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Quantification of polyethylene terephthalate micro- and nanoplastics in domestic wastewater using a simple three-step method



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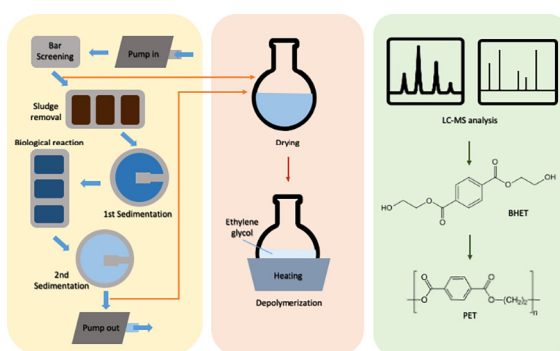
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HIGHLIGHTS

- PET MNP was detected in all the influents (ranging from 24.9 µg/L to 680 µg/L).
- PET MNP was detected in most of the effluents (ranging from <LOD to 23.1 µg/L).
- A wastewater treatment plant cannot prevent PET MNP entering the environment.

GRAPHICAL ABSTRACT



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ABSTRACT

Concerns about impact of small plastic particles, known as microplastics (<5 mm) and nanoplastics (<1 µm), together abbreviated as MNP, on the environment and on human health have increased in recent years. Polyethylene terephthalate (PET) microplastics have been detected previously in different environmental samples including freshwater and wastewater sludge. In the present study, we target all small plastic particles of PET with a diameter smaller than 5 mm (PET MNP). A simple three-step method of drying, (in matrix) PET depolymerization in ethylene glycol and liquid chromatography-mass spectrometry (LC-MS) analysis, was applied for the quantification of PET MNP in influents and effluents collected from ten Dutch wastewater treatment plants. The PET recovery was 98% in the wastewater matrix. The limits of detection (LOD) for PET in influents and effluents were 2.0 µg/L and 1.2 µg/L, respectively. PET MNP was detected in all the influents (ranging from 24.9 µg/L to 680 µg/L) and most of the effluents (ranging from <LOD to 23.1 µg/L). The results of the present study confirmed that WWTP effluents can be a source of PET MNP in the environment.

1. Introduction

The issue of plastic waste in the environment has been obvious and problematic for some time. In recent years, concerns about the impact of small plastic particles on the environment and on human health have increased (Rochman, 2018). These small plastic particles are defined as

microplastics (<5 mm) and nanoplastics (<1 µm) (GESAMP, 2016; Okoffo et al., 2020). In the present study, the abbreviation “MNP” is used to represent all small plastic particles with a diameter smaller than 5 mm, including both microplastics and nanoplastics.

The sources of MNP in the environment can be primary (direct release from commodities containing microbeads or microfibers) or secondary

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(degradation products or fragments of larger plastics). The MNP, from different pathways (direct release, washed by surface run-off, blown by wind, etc.), may eventually end up in the aquatic environment, including rivers, estuaries and oceans. Furthermore, MNP have been detected in hundreds of species of aquatic organisms (Van Cauwenberghé and Janssen, 2014; UNEP and GRID-Arendal, 2016), and it is estimated that humans ingest up to several thousand MNP particles per year through seafood consumption, with numbers varying depending on eating habits and living locations (Cox et al., 2019). Various types of microplastics have been detected in influents of wastewater treatment plants (WWTP). According to Li et al. (2018) and Xu et al. (2019), microplastics are most frequently detected in fiber shape in both influent and effluent. Polyethylene terephthalate (PET) accounts for a large proportion of manufacturing textile microfibers (Napper and Thompson, 2016). It is reported that PET textile garments lose thousands of microfibers in each washing cycle (Napper and Thompson, 2016; Barrows et al., 2018). PET MNP are not degraded in WWTPs, and the MNP in effluents are emitted to the surface water (Napper and Thompson, 2016). It has been reported that the wastewater treatment processes (e.g. sedimentation) can remove the majority of microplastics in the wastewater (Xu et al., 2019; Jiang et al., 2020). However, concerning the huge volume of effluent being constantly discharged to the aquatic environment, WWTPs can still be one of the main routes for MNP to enter the aquatic environment (Xu et al., 2020). On top of that, globally, 44 % of all wastewater flows generated by households is not safely treated (treated by secondary or higher processes or with effluent discharges meeting relevant standards) (UN, 2021).

