





Identification of microplastic pathways within a typical European urban wastewater system

Korinna Altmann¹  | Caroline Goedecke¹ | Claus-Gerhard Bannick²  |
Attaallah Abusafia³  | Christian Scheid³ | Heidrun Steinmetz³ | Andrea Paul¹ |
Claudia Beleites^{4,5} | Ulrike Braun² 

¹Bundesanstalt für Materialforschung und -prüfung, Berlin, Germany

²Umweltbundesamt, Berlin, Germany

³Technische Universität Kaiserslautern, Kaiserslautern, Germany

⁴Chemometrix GmbH, Wölfersheim, Germany

⁵Julius Kühn Institute, Federal Research Centre for Cultivated Plants, Institute for Ecological Chemistry, Plant Analysis and Stored Product Protection, Berlin, Germany

Correspondence

Korinna Altmann, Bundesanstalt für Materialforschung und -prüfung, Unter den Eichen 87, Berlin 12205, Germany.
Email: korinna.altmann@bam.de

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Abstract

In recent years, thermoextraction/desorption-gas chromatography/mass spectrometry (TED-GC/MS) has been developed as a rapid detection method for the determination of microplastics (MP) mass contents in numerous environmentally relevant matrices and, in particular, for the measurement of polymers in water samples without time-consuming sample preparation. The TED-GC/MS method was applied to investigate a typical European municipal wastewater system for possible MP masses. Such investigations are important in view of the recent revision of the Urban Wastewater Treatment Directive. Four different representative sampling sites were selected: greywater (domestic wastewater without toilet), combined sewer, and influent and effluent of a wastewater treatment plant (WWTP). All samples were collected by fractional filtration. Filtration was carried out over mesh sizes of 500, 100, 50, and in some cases, 5 µm. Polyethylene (PE), polypropylene (PP), and polystyrene (PS) were detected in all samples, with the PE fraction dominating in all cases. Styrene-butadiene rubber which serves as an indication of tire abrasion, was only found in the influent of the WWTP. The highest MP mass contents were found in the combined sewer, so MP can become a source of pollution during heavy rain events when the capacity limits of the effluent are reached, and the polluted effluent is released uncontrolled into the environment. Based on the studies, MP retention from the WWTP could be estimated to be approximately 96%. Few trends in polymer type or mass contents were detected within the different fractions of the samples or when comparing samples to each other.

KEYWORDS

mass contents, microplastic, microplastic analysis, microplastic pathways, TED-GC/MS

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INTRODUCTION

The presence of microplastics (MP) is a phenomenon recognized worldwide, [1], where MP particles are found not only in water and soil but also in air and biota [2, 3]. MP consists of synthetic polymers, mainly with some additives, with a size of 1 to 1000 μm [4] and are mostly caused by the degradation of macroplastics, for example, from littering [5, 6]. Macroplastics are exposed to UV radiation, oxygen, and moisture in the environment, which causes them to age and, due to mechanical abrasion, finally break into small MP fragments [7–9]. The smaller the fragments, the more they are airborne and spread to the most remote areas all over the earth [10]. So far, there is a huge need for further research in the field of MP. Possible pathways into the environment have not been clarified [11]. Next to littering, MP can, for example, enter the environment via abrasion of textile clothing or compost/sewage sludge used as fertilizer in agriculture [12, 13]. It is transported into the world's oceans via rivers and terrestrial ecosystems, possibly entering the groundwater reservoirs and hence entering the human food chain. In many countries around the world, urban wastewater systems are in place to reduce the human impact on the natural environment and to treat raw sewage before it is flushed back into rivers [14]. These urban sewage systems may represent other possible pathways for MP to enter the environment [15–17].

Most of the investigations so far have been carried out with time-consuming micro-spectroscopic methods yielding information on the MP particles since particle number and size are essential for the assessment of (eco-) toxicological issues. For regulatory questions, in contrast, it is necessary to have fast and robust methods. To make first estimations regarding further measurements, total mass contents are sufficiently suitable.

On the other hand, there are currently no harmonized procedures for sampling, sample preparation, and detection. The first approaches in standardization are recognizable in the ISO/TR 21960:2020 [4]. Here, two basic methodologic approaches can be identified depending on the information requested [18]. Polymer type, form, shape, and number of particles can be determined by imaging methods like μ -Fouriertransform infrared (μ -FTIR) and μ -Raman spectroscopy, while integral methods detect the polymer type and the corresponding mass content such as Pyrolysis-GC/MS or ThermoExtraction/Desorption-GC/MS [19, 20].

Scientific publications often report frighteningly high particle numbers in the environment, but unfortunately, due to the time-consuming spectroscopic analysis procedures and difficulties with overlaying, small or colored particles, [21] these numbers are based on only a few real data and have usually been extrapolated. [22] At the same time, initial intercomparison tests show in the first results that the reliability of the experimentally measured data is difficult and hardly comparable. [23, 24] There is an urgent need for research in this area in the future [25, 26].

