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# Microplastics in urban and highway stormwater retention ponds

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HIGHLIGHTS

- An assessment of MP in stormwater retention ponds across various landscapes.
- Commercial and industrial areas emitted more MP than residential and highway areas.
- PVC, PS, PP, PE and polyester were the most common polymers found.
- PVC had the largest particle size and mass.

GRAPHICAL ABSTRACT
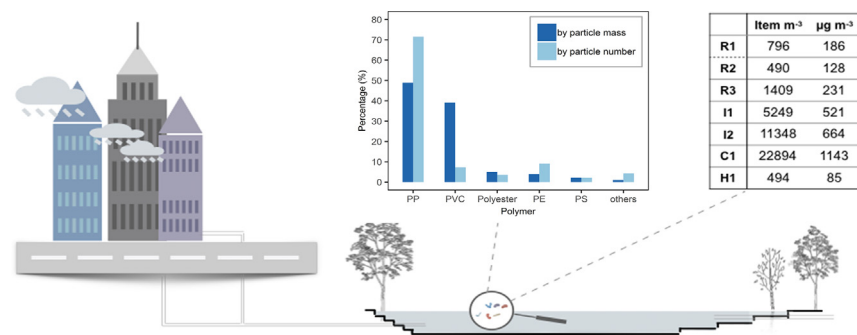
ARTICLE INFO

ABSTRACT

Urban and highway stormwater runoff seems an obvious pathway for conveying microplastics from land-based sources to the aquatic environment. The significance of this pathway is however largely unknown. This study presents first results on microplastics in urban and highway stormwaters by analysing microplastics (10–2000 μm) in the water phase of stormwater treatment ponds in the North of Jutland, Denmark. The stormwater of seven ponds treating runoff drained from different landscapes were studied. Ponds serving highway and residential areas had the lowest microplastic concentration, while ponds serving areas with industry and commerce had the highest. The stormwater of the ponds contained 490–22,894 items m^{-3}, corresponding to an estimated 85–1143 μg m^{-3}. The dominating polymers were polypropylene, polyvinylchloride, polyester, polyethylene and polystyrene. There was a tendency towards polyvinylchloride particles being the largest in size, while a mixed group of less common polymers were the smallest. Residential ponds generally held the largest microplastic particles and were in many aspects quite comparable to one another, in terms of concentrations, sizes, and polymer composition. This study shows that land-based sources are significant contributors to MP in the environment. It demonstrates that the stormwater retention ponds act not only as pollution hotspots, but also play a role in the transport of MP from land to the aquatic environment.

1. Introduction

Plastic pollution is highly visible in the environment, and an emerging topic on the public, scientific, and political agenda. Reports on marine plastic pollution and concerns about its impact on marine vertebrates (Thompson et al., 2004; Lusher et al., 2013; Marques et al., 2018) have driven the awareness of the plastic-issue. Microplastics (MP), most frequently defined as plastic particles <5 mm in length (Arthur et al., 2009), have been found throughout the marine environment, including the gut of fish (Beer et al., 2017), zooplankton (Cole et al., 2013), as well as deposit and filter feeders (Van Cauwenberghe et al., 2015). Negative

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effects have been documented at the individual level in copepods (Cole et al., 2015), worms (Wesch et al., 2017), and bivalves (Bour et al., 2018). Meanwhile, concerns regarding human exposure through seafood (Van Cauwenberghe and Janssen, 2014; Lusher et al., 2017) and potable water (Pivokonsky et al., 2018) have also been raised.

The MP originate from sea-based and land-based sources, where urban areas are identified as important contributors of MP into the environment (Horton et al., 2017). The discharge of MP from urban wastewater treatment plants (WWTPs) has been studied in quite some detail (Mintenig et al., 2017; Talvitie et al., 2017; Simon et al., 2018). However, advanced WWTPs that include carbon and nutrient removal are efficient at retaining MP, and their discharge seems only to explain part of the MP in the receiving waters. For example did Simon et al. (2018) find that Danish WWTPs released on average 0.56 g of MP per capita and year, which does not seem to explain the occurrence of MP in Danish waters. For a British watershed, Kay et al. (2018) reported that the studied WWTPs led to elevated levels of MP, but that other sources also played an important role in explaining the detected levels of MP contamination. Estahbanati and Fahrenfeld (2016) measured MP upstream and downstream of four municipal WWTPs and found that downstream MP levels were elevated. However, the waters also contained MP at the upstream locations which, was contributed by other sources. Lasee et al. (2017) studied MP in three connected urban lakes receiving treated wastewater. Their results indicated that the treated wastewater alone could not explain the levels of MP and they reasoned that urban stormwater runoff must also have contributed to the levels in the lakes. Recent work has also shown that the ingestion of MP by river invertebrates downstream of WWTPs was not significantly higher than upstream (Windsor et al., 2019a). Sutton et al. (2016) drew a similar conclusion when investigating MP in the San Francisco Bay, concluding that there must have been other sources for MP than the treated wastewater, and suggested stormwater runoff as one of these. Horton et al. (2017) also presented data indicating that stormwater runoff might contribute to MP in the environment. They observed fragments of thermoplastic road markings downstream of a storm drain outfall and attributed these to road runoff.

Urban and highway stormwater runoff are direct pathways for land-based microplastics into freshwaters (Horton et al., 2017; Vaughan et al., 2017), which have been pointed out to be important in terms of microplastics impacts and pathways for emissions to the oceans. At the same time, they have been rather under-studied and only to a limited degree pinned down in terms of actual measurements (Rochman, 2018). Blettler et al. (2018) also identified freshwater environments as an area with little data and a need for investigations covering plastics in all size fractions and pointed out that the large rivers in countries with rapid growth and poorly managed waste have been especially overlooked. For the freshwater systems of the developed world, there is some data on MP in river systems (Liedermann et al., 2018; Rodrigues et al., 2018) and lakes (Hendrickson et al., 2018; Sighicelli et al., 2018) showing that MP are also ubiquitous in these waters. All these observations indicate that stormwater runoff is important for the conveyance of MP into the aquatic environment, but that the understanding of magnitudes and processes remains poor, as the field-based measurements are limited (Windsor et al., 2019b).

