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Traffic-related microplastic particles, metals, and organic pollutants in an urban area under reconstruction

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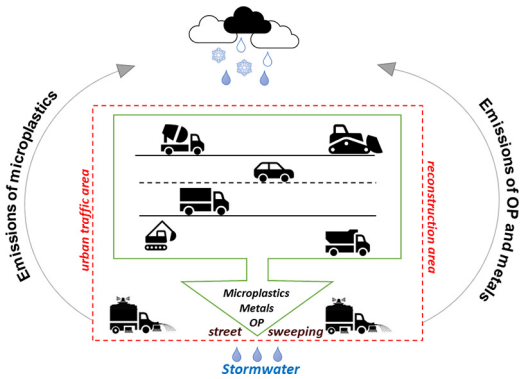
HIGHLIGHTS

- MP, OP, and metals were analyzed simultaneously in stormwater and road material.
- MP were analyzed with stereo microscopy, melt-tests and tactile identification.
- High amounts of TWP, OP, and metals were emitted in the reconstruction area.
- New OP was identified with a non-targeted screening deconvolution GC/MS method.
- Street sweeping is a promising measure to reduce transport of MP, OP and metals.

GRAPHICAL ABSTRACT



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ABSTRACT

In urban environments, particularly areas under reconstruction, metals, organic pollutants (OP), and microplastics (MP), are released in large amounts due to heavy traffic. Road runoff, a major transport route for urban pollutants, contributes significantly to a deteriorated water quality in receiving waters. This study was conducted in Gothenburg, Sweden, and is unique because it simultaneously investigates the occurrence of OP, metals, and MP on roads and in stormwater from an urban area under reconstruction. Correlations between the various pollutants were also explored. The study was carried out by collecting washwater and sweepsand generated from street sweeping, road surface sampling, and flow-proportional stormwater sampling on several occasions. The liquid and solid samples were analyzed for metals, polycyclic aromatic hydrocarbons (PAH), oxy-PAH, aliphatics, aromatics, phthalates, and MP. The occurrence of OP was also analyzed with a non-target screening method of selected samples. Microplastics, i.e. plastic fragments/fibers, paint fragments, tire wear particles (TWP) and bitumen, were analyzed with a method based on density separation with sodium iodide and identification with a stereo microscope, melt-tests, and tactile identification. MP concentrations amounted to 1500 particles/L in stormwater, 51,000 particles/L in washwater, and 2.6×10^6 particles/kg dw in sweepsand. In

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stormwater, washwater and sweepsand, MP ≥20 μm were found to be dominated by TWP (38%, 83% and 78%, respectively). The results confirm traffic as an important source to MP, OP, and metal emissions. Concentrations exceeding water and sediment quality guidelines for metals (e.g. Cu and Zn), PAH, phthalates, and aliphatic hydrocarbons in the C_{16}–C_{35} fraction were found in most samples. The results show that the street sweeper collects large amounts of polluted materials and thereby prevents further spread of the pollutants to the receiving stormwater.

1. Introduction

In high-density traffic areas, microplastics (MP) (Knight et al., 2020), metals (Huber et al., 2016; Kabir et al., 2014), polycyclic aromatic hydrocarbons (PAH), and other organic pollutants (OP) (Markiewicz et al., 2017) are emitted in large amounts. Road runoff is a major transport route for urban pollutants and a significant contributor to a deteriorated water quality in receiving waters (Masoner et al., 2019; Müller et al., 2020). Only a small portion of the stormwater from urban areas is treated, and the treatment systems currently in use are not designed to effectively remove all types of pollutants.

Microplastics are generally defined as solid particles insoluble in water, within the size range 1–5000 μm, and composed of synthetic polymers with thermoplastic or thermoset properties. Due to their chemical and physical properties, tire wear particles (TWP) are often included in the microplastic definition (Andrady, 2011; GESAMP, 2016; Verschoor et al., 2016). Tires consist of styrene-butadiene rubber (SBR), natural rubber, carbon black, and additives (Sommer et al., 2018). Recent studies have indicated that TWP constitutes a major source of MP in the environment (Baensch-Baltruschat et al., 2020; Hann et al., 2018; Lassen et al., 2015; Sommer et al., 2018). In the Nordic countries, it is estimated that TWP contributes to more than half of the MP emissions to the environment (Magnusson et al., 2016; Sundt et al., 2014). TWP are generated during the interaction between the tire and the road surface, creating particles that are generally black and elongated, often with encrustations of pavement and minerals (Abbasi et al., 2017; Järlskog et al., 2020; Panko et al., 2012). TWP are mainly detected in the size range between 10 nm to several 100 μm (Kole et al., 2017). The size and shape depend on the wear process (e.g. vehicle speed and driving patterns, meteorological conditions, the structure of the road surface, and road maintenance), different tire compositions, as well as differences in sampling procedures, sample preparation, and different size limits for the analytical methods (Eisentraut et al., 2018). There are still knowledge gaps that need to be closed to develop risk assessments and transport models both regarding the occurrence, distribution, and toxicology of TWP, but also concerning the quantitative importance of TWP reaching rivers and oceans from road surfaces via stormwater. Most studies on metals in stormwater focus on copper (Cu), lead (Pb), and zinc (Zn) (Müller et al., 2020), because of their frequent occurrence at concentrations that may lead to acute and chronic effects on aquatic life, including survival, growth, and reproduction. Tire and brake wear have been identified as one of the most important non-exhaust pollution sources of Cu, Pb, and Zn. Zinc is commonly found in tires, about 1.5 w% (Degaffe and Turner, 2011; Kreider et al., 2010), and in metal parts of vehicles, e.g. brakes (Hjortenkrans et al., 2007; Ma et al., 2021). Zinc is also used in crash barriers, lampposts, and other road furniture made of galvanized steel (Folkeson, 2005). Copper is used in tires and brake-linings (Ma et al., 2021). Nickel, chromium, and lead can be found in brake wear, engine wear, metal plating, and leakage from the wheel balancing weights. Similar to the release of MP, the release of metals depends on driving behavior (i.e. speed, use of brakes, start-stop), traffic intensity, surface structure, and road maintenance (Müller et al., 2020).

In a literature screening of OP present in road-related sources, over 1100 specific pollutants were identified (Markiewicz et al., 2017). Based on compound fate and toxicity, the following OP were prioritized among the 1100 pollutants: polycyclic aromatic hydrocarbons (PAH) > alkanes C_{20}–C_{40} > alkylphenols > phthalates > aldehydes > phenolic antioxidants > bisphenol A > oxygenated PAH (oxy-PAH) > naphtha C_5–C_{12} > amides > amines. Markiewicz et al. (2017) also showed that tire wear is a major source of road-related emissions of particles and associated contaminants, particularly PAH, and that wear of the road surface and markings makes up a considerable proportion of these emissions. Further, Alves et al. (2020) identified a large number of different OP, e.g. aliphatic hydrocarbons, PAH, thiazoles, fragrant compounds, triterpenoids, phenolic compounds, and phthalate plasticizers, in PM_{10} collected in a tire wear simulator study. Concentrations of PAH in tires may vary from 10 to 230 μg/g, with pyrene being the most abundant compound (20–50% of total measured PAH), followed by benzo(*ghi*)perylene and coronene (Hwang et al., 2019). However, Kreider et al. (2010) found considerably higher PAH concentrations in road particles than in tire wear itself. In a recent road simulator study (test chamber, only road surface and tires, no exhaust emissions) a large numbers of different OP, e.g. aliphatic hydrocarbons, PAH, thiazoles, alcohols, fragrant compounds, sugars, triterpenoids, sterols, phenolic compounds, phthalate plasticizers and several types of acids, were identified in PM_{10} samples from tire wear (Alves et al., 2020).

Two previous studies investigated the potential of street sweeping as a measure to prevent or reduce OP, MP, and metal emissions to stormwater. The material collected by the sweeper showed high concentrations of aliphatics, phthalates, aromatics, PAH (Polukarova et al., 2020), and TWP (Järlskog et al., 2020). The sweeper collected large amounts of TWP, fine particles, and associated pollutants, leading to the conclusion that sweepsand and washwater from street sweeping should be treated before disposal. This agrees with a study of road dust collected in northern Vietnam, where more than 100 specific OP were quantified in high concentrations with a target GC/MS screening method (Anh et al., 2019b). The most predominant OP found were n-alkanes, PAH, and current-use chemicals such as phthalate plasticizers. Although several studies have investigated the occurrence of various OP and metals in road-related matrices, such as road dust and stormwater, more information is still needed on sources, occurrence and fate of MP in the environment (Dröge and Hulskotte, 2018; Dröge et al., 2019; GESAMP, 2016). For on-road emissions of TWP and other MP, as well as associated pollutants, runoff is a major vector for further transport to the aquatic environment (Baensch-Baltruschat et al., 2020).

Due to urbanization (EC, 2020), extensive infrastructure projects are underway in many cities, resulting in erosion and sediment release, which is then transported off-site with runoff or heavy vehicles frequenting the construction areas. In this study, we have studied an area under reconstruction in the central parts of Gothenburg, Sweden (population 600,000). The aim of the study was to: (1) investigate the occurrence of different categories of MP on road surfaces within an urban reconstruction area, and in stormwater from the same area; (2) estimate the amounts of MP, metals, and OP in road-related matrices; (3) determine any correlations between TWP, PAH, and metals in stormwater and street sweeping washwater collected in different

seasons; and (4) investigate the content of TWP, OP and metals in the material collected by the street sweeper.

The hypothesis is that pollutants such as MP, OP, and metals are generated from traffic within the reconstruction area and emitted in large amounts to the environment. Since the annual average daily traffic within the area is rather low, but the number of heavy vehicles is high, a high number of TWP is expected. However, large amounts of different plastics are used at construction sites, hence it is not obvious whether TWP is the largest category of MP. Another hypothesis is that street sweeping will effectively collect contaminated material from the roads, with MP, metals, and OP in high concentrations. The novelty of this study is that MP, OP, and metals were simultaneously studied in different environmental matrices. Also, the study focuses on TWP since most published studies on MP have not included analysis of these particles.

The results can be applied as input to environmental compartment (air, soil, water) transport models or risk assessments of traffic-derived particles. Results can also be compared with theoretical values to confirm/disprove previous assumptions on emissions. Further, the results can be used as a support for stakeholders or decision-makers when the impact on the environment should be evaluated.

2. Method

This is an experimental study based on real-life samples collected in a reconstruction case study area in the central parts of Gothenburg, Sweden, and includes a site for reconstruction of existing buildings and roads, a new tunnel, and a new bridge. Metals, MP, and OP (PAH, aromatics, aliphatics, phthalates, and screening) were analyzed in samples collected from 2018-07-05 to 2019-06-14. Samples were collected from stormwater, road surfaces, and from the material collected by a street sweeper (sweepsand and washwater). Fig. 1 illustrates the hypothesis, sampling matrices, the analytical methods, the main results from this study as well as some of the potential applications of the results.

2.1. Case study area

The study catchment, selected to represent an urban densification and reconstruction area, is situated in Gullbergsvass in the central parts of Gothenburg, Sweden (Fig. 2). The total catchment area is 9 ha and is dominated by impervious surfaces, comprising 4.7 ha roads and parking and 4 ha roofs of commercial buildings. The Göta River is classified as a *less sensitive recipient* (Environmental Quality Guidelines for water recipients) within the area of Gullbergsvass, meaning that only a simple stormwater treatment method is necessary prior to release into the river is necessary (e.g. catch basin filters or sand traps) according to local guidelines.

