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Occurrence of tire and bitumen wear microplastics on urban streets and in sweepsand and washwater

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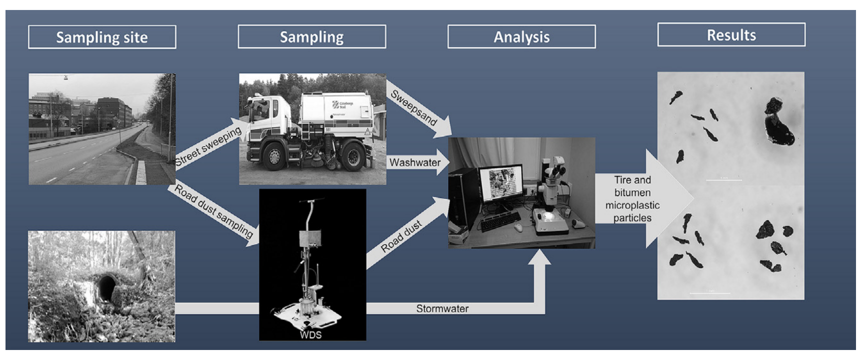
HIGHLIGHTS

- Traffic related microplastics is one of the major sources to microplastic emissions
- Samples were collected at the road surface, in the stormwater and from a sweeper
- Microplastics have been analysed and quantified with light microscopy
- The majority of identified microplastics in all media consist of tire and bitumen
- The measured emissions seem to be we`ll in line with theoretical values

GRAPHICAL ABSTRACT



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ABSTRACT

Tire and road wear particles have been identified as a potential major source of microplastics in the environment. However, more knowledge of the emissions and their further fate in the environment is needed, and the effectiveness and benefits of potential measures must be investigated to support future risk management efforts. Here the concentrations of tire and bitumen microplastic particles (TBMP) on roads and in nearby in stormwater, sweepsand and washwater were measured for the first time within the same area and time period. The analysis also included plastic, paint and fiber particles. Road dust was sampled on the road surface using a wet dust sampler, before and after street sweeping on two occasions. On each of these occasions, and several occasions during a four-month period with frequent street sweeping, sweepsand and washwater, as well as flow-weighted sampling of stormwater, were collected. TBMP concentrations were operationally defined, using density separation for some samples, followed by analysis by stereo microscopy. Sodium iodide (NaI) was found to be effective for density separation of TBMP. The largest proportion of anthropogenic microplastics detected consisted of tire tread wear and bitumen. The number of TBMP ≥100 μm in the WDS samples was up to 2561 particles/L. Sweepsand and washwater contained high amounts of TBMP ≥100 μm, up to 2170 particles/kg dw and 4500 particles/L, respectively. The results show that the sweeper collects considerable amounts of TBMP, and thus weekly sweeping might prevent further transport of TBMP to the receiving stormwater. In stormwater the number of

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particles ≥100 μm was up to 3 particles/L and ≥ 20 μm was up to 5900 particles/L showing the importance of analysing smaller microparticle sizes than 100 μm in all samples in future studies. This study also confirms that there is a substantial volume of TBMP generated from traffic that enters the environment.

1. Introduction

Increased concentrations of plastic in oceans, lakes and watercourses is a growing environmental problem that has been recognized in the past decade (Auta et al., 2017; Barboza and Gimenez, 2015; Jiang, 2018). Microplastics are generally defined as particles in the size range 1–5000 μm, composed of synthetic polymers with thermoplastic or thermo-set properties, elastomers (e.g. styrene-butadiene rubber, SBR), and polymer modified bitumen (Andrady, 2011; GESAMP, 2019; Verschoor et al., 2016). Road wear particles from asphalt with bitumen, a viscoelastic binding agent that consist of highly heterogenous mixtures of hydrocarbons, are not explicitly mentioned in the definition. However, for practical reasons bitumen has often been included when traffic related particles are studied. Recent studies indicate that tire and road wear particles (TRWP), may be one of the major sources of microplastics in the environment (Kole et al., 2017; Sommer et al., 2018). For example, in Sweden alone, 8700 tons of TRWP are theoretically released into the environment every year (Magnusson et al., 2016; Sundt et al., 2014). Based on a compilation of results from existing models, Sommer et al. (2018) suggested that 30 vol% of all microplastics that pollute rivers and oceans relate to tire wear. Another study from Panko et al. (2013) indicated that only 0.84% of the total amount of PM10 (i.e. airborne particulate matter with an aerodynamic diameter < 10 μm) is TRWP, which suggests that almost all TRWP (>99%) remain on the ground. TRWP that end up in waterways may be transported large distances from their source, and Kole et al. (2017) have estimated that 5–10% of all plastics in the oceans originate from TRWP.

