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Removal and fate of microplastics in permeable pavements: An experimental layer-by-layer analysis

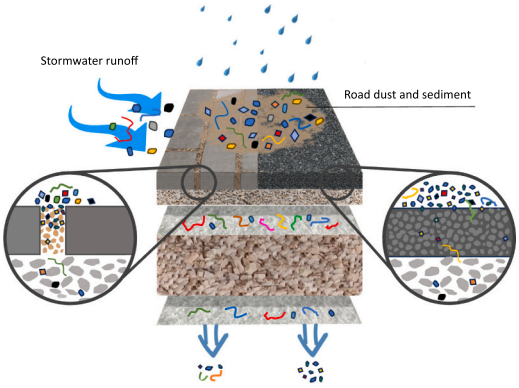
Eduardo García-Haba [a,*], Alba Benito-Kaesbach [b], Carmen Hernández-Crespo [a], Carlos Sanz-Lazaro [b,c], Miguel Martín [a], Ignacio Andrés-Doménech [a]

[a] Instituto Universitario de Ingeniería del Agua y Medio Ambiente, Universitat Politècnica de València, Camí de Vera s/n, 46022 València, Spain
[b] Departamento de Ecología, Universidad de Alicante, 99, E-03080 Alicante, Spain
[c] Multidisciplinary Institute for Environmental Studies (MIES), Universidad de Alicante, 99, E-03080 Alicante, Spain

HIGHLIGHTS

- Permeable pavements retain microplastic and tire wear particles efficiently.
- Fibres and fragments <0.1 mm represent challenging particles to retain.
- Geotextile and surface layers are key in microplastic capture.
- Geotextiles do not contribute to microplastic pollution.
- Frequent surface cleaning is key to control microplastic pollution.

GRAPHICAL ABSTRACT



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ABSTRACT

The increasing prevalence of microplastics (MP) in urban environments has raised concerns over their negative effects on ecosystems and human health. Stormwater runoff, and road dust and sediment, act as major vectors of these pollutants into natural water bodies. Sustainable urban drainage systems, such as permeable pavements, are considered as potential tools to retain particulate pollutants. This research evaluates at laboratory scale the efficiency of permeable interlocking concrete pavements (PICP) and porous concrete pavements (PCP) for controlling microplastics, including tire wear particles (TWP) which constitute a large fraction of microplastics in urban environments, simulating surface pollution accumulation and Mediterranean rainfall conditions. Microplastic levels in road dust and sediments and stormwater runoff inputs were 4762 ± 974 MP/kg (dry weight) and 23.90 ± 17.40 MP/L. In infiltrated effluents, microplastic levels ranged from 2.20 ± 0.61 to 5.17 ± 1.05 MP/L; while tire wear particle levels ranged between 0.28 ± 0.28 and 3.30 ± 0.89 TWP/L. Distribution of microplastics within the layers of PICP and PCP were also studied and quantified. Microplastics tend to accumulate on the pavements surface and in geotextile layers, allowing microplastic retention efficiencies from 89 % to 99.6 %. Small sized (< 0.1 mm) fragment shaped microplastics are the most common in effluent samples. The results

* Corresponding author.
E-mail address: edgarha@upv.es (E. García-Haba).

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indicate that permeable pavements are a powerful tool to capture microplastics and tire wear particles, especially by surface and geotextile layers. The study aims to shed light on the complex mobilisation mechanisms of microplastics, providing valuable insights for addressing the growing environmental concern of microplastic pollution in urban areas.

1. Introduction

Microplastics are becoming of increasing concern due to their environmental and human threats. These tiny plastic particles, which can measure up to 5 mm in their longest dimension, have negative impact on the physiology of both humans and marine organisms, causing genetic alteration or behavioural changes (Sharma et al., 2023; Prata et al., 2021; Thompson et al., 2004).

Urban areas constitute an important diffuse source of microplastics as consequence of anthropogenic activities, including wear processes, landfills or sewage treatment among others (Österlund et al., 2023; Qiu et al., 2020). The substantial growth of cities, coupled with the expansion of impervious surfaces, lead to significant alteration of urban catchments and hydrology (Walsh et al., 2012). During rain events, stormwater runoff can mobilise the accumulated microplastic particles (i.e. plastic fragments and fibres, and tire wear particles among others), into receiving ecosystems (Cho et al., 2023; Werbowski et al., 2021), acting as the main driver of this pollutant to water bodies, including rivers ending up in the oceans (Sugiura et al., 2021; Bilkovic et al., 2019; Andrés-Doménech et al., 2018; Pal et al., 2014). Tire wear particles, classified as a subset of microplastics due to their physical and chemical properties, are the result of tread abrasion against the road surface, and represent a major contributor of microplastic emissions to the environment (Knight et al., 2020).

Microplastic removal from stormwater is crucial to mitigate the associated environmental and human impacts (Stang et al., 2022). Conventional drainage systems collect and transport microplastics via storm and combined sewers to the wastewater treatment plant (WWTP). Although microplastics are efficiently captured at this stage, they are transferred into sewage sludge and reach the natural environment via agricultural applications (Calzadilla et al. 2023; van den Berg et al., 2020). In some cases, when the amount of runoff exceeds the capacity of the drainage system, combined sewer overflows (CSOs) occur, and microplastics can be discharged into water bodies without treatment (Zhou et al., 2023; de J Piñon-Colin et al., 2020).

To reduce these negative impacts, stormwater should be treated before it reaches the aquatic ecosystem (Walsh et al., 2012). Sustainable urban drainage systems (SUDS) are engineered nature-based solutions for sustainable urban stormwater management that deliver multiple benefits, such as water quality improvement, or prevention from urban diffuse pollution among others, promoting several processes such as infiltration, filtration, retention/detention and treatment (García-Haba et al., 2022; Andrés-Doménech et al., 2021). SUDS comprise a wide variety of structures, e.g. permeable pavements, vegetated swales, bioretention structures or constructed wetlands, etc. (Ballard et al., 2015). Some of them have been studied for their capacity to capture microplastics (Bodus et al., 2024). Sedimentation-based systems, such as wetlands or ponds, and filtration-based systems, including bioretention cells or gardens, have been shown to effectively retain a significant number of MP within the sediment and the filter media. However, there is still lack of knowledge in the application of SUDS to tackle the problem of microplastics (García-Haba et al., 2023b).

Permeable pavements allow water to soak into its layered structure, where it is temporarily held before it infiltrates into the ground, enters the drainage network, or is harvested for a possible later use (Ballard et al., 2015). It is widely demonstrated that permeable pavements significantly improve water quality through filtration processes (Hernández-Crespo et al., 2019; Fernández-Gonzalvo et al., 2021; Razzaghmanesh and Borst, 2019). Despite the limited research to date, these structures seem a promising solution for microplastic retention (Kong et al., 2024; Mitchell and Jayakaran, 2024; Rasmussen et al., 2023). The incorporation of adsorbent materials within the structure of a permeable pavement could help in the retention of microplastics due to their immobilisation capacity in the aqueous phase (Chen et al., 2022b). However, uncertainty remains about the fate of microplastics in the inner layers of permeable pavements, and about the effectiveness of this technology compared to other solutions. Additionally, the underlying migrating processes within the permeable structure are not fully understood (Österlund et al., 2023).

