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Research article

An integrative analysis of microplastics in spider webs and road dust in an urban environment–webbed routes and asphalt Trails

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1. Introduction

Microplastics (MPs), typically characterised as plastic fragments smaller than 5 mm (Barnes et al., 2009) or particles between 1 and 1000 μm (Hartmann et al., 2019), have been identified as an environmental concern due to their pervasive presence in many ecosystems around the globe (Andrady, 2011). Initially discovered in marine environments, subsequent research has elucidated their occurrence in multiple settings, such as freshwater bodies (Mintenig et al., 2020), terrestrial ecosystems (Büks and Kaupenjohann, 2020), and the atmosphere (Allen et al., 2021; Dris et al., 2017; Song et al., 2021). Studies have demonstrated that MPs can act as vectors for pollutants due to their sorption capacity (Pham et al., 2022; Torres et al., 2021) and have the potential to affect the fauna, for example by ingestion (Barnes et al., 2009). They are known to permeate through the food chain, causing detrimental ecological and potentially human health impacts (Browne et al., 2011). MPs' persistence and bioaccumulative nature have stimulated extensive research to understand their sources, fates, impacts, and solutions.

Urban areas have been identified as significant contributors to the release of MPs into the environment, primarily due to their dense population and concentration of human activities (Horton et al., 2017). These MPs find their way into aquatic ecosystems through various pathways, including wastewater discharges, combined sewage overflows (CSO), and the wind and rain-aided transport of road dust. In some matrices, such as wastewater, the concentrations of MPs have been extensively studied in this context (Mintenig et al., 2017; Murphy et al., 2016; Rasmussen et al., 2021; Roscher et al., 2022; Simon et al., 2018; Talvitie et al., 2017), while others, such as road dust, have been less researched (Monira et al., 2022; Morioka et al., 2023; O'Brien et al., 2021).

Road dust generally refers to the fine particulate matter generated from the abrasion of vehicle components, road surfaces, and the resuspension of previously deposited particles on roadways. Road dust can encompass many hazardous materials, including MPs, as highlighted by Monira et al. (2022), who, in their comprehensive review, indicated that road dust contributes to approximately half of the total MP load entering aquatic systems via stormwater runoff. This assertion is further corroborated by other studies examining MPs in stormwater pond sediments. Specifically, research by Liu et al. (2019a) and Molazadeh et al. (2023a,b) reported average concentrations of 3 μg MP g^{-1} and 11.8 μg MP g^{-1}, respectively in stormwater pond sediments.

There are several techniques used to quantify MPs in environmental matrices. Notably, μ-Raman spectroscopy and μFTIR are prominent methods that provide detailed information about particle counts and morphology and other research estimated as well a mass (Maurizi et al., 2023; Simon et al., 2018). Additionally, Pyrolysis GC-MS is used to directly assess the mass of MPs (Ivleva, 2021).

MPs have been detected in both indoor (Vianello et al., 2019) and outdoor (Dris et al., 2016) air. Further studies (Choi et al., 2022; Gaston et al., 2020; Perera et al., 2022), have found that indoor air typically contains larger quantities of MPs. This is of particular concern for human exposure, as the sampled air directly correlates with the air that individuals breathe. Recently, Goβmann et al. (2022) introduced spider webs as an innovative indicator of MP pollution in urban air. Their research revealed that every spider web sampled contained MPs, constituting up to 10% of the total weight of the webs. The extent of MPs caught by the individual spider webs ranged from 11,400 μg MP g^{-1} to 108,000 μg MP g^{-1}. In prior studies, spider webs have served as tools for monitoring air quality, particularly in the detection of contaminants such as heavy metals (Rybak et al., 2015). The advantages of using spider webs lie in their ease of sampling and their widespread presence across various global regions. Importantly, data derived from spider webs have proven to be on par with other established biomonitors and air sampling techniques (Rutkowski et al., 2020).

The aim of the present study was to assess the dynamics of MPs in road dust from urban settings, using car parks as the sampled sites. For

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this purpose, a custom-made novel sampling device was specifically designed to collect synthetic particles and was utilized to gather road dust from the surface of the parking lots. Additionally, spider webs were gathered and analysed to estimate the presence of airborne MPs. This integrated approach, employed for the first time in this study, allows for a comprehensive analysis of MPs (excluding car tyre rubber), evaluating aspects such as size, polymer composition, counts, and mass, employing FPA-μFTIR-imaging.

2. Materials and methods

2.1. Sampling location

Road dust samples were collected from seven distinct parking areas (Table 1, Fig. 1). Car parks were selected for sampling due to their controlled environment. They predominantly service standard-sized cars and offer ease of sampling due to the reduced traffic and consistently low speeds. The areas were within Uppsala City, Sweden, situated 71 km to the north of Stockholm and home to a population of 177,074 inhabitants. The selected parking areas included a variety of both indoor and outdoor spaces spread across different urban catchments. These represented a range of urban functionalities: residential, commercial, city centre, industrial, and recreational zones as seen in Table 1.

1. Recreational Area Parking (REC): Located near an artificial grass field within a recreational area that includes green spaces, a gymnasium, and larger gym facilities, this parking lot serves a community surrounded by residential areas.
2. City Centre Hotel Parking (Hotel): Near a hotel in the city centre, this lot is strategically placed close to restaurants, the Uppsala train station, the concert hall, and high-rise residential buildings, reflecting its central urban location.
3. Gränbystaden Shopping Mall (Indoor and Outdoor Parking, No. 3, COM and COM indoor): As one of Sweden's largest malls with over 10 million annual visitors, Gränbystaden provides extensive parking spaces (more than 3000) to accommodate its large customer base.
4. Libroback Industrial Region Parking (Industrial): Located on the city's outskirts along the Fyris River, this parking lot serves an industrial area with small industries and businesses, including building supply stores and auto repair shops.
5. Retailer Parking in Boländerna Industrial Precinct (Retailer): Adjacent to a large home furnishings retailer, this parking lot offers substantial space and is located near heavily frequented roads, indicating its high commercial activity.
6. Residential Parking in Kvarngärdet (RES): This outdoor parking space in a centralized residential area is characterized by higher-density living spaces, including apartment houses of various sizes.

2.2. Sampling

2.2.1. Road dust collection

The samples were collected after two days with less than 1 mm of precipitation during dry conditions on the parking lots. The sampling spanned from May 21, 2022, to June 21, 2022, where a total of 56 mm of rain fell in Uppsala City (SMHI, station number 97510). Dust samples were collected within 1–2 days post-rain events. These prior rain events exhibited cumulative precipitation ranging between 9 and 12 mm over a three-day period and had a maximum daily precipitation of 6.6 and 5.7 mm (see also SI1 Fig. S1).

