

## Permeable pavements

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Rasmussen, Lasse Abraham; Lykkemark, Jeanette; Andersen, Theis Raaschou; Vollertsen, Jes

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# Permeable pavements: A possible sink for tyre wear particles and other microplastics?

Lasse Abraham Rasmussen<sup>a,\*</sup>, Jeanette Lykkemark<sup>a</sup>, Theis Raaschou Andersen<sup>b</sup>, Jes Vollertsen<sup>a</sup>

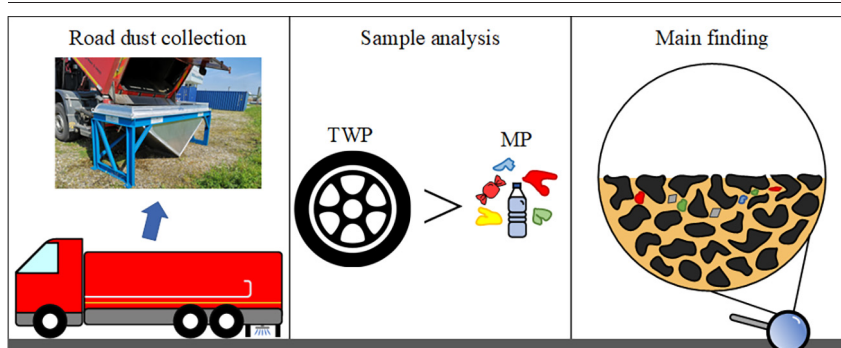
<sup>a</sup> Department of the Built Environment, Aalborg University, 9220 Aalborg Øst, Denmark

<sup>b</sup> VIA University College, Research Centre for Built Environment, Energy, Water and Climate, Banegårdsgade 2, 8700 Horsens, Denmark

## HIGHLIGHTS

- Road dust from permeable pavements was analysed for microplastics.
- Microplastics down to 10 µm were identified using µFTIR imaging.
- Tyre wear particles down to 10 µm were determined by Pyrolysis-GC-MS.
- Tyre wear was on average 49 times more abundant than other microplastics.
- Permeable pavements might act as a sink for microplastics including tyre wear.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this study, seven roads and parking lots were sampled by a road surface cleaning truck and approximately 100 kg of particulate material was collected per site. Thereafter, the samples were analysed for microplastics, including tyre wear particles. The analyses revealed that tyre wear constituted 0.09 % of the dry mass of the samples on average. Other plastic types were also identified in the samples, but at on average 49 times lower concentrations compared to tyre wear particles. Although the roads and parking lots were used for residential, industrial, and commercial purposes, no correlation between land use and the total concentrations of microplastics was identified. Of microplastics other than tyre wear particles, polypropylene constituted an important fraction in all samples, whereas other polymers were present at various degrees. The contents of heavy metals, sulphur, and total organic carbon were also measured in the samples, but no correlation between them and microplastics was determined. A back-of-the-envelope estimation indicated that the tyre wear material retained by permeable pavements constituted a non-negligible fraction of the total mass of microplastics released on roads and parking lots. Therefore, permeable pavements can serve as a tool for the management of this pollutant.

## 1. Introduction

Microplastics (MP) are recognised as one of the major global pollution threats, potentially constituting a risk to both ecosystems and humans. While no generally accepted definition of MP exists, MPs are commonly de-

fined as particles of, or particles containing a significant amount of, synthetic or semi-synthetic organic polymers, and having a largest dimension between 1 µm and 5 mm. This includes rubbers, which in a strict chemical sense are not plastics. While the degree of risk entailed and the conditions under which MP pose a threat are still unclear, there are strong indications that at least a few MP types can be harmful under certain conditions. One example of harmful MP is particles from tyres. For example, Khan et al. (2019) and Simon et al. (2021) reported toxic effects of tyre particles on a

\* Corresponding author.

E-mail address: [lar@civil.aau.dk](mailto:lar@civil.aau.dk) (L.A. Rasmussen).

freshwater crustacean and a freshwater algal species, respectively. Kim et al. (2022) noted that these particles could also have detrimental effects on soil invertebrates. However, the potential effects of tyre particles on humans have received limited scientific attention, but initial studies implied that these impacts are plausible (Li et al., 2022). Laboratory studies have also indicated the potential impacts of other plastic types on humans (Dong et al., 2020; Djouina et al., 2022) and on ecosystems (Khalid et al., 2021).

Road transport results in the formation of MP from the wear and tear of tyres, road markings (paints), and vehicle parts. The contribution of particles containing tyre material has for some countries been argued to be the largest sources of MP discharged into the environment (Lassen et al., 2015; Sundt et al., 2014). Particles containing tyre wear are formed by abrasion with the road surface when the tyre tread partly melts and resolidifies. In this process, agglomerates containing tyre tread, road dust, and road material are formed, commonly termed 'tyre and road wear particles' (TRWP). Jung and Choi (2022) determined that the density of TRWP was  $1.2\text{--}1.7\text{ g cm}^{-3}$ , and Klöckner et al. (2019) concluded that  $>90\%$  of all TRWP was collected at a density of  $1.9\text{ g cm}^{-3}$ . As tyre tread by itself has a density of  $1.15\text{--}1.20\text{ g cm}^{-3}$ , this indicates that quite significant amounts of inorganic material are picked up in the process. Chemical quantification of tyre material, for example by pyrolysis GC-MS, however, quantifies the content of tyre tread related polymers, from which a tyre material content is deducted. A common way to represent the amount of tyre wear related particles is to stipulate the amount of tyre wear material in a sample and term it 'tyre wear particles' (TWP). It is hence important to distinguish between the mass of tyre wear material (TWP), and the mass of tyre and road wear particles (TRWP), where the latter cannot be directly deducted from the first. In some cases, asphalts are also modified by adding polymers such as styrene butadiene rubber (SBR) (Zofka et al., 2021), which can also be part of the tyre tread. Likewise, recycled tyre material is sometimes added to the asphalt (Hernandez-Olivares et al., 2009). Whether or not this turns particles of road material into MPs is up for discussion, but both practices can for sure interfere with the chemical identification of TWP.