By means of a laboratory approach, this investigation aims to evaluate the efficiency of permeable pavements for the control of microplastic and tire wear particles. The novelty of this study lies in the layer-by-layer analysis of retained microplastics, providing new insights into the mechanistic understanding of micropollutant retention by permeable pavements. This was achieved by simulating synthetic rainfall and percolation processes of surface-accumulated dust and sediment, and the application of real runoff. The incorporation of adsorbent material within a permeable pavement was tested to analyse its influence on the retention of microplastics. Drinking water treatment sludge, combined with granular filter media, was used with the objective of laying the basis for future research.

2. Materials and methods

Two experiments (E1 and E2) were accomplished in this research. Information regarding the setup and the specific objectives pursued in each experiment is summarised in Table 1 below. The details of each experimental configurations are explained within the next subsection.

2.1. Materials

2.1.1. Infiltrometer 1, 2 and 3 setups (experiment E1)

Each infiltrometer was composed of three main parts: input setup, permeable pavement structure, and output setup (Fig. 1).

The input setup comprises a rainfall simulator (Rodriguez-Hernandez et al., 2015), composed of a water supply tank, two peristaltic pumps (LLG-uniPERISTALTICPUMP 3), and a grid of drip irrigation pipes with a plastic mesh to break rainfall drops (Naves et al., 2020).

The permeable pavement structure (50 cm wide and 50 cm long) is placed 40 cm below the rain simulator and is assembled inside a 0.25 m^2 bottomless plastic chamber. The cross section was defined according to Ballard et al. (2015): surface, base, and sub-base. Three different surface configurations were tested. Configuration 1 consists of permeable interlocking concrete pavement (PICP) using fine gravel (3–6 mm) as joint filler; configuration 2 consists of PICP but considering a mixture of 80 % fine gravel and 20 % drinking water treatment sludge (DWTS) in the joint filler, hereafter PICP-DWTS (García-Haba et al., 2021a); and configuration 3 consists of porous concrete pavement (PCP). Each permeable pavement structure had the same base and sub-base layers, consisted of fine washed gravel (2–4 mm), and coarse washed gravel (25–40 mm) respectively. The permeable pavement structure is supported on a 52 mm thick drain cell and includes nonwoven polypropylene-based geotextile (thickness of 0.15 mm, opening size of 0.09 mm, and vertical permeability of 90 mm/s) between base and sub-base, and between sub-base and drain cells for separation/retention purposes (Fig. 1). For the output setup, infiltrated water is collected through a receiving stainless-steel hopper, and a precision balance (611–2299 VWR) is placed below each infiltrometer to measure the

infiltrated water volume. Distance between hopper and sampling glass was approximately 20 cm.

2.1.2. Storage tank setup (experiment E2)

The experimental setup of the storage tank follows the same pattern as configuration 2 in experiment E1, except that only 1 geotextile was placed between fine gravel and coarse gravel layers, as no free infiltration was allowed (Ballard et al., 2015). A drainpipe was included at the bottom of the system to allow water extraction and level regulation inside the storage tank (García-Haba et al., 2021b) (Fig. 2).

2.1.3. Road dust and sediment (experiment E1)

To simulate street pollution build-up in E1, real dry dust and sediment was collected from an intense traffic road within the campus of the Polytechnic University of Valencia (UPV) (Fernández-Gonzalvo et al., 2021; Andrés-Doménech et al., 2018). Samples were collected by mechanical vacuum-sweepers (Yang et al., 2022) and sieved to a size of 5 mm.

2.1.4. Rainwater (experiments E1 and E2) and stormwater runoff (experiment E2)

To simulate rainwater in experiments E1 and E2, deionised water was used (10 μS/cm and pH of 7.1), given its analogous properties to the analysed rainwater in the Valencian region (Perales-Momparler et al., 2017). Regarding runoff, real samples from rain events were collected by an automatic sampler (ISCO3270), equipped with a liquid detector and a set of 24 1-l bottles from a small impervious urban catchment (Andrés-Doménech et al., 2018). This runoff was a direct surface input in experiment E2.

2.2. Methods

2.2.1. Rainfall simulation (experiments E1 and E2) and pollution build-up (experiments E1)

A long-term simulation period encompassing progressive pollution build-up in the permeable pavements under dry-rain conditions was conducted (Fernández-Gonzalvo et al., 2021). The simulated rainfall reproduced the average regime of Valencia (Spain): events with a rainfall volume of 16 mm and 30 min duration (32 mm/h) (Andrés-Doménech et al., 2010), whereas pollution build-up was simulated by means of manual dispersion of dry dust and sediment on the pavement surface. A visually uniform and homogeneous layer of sediment was formed, so that the sediment percolation was as regular as possible. A deposition rate of 5 g/m^2/d was selected according to Hernández-Crespo et al. (2019). The simulation process consisted of the application of dust on each permeable pavement and later the rainfall simulation. This process was carried out every 2 weeks for a 2-year period for PICP-DWTS and PICP infiltrometers, and a 5.5-year period for PCP. After each simulation process, the stainless-steel hopper was thoroughly rinsed with deionised water to ensure that any potential adhering microplastics were included in the infiltrated water.

2.2.2. Operation of the storage tank (experiment E2)

During a four-month period, the operation procedure consisted of two main processes: filling of the storage tank with direct rainwater and real runoff, and water extractions from inside the storage tank (Fig. S4). The filling process took place when a rain event occurred. The methodology consisted of gradually application of runoff and rainwater volumes onto the pavement bricks. A glass vessel was used to ensure uniform distribution, thus mitigating the risk of excessive washing of sediment accumulated on surface during previous filling processes. Entered rainfall and runoff volumes corresponded to those monitored within the campus catchment (Andrés-Doménech et al., 2018). Water extractions represented filtered effluents through the permeable pavement structure. Water extractions simulated a fictitious regular and cumulative demand for irrigation water, alongside potential overflows that may occur as consequence of a complete filling of the tank. Water abstractions for irrigation were carried out every 2 weeks, and represented the accumulated water volume demand for irrigation over the two-week interval. The irrigation volumes were estimated as function of rainfall records and the potential evapotranspiration rate recorded at a nearby meteorological station. Occasionally, overflows occurred when the water level exceeded the surface layer after the filling of the tank. The excess water was then withdrawn through the drainpipe until it equilibrated with the uppermost level of the surface layer (see Fig. S4). The water extractions were carried out by gravity through the drainpipe, by simulating small discharge flows to avoid the dragging of solids that could enter a hypothetical pipe network, and consequently, cause blockages.

2.2.3. Sample collection

The microplastic sampling campaign for E1 started in January 2022 which means 1 year after the start of the experiment for PICP-DWTS and PICP infiltrometers, and 5 years after for PCP. Samples of infiltrated water were immediately taken after a simulation process, using 2 L glass beakers for collection. In total, 6 effluent samples were taken from PICP-DWTS and PICP configurations, and 9 from PCP. Volumes of sampled effluents ranged between 300 and 700 mL.