Under normal circumstances, the average annual daily traffic (AADT) in the catchment area is 5500 vehicles. Runoff within the catchment is drained via separate storm sewers and discharged untreated into the Göta River. During the sampling period, different construction works took place, including both renovation and transportation infrastructure projects, such as the construction of a new bridge. Each construction site has access routes to the surrounding road network (Fig. 2), at which an appreciable number of trucks enter and leave on a daily basis. The truck traffic through the study area varied considerably from week to week, but during the autumn approximately 500 trucks/week passed through the construction site. On a few occasions, one driving lane from E45, the major throughway, was redirected through Gullbergs strandgata: two weeks in September 2018 (nighttime 21-05 only) and two weekends in November 2018 (nighttime 21-05 only).

2.2. Street sweeping routines

For this study, street sweeping was performed on ten occasions in 2018. The sweeping campaign started 2018-07-05, and the final sweeping event took place 2018-12-13. (Fig. 3).

Sweeping was performed during daytime, and the streets were not closed for traffic which resulted in slight variations in the sweeping performance and distance covered (from 3.1 to 4.3 km). The sweeper's travel time and distance were recorded using a GPS-based smartphone app called Strava. The sweeper's mean driving speed varied between 4.7 and 10 km/h. Different driving speeds are expected to result in variations in the capacity to collect road dust; the lower the speed, the higher the capacity to collect road dust, and the other way around (Järlskog et al., 2017; Snilsberg et al., 2018). As the catchment area included parallel parking spaces along the road, the dust accumulated in these spaces was collected only on occasions when the spaces were not occupied. The sweeper used in this study was a vacuum sweeper with metal brushes (Johnston Beam VTJB651, Fig. S4). In dry weather conditions, the sweeper sprayed water onto the road to prevent fine dust from being emitted into the air, in line with regular sweeping routines. The amount of water sprayed on the streets during sweeping varied depending on weather conditions.

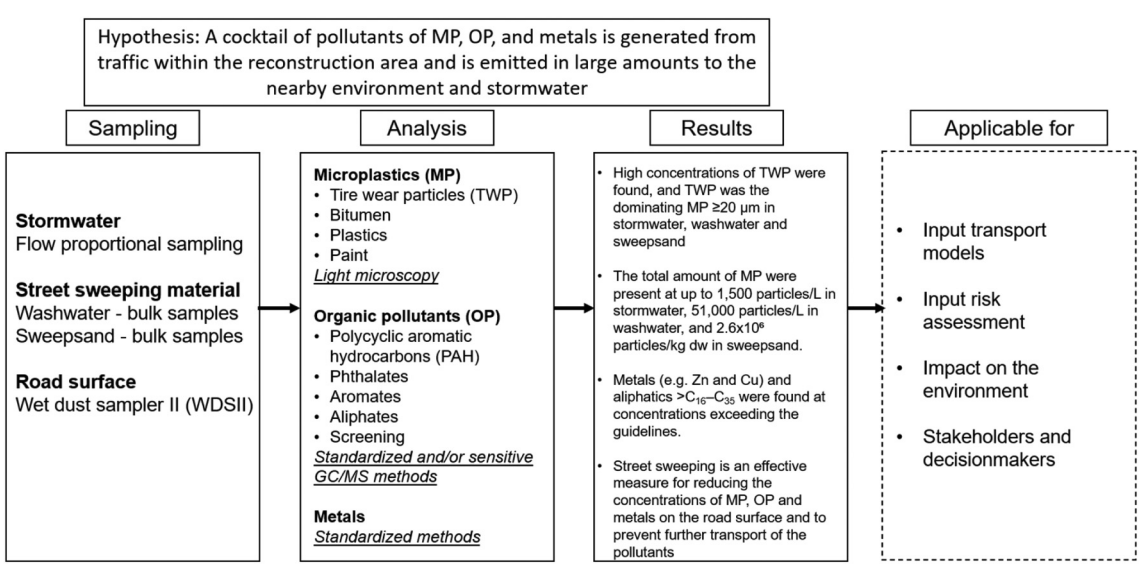

Fig. 1. Schematic illustration of the sampling matrices, analytical methods and main results from this study complemented with potential applications of the results.

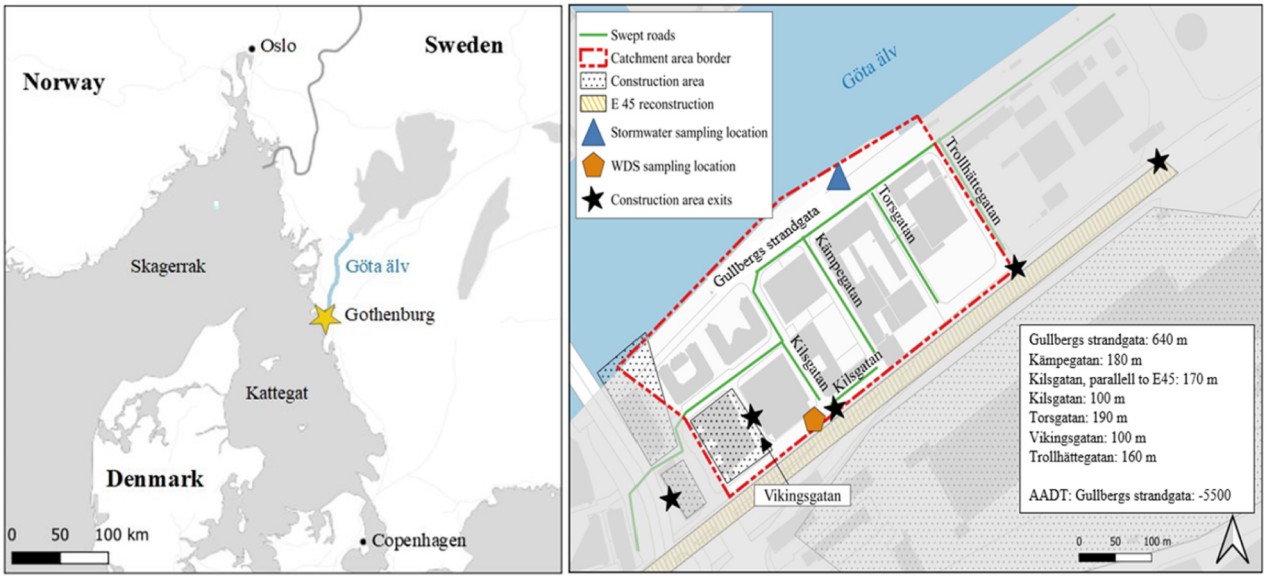

Fig. 2. Location of Gothenburg (left) and Gullbergsvass study area (right). The image on the right shows the construction areas, exits to the road network, catchment area, and the sampling locations. Precipitation was measured in parallel by a national monitoring station, located just outside the map to the east.

2.3. Sampling procedures

2.3.1. Stormwater sampling

Six runoff events (Fig. 3) were sampled during the period when street sweeping was carried out weekly, and three events were sampled during the period without weekly street sweeping (Table S2). The samples were analyzed for general water quality parameters, MP, metals, and OP. Samples for metals and general water quality parameters were collected in plastic bottles, and samples for OP and MP in glass bottles. The samples were stored cool and dark, and sent to the analyzing laboratories short after sampling. Metals were analyzed on both filtered (membrane filter, pore size 0.45 μm) and unfiltered samples.

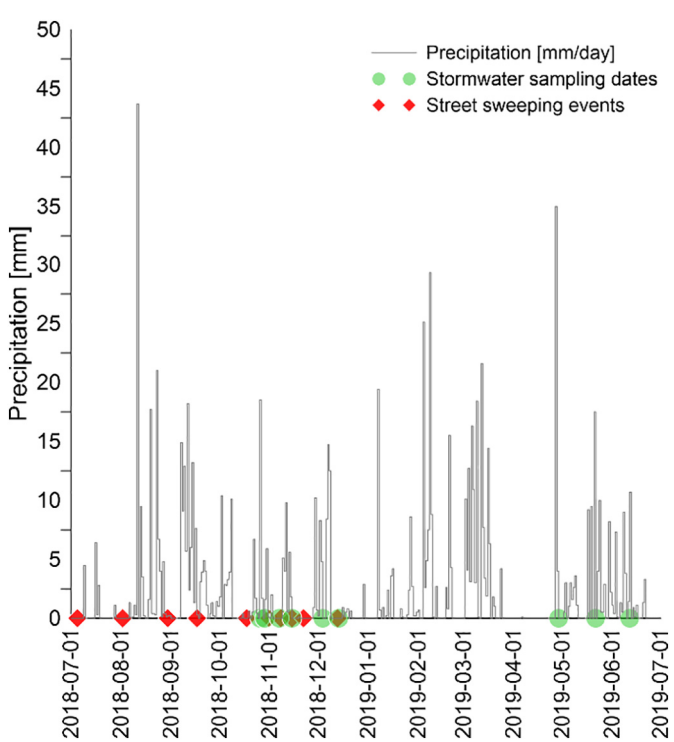

Fig. 3. Precipitation, stormwater sampling dates, and street sweeping events in the case study area in Gullbergsvass, Gothenburg (Tables S1 and S2).

The selected sampling point was located downstream in the stormwater system, at the point to which all the stormwater drained from the catchment is diverted before discharge to the river (X: 148 419.030, Y: 6 399 640.753). Two automatic samplers (model ISCO 6712) were connected to velocity flow meters (Triton+) and installed in a stormwater manhole located at the sampling point, allowing flow-weighted, composite sampling of stormwater throughout the runoff event. In 2018, the samplers were programmed to start collecting sub-samples at flows exceeding 5 L/s, but due to fluctuating flows, this was changed to water levels in the pipe (Ø 1200 mm) exceeding 75 mm. The level was chosen to discriminate against small rain events (<3 mm rainfall); this increased the likelihood of collecting sufficient sample volumes from each storm event. A tipping bucket rain gauge (type MJK, tipping volume 0.2 mm) was installed on one of the adjacent buildings to monitor rainfall. A national weather monitoring station is located close to the sampling point, and its open-source data were used to validate the rain gauge and estimate the precipitation.

2.3.2. Washwater and sweepsand from the street sweeper

Street sweeping sand and street sweeping washwater were sampled at a local deposition site shortly after sweeping of the streets in the reconstruction area. Samples were collected on seven occasions during autumn 2018, when weekly street sweeping was taking place. Washwater samples were collected directly from the sweeper's container in two 10 L glass bottles. After emptying of the sweeper's container, sweepsand was manually collected in two 10 L stainless-steel buckets. These samples were representative composite samples for the total volume of washwater and sweepsand collected from the whole area. The washwater included stormwater collected by the sweeper during sweeping (moist weather), water used by the sweeper to suspend fine particles during sweeping (dry weather), and suspended solids that did not settle inside the sweeper. The dry weight of the sweepsand was determined by drying triplicate sample aliquots of 1 to 2 g of the homogenized wet sediment at 105 °C to constant weight. Several liters of washwater were also filtered through 0.7 μm glass-fiber filters (Whatman GF/F), after which the solid materials on the filters were mixed to produce a composite and representative sample, henceforth referred to as washwater particles <0.7 μm. Directly after sampling, both washwater and sweepsand were divided into

plastic bottles/containers for analysis of general quality parameters and metals, and glass bottles/containers for OP and MP analysis. The samples were stored cool and dark, and sent to the analyzing laboratories short after sampling. Metals were analyzed in unfiltered and acid-digested washwater samples.