Having all this in mind, Baensch-Baltruschat et al. (2020) estimated that the per capita discharge TWP was between  $0.2$  and  $5.5\text{ kg y}^{-1}$ . Furthermore, Kole et al. (2017) estimated that tyre wear particles (TWP) constituted  $5\text{--}10\%$  of the MP influx to the oceans, but models estimate indicated that  $<5\%$  of the TWP generated by traffic reached the oceans (Baensch-Baltruschat et al., 2020; Sieber et al., 2020). Similarly, Goßmann et al. (2021) estimated that  $5\text{ mg g}^{-1}$  and  $0.3\text{ mg g}^{-1}$  of road dust was composed of TWP and other MP types, respectively. However, in a few cases, this ration decreased below the detection limit of the study while moving away from the source and into the ocean. Moreover, Müller et al. (2022) found that the content of TWP reduced with the distance from the source (a road) and soil depth. The authors estimated  $1.8\text{ g kg}^{-1}$  of SBR, an important constituent of TWP, closest to the road ( $0.3\text{ m}$ ). While there have been a few studies on the occurrence of TWP, studies estimating actual emissions from roads are still scarce (Mennekes and Nowack, 2022). One of these is Klöckner et al. (2019) who estimated  $380\text{--}150,000\text{ mg kg}^{-1}$  TRWP in samples collected from two road runoff treatment systems, whereas Klöckner et al. (2020), who analysed road dust and material from sweeping trucks, found  $3700\text{--}480,000\text{ mg kg}^{-1}$  TRWP in the samples. Similar estimates were noted by Røddland et al. (2022) who characterized TRWP in a sample collected from a road tunnel. They found  $25.3\text{--}4820\text{ mg m}^{-2}$  TRWP in samples collected from the road surface while the material from gully pots contained  $31,400\text{ mg kg}^{-1}$  TRWP on average. In the sediments of seven stormwater treatment ponds, Liu et al. (2019b) reported the concentrations of MP other than TWP to be  $0.12\text{--}28.7\text{ mg kg}^{-1}$ , and Olesen et al. (2019) found  $0.40\text{ mg kg}^{-1}$  sediments in one pond. Moreover, Eisentraut et al. (2018) analysed the solids in street runoff (grab sample) and sediments from a runoff treatment facility for SBR, polyethylene (PE), polypropylene (PP) and polystyrene (PS) and depicted that SBR, an indicator of TWP, was the dominant MP in both matrices.

To identify MP types other than TWP, Liu et al. (2019a, 2019b) investigated seven Danish stormwater ponds receiving runoff from several roads and buildings and found that the pond water between runoff events contained between  $490$  and  $22,894\text{ counts m}^{-3}$  (i.e., pieces of MP per  $\text{m}^{-3}$ ), with an estimated mass of  $85\text{--}1143\text{ }\mu\text{g m}^{-3}$ . In the sediments of the same ponds, the authors estimated  $1511\text{--}127,986\text{ counts kg}^{-1}$  dry sediments, corresponding to  $115$  to  $28,732\text{ }\mu\text{g kg}^{-1}$ . The dominant polymers in both matrices were the common ones, which in order of abundance were PP, polyvinylchloride (PVC), polyester, PE, PS, acrylic, and polyamide (PA). Similar observations regarding stormwater runoff were reported by Herath et al. (2022) and Lange et al. (2022). Nonetheless, the analytical method applied by Liu et al. allowed the quantification of MP of sizes equal to  $11\text{ }\mu\text{m}$ , in addition to mass estimation. Using another analytical method that allowed quantifying MP of sizes equal to  $250\text{ }\mu\text{m}$  and targeted only low-density polymers, Boni et al. (2022) estimated the average content of MP for three stormwaters outfalls at  $300$ ,  $370$ , and  $800\text{ counts m}^{-3}$ . The difference in concentrations probably resulted from the use of different methods and their detection limits. Overall, these studies suggested that road transport generated significant amounts of MP, of which most likely only a small fraction ended up in the ocean while the rest stayed on land or in freshwater systems.

MPs are also present in the air, some of which comes from roads. Kole et al. (2017) estimated that the particulate matter ( $\text{PM}_{2.5}$ ) in the air contained  $3\text{--}7\%$  TWP, whereas Sun et al. (2022) found that  $\text{PM}_{2.5}$  in several Chinese megacities consisted of  $>0.1\text{ mg g}^{-1}$  TWP. This indicated that TWP become airborne to some degree and spreads via air. This was confirmed by Goßmann et al. (2022), who investigated urban outdoor spiderwebs and reported that approximately  $10\text{--}40\text{ mg g}^{-1}$  of the collected spiderweb mass contained TWP, in addition to significant amounts of other MP types. While the TWP accounted for  $40.8\%$  of the total amount of MP they found, PET (poly(ethylene terephthalate)) accounted for  $36.0\%$  and PVC (poly(vinyl chloride)) for  $12.0\%$ . Other plastic types have also been found occasionally in similar samples. The first study on MP in outdoor air (Dris et al., 2017) estimated an MP concentration of  $0.3\text{--}1.5\text{ counts m}^{-3}$ , with small fibres dominating the air. Although studies suggest that the current outdoor air contains MP, the reported concentrations of MP vary between studies, with several uncertainties (Beaurepaire et al., 2021). This prompts the question of where the MP types other than TWP and road paints, found on roads and various other impervious surfaces, originate from. Solid data on this is unfortunately scarce (Beaurepaire et al., 2021), but it stands to reason that mismanaged waste breaking down in the urban environment would be a contributor.