In relation to E2, a total of 11 water extractions took place. Therefore, 11 samples were taken. It is important to notice that only 2 of them corresponded to overflows. The small number of overflow samples represent a limitation of the study. A 1 L sample from every water extraction was taken and analysed. A metal bucket was used for water collection. Samples of water extractions were immediately taken after the discharge process.

At the end of the experiments, the permeable pavement structures were disassembled, layer by layer. Retained dust and sediment on the surface was collected with a vacuum cleaner and processed as solid matrix. To mitigate the risk of cross-contamination between samples, the vacuum cleaner was rinsed with ultrapure water following each sampling to ensure that any adhering microplastic was included in the samples. MP retained in granular layers were washed-off using deionised water. Washing water was then filtered through a 40-μm sieve. If

Table 1
Summary of characteristics and objectives of experiments.

Experiment	Device tested	Permeable pavement configuration	Water management system	Microplastic input source	Rainfall input	Direct runoff input	OBJECTIVES
E1	Infiltrometer 1	Configuration 1	Infiltration	Dry road dust and sediment	Yes	No	To study the influence of the type of surface pavement layer, and an adsorbent material (DWTS) on microplastic retention.
	Infiltrometer 2	Configuration 2			Yes	No	
	Infiltrometer 3	Configuration 3			Yes	No	
E2	Storage tank	Configuration 1	Harvesting	Real urban runoff	Yes	Yes	To study the influence of the microplastic input source, and the structural configuration on microplastic retention.

the solid content intercepted in the sieve was abundant, it was processed as solid matrix. Otherwise, the sieve was washed with deionised water and the sample was processed as liquid matrix. Geotextile layers were wrapped in aluminium foil for subsequent visual inspection.

2.2.4. Sample processing and analysis

2.2.4.1. MP extraction. Liquid samples (stormwater runoff and, infiltrated and extracted water) were filtered through glass fibre filters with a nominal pore size of 1.2 μm and diameter 47 mm. For solid matrices, a pre-treatment was applied to remove organic matter, while minimizing alteration of MP. The pre-treatment consisted in two digestions. For the first digestion, 150 mL of an acidic oxidant solution (H_2SO_4 1:1 H_2O_2), that do not alter the chemical structure of microplastics (Hernández-Arenas et al., 2021), was slowly added to sediment subsamples (5–10 g). To prevent the sample from burning due to the exothermic reaction produced when the solution contacts organic matter, it was added in 10 mL fractions every 30 min. Once all the reagent was added, it was left to react for 72 h in a gas chamber. Then, the solution was filtered, and a second digestion was performed with Fenton reagent, which produces an advanced oxidation process using H_2O_2 in the presence of a catalyst (Fe^{2+}). The Fenton reagent does not produce chemical or morphological changes in different polymers (Pfohl et al., 2021), being a suitable reagent for the extraction of microplastics from a solid matrix. During the addition of the reagent, an ice bath was used to keep temperature below 40 °C (Hurley et al., 2018). Once the Fenton was added, the samples were mixed at 100 rpm for 5 days to ensure that all the organic matter of the sample was in contact with the reagent. Subsequently, they were left in the oven at 45 °C for 4 days.

Afterwards, MP were extracted by density separation using NaCl at saturation. The selected salt was NaCl, (with a concentration of 322 g/L and a density of 1.2 g/cm^3) due to its low cost and lack toxicity (Bellasi et al., 2021). This method allows settlement of the sediment to the bottom of the beaker and promotes microplastic flotation. Once the saturated solution was added to the previously digested sample, the mixture was stirred with a stirring rod to ensure a homogeneous distribution of microplastics. After that, the sample was left to settle for 24 h to ensure that separation between inorganic matter and MP occurred. Then, the upper part of the water column was filtered ensuring their extraction, following the liquid matrix process. The extraction process was repeated four times. To improve the recovery rate of MP of high dense particles in every sample, sodium bicarbonate was added only in the fourth extraction, promoting flotation of microplastics via CO_2 bubbles release (Hernández-Arenas et al., 2021). This method was validated in previous works, with a recovery percentage of 87 ± 2 % (mean ± SE) (Hernández-Arenas et al., 2021), being efficient in the extraction of microparticles of Polypropylene (PP), Low Density Polyethylene (LDPE), High Density Polyethylene (HDPE), and Polyethylene Terephthalate (PET), which are the most commonly used plastics worldwide (Plastics Europe, 2022) and expected to be found in this study.

2.2.4.2. Polymer identification and size estimation. All filters (from both liquid and solid matrices) were visually analysed using a stereomicroscope (MOTIC SMZ 143 N2GG, Wetzlar, Germany) coupled to an integrated camera to select potential microplastics. This combined equipment achieves a maximum resolution of 10–20 μm. Identified particles counted as possible microplastic were analysed with Raman spectroscopy. The laser excitation frequency and intensity used were 784.79 nm and 11.8 mW, respectively. Raman spectra were recorded with a charge-coupled device camera (UV-NIR range, 1024 × 255 pixels) electrically cooled to −70 °C. Raman spectra were obtained in the range 162–1886 cm^{-1} with a spectral resolution of 2.47 cm^{-1}.

To determine whether the material of identified particles was plastic, polymer spectra obtained from the Raman spectrometer (NRS-5100, Jasco) and from the Open Specy 55 spectral library were compared (Cowger et al., 2021). Special attention was paid to tire wear particles due to its high presence in the samples. To ensure the correct identification of this type of microplastic, samples from different tires and different levels of wear were previously analysed by means of Raman spectroscopy and added to the library used for polymer identification. Two factors were considered to perform the classification: the similarity degree between the two spectra, expressed as the coincidence factor R (Huppertsberg and Knepper, 2018), and the coincidence between the main bands of both spectra. The closer the value of R is to 1, the closer the spectrum of the analysed particle resembles the spectrum of a plastic type in the database. When $R > 0.7$ the correlation between the two spectra is considered as high. However, it might occur that although the R value is low (< 0.7), the particle is made of the plastic material of the reference spectrum in the database. Microplastics in the environment