Tires consist of 60% styrene butadiene rubber (SBR), which is combined with different additives and natural rubber (Sommer et al., 2018; Sundt et al., 2014). Analyses using scanning microscopy have revealed that traffic generated tire wear contain incrustations of minerals from the road surface (Kreider et al., 2010). Several metals, e.g. aluminium, iron, magnesium, titanium and silicon, are also found in higher concentrations in traffic generated tire wear than in unused tires. Particles from tire wear have been detected in the size range from 10 nm to several 100 μm (Kole et al., 2017). The variation in wear sizes may depend partly on the wear process (e.g. different driving situations affecting the tire surface, temperature, meteorological and road surface properties), and partly on different tire compositions. It may also be a result of differences in sampling procedures, sample preparation, analytical methods, and sampled particle sizes. In addition, tire and road wear particles are especially hard to analyse, as filler materials, such as carbon black, in particles from tires cause disruptive fluorescence phenomena upon irradiation, resulting in almost complete absorption of IR light (Eisentraut et al., 2018; Wagner et al., 2018). Although the estimated emissions of TRWP are very large (e.g. (Magnusson et al., 2016; Sundt et al., 2014)), there is still limited information on the quantities present in the environment. Furthermore, only a few environmental samples have been analysed for TRWP, because the analysis methods are complicated, costly and time consuming, which makes it difficult to compare and draw conclusions regarding sources and transportation mechanisms, as well as to carry out risk assessments of TRWP emitted from roads.

TRWP and other forms of plastic debris in the marine environment originate from several sources and are transported to the oceans by wind, stormwater, road runoff, snow dumping, wastewater treatment plants, and mismanaged waste (Andrady, 2011). Only a few percentages of the stormwater volume in Sweden is treated in wastewater treatment plants or in local treatment facilities as for example stormwater ponds. Investigations in seven stormwater ponds in Denmark show that they act as pollution hotspots for microplastics, and they are important for the transport of microplastics from land to the aquatic environment (Liu et al., 2019). There are only a very few studies where TRWP have been quantified in environmental samples such as road runoff Eisentraut et al. (2018), sediments in streams Unice et al. (2013) and road dust (Sommer et al., 2018; Unice et al., 2013). Most previous studies have used Zn concentration as a marker for TRWP (Wagner et al., 2018). Among the latest ones is Klöckner et al. (2019) who estimated road runoff to contain 0.38 to 150 mg TRWP/g.

Previous studies indicate that the main part of the TRWP deposits adjacent to the kerb and/or close to the road settles in the soil nearby (Wagner et al., 2018). Modelling of further spreading of tire wear from traffic indicates that around 50% of the TRWP mass fraction can be expected to remain in the roadside soil, while the rest is transported away from the road by runoff (Hann et al., 2018; Unice et al., 2019a). Depending on how much of the urban runoff is treated in waste water treatment plants, and the plant effectiveness, previous modelling studies indicate that 1–25% of the total tire wear emitted from roads reaches nearby watercourses (Hann et al., 2018; Unice et al., 2019a; Unice et al., 2019b; Wagner et al., 2018). Simulations on TRWP in receiving waters by hydrodynamic modelling show that peak concentrations of TRWP have a short duration but elevated concentrations may be present for hours after rainfall, and there is a high risk that high loads of TRWP from the city will reach the marine environment (Bondelind et al., 2019). There are large variations in the current estimates of further transportation as several properties, in particular size and density (Unice et al., 2019a; Unice et al., 2019b), in addition to the strengths and locations of the sources and the occurrence and effectiveness of stormwater treatment facilities, that must be known to enable us to understand and estimate how TRWP and other microplastics are transported in the environment. However, only a few percent of the stormwater generated in our urban environment is treated, and the treatment systems available today are not designed to effectively remove pollutants as TRWP.

Despite the incomplete knowledge, there have been calls to reduce the recipient loads of microplastics from all sources (e.g. European-Commission (2019)). Reduced amounts of traffic and decreased speed limits are measures that could be used to reduce the release of TRWP (Verschoor and Valk, 2018). Since the use of rubber tires would not discontinue, it is of interest to identify and evaluate other potential measures. Since the 1980s, street sweeping has been used to reduce the contaminants from roads and highways (e.g. (U.S.EPA, 1983)). Modelling studies investigated the efficiency of various brushes (Vanegas-Useche et al., 2018; Vanegas-Useche et al., 2019) and field studies have investigated if street sweeping can reduce the amounts of inhalable particles (Bogacki et al., 2018; Järlskog et al., 2017; Norman and Johansson, 2006; Snilsberg and Gryteselv, 2017; Snilsberg et al., 2017). Amato et al. (2010) stated that most studies have focused on sweeping as a measure to reduce the transportation of particle-associated contaminants (PAH and metals) and fine particles to surrounding recipient waters. Polukarova et al. (2020) concluded that sweepers can collect large amounts of particles including nanoparticles and organic pollutants. Also, recent compilations indicate that street sweeping may be relevant measure to reduce the spreading of microplastics from urban streets (Hann et al., 2018; Vogelsang et al., 2018),however, there are no field studies available on the efficiency and uptake of microplastics by street sweepers.