The abovementioned studies indicated that activities on the roads produced significant amounts of TWP, and that other MP types were also present on road surfaces. It is well-known from urban hydrology, that particles accumulate on these surfaces during dry weather, reaching a saturation concentration depending on various factors, including traffic load and traffic behaviour (Goonetilleke et al., 2009; Djukic et al., 2018). This suggests that MP would follow similar build-up and wash-off processes. Nonetheless, the runoff water from these surfaces is either treated using stormwater best management practices (BMPs) in sustainable stormwater drainage systems (SuDS) or discharged untreated (Hvitved-Jacobsen et al., 2010).

The most common stormwater treatment approach is to allow the runoff to pass through an artificial pond or infiltrate the soil. A promising, emergent treatment approach includes permeable pavements such as permeable asphalt and concrete. Similar to most BMPs, permeable pavements can manage both peak flows and pollutants, as illustrated by the findings of Hernandez-Crespo et al. (2019), who found that suspended solids, organic matter, and nutrients could be reduced by the passage of runoff water through a porous concrete pavement. Jayakaran et al. (2019) found that porous asphalt could retain as much as  $93\%$  of the total suspended-solids load. However, porous asphalt was significantly less efficient at retaining dissolved substances. In contrast, Dreelin et al. (2006) observed that porous pavements were

effective at retaining fine particles in stormwater runoff, including clay. Therefore, these pavements could also retain MP present in the runoff water, albeit to which degree remains unknown. A porous pavement must be maintained to ensure its function as a drainage and treatment infrastructure, which is typically done by pressure washing and vacuuming up the released particles. Without regular maintenance, for example once a year, the permeable surface is likely to get clogged and finally sealed (Razzaghmanesh and Beecham, 2018; Simpson et al., 2019; Andersen et al., 2022). Hence, their maintenance harvests the particles accumulated in the porous pavement, after which they can be safely disposed of.

The present study aimed at improving the understanding of how porous asphalt can retain MP, in addition to suspended solids and other pollutants. For this purpose, we investigated the particles collected during routine pressure washing of seven existing roads and parking lots equipped with porous asphalt. The study also presented a back-of-the-envelope estimate on whether the retainment of MP was significant compared to the amount of MP that can be expectedly produced on the roads.

## 2. Material and methods

### 2.1. Site description

The sampling sites were located on the peninsula of Jutland and the island of Funen in Denmark. All sites were constructed by NCC Industry A/S, Denmark, and comprised of a permeable asphalt surface course, a permeable asphalt binder course, and an unbound open-graded base course. The latter was designed to store water until it infiltrated the underlying soil. The surface course (PermaSlid®) consists of crushed granite of 11–16 mm maximum grain size bound by polymer modified bitumen. The binder course (PermaGAB®) consists of 16–22 mm maximum grain size gravel, also bound by polymer modified bitumen. Both courses have a pore volume of 19–23 %. The underlying unbound open-graded base course has a pore volume of 30 %. At the time of sampling, a total of 118 roads and parking lots equipped with these permeable pavements had been constructed in Denmark by NCC Industry A/S. Of these, seven sites were selected for sampling, covering different land use including residential ( $n = 3$ ), industrial ( $n = 2$ ) and commercial ( $n = 2$ ) (Table 1). The pavements were cleaned approximately once a year by pressure washing and wet vacuuming.

**Table 1**  
Sampling sites.

ID	Type	Description
ALV	Residential	Agerlandsvej, Odense: a dead-end road in a suburban residential area with 16 detached family houses.
PKV	Residential	Porskervevej, Galten: a main road with bus traffic and nine houses on the side. One of these properties includes a truck parking lot.
HDS	Residential	Dalbyvej, Hedensted: an arterial road in the suburban area of Hedensted, with no houses directly adjacent to the road. However, the road connects approximately 100 houses, as well as the rural area outside to the city
LHV	Industrial	Lufthavnsvej, Nr. Sundby: an access road for bus traffic to Aalborg Airport – the third largest airport in Denmark, with 1.4 million annual passengers (2019), and an access road to an industrial area containing a broad range of industries such as metalworking, recycling and production.
EPV	Industrial	Eksportvej, Billund: a road in an industrial area with a shipping and taxi company, in addition to a cargo handling space for the Billund Airport, the second largest airport in Denmark, with 3.7 million annual passengers (2019).
LDL	Commercial	Parking lot, Dalbyvej 20, Hedensted: a parking lot for a grocery store (Lidl) with 90 parking spaces.
JFR	Commercial	Parking lot, Grenåvej 425, Egå: a parking lot for a hardware store (Jem&Fix) with 49 parking spaces.

### 2.2. Sample collection

Road dust was collected using a designated road surface cleaning truck (Beam S14000), which was used for regular maintenance of permeable surfaces, including the pavements sampled in of the present study (Supplementary Information (SI), Fig. S1). The accumulated road dust was first loosened from the porous surface using a rear-mounted rotor cleaning system, which pressures washed the road at 300 bar. The mix of water and road dust was then sucked into the hopper. This cleaning approach allowed the removal and collection of materials from a depth of a few centimetres into the permeable road surface.