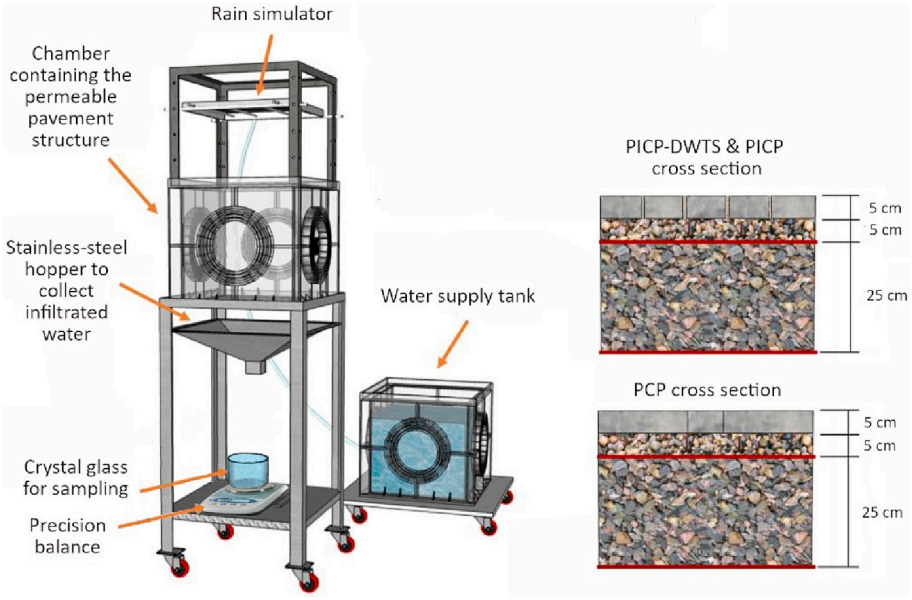


Fig. 1. Scheme of permeable pavement infiltrometer setup (input setup, permeable pavement structure and output setup) and cross sections. Geotextiles are represented as horizontal red lines (adapted from Fernández-Gonzalvo et al., 2021).



Fig. 2. Details of the storage tank setup and cross section. Geotextile is represented as horizontal red line.

suffer weathering which results in the modification of their original spectrum, which is used as their fingerprint by spectroscopic equipment such as Raman to identify its polymeric composition. Additionally, some materials may be made of a composition of several types of plastic. In these cases, the assignment of the tentative particles to virgin (non-weathered) plastics is hindered. To overcome this issue, SLoPP and SLoPP-E databases, which contain spectra of degraded polymers (Munno et al., 2020), were used. Spectra of all microplastic particles detected in the study are available upon demand.

To estimate the size of all the particles analysed, a Java-based public domain image processing software developed by the U.S. National Institutes of Health (ImageJ) was used (Rosal, 2021). Size ranges of <0.1 mm; 0.1–0.5 mm; 0.5–1 mm and 1–5 mm were considered for microplastic identification. The smallest identified microplastics measured 10 μm in size.

Microplastic identification from geotextile samples was visually performed using a stereomicroscope with augmentations ranging from x20 to x40 (SZ30, Olympus, Japan). Only the morphology of the particles detected was estimated. It is worth noting that the microparticles detected in the geotextiles were not confirmed as microplastics through spectroscopy, unlike those detected in other layers of the permeable pavement. Instead, "The Hot Needle Test" was used as an alternative identification method (De Witte et al., 2014).

2.3. Quality assurance and quality control

Quality control management was carried out during the whole experiments. Cross-contamination of samples was minimised by using aluminium and glass material that had been cleaned with ultrapure water, cotton laboratory coats and, whenever possible, clothes that were made of natural fibres. Additionally, containers holding the samples were covered to prevent airborne contamination of microplastics (Krüger et al., 2020). The NaCl solution was always previously filtered to remove any possible microplastics in the salt. Blanks from the laboratory environment and from component materials of permeable pavements were analysed (Table S6). However, they have not been considered to correct the sample results due to uncertainty regarding whether all samples were equally contaminated. Samples of plastic materials involved in the experiments, including the geotextile, were analysed with Raman and spectra were added to the database in order to detect contamination generated by them. No particles from these materials were found.

2.4. Statistics

Statistical analyses were performed using STATGRAPHICS Centurion 19 (version 19.1.2). The effects of the permeable pavement configuration (PICP-DWTS, PICP and PCP) on microplastic levels at infiltrated water was studied. ANOVA test was used if normality was satisfied whereas Kruskal-Wallis was used for non-normal distributions. Furthermore, the assessment of the impact of permeable pavement layers on the internal accumulation of MP was accomplished by comparing microplastic retention percentages within each layer, considering that vertical profiles were related samples. In this case, t-student test was used if normality was met and Wilcoxon model otherwise. Statistical significance was indicated by a probability of a type I error of 5 % or less ($p \leq 0.05$). No comparison was undertaken between storage tank and other setups due to dissimilarities in their operational procedures. To measure the relationship between accumulated sediment on the pavement surface and microplastic levels in infiltrated water, linear correlations between variables were obtained using either Pearson correlation coefficient (for normal distribution variables) or Spearman correlation coefficient (for non-normal distribution variables). It was assumed that correlation coefficients were statistically significant with a probability of a type I error of <5 % ($p \leq 0.05$).

3. Results and discussion

3.1. Levels and characterisation of MP in road dust and sediment

Average level of microplastics in road dust and sediment was 4762 ± 974 MP/kg ($n = 3$). Similar results were reported by other studies that also studied microplastics in road dust: 2279 ± 914 MP/kg, n = 3 (Patchaiyappan et al., 2021); and from 2777 ± 333 to 20,167 ± 333 MP/kg (Dehghani et al., 2017). Polymers found were PET, Polymethylstyrene (PMS), Polyester, PP, Polyvynyl Chloride (PVC) and Polyvinyl alcohol (PVOH), where PP (75 %) was the most prevalent (Fig. 3). This is consistent with the polymers demanded by markets, mainly for household and industrial purposes (Plastics Europe, 2022; Monira et al., 2021). In relation to morphology, fragments were the most common shape (90 %). This finding is also in line with Patchaiyappan et al. (2021) (92.5 %) and Dehghani et al. (2017) (65.9 %). Fragment type microplastics usually originate from the decomposition plastic waste, or from the wear and tear of plastic products (Grbić et al., 2020). Looking at size levels, most of the particles were classified in the smallest size fraction (86 %).

3.2. Levels and characterisation of MP in stormwater runoff

Microplastic levels in the stormwater runoff samples greatly varied from 2.00 to 110.59 MP/L (Table S4). Previous studies that also investigated the presence of microplastics in stormwater runoff, show a significant variation in microplastic levels, ranging from <1.00 to 8550 MP/L, due to differences in catchment land uses, rainfall intensities and duration, and methodological procedures, making comparison difficult (C. Wang et al., 2022). In average, runoff MP level was estimated at 23.90 ± 17.40 MP/L ($n = 6$), which is similar to that determined by Ross et al. (2023), who obtained a mean of 31.90 ± 34.00 MP/L ($n = 7$), in the analysis of stormwater runoff samples from different types of urban catchments. Polymers detected were PP (62 %), PVOH (23 %), PVC (6 %), Polyester (4 %), PMS (2 %), HDPE (2 %), PET (1 %) and Acrylonitrile Butadiene Styrene (ABS) (0.1 %) (Fig. 4). With respect to morphology, fragments represented the highest fraction (84.7 %). Other authors such as Chen et al. (2022a) and Sang et al. (2021) also detected fragments as the most common shape in road runoff. Regarding to size, most of the particles were within the 0.1–0.5 mm range (61.3 %).