This paper describes a case study where the occurrence of microplastics, focusing on tire and bitumen microplastic particles (TBMP), on road surfaces and in nearby stormwater samples have been analysed. The uptake of TBMP by street sweeping was estimated at the same location, by analysing sand and washwater from the sweeper. Also, the TBMP on the road was measured before and after street sweeping by a combination of visual and tactile identification, using stereomicroscopy. This is the first study where several types of microplastics (i.e. plastics, fibres, paints, bitumen and rubber particles) are analysed in samples from the same location in four different medias (i.e. sweepsand, washwater, stormwater and WDS). The study was also performed during different seasons providing samples of both winter and summer tires. The aim of the study was to: 1) investigate the concentrations of microplastics/TBMP on road surfaces in an urban area, and in stormwater collected from the catchment area during a period with weekly street sweeping; 2) estimate the potential contribution of micro-sized tire wear from an urban street area to water recipients (nearest watercourse and estuary); and 3) to investigate the uptake of TBMP by street sweeping.

2. Method

This study is a case study of TBMP samples collected during the period from 2017 to 09-17 to 2018-04-19 in the central parts of Gothenburg, Sweden. Gothenburg, the second largest city in Sweden, with 600,000 inhabitants (1,000,000 in the larger urban area) and is located at the mouth of the river Göta älv, which feeds into the Kattegat, an arm of the North Sea, Fig. 1.

The study catchment area of Vitsippsbäcken is located in the outer part of the city centre of Gothenburg (Fig. 1), and covers approximately 55 ha and consists of vegetation (60%), paved surfaces (17%), buildings (13%) and roads (10%). The annual average daily traffic (AADT) on the streets within the catchment area is approximately 5500–13,000 vehicles/day. The AADT at the WDS sampling street, Ehrenströmsgatan, is 11,500 vehicles/day and the road width are approximately 7 m.

The length of the sweeping route was approximately 5 km. The climate is mild with an average annual precipitation of 776 mm. The data applied in the study is open source data that relates to the weather station "Gothenburg A 71420" (SMHI, 2019). The wind speed was generally low during the measurement period, with a mean value of 2.7 m/s (max: 12 m/s and min: 0 m/s), which may indicate that the amount of TRWP transported by wind was relatively small. The wind speed was calculated as the mean value of 24 measurements/day and the amount of rain was measured once a day.

In the case study area, the stormwater sewer system is separate from the wastewater pipes, which means that all runoff from impervious surfaces in the area drains, without treatment, into a small creek Vitsippsbäcken. The creek is classified as a sensitive water recipient with a high pollution load of Cu, Zn, Cr and Co, due to traffic, and corroded metal roofs in the catchment. It discharges through a natural reserve and the Botanical Gardens of Gothenburg, into the river Göta älv (Fig. 1), the main watercourse of the city.

2.1. Street sweeping

Street sweeping was performed on the roads Ehrenströmsgatan, Guldhedsgatan, Doktor Allards gata and Medicinaregatan (Fig. 1). Street sweeping of paved roads within the catchment area of Vitsippsbäcken is normally performed once a month between May and November. In this study, the sweeping was more frequent. The streets were swept once a week in the autumn, between the 2017-08-17 and the 2017-12-07, with a break in November due to the autumn defoliation.

The last sweeping event in 2017 was performed to investigate the contribution of studded tires to the tire and road wear process. The sweeping was paused during the winter (December–March), due to sub-zero temperatures, snow, and other types of operations, such as ploughing, salting, and sanding. The sweeping was resumed in the spring when it was carried out twice; on 2018-03-26 and 2018-04-19. The machine used in the study was a Johnston Beam VTJB651 (see Fig. S3). The brooms were made of steel, so as not to contaminate the road surface with wear from plastic brooms.

2.2. Sampling procedures

Four different types of samples were collected: road dust from the road surface, stormwater, and two materials from the sweeper, hereafter called sweepsand and washwater.