A total of 1000 L of water was used for sample collection at each site, corresponding to 800 m<sup>2</sup> of road for six sites and 500 m<sup>2</sup> for one site (Table 2). After collection, the samples were delivered by the cleaning truck to Aalborg University, Aalborg, Denmark, over travel distances ranging from 12 km to 255 km. The samples were then unloaded into a custom-made settling tank (SI Fig. S2). A schematic of the procedure from sampling to final dataset is given in Fig. 1.

### 2.3. Sample preparation

Upon transfer to the settling tank, the samples were allowed to settle for 48 h, after which the bulk of the water was removed through filtration using a plastic-free filtration system to collect particles on two 10 µm stainless steel filters of 167 mm diameter (Rist et al., 2020; Rasmussen et al., 2021). The retentate on the filters was removed from the filters by ultrasonication and collected in particle-free demineralized water and transferred back into the settling tank and allowed to settle for an hour.

As the amount of material in the settling tank was large, subsamples were collected using glass sediment cores and stored for the analysis of MP, including TWP and heavy metals. Three separate sediment cores were used for sampling from the left, middle and right sides of the settling tank to ensure adequate representation. These cores were pooled into a glass container and oven-dried at 50 °C. After sample extraction, the remaining sediment in the settling tank was weighed and its water content was measured to obtain the total amount of sediment recovered at each sampling.

To prepare for MP and TWP analyses, the samples underwent a multistep polymer-preserving sample preparation procedure. Initially, a 30 g subsample of dried road dust was homogenized using a mortar and added to a beaker containing sodium polytungstate (SPT) with a density of 1.9 g cm<sup>-3</sup>. This density probably allowed the extraction of most of the TRWP, which have a higher density than the tyre tread itself (Klößner et al., 2019). Thereafter, the homogenized sample was loaded into a separation funnel and thoroughly mixed with compressed air for 30 min to avoid aggregates followed by separation for 24 h. The sink-fraction was drained away while the float-fraction was filtered onto a Ø47 mm stainless steel filter of 10 µm nominal pore size, transferred into a 5 % sodium dodecyl sulphate (SDS) solution, and incubated while stirring at 50 °C for 48 h.

To further degrade the organic material, the samples were subjected to enzymatic treatment. First, the samples were transferred into a 0.2 M tris (hydroxymethyl) aminomethane buffer solution (pH 8.2) with 1 mL of protease (Alcalase® 2.4 L FG, Novozymes, Bagsværd, Denmark) and incubated at 50 °C for 48 h. Afterwards, the buffer solution and enzyme were changed to acetate buffer (pH 4.8) with 0.5 mL of Cellulase enzyme blend® and the cellulolytic enzyme mixture Viscozyme® L (Merck KGaA, Darmstadt, Germany), respectively, and further incubated for 48 h. Subsequently, the organic material was further removed by a Fenton's reaction wherein H<sub>2</sub>O<sub>2</sub> was catalysed using Fe under acidic conditions to enhance its oxidative potential. The samples were then transferred into 200 mL of Milli-Q water, followed by the addition of 145 mL of 50 % H<sub>2</sub>O<sub>2</sub> and 62 mL of 0.1 M iron sulphate (FeSO<sub>4</sub>) were added. The pH of the mixture was adjusted to 3 by adding 0.1 M sodium hydroxide (NaOH). As the Fenton's reaction is exothermic, the temperature was controlled by keeping the



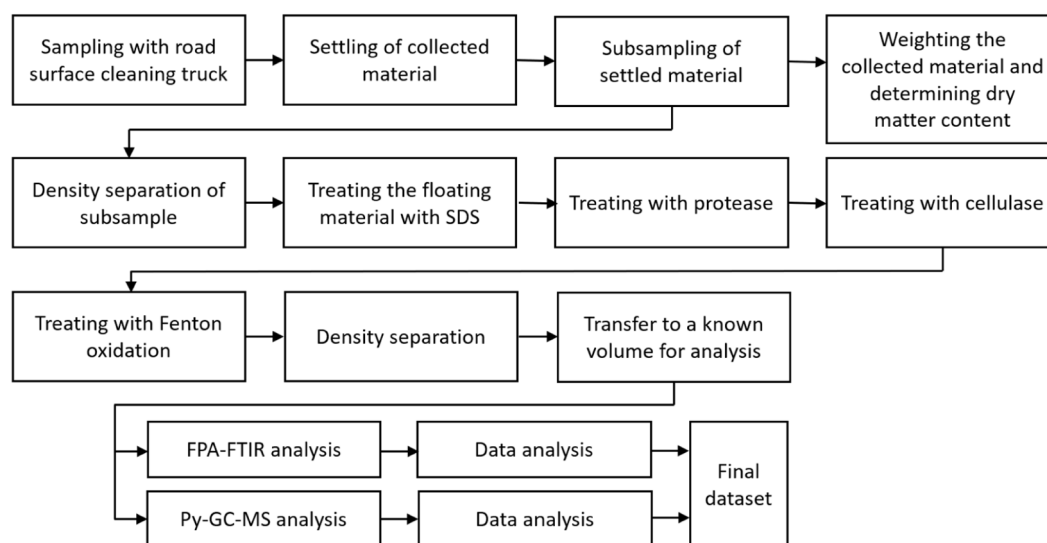
**Table 2**

Sampled material and the measured concentrations.