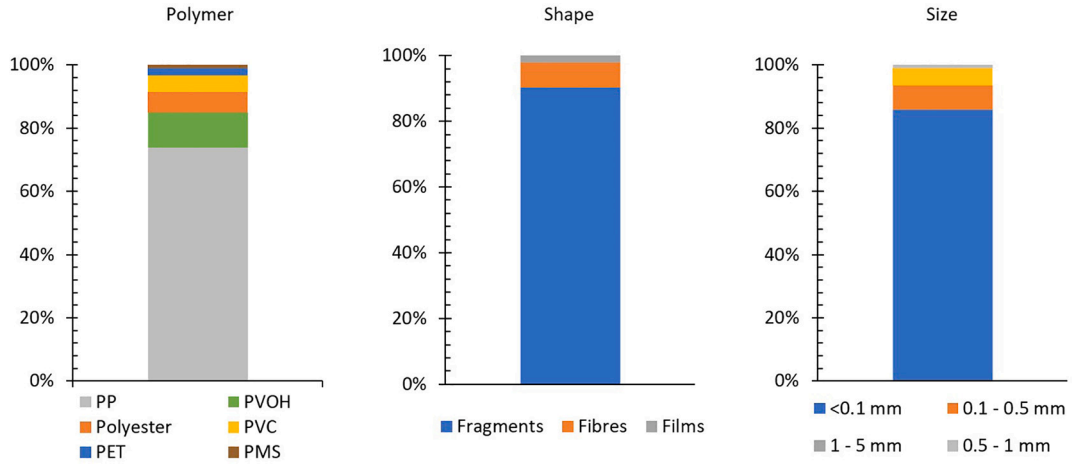


Fig. 3. Microplastic distribution in road dust and sediment, according to polymer, shape and size in average percentage. Total sampled dry weight = 19.35 g (n = 3).

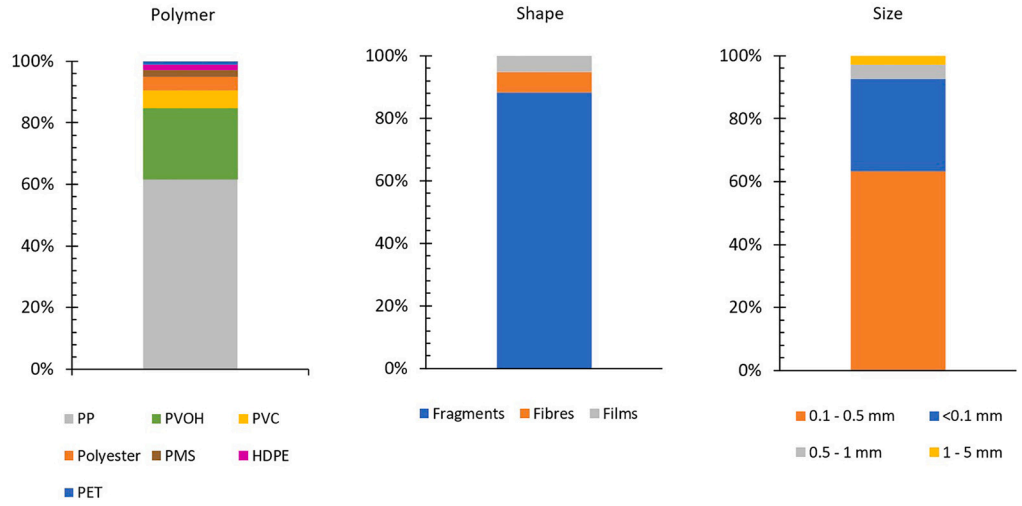


Fig. 4. Microplastic distribution in stormwater runoff according to polymer, shape, and size in average percentage. Total sampled runoff = 153 L (n = 6).

3.3. Levels and characterisation of MP in infiltrated water

3.3.1. Infiltrometers (experiment E1)

Estimated average microplastic levels in the infiltrated water through PICP-DWTS, PICP and PCP were 2.22 ± 1.03 MP/L (n = 6), 2.99 ± 1.00 MP/L (n = 6) and 5.17 ± 1.05 MP/L (n = 9) respectively. No significant differences in output microplastic levels between infiltrometers were detected ($p > 0.05$), and no correlation between surface dust and sediment accumulation and microplastic abundance in the effluent of every infiltrometer was found: PICP-DWTS ($r = 0.10$; $p > 0.05$); PICP ($r = -0.19$; $p > 0.05$); PCP ($r = 0.44$; $p > 0.05$).

Similar percentages of fibres, films and fragments were released from PICP-DWTS (39.3 %, 35.7 % and 25 %) (Table 1). In PICP, fragments and fibres were prevalent (46.4 and 44.3 %). In case of PCP samples, the highest microplastic shape proportion corresponded to fragments (89 %). Regarding to size, most of the particles released in PICP and PCP effluents were below 0.1 mm (55.7 and 81.4 % respectively), while 0.5–1 mm particles were prevalent in PICP-DWTS (64.3 %). The higher proportion of intermediate-sized particles escaping from PICP-DWTS, mostly represented by fibres, can be attributed to their perpendicular alignment with the filtering matrix, enabling passage through the layers due to their smaller diameter compared to their length (Choi et al., 2021).

From the results, it seems that permeable pavement structures present difficulties at retaining the smallest size fraction and fragment shaped particles (Table 2). This is particularly noticeable in PCP configurations. This fact suggests that the permeable surface configuration could influence on retained particle characteristics. For example, PCP are much better than PICP at stormwater treatment as consequence of a smaller porous structure (Kuruppu et al., 2019; Lucke and Beecham, 2013).

Comparable microplastic levels at PICP-DWTS and PICP effluents might suggest that DWTS does not have influence in microplastic capture. This result is surprising since recent studies reported that reactive porous media have positive influence on the removal and immobility of microplastics (Abuwatfa et al., 2021). Retention efficiencies higher than 95 % were obtained for Polystyrene (PS) spheres (1–10 μm), Polyethylene (PE) cylindrical fragments (2–3 mm) and fleece fibres, using biochar with pore sizes ranging from <2 to 50 nm (J. Wang et al., 2021; Siipola et al., 2020; Z. Wang et al., 2020). It is plausible that the proportion of DWTS used was insufficient to capture significant amount of particles.

3.3.2. Storage tank (experiment E2)

Average MP level in water extractions for irrigation was estimated at 2.00 ± 0.73 MP/L (n = 9). PP particles represented once again the highest fraction (44 %) (Fig. 5). Fragments represented 61.6 % of the particles, followed by fibres (31 %) and films (7.3 %). In relation to size,

Table 2
Microplastic proportions and levels according to polymer, shape and size identified in permeable pavement effluents (percentage and mean concentration ± standard error). Percentage refers to sampled effluent volumes. Empty cells represent that no MP were detected.