One issue in pinning down the sources of MP contamination is the use of widely different sampling approaches and analytical methods, combined with different MP size-ranges addressed. Furthermore, the vast majority of the published studies refer to MP in terms of particle number within some size range (Löder et al., 2017). Such data are highly important when assessing the environmental impacts of MP, but insufficient when assessing the load of MP on the environment or its fate therein. Here, the mass of the MP must also be quantified. This creates some analytical problems. The spectroscopic methods typically applied for MP analysis are well suited for the quantification of size and shape of MP particles. The current state of the science, they do however only allow MP mass to be estimated (Simon et al., 2018).

A further issue when quantifying MP in stormwater discharge from urban areas is the difficulty in obtaining a representative sample during a storm event. All cities drain stormwater by natural and constructed systems, where the latter typically is a piped drainage system. Some of the drainage systems are combined – where domestic and industrial wastewater and stormwater are conveyed in the same pipe, while others are separated – where one pipe conveys wastewater and another stormwater. Contrary to dry-weather WWTP discharges, the discharges during rain events are of intermittent nature. A combined drainage system, for example, only discharges when its hydraulic capacity is exceeded during a large storm event. A separate storm sewer, on the other hand, discharges at any rain event above a certain initial hydrological loss. The general pollutant concentration in the stormwater runoff is known to vary substantially between sites, and also between events at the same site (Bertrand-Krajewski et al., 1998). Furthermore, part of the stormwater of a modern city is not discharged directly, but undergoes some sort of treatment, often in artificial basins, called retention ponds, that mimic the natural treatment processes of small freshwater lakes (Hvitved-Jacobsen et al., 2010). Here, the stormwater stays for a period of days to weeks, allowing many of the particulates to settle, upon which part of the treated water is discharged by displacement during the following rain event.

The objective of the present study is to quantify microplastics in stormwater from retention ponds, partly because these have many similarities to small natural lakes subject to high pollution loads, and partly because stormwater is an important pathway for MP into the aquatic environment. Seven ponds were chosen as having different catchment characteristics, in order to evaluate whether the land use in the catchment influenced the amount of MP in the water of the ponds.

2. Material and methods

2.1. Sampling

Stormwater was sampled from September to October 2017 from seven retention ponds in Denmark (Table 1) – three ponds with residential catchments (R1, R2, R3), two with industrial ones (I1, I2), one with a commercial catchment (C1), and one with a highway catchment (H1). Three sampling rounds were performed during dry weather, with at least two weeks between them. Dry weather was defined as no rain occurring at least two days prior to the sampling. Samples were collected using a filtering device equipped with a removable filter cut from a 10 μm stainless steel mesh (100 mm in diameter) (Fig. S1). Water was pumped through the device by a positive displacement pump (Creusen Roermond, Netherlands). The filtering device was made in stainless steel, with the inflow and outflow pipes made in PVC. The suction hose was connected to a cuboid cage ($10 \times 10 \times 20$ cm) made of a 2 cm stainless steel mesh to filter out large debris. The flow was logged with an electronic flowmeter (IFM SM7000, Germany). When sampling, the suction hose was held by a floater approx. 20 cm below the water surface. The cage was placed in the pond approx. 5–10 m from the shoreline, and the corresponding water depth was approx. 1 m. For each sampling of a pond, water was filtered until the filter clogged, which typically happened after 201 to 454 L. For each pond, the filters from the three sampling rounds were lumped and analysed as one sample. The total volumes sampled per pond are given in Table 1. The filters were stored in glass jars and soaked in a sodium dodecyl sulphate solution (SDS, 0.15 g L^{-1}) immediately after use. After transferring to the lab, filters from the same pond were combined into one beaker and soaked with 500 mL of the SDS solution in order to detach MP from other solids, hence to open up the surface space for subsequent MP extraction.

2.2. Sample processing

Samples were wet-oxidised on the filters for 2 days by adding 50 mL of 50% H_2O_2 to the 500 mL of SDS-solution holding the filters. The H_2O_2 was hence diluted in the final solution, to minimize the potential of

Table 1
Pond locations, catchment characteristics and sample volumes.

Site	City	Coordinate (N, E)	Pond area (m²)	Impervious catchment area (ha)	Sample volume (L)	Catchment type
R1	Silkeborg	(56°11′39.8″, 9°32′57.9″)	4000	7.1	1016	Residential with single family houses and part of an orbital road
R2	Aarhus	(56°10′18.4″, 10°05′49.9″)	11,880	42.6	1139	Residential with single family houses
R3	Aarhus	(56°10′18.4″, 10°06′22.6″)	11,900	46.6	958	Residential with single family houses
I1	Aarhus	(56°10′49.0″, 10°07′58.4″)	7460	10.9	722	Light industry
I2	Viborg	(56°28′29.3″, 9°24′43.3″)	6500	69.8	1078	Light industry, commerce, do-it-yourself shops
C1	Aarhus	(56°08′41.8″, 10°08′12.2″)	6050	19.0	850	Commercial (shopping centre), some residential use
H1	Aarhus	(56°13′13.9″, 10°07′43.5″)	5540	4.8	979	Highway

polymer degradation (Hurley et al., 2018). This step partly removed some organic matter, and partly opened up the matrix for a subsequent enzymatic digestion (Enders et al., 2015). Particles were removed from the filters by ultrasonic treatment and collected into 200 mL of SDS solution. The samples were incubated with the enzymes Cellubrix (500 μL, Sigma-Aldrich) and Viscozyme (350 μL, Sigma-Aldrich) at 50 °C for 3 days, upon which Alcalase (500 μL, Novozymes) was added and the solution incubated for another 3 days, also at 50 °C. Finally, the sample underwent a Fenton reaction by adding 146 mL 50% H_2O_2, 63 mL of 0.1 M $FeSO_4$ and 65 mL of 0.1 M NaOH to further remove organic matter (Simon et al., 2018). As the process is exothermic, the vessel was kept on ice, targeting a temperature of 15–19 °C. Lares et al. (2018) carried out a study where MP and fibers were successfully extracted from wastewater through a rigorous oxidation treatment (30% H_2O_2 with catalyst, followed by an enzyme treatment). The present study applied relatively lower concentrated reagents, therefore the degradation of MP and fibers were neglected. After 4 h, the reaction had slowed down sufficiently and the vessel was left for 2 days at room temperature. The samples were filtered on 10 μm steel filters (47 mm in diameter), the filters transferred to a 50 mL beaker with a 1.7 g cm^{-3} zinc chloride solution, removed from the filters by ultrasonic treatment and transferred to a 200 mL funnel for density separation. The funnel was topped off with zinc chloride solution.