ID	Sampled road area [m <sup>2</sup> ]	Total dry mass of sampled particles [kg]	MP count concentration without TWP [counts kg <sup>-1</sup> ]	MP mass concentration without TWP [mg kg <sup>-1</sup> ]	TWP mass concentration [mg kg <sup>-1</sup> ]	Area-specific amount of MP without TWP [mg m <sup>-2</sup> ]	Area-specific amount of TWP [mg m <sup>-2</sup> ]
LHV	500	108.57	322,000 (±14,000)	14.8 (±1.7)	74.95 (±1.38)	3.21	16.27
PKV	800	66.05	418,000 (±27,000)	10.0 (±2.5)	1204.64 (±693.78)	0.83	99.46
LDL	800	88.17	375,000 (±70,000)	8.1 (±2.3)	748.22 (±192.38)	0.89	82.46
HDS	800	98.87	133,000 (±13,000)	8.7 (±2.5)	404.15 (±47.63)	1.08	49.95
ALV	800	169.95	718,000 (±372,000)	16.2 (±9.2)	0.00	3.44	0.00
EPV	800	133.80	1,075,000 (±386,000)	122.3 (±17.9)	2868 (±548.40)	20.45	479.70
JFR	800	85.00	236,000 (±27,000)	23.7 (12.0)	68.66 (±21.16)	2.52	7.29

reaction vessel in an ice bath to avoid degradation of polymers which might

deposition area was scanned (10 × 10 mm<sup>2</sup>) at a spectral range of

**Fig. 1.** Schematic of the process from sampling to final dataset.

occur at high temperatures. Finally, the samples underwent another density separation using the SPT solution of 1.9 g cm<sup>-3</sup>, upon which the float-fraction was concentrated in 10 mL 50 % (v/v) ethanol.

#### 2.4. Microplastic analysis

Focal plane array-based Fourier transform-infrared (FPA-FTIR) spectroscopy was performed using a Cary 670 FT-IR spectrometer (Agilent Technologies, Santa Clara, CA, USA) coupled with a Cary 620 FT-IR microscope to identify MP other than TWP in the particle concentrates. This technique creates a hyperspectral image of deposited particles, which can be interpreted to identify MPs contained herein. First, 10 mL of concentrate was homogenized by vortexing, after which a subsample of 50–200 µL was deposited onto an infrared transparent, Ø13 × 2 mm<sup>2</sup> circular zinc selenide window using a glass-capillary micropipette. The windows were placed in a compression cell (Pike Technologies), limiting the effective area of deposition to Ø10 mm. After particle deposition, the subsamples were dried on a heating plate at 50 °C. Then, subsamples ( $n = 3$ ) were collected from each sample to account for heterogeneity during subsampling and improve the representation of the samples.

The FTIR microscope was equipped with a 128 × 128 FPA mercury–cadmium telluride detector, and images were acquired using a 15 × Cassegrain objective with a pixel resolution of 5.5 µm. The whole

3750–850 cm<sup>-1</sup> and a resolution of 8 cm<sup>-1</sup>. To reduce the signal-to-noise ratio, 30 co-added scans were acquired for the samples and 120 co-added scans were used for the background.

As FPA-FTIR could not determine the content TWP due to the presence of carbon black, we used pyrolysis–gas chromatography–mass spectrometry (Py-GC/MS) for this purpose. A total of 50 µL of subsamples from the particle concentrates were deposited into sample cups using a glass-capillary micropipette and dried on a heating plate at 50 °C. Then, the subsamples were analysed using a Py-GC/MS system composed of a micro-furnace pyrolyser EGA/Py-3030D (FrontierLab, Japan) and an auto-shot sampler AS-1020E (FrontierLab, Japan) connected to a Thermo Scientific TRACE 1310 GC and an ISQ™ single quadrupole GC/MS system with helium as the carrier gas. A solution of 0.2 g L<sup>-1</sup> deuterated anthracene (A-d10) in n-hexane was used as the internal standard. Both d-anthracene and d-PS were considered as internal standard. Their performance was evaluated against the investigated matrix, and it was found that both performed well, and d-anthracene chosen in this project. The method for the Py-GC/MS included pyrolysis at 600 °C, with an interface at 280 °C. Helium gas was used as the carrier (1 mL min<sup>-1</sup>) and injected in split mode at a ratio of 30:1. The temperature program involved heating at 40 °C for 2 min, with a temperature gradient of 10.5 °C min<sup>-1</sup> and increasing to a final temperature of 300 °C for 5 min. The mass spectrometer was operated in the EI positive mode (70 eV;  $m/z$  range: 35–500), and the transfer line and ion source were at maintained 250 °C and 200 °C, respectively.

## 2.5. Estimation of elements and total organic content

Subsamples of dried road dust ( $n = 3$ ) were prepared for heavy metal analysis using microwave assisted acid digestion (Anton Paar, Multiwave 7000) and subsequently analysed using an Inductively Coupled Argon Plasma with Optical Emission Spectrometry (ICP-OES) detection system (ICAP 6300 Duo View, Thermo Scientific) to determine the contents of Cd, Cr, Cu, Fe, Mn, Ni, Pb, S, and Zn. The total organic carbon (TOC) content was also determined using a TOC analyser (Vario TOC select, Elementar Analysensysteme GmbH) ( $n = 3$ ). Freeze dried subsamples were weighed before and after acidification with 1 M HCl before analyses.

## 2.6. Data analysis

The FPA-FTIR analysis resulted in a spectral mapping of the particles on the window, containing 3.2 million individual spectra per scan. This data was processed using the freeware *siMple* developed for the semi-automated analysis of MP other than TWP. The software correlated the acquired spectra with a reference library (Primpke et al., 2020), which presently contained >100 spectra of polymers and natural materials. Thereafter, the mass of the particles was estimated from the 2-dimensional size measurements and material density of the identified MP (Simon et al., 2021).