Conventional methods for MNP identification, including visual observation (microscope), and/or analyses based on Fourier-transform infrared (FTIR) and Raman, require sample pretreatment to remove the organic matter (Zarfl, 2019). Visual observation is time-consuming and also not always reliable. For example, it was reported that as much as 70 % of the small particles that were characterized visually as microplastics were not confirmed as plastics by FTIR spectroscopy (Hidalgo-Ruz et al., 2012). Though new identification method based on holographic imaging and machine learning provides high accuracy, it is not validated for small-size (20 μm) particles (Bianco et al., 2020). Another method for MNP detection is fluorescent dyeing, but this method has not yet been validated for samples with high organic matter (Shim et al., 2016). Visual counting was also widely used for the quantification of microplastics (Gimiliani et al., 2020). In recent years, several thermoanalytical methods have been reported for the quantification of MNP. For example, thermal decomposition followed by mass spectrometry (MS) detection such as pyrolysis gas chromatography-MS (Okoffo et al., 2020; Liu et al., 2021), a thermoanalytical method based on elemental analysis of the parameters C, H, N, S and O (Mallow et al., 2020) and thermal depolymerization methods followed by liquid chromatography (LC) or LC-MS analysis (Tian et al., 2022; Wang et al., 2017).

Early use of the thermal hydrolysis method took the approach of the depolymerization of PET MNP followed by a water extraction, centrifugation, a cleanup procedure and LC-MS analysis (Wang et al., 2017; Zhang et al., 2019). A simplified three-step thermal glycolysis method for PET MNP quantification in environmental samples (based on the depolymerization using an excess of ethylene glycol) was developed in our previous study (Tian et al., 2022). This type of thermal depolymerization method has not been applied to wastewater matrices. The present study applied the simplified three-step method for the quantification of PET MNP in domestic wastewater to understand the mass concentrations of PET MNP in WWTP influents and effluents, and also to explore the fate of PET MNP during the wastewater treatment process.

2. Materials and methods

2.1. Reagents and standard preparation

LC-grade water and methanol were purchased from Biosolve (Valkenswaard, the Netherlands). Zinc acetate (purity $\geq 99\%$) and PET fiber (purity $\geq 99\%$, diameter 0.1 mm) were purchased from Fisher

Scientific (Landsmeer, the Netherlands). Bis(2-hydroxyethyl) terephthalate (BHET, purity $\geq 99\%$), terephthalic acid (TPA, purity $\geq 99\%$), TPA- D_4 (purity $\geq 99\%$) and ethylene glycol (purity $\geq 99\%$) were purchased from Sigma-Aldrich (Zwijndrecht, the Netherlands). Stock solutions of BHET and BHET- d_4 were prepared in ethylene glycol (500 mg/L) just before the experiments and stored at 4 °C.

2.2. Synthesis of BHET- D_4

BHET- D_4 (BHET-2,3,4,5- D_4) was used as internal standard (Section 2.3). BHET- D_4 was synthesized using TPA- D_4 , as the chemical standard was not available at the time of the experiment. The detailed procedure of BHET- D_4 synthesis can be found in the Supplemental material under Fig. S1.

2.3. Quality control

Several measures were taken to avoid and control background contamination during the experiments. No PET plastic tools nor containers were used during the experimental procedure, including sampling. Lab coats (100 % cotton) were worn during the experiment. Lab tools including stainless steel tools, glass jars for sampling and laboratory glassware were cleaned and baked at 420 °C for 4 h before use. The depolymerization reactions were performed in a fume hood which was vacuum cleaned before each reaction. Reagents and materials were used exclusively for the depolymerization experiments in this study. Solvent blanks and procedural blanks were carried out. Solvent blanks and procedure blanks were analyzed in duplicate with each sample batch to monitor and (if necessary) correct for background contamination. TPA and BHET have not been reported to be naturally present in the environment, and even though TPA is present in domestic wastewater, the concentration of TPA is expected to be at trace level and multiple orders of magnitude lower than PET (Wang et al., 2009). Thus, BHET quantified in samples in the present study can be assumed that exclusively come from the PET in the wastewater. BHET- D_4 was used for quantification and monitoring retention time (RT) shifts and signal drift.

2.4. Sampling

Wastewater samples (both influents and effluents) were collected from ten different domestic WWTP in the Netherlands between April 2021 and July 2021 during dry weather conditions. The detailed information of the sampling sites is shown in Table 1. Influent samples were collected from each WWTP after the bar screening step (in which large pieces of solid waste are removed). All effluent samples were collected from the WWTP after the complete wastewater treatment process at the point of discharged to the environment (the same day as influent, within 24 h). About 1 L of each sample was collected and stored in a glass jar sealed with a polypropylene cap. For each sampling location, both influent and effluent were sampled twice within a day. At the laboratory, the two samples (e.g. two influent samples from the same location) were stir-mixed to a pooled sample (about 2 L) before the analysis. Then each pooled sample was passed through a 5 mm stainless-steel sieve to check for the presence of macro-

Table 1
Sampling information.