	PICP (n = 6) Experiment E1, configuration 1		PICP-DWTS (n = 6) Experiment E1, configuration 2		PCP (n = 9) Experiment E1, configuration 3	
Polymer	MP proportion (%)	MP level [MP/L]	MP proportion (%)	MP level [MP/L]	MP proportion (%)	MP level [MP/L]
ABS	9.3	0.27 ± 0.30			3.5	0.18 ± 0.18
LDPE			12.5	0.27 ± 0.27		
PET	9.3	0.27 ± 0.30	12.5	0.27 ± 0.27	3.6	0.18 ± 0.18
Polyester	9.3	0.27 ± 0.30	26.8	0.59 ± 0.37	3.5	0.18 ± 0.18
PMS	9.3	0.27 ± 0.30			10.7	0.55 ± 0.39
Polyphenylsulfone			12.5	0.27 ± 0.27		
PP	27.9	0.83 ± 0.62	35.7	0.79 ± 0.55	54.3	2.80 ± 0.67
PVC	9.3	0.27 ± 0.30				
PVC + PP	16.4	0.49 ± 0.53				
PVOH	9.3	0.27 ± 0.30			24.3	1.25 ± 0.91
Shape						
Films			35.7	0.79 ± 0.55		
Fibres	44.4	1.32 ± 0.69	39.4	0.87 ± 0.39	11.4	0.5 9 ± 0.29
Fragments	46.5	1.38 ± 0.56	24.9	0.55 ± 0.35	88.6	4.58 ± 1.08
Pellets	9.1	0.27 ± 0.30				
Size						
< 0.1 mm	55.7	1.66 ± 0.66			81.4	4.21 ± 1.05
0.1–0.5 mm	9.3	0.27 ± 0.30	35.7	0.79 ± 0.55	3.6	0.18 ± 0.18
0.5–1 mm	9.3	0.27 ± 0.30	64.3	1.42 ± 0.79	11.5	0.59 ± 0.29
1–5 mm	25.7	0.76 ± 0.56			3.6	0.18 ± 0.18

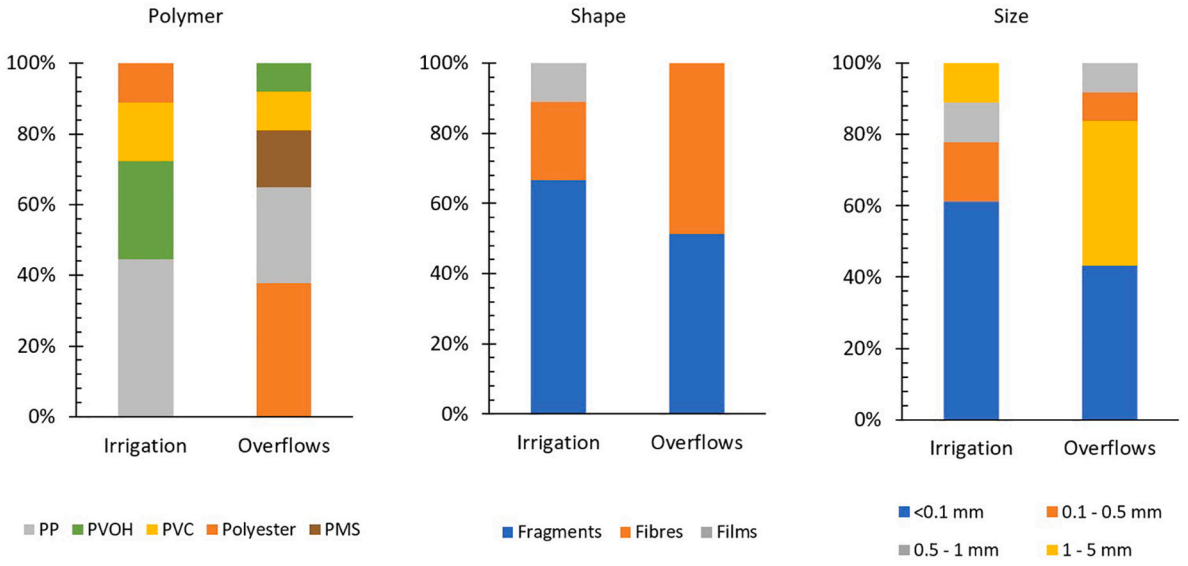


Fig. 5. Microplastic distribution in water extractions from the storage tank for irrigation (n = 9) and in storage tank overflows (n = 2) according to polymer, shape, and size in average percentage.

small MP (< 0.1 mm) were predominant (55.2 %). Similar percentages were estimated between the size ranges 0.1–0.5 mm (13.7 %), 0.5–1 mm (10.1 %) and 1–5 mm (21 %). In case of water overflows, average MP level was estimated at 3.08 ± 0.42 MP/L (n = 2). Polyester was the most frequent polymer (38 %), followed by PP (27 %). Only fragments and fibres were present in the overflows (51 % and 49 %), and predominant sizes were < 0.1 mm (43 %) and 1–5 mm (41 %).

3.4. Microplastic retention efficiency, accumulation and fate of MP retained within the permeable pavement layers

3.4.1. Infiltrometers (experiment E1)

A total of 4345 microplastic particles were estimated as input in both PICP-DWTS and PICP infiltrometers, and 11,030 MP in PCP. No significant differences were detected in the quantity of MP retained within all permeable pavement infiltrometers ($p > 0.05$). The number of MP in the permeable pavement outputs were estimated at 145 (PICP-DWTS), 186 (PICP) and 1240 (PCP). Hence, estimated retention efficiencies of the permeable pavements were 96.7 % (PICP-DWTS), 95.7 % (PICP), and 88.8 % (PCP). The lack of similar studies to compare these results with other authors findings, suggests looking at similar indicators such as TSS, which have a strong correlation with MP (Q. Wang et al., 2020). TSS retention efficiencies in permeable pavements have been reported from 91 to 96 % (Zheng et al., 2022); 70 to 80 % (Liu et al., 2020); up to 99 % (Hernández-Crespo et al., 2019); and 100 % (Kamali et al. (2017).

Significant amount of MP was accumulated on permeable pavement surfaces over the other layers (Fig. 6). This finding is directly related to sediment accumulation, as consequence of the formation of an impermeable matrix or "crust", which is generally limited to the upper few centimetres of the surface structure (Brown et al., 2009; James, 2004; James and Gerrits, 2003; Pratt et al., 1995; Baladès et al., 1995). Therefore, surface sediment accumulation might be considered as an integral component of the permeable pavement system's retention efficiency as it can be effectively removed by cleaning services. Although