An estimation of the content of TWP was obtained by fitting the results to calibration curves derived from pyrolyzing standards consisting of a cryo-milled mix of tyre tread from cars and trucks from Denmark and Sweden (see SI). Car and truck tyres have highly variable contents of natural and synthetic rubber, and whatever approach for calibration is chosen will present issues when applying specific markers for quantification (Rauert et al., 2021). Miller et al. (2022) suggested to overcome this by an empirical approach where tyre tread materials representative for the region were used, which was the approach used in the present study. Extracted ion chromatograms for 4-vinylcyclohexene were used for quantification, which was found as omnipresent in both car and truck tyre treads (Goßmann et al., 2021) and served as a marker for styrene butadiene rubber (SBR) and butadiene rubber (BR).

## 2.7. Quality control and contamination

To avoid cross-contamination between samplings, the hopper of the road surface cleaning truck was pressure-washed prior to each sampling. Sediment cores and other utensils for sampling from the sediment tanks, as well as glassware used in sample preparation, were flushed using filtered demineralized water. Smaller items such as steel filters were muffled at 500 °C prior to use. Sample preparation was done inside a fume hood and samples kept covered by aluminium foil whenever possible. No plastics utensils were used during sample preparation. The only exception was Teflon covered magnetic stir bars. However, Teflon was not included in the analysis, and this would hence not have interfered with the results. All liquids were prefiltered on GF filters. The air of the analytical lab was continuously filtered by an industrial size HEPA filter while only cotton lab coats were always worn.

However, as some degree of contamination is expected due to the ubiquitous presence of plastics, triplicate laboratory blank were prepared alongside the other samples.

## 3. Results and discussion

### 3.1. Contamination

A subsample of 1 ml of each laboratory blank was analysed and revealed a total of 11 MP particles. On average each sample could possibly contain 3.7 items with a corresponding average mass of 118.5 ng. Considering the average number of MPs and their mass from all of the samples, the level of contamination equates to 2.8 % and 1.7 %, respectively. Of the two polymer type were found in the blanks, polyester accounted for 64 %

of the total mass while PS accounted for the remaining 36 %. The blanks were not analysed for TWP. Due to the relatively low level of contamination within the blanks, the results were not in any way corrected for contamination.

### 3.2. MP and TWP concentrations

The FPA-FTIR analysis identified MP other than TWP in all samples (Table 2). The scans identified a total of 2755 MP and were used to extrapolate their concentrations (Table 2). The MP concentrations ranged from 133,000 to 1,075,000 counts  $\text{kg}^{-1}$ , corresponding to 8.1 to 122.3  $\text{mg kg}^{-1}$ . Average values were 468,000 counts  $\text{kg}^{-1}$  and 29.1  $\text{mg kg}^{-1}$ , respectively. TWP was identified in all but one sample at concentrations ranging from 68.66 to 2868.19  $\text{mg kg}^{-1}$  with an average of 894.80  $\text{mg kg}^{-1}$ . Moreover, the TWP concentration was, on average, 49 times higher than that of other MP. In the sample from PKV, it was upwards of 120 times more abundant. Where the concentration of TWP was highest (site EPV), it corresponded to 0.29 % of all particulate material being TWP. The concentration of TRWP, i.e., the agglomerates of tyre material, road dust, and road materials, must per definition have been even higher, albeit it cannot be estimated from the present data how much higher. The corresponding mass of MP other than TWP at this site was 0.012 % of all particulate material. One caveat is that the bitumen of the permeable asphalts was modified by elastomers. Polymer modified asphalt particles could hence to an unknown degree have been interpreted as TWP.

Considering the road areas sampled and the total weights of dry materials collected, the measured concentrations corresponded to area-specific concentrations of 0.83 to 20.45  $\text{mg m}^{-2}$  for MP other than TWP and 7.29 to 479.70  $\text{mg m}^{-2}$  for TWP. In both cases the concentrations failed a normal distribution test. The medians for the area-specific concentrations of MP without TWP, TWP, and MP including TWP were 1.71, 49.95, and 51.02  $\text{mg m}^{-2}$ , respectively, while the means were 4.52, 105.02, and 109.54  $\text{mg m}^{-2}$ , respectively.

MP concentrations did not exhibit any trend with land use (residential, industrial, commercial). While the site with the highest MP concentrations (EPV) was industrial, the second industrial site (LHV) exhibited less than average MP concentrations. Similarly, for residential land use: While the residential site PKV had lowest concentrations of MP other than TWP, it held the second highest TWP concentration. On the other hand, the site ALV, also residential, held zero TWP but the second-highest concentration of MP other than TWP. Somewhat similar for the commercial sites, where LDL held the second-lowest concentration of MP other than TWP, while it held the third highest TWP concentration. The data obtained in the present study was consistent with the studies on TWP and TRWP mentioned in the introduction, indicating that TWP was the dominant type of MP on road surfaces.

Table 3 illustrates the concentration of selected heavy metals, TOC and sulphur in the sampled materials. Comparing to the content of MP other than TWP and TWP yielded no correlation. Despite zinc being present in tyre material at high concentrations and being considered as a marker for TWP (Müller et al., 2022), zinc content was not correlated with TWP in this study (Klößner et al., 2019). Additionally, zinc can leach out of TWP and TRWP (Rhodes et al., 2012; Halle et al., 2021); although the leaching

**Table 3**

The contents of TOC, chromium, copper, lead, sulphur, and zinc in the collected samples.