Sampling site	Population served	Daily discharge after treatment (L)
Location 1	6×10^4	8.8×10^6
Location 2	5.5×10^4	8.2×10^6
Location 3	3.5×10^4	5.6×10^6
Location 4	4×10^4	6.8×10^6
Location 5	1×10^4	1.5×10^6
Location 6	5×10^4	7.5×10^6
Location 7	1.4×10^5	1.8×10^7
Location 8	4.5×10^4	7.1×10^6
Location 9	1×10^6	1.5×10^7
Location 10	1.8×10^5	5×10^7

particles (the solid content of each sample was shown in Supplementary Table S1).

2.5. Sample drying

Two drying methods were evaluated in the present study: evaporation with and without a cover (See Section 3.2). Four samples of influent (150 mL) and four samples of effluent (250 mL) were dried using both methods, in duplicate. For the open drying method, each sample was transferred to a round-bottomed flask and subsequently dried in a drying oven (Carbolite Gero, Derbyshire, United Kingdom) at 60 °C till dryness. The covered drying method was conducted at the same time: a binder-free GF/B glass microfiber filter (1 µm mesh size) (Fisher Scientific, Landsmeer, the Netherlands) was used to cover the round-bottomed flask by clamping it over the opening during drying. After evaporation, the flasks were immediately used for the depolymerization reaction (see Section 2.7). For the two drying methods, LC-grade water was used as the blank sample. The effluents were treated with the same procedure as the influents. To compare the difference between drying with and without a cover, samples from four locations were tested and the analysis for each location was replicated twice.

2.6. Depolymerization reaction in dried wastewater samples

The depolymerization reaction of PET using excess ethylene glycol to yield BHET has been described in our previous study (Tian et al., 2022). In brief, 100 mL ethylene glycol containing 10 mg zinc acetate was added to a round-bottomed flask containing the dried wastewater samples (or dried blank samples) described in Section 2.6. After the reaction, the mixture was cooled down to room temperature and spiked with BHET-D₄ (internal standard) to yield a BHET-D₄ concentration of 0.3 mg/L. Subsequently, the mixture was filtered through a 0.2 µm polytetrafluoroethylene (PTFE) filter (Fisher Scientific, the Netherlands) directly into 2 mL glass LC vials and stored at 4 °C until LC-MS analysis.

An excess amount of ethylene glycol was used to ensure the full depolymerization of PET (Tian et al., 2022). The efficiency of the depolymerization reaction for PET MNP in the wastewater sample matrix was evaluated using a wastewater effluent sample (from location 9, which showed a minor variation in PET MNP content among each aliquot in our pre-experiment test) and a PET analytical standard. In brief, 250 mL of the effluent was spiked with 0.2 mg PET analytical standard and dried using the open drying method (Section 2.6). Then the depolymerization was conducted following the procedure mentioned above (Section 2.7). Another 250-mL effluent sample (from location 9) without added PET was used as a blank. As a mass-labeled PET standard is not commercially available and the wastewater matrix is not homogeneous, the recovery result of PET MNP in wastewater matrix can be slightly overestimated or underestimated. The efficiency test was performed in triplicate and the average recovery is reported in Section 3.1.

2.7. Instrumental analysis

Sample extracts were analyzed using a Bruker Elute LC system coupled with a Bruker EVOQ triple quadrupole mass spectrometer equipped with an atmospheric pressure chemical ionization (APCI) source. The LC separation was conducted on a Poroshell 120 Phenyl Hexyl column (Agilent Technologies; 2.7 µm × 3.0 mm × 100 mm) fitted with a Poroshell 120 Phenyl Hexyl guard column (Agilent Technologies; 2.7 µm × 3.0 mm × 10 mm). The LC gradients started from 90 % water (solvent A) and 10 % ethanol (solvent B). The percentage of solvent B increased linearly to 100 % at 4.75 min, and maintained at 100 % during 4.75–10 min; at 10.1 min, the eluent was restored to the initial conditions for 2 min to re-equilibrate the column for the next injection. Samples were analyzed under APCI negative mode at a probe temperature of 600 °C. The acquisition MS method was multiple reaction monitoring. The quantifiers for BHET are 209 *m/z* and 165 *m/z*, and the quantifiers for BHET-D₄ are 212.9 *m/z* and 169.2 *m/z*.