Road dust was collected using a custom-designed stainless steel sampling device named 'Dusty.', which was constructed with a removable aluminium bucket securely fastened to the frame of a sack truck, providing mobility and ease of use during sampling. The bucket was designed to be easily detached and replaced for a fast and efficient collection of samples, and affixed to the bucket was a specialised lid engineered to accommodate a 10 μm steel filter, Ø 167 mm (www.filtertek.dk). This filter, which was secured by a metal lid, was crucial in capturing the fine particles while larger debris was deposited into the bucket. Extending from this lid was a rigid pipe connected to a floor nozzle designed for practical surface sampling. An industrial-grade vacuum cleaner was connected to the opposite side of the lid, providing the suction necessary to lift and collect road dust samples through the nozzle. All components of the 'Dusty' sampling device were fabricated from metal to ensure durability and minimise contamination risk. A silicone gasket was used to achieve a secure seal between the bucket and the lid, preventing sample loss and external contamination. The sack truck's wheels were initially plastic but were replaced with wood to mitigate potential contamination sources during sampling. An illustration of the device can be seen in Fig. 2.

Before the sampling, the particles resting on the tarmac were manually displaced using a steel bristle broom. This preparatory step ensured that particles adhering to the surface were loosened, making them accessible for collection. This step was deemed unnecessary at the indoor parking lot because the floor was smooth and glossy. Additionally, samples from the floor, walls, and other plastic materials were gathered from the indoor parking lot to assess any potential sources of contamination.

The sampling strategy was designed to encompass three distinct zones within each parking lot: the curb, middle, and drive-in (inlet) areas (Fig. 1). For each of these zones, three one-square-meter sections were delineated and sampled. The collected samples from these three separate squares were then pooled into a single bucket. Subsequently, the filter and the fine particles it retained were added to the same bucket, creating a composite sample for each transect within each parking lot – three samples were collected for each parking lot.

2.2.2. Spider webs

In addition to road dust, spider webs were systematically collected from locations corresponding to the road dust sampling sites. Wooden

Table 1

Geographic and descriptive data for the studied urban car parks. The table presents the type, ID, coordinates, area of sampling, total parking area and the number corresponding to the map of Fig. 1.

Type	ID	WGS84 decimal (lat, lon)	Type of parking	Area of sampling (m^2)	Total area of parking lot (m^2)	Corresponding map number
Artificial grass field/recreation area	REC	59.846968, 17.607948	Outdoor	9	1027	1
City central	Hotel	59.860563, 17.646441	Outdoor	9	702	2
Commercial	COM	59.877352, 17.676267	Outdoor	9	5275	3.a
Commercial	COM Indoor	59.877352, 17.676267	Indoor	9	–	3.b
Industrial	Industrial	59.881761, 17.588446	Outdoor	9	2331	4
Industrial/Commercial	Retailer	59.847863, 17.693326	Outdoor	9	6892	5
Residential	RES	59.868779, 17.647817	Outdoor	9	545	6

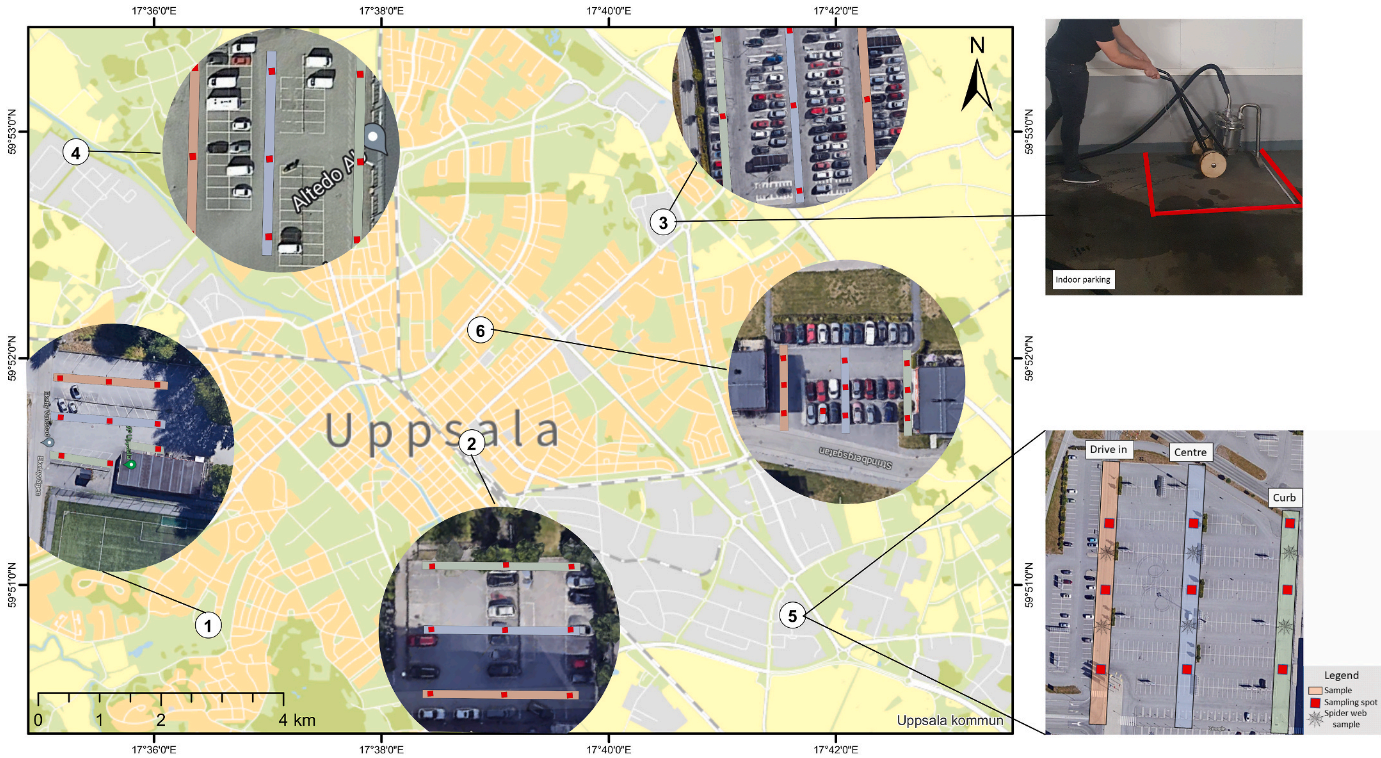

Fig. 1. Map of the selected car parks for the sampling of road dust and spider webs.