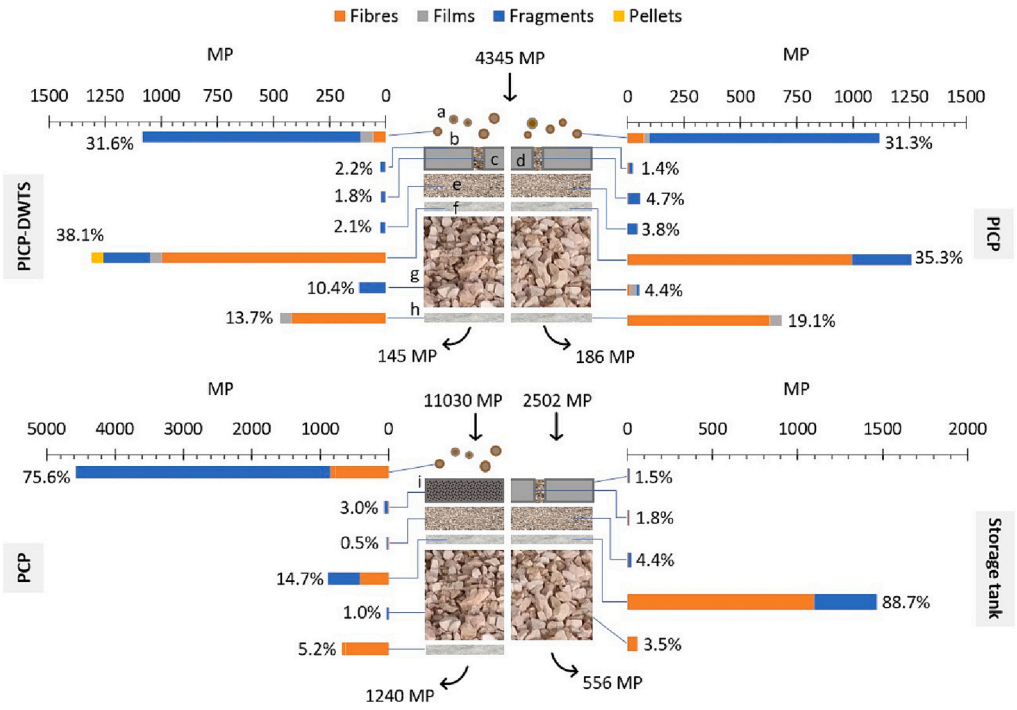


Fig. 6. Total MP estimated at input and output of the permeable pavements, MP fate (%) and shape distribution and accumulation within the permeable pavement layers (a. Dust and sediment accumulated on surface; b. Concrete brick; c. Filler+DWTS; d. Filler; e. Fine gravel; f. Upper geotextile; g. Coarse gravel; h. Lower geotextile; i. Porous concrete brick). Microplastics found in geotextiles lack spectroscopic confirmation, unlike those identified in other layers which have been spectroscopically confirmed.

this study did not characterise the porosity of the permeable joint in the PICP or the porous surface of the PCP, it is thought that the pore size of the latter is generally smaller than in the permeable joint (Fig. S3). This fact suggests that PCP can be more easily clogged by small particles and contributes more than PICP to the formation of the "crust" previously commented. This hypothesis is plausible since fine sediments are prone to cause permeability loss in porous asphalt (García-Haba et al., 2023a). In this study, identified microplastic sizes are grouped in the mentioned size ranges, and consequently, might contribute to the clogging process of PCP (Rasmussen et al., 2023). The high proportions of fragments observed in the accumulated dust and sediment (89.7 % in PICP-DWTS; 91.4 % in PICP; and 81.4 % in PCP) could be attributed to the morphology itself. Fragments, which are generally angular shaped, might be susceptible to get trapped by other particles contained in the sediment, resulting in a barrier effect on MP to enter the system.

In the inner structure, geotextiles play a major role in microplastic retention compared to other layers, more specifically for fibres capture. Upper geotextiles captured 38.1 % (PICP-DWTS), 35.3 % (PICP), and 14.7 % (PCP), whereas lower geotextiles retained 13.7 % (PICP-DWTS), 19.1 % (PICP), and 5.2 % (PCP). Fibres were the most frequent shape retained in the geotextiles installed in PICP based infiltrometers, while fibres and fragments were found in equal proportions in PCP. This result agrees with previous studies wherein geotextiles improved TSS removal efficiency in permeable pavements by about 30 % (Kamali et al., 2017; Rowe et al., 2009). Caution should be exercised regarding the obtained results, as the particles suspected to be microplastics were not verified by spectroscopy, but The Hot Needle Test, which may be less precise than the former. This fact could potentially affect the reported particle quantities. Looking at other permeable pavement layers above the upper geotextile (filler and fine gravel) and below (coarse gravel), they had very little influence on retaining microplastic particles (Fig. 6).

A high variability in the retention capacity of the rest of the layers was observed in the three infiltrometers, being fragments and sizes from <0.1 mm up to 0.5 mm the most common particles retained. Likely, fibres can fit and adapt to void spaces of the granular layers, being susceptible to be washed off.

Recovery percentages, estimated as the relation between MP collected, encompassing particles recovered from the permeable pavement layers and contained in effluent, and the total amount of input MP applied via dust and sediment were 82 %, 86 % and 66 % for PICP-DWTS, PICP and PCP infiltrometers. These recovery percentages are within the range reported in other studies assessing the vertical fate of MP (Q. Wang et al., 2021), except for PCP. An important fraction of particles might have been retained within the inner pores of this type of pavement, rendering them inaccessible for analysis.

3.4.2. *Storage tank (experiment E2)*

At the end of the experiment E2, a total of 2503 microplastic particles were estimated as input and 556 MP as output. This results in a retention efficiency of 77.8 % (Fig. 6), which is within the range of other studies that reported TSS retention efficiencies of 54 % (Ghisi et al. (2020) and 98.8 % Winston et al. (2020). The smaller retention efficiency of the storage tank in comparison to the other infiltrometers, might be supported by the presence of only one geotextile. These findings provide evidence that the retention of MP can be effectively achieved in permeable pavements when more than one geotextile is incorporated into their structure.

Microplastic distribution in the storage tank layers follows a similar pattern than in the infiltrometers. As an exception, no sediment was accumulated on the surface. This fact can be explained as the total sediment mass contained in stormwater runoff samples was 25 times lower than the total dust and sediment used in E1. Another reason that could explain this situation would be the way in which the runoff was applied to the permeable pavement surface. Particles accumulated on the surface could have been washed-off by the runoff flow, entering the system through the joints between bricks. According to the results, approximately 30 % more microplastics entered the system via runoff. Geotextile is once again revealing itself as the main microplastic trapping layer (88.7 %), mainly fibres, followed by fine gravel (4.4 %), coarse gravel (3.5 %), filler (1.8 %), and concrete brick (1.5 %). In the

layers above the geotextile, fragments (75 % concrete brick; 43 % filler) and films (43 % filler) represented the major retained fraction, whereas only films were found in the coarse gravel layer. The particle recovery percentage was estimated at 88 %.

3.5. Tire wear particles

3.5.1. Levels and characterisation of TWP in road dust and sediment, and stormwater runoff

Tire wear particles constituted a significant fraction of the total microplastics accounted in this study (51 % in road dust and sediment, and 40 % in stormwater runoff). This finding confirms that TWP make substantial contribution to microplastics accumulated on urban surfaces, susceptible to being washed-off (Österlund et al., 2023), and that stormwater runoff represents a major source of TWP, contributing to microplastic pollution, as they are estimated to account for 5–10 % of the total plastic entering the global ocean (Jan Kole et al., 2017). All the particles detected in road and runoff samples measured <0.5 mm, with 80 μm being the smallest size identified.