The sample underwent density separation twice. The first time the liquid was left to settle for 2 h and the second time it was left overnight. Settled particles were drained off and the remaining liquid was filtered through a 10 μm stainless steel filter. Collected particles were transferred into HPLC grade ethanol using ultrasonic treatment. The suspension was transferred into a glass vial and evaporated until it was dry by flushing with N_2 (N5.0). Five mL of 50% ethanol was added for quantification by μFTIR imaging (Fourier-transform infrared spectroscopy). Initial tests had shown that such a solution minimized particles sticking to the inside of the glass vial.

2.3. MP identification and quantification

The suspension was homogenised and sub-samples were deposited onto a 13 mm diameter zinc selenide transmission window of 2 mm thickness, using disposable glass capillary pipettes. The surface of the window was restricted by a compression cell (Pike Technologies, USA) to 10 mm diameter. The windows were covered with a glass beaker and dried at 55 °C. Depositing a too large sample volume on a window would impede the subsequent interpretation of the data obtained by the μFTIR imaging. Sub-samples were hence deposited and dried in increments of 100 μL until a suitable amount was reached. The latter was verified under a microscope. For each sample, three individual windows were prepared and analysed, corresponding to analysing 6–12% of the entire sample volume. The final MP concentration of a sample was determined from the sum of MP found on the three windows.

The whole of all 10 mm diameter windows were scanned with an FPA-based (Focal Plane Array) FTIR imaging technique at 5.5 μm pixel resolution (μFTIR imaging). This resulted in three IR-maps of approx. 3.2 million spectra each, as the equipment scans rectangular areas. The equipment was a Cary 620 FTIR microscope coupled with a Cary

670 IR spectroscopy (Agilent). The microscope used a 15× Cassegrain objective and had a 128 × 128 Mercury Cadmium Telluride (MCT) FPA detector. The scans were performed in transmission mode in the spectral range of 3750–950 cm^{-1} at 8 cm^{-1} resolution, applying 30 co-added scans (Simon et al., 2018).

The maps from the μFTIR imaging were analysed using MPhunter, a software for the automated detection of MP from μFTIR chemical imaging datasets developed at Aalborg University, Denmark, in collaboration with Alfred Wegener Institute, Germany. MPhunter builds upon the work of Primpke et al. (2017) and compares the IR-spectrum of each map pixel with the spectra of a reference database and assigns a score to the quality of each match. The algorithm generating the score correlates the raw spectra, their 1st derivatives and their 2nd derivatives by a Pearson correlation, yielding three Pearson's correlation coefficients (r_0, r_1, r_2) per combination of map spectrum and reference spectrum. The r-values are squared, and the user assigns them global weights (k_0, k_1, k_2). A score between 0 and 1 is calculated, $S_{d(i,j)}$, where (i,j) are the coordinates of the pixel on the map and d the number of the reference spectrum (Eq. (1)). Negative correlations are omitted, i.e. if an r-value is below zero, it is set to zero.

$$S_{d(i,j)} = \frac{k_0 r_0^2 + k_1 r_1^2 + k_2 r_2^2}{k_1 + k_2 + k_3} \tag{1}$$

The analysis yields a correlation map per reference spectrum in the database. The applied database contained 113 reference spectra of plastics as well as natural organic materials that can be misinterpreted as plastics. The spectra were organised into material groups covering the plastics: ABS (acrylonitrile butadiene styrene), acrylic, acrylic paints, alkyd, aramid, cellulose acetate, diene elastomer, EPDM (ethylene propylene diene monomers), epoxy, EVA (ethylene vinyl acetate), PA (polyamide), PAN Acrylic fibre (polyacrylonitrile), PE (polyethylene), Pebax®, PEG (polyethylene glycol), phenoxy resin, PLA (polylactic acid), polycarbonate, polyester, POM (polyoxymethylene), PP (polypropylene), PS (polystyrene), PTFE (polytetrafluoroethylene), PU (polyurethane), PU paints (polyurethane paints), PVA (polyvinyl alcohol), PVAC (polyvinyl acetate), PVC (polyvinyl chloride), SAN (styrene acrylonitrile), SBR (styrene butadiene rubber) and vinyl copolymer. Even though SBR was included, this does not mean that car tyre rubber containing SBR could be quantified as FTIR in transmission or reflection mode is not able to determine tyre material, due to the carbon black added to the tyre material as a filler (Kole et al., 2017). Carbon black is absorbent throughout the infrared region, making the acquisition of transmission or reflection spectra problematic. The included non-plastic groups were cellulose and proteinaceous material.

MPhunter allocates particles to a material group through an algorithm based on score thresholds, which were set prior to the analysis by manually assessing which thresholds produced trustworthy particle recognition. Applying MPhunter, the user first sets three thresholds for each reference spectrum ($t_1 > t_2 > t_3$). To qualify as a particle belonging to a material, at least one pixel must have a score of $S_{d(i,j)} \geq t_1$. If several materials have a score above their respective t_1 value, the pixel is associated with the material with the highest score. As an example, if a particle exceeds both the threshold of polyamide and proteinaceous

materials, but has a score for proteinaceous materials that is higher than that for polyamide, then that particle is associated with proteinaceous materials.