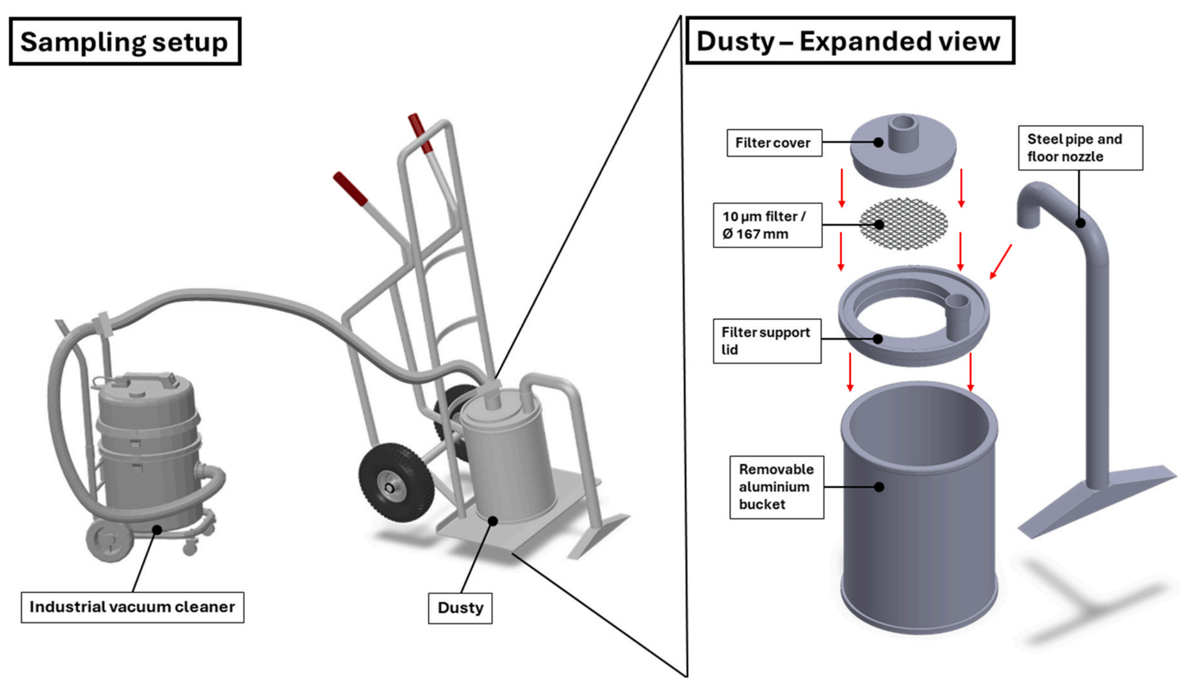

Fig. 2. Graphical representation of the road dust sampler 'Dusty'. The assembled setup is depicted on the left, while the expanded components of 'Dusty' are visualised on the right. The red arrows indicate how the components would be assembled. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

sticks were utilized to collect the webs to avoid contact contamination. All visible spider webs within the defined transects – curb, middle, and drive-in (inlet) – were sampled. Outdoor spider webs were collected from, among others, post signs and light poles at heights of one to 2 m. Indoor webs were obtained from the ceilings of underground parking lots, approximately 3 m high. The collected amounts of webs can be seen in Appendix S1. Following collection, the web samples were carefully placed into 20 mL headspace glass vials (sealed with a Teflon-coated septum in the inner part with a synthetic rubber lid) for transport and subsequent analysis.

2.3. Sample preparation

2.3.1. Road dust

Upon receipt in the laboratory, the road dust samples were processed according to the following MP extraction protocol, slightly modified from Rasmussen et al. (2024): Coarser particles settled in the collection bucket were initially sieved via a 500 μm mesh, removing macro debris. The larger particles (>500 μm) were stored for later inspection. The residual <500 μm fraction was transferred to 100 mL of sodium polytungstate (TC-Tungsten Compounds) (SPT) at a 1.9 g cm^{-1} density (Klöckner et al., 2020). Concurrently, filters containing the finer particles were submerged in SPT within a crystallising dish and subjected to sonification to dislodge adhered particles. The resultant SPT solution, now laden with these particles, was merged with the previously sieved fraction. Between 3 and 15 g of road dust per sample (particles <500) μm were analysed (SI 1, Table S2).

The integrated samples were transferred to a separatory funnel, agitated with compressed air, and subsequently allowed to settle. After settling overnight, the settled matter was removed. This settling and removal cycle was repeated over several consecutive days. The SPT was then separated via a 10 μm steel filter, retaining the sample. This sample was immersed in a 5% w/w sodium dodecyl sulphate (Sigma Aldrich, purity >99%) (SDS) solution, incubated at 55 °C for 24 h, and subsequently filtered. The samples were then combined with 200 mL of 0.7 μm pre-filtered demineralised water and subjected to catalysed oxidation using Fenton's reagent (Tagg et al., 2016), incorporating 145 mL of 50% H_2O_2 (TH. Geyer), 65 mL of 0.1M NaOH, and 62 mL of 0.1M $FeSO_4$. After 24 h, the samples underwent another filtration and were reintroduced to the SPT, undergoing a density separation analogous to the initial step. The final concentrated sample was placed into 5 mL of HPLC-grade 50% ethanol (TH. Geyer, purity >99.9%) and transferred into a 10 mL headspace vial, from which ethanol was evaporated at 50 °C under a gentle flow of N_2 in a TurboVap® LV bath. The sample volume was standardised by supplementing with 5 mL of HPLC-grade 50% ethanol.

2.3.2. Spider webs

Spider webs were carefully detached from the wooden sticks and positioned in small aluminium cups. Their precise weights were determined using an Ultra-Micro balance (Cubis® II Micro and Ultra-Micro Configurable Lab Balance). Between 0.15 and 26 mg of webs were analysed (SI 1, Table S1). Subsequently, the weighed samples were transferred to a 150 mL beaker containing 50 mL of H_2O. A 50% hydrogen peroxide (H_2O_2) solution was added in 5 mL increments and monitored until no effervescent reaction was indicated by foaming. This methodology is similar to that employed in Goβmann et al. (2022), albeit with Fenton's reaction as the alternative oxidative process. Following this oxidative treatment, the samples were concentrated in 50% ethanol using the same approach as the road dust samples.