2.3.3. Road dust sampling

A Wet Dust Sampler II (WDS II, Fig. S3) was used to collect samples from the road surface. The WDS II methodology has previously been described in (Gustafsson et al., 2019; Jonsson et al., 2008; Lundberg et al., 2019). The samples were collected from the road surface directly after street sweeping at a construction area exit at Kilsgatan (Fig. 2). The WDS II uses an automated sampling procedure, where a known amount of high-pressurized de-ionized water (340 mL) is used to clean a road surface area of 20.43 cm^2. Each sampling is referred to as a "shot". The sample (containing the water and the road dust of the sampled surface area) is then transferred from the sampling chamber to a glass bottle using compressed air. WDS samples were collected in the area adjacent to the curb, where most of the road dust accumulates (Järlskog et al., 2020). Composite samples of 1 L, consisting of three shots each, were accomplished by moving the WDSII approximately 20 cm forward between shots, to cover a previously unsampled portion of the surface. The same sampling procedure was performed for all samples sent for analysis (metals, MP, and OP). Samples were stored cool and dark until analysis.

2.4. Microplastic analysis

Microparticles also referred to as micro-litter, includes microplastics, presented as the sub-categories: plastic, paint, tire wear, and bitumen, but henceforth referred to as microplastics. All sweepsand samples, and some washwater samples, contained large amounts of mineral particles, which would interfere with the detection of microplastics. For this reason, these samples had to undergo a density separation step prior to analysis. The samples were thoroughly mixed with saturated sodium iodide (NaI) solution (density 1.85 g cm^{-1}), after which the particles were left to separate, allowing heavier particles to sink to the bottom and lighter ones to float to the surface, where they could be collected. A more detailed description of the separation step can be found in Järlskog et al. (2020). Unused tire tread has an approximate density of 0.8–1.2 g/cm^3 (Kreider et al., 2010; Rhodes et al., 2012; Sofi, 2018), and the density of bitumen from pavements is 0.9–1.1 g/cm^3 (Nynas, 2017). These particles can therefore be expected to float. However, TWP often contain mineral encrustations, resulting in a higher density than the tire material itself. The material that settled on the bottom (particles with a density > 1.85 g cm^{-1}) was not analyzed in this study, why heavy TWP may have been dismissed and the concentrations in the samples somewhat underestimated. The densities of the most commonly used synthetic polymers are well below the density of the NaI solution used in the present study. Most plastic particles present in the samples could therefore be expected to end up in the surface water after separation. No density separation was needed for the stormwater samples since they contained only small amounts of mineral particles. The samples were filtered in sequence over nylon filters with mesh sizes of 300 and 100 μm; in some cases, also 20 μm (Table S10). All filters were analyzed using a stereomicroscope (Leica M205 C 80–160×) and the identified anthropogenic particles were categorized into TWP, paint particles, other microplastic particles, and bitumen, see further description in (Järlskog et al., 2020). The results are presented as number of particles ≥100 μm, (includes the sum of particles collected on both the 300 and 100 μm filters), and as number of particles ≥20 μm, (the total sum of particles collected on the 300, 100, and 20 μm filters). Procedural blanks were prepared by running the protocols for sweepsand, washwater, and stormwater (density separation and filtration) using only saturated NaI or Milli-Q water. Two blanks were carried out for each sample category. The only anthropogenic particles detected were non-synthetic fibers at concentrations between 0 and 4 per blank sample. Sample contamination during laboratory work was therefore not considered to have had any effect on the evaluation of the MP data.

2.5. Analysis of metals and organic pollutants

The studied OP were selected from the list of priority pollutants suggested by Markiewicz et al. (2017) and were analyzed with sensitive, targeted GC/MS methods. A few samples were also analyzed with a new sensitive, non-targeted screening method to enable identification of additional groups of OP. Stormwater, sweepsand, washwater and road dust samples were analyzed for the presence of metals, 16 specific US EPA PAH (PAH), nine oxy-PAH, 13 phthalates, five fractions of aromatic hydrocarbons C_8–C_{35} (aromatics), BTEX (benzene, toluene, ethylbenzenes and xylenes), six fractions of aliphatic hydrocarbons (aliphatics) C_5–C_{35}, and alkylated PAH as aromatics >C_{16}–C_{35} (sum of methyl pyrenes, methyl fluoranthenes, methyl chrysenes, and methyl benzo(*a*)anthracenes). The analyses were carried out at commercial laboratories following standardized methods (Table 1). The PAH was

Table 1
Summary of the analytical methods used for each sample matrix.

Analyzed parameter	Sample matrix	Analytical approach	Method
Concentrations of metals	Stormwater (total and filtrate 0.45 μm)	ICP-MS	SS-EN ISO 17294-2
	Washwater and WDS-samples	ICP-SFMS	SS EN ISO 17294-1, 2 (mod) and EPA-method 200.8 (mod).
		ICP-AES	SS EN ISO 11885 (mod) and EPA-method 200.7 (mod)
	Sweepsand	ICP-SFMS	SS EN ISO 17294-1, 2 (mod) and EPA-method 200.8 (mod).
Concentration of specific OP	Stormwater	GC–MS for analysis of US EPA PAH (PAH-16) and aliphatics	PAH-16: commercial lab method; oil index(aliphatics): SS-EN ISO 9377-2 modified.
	Washwater (water and filtrate 0.7 μm) and WDS-samples	GC–MS for analysis of PAH-16, oxy-PAH, aromatics, and aliphatics	PAH-16, alkane and aromatic substances: commercial lab method modified after SPIMFAB US EPA PAH (PAH-16): US EPA8270, CSN EN ISO 6468 Phthalates: DIN EN ISO 18856
	Sweepsand	GC–MS for analysis of PAH-16, oxy-PAH, aromatics, aliphatics, and phthalates	PAH-16, alkane and aromatic substances: commercial lab method modified after SPIMFAB US EPA PAH (PAH-16): SS EN ISO 18287:2008 Phthalates: E DIN 19742 (2012-04) Sensitive PAH-method, not standardized, see 2.6
Screening of OP	Stormwater, Washwater and Sweepsand	Spectral deconvolution GC/MS	Not standardized (see Section 2.6)

divided into PAH-L (naphthalene, acenaphthylene, and acenaphthene), PAH-M (anthracene, fluorene, phenanthrene, fluoranthene, and pyrene), and PAH-H (benzo(*a*)anthracene, chrysene, benzo(*b*)fluoranthene, benzo(*k*)fluoranthene, benzo(*a*)pyrene, dibenzo(*ah*)anthracene, benzo(*ghi*)perylene, and indeno(123*cd*)pyrene). The sweepsand was not sieved (total fraction) prior to analysis of OP and metals.

2.6. Complementary analyses, organic pollutants

One sweepsand, two washwater, and one stormwater sample were analyzed with a particularly sensitive method for PAH (Table S7) and oxy-PAH but also screened for non-targeted OP, using a mass spectra enhancing method based on spectral deconvolution for GC/MS

Table 2
Concentrations of microplastics, metals, and organic pollutants in stormwater, washwater, washwater particles, sweepsand, and WDS-samples. Microplastics are presented as the sub-categories plastic, paint, and tire wear and bitumen. The concentrations are presented as mean values (max, min, and STD values are available in Table S8). Note that Σ particles ≥20 μm includes also particles ≥100 μm. In the microplastic analysis, a total of n samples was analyzed for microplastics ≥100 μm, after which n of these samples were also analyzed for microplastics <100 μm–≥20 μm). Metals and OP were analyzed using the commercial methods listed in Table 1, except for the sweepsand sample labeled "Sensitive method".

	Stormwater	Washwater	Washwater particles < 0.7 μm	Sweepsand		WDS
	[No. of particles/L]	[No. of particles/L]	[No. of particles/kg dw]			[No. of particles/L]
	≥100 μm n = 9, ≥20 μm n = 6	≥100 μm n = 6, ≥20 μm n = 2	n = 1	≥100 μm n = 7, ≥20 μm n = 3		n = 2
∑Plastic[a] ≥100 μm	18	41	N.A	1500		28
∑Plastic ≥20 μm	69	97	N.A	2500		N.A
∑Paint[b] ≥100 μm	0	44	N.A	3600		2
∑Paint ≥20 μm	14	38	N.A	10,000		N.A
Tire wear ≥100 μm	9	4200	N.A	150,000		1200
Tire wear ≥20 μm	220	24,000	N.A	850,000		N.A
Bitumen ≥100 μm	12	2400	N.A	95,000		N.A
Bitumen ≥20 μm	280	4700	N.A	230,000		N.A
	[μg/L]			[μg/kg dw]		[μg/L]
	n = 9	n = 6	n = 1	n = 7		n = 2
As	1.6	27	BQL	650		2.4
Cd	1.1	3.4	290	240		0.3
Co	3.3	120	5000	4500		13
Cr	6.5	300	19,000	13,000		30
Cu	210	1400	31,000	42,000		92
Mo	BQL	31	2200	3200		1.7
Ni	5.2	210	8800	8500		30
Pb	25	540	13,000	12,000		50
Zn	340	5900	110,000	130,000		470
V	BQL	520	19,000	14,000		58
					Sensitive method[c]	
Naphthalene, NAP	0.27	0.10	BQL	BQL	BQL	0.01
Acenaphthylene, ACY	0.02	0.08	BQL	BQL	BQL	BQL
Acenaphthene, ACE	0.03	0.13	BQL	BQL	BQL	BQL
Fluorene, FL	0.03	0.18	BQL	BQL	BQL	0.01
Phenanthrene, PHE	0.15	2.2	BQL		57	0.06
Anthracene, ANT	0.05	0.40	BQL	BQL	BQL	0.03
Fluoranthene, FLR	0.44	4.6	BQL	140	96	0.18
Pyrene, PYR	0.32	4.1	BQL	140	100	0.17
Benzo(*a*)anthracene, BaA	0.15	2.1	BQL		90	0.08
Chrysene, CHY	0.17	2.5	BQL		56	0.20
Benzo(*b*)fluoranthene, BbF	0.22	3.4	BQL	110	66	0.13
Benzo(*k*)fluoranthene, BkF	0.09	0.89	BQL	BQL	59	0.03
Benzo(*a*)pyrene, BaP	0.12	2.5	BQL	BQL	61	0.06
Dibenzo(*ah*)anthracene, DBahA	0.03	0.50	BQL	BQL	24	0.02
Benzo(*ghi*)perylene, BPY	0.13	2.2	BQL	BQL	71	0.07
Indeno(123 *cd*)pyrene, INP	0.09	2.1	BQL	BQL	49	0.06
∑PAH-L[d]	0.32	0.30	BQL	BQL	BQL	0.01
∑PAH-M[e]	0.98	12	BQL	310	260	0.43
∑PAH-H[f]	1.0	16	BQL	200	480	0.64
∑PAH16	2.3	28	BQL	490	730	1.1
Aliphatics >C_{10}-C_{12}	8.0	20.0	BQL	BQL		BQL
Aliphatics >C_{12}-C_{16}	25.0	80.0	BQL	BQL		BQL
Aliphatics >C_{16}-C_{35}[g]	760	3100	96,000	170,000		150
Aromatics >C_8-C_{10}	N.A	0.52	BQL	BQL		BQL
Aromatics >C_{10}–C_{16}	N.A	2.3	BQL	BQL		BQL
Methyl pyrenes + methyl fluoranthenes = ∑MP + MF	N.A	4.2	BQL	BQL		BQL
Methyl chrysenes + methyl benzo(*a*) anthracenes = ∑MC + MB(*a*)A	N.A	5.2	BQL	BQL		BQL
Aromatics >C_{16}-C_{35} = ∑MP + MF + MC + MB(*a*)A	N.A	9.4	BQL	BQL		BQL
Di ethyl phthalate	N.A	3.2		BQL		BQL
Di-n-butyl phthalate	N.A	1.3		70		BQL
Di-iso-butyl phthalate	N.A	1.2		BQL		BQL
Di-(2-ethyl hexyl)phthalate (DEHP)[h]	N.A	17.1				5.3
Di-iso-nonyl phthalate (DINP)	N.A	98		BQL		BQL