Calculated mean level of TWP in road dust and sediment was 4935 ± 1040 TWP/kg (19.35 g of total sampled dry weight; $n = 3$), which is in the range of other reported TWP levels: 340 ± 45.5 TWP/kg dw, n = 3 (Worek et al., 2022); 3600–9100 TWP/kg dw, (Järlskog et al., 2020); and 854,832 ± 1,071,314 TWP/kg dw, n = 3 (Järlskog et al., 2021). Wide variation in TWP levels in road dust and sediment is influenced by various factors, including maintenance practices, weather conditions and traffic level (Wagner et al., 2018; Jan Kole et al., 2017). Most of previous studies reported TWP levels in units of particle mass/wet sediment volume and/or particle mass/dry sediment mass (e.g., Rasmussen et al., 2023; Knight et al., 2020; Klöckner et al., 2019), making it difficult to compare between results. This study provides more reliable data as they are reported in number of particles per dry weight, which is a more comparable unit of measurement.

In stormwater runoff samples, TWP average level was estimated at 7.95 ± 1.75 TWP/L (total sampled runoff = 153 L, $n = 4$), which falls within the range of other TWP levels reported by previous studies: 20.40 ± 5.70 TWP/L, $n = 23$ (Mahjoub et al., 2023); 31.62 ± 29.41 TWP/L, $n = 13$, (Smyth et al., 2021); 219 ± 129 TWP/L, $n = 6$ (Järlskog et al., 2021); 0.064 TWP/L, n = 3 (Gilbreath et al., 2019). Several factors such as traffic load, catchment features, and rainfall conditions can be attributed to have significant influence on large variations of the TWP levels reported (Järlskog et al., 2020; Liu et al., 2019).

3.5.2. Levels and characterisation of TWP in infiltrated and extracted water

No significant differences in output TWP levels between infiltrometers were detected ($p > 0.05$). However, significant correlation between surface dust and sediment accumulation and TWP abundance in the effluent of PICP-DWTS ($r = 0.9$; $p < 0.05$) and PICP ($r = 0.65$; $p < 0.05$) was found. Similar average levels of TWP compared to those obtained in the analysis of MP: 0.28 ± 0.28 TWP/L (PICP-DWTS; n = 6), 2.71 ± 0.82 TWP/L (PICP; n = 6)), 3.30 ± 0.89 TWP/L (PCP; $n = 9$), 2.67 ± 0.65 TWP (Irrigation water extracted from the storage tank; n = 9) and 4.67 ± 3.33 TWP/L (Storage tank overflows; n = 2). Identical pattern of the TWP size distribution to that of MP was detected: prevalent sizes corresponded to 0.1–0.5 mm fraction in PICP-DWTS (100 %), and < 0.1 mm in PICP (56 %), PCP (65 %), and Storage tank (69 %). The substantial difference in TWP levels between PICP-DWTS and PICP, coupled with the absence of particles smaller than 0.1 mm, might suggest that DWTS has an influential role on the removal mechanisms of these particles (Abuwatfa et al., 2021).

3.5.3. TWP retention efficiency of the permeable pavement

Input TWP applied on PICP-DWTS and PICP infiltrometers was 4503 particles, and 11,430 on PCP. Regarding to the infiltrated water, a total of 18, 169 and 791 TWP were estimated at PICP-DWTS, PICP and PCP effluents. Consequently, the experiment yielded remarkably high retention efficiencies of 99.6 %, 96.3 % and 93.1 %. The results obtained demonstrate the good performance of permeable pavements in the capture of TWP, preventing their access to natural ecosystems. A recent study also tested PCP structures and showed a TWP retention of over 96 % in terms of mass, which corroborates its high retention efficiency (Mitchell and Jayakaran, 2024). Regarding the storage tank, a total of 1035 TWP were applied as input, while 775 TWP were accounted in all water extractions. Therefore, the TWP retention capacity of the storage tank was 25 %. The observed reduction in efficiency, when compared to estimations from other infiltrometers, could be attributed to the relatively smaller number TWP inputs in comparison to number of MP. Furthermore, apart from only one geotextile layer was considered, all particle sizes were <0.5 mm, which are the most likely to escape from the system.

4. Conclusions

This study represents the first attempt of a complete estimation of the efficacy of permeable pavements in retaining microplastics, on a layer-by-layer focus. Moreover, it offers a comprehensive analysis of the characteristics, fate, and distribution of these microparticles present in road dust and sediment, stormwater runoff, and within the structure of a permeable pavement. The investigation demonstrates that permeable pavements represent a feasible and effective solution for retaining MP from urban stormwater runoff, including tire wear particles, thereby reducing their discharge into the natural environment. However, fragments and small particles (< 0.1 mm) represent a challenge, as these are the most common particles capable of eluding permeable pavements.

In general, when large dust and sediment contributions reach the permeable pavement surface, MP tend to accumulate on the permeable pavement surface (generally fragments) and in geotextile layers (mostly fibres). Despite clogging of permeable pavements is not desirable, the surface layer itself acts as a potential barrier for MP, which can be easily collected through appropriate mechanical maintenance tasks. In particular, PCP surface configurations captured more MP than PICP and geotextiles due to a smaller porous structure, which is more susceptible to clogging.

Geotextiles play a significant role in microplastic interception. Permeable pavement structures that comprise 2 geotextiles show higher microplastic and TWP retention efficiencies than permeable pavements with 1 geotextile layer. Optimal design of a permeable pavement should incorporate a minimum of two geotextile layers. In scenarios where infiltration is not feasible or when permeable pavements are designed for water storage and reuse, the drainpipe should be wrapped with geotextile to ensure particle retention, whenever high permeability is guaranteed.

It is important to highlight that after the whole testing period, no particles dislodged from the geotextiles, nor from other plastic parts of the experimental setups were observed in the effluents of the permeable pavements tested. Nonetheless, further research and implementation of non-polymeric materials such as natural fibres are recommended to potentially substitute this layer in order to reduce plastic use.

Further research should investigate the effect of rain intensity and duration, as well as the porosity of the materials, on microplastic mobilisation and fate under severe weather conditions. Additionally, although adsorbent materials have started to be studied for microplastic retention, it would be interesting to investigate on the incorporation of this material in permeable pavement structures to deliver additional and efficient retention capacity, particularly concerning particles smaller than 0.1 mm. Special efforts should be put on TWP to assess its fate and accumulation within filtration layers. This crucial step will enable the enhancement of permeable pavement design to prevent this widespread pollutant reach natural environments.

CRediT authorship contribution statement

Eduardo García-Haba: Writing – original draft, Investigation, Funding acquisition, Conceptualization. **Alba Benito-Kaesbach:** Writing – original draft, Methodology. **Carmen Hernández-Crespo:** Validation, Supervision. **Carlos Sanz-Lazaro:** Validation, Supervision, Methodology. **Miguel Martín:** Formal analysis. **Ignacio Andrés-Doménech:** Validation, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

Eduardo Garcia-Haba reports financial support was provided by Spain Ministry of Science and Innovation. Ignacio Andres-Domenech reports financial support was provided by Spain Ministry of Science and Innovation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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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.2024.172627.

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