2.4. 2.4contamination prevention and assessment

Rigorous precautions were taken to avoid contamination throughout the sample handling process. These included: (1) Rinsing all glassware, utensils, and tools interacting with the samples thrice using demineralised water pre-filtered through a 0.7 μm glass fibre filter. (2) Ensuring all sample handling personnel wore cotton-only lab coats and t-shirts. Moreover, (3) the samples were covered with aluminium foil to shield them from external contaminants. Prior to use, all reagents were passed through 0.7 μm glass fibre filters to ensure contaminant-free processing.

2.4.1. Blank samples

Despite these meticulous precautions, the potential for contamination from airborne MPs remains. Hence, a Petri dish (Ø 167) was exposed during each sampling event by placing it adjacent to the sampling points, simulating the exposure the sample containers underwent when opened. This allowed for the assessment of ambient contamination levels. Petri dishes from both an outdoor (no. 6 in Table 1 and Fig. 1) and an indoor parking lot (no. 3 in Table 1 and Fig. 1) were subjected to the same sample preparation procedure as the road dust samples and subsequently analysed to assess the extent of potential contamination.

2.5. μFTIR analysis

For chemical characterisation, a portion of the particle concentrate was placed on a circular zinc selenide transmission window measuring 13 × 2 mm. This window was set within a compression cell (Pike Technologies), effectively reducing its active viewing area to Ø10 mm. Each sample was deposited on a zinc selenide window in 100 μl increments using a glass capillary micropipette and then left to dry at 50 °C on a heating plate. This step was repeated until the window displayed an optimal distribution of particles, ensuring minimal particle aggregation.

FPA-μFTIR determined the chemical identity of the particles. The equipment comprised a Cary 620 FTIR microscope paired with a Cary 670 IR spectroscope (Agilent Technologies, USA). The complete functional area of the zinc selenide window (Ø 10 mm, equating to an area of 78.5 mm^2) underwent scanning with a 15x magnification Cassegrain objective, coupled with a mercury cadmium telluride (MCT) detector equipped with a 128 × 128 FPA producing a pixel resolution of 5.5 μm. Executed in transmission mode, these scans spanned a spectral range of 3750–850 cm^{-1} at 8 cm^{-1} resolution by accumulating data from 30 scans per pixel. Prior to every sample analysis, a background scan was taken, aggregating data from 120 individual scans.

2.6. Data handling and statistics

After conducting an in-depth visual assessment of the particles above 500 μm collected on the 500 μm mesh, it became apparent that synthetic particles of this size were only found infrequently and were not consistently present across all samples. Due to their intermittent presence, these particles were not considered in the subsequent data evaluation and processing phases. For particles sized between 10 and 500 μm, the software siMPle (previously known as MPhunter) (Primpke et al., 2020) was used to analyse the derived infrared imagery. Employing such an automated approach diminishes the potential for human-induced biases typically associated with the manual interpretation of spectral data. siMPle identifies and quantifies MPs in each sample, yields the polymer makeup of the particles and their dimensions and estimates the volume and mass of each particle. The major dimension was calculated as the longest distance between the pixels of the particle, while the minor dimension was the longest axis perpendicular to the major dimension. The thickness was assumed as 60% of the minor dimension (Simon et al., 2018; Liu et al., 2019b). The mass of each particle was estimated from the volume of the particle assuming an ellipsoid shape and it's density. Based on these two dimensions, fibres were classified based on having a ratio >3 between the major and minor dimensions (Vianello et al., 2019).

All statistical analysis was conducted in R (version 4.2.2). A Shapiro-Wilk test was used on the major dimension to assess the normality of the dataset. A non-parametric Kruskal-Wallis test was applied to check for differences between the road dust and spider webs. Furthermore, a pairwise Wilcoxon test was used to assess differences between the samples. The significance level (p-value) was set to 0.05 for all the statistical tests.

3. Results and discussion

3.1. Contamination

Analysis of the blank samples indicated low contamination, with three MPs identified at the outdoor retailer parking lot (no. 6 in Table 1 and Fig. 1) and two particles found at the COM Indoor parking facility (4). The average count of MP in all samples was 101.5, which indicates that contamination on average accounted for 2 and 3% of the particles detected in the blank samples from the two sites, respectively. Blank correction was deemed inappropriate for this study due to the low presence of particles in the blank samples.

3.2. MP concentrations

FPA-μFTIR successfully identified MPs in all samples. A total of 4264 MP counts were identified and used for calculating concentrations. The concentration in road dust exhibited considerable variation, with counts ranging from 5.78 to 4951 counts g^{-1} and a median value of 156 counts g^{-1} (Fig. 3). When quantified by mass, the concentration spanned between 0.06 and 95.3 μg g^{-1} with a median of 13.7 μg g^{-1}. These findings align with those of Rasmussen et al. (2023), where road dust was collected from the surface of permeable pavements and analysed using the same methods as in the current study. In their study, concentrations ranged between 236 and 1075 counts g^{-1} and 14.8–122.3 μg g^{-1}.

Zhang et al. (2022) reported concentrations ranging from 35.4 to 1329.2 counts g^{-1} in fugitive road dust collected from two urban mining bases. Their analysis involved visual sorting followed by FTIR characterisation of the isolated particles. These findings appear, at first examination, to be consistent with the results presented in our study. However, when comparing the values, one has to keep in mind that the two studies were conducted applying quite different methodologies, and results hence may or may not be comparable.

Morioka et al. (2023) applied Pyr-GSMS to quantify the mass of MP in road dust and identified concentrations of 2429 μg g^{-1} within the size fraction of 1.1–500 μm. Adopting a similar methodology, O'Brien et al., 2021 found concentrations that varied from 500 μg g^{-1} in rural regions to 6000 μg g^{-1} in urban areas. These findings indicate significantly larger amounts of MPs in road dust compared to the mass estimations in the current study. Nonetheless, the variability in analytical techniques, targeted size ranges, and differences in sampling methodologies makes comparison a difficult challenge.

This being said, in stormwater pond sediments, using an almost identical methodology as in the current study, Liu et al. (2019a) found an MP concentration of 17.5 counts g^{-1}, corresponding to 3 μg g^{-1}, while Molazadeh et al. (2023a,b found on average 44.4 counts g^{-1}, corresponding to 11.8 μg g^{-1}. These results from stormwater pond sediments align with those from road dust. While one might anticipate higher concentrations in stormwater pond sediments due to the continuous runoff they receive from roads, the low concentrations suggests otherwise. It seems only a fraction of the MPs from roads are mobilised and carried with the runoff water, the rest of the MPs are taking alternative routes.