Our approach to MP detection is different. In recent years, we have developed a rapid method that is able to determine the polymer

type and the corresponding mass content in real environmental samples. This method is called thermoextraction/desorption-gas chromatography-mass spectrometry (TED-GC/MS) and combines the advantage of the large sample volume of thermogravimetric analysis with the very low detection limits of GC/MS technology. [27] The TED-GC/MS method can identify and quantify representative sample amounts of real environmental samples according to the most common polymers found in the environment, such as polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), polyamide (PA), and acrylates. TED-GC/MS, as a thermo-analytical and integral method, has the advantage that styrene-butadiene rubber (SBR), often associated with tire abrasive, can also be detected in real environmental samples. [28] Even though SBR is by definition not an MP, it is often mentioned in the context of MP because it is a polymer that forms particles in the size of MP dimension. It is even assumed that 1/3 of the MP in the environment could result from tire abrasion. [29] Furthermore, the TED-GC/MS measurement includes all particles and even the sizes down to the nanometer scale, which is a big challenge for microscopic methods such as μ -Raman.

The following article presents the functionality of the TED-GC/MS as a detection method for wastewater analysis for routine analysis for MP type and mass content determination. The article presents data on MP contents in municipal sewerage systems of a bigger European city. There are some data on different wastewater treatment plants (WWTP) available in the literature, [30–32], but a more comprehensive analysis of the entire municipal wastewater system often not exists. The entire wastewater system must be considered to track down undesirable entry paths into the environment. In addition to the WWTP with its influent and effluent, further components must also be considered, for example, the inflow of grey and black water, rainwater inlets, especially from road gullies, and stormwater retention tanks. To detect these possible entry pathways in a local European sewage system, we sampled four different sewage locations and will thus get the chance to derive avoidance strategies in the future.

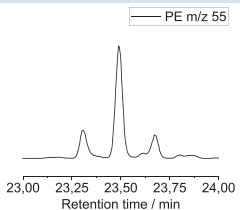
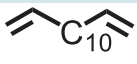
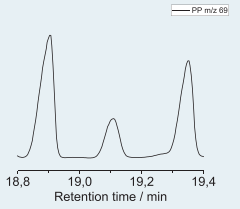
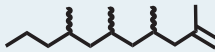
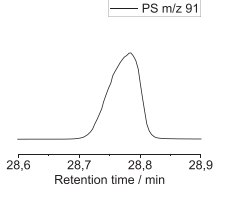
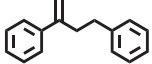
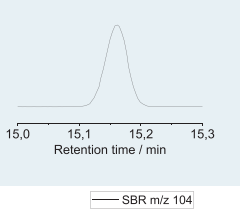
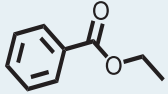
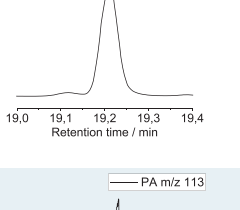
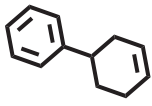
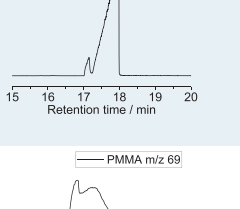
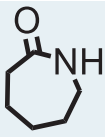
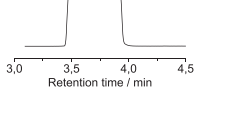
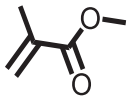
METHODS

Materials

The city of Kaiserslautern, with 100,000 inhabitants, was selected for the investigation of a typical urban wastewater system in Germany. Sampling locations have been grey water as wastewater from a household from bath and kitchen without the toilet water (two dates), a combined sewer in a stormwater retention tank (six dates), and the WWTP with the influent (four dates) and the effluent (three dates).

Pristine polymers, which are needed as reference for quantification by TED-GC/MS, are as free as possible of additives and kindly provided by PlasticsEurope as granulate. The pellets are cryo-milled into MP powders.

TABLE 1 Summary of polymers routinely screened with polymer markers for quantification and limit of detection (LOD)

Polymer	Polymer marker Peak shape	Name	Structural formula	m/z	LOD μg
Polyethylene (PE)		1,14-Pentadecadiene		81, 55, 95	2.2
Polypropylene (PP)		2,4,6,8-Tetramethyl-undec-10-en		111, 69, 154	0.14
Polystyrene (PS)		2,4-Diphenyl-1-buten		91, 130, 104, 208	0.08
Polyethylene terephthalate (PET)		Ethylbenzoat		105, 77, 122, 150	0.24
Styrene-Butadiene Rubber (SBR)		Cyclohexenyl-benzen		104, 158	0.06
Polyamid 6 (PA)		Caprolactam		113, 85, 55	0.24
Acrylate		Acrylate		69, 100, 41	0.12