(Du and Zeisel, 2013) as a complementary analysis. Extraction was performed using a mixture of cyclohexane/ethyl acetate (75/25). A known mass of deuterated PAH was added as an internal standard. For the analysis of PAH, the mass spectrometer was run in the single-ion monitoring mode and each specific PAH was quantified using known masses, with three different deuterated PAH as quantification standards. The non-target screening was done by separating the compounds in the extracts in the GC-column, and each eluted compound was scanned with the MS in total-ion current mode, with mass-to-charge ratios (m/z) of 33–300. The largest peaks were identified using the mass spectra library NIST 2017 after the application of an advanced background subtraction algorithm, i.e. deconvolution (AMDIS). Sample spectra correspondence with NIST database spectra were evaluated based on the "hit rate", where a higher value (maximum 100) indicates higher correspondence with the database reference spectra. Hit rates below 80 are considered uncertain. The concentrations were calculated using the phenanthrene-d_{10} as quantification standard for all compounds (i.e. semi-quantification), which means that the calculated concentrations are approximate, but demonstrate the order of magnitude. Heavier aromatics and aliphatics can have several different isomers, which cannot be distinguished by their mass spectra. These isomers may be separated in the GC column, resulting in similar mass spectra, but different retention times. Also, the mass spectra cannot provide information on the exact position of detected double bonds and alkyl groups. The identified compounds were manually classified into the following groups of OP: alkanes, alkenes, PAH, ketones, alcohols, acids, esters, ethers, phthalates, amines + amides + N_{org} (nitrogen-containing organic compounds), halogenated organic compounds, siloxanes + S_{org} (sulfurous organic compounds), and others. Note that most of the individual OP may belong to more than one group of compounds but are only reported in one of the groups.

2.7. Other water quality analysis

Total/dissolved organic carbon (TOC/DOC) was measured following the method CSN EN 1484/13370 with a TOC analyzer and IR detection. Total/volatile suspended solids (TSS/VSS) were measured following EN ISO 872:2005 with filtration through 1.2 μm glass fiber filters (Table S3). For stormwater, additional analyses (pH, turbidity, conductivity, phosphorous, and nitrogen) were carried out using a HANNA Multiparameter (HI9829) (Table S6).

2.8. Statistical analyses

To identify the relationship between PAH, metals and TWP, Pearson correlation coefficients were calculated in Excel (Table S9).

3. Results and discussion

Results from the analysis of all sample matrices: stormwater, washwater, washwater particles >0.7 μm, sweepsand, and WDS samples are presented in Table 2 (and Tables S3–S8), and include investigated MP, metals, and OP detected at concentrations above the quantification limit in at least one of the investigated samples. The results for samples analyzed by the more sensitive method for PAH, oxy-PAH, and BTEX are presented in Table S7 and the results from the screening of OP is presented in Chapter 3.5. Microplastics, metals, and all analyzed OP, except oxy-PAH, BTEX, and short-chain aliphatics up to C_{10}, were quantified in most samples (Table 2). The high concentrations of MP, TWP, metals, PAH, alkylated PAH, aliphatics >C_{16}–C_{35}, and phthalates found in the washwater and sweepsand indicate that street sweeping may be an effective measure to reduce the spread of TWP and pollutants. The results from the analysis of general water quality parameters, such as pH, conductivity, turbidity, TOC/DOC, and TSS are presented in Tables S3–6.

3.1. Stormwater

All MP categories, i.e. plastic particles, paint particles, TWP, and bitumen particles, in the size fraction ≥20 μm were quantified in six out of nine stormwater samples, and the relative distribution was found to be: bitumen 47%, TWP 39%, plastics 12%, and paint 2% (Fig. 4). The total number of MP particles was up to 1500/L, which was lower than the results from a previous study in Gothenburg (4400 MP/L) (Järlskog et al., 2020), but higher than what was found in stormwater collected in Tijuana in Mexico, where the mean MP (≥25 μm) concentration varied between 66 and 191 particles/L, with the highest mean concentrations found at an industrial site (Piñon-Colin et al., 2020). Further, the study found that polyethylene was the most abundant MP, however, neither TWP nor black particles were analyzed specifically. Similar results were found by Pramanik et al. (2020), who also did not analyze TWP, but found that MP were present in all stormwater samples and that microfibers were most frequently found among the analyzed MP (50%).

Of MP in the size fraction ≥100 μm, bitumen made up 24% (analyzed in six out of nine samples), 27% TWP, 48% plastics, and 1% paint (Fig. 4). As shown in Table 2, there were 4, 44, 24, and 23 times more plastics, paints, TWP, and bitumen, respectively, in the smaller fraction, ≥20 μm, than in the fraction ≥100 μm. The finding that the majority of the TWP belonged to the smaller fraction can be explained by the fact that TWP ≥100 μm are less likely to be found in the water phase. This is in line with studies by (Borg Olesen et al., 2019; Dröge et al., 2019; Liu et al., 2019a; Liu et al., 2019b) which analyzed MP in stormwater and sediment ponds, and with Ziajahromi et al. (2020), who analyzed microplastic pollution in a stormwater floating treatment wetland, with focus on TWP. The microplastic concentrations in the water/floating phase (inlet and outlet) were low, however more microplastics were found in the sediment phase (samples collected at the bottom of the wetland), and 15–38% of the particles were identified as TWP (ibid). Previous findings in sediments in the Alto-Tietê catchment area in Poá City, São Paulo, Brazil, found that tire wear constituted around 30% of the total MP (Braga Moruzzi et al. (2020), confirming the findings of this study that tire wear is a major source of MP both in stormwater and in receiving environments.

Previous calculations and studies indicate that theoretical estimates of TWP emissions entering the stormwater are many times higher

Notes to Table 2:
BQL – below the quantification limit.
N.A – not analyzed.
[a] ∑Plastic – identified polymer particles of other origins than paint, tire wear, or bitumen (fibers, fragments, and films).
[b] ∑Paint – identified paint particles (fragment and flakes).
[c] Sensitive method: Sweepsand sample (2018-10-18) analyzed using a more sensitive method (see chapter 2.6). NB: All PAH in the sweepsand sample analyzed with the commercial method was BQL 2018-10-18.
[d] ∑PAH-L – PAH with low molecular weight (naphthalene, acenaphthylene, and acenaphthene).
[e] ∑PAH-M –PAH with medium molecular weight (anthracene, fluorene, phenanthrene, fluoranthene, and pyrene).
[f] ∑PAH-H PAH with high molecular weight (benzo(*a*)anthracene, chrysene, benzo(*b*)fluoranthene, benzo(*k*)fluoranthene, benzo(*a*)pyrene, dibenzo(*ah*)anthracene, benzo(*ghi*)perylene, and indeno(123*cd*)pyrene).
[g] ∑aliphatics C_5-C_{35} – Aliphatics C_5-C_{10} were also analyzed, but the concentrations were below the limit for quantification.
[h] Additional phthalates (di methyl phthalate, di-n-propyl phthalate, di-pentyl phthalate, di-n-octyl phthalate (DNOP), butyl-benzyl phthalate, di-cyclo-hexyl phthalate, di-iso-decyl phthalate (DIDP) and di-n-hexyl phthalate) were also analyzed, but the concentrations were below the limit for quantification.

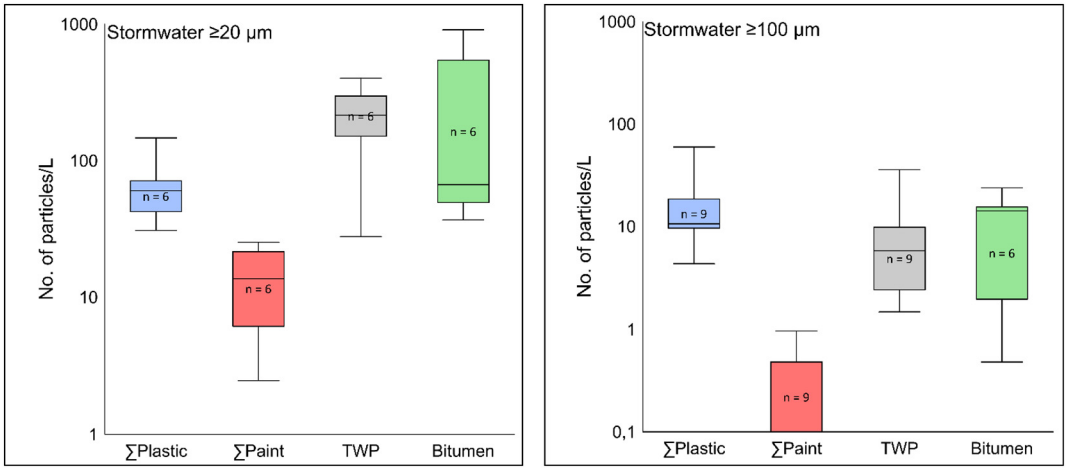

Fig. 4. Event mean concentrations of different categories of microplastics (≥20 µm left and ≥100 µm right) identified in stormwater. n = number of samples. Nine stormwater samples were analyzed for particles ≥100 µm, six of which were also analyzed for particles ≥20 µm.

(10–100) than the observed concentrations of particles in stormwater (Järlskog et al., 2020; Tumlin and Bertholds, 2020). A possible explanation for this is that the bigger particles remain on the road surface longer than expected and end up on roadsides and in the soil (Baensch-Baltruschat et al., 2020). This is well in line with the results from this study (more heavy traffic) where the WDS samples contain much more TWP than the previous study by Järlskog et al., 2020. Smaller particles may be more commonly found in, and transported via, the air than expected. It may also be due to the runoff not entering the stormwater pipes correctly, or because of leakages from the pipes. The bigger size fractions may also be deposited on the sides or at the bottom of the pipes. Notice that TWP also may occur largely in fractions <20 µm which was not analyzed in this project. The results indicate that there is a need to better understand the initial spreading patterns, using field, laboratory, and modeling studies. The mean values ($n = 9$) of the metal analysis carried out on the total and filtrated (<0.45 µm) stormwater samples taken at 9 rain events are shown in Fig. 4. The Event Mean Concentrations (EMC) of the metals were several times higher in the total samples than in the filtrated samples. This was particularly true for Pb, which like Cu, readily binds to organic matter due to the formation of strong complexes with humic acids (Kalmykova et al., 2008). The metal EMCs were of the same order of magnitude throughout the study period, which meant that it was not possible to identify a clear trend over time. A comparison with the local guideline values for the release of polluted water to recipients (black cross in Fig. 5) showed that the guideline values were exceeded for all metals except Cr. Possible explanations for the high metal concentrations in the stormwater include leaching from construction materials, copper roofs on the surrounding buildings, and traffic (Huber et al., 2016; Hwang et al., 2016; Johansson et al., 2009; Müller et al., 2020).