The highest concentration of MP, by both mass and counts, was detected at the COM indoor parking lot (4). The indoor parking exhibited a median concentration that was notably elevated compared to the outdoor sites, being 31 times higher in MP counts and 9.4 times higher in MP mass compared to the median value found in road dust collected outdoors. The notable accumulation of MPs in an indoor parking lot, significantly exceeding that in outdoor areas. This is likely due to reduced dispersion as a result of limited air movement. With restricted air movement, MPs have fewer opportunities to disperse, effectively trapping them within the enclosed space. Consequently, these particles accumulate over time due to the lack of alternative pathways for dispersion.

No discernible trend existed between location and MP counts or between the different sampling positions. However, in terms of mass, the median value for all samples collected at the curb was 26.24 μg g^{-1}, exceeding the medians of the midway and entry samples of 16.46 and 6.36 μg g^{-1}, respectively. Interestingly, this pattern aligns with the findings of Järlskog et al. (2022), who similarly reported elevated concentrations of traffic-derived particles along curbs. It should be emphasised that the concentration of MPs at the curb is likely underestimated in this study, as the samples from this location exhibited a notably higher debris content. This increased presence of debris could potentially have masked the MPs during the μFTIR analysis.

In the spider webs (Fig. 4), MP counts ranged from 2500 to 505,000 counts g^{-1}, with a median value of 20,830 counts g^{-1}. A similar trend

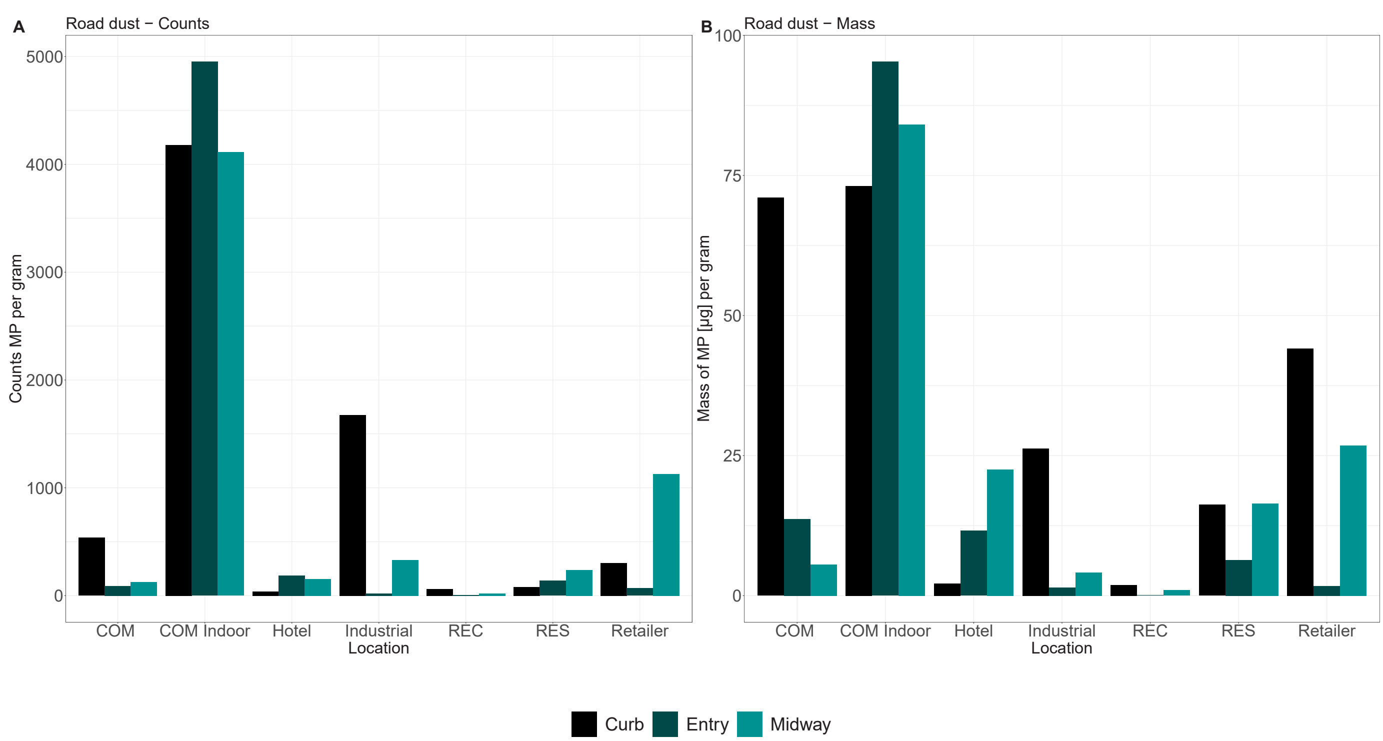

Fig. 3. Concentration of MP in the road dust at the investigated parking lots. The data has been adjusted to dispay the concentration by counts MP per gram (A) and by micrograms MP per gram (B).