Sampling

Sampling was performed via fractionated filtration in dry weather. A representative amount of wastewater was filtered over stainless steel ring sieves with mesh sizes of 500, 100, and 50 μm [33]. As sampling development progressed, our methods improved, and we were also able to sample with a 5 μm sieve since 2019. Particles in the fraction $5 < 50 \mu\text{m}$ will be numerous, but due to their small size, they have a limited influence on the total mass contents, so we consider the total contents of sampled days with and without the 5 μm fraction equally. The filtered water volumes are based on the particle content of the different types of wastewater, [34] whereby 1000 L per sample could be filtered in the case of water from the WWTP effluent, but only about 20 L was filtered of water from the influent to the WWTP, as the sieves then clogged up and a filter cake is formed. For the same reason, only about 20 L was sampled for the other two wastewaters, grey water, and combined sewer.

Detection

Measurements with the TED-GC/MS are a multi-stage process. [27, 28] First, a representative sample quantity, which can be more than 500 mg depending on the inorganic content of the matrix, is filled into a crucible and heated from 25°C to 600°C (10 K/min) under inert N_2 atmosphere (50 ml/min) in a single arm thermobalance (TGA2, Mettler-Toledo GmbH). For these experiments, a 10 mg intake was used. This corresponds to about 0.5%–3% of the sieve residue mass for the fraction 1000–500, 500–100, and 100–50 μm . The sampling mass of the 50–5 μm fraction was often lower than 10 mg than all samples measured. The resulting decomposition gases are deposited on an adsorber (length: 20 mm, diameter: 2 mm) consisting of polydimethylsiloxane (Sorb-Star©, Mercury Instruments), before this fully loaded adsorber is transported to the thermodesorption GC-MS coupling in an automatic process. There, the adsorber is slightly heated with 40 K/min from 50°C to 200°C, so that the decomposition gases bound are released again. The decomposition gases are then collected in a cold injection system (CIS4, Gerstel) working as cryotrap at -100°C and hence transferred to the gas chromatograph (Agilent 7890, Agilent Technologies), which is equipped with an HP-5MS column (length: 30 m \times inner diameter: 0.25 mm \times film thickness: 0.25 μm , Agilent Technologies). The decomposition gases are separated and detected in a mass-sensitive detector (5977B, Agilent). The spatial separation between the pyrolysis of the environmental sample, which is always a mix of matrix and polymer, and the GC/MS with the bypass via the adsorber has the advantage that even large, representative sample quantities can be measured due to the fact that there is no contamination or clogging of the GC/MS system. At the same time, the loading volume of the adsorber is large enough that the decomposition products are representative of the sample. Most water samples can be analyzed without time-consuming sample preparation. One single TED-GC/MS run for one sample lasts

between 69 min and 2 h 30 min. Up to 100 measurements (including blank values) can be done per week as the TED-GC/MS runs automatically with 18 samples in the autosampler for one sequence.

Due to the complex composition of environmental samples, the chromatograms resulting from mass spectrometry are also very complex, with plenty of different peaks representing various molecules.

The obtained chromatogram is routinely screened for specific decomposition products, called polymer markers, for the respective polymers. To evaluate whether the decomposition product found really corresponds to one of the polymers in question, various mass traces within the individual peak spectra are considered. For the detection of polystyrene, for example, the decomposition product 2,4-diphenyl-1-butene should be present with the mass traces m/z 91, m/z 130, and m/z 208. More polymer markers and mass traces are explained in detail in Table 1. Some marker peaks used have irregular shapes. In the case of methyl methacrylate, the combination of short retention time and dynamically heated injection system causes deviation from the Gaussian shape. However, since the detector operated in its dynamic range and the borders of the peak integration are clear, quantification should—in contrast to peaks with strong fronting or tailing—not be compromised. The same is applicable to the PA6 decomposition product caprolactam, whose peak exhibits a triangular shape.

A blank value is measured before each sample measurement to ensure that no polymer marker molecule has been carried over from the previously measured sample, which would lead to a false positive effect. Moreover, an internal standard consisting of five-times deuterated polystyrene is used for quality control. Polystyrene is very well suited as a standard because it can be dissolved easily in toluene, and thus very small quantities can be added very precisely to the actual sample. The deuterated PS has an m/z ratio of 96, which differs well from the PS from environmental samples with m/z 91.

RESULTS

The mass contents detected in the wastewater depending on the different size fractions are presented in Table 2. To compare results, all data are standardized to 1000 L and thus presented in $\text{mg}/1000 \text{ L}$ (or mg/m^3).