The EMCs of PAH varied between 0.2 and 6.4 µg/L in the stormwater samples, which is above the City of Gothenburg's guideline value for polluted waters emitted to recipients, which is a benzo(a)pyrene concentration of 0.27 µg/L, as well as the Canadian Water Quality Guidelines for protection of aquatic life in freshwater. The PAH concentrations were of the same order of magnitude as reported for stormwater affected by traffic in other studies (Hou and Li, 2018; Pettersson et al., 2005; Zgheib et al., 2012), but lower than reported in an extensive study of stormwater at 21 sites in the US, where total PAH concentrations were up to 180 µg/L, with concentrations >10 µg/L at 42% of the sites (Masoner et al., 2019). Potential sources of the PAH at this construction site include construction machines (Zhang et al., 2020) and the relatively high occurrence of heavy traffic (although the total AADT was fairly low) leading to high emissions due to tire and road wear (Baensch-Baltruschat et al., 2020). For the larger PAH, the highest EMCs were seen in May 2019, however, the same was not true for PAH-L (Fig. 6 and Table S6). The highest concentration of PAH-L was observed in December 2018, coinciding with the second-highest concentrations for the other PAH. The concentrations of the most volatile PAH-L fraction were expected to be temperature-dependent, and therefore season dependent, which is well in line with their peak occurrence being observed in December. The high EMCs of the PAH in December and May can be explained by intensive construction activities during the periods prior to those sampling dates. In addition to variations in construction activities, differences between the PAH measurements may also be due to meteorological variations, such as wind speed, wind direction, precipitation, and temperature. The pollutant loads in stormwater depend on antecedent dry days, rain depth, rain intensity and duration (Murphy et al., 2015). Furthermore, deviations may be due to variations in street sweeping activities (Amato et al., 2010). A possible explanation for the high concentrations found in May 2019 is that no street sweeping had been performed since December 2018, allowing PAH to accumulate on the road surface and be further transported to, and with, stormwater. In most stormwater samples,

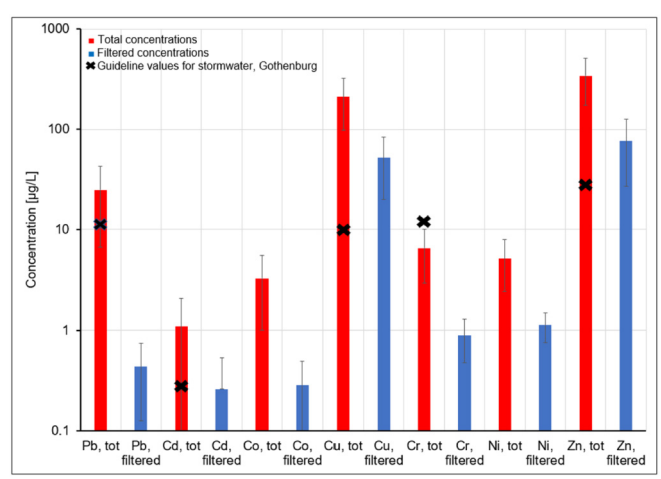

Fig. 5. Event mean concentrations (n = 9) of total and filtered toxic trace metals in stormwater collected in Gullbergsvass during autumn 2018 and spring 2019. Note that the y-axis has a 10 logarithmic scale and concentrations are event mean concentrations. Black crosses represent guideline values for the release of polluted water into recipients (Miljöförvaltningen, 2013).

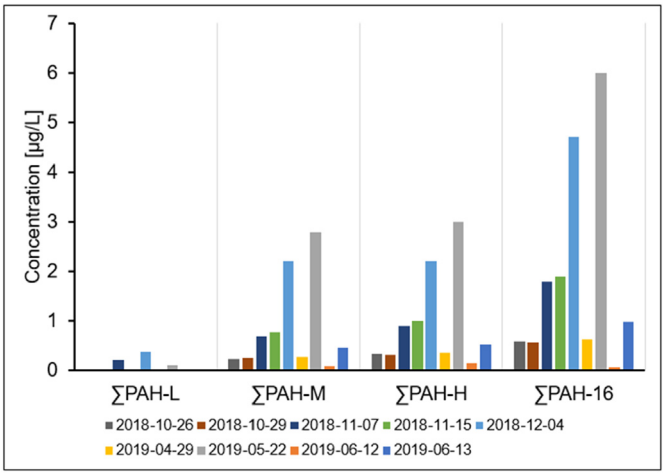

Fig. 6. Event mean concentrations of PAH in stormwater collected in Gullbergsvass during a period of weekly street sweeping in autumn 2018 ($n = 5$) and a period without street sweeping in spring 2019 ($n = 4$).

the PAH-M and PAH-L exceeded the guideline values by more than a factor of 10 (Table S6) (CCME, 2010; Miljöförvaltningen, 2013).

The highest EMC of the total petroleum hydrocarbons, fractions $>C_{10}$–C_{40}, in stormwater was 3000 µg/L, found 2018-12-04, when the EMC for the fraction $>C_{16}$–C_{35} was 2500 µg/L. (Fig. 7). This is well above the local guideline for the release of water polluted with aliphatics C_{10}–C_{40} to recipient waters, which is 1000 µg/L. It is not uncommon to find total petroleum hydrocarbon concentrations exceeding 1000 µg/L in road runoff (Flanagan et al., 2018; Snilsberg and Gryteselv, 2016). Additionally, in December 2018 the weather was dry for a period, but heavy rain fell a few days before the sampling, likely resulting in both high runoff volumes and high amounts of road dust, as well as the release of higher amounts of aliphatics with higher molecular weight and lower water solubility into the stormwater.

3.2. Washwater and sweepsand

3.2.1. Microplastics

The total number of MP particles (plastics + paints + TWP + bitumen) ≥20 µm in washwater from the sweeper was up to 5.1×10^4/L (mean 2.9×10^6/L) and, correspondingly, in the sweepsand was up to 2.6×10^6/L (mean 1.1×10^6/L). The high concentration in the sweepsand agrees with the high concentrations found in sediments

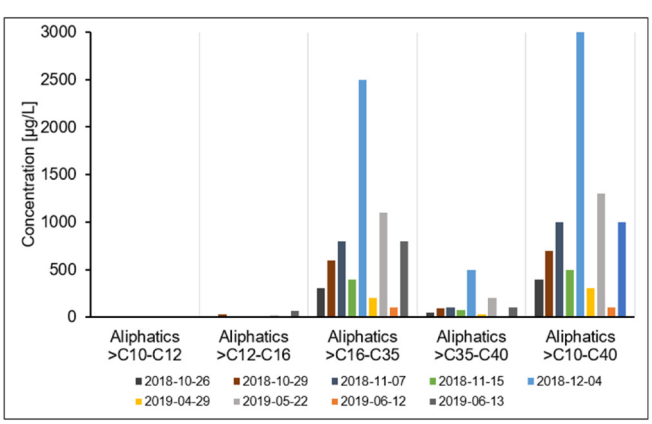

Fig. 7. Event mean concentrations of aliphatic petroleum hydrocarbons, fractions C_{10}–C_{40}, in stormwater collected in Gullbergsvass during a period of weekly street sweeping in autumn 2018 (n = 5) and a period without street sweeping in spring 2019 (n = 4).

compared to the water/floating phase, as discussed in (Borg Olesen et al., 2019; Liu et al., 2019a; Liu et al., 2019b; Ziajahromi et al., 2020). The ratios of 20 µm–100 µm / ≥100 µm (small particle fraction/large particle fraction) were 13.9 for stormwater, 3.3 for washwater and 3.7 for sweepsand. A ratio over 1 in all matrices indicate that the majority of the particles are small found in the smaller fraction.

A high MP 20–100 µm / ≥100 µm ratio was also found for TWP in stormwater in a recent study by Järlskog et al. (2020) and the higher ratios (i.e. more of the smaller particles) found in the stormwater compared to the washwater and sweepsand can be explained by the fact that lighter and smaller particles enter the stormwater deposit more easily than larger particles, and that larger/heavier particles are less mobile and therefore remain on the street, where the sweeper can collect them, for a longer period of time than smaller particles. The concentration of paint particles in the sweepsand was relatively high (Table 2). This strongly indicates that the paint particles found on the road have a higher density than the TWP and are therefore more likely to remain settled on the ground. However, during rain events, smaller paint fragments are more easily resuspended from the road than larger ones. Also, plastics, TWP, and bitumen seem to have a low tendency to be transported in water, but less pronounced than the paint particles ≥100 µm. In agreement with a previous study by Järlskog et al. (2020), the occurrence of TWP ≥100 µm in the washwater was higher than the occurrence of plastics ≥100 µm (more than 100 times) and paint ≥100 µm (more than 100 times) (Fig. 8, left). This was also the case in the sweepsand, where TWP concentrations were found to be 100 and 40 times, respectively, higher than those of plastics and paint particles in the size fraction ≥100 µm (Fig. 8, right). This confirms that tire wear is a dominant source of microplastics in urban and trafficked areas. The total concentrations of TWP in the washwater were within the same range as previously found in the more traffic dense area studied in Järlskog et al. (2020), which may be due to the higher proportion of heavy traffic in the present study. This study also found greater variation in the TWP concentrations (based on the number of particles), which may be explained by more varying activities and traffic intensities in the investigated area during the sampling period. The number of TWP particles in washwater ≥100 µm in Järlskog et al. (2020) varied between 17 and 4200 particles/L compared to 1–23,000 particles/L in the present study.

The smaller size fraction (≥20 µm) and bitumen were analyzed in two of the washwater samples (2018-08-02 and 2018-11-15, see Fig. S1). In these samples, the number of plastic particles was up to 120/L (mean 97/L), the number of paint particles was up to 31/L (mean 19/L), the number of TWP particles was up to 45,000/L (mean 24,000/L), and the number of bitumen particles was up to 6500/L (mean 4700/L). The relative contribution of bitumen to the total number of microplastic particles in the washwater was 16%, the corresponding pattern for TWP was 83%, and <1% of the samples contained plastics or paint. Plastics and paint were only found in the form of larger particles, not yet fragmented, for which the source is expected to be close in time and space. It is likely that the plastic and paint had been emitted relatively recently from a nearby source, as the degradation and fragmentation increase with time and thereby also with distance from the emission source (Min et al., 2020). The total number of MP particles ≥20 µm was 8 times higher in August 2018 than in the middle of November 2018. One possible explanation is that the roads had not been swept during the summer, and 2018-08-02 was the first sweeping event since May, indicating a positive impact of the street sweeping during autumn 2018.