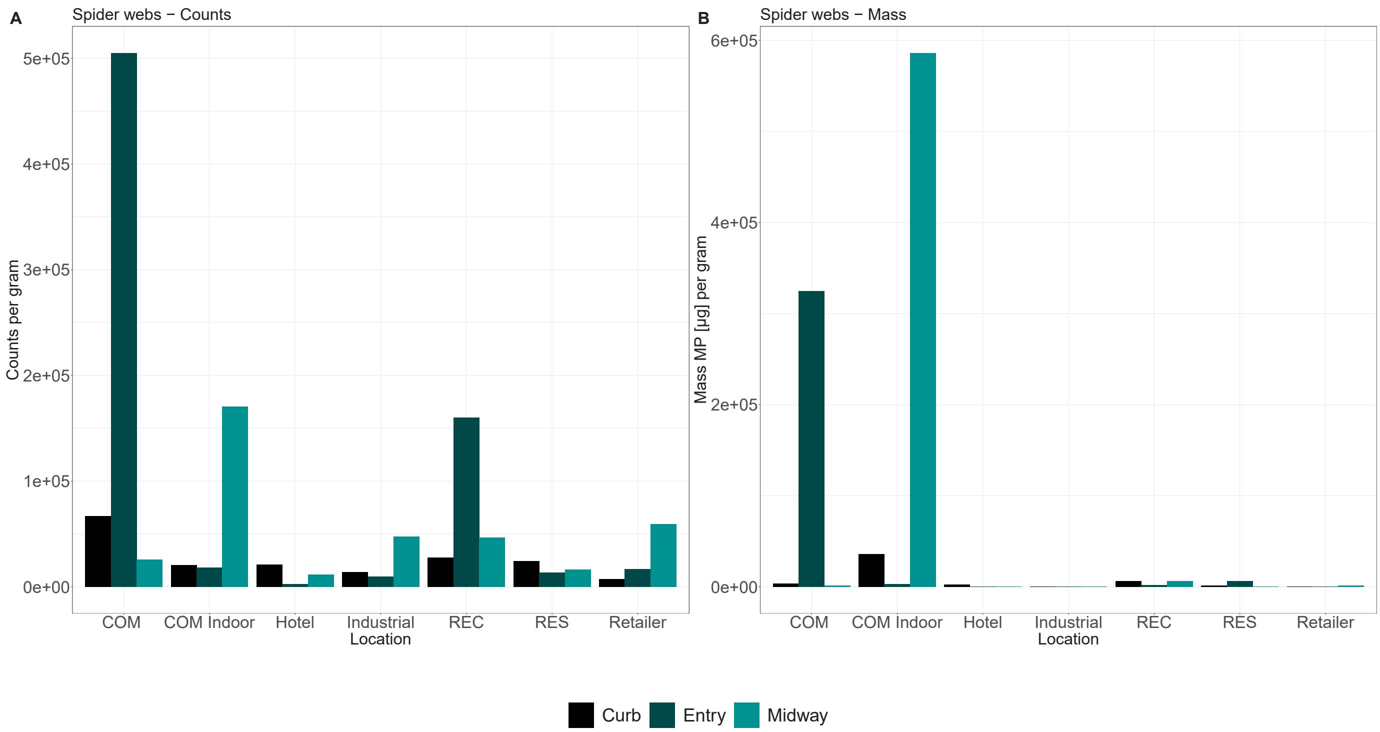

Fig. 4. Concentration of MP in the spider webs at the investigated parking lots. The data has been adjusted to dispay the concentration by counts MP per gram (A) and by micrograms MP per gram (B).

was seen in the mass estimation, where values ranged from 40 to 586,000 μg g^{-1}, and the median was 1360 μg g^{-1}. The COM shopping centre (3) had the highest concentration of MP counts and the second highest MP mass. Conversely, the highest concentration in terms of mass was identified in the indoor parking lot, aligning with the patterns from the road dust. The patterns observed in our research align with findings from earlier studies. Notably, Choi et al. (2022), Gaston et al. (2020), and Perera et al. (2022) all reported markedly higher concentrations of MP in indoor air compared to outdoor air.

The pioneering study by Goβmann et al. (2022) employed spider webs as a biomonitor for MPs and reported concentrations between 11, 400 μg^{-1} g and 108,000 μg^{-1} g using Py-GC-MS. These values align closely with the range observed in our study, further underscoring the viability of spider webs as an effective tool for monitoring MP contamination in outdoor air.

The variability in MP concentrations between spider webs was extensive, and while a significant portion of this variation can be attributed to the location and nearby sources, the duration over which the spider webs accumulated MPs also plays a crucial role. The longevity of spider webs is subject to external conditions such as wind, temperature, and precipitation, each contributing to the varied lifespan of the webs and, consequently, the accumulation of MPs. Webs situated indoors probably have longer lifespans than those exposed to open air, providing a plausible explanation for the observed variation in MP concentrations. In outdoor settings, it is plausible to infer that the MPs might be carried by wind from distant locations, while in the indoor parking, particles are directly associated with the immediate surroundings and become airborne due to vehicular movements during entries and exits.

Furthermore, the discrepancies in MP concentrations between road dust and spider webs could stem not only from these environmental and locational factors but may also be associated with the inherent characteristics of the MPs themselves, including their size, shape, and polymer types. These inherent properties could significantly affect the behaviour of MPs, their entrapment efficiency by the spider webs, and their deposition on the asphalt, adding another layer of complexity to the observed variations in MP concentrations across different environments.

3.3. Polymer composition

The polymer composition by counts of the entire dataset was as follows: polyurethane (48.5%), polypropylene (16.8 %), polyester (12.59 %), acrylic (7.8 %), PE (6.3 %), PS (4 %), PA (2.6%) and cellulose acetate (0.6 %). The four least abundant polymers – polyvinyl chloride, acrylonitrile butadiene styrene, pan acrylic fibre and polyvinyl acetate – were grouped as Others. The polymer composition is shown in Fig. 5.

The most pronounced distinctions between samples (Fig. 5 A and B) are those from COM indoor and Industrial (4, 5). In the COM indoor samples (4), polyurethane was the predominant polymer, constituting over 70% of the total composition by count. However, when evaluating the distribution of polymer mass, polyester emerged as the dominant component, comprising nearly the entirety of the composition. At Industrial (5), acrylic was the most abundant polymer, both by counts and mass. This prevalence of acrylic is potentially indicative of the surrounding industrial land use at this parking lot.

PP was predominant in road dust from outdoor parking lots. Rasmussen et al. (2023) also identified PP as the leading polymer in road dust when excluding car tire rubber, a finding echoed by both Molazadeh et al. (2023a,b and Liu et al. (2019a). The consistently high prevalence of PP in road dust and related environments like stormwater pond sediments poses questions about its source, a topic warranting further investigation.

A PCA analysis, which classified road dust and spider webs based on their sample type, revealed distinct chemical signatures for both groups in terms of MP counts and mass (see also SI2 Figs. S2 and S3). The variance between the two sample types can be attributed to the predominance of PP and polystyrene PS in road dust, whereas spider webs showed a higher occurrence of polyester and PE. Additionally, a separate cluster emerged for road dust samples collected from indoor parking, distinguished by a significant presence of PU. This significant quantity of PU particles prompted an investigation into their origin. Subsequent sampling involved collecting potential sources of these