Sample	TOC [ $\text{mg kg}^{-1}$ ]	Cr [ $\text{mg kg}^{-1}$ ]	Cu [ $\text{mg kg}^{-1}$ ]	Pb [ $\text{mg kg}^{-1}$ ]	S [ $\text{mg kg}^{-1}$ ]	Zn [ $\text{mg kg}^{-1}$ ]
LHV	6.4	12.80	19.31	4.62	637.36	86.08
PKV	275.4	70.05	76.05	18.25	1748.78	328.90
HDS	40.3	16.27	29.11	9.23	891.35	142.64
LDL	110.7	12.70	34.58	6.33	941.82	111.87
ALV	78.8	22.71	95.25	16.10	1792.86	204.43
EPV	45.8	23.76	69.54	12.16	2128.10	228.47
JFR	68.3	17.53	94.05	16.02	1685.63	414.69

rate is poorly understood, zinc leaching may be enhanced by particle weathering (Simon et al., 2021). Therefore, leaching of zinc from the TRWP on roads and through the pores of asphalt might be why zinc content was not correlated with TWP in this study.

### 3.3. Size distribution of MP

Only MP identified by FPA-FTIR could be distinguished in terms of size as py-GC/MS solely yields information on the bulk mass. The largest particles were found in the samples collected from the industrial site EPV, which also contained the highest mass of MP (Table 2). In contrast, the smallest particles were found at PKV, where also the lowest mass was found. Overall, there was a weak correlation between the median MP size and the total mass ( $r^2 = 0.879$ ). A similar observation was recorded while comparing the distributions of estimated particle masses. All residential sites (PKV, HDS, ALV) held comparable and small MP sizes while three out of the four sites serving commercial (LDL, JFR) and industry (LHV, EPV) purposes held MP larger than the rest. As a general trend, few large particles tended to hold a large part of the total mass (SI Fig. S3). On average the 6.2 % most massive particles held half the total MP mass found by FPA-FTIR (Fig. 2).

The polymer composition of MP excluding TWP is shown in Fig. 3. Overall, PP dominated the samples, which aligns with several other studies on urban MP (e.g., Liu et al., 2019a, 2019b). However, the sample collected from the parking lot outside a grocery store (LDL) exhibited a high concentration of cellulose acetate, which could be attributed to cigarette buds. Samples collected from the other parking lot outside a hardware store (JFR) and the residential road (PKV) also exhibited significant amounts of cellulose acetate.

### 3.4. Estimating retainment efficiency

The permeable asphalt clearly retains MP in its pores, and hence has some effect as a stormwater treatment unit, a BMP. However, how much of the MP produced on the site is retained? For this the input of MP to the road surface must be known for each site. This is though difficult to measure directly. A very rough first estimate for TWP can be done by assuming that the average retained mass per  $m^2$  of road found is representative of all roads, and that regional estimates on TWP release from vehicles can be used as the input for a region or country. For Denmark, Lassen et al. (2015) estimated that some 4200–6600 ton of TWP is released by vehicles per year. According to Danish national statistics, the total area of Danish paved roads is approx. 1400  $km^2$ . On top of this comes an unknown area of other surfaces experiencing traffic, such as parking lots. Assuming that the total area on which tyres are abraded in Denmark is 1500  $km^2$ , this leads to an annual release of around 3.6  $ton\ km^{-2}$  of road, or 3600  $mg\ m^{-2}$ . In comparison, the average mass of TWP retained was 105.02  $mg$

$m^{-2}$  which in terms of total mass, would be 157.53  $ton\ y^{-1}$ . While there indeed are very large uncertainties in such a back-of-the-envelope estimate, it nevertheless indicates that properly maintained permeable pavements can play a role in mitigating pollution from car tyres. With respect to other MP than TWP, a similar calculation yields that the permeable pavements could retain in the order of 7  $ton\ y^{-1}$ . Applying estimates from Lassen et al. (2015) on other plastic types and excluding rubber granules from for example artificial turfs, yields an annual yearly release of microplastics to the Danish environment of approx. 3300  $ton\ y^{-1}$ . On top of this comes an unknown amount MP formed from larger plastic items breaking down in the environment. Whatever the true numbers are, the total retainment of MP other than TWP by pervious pavements seems small compared to the overall load on the environment. Whether it is significant compared to the actual load on the road can unfortunately not be estimated from the current data.

### 3.5. Strength and limitations

Moving out of the lab and into the field introduces many uncertainties as conditions cannot be controlled and often stay largely unknown. This is also the case in this study, where material was collected from permeable asphalt using a road surface cleaning truck. Questions like ‘how representative is the sample’ are difficult to answer as results might differ when sampling the same road at another time. It is also difficult to know how deep into the road surface the cleaning reaches, and hence if all or only part of the material was collected. On the other hand, the amount of material sampled was quite huge, and covered a rather large area, giving confidence in the representativeness of the sample for that road at that point in time. Something which cannot be ensured if a more limited approach had been used, such as sampling a small area manually. Furthermore, a controlled lab study would struggle to reproduce the dynamics of activities and loadings of a real road surface.

Once the sample is received by the lab, uncertainties continue. It begins when subsampling the approx. 100 kg of material collected per site (Table 2) for further analysis, where it is notoriously difficult to ensure that the sample is representative, an issue well-known from sampling solid matrices (Gustavsson et al., 2006). The sample preparation and the subsequent analysis also introduce errors, which though are difficult to quantify for MP where results tend to depend heavily on the targeted size range (Haave et al., 2019; Chand et al., 2021). They are also affected by the sample preparation procedure and the analytical method applied (Dimante-Deimantovica et al., 2022). It can, though, be difficult to assess exactly how much, as it is difficult to determine the ground truth of MP concentration in an environmental sample. Hence the many uncertainties undoubtedly present in a study like this will affect the specific numbers obtained. All this being said, the introduced analytical errors probably tend to be systematic, and conclusions on relative values hence more solid than absolute numbers. Furthermore, the uncertainty on specific numbers will not affect the overall conclusions that porous asphalt can play a significant role when treating stormwater runoff for MP, a conclusion which would be difficult to draw by other means than a large-scale field study conducted under environmentally realistic conditions.