The so-selected pixel $S_{d(i,j)}$ is used as the nucleus of a particle, where both MP and natural particles are identified. The algorithm develops the particle by evaluating the adjacent pixels (Eq. (2)). If any of them have a score of $S_d \geq t_2$ for any reference spectrum belonging to the same material group as the nucleus pixel, it becomes associated with the particle created around it. After having developed the particle around $S_{d(i,j)}$, the procedure is repeated around all pixels that have been added to the original particle nucleus. The above approach can give rise to particle sizes from one pixel upwards. However, practical experience has shown that particles of only one pixel tend to be artefacts. Hence, a minimum particle size of 3 pixels was set. The third threshold is solely used to make the dataset more manageable, where any correlation below t_3 is ignored in the visualisation. Setting the threshold t_1 low will increase the number of false positive detections but decrease the number of false negative detections. Setting it high will have the opposite effect. Setting t_2 low will increase the size of the particle, setting it high will decrease it.

$$\begin{bmatrix} S_{d(i-1,j+1)} & S_{d(i,j+1)} & S_{d(i+1,j+1)} \\ S_{d(i-1,j)} & S_{d(i,j)} \geq t_1 & S_{d(i+1,j)} \\ S_{d(i-1,j-1)} & S_{d(i,j-1)} & S_{d(i+1,j-1)} \end{bmatrix} \tag{2}$$

A so-constructed particle is characterised by a major dimension and a minor dimension. The major dimension was calculated as the longest linear distance between pixels constituting the particle. The minor dimension was estimated from the area of the particle and its major dimension – assuming an elliptical shape. The volume is estimated by assuming the particle is an ellipsoid with its third dimension being 0.6 times its minor dimension (Simon et al., 2018). The particle mass is calculated from the estimated volume and density of the polymer. Further details and graphics on MPhunter can be found in supplementary materials, S1.

Applying an automated approach to identifying MP from μFTIR imaging datasets is vastly superior to using the alternative: a manual checking of each of the thousands of possible particles of a scanned sample (Primpke et al., 2017). Not only is this approach much less time consuming, it also reduces the risk of overlooking particles as every single pixel of a scan is analysed, not just visually selected particles. However, the approach is not free of human bias, as the operator has to set the thresholds for the analysis as well as select the reference spectra dataset – the latter being quite critical when identifying MP (Primpke et al., 2018). During the development of MPhunter, the program was validated by analysis of MP standards created from pure polymers. For the present study, the identification of MP in real samples was evaluated to take into account that real MP might behave differently from pure polymer standards, as well as to take into account possible matrix effects. This was done by first setting the threshold t_1 rather low and manually walking through a limited number of particles belonging to the targeted material groups. If the automated detection caused a significant number of false positive detections, the threshold was increased until the number of false positive and false negative detections were minimized. Secondly, the threshold t_2 was adjusted by walking through the boundaries of a limited number of particles to evaluate if the particle boundaries were well identified. In this process, the grouping of reference spectra into polymer groups was also adjusted. For MP where the μFTIR imaging revealed a major dimension >500 μm, the actual size of the MP was further confirmed using the visual image. Large particles can be of substantial thickness, leading to saturation in μFTIR imaging spectra. Visual inspection was needed in these cases to ensure that particle size was not underestimated.

2.4. Experimental quality control

The use of a plastic suction hose (PVC) for the filtering device could not be avoided. Hence, potential contamination during sampling was assessed. To do so, the spectrum of the hose was collected using Attenuated Total Reflectance – FTIR (Cary 630 with a diamond ATR crystal, Agilent) and compared to all particles identified as PVC in the μFTIR imaging of the samples. The spectra comparison was conducted in the OMNIC software (Thermo Fisher, USA). In case the two spectra matched, there was a risk that this PVC particle could have originated from the hose, and it was excluded from the results.

Precautions were taken to reduce contamination during sample processing. Only glass, or Teflon-coated labware was used. Teflon is excluded from the data analysis as this plastic has a density of about 2.2 g cm^{-3}. It would, hence, be removed together with inorganic particles in the density separation. All labware was rinsed with Milli-Q water and steel filters were muffled at 500 °C before use. All open containers were covered with Watch glass dishes (Sigma-Aldrich) immediately after use. To counteract contaminations from air deposition, which may originate from clothes, human skin, or be present in the common dust of laboratories (Woodall et al., 2014; Dris et al., 2017), the air of the μFTIR scanning room was continuously filtered by an air treatment device (Dustbox® Hochleistungsluftreininger, Germany) with a HEPA filter (H14, 7.5 m^2).

However, MP can still occur due to their ubiquitous presence. Therefore, the background contamination from the sample preparation was assessed by three blanks: a total of 100 L filtered Milli-Q water (1.2 μm GF filters) was filtered on three 10 μm steel filters by the previously described sampling device and the filters processed as described for the stormwater samples.

2.5. Statistical analysis

The normality of datasets was tested by a Shapiro-Wilk normality test (Ašmonaite et al., 2018). To check if there were differences on MP sizes among the ponds, a non-parametric Kruskal-Wallis test was applied. If the difference was statistically significant, then a Wilcoxon rank sum test was used afterwards to assess the differences in the pairwise comparison between ponds (Brandon et al., 2016). Principle Component Analysis (PCA) was carried out to estimate the variables which explained most of the variations. All statistical analysis was performed in R (v3.5.1), and the significance level was set to 0.05 in all cases.

3. Results and discussion

3.1. Background contamination

None of the particles identified in the stormwater matched the PVC-material of the sampling hose, meaning that the suction hose did not cause contamination. During sample preparation and analysis the blanks did, however, revealed a slight contamination by PE, PA, polyester, and acrylic MP. For each of the three blanks, a sub-sample of 600 μL was deposited. In these, a total of eight MP were found, where three were of PE, two of PA, two of polyester and one of acrylic. The mass of the eight MP was estimated as 339 ng. The MP contamination could have come from the equipment and surrounding air, but presumably not from the filtered water used for the blanks. For instance, Lin et al. (2018) performed one procedural blank simultaneously throughout their experiment and suggested that lab-coats were the main origin of airborne contamination. Hence, the contamination is believed to be related to the lab processing of the sample, and not the volume filtered. Correspondingly, the contamination was 22.2 items per processed sample, corresponding to an estimated 942 ng per processed sample. On average, 954 L of stormwater was collected per sample, leading to an approximate contamination of 23.3 items m^{-3} or 987 ng m^{-3}. Due to the low level of contamination, and the corresponding statistical uncertainty on the actual numbers, the results were not corrected for background contamination.