The smaller size fraction (≥20 µm) and bitumen were analyzed in three of the sweepsand samples (2018-08-02, 2018-11-15, and 2018-12-13, see Fig. S2). In these samples, the number of plastic particles was up to 3300/kg dw (mean 2500/kg dw), the number of paint particles was up to 12,500/kg dw (mean 10,300/kg dw), TWP particles up to 2.1×10^6/kg dw (mean 850,000/kg dw), and bitumen particles up to 540,000/kg dw (mean 230,000/kg dw). The relative contribution of

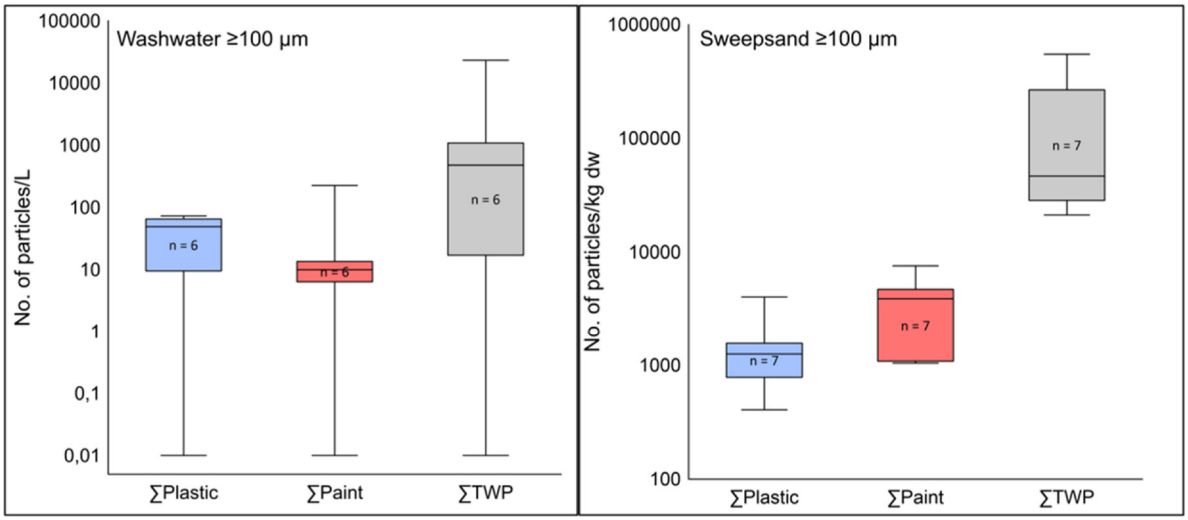

Fig. 8. Left: Concentrations (particles/L) of microplastics ≥100 μm identified in washwater. Right: Concentrations (particles/kg dw) of microplastics ≥100 μm identified in sweepsand.

TWP to the total number of microplastic particles in sweepsand was 78%, for bitumen 21%, and 1% of the particles consisted of plastic and/or paint. In a study by Dehghani et al. (2017), the authors collected road dust by manual street sweeping and found 2900–20,200 MP particles/kg dw, however the size limit in that study was 50 μm, and a lower size limit would probably have resulted in a higher number of microplastic particles.

Considerable work is ongoing globally to develop and validate analytical methods for identifying and quantifying TWP. Progress have been made, and several promising analytical methods have been published recently (Eisentraut et al., 2018; Kovochich et al., 2021; Mengistu et al., 2019; Tian et al., 2021). When this study was conducted, no suitable analytical method for TWP was available, why stereo microscopy in combination with melt-tests and tactile identification were used. Parallel to the method development work for this study, several samples were analyzed using novel methods, i.e. Fourier-transform infrared spectroscopy (FTIR) and pyrolysis-GC/MS. Results from those analyses are not presented here, but they indicated that TWP were in the same size range as the results from stereo microscopy.

3.2.2. Metals

The concentrations of Cu and Zn in washwater were at their lowest in August 2018, after which they increased over the autumn to a peak of 1700 μg/L and 8100 μg/L in December, respectively (Table S3). These concentrations are extremely high and several hundred times higher than the City of Gothenburg's guideline values for release of polluted water to recipients, which are 10 μg/L for Cu and 30 μg/L for Zn. The rest of the analyzed metals (i.e. As, Cd, Cr, Ni, and Pb) were also found in concentrations several times higher than the guidelines. The concentrations of Cu and Zn were consistently high and relatively stable in the sweepsand, however, concentrations of up to 69,000 μg/kg for Cu and 190,000 μg/kg for Zn were found (Table S4). These concentrations exceed the Canadian interim sediment quality guidelines for freshwater, which are 35,700 μg /kg for Cu and 123,000 μg /kg for Zn. Cu and Zn are the most commonly measured metals in road runoff and two primary contributors to metals in road-deposited solids, however, both Zn and Cu could originate from several sources, including construction works in the area, but both are also ubiquitous in urban environments (Huber et al., 2016; Hwang et al., 2016; Johansson et al., 2009; Müller et al., 2020). The connection with the construction work is in line with the lowest concentrations of Zn being measured immediately after a holiday period, during which there was less activity in the construction area. For comparison, in a study by Nawrot et al. (2020) the authors analyzed metals in sweepsand (road sweeping waste) and found concentrations of the same magnitude, or even higher, than in the present study. In two studies, one from Australia and another from Korea, it was also shown that road deposited solids in highly trafficked areas had about the same metal concentrations as found in this study (Choi et al., 2020; Ma et al., 2021). These findings indicate that streets in different parts of the world are contaminated with high concentrations of toxic metals and need treatment. Due to the high concentrations of metals in the washwater, it should not be permitted to deposit material from the sweeper without treatment. If the metals in the washwater could be trapped in filters, the material may be of interest for metal recovery (Norén et al., 2020).

3.2.3. Organic pollutants

In this study, only a few OP were quantified in the sweepsand using a commercial analytical method. However, the total PAH concentrations were as high as 610 μg/kg dw (phenanthrene, fluoranthene, pyrene, benzo(*a*)anthracene, chrysene, and benzo(*b*)fluoranthene detected) (Table 2 and Table S8). Compared to the Canadian Sediment Quality Guidelines for specific PAH, which are 7–100 μg/kg dw, the concentrations are high. In the sample from 2018-10-18 (Table S4), the commercial analysis method could not quantify any of the PAH, however, the more sensitive method found the PAH concentration to be 732 μg/kg dw (all PAH quantified except PAH-L). Environmental concentrations are often close to the quantification limits offered by commercial laboratories, why it is important to develop more sensitive analytical methods for PAH. However, these results are well in line with analysis of road dust in Iran where total PAH concentrations varied between 170 and 5300 μg/kg, with an average of 770 μg/kg (Abbasnejad et al., 2019), but also with results from a study in northern Vietnam where the concentrations in road dust varied from 170 μg/kg in suburban/rural till 5000 μg/kg in urban/industrial areas (Anh et al., 2019a). In the washwater, the mean concentration of PAH was 28 μg/L (Table 2), which is high compared to the local guideline value for benzo(*a*)pyrene, which is 0.27 μg/L. In contrast to the stormwater samples, the changes in the concentrations of PAH in washwater showed no clear trend over time, indicating that the sources of PAH found in the road are near and recent. The relative concentrations of PAH indicating that traffic is the most dominating source to the PAH pollution in washwater, see Chapter 3.5 Composition of polycyclic aromatic hydrocarbons. In the washwater samples, the aromatics C_8–C_{10} and C_{10}–C_{16} were also found in concentrations of up to 1.4 μg/L and 3.3 μg/L, respectively (Table 2). The concentration of alkylated PAH (aromaticsC_{16}–C_{35} =

sum of methyl pyrenes, methyl fluoranthenes, methyl chrysenes, and methyl benzo(*a*)anthracenes) was up to 15 μg/L (Tables 2 and S8), but below the limit of quantification in the sweepsand samples. In the study from northern Vietnam, 15 methylated PAH varied in concentrations between 170 and 510 μg/kg with a mean of 270 μg/kg in road dust from an urban area (Anh et al., 2019a).

The highest concentrations of petroleum hydrocarbons related to the aliphatic C_{16}–C_{35} fraction in both washwater (≤78,000 μg/L) and sweepsand (≤390,000 μg/kg dw) (Table 2 and Table S8). These concentrations were up to several hundred times higher than the City of Gothenburg's guidelines for release of polluted water to recipients, which is 1000 μg/L, and higher than the general quality guideline for contaminated soil in Sweden, which is 100 mg/kg. The concentrations of aliphatics found in washwater and sweepsand were in the same order of magnitude as found in (Polukarova et al., 2020), but much higher than the aliphatics C_1–C_{33} measured in northern Vietnam at ≤63,000 μg/kg (Anh et al., 2019b). The relative composition of the aliphatics in washwater and sweepsand was consistent with the low water solubility of heavy aliphatic compounds (i.e. a relatively higher percentage of the higher aliphatics was found in the sweepsand). As for the PAH, the concentrations of petroleum hydrocarbons were high and relatively constant over time, both in the washwater and the sweepsand. The most likely sources to the high concentrations of aliphatics in washwater and sweepsand are exhaust and diesel fuel emitted in gas and particulate phase from diesel engines (Alam et al., 2019; Anh et al., 2019b), as well as engine oil and asphalt wear (Hwang et al., 2019).

Of the analyzed phthalates (Table S4), the following occurred in amounts exceeding the quantification limit in washwater: DEP, DBP, DIBP, DEHP, and DINP. Correspondingly in sweepsand: DBP and DEHP (Table 2). The highest concentrations were found for DEHP, at 24 μg/L in washwater and up to 3400 μg/kg dw in sweepsand. These concentrations exceed the Canadian Quality Guideline for the protection of aquatic life in freshwater, which is 16 μg/L for DEHP. The concentrations of DBP and DEHP in the sweepsand were of the same magnitude as found in previous studies, i.e. DEHP 340–504 μg/kg dw and DBP 70–5700 μg/kg dw (Anh et al., 2019b; Björklund et al., 2009; Polukarova et al., 2020; Škrbić et al., 2016), indicating that these compounds are commonly occurring in urban dust and sediment. The relatively high concentrations of DEHP in some of the sweepsand samples (Table S4) indicates the presence of additional sources, other than traffic, such as the release of plastic materials during the extensive construction of a road tunnel, bridge, roads, and new buildings, which was ongoing in the area. DEP, DIBP, and DINP were found in the washwater but not in the sweepsand. These results may indicate that they are emitted and transported in the form of smaller particles, as emulsions or colloidal particles in the washwater (Markiewicz et al., 2017). A recent study found that these phthalates are released from vehicles during car wash and simulated rain, although in the order DINP ≫ DEHP ≫ DEP (Markiewicz et al., in prep). This suggests that their origin is often, but not solely, traffic-related. DINP is also used in various types of plastic consumer products, such as PVC flooring, automobile interiors, wire and cable insulation, gloves, tubing, garden hoses, and shoes, as well as in non-PVC products, such as some inks and pigments, adhesives, sealants, paints, and lacquers. Large amounts of DBP and DEHP have been used as plasticizers in PVC, paint, sealants, and car care products, and the compounds are commonly found in urban runoff and road dust (Anh et al., 2019b; Björklund et al., 2009; Flanagan et al., 2018; Škrbić et al., 2016).