The highest polymer contents are found at the beginning of the wastewater chain with the raw wastewater of the greywater and in the combined sewer to the treatment plant. Three different polymer types were found: PE, PP, and PS. The detected masses in the greywater varied strongly from 32.0 to 243.1 mg/m^3 (PE) down to 2.1 to 6.2 mg/m^3 (PP), and 0.1 to 0.3 mg/m^3 (PS). The same picture with even higher masses is found in the combined sewer with 0.6 to 992.2 mg/m^3 (PE), 0.8 to 75.7 mg/m^3 (PP), and 0.3 to 46.7 mg/m^3 (PS). The influent of the WWTP comprises the highest diversity of polymer types. Besides the dominant PE with 5.7–114.9 mg/m^3 , only lower masses of 1.5–11.7, 0.2–15.6, and 1.8–18.0 mg/m^3 were detected in individual fractions for PP, PS, and SBR. Minor polymer

TABLE 2 Polymer masses found in the greywater from the organic farm, the combined sewer, and the influent and effluent of the wastewater treatment plant (WWTP) according to 5, 50, 100, and 500 μm size fraction

Sampling location	Sampling date	Fraction μm	Polyethylene mg/1000 L	Polypropylene mg/1000 L	Polystyrene mg/1000 L	Styrene-butadiene rubber mg/1000 L	
Greywater	19th September 2018	500–1000	32.0	2.1	0.1	0	
		100–500	158.7	6.2	0.4	0	
		50–100	147.6	4.2	0.4	0	
	11th October 2018	500–1000	136.2	0	0	0	
		100–500	154.4	3.1	0	0	
		50–100	243.1	6.1	0.9	0	
Combined sewer	17th August 2018	500–1000	38.9	0	0	0	
		100–500	188.8	8.0	3.3	0	
		50–100	21.8	0.8	0.3	0	
	20th February 2019	500–1000	0	34.5	0.5	0	
		100–500	0	45.2	4.2	0	
		50–100	297.9	25.3	2.5	0	
	25th March 2019	5–50	0	0.8	0	0	
		500–1000	0	0	0	0	
		100–500	320.1	23.7	3.5	0	
		50–100	0	13.0	1.5	0	
	4th December 2019	5–50	25.4	1.1	0.3	0	
		500–1000	193.0	0	4.7	0	
		100–500	992.2	75.7	16.8	0	
		50–100	358.0	45.7	0	0	
	21st January 2020	5–50	218.8	30.6	0	0	
		500–1000	0	0	6.8	0	
		100–500	621.7	53.5	46.7	0	
		50–100	257.0	0	6.4	0	
	5th February 2020	5–50	10.8	17.4	0.5	0	
		500–1000	72.9	0	0	0	
		100–500	296.6	0	0	0	
		50–100	152.1	0	0	0	
	Influent WWTP	5th November 2018	5–50	0.6	0	0	0
			500–1000	50.2	1.5	0.5	0
100–500			44.7	5.1	0.4	3.0	
15th November 2018		50–100	72.0	6.9	2.4	9.0	
		500–1000	114.0	3.8	2.2	5.3	
		100–500	112.5	11.2	1.9	9.6	
2nd February 2019		50–100	92.4	0	2.2	6.5	
		500–1000	114.7	8.2	0.9	0	
		100–500	112.7	9.6	3.0	18.0	
50–100		53.1	11.7	1.1	9.3		

(Continues)

TABLE 2 (Continued)

Sampling location	Sampling date	Fraction μm	Polyethylene $\text{mg}/1000\text{ L}$	Polypropylene $\text{mg}/1000\text{ L}$	Polystyrene $\text{mg}/1000\text{ L}$	Styrene-butadiene rubber $\text{mg}/1000\text{ L}$
		5–50	5.7	2.1	0.2	1.8
	25th February 2019	500–1000	70.1	1.6	1.0	0
		100–500	114.9	5.4	15.6	0
		50–100	19.2	4.4	1.1	2.3
Effluent WWTP		12th February 2019	500–100	0	1.1	0
	18th February 2019	100–500	4.7	0.5	0	0
		50–100	0	0.5	0.1	0.2
		500–100	0	0.4	0.1	0
	4th March 2019	100–500	5.9	0.9	0	0
		50–100	0	0.2	0	0.3
		500–100	2.7	1.0	0.1	0.1
		100–500	11.8	1.6	0.2	0
		50–100	0	0.4	0.1	0.3

Note: Maxima within each sampling day and polymer are marked bold.

contents of the same polymer types are found in the effluent of the WWTP with 2.7–11.8 mg/m^3 (PE), 0.2–1.6 mg/m^3 (PP), 0.1–0.2 mg/m^3 (PS), and 0.1–0.3 mg/m^3 (SBR). PE is predominant in all sampling locations.

When comparing all measured samples, it becomes clear that despite the universal dominance of PE, the different samples show a high variance. For example, although the polymers PP and PS are found in principle in all sampling locations, they are not necessarily found in all fractions or samples, nor are they found with similar contents.

For an extended evaluation, further sampling campaigns, especially for seasonal fluctuations, are necessary. So far, no annual trends for MP particles (type and mass) within an urban WWTP exist.