2.8. Instrument performance validation

To validate the instrument performance, a set of calibration standards (5 µg/L to 500 µg/L of BHET, 300 µg/L for BHET-D₄) was assessed. The linearity of the instrument response was assessed using the response of the calibration standards prepared in ethylene glycol. The instrumental limit of detection (LOD) was determined as the concentration of the target analyte in ethylene glycol that yielded a peak signal-to-noise ratio of 3. The limit of quantification (LOQ) was determined as the concentration of the target analyte in ethylene glycol that yielded a peak signal-to-noise ratio of 10. The relative standard deviation (RSD) for the inter-day precision was calculated based on the analysis of three replicates of standard sample on different days. An inter-day precision (RSD) lower than 15 % was deemed acceptable.

2.9. Calculations

The PET amounts in environmental samples were calculated through the analytical quantification of BHET as described in Tian et al. (2022). A *t*-test was used to evaluate the differences between two groups (drying with and without a cover) (IBM® SPSS® statistics 24).

3. Results and discussion

3.1. Instrument performance validation

The instrument responses for BHET were linear, with the coefficient of determination (*r*²) higher than 0.99. The inter-day precision (RSD) for LC-MS analysis was below 10.1 %. The LOD for PET in influents and effluents was 2.0 µg/L and 1.2 µg/L, respectively. The LOQ for PET in influents and effluents was 6.7 µg/L and 4.0 µg/L, respectively. The LOD and LOQ of PET were calculated based on the BHET quantified in wastewater sample (Tian et al., 2022). The depolymerization reaction yielded a mean recovery higher than 98 % for the spike test in wastewater matrix (Supplementary Table S2).

No macro-particles were visually observed in the sieving step (Section 2.5). TPA was not detected in solvent blanks nor in procedural blanks. BHET was not detected in solvent blanks but was detected in several procedural blanks (at very low levels, not detected-2 µg/L), and in those cases, the blank value was subtracted in the quantification calculation (see Section 3.3).

3.2. Quantification of PET MNP using two drying methods

The sample pretreatment process required separation (e.g. sieving, filtration or evaporation) and extraction/digestion (Zhang and Chen, 2020; Xu et al., 2020). Filtration has been widely applied when studying microplastics in wastewater and sludges (Lusher et al., 2017; Lares et al., 2019; Zhang and Chen, 2020). Lares et al. (2019) concluded that filtration is the best method compared to the alternatives (wet peroxide oxidation, density separation, drying, digestion, etc.) for the separation of multiple types of microplastics with high recovery and reduced processing time. However, the filtration method led to low recoveries (23.3–63.3 %) for PET microfibers (Lares et al., 2019). As fiber is the main shape of microplastics observed in wastewater (Li et al., 2018; Xu et al., 2019), drying is preferred in the present study.

The quantification of PET MNP in samples after using the open and covered drying methods is shown in Table 2. Eight wastewater samples from 4 locations were analyzed to compare the two methods. The results show that there is no significant difference between drying with and without a cover (*p* > 0.1). A similar result was also reported by Lares et al. (2019), who suggested that drying in an oven using an open flask can accelerate the drying procedure without a significant impact on the microplastics recovery. This is of course dependent on the quality of air in the laboratory and time period exposed to the air, factors which are checked when using procedural

Table 2
Measured concentrations of PET MNP using different drying methods ($\mu\text{g/L}$).

Samples	Replicate	Dried without a cover	Dried with a cover
Influent (location 1)	1	680	670
	2	540	586
Influent (location 4)	1	48.9	50.3
	2	73.5	59.1
Influent (location 9)	1	136	128
	2	142	111
Effluent (location 5)	1	23.1	20.1
	2	15.8	11.9

blank samples that have gone through all pretreatment, drying and analysis steps of the method.

3.3. PET in influents and effluents

The PET MNP quantification results for influents and effluents from different sampling locations are shown in Table 3 (Fig. S2). The results in Tables 2 and 3 demonstrate that, for most of the cases, the difference in the level of PET MNP between each replication is not big. Thus, the results in Table 3 can, to some extent, reflect the mass level range of PET MNP in the sampling location at the sampling time point.

PET MNP was detected in all the influent samples, with the concentrations ranging from 24.9 $\mu\text{g/L}$ to 680 $\mu\text{g/L}$. The concentrations of PET MNP in effluents range from <LOD to 23.1 $\mu\text{g/L}$. The results suggest that waste water treatment can remove a large portion of PET (approximately between 70 % and 100 %), which is in accordance with the information reported in literature (up to 100 % removal) (Jiang et al., 2020; Edo et al., 2020).