Although precautions are commonly taken and researchers are aware of the issue, background contamination is not always assessed and quantified (Browne et al., 2011; Cheung et al., 2016; Kalčíkoá et al., 2017; Di and Wang, 2018; Sighicelli et al., 2018). The contamination level reported in other studies ranged from 0.4 to 2110 items L^{-1}, or 4.4 ± 1.4 items per sample (Mintenig et al., 2017; Talvitie et al., 2017; Lares et al., 2018; Simon et al., 2018). Compared to these numbers, the values of the present study are deemed similar or even lower.

3.2. MP abundance

A total of 3436 particles were identified as MP in the samples from the seven ponds. The lowest value was from H1 with 58 MP, and the highest value was from C1 with 1946 MP. The concentration per pond ranged from 490 to 22,894 item m^{-3} and 85 to 1143 µg m^{-3}, with a median value between all ponds of 1409 (interquartile range (IQR) = 11,348–494) item m^{-3} and 231 (IQR = 664–128) µg m^{-3} (Table 2). Comparing to the contamination of the blanks, it is seen that H1 was close to the level of contamination (58 MP found in the analysed sample and 22.2 MP in the blanks). For the other ponds, the contamination was deemed acceptable or low (Table 2). Normality tests showed that the MP mass concentrations (µg m^{-3}) were normally distributed between the seven ponds (p = 0.144) but not the number concentrations (item m^{-3}) (p = 0.014). Hence, the overall concentration of MP in the ponds was described by the median value with an IQR (interquartile range) both for MP mass and number.

MP were detected in all ponds. The commercial pond (C1) had the highest concentration in terms of both number and estimated mass (22,894 item m^{-3} and 1143 µg m^{-3}), while the lowest numbers were found in R2 (490 item m^{-3}) and the lowest mass in H1 (85 µg m^{-3}). The residential ponds (R1, R2 and R3) had an average MP concentration of 898 ± 468 item m^{-3} and 182 ± 52 µg m^{-3}, while the industrial ponds (I1 and I2) had an average MP concentration of 8299 ± 4313 item m^{-3} and 593 ± 101 µg m^{-3}.

As this study is among the first peer-reviewed study to have reported MP in stormwater ponds, so the obtained results were compared the data to other measurements in freshwater systems and treated wastewater. However, due to a lack of generally accepted definition of the sizes making up the analysed MP (Hartmann et al., 2019), together with the different sampling methods and analytical techniques applied, the comparison is challenging. A typical example is a study from Rodrigues et al. (2018), where the MP number concentration (58–1265 item m^{-3}) was comparable with that of the present study, but the mass concentration was over 10–100 orders of magnitude higher (5–51.7 mg m^{-3}). This could be attributed to the larger size range they used (55–5000 µm), resulting larger particles tend to be found. With a more similar approach in the sampling and MP extraction process, the MP concentration in the present study was lower than that reported in treated wastewater (Simon et al., 2018), where 54,000 item m^{-3} and 4200 µg m^{-3} as the average value was found in the outlet of ten Danish wastewater treatment plants.

3.3. Overall polymer composition

Lumping together all data from the retention ponds, the MP could be allocated to 17 polymer groups: PE, PS, PVC, PP, PA, PU, polyester, epoxy,

PVAC, acrylic, SAN, vinyl copolymer, EVA, PVA, phenoxy resin, alkyd and acrylic paints. Some of these were only sparsely detected throughout the samples and were gathered into a group of "others" (PA, PU, epoxy, PVAC, acrylic, SAN, vinyl copolymer, EVA, PVA, phenoxy resin, alkyd and acrylic paints). The group of "others" contained 148 MP, equivalent to 4.3% of the total number of MP identified. Further details on MP in the polymer groups is given in Table S2.

PP was the dominant polymer both by mass and number (Fig. 1). When ranked according to number concentrations, the order of polymers was PP, PE, PVC, polyester and PS (71.5%, 9.1%, 7.4%, 5.5%, and 2.2%, respectively). When ranked according to mass concentrations, the order changed to PP, PVC, polyester, PE, and PS (48.9%, 39.1%, 5.0%, 3.9%, and 2.2%, respectively). The difference in the abundance of polymers when measured as number and mass concentrations is related to the difference in polymer density and particle size distributions. The most pronounced example is PVC versus PE. Although PVC compared to PE was not high in terms of particle numbers, its higher density and a tendency towards larger particles (Fig. 3B) led to its mass concentration being significantly higher than that of PE (Fig. 1).

The overall polymer composition in this study was comparable with other freshwater-related studies, for instance, Rodrigues et al. (2018) found PP, PE, and PS made up 68% of the MP counted in water and sediments from the Antuã River. Sighicelli et al. (2018) reported that PE, EPS (expanded polystyrene), and PP dominated (in total 78% in terms of MP counts) in the surface waters of Italian Subalpine lakes. PVC was also detected in the coastal waters of China by Qu et al. (2018), but at a much lower concentration in terms of MP number (2%). The occurrence of these polymers was consistent with the polymers demanded by market, where PP, PVC, Polyester, PE, and PS together made up more than half of the plastics produced (PlasticsEurope, 2017).

The polymer distribution differed significantly between ponds. For example was C1 dominated by PP with both mass and number concentrations above 90%, while other ponds showed much less PP (Fig. 2); PVC dominated in R1, R2, R3, and I1 in terms of mass, and polyester was abundant in H1. While the cause of these differences is unknown, it seems reasonable to assume that it has something to do with the land use in the catchment. C1 holds a large shopping centre where the handling of plastic wrappings, food packaging and littering might be an important source (PlasticsEurope, 2017). This pond is, furthermore, known to receive significant amounts of illicitly connected wastewater, which also could be a contributing factor. It is also noteworthy that C1 was the pond holding the most MP (Table 2). I2 held the second-highest concentration. Its catchment also holds many retail outlets, but of a somewhat different nature (do-it-yourself shops), which might lead to similar issues regarding plastic pollution, albeit with a
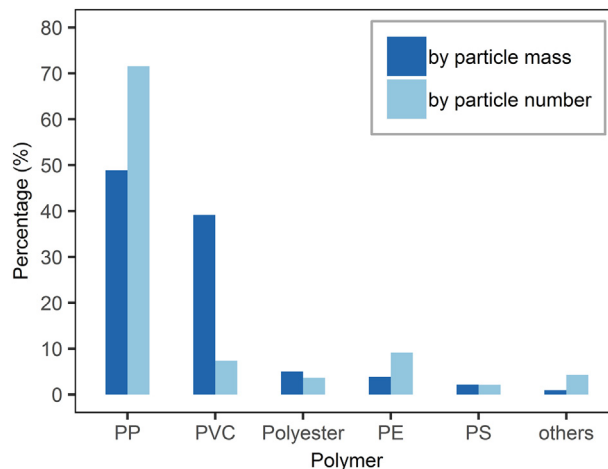
Table 2
The abundance of MP in the water phase from seven stormwater retention ponds in terms of counted MP number and estimated MP mass.