A special note refers to SBR. The detection of SBR is often associated with tire abrasion in the environment, [25] because it is present in almost all tire types of all manufacturers, but unlike natural rubber has specific polymer markers. When evaluating SBR masses, one should always be aware that these are mainly introduced to the urban wastewater system during wet weather conditions (precipitation). Especially in the case of heavy rain, a “first flush” at the beginning of the rain event would presumably mobilize and transfer most of the deposited SBR into the urban wastewater system. To evaluate the SBR content, the weather conditions before sampling must always be taken into account. Although high SBR masses are not to be expected during dry weather conditions, traces of SBR can still be detected. An explanation for this phenomenon would be that SBR particles are captured in sinks within the sewer system or in stormwater retention tanks and then released gradually again by dry weather flow.

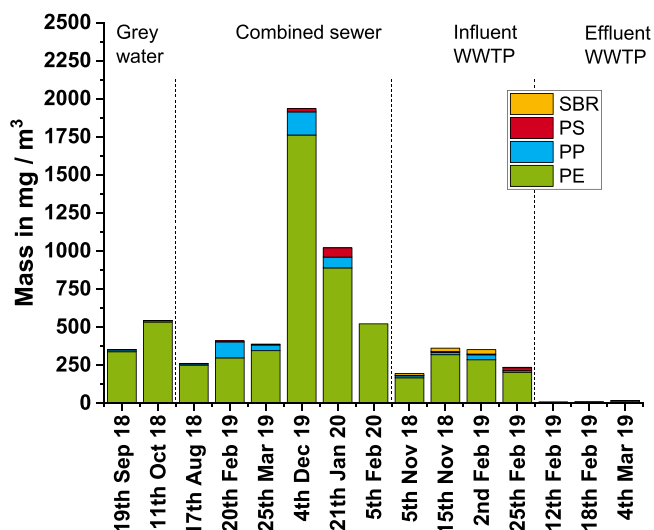


FIGURE 1 Total masses of the various sampling locations dependent on the polymer types found and the sampling day in mg/m^3

To get a better overview of the water pollution of the different sampling sites, the totals of the polymer contents regarding the sampling days were calculated by summarizing the individual polymer fractions per day, which is given in Figure 1. We are aware of the fact that due to the small number of sampling dates, only a limited discussion of the observed polymer mass fractions is possible, and merely the observed trends can be described:

- (i) It is obvious that the masses of the sampling locations' grey water, combined sewer, and influent of the WWTP are much

higher compared to the effluent of the WWTP. The total mass content per sampling day varies with a magnitude of 10 between 195 mg/m^3 (influent WWTP, 5th November 2018) and 1935 mg/m^3 (combined sewer, 4th December 2019), although all sampling occurred in dry weather.

- (ii) All samples are dominated by PE. On the one hand, PE is very much used in industry, and daily live products are why high mass loads are reasonable, on the other hand, high PE fractions could be explained by the abrasion of PE water pipes, which applies to the entire sewage system. However, the high PE content in the greywater in relation to the other wastewater is questionable. The grey water location is an organic farm, where the residents pay much attention to a sustainable and healthy way of life hardly compatible with the high amount of PE. As the graywater samples were taken in a cement tank, an accumulation of biological and domestic substances, such as fatty acids from cooking or other organic compounds or biofilms, could possibly cause the observed effect. These may produce in TED-GC/MS the same polymer marker molecules as PE, and hence, PE would be overestimated.
- (iii) In addition to the general high PE, there is mostly PP ranging from 0.8 to 75.7 mg/m^3 in the combined sewer and smaller amounts of PS with 0.1 mg/m^3 in gray water to 46.7 mg/m^3 in the combined sewer.
- (iv) While the wastewater from the grey water and the inflow into the WWTP only differ by a factor of two in terms of the total masses per day, more variation can be seen in the case of the combined sewer.
- (v) It is also noticeable that neither PET nor PA was found in any of these samples. These polymers are extensively used in textiles and should be sampled directly with the washing machine wastewater in the case of grey water. However, due to the special nature of the sampling site of an organic farm, one may assume that the inhabitants wear mainly textiles made of natural materials such as cotton or linen.
- (vi) In the case of PE, it does appear that the fractions $100\text{--}500 \mu\text{m}$ most often have the highest masses overall, even if PE is sometimes only found in one fraction. In contrast, SBR is preferably observed in the smallest fraction, $50\text{--}100 \mu\text{m}$. For the other polymers, hardly, any trend is observed.

DISCUSSION

A direct comparison of the total polymer masses for the different types of wastewater is provided in Figure 2. The mean polymer content determined for the combined sewer is 756 mg/m^3 , three times as high as the inflow to the WWTP (286 mg/m^3). It is remarkable that a large proportion of the MP particles detected in the combined sewer ($\sim 62.2\%$) do not reach the WWTP. Some of these particles will sediment in the sewage system on their way to the WWTP, some might be flushed out during heavy rainfall. However, there is also an inflow of fresh water in a mixed water system, so a dilution of the MP mass contents can occur.