Although this study indicates that street sweeping may be an effective measure, maintenance of pipes and other maintenance measures should also be further investigated as measures to limit the spread of pollutants. The treatment of the collected materials also needs further investigation, as previously concluded by (Baensch-Baltruschat et al., 2020; Dröge and Hulskotte, 2018).

3.3. Samples collected with the Wet Dust Sampler

The particle size distribution in the WDS II samples (2018-09-17) indicates that the mean size for all particles was approximately 50 μm, which is well in line with previous studies of road dust in Sweden (Gustafsson et al., 2019). The concentrations of plastic and paint particles were within the same range as at another location in Gothenburg (Fig. 9), studied by Järlskog et al. (2020), 30 particles/L compared to 48 particles/L. The number of TWP per liter was more than 10 times higher in the present study, 1200 particles/L compared to 112 particles/L. A possible explanation is that the high ratio of heavy traffic in the current study area led to greater wear, as the vehicles were heavier, the number of wheels per vehicle was higher, and the driving patterns were different. In this study, the samples were collected near a construction site exit, where vehicles performed many starts and stops and heavy turns, resulting in more tire wear. Another explanation is that the samples in the previous study were collected during the winter/early spring, when studded tires are in use, resulting in less tire wear and more road wear being released. An interesting observation in the comparison between this study and the previous study by Järlskog et al. (2020) is that the behavior of the stormwater concentrations is the opposite of the WDS results. The most likely explanation is that the runoff was lower in this study due to meteorological factors, such as wind and precipitation. It may also be the case that the particles were larger and less mobile, due to more heavy traffic in this study than in the previous study.

In the present study, TWP stands for 97% of all identified particles in the size range ≥ 100 μm in the WDS samples. The high percentage of TWP agrees with the percentage found in other sample matrices, confirming that traffic is the major source of MP in the area.

In the WDS samples collected after sweeping in Gullbergsvass, the concentration of DEHP was 5.3 μg/L, which is within the same range as in the previous study by (Polukarova et al., 2020), where it was 3.7 μg/L. Total aliphatics C_{16}–C_{35} was 150 μg/L, which is five times lower than the 783 μg/L found in the previous study.

3.4. Correlations

For the year overall, no correlation was found between the EMC of TWP (≥100 μm) and PAH or metals ($r = -0.034$) in the stormwater samples, however, there was a correlation when samples were divided into a non-studded tire season (16/4–30/9) (e.g. TWP and PAH $r = 0.96$, $p < 0.05$; TWP and Zn $r = 0.88$) and a studded tire season (1/10–15/4)

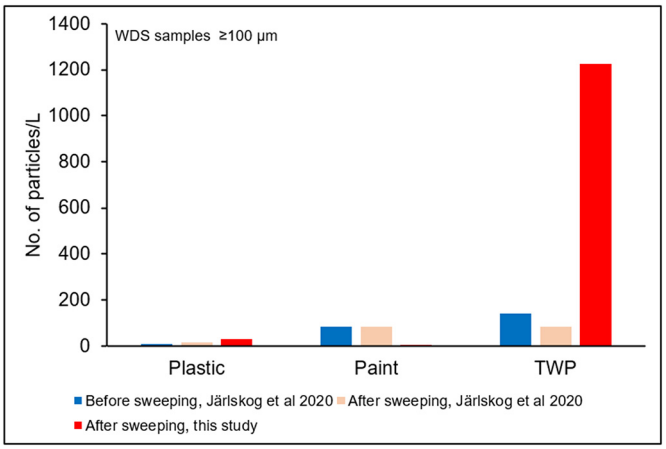

Fig. 9. Concentrations of plastics, paint and TWP (particles/L) in WDS samples collected after sweeping in Gullbergsvass 2018-09-17 (mean of six shots over a total area of 123 cm^2). These are compared to the mean value of samples (mean of 12 shots over a total area of 245 cm^2 before and after sweeping, respectively) collected in Vitsippsbäcken during March and April 2018, as reported in Järlskog et al. (2020).

(e.g. TWP and PAH, $r = 0.88$; TWP and Zn $r = 0.46$). The dates have been selected according to the periods when the use of studded tires is permitted in Sweden. Zn is often used as a tracer for tire wear (Councell et al., 2004; Degaffe and Turner, 2011; Rhodes et al., 2012), the lower correlation with tires during the non-studded season may indicate that the generation of tire wear is lower. This is well in line with previous research (Järlskog et al., 2020). The relatively high correlation between TWP and PAH during the period when studded tires are used may be due to sources and meteorological conditions. A high positive correlation between individual PAH in stormwater samples collected during the winter was also found ($r > 0.9$), indicating that many of the PAH may originate from the same sources (e.g. phenanthrene, fluoranthene, and pyrene). Due to the small number of samples of PAH (Table S6), additional conclusions cannot be drawn, and further investigations are needed to confirm the sources. During the period when studded tires are not used, TRWP emissions are expected to be dominated by tire wear. During the period with studded tires (winter), TRWP emissions are expected to be dominated by road wear (Gustafsson et al., 2008), containing higher amounts of bitumen-related PAH, and the temperature is lower. The lower temperature may contribute to a higher proportion of PAH-L in the stormwater than during warmer periods with higher evaporation.

A high positive correlation between the traffic-related metals Zn, Ni, Cu and Cr, and PAH was found in the stormwater samples, both during the studded tire season (0.51–0.98), and the non-studded tire season (0.3–0.76) (Table S9). Potential reasons for the lower correlation found in the spring include different emissions and changes in the sources of emissions from the construction activities. Variations in the weather conditions on the sampling occasions and the frequent street sweeping may also have impacted the results.

Similar correlation tests were performed for the washwater samples; however, no correlation was found neither between TWP and PAH, nor between TWP and metals. A high positive correlation between individual PAH was identified, although the number of analyzed samples was too small (four samples from the summer and two samples from the winter) to draw any definitive conclusions.

3.5. Composition of polycyclic aromatic hydrocarbons

The relative compositions of PAH-M and PAH-L in washwater, sweepsand (based on results using the more sensitive analytical method), and stormwater from the Gullbergsvass area were very similar, with the following dominating PAH: fluoranthene, pyrene, ≈benzo(*a*)anthracene, benzo(*k*)fluoranthene and phenanthrene (Fig. 9), indicating that they originate from the same source. These PAH are about the same as analyzed in road dust in Iran where the most abundant PAH found were fluoranthene, phenanthrene, pyrene, and chrysene which indicate the same sources of pollution (Abbasnejad et al., 2019), and in the urban area in northern Vietnam where the dominating PAH were phenanthrene, pyrene, fluoranthene and chrysene (Anh et al., 2019a). The PAH composition in this study suggests a mixture of many traffic-related sources: tire wear, vehicle exhausts, brake linings, motor lubricant oils, and road surface wear (Markiewicz et al., 2017; Zhang et al., 2020). PAH-H made up approximately 50–65% of the PAH in the samples in this study, which is slightly higher than the 40–55% previously found by Polukarova et al. (2020). This deviation can be explained by meteorological differences but may also be due to the higher proportion of heavy traffic in Gullbergsvass than in the previous study (ibid). Benzo(*a*)anthracene found in the Gullbergsvass samples may originate from road surface wear (Markiewicz et al., 2017). To investigate the relative PAH compositions in different matrices, the results from this study were compared to a profile of PAH present in laboratory-tested bitumen, the mean PAH profile from analyzed summer and winter tires (Gustafsson et al., 2009), and data profiles from Markiewicz et al. (2017), see Fig. 10. Notice that the selected washwater, stormwater, and sweepsand samples illustrated in Fig. 10 are those with the highest PAH concentrations (i.e. all the PAH quantified). PAH-L was excluded, as these PAH are mainly emitted into the air in the traffic environment (Markiewicz et al., 2017).

When comparing PAH profiles for bitumen and stormwater samples, several differences were observed. Pure bitumen has a high content of benzo(*ghi*)perylene and chrysene, and a low content of anthracene and fluoranthene, whereas phenanthrene and fluoranthene were two

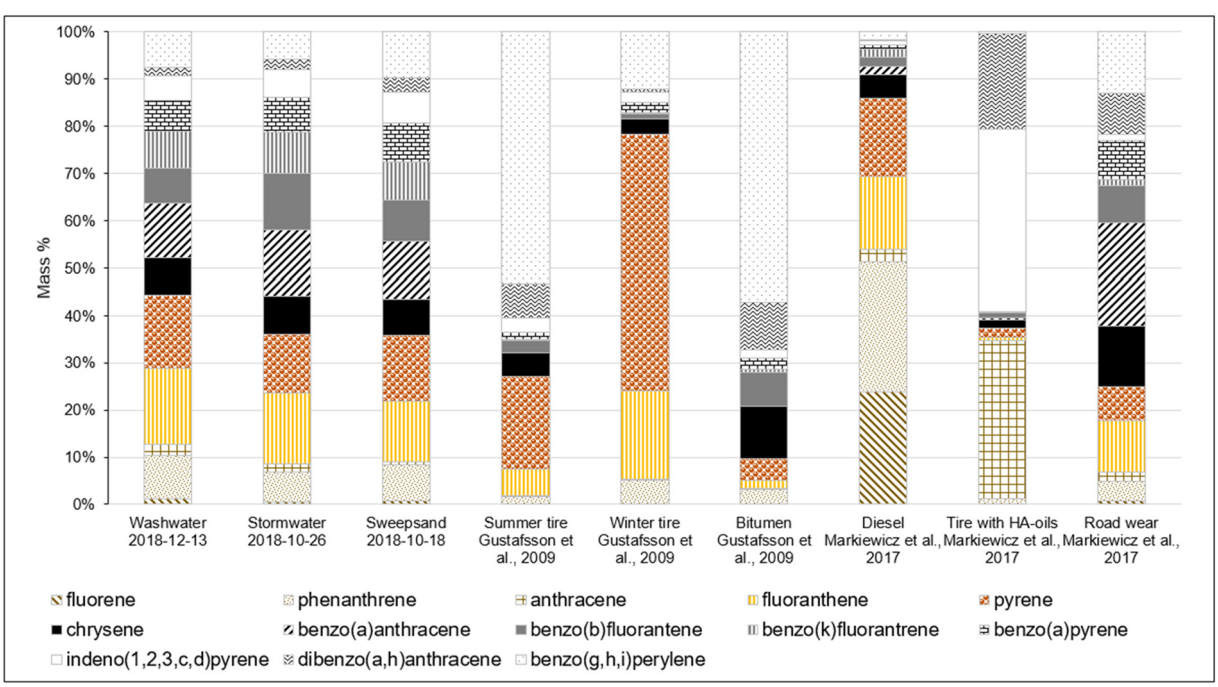

Fig. 10. The relative composition, in mass percentage, of PAH-M and PAH-H in different sample matrices, analyzed tires, and bitumen (Gustafsson et al., 2009), compared to data on diesel, tire, and road surface wear in (Markiewicz et al., 2017).

of the most frequently found PAH in the stormwater. A possible explanation is that the PAH in stormwater are a mix of those present in road and tire wear –winter tires, in particular, contain high amounts of phenanthrene – as well as diesel exhausts, which contain high amounts of fluoranthene (Fig. 10). Phenanthrene and fluoranthene may also originate from other primary sources within the construction area. The lack of correlation between TWP and PAH between seasons can, as discussed in Section 3.4, be explained by road wear being more pronounced in winter due to studded tires being used, and tire wear being more pronounced during the rest of the year. The differences may also be explained by higher temperatures and more evaporation of particularly PAH-M during the non-winter seasons. Benzo(*k*)fluoranthene, which is present in low amounts in diesel, was also found in the stormwater, washwater, and sweepsand. The relative composition on the street and in the stormwater will change over time, however, as the lighter PAH evaporates to a much higher extent than the heavy benzo(*k*)fluoranthene, which accordingly will become relatively more important.