	R1	R2	R3	I1	I2	C1	H1	Average
Item m^{-3}	796	490	1409	5249	11,348	22,894	494	1409 (IQR = 11,348–494)
µg m^{-3}	186	128	231	521	664	1143	85	231 (IQR = 664–128)

Fig. 1. Polymer composition of the main polymers detected in terms of the relative MP mass and MP number concentrations.
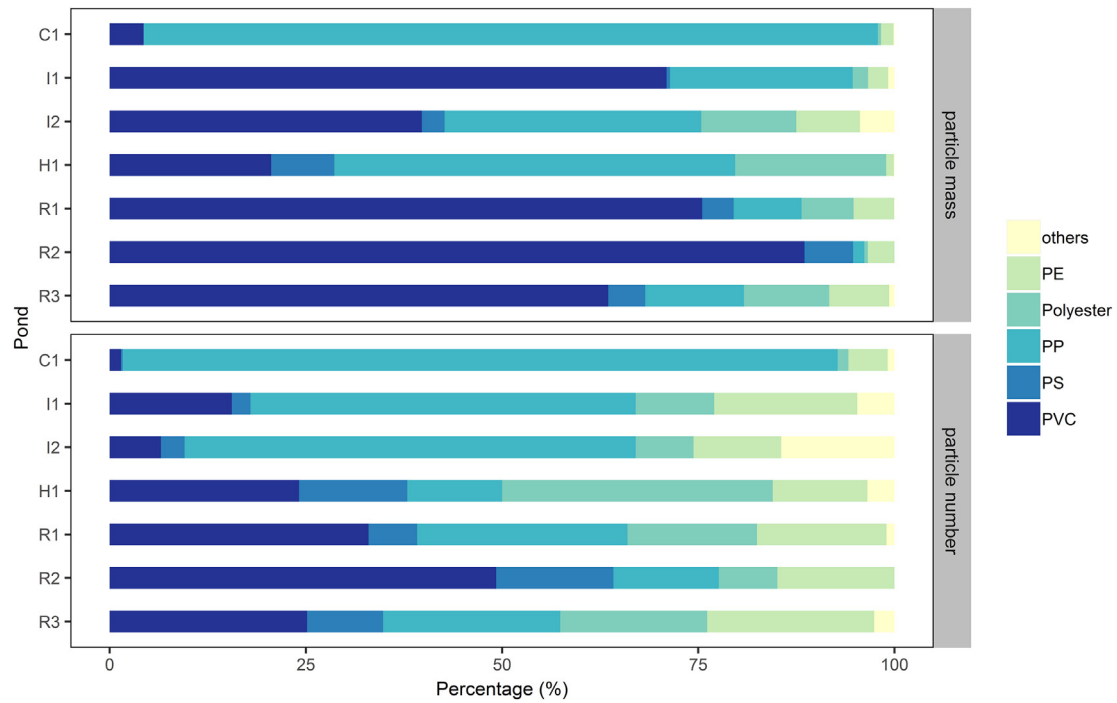
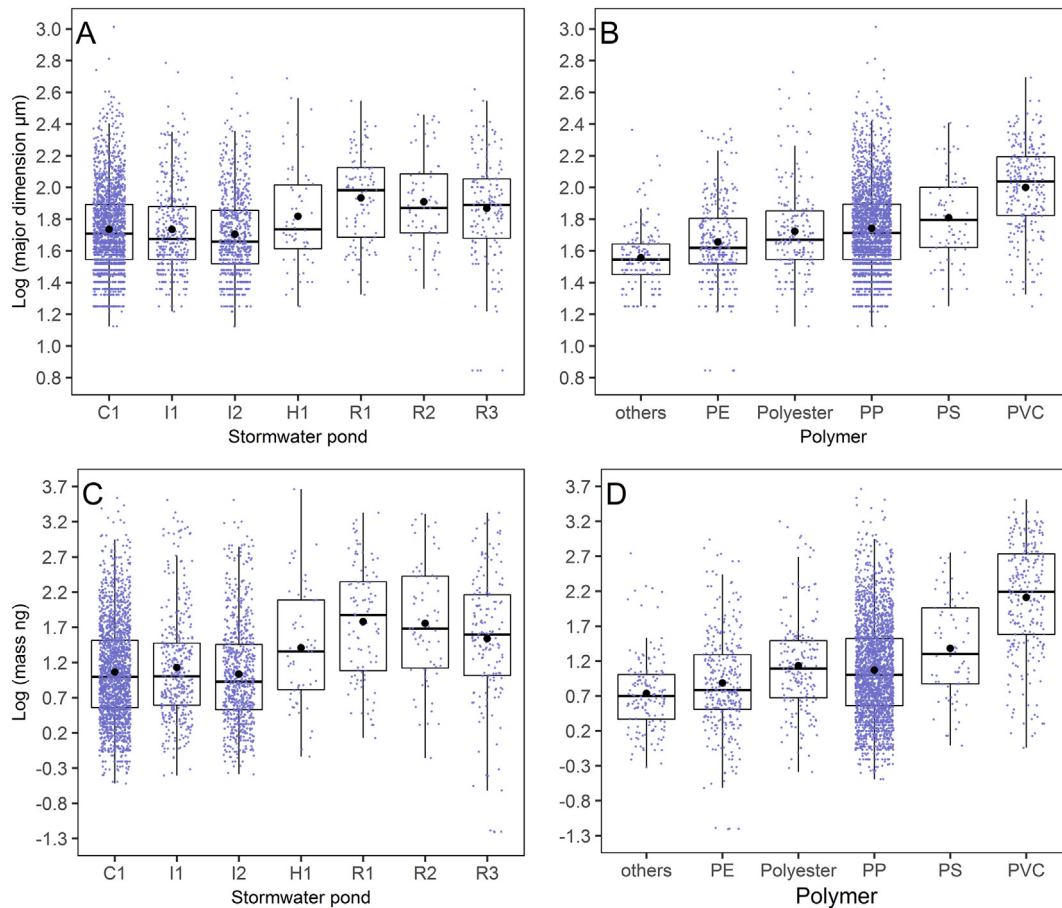**Fig. 2.** Polymer distribution between ponds.