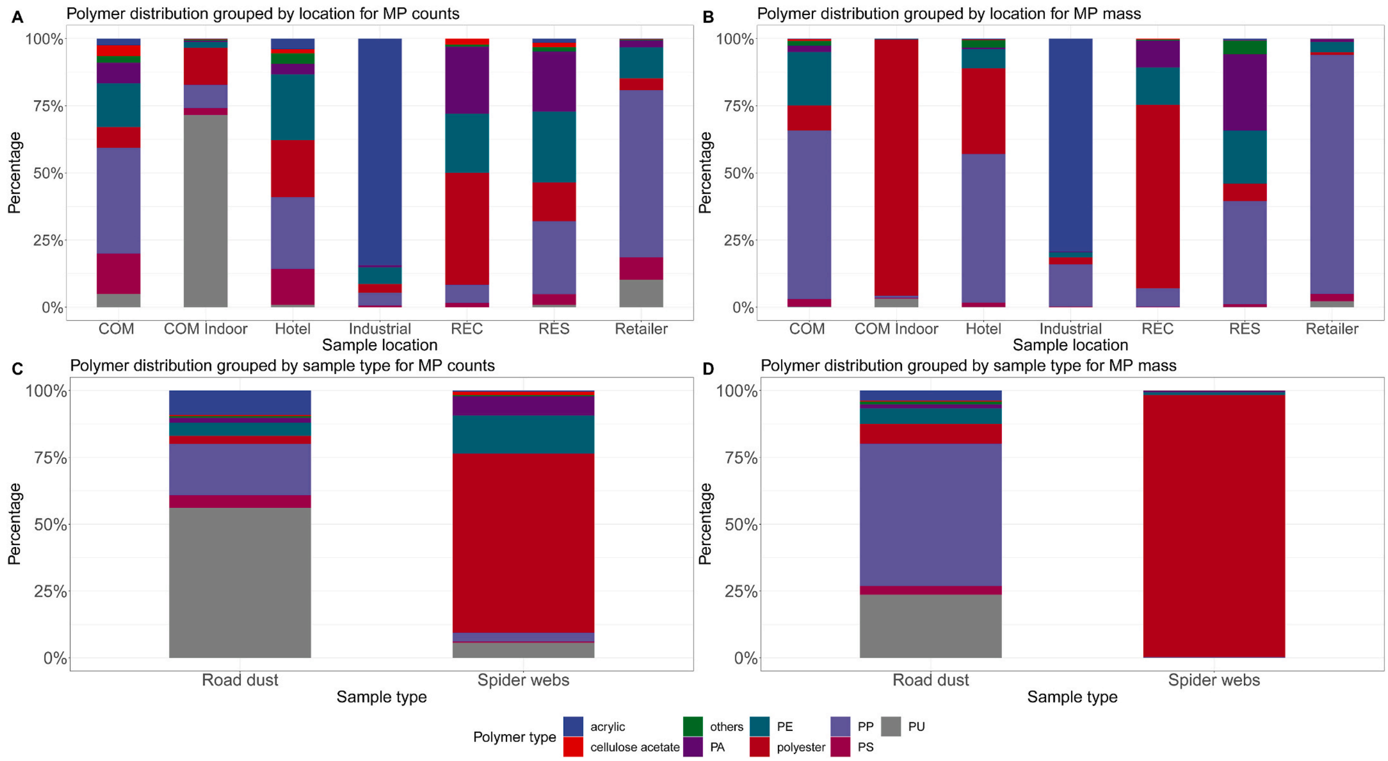

Fig. 5. Polymer distribution in the entire dataset by counts (A, C) and mass (B, D) of MP. The dataset was grouped by sample location (A, B) and sample type (C, D).

particles for analysis. Samples from the floor, walls, and shopping cart wheels were analysed using μFTIR. The findings revealed the presence of polyurethane in the parking lot floor, suggesting it might be a component of the applied coating (see also SI2, Fig. S4).

Polyester was the prevalent polymer detected by counts and mass in spider webs. Even in the indoor parking lot – where a major PU contamination occurred – polyester was still the polymer most likely to get trapped in the spider webs. Polyester is often predominantly found in

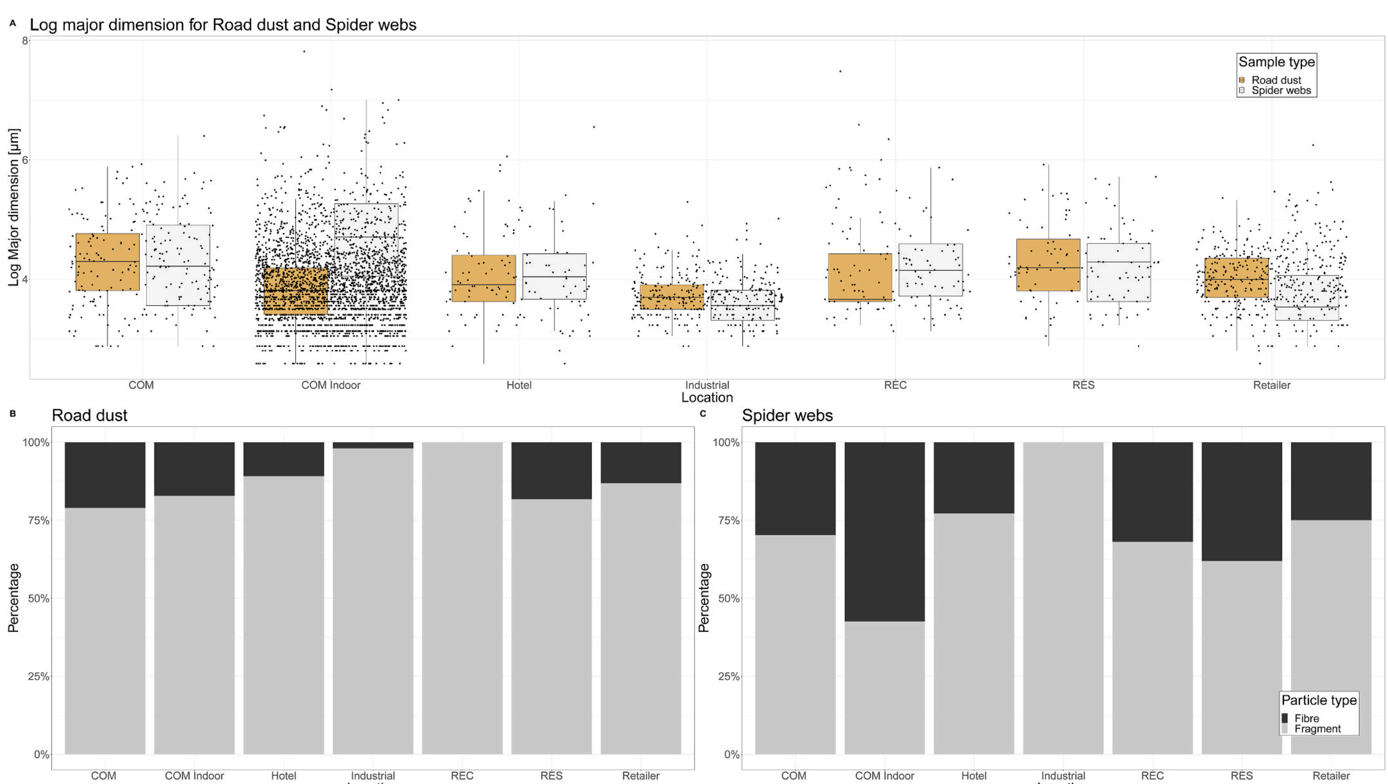

Fig. 6. Boxplot and whiskers plot (A) of the major dimension on a logarithmic scale of the particles in each parking lot for the road dust and spider webs. Plots B and C illustrate the fibre-to-fragment ratio at each location for MPs in road dust and spider webs, respectively.

