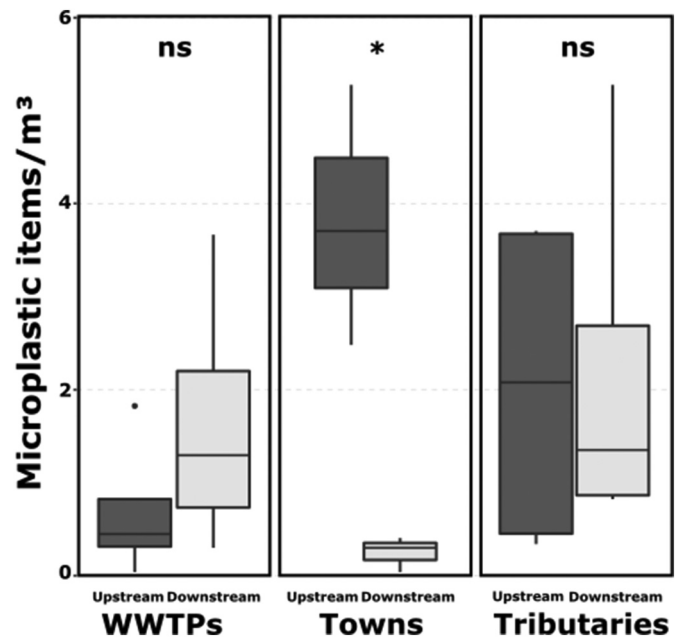


Fig. 4. Detected amounts of items per  $m^3$  upstream (darker grey) and downstream (light grey) of WWTP, towns and small tributaries. The concentration of microplastics was significantly lower downstream towns ( $p = 0.012$ ). No significant differences between concentrations upstream and downstream of WWTP or tributaries were found, respectively (ns = not significant, \* =  $p < 0.05$ ).

office A. Vollmer, unpublished). Hence, the weirs and the resulting reduction of flow velocities upstream of the weirs may lead to an increased sedimentation of floating microplastic. Transport of floating microplastic was shown to increase with increasing discharge (van Emmerik et al., 2019; Wagner et al., 2019) and to decrease with decreasing flow velocities in reservoirs or dams (Hübner et al., 2020; Watkins et al., 2019; Zhang et al., 2015). Furthermore, biofouling can burden buoyant particles so that also particles with smaller densities than water may sink and deposit at the riverbed (Lagarde et al., 2016; Leiser et al., 2020; Leiser et al., 2021). Hence, the weirs in the towns may act as potential sinks for microplastic leading to lower amounts of floating microplastic. To verify this hypothesis sediment samples should be evaluated or sediment traps could be employed (Saarni et al., 2021). In addition, cities could still be entry pathways for plastic into rivers, as especially larger litter was often sighted on the banks near cities in this study.

Concerning sewage waste contamination, WWTP effluents contributed to microplastic entry but concentrations downstream of WWTPs were not significantly higher than upstream. WWTPs can remove up to 99% of microplastic (Magnusson and Norén, 2014). Nevertheless, WWTPs were shown to emit between  $1 \times 10^7$  and  $5 \times 10^9$  particles per year (Minténig et al., 2017) and reported as important entry paths of microplastics due to the entry of synthetic fibres during each load of laundry (Frei et al., 2019; Valine et al., 2020). Two sampling sites at the river Ems downstream inflowing sewers are contaminated with microplastics above average and may hence increase the microplastic loads. However, sewage discharge cannot constitute the only pathway for microplastic debris as a lot of sampling sites upstream the first sewers are already contaminated with microplastic debris and there was no consistent and significant trend of higher microplastic concentrations downstream of WWTP.

Tributaries represent a further potential medium for transporting microplastics into rivers. The highest amounts of microplastic debris with  $5.28 \pm 2.17$  items  $m^{-3}$  was detected downstream a small tributary with a comparable land use in the catchment. Compared to the low contamination upstream this tributary it is very likely that this inflow is

polluted with microplastics acting as an additional load. However, we did not find a general or significant trend downstream of tributaries. In addition to microplastic input, inflows from tributaries could also cause turbulences or increase local flow velocity, which might lead to resuspension of already sedimented particles or in contrast dilute microplastic concentrations. Local water currents or wind-induced currents have been shown to play an important role regarding heterogeneous contamination patterns in lakes (Imhof et al., 2013; Eriksen et al., 2013; Zbyszewski and Corcoran, 2011), which might also apply to fluvial systems. To analyse the exact relation between tributaries and the distribution pattern, the tributaries themselves would have to be tested for their microplastic load before they enter the River Ems.

The results of this survey indicate a considerable contamination of streams and small rivers despite its rural catchments. We detected an average amount of  $1.54 \pm 1.54$  items  $m^{-3}$  in the first 70 km of the river Ems. Hence, this small and rural river shows a similar contamination with microplastic particles as large European freshwater streams, such as the Danube ( $0.316 \pm 4.664$  items  $m^{-3}$ ; Lechner et al., 2014), at least as long as only the sheer abundance of particles is considered. This indicates a relatively intensive pollution of the river Ems, especially considering its smaller drainage basin compared with those rivers. Furthermore, great industrial plants or inland navigation, identified as potential sources of microplastics at the Danube and the Rhine (Lechner et al., 2014; Mani et al., 2015), are lacking in the study area. However, plastic particles found in this study in the river Ems were rather smaller than the particles found mainly in the Danube. This shows that a direct comparison as well as transfer of possible input pathways between large and small rivers is difficult.

Properties of most items found within the river Ems differ from those described in larger streams, which are strongly affected by industry. For example, no spherules could be detected in the Ems, but flakes dominated the river samples, mostly blue and red in colour. The detected polyethylene items, such as coloured flakes or films, are secondary fragments of larger plastic products. This corresponds to other investigations describing polyethylene as the most abundant polymers in their studies (Sadri and Thompson, 2014; Zbyszewski and Corcoran, 2011). The upper reach of the river Ems is dominated by agricultural land use. This could explain the absence of primary particles, which are mostly connected to industrial production. Considering the high abundance of fibres within the Ems river samples agricultural material such as plastic strings could be taken into consideration, besides the already mentioned potential source of synthetic fibres in sewage from laundry. Fishing-related sources like in Lake Garda (Imhof et al., 2013) are unlikely at the river Ems since the river is neither impacted by inland navigation nor by commercial fishing.

Several agricultural techniques such as sewage sludge application (Corradini et al., 2019) which was deployed until 2018 in these districts (Guhlke, Neue Westfälische, 2019) or plastic mulching (Huang et al., 2020) were shown as entry paths for microplastics into soils. Soil erosion through heavy rainfall events or winds could subsequently transport these particles and films into rivers. Besides the potential impact of agricultural techniques, further pathways for microplastic litter such as direct dumping, indirect discharge through wind-blown matter or tyre abrasion, airborne deposition and leaching of litter at riversides through precipitations cannot be excluded.

Finally, our study shows that also smaller rivers and streams with an agriculturally dominated catchment and an absence of industry can contain considerable concentrations of microplastic. Furthermore, these concentrations do not increase along the river course but for example towns with weirs may act as sinks for floating microplastic. This needs to be taken into account for modelling microplastic distribution and transport (into the ocean). Furthermore, investigations especially in smaller rivers are an urgent need to identify sources and to find possibilities to decrease the pollution through plastic litter.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.151641>.

## CRediT authorship contribution statement

**Pia Eibes:** Data collection, Formal analysis, Investigation, Writing – Original draft preparation. **Friederike Gabel:** Conceptualization, Supervision, Writing – Review & editing.

## 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.

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