Fig. 3. Boxplot of the measured major dimension of MP on a logarithmic scale versus stormwater pond (A) and versus polymer types (B). Estimated mass of MP on a logarithmic scale versus stormwater pond (C) and versus polymer types (D). Each point represents one of the identified MP. The solid black dots represent the mean values.

different material composition. Another cause for the differences could be the materials used to construct the drainage systems and the buildings in the catchment. For example are PVC, PP, and PE common materials used for urban drainage pipes (Vahidi et al., 2016). Finally, it cannot be excluded that atmospheric deposition can be a cause of MP in ponds where one otherwise would not expect it. For example the presence of polyester in the highway pond (H1) is unlikely to originate from the road drainage system or the traffic.

3.4. Size, mass and polymer type

Among all the MP identified, only five particles had major dimension of >500 μm, and three of them were from C1 and two from I1. Their major dimensions ranged from 532 μm to 1030 μm. Four were of PP and one was of polyester. The rare presence of MP in this size range was also observed in the effluent of 12 German wastewater treatment plants (Mintenig et al., 2017), where MP > 500 μm were visually sorted and identified by ATR-FTIR, and the concentration ranged from 0 to 40 items m^{-3}. A likely reason for the absence of large MP in the stormwater ponds is partly that small particles, in the nature of things, must be more abundant than large particles. The large particles will furthermore have a higher tendency to either float to the water surface or settle to the bottom of the pond as the terminal velocity (rising or sinking) of a particle in a fluid is proportional to the square of its diameter (Stokes Law). In both cases, the particles would avoid being sampled with the approach used in the present study (extracting water 0.2 m below the water surface).

Comparing all MP from the ponds, particles in the commercial and industrial ponds tend to be on average smaller in the size, and lower in the mass (Fig. 3A, C). The residential ponds (R1, R2, and R3) contained MP with an average particle size of 82.7 ± 11.8 μm and particle mass of 54.4 ± 18.6 ng (see Table S1 for details). These ponds ranked the highest, followed by the highway pond (H1). The industrial ponds (I1 and I2) held MP with an average particle size of 46.5 ± 1.3 μm and particle mass of 9.3 ± 1.1 ng, and ranked the lowest among all pond types. This was statistically significant (p < 0.05) for both size and mass of the individual MP when comparing C1, I1, and I2 to R1, R2, and R3. The highway pond was somewhere in between these two groups (see Table S4 for details). The three residential ponds did not differ significantly among themselves in terms of MP number and mass (p < 0.05). For commercial and industrial ponds, the picture was similar, with the exception of the MP size distribution between C1 and I2.

The median size of MP related to their polymer type followed the trend: PVC > PS > PP > polyester > PE > others (Fig. 3B). For the median mass, the order was slightly different: PVC > PS > polyester > PP > PE > others (Fig. 3D). In other words, PP and polyester changed place in the ranking (see Table S2 for details). A contributing factor for this shift is that polyester has a density of 1.3–1.4, while PP's density is slightly below one. However, differences in density cannot alone explain the difference. The second contributing factor is the shape and size distribution of the MP, where small differences in shape and size cause large differences in estimated MP volume, and hence mass.

A PCA analysis was applied to further assess what differentiated the ponds. The polymer composition of each pond was compared to the MP number concentrations (Fig. 4A) and the MP mass concentrations (Fig. 4B), as was the MP size of all the MP in each pond (Fig. 4C). For the latter, the logarithm of the measured major dimension was sorted into bins of 0.2 widths.

With respect to polymer composition, the ponds with residential catchments were grouped for MP number concentrations, and to some degree, for mass concentrations. The other ponds did not show any grouping and, all in all, the catchment type could not by itself explain the polymer composition in the water of a pond. The picture was clearer with respect to the MP size, where ponds serving areas with different land use could be differentiated (Fig. 4C). The MP in the highway pond tended to hold the largest size MP, while the commercial and industrial ponds had the smallest.

The PCA on the polymer composition with respect to MP number concentration showed that the first principle component (PC1) explained 49.7% of the variation, but that the differentiation was mainly driven by the industrial pond I2. Less abundant polymers (alkyd, phenoxy resin, PVAC, PVA, vinyl copolymer, acrylic, epoxy) had a high loading on PC1 (all above 0.9). PC2 explained 21.1% of the variation of the dataset, and most of the separation along it referred to PS (0.7), PP (−0.66), PU (−0.66), PA (0.63), and polyester (0.61). All the residential ponds (R1, R2, and R3) and the highway pond (H1) could be grouped based on the low PC1 value, while a rather high PC1 value was found for the industrial pond I2. The commercial pond (C1) and the industrial pond I1 tended to cluster at low PC2 values.