## 4. Conclusions

Permeable pavements have been increasingly used as a measure to manage stormwater runoff from urban areas and rural roads. The pores of the pavements retain particulate material, including MP. In this study, seven similar pavements were analysed for MP, including TWP. The concentration of TWP was highly variable, ranging from 0  $g\ kg^{-1}$  to 2.87  $g\ kg^{-1}$ , and no correlation to land use could be identified. MP other than TWP were also present in the sample, but at concentrations 49-times lower than that of TWP. The average TWP concentration was 0.89  $g\ kg^{-1}$ , i.e. 0.09 % of all particulate material collected was contributed by tyre wear. Tyre wear particles would not only have been present as tyre fragments but also as TRWP, which result from the melting of dust particles and

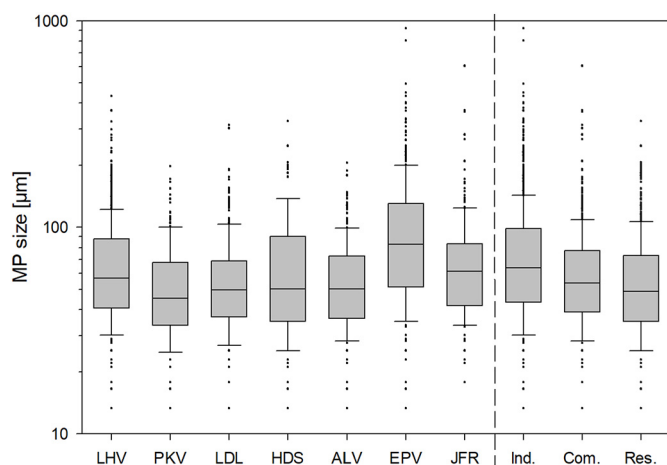


Fig. 2. MP size distribution per site and land use.

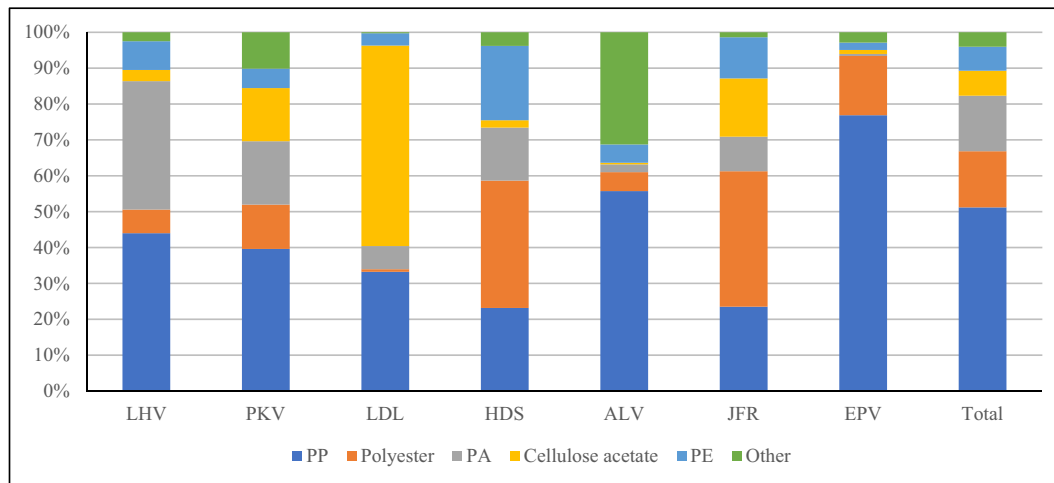


Fig. 3. Polymer type distribution in terms of relative mass.

road materials into a tyre polymer matrix. Hence, the amount of the latter would also be high, which was indicated by the fact that permeable pavements accumulated high concentrations of this type of MP. Furthermore, a back-of-the-envelope estimation indicated that permeable pavements could retain a significant fraction of the TWP generated on roads and hence these pavements can play a role in developing solutions to manage MP in stormwater runoff from urban areas and rural roads.

Permeable pavements as a solution to treat stormwater for MP seem promising. However, there are still numerous research gaps to be covered to fully understand how it functions and how efficient it is compared to other treatment solutions. One example of such research gap is the downwards migration of MP in the permeable asphalt. Do the MP stay in the top layers where they can be readily removed by a cleaning truck? Do they migrate into the deeper layers of the road construction? Or maybe even into the underlying groundwater? Another important knowledge gap is to understand the fate of MP on the road surface. Do MP on the road surface predominantly move into the asphalt pores, or are they predominately thrown off the road surface by vehicle movement? These and many other questions need to be addressed by future studies on best management practices for stormwater MP pollution.

#### CRedit authorship contribution statement

Lasse Abraham Rasmussen: conceptualized the project, developed methodology, carried out the experiments, analysed data and wrote the original draft. Jeanette Lykkemark: developed methods, analysed data and wrote the original draft. Theis Raaschou Andersen: conceptualized the project, reviewed-, and edited the final draft. Jes Vollertsen: Supervised the project, wrote and the original draft. All authors contributed equally the final manuscript.

#### Data availability

Data will be made available on request.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.161770>.

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