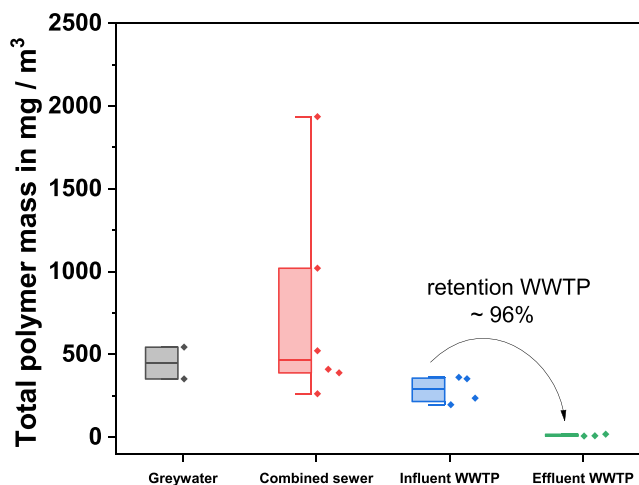


FIGURE 2 Variation of the total polymer masses found at the different sampling locations

A strongly reduced number of polymers with an average polymer mass content of 11 mg/m^3 was found in the wastewater of the WWTP, which corresponds to a general MP reduction of about 96%. This may indicate efficient retention of the polymers in the WWTP. The boxplot in Figure 2 shows that the total polymer masses of the individual wastewaters lie within a limit of 25%–75% of the mean value.

However, as can be deduced from Figure 3, the data of the individual sampling days do not scatter around a mean value because there is no normal distribution, but a distribution that decreases from zero to high values. This means that mean value and standard deviation do not describe these data correctly. Except for the WWTP effluent, where all polymers exhibit a quite homogeneous pattern, all other sampling sites show variable contents over sampling dates.

Scaling the mass content of each polymer from zero to one allows a comparison of the importance of each tested wastewater compartment with respect to polymer types (Figure 4).

Whereas the highest amounts of PE, PP, and PS were found in the combined sewer (cf. storm in Figures 3 and 4), SBR dominates in the wastewater influent, followed by PS and PP. Greywater is dominated by PE, and the wastewater effluent, in general, is characterized by strongly reduced polymer masses, not detectable on this scale. In the combined sewer, which also contains the largest amounts of PS, PP, and PE, each sampling day seems to exhibit its own pattern regarding the individual polymer composition.

Sampling locations with a higher number of sampling dates (combined sewer and the influent of the WWTP) are compared according to their occurrence and frequency in Figure 5. The highest sample numbers are available for the combined sewer. Here PE, PP, and PS were found under dry weather flow conditions. However, the results varied strongly depending on the sampling day (Table 2). On 5th of February 2020, only PE has been detected. On other days, however, PE levels exceeded 1000 mg/m^3 (4th December 19 and 21st January 2020). PP and PS could not be detected within all size