Numerous publications have reported the occurrence and the particle concentration (number) of PET microplastics in influents and effluents from WWTP (Edo et al., 2020; Leslie et al., 2017; Jiang et al., 2020; Xu et al., 2020), but the mass of PET MNP in influents and effluents has not been widely studied (Okoffo et al., 2020; Okoffo et al., 2022). The concentrations of PET MNP in effluents are relatively low (Table 3), but once taking into account of the effluent volumes continuously discharged to the environment (Table 1), the amount of PET MNP entering into the aquatic environment from WWTP is not negligible. Additionally, these PET MNP are persistent and therefore accumulate in the environment over time, which may pose health threats to both humans and biota (Napper and Thompson, 2016; Nelms et al., 2018; Fan et al., 2019). Normally when

Table 3
PET MNP concentrations measured in different Dutch WWTP influent and effluent ($\mu\text{g/L}$).^a

Sampling site	Replicate	PET MNP in the influent	PET MNP in the effluent
1	1	680	7.3
	2	539	5.4
2	1	152	<1.2 (LOD)
	2	162	<1.2 (LOD)
3	1	48	2.6
	2	24.9	7.1
4	1	48.9	11.2
	2	73.5	11.9
5	1	208	23.1
	2	185	15.8
6	1	442	3.4
	2	209	2.3
7	1	348	18.2
	2	370	7.2
8	1	68	12.1
	2	70.1	9.2
9	1	136	12
	2	142	14.3
10	1	87.8	<1.2 (LOD)
	2	81.7	<1.2 (LOD)

^a Values were corrected for the procedural blank.

introducing materials or chemicals into society research is performed to confirm if there are health and/or environmental risks related to these products. Initially PET microfibers were not taken into account as a product of the use of PET, which means that the adverse health effects were not widely studied (Leslie and Depledge, 2020).

3.4. Perspectives for future research

There are still limitations and knowledge gaps in the quantification of MNP, especially for nanoplastics. Firstly, there is no standard protocol for the identification and mass quantification of nanoplastics, and the data on their occurrence in the environment is limited. The present study successfully quantified PET MNP in wastewater using a simple and quick method and this method has the potential to be applied for PET MNP monitoring in wastewater as it can provide quantitative mass based concentrations without the need for a high-end instrument. A similar experimental procedure could possibly be applied for quantifying other condensation polymers (e.g. polycarbonates, polyamides such as nylon-6 and nylon-6.6).

Secondly, there is a lack of automated tools for nanoplastic separation due to its tiny size (<1 μm) and variability in size (between 1 nm and 1 μm). Several studies have reported good recoveries (>85 %) for nanoplastics using filtration (e.g. membrane filter) and extraction (e.g. cloud-point extraction) methods (Ter Halle et al., 2017; Zhou et al., 2018; Cai et al., 2021). Similar studies in literature reported the separation method that can separate nanoplastics with sizes as small as 25 nm, but not smaller (Cai et al., 2021). In this case, by quantifying the total MNP and deducting the microplastic quantity may be a way to quantify the total nanoplastics. However, for mass-based detection methods, a 25-nm PET particle size would have to be present in large numbers to reach mass detection limits, considering a PET sphere of that size (and density of 1.38 g/cm^3) has a mass of only around 11 atto-grams. Probably, using pooled samples or high sample volumes can be a way to overcome the mass detection limit issue.

Efforts are still needed to further develop quantification methods for other types of MNP, especially for polyolefins such as polyethylene, polypropylene, polyvinylchloride and polystyrene. Quantification of MNP by mass is particularly useful when assessing removal efficiency in a WWTP because a particle numbers may change during the treatment process (e.g. fibers and fragments may break during processing), in ways that are not linearly related to the polymer mass present in influents and effluents. Overall, the MNP quantification approaches for mass and particle number analysis can complement each other in the quest to gain a full understanding of the dynamics of MNP pollution and improve risk assessment.

4. Conclusions

PET MNP was detected in all the influents and most of the effluents from WWTPs in this study, indicating that WWTPs are a point source of PET MNP in the environment. Results of the present study revealed that the presented quantification method is simple and effective and can be applied for monitoring the PET MNP level in wastewater. The reported experimental procedure shows the potential to be applied for the MNP quantification of other condensation polymers.

CRediT authorship contribution statement

Lei Tian: Data curation, Writing – original draft. **Ewa Skoczynska:** Data curation, Writing – original draft. **Robert-Jan van Putten:** Supervision, Writing – review & editing. **Heather A. Leslie:** Writing – review & editing. **Gert-Jan M. Gruter:** Funding acquisition, Supervision, Writing – review & editing.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Lei Tian and Ewa Skoczynska report financial support by the Dutch Research Council and Avantium Support BV.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.159209>.

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