the environment as fibres (Browne et al., 2011) and is one of the most frequently produced fibre types (European Environment Agency, 2021). Even though polyester has a higher density (1.38 g cm^{-1}) – indicating a propensity to remain grounded – it might still become airborne and entangled in webs due to its fibrous nature (Browne et al., 2011).

3.4. Size distribution

The Shapiro-Wilks test revealed that the size of particles from road dust and spider webs was not normally distributed. Significant differences ($p < 0.05$) were revealed by the Kruskal-Wallis test between the two matrices (see also Table S2).

While the particle sizes from road dust and spider webs were comparable in most parking lots, distinct differences were evident (Fig. 6, A). Specifically, at the COM indoor parking lot (4), the major dimension of the particles captured in the spider webs surpassed those found in road dust. Conversely, particles retrieved from spider webs were smaller than from road dust at the retailer parking lot (6).

Employing a ratio between major and minor dimension >3 as thresholds for fibres (Vianello et al., 2019) yielded that all spider webs contained more fibres than road dust. Notably, the highest concentration of fibres was identified in COM indoor (4), comprising over half of the total MPs detected.

In the open environments, particle size had minimal influence on whether particles became airborne or stayed grounded. However, in enclosed spaces like the COM indoor parking lot (4), where environmental elements are controlled or absent, the particles trapped in webs were noticeably larger – mostly fibres – compared to those that settle on the floor, as expected. Conversely, in outdoor settings, weather conditions are likely to be the determining factor influencing the airborne status of particles.

3.5. Perspectives

This study employed a custom-made device to collect road dust from 9 m^2 per parking lot. Given the substantial average size of the parking lots – approximately 2800 m^2 – it is crucial to consider the potential implications for data interpretation. The intricate and time-intensive nature of extracting MPs from road dust implies that only a small fraction of the total sample was analysed using μFTIR (Molazadeh et al. (2023a,b). This selective analysis may introduce uncertainties, necessitating cautious interpretation of the resultant data (Kooi et al., 2021).

It is crucial to highlight that this study adopted a meticulous approach to sample collection, involving the intentional dislodgement of particles from the asphalt using a metal brush. This step was paramount to include particles that would generally resist natural relocation, ensuring a more encompassing representation of particle distribution within the sampled areas. Incorporating loosely attached and firmly adhered particles on the asphalt surface provides a more accurate reflection of the diversity and concentration of particles present. However, it is essential to consider that this approach may lead to a potential overestimation of the concentration of MP that might typically enter the environment under natural conditions due to the inclusion of particles that would otherwise remain stationary, thereby implying a potentially higher assessment of environmental exposure to MPs. However, at the curb, particles accumulated were seldom firmly attached to the asphalt, making them easily collectable using 'Dusty'.

The substantial quantities of MPs discovered in both spider webs and road dust within the indoor parking lot illustrate the potential inhalation risks to humans. However, it is crucial to consider that individuals typically only traverse through such indoor parking lots for a few minutes, thereby substantially limiting exposure time. Additionally, according to the shopping centre administration, the indoor parking floor is cleaned every three weeks, while it is probable that the spider webs have never been cleaned, potentially impacting the observed concentrations of MPs.

Using a back-of-the-envelope calculation with the average MP values obtained from outdoor parking areas — 19.5 μg m^2 or 310 counts m^2 — and applying these to the city of Uppsala, where around 32% of the 47 km^2 area is paved, the daily accumulation of MPs can be estimated. Assuming a runoff coefficient of 100% (Butler et al., 2018), this would correspond to an average runoff from the paved surfaces of approximately 293 g d^{-1} of microplastics into the environment, translating to around 4.7 billion particles every day there is significant rainfall. This estimate underscores the considerable impact that urban areas may have on microplastic pollution of receiving waters via surface runoff.

Analysing road dust is inherently complex, requiring specialised equipment and substantial time investment. In contrast, spider webs, due to their ubiquity and the simplicity associated with their collection and preparation, emerge as an efficient tool for monitoring urban airborne MPs (Goβmann et al., 2022). Their practicality and accessibility offer a stark and advantageous contrast to the complexities involved in road dust analysis, emphasising their potential utility in tracking urban MP contamination.

4. Conclusion

This study employed μFTIR imaging to analyse MPs in road dust and spider webs in a novel integrative manner, unveiling the significant and variable concentrations of MPs present in urban environments. A new, custom-made device was built for efficiently collecting road dust, revealing the highest concentrations in an indoor, enclosed parking area. The analysis of spider webs revealed a trend consistent with that observed in road dust, supporting the identification of an MP pollution hotspot in the underground parking lot across both matrices. While polyurethane emerged as the predominant polymer in road dust, polyester was found to be the most prevalent in spider webs. Moreover, the fibre percentage was three times higher in spider webs than in road dust.

Substantial rainfall events can displace nearly five billion microplastics daily, channelling them from urban pavements directly into the environment. This significant transport mechanism underscores the profound impact urban areas might have on the nearby water bodies, highlighting the need for effective management and source control to safeguard our waterways from the relentless influx of MPs.

While this investigation offers a glimpse into the dynamics of MP presence in urban areas, it underscores the need for more comprehensive studies to understand the movement and impact of MPs in city landscapes. MPs are prevalent in urban environments and are subtly and continuously migrating.

CRediT authorship contribution statement

Lucian Iordachescu: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Resources, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Gabriella Rullander:** Writing – review & editing, Visualization, Resources, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jeanette Lykkemark:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Sahar Dalahmeh:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **Jes Vollertsen:** Writing – review & editing, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the authors used CHATGPT-4 in order to improve the grammar and readability of the manuscript. After using this tool, the authors reviewed and edited the content as needed and take full responsibility of the content of the publication.

Declaration of competing interest

The authors declare that they have no financial interest that could have influenced 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.jenvman.2024.121064.

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