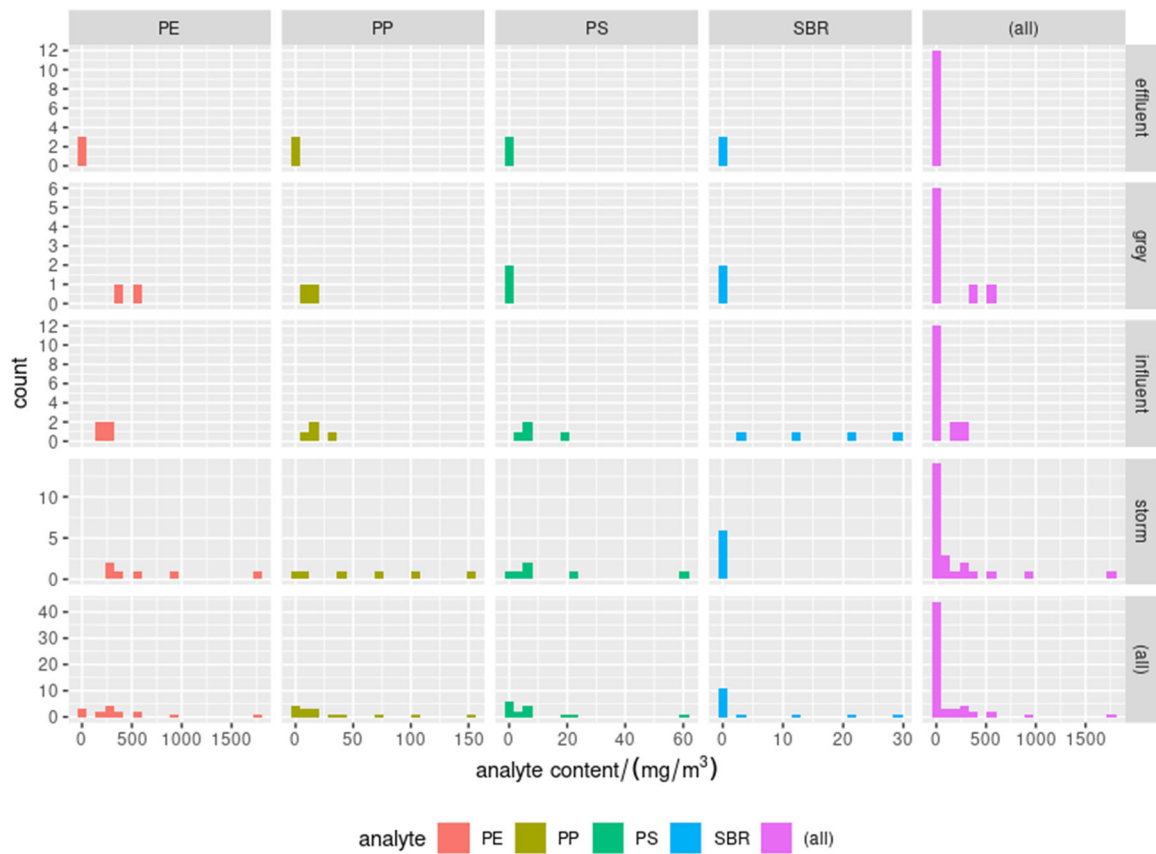


FIGURE 3 Histograms of the encountered analyte contents for the different analytes in columns and sampling locations in the rows. (Storm) denotes the combined sewer, and (all) is the marginal (total) distributions for analytes and sampling locations, respectively.

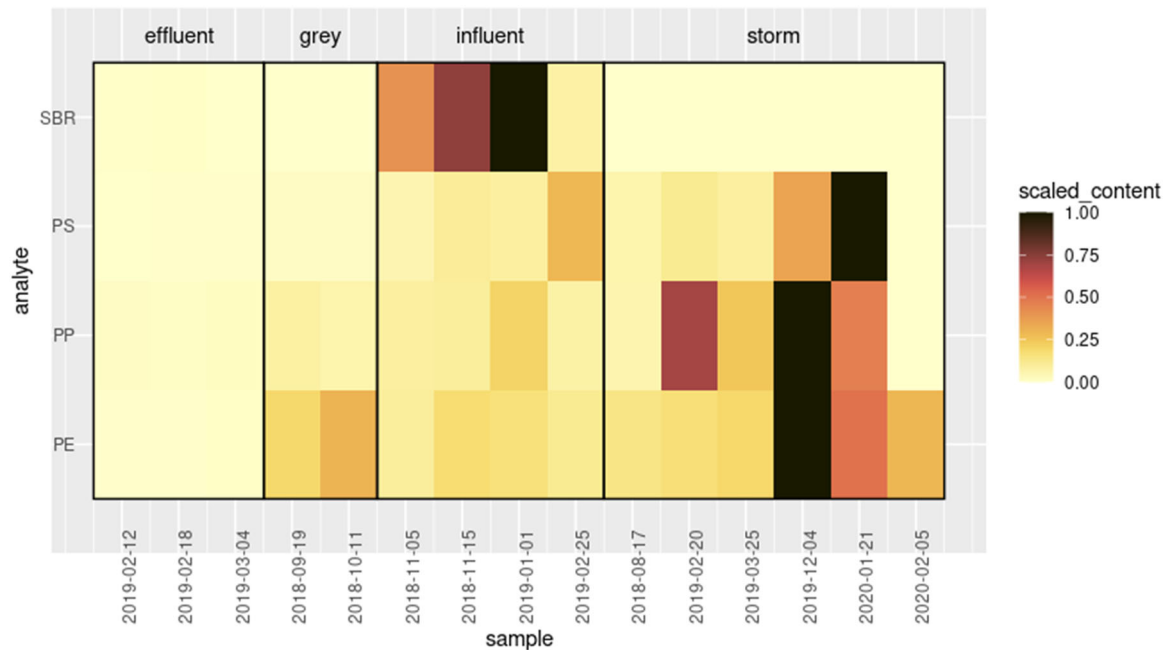


FIGURE 4 Concentrations as a fraction of the maximum encountered concentration of the respective analyte. (Storm) here denotes the combined sewer.