3.6. Screening of organic pollutants

In the non-target screening of organic compounds with expected anthropogenic origin in stormwater, a total of 87 organic compounds were tentatively quantified, with an approximate total EMC of 310 μg/L. This is in the same order of concentrations as in a study of 50 stormwater samples collected at 21 sites across US, which analyzed total concentrations of organic pollutants up to 260 μg/L in the stormwater samples (Masoner et al., 2019). In this study, eight analytical methods were used to determine concentrations of more than 400 hundred organic chemicals, The predominant compounds in this study were branched and straight alkanes, as well as cycloalkanes (Fig. 11), with a combined EMC of 60 μg/L. The alkanes are typical of vehicle exhaust emissions, petrol and diesel spills, and oil residues (Alam et al., 2019; Anh et al., 2019b; Yang et al., 2017), confirming the likelihood that traffic is a major source of the measured pollutants in this study.

Phthalates were also prevalent, with a total EMC of 59 μg/L, which can be compared with the Environmental Quality Standards in the EU Water Framework Directive, which has a guideline value of 1.3 μg/L for DEHP. Unfortunately, the stormwater samples were not analyzed for quantification of phthalates (Table 2), however, the results show that road runoff may be an important source of phthalates in stormwater. Potential emission sources for phthalates include tire wear, PVC, undercoating, and paints, which may all contain phthalates and are released from vehicles to runoff (Björklund et al., 2009; Markiewicz et al., 2017). The abundance of oxygen-containing compounds, dominated by alcohols with a total concentration of 37 μg/L, and ethers with a total concentration of 25 μg/L, is a clear sign of the presence of decomposed hydrocarbons. The specific organic compounds occurring in the highest concentrations were the solvents tetrachloroethylene and toluene, followed by 7-oxabicyclo[4.1.0]heptane, methyl-cyclohexane, and 5-methyl-1-heptanol.

In the sweepsand sample, a total of 99 organic compounds with an expected anthropogenic origin corresponded to an approximate total concentration of 36,000 μg/kg. As was the case for stormwater, branched and straight aliphatic compounds, in concentrations of up to 9400 μg/kg, and phthalates, in concentrations of up to 6700 μg/kg, were predominant in the sweepsand. The corresponding sweepsand sample (2018-10-18, see Table S4), were also analyzed for the same thirteen phthalates, however, all the specified compounds were below the quantification limit. These results show that the methods for analyzing phthalates should be further developed and made much more sensitive. There is also a need for the analytical methods to cover a greater number of phthalate isomers, to enable an analysis of all phthalates that are released in traffic and road environments. Siloxanes and sulfurous acids were abundant, with a total concentration of 4500 μg/kg dw, indicating a new group of emerging pollutants to investigate incoming studies, as methyl siloxanes may have environmental impacts due to their persistence and bioaccumulation potential (Wang et al., 2013). The abundance of oxygen-containing compounds, dominated by ketones with a total concentration of 3400 μg/kg dw, and acids with a total concentration of 3300 μg/kg, is a clear sign of the presence of decomposed hydrocarbons. Several different groups of nitrogen-containing organic compounds, including pyrrolidinones, pyrazoles, triazines, and amines were also present in the sweepsand, in the concentration 2400 μg/kg dw. The specific organic compounds occurring in the highest concentrations were: DEHP, 3-methyl-5-propyl-nonane, didecan-2-yl phthalate, 2,6,11-trimethyl-dodecane, and tridecanoic acids.

In the two washwater samples, 106 and 123 organic compounds with expected anthropogenic origins were found, with a total concentration of approximately 1500 μg/L and 78,000 μg/L, respectively. The extremely high concentrations in one of the samples (2018-12-13, Table S3) were due to a sampling error, and only the relative concentration of the compounds should be taken into consideration. Like the stormwater and sweepsand samples, the predominant substances in the washwater samples were alkanes (Fig. 11) with a concentration of 710 μg/L. Although the proportion of phthalates was lower in these samples, the concentration was still 99 μg/L, which is nearly 100 times higher than the guideline value for DEHP (see above). Relatively high concentrations of siloxanes and sulfurous acids were also found in the washwater, with a total of 160 μg/L. Most of the compounds detected in the washwater contained hydroxide and oxide groups (but no double bonds), which makes them water-soluble and may indicate oxidation of organic compounds. Many aromatic compounds were also present, including both PAH and branched monoaromatics. The toxic *N*-(1,3-dimethylbutyl)-*N*′-phenyl-p-1,4-benzenediamine (6PPD) was identified in both washwater samples (concentrations of 6.9 μg/L in the sample from 2018-10-18 and 840 μg/L in the sample from 2018-12-13, with a sampling error). Note that the quantification is approximate since compounds in the screening were quantified towards phenanthrene-d_{10} as the quantification standard. 6PPD is used in tire rubber as an antioxidant and was shown to cause acute mortality of adult salmon (*Oncorhynchus kisutch*) in the US due to stormwater exposure (Tian et al., 2021). The median lethal concentration of this substance is as low as 0.8 μg/L. In the US study, 6PPD was measured in runoff from a

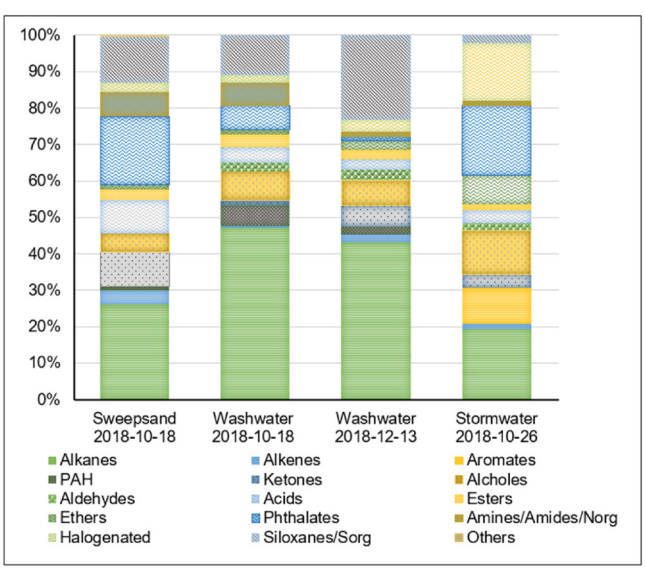

Fig. 11. The relative composition, in mass percentage, of organic pollutant groups quantified in sweepsand (99 specific compounds, total concentration 36,000 μg/kg), street sweeping washwater (106 specific compounds, total concentration 1500 μg/L; 123 specific compounds, total concentration 78,000 μg/L, respectively) and stormwater (87 specific compounds, total concentration 310 μg/L).

multilane highway at concentrations between 0.8 and 19 μg/L. Results from the present indicate alarming concentrations of 6PPD in washwater from the sweeper.

The specific organic compounds occurring in the highest concentrations were: nonadecane, 3,8-dimethyl-undecane, di(2-propylpentyl) ester phthalic acid, 3-methyl-5-propyl-nonane, 6-methyl-tridecane and heneicosane, 1-octadecanethiol, n-tridecan-1-ol, nonadecane, and tetracosane.

4. Conclusions

This study confirms the hypothesis that TWP, OP and metals are released in large quantities to the environment within the studied reconstruction area. From the contaminant composition, it is indicated that heavy vehicles are the most important source of the contaminants, but also that the ongoing construction could contribute to the contaminant load in the area. Extensive infrastructure projects are underway in many cities in Scandinavia, and for that reason, it is very important to consider the increased load of pollutants on streets and in stormwater when planning reconstruction in urban areas.

The concentrations of metals and PAH in stormwater, sweepsand and washwater were well above the local environmental quality guidelines. The number of TWP quantified in investigated media confirms that traffic is an important source of pollutants. Further, the non-target screening of organic compounds in stormwater, washwater, and sweepsand showed that approximately 100, mainly traffic-related, OP occurred. Among the compounds found in washwater, was the highly toxic *N*-(1,3-dimethylbutyl)-*N*′-phenyl-1,4-benzenediamine (6PPD) originating from tires. These results indicate that measures to reduce the spreading of contaminants are needed and that stormwater should be treated before discharge to receiving waters. Measures should also be taken to decrease the emissions of road-related particulate matter to the environment. This study has shown that street sweeping is an effective measure for reducing the load of TWP, metals, and PAH on the road surface, and for preventing these pollutants from reaching stormwater. Still, sources and dispersal need to be better understood to enable the most effective measures and maintenance of pipes and other maintenance measures.

The results indicate that larger amounts of TWP are generated during summer than during winter. These results are important to take into consideration in the environmental classification of rubber tires, but also in the development of new qualities of rubber used for tires. Studded tires cause air pollution issues due to increased asphalt wear, and summer tires cause stormwater pollution because the rubber wears more easily. The reasons for the relatively low TWP concentrations found in stormwater compared to theoretically estimated emissions need to be better understood by further field, laboratory, and modeling studies. A high positive correlation between TWP, metals, and PAH was found when the stormwater samples were divided into summer and winter seasons. The results showed that most of the TWP occurred in the smallest particle fraction 20–100 μm. It is assumed that there are relatively large amounts of TWP in the fraction <20 μm, hence developing analytical methods to measure even smaller particles is necessary.

CRediT authorship contribution statement

Ida Järlskog: Writing – original draft, Writing – review & editing, Resources, Visualization. **Ann-Margret Strömvall:** Supervision, Writing – original draft, Writing – review & editing, Conceptualization. **Kerstin Magnusson:** Writing – original draft, Writing – review & editing, Resources, Investigation, Validation. **Helén Galfi:** Conceptualization, Funding acquisition, Writing – review & editing. **Karin Björklund:** Writing – original draft, Writing – review & editing, Validation. **Maria Polukarova:** Writing – review & editing, Resources. **Rita Garção:** Conceptualization, Resources, Writing – review & editing. **Anna Markiewicz:** Writing – review & editing, Resources, Visualization. **Maria Aronsson:** Conceptualization, Funding acquisition, Writing – review & editing. **Mats Gustafsson:** Writing – review & editing, Supervision. **Malin Norin:** Conceptualization, Resources, Writing – review & editing. **Lena Blom:** Conceptualization, Resources, Writing – review & editing. **Yvonne Andersson-Sköld:** Supervision, Writing – original draft, Writing – review & editing.

Declaration of competing interest

We wish to confirm that there are no known conflicts of interest associated with this publication and there has been no financial support for this work that could have influenced its outcome.

We confirm that the manuscript has been read and approved by all named authors and that there are no other persons who satisfied the criteria for authorship but are not listed. We further confirm that the order of authors listed in the manuscript has been approved by all of us.

We confirm that we have given due consideration to the protection of intellectual property associated with this work and that there are no impediments to publication, including the timing of publication, with respect to intellectual property. In so doing we confirm that we have followed the regulations of our institutions concerning intellectual property.

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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.2021.145503.

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