When the PCA analysis was performed on polymer composition related to polymer mass, PC1 explained more variation (61.2%), which was again mainly driven by pond I2. Again, less common polymers (PA, PU, epoxy, acrylic, SAN, vinyl copolymer, PVA, PVAC, phenoxy and alkyd) all had loadings above 0.9. The second principal component
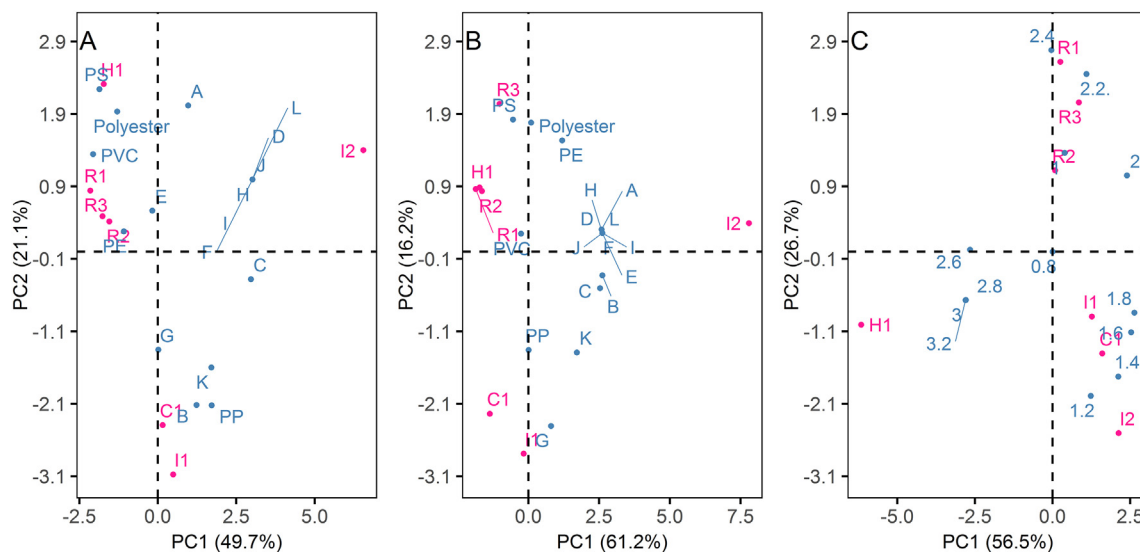

Fig. 4. Biplot of PCA on polymer composition of each pond based on particle number (A) and particle mass (B) concentration. Biplot of PCA on particle size (major dimension, logarithmic scaled) of the MP (C). Capital letters represent the uncommon polymers: A – PA; B – PU; C – epoxy; D – acrylic; E – SAN; F – vinyl copolymer; G – EVA; H – PVA; I – PVAC; J – phenoxy resin; K – acrylic paints; L – alkyd.

(PC2) explained 16.2% of the variances, and the separation was mainly determined by EVA (−0.91), PS (0.69), polyester (0.67), and PE (0.58). Similar to the first PCA analysis, the residential ponds (R1, R2, and R3) and the highway pond (H1) had low PC1 values, while the industrial pond I2 stood alone with a distinguishably high PC1 value. The commercial pond (C1) and the industrial pond I1 were again grouped based on the low values of both PC1 and PC2.

PC1 regarding MP size accounted for 56.5% of the total variance, which was mainly driven by the highway pond (H1), and to a lesser degree by the industrial and commercial ponds. The large MP size bins (bins from 2.6 to 3.2, or 398 μm to 1585 μm) had low loadings on PC1 (all below - 0.9). Some medium MP size bins (bins from 2 to 2.4, or 100 μm to 251 μm) had high loadings on PC2, while smaller MP size bins (bins from 1.2 to 1.8, or 16 μm to 63 μm) had low loadings. The residential ponds tended to cluster at high PC2 values, while the industrial and commercial ponds also clustered at low PC2 values. The highway pond held MP with large particle sizes, which differentiated it from other ponds.

Overall, the study pointed towards some relation between land use and the content of microplastics in the stormwater of the treatment ponds. This was clearest for the ponds serving areas of industrial and commercial land use, versus those with residential and highway usages. The latter group held roughly an order of magnitude less MP. This clearly demonstrates that land use does play an important role for the amount of MP. With respect to the highway pond, it seems reasonable that it had the lowest MP concentrations, as there likely are fewer direct sources for plastic pollution – excluding tyre particles, as these could not be measured in the present study. The sources of the identified MP in the highway pond are unknown, but atmospheric deposition probably plays some role.

While the industrial and commercial ponds showed much variability among themselves, all three PCA analyses (Fig. 4), the size and mass relations (Fig. 3A, C), and the polymer compositions (Fig. 2) all indicated that ponds serving residential catchments held MP concentrations of comparable size and composition. This leads to some confidence in these data being representative of the microplastic concentration of the water phase of residential stormwater retention ponds sized similar to the studied ones.

The measured MP concentrations are likely to be somewhat indicative of the concentration of microplastics in the outflow during small storm events as retention ponds in such cases behave as plug-flow reactors, where incoming water displaces part of the standing pond water (Hvitved-Jacobsen et al., 2010). However, this will depend on local conditions and the actual rain event, and hence needs verification. Furthermore, the data should not be extrapolated to separate stormwater runoff from residential areas per se, partly because not all runoff undergoes treatment, and partly because the efficiency of treatments solutions might vary.

4. Conclusion

Microplastics was present in the water phase of all seven studied stormwater retention ponds, albeit with substantial differences in concentrations and polymeric composition. Land use was identified as an important parameter for explaining the differences between the investigated retention ponds. The ponds holding the highest microplastics concentrations served industrial or commercial catchments, and showed little similarity in terms of microplastics concentrations and polymer composition. Ponds with residential catchments, on the other hand, were more comparable in terms of the microplastics they held.

The most common polymers belonged to PVC, PS, PP, PE, and polyester, while a large number of other materials were found at low levels. The median particle sizes depended on the polymer groups, with PVC particles being the largest while particles belonging to the group of various less common polymers had the smallest size and estimated mass. In terms of particle mass, this difference was quite profound, with PVC

microplastics being more than an order of magnitude heavier than microplastics of, for example, PE, polyester, and PP.

This study of seven stormwater ponds is among the first experimental investigation on the abundance of MP in stormwater runoff from urban and highway areas. It shows that the urban and highway runoff contributes MP to the aquatic environment even though it is treated in stormwater ponds, and that the stormwater runoff needs to be included as a pathway of MP from land to sea. It also shows that the land use of the contributing area should be taken into account when assessing MP loads to the aquatic environment.

Author contributions

Fan Liu carried out the experiments, interpreted the results, and wrote the initial draft. Kristina B. Olesen and Amelia R. Borregaard contributed to the sampling and sample preparation. Jes Vollertsen developed the MPhunter software and supervised the project. All authors contributed to the final manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.03.416.

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