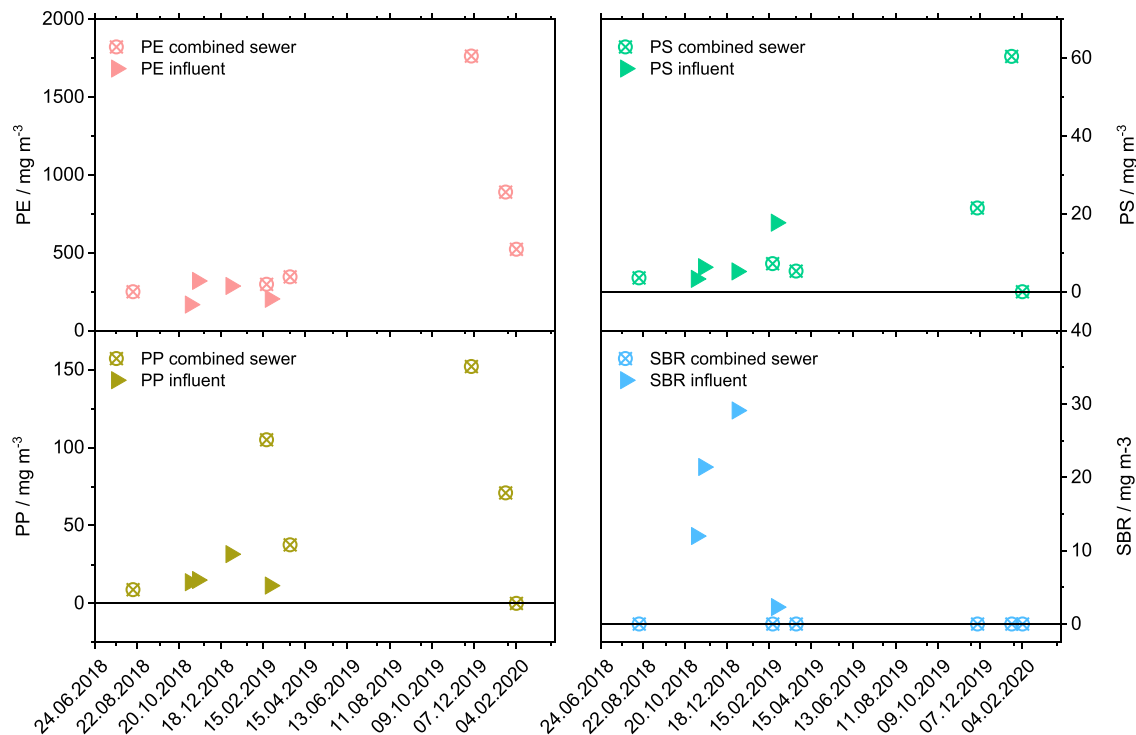


FIGURE 5 Cumulated polymer mass of sampling days of the combined sewer and the wastewater treatment plant influent

classes. Rather, the detection appears random and unpredictable, with 8.8–152.0 mg/m³ for PP and 3.6–60.4 mg/m³ for PS.

To check, whether the sampling locations combined sewer and WWTP influent are correlated, the temporal order of sampling can be compared in the roughly overlapping period from 5th November 2018 to 25th March 2019. It is known that processes such as deposition and dilution of particles both will affect the resulting polymer load detected. However, from a phenomenological point of view, similar amounts of PE are detected in both wastewaters (Figure 5). In contrast, PP masses in the combined sewer are much higher than on 15th February 2019, where PP predominates in the WWTP influent, possibly indicating dilution by other water bodies. A clear trend, however, is observed for SBR in autumn 2018, where it is exclusively found in the WWTP influent, suggesting other sources of entry pathway or very low amounts in the combined sewer that does not exceed the limit of detection due to long dry weather periods.

SUMMARY AND CONCLUSION

A sampling of different types of municipal wastewater in the city of Kaiserslautern was technically possible in dry weather using filter cascades to identify possible pathways of MP into the aquatic environment.

The polymers PE, PP, and PS were detected in all samples at all sampling locations, whereby the proportion of PE always dominated. We could further prove that TED-GC/MS, as an analytical method, is, in principle, suitable for the investigation of plastics in wastewater.

A comparison of the average total polymer mass contents of the different sampling points shows that the WWTP has an MP retention of about 96% and that the highest MP mass was found in the combined sewer. This appears to be a major potential source of environmental pollution when runoff rates are exceeded the WWTP during heavy rains, resulting in an uncontrolled discharge of effluent from the stormwater retention basin into the environment.

The limited number of samples makes reliable statements on the quantity and occurrence of the polymers difficult. No clear trends could be detected. However, this study allows an insight into the fundamental problems of environmental sampling, which is characterized by random sampling. Although representative sample volumes were taken, only a snapshot can be generated, which is hard to interpret. As can be seen from the open question, whether the high PE fraction is real or perhaps the result of a long-chained interferent that occurs in this particular type of samples by environmental matrix. Hence, analytical approaches must be critically questioned and readjusted. Future studies need to address both the clarification of this phenomenon and the understanding of MP entry, transport, and loss through systematic sampling strategies.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

ORCID

Korinna Altmann  <https://orcid.org/0000-0003-3467-4882>

Claus-Gerhard Bannick  <https://orcid.org/0000-0003-3685-9477>

Attaallah Abusafia  <https://orcid.org/0000-0003-1740-2631>

Ulrike Braun  <https://orcid.org/0000-0002-3682-3637>

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