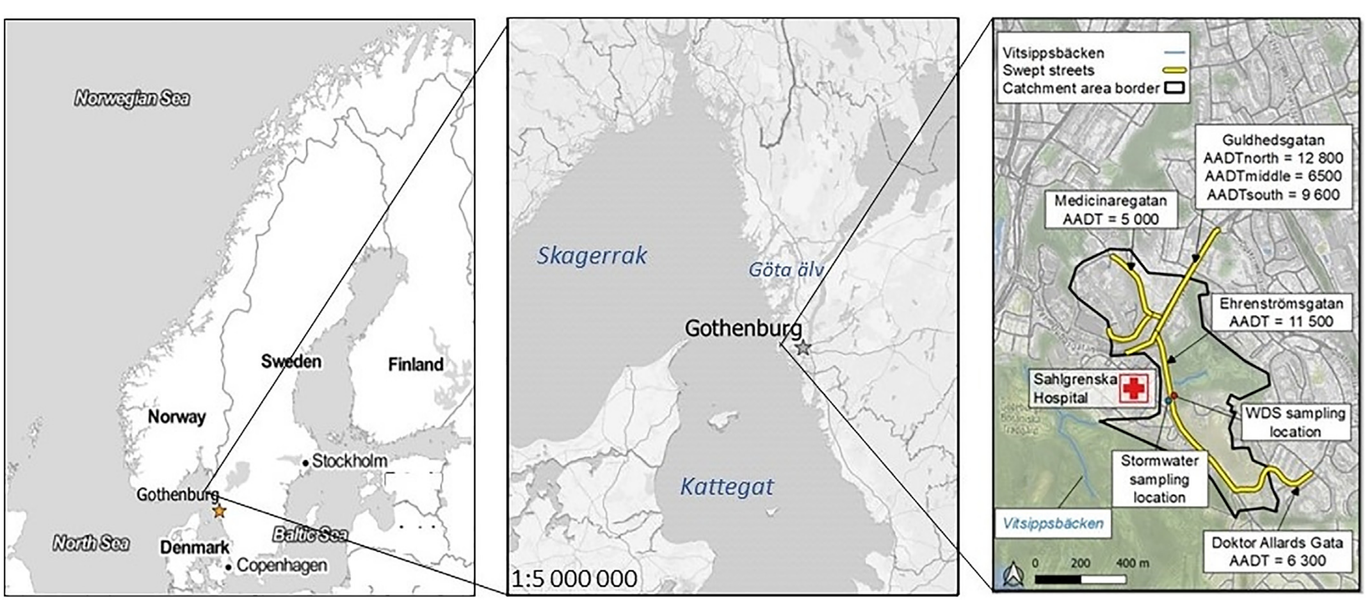

Fig. 1. The location of Gothenburg (left panel), Kattegat and the basin of the river Göta älv (centre panel) and the studied catchment area (right panel) with the swept streets (yellow) and the locations where WDS and stormwater samples were collected (arrows). Map tiles to the right by Stamen Design, under, CC BY 3.0. Data by OpenStreetMap, under, ODbL. 2.1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.2.1. Road surface

Samples from the road surface were collected with a Wet Dust Sampler II (WDSII), Fig. 2, (Gustafsson et al., 2019; Jonsson et al., 2008; Lundberg et al., 2019). In an automated sampling procedure, a known amount of de-ionised water (340 mL) was used to clean an area of 20.43 cm^2 with high pressurized water, and the water-road dust sample was transferred to a sample bottle using compressed air. A single sampling procedure will hereafter be referred to as a "shot". Road dust samples were collected before and after street sweeping of Ehrenströmsgatan twice during the spring of 2018. The first sampling occasion took place on the night between 26 and 27 March. At this time, no street sweeping had been performed since the late autumn, the pavement was soiled, and 50% of all cars were estimated to be fitted with studded tires, in line with a previous investigation by the Swedish Transport Administration. The temperature was approximately 0 °C and the pavement was moist. A light snowfall started when the sampling was finished. The second sampling took place during the night between 18 and 19 April. On this occasion the temperature was around 11 °C, the pavement was dry and looked much cleaner, and almost all cars had switched to summer tires.

Samples to be used for particle size distribution, determination of road dust load and loss of ignition were collected. Three bottles were collected from the left wheel track (WT1–3) and between the wheel tracks (BW1–3) each. One bottle was collected adjacent to the kerb (K1). Samples for microplastic analysis were collected in a glass bottle adjacent to the kerb (MP). The WDS was moved slightly between each shot to make sure that the surface was unsampled (Fig. 2). The same number of samples were collected before and after street sweeping.

2.2.2. Sweepsand and washwater

Sweepsand for microplastic analysis was collected from the sweeper on ten occasions during the investigation period (Table 1). On four occasions, washwater samples were collected for microplastic analysis; 2017-08-17 (no studded tires in use), 2017-12-07 (studded tires in use), 2018-03-26 (studded tires in use and the date of the first WDS sampling), and 2018-04-19 (almost no studded tires in use and the date of the second WDS sampling) (Table 1).

After each sweeping event, the sweeping machine was driven to a local site for temporal deposition of the collected sweepsand (~1 m^3) and washwater (~1 m^3). The washwater was made up of rainwater/road runoff sucked up by the machine during sweeping (wet weather), water used by the sweeper to suspend dust during sweeping (dry weather), and road dust that did not settle inside the sweeper. The washwater was systematically collected in 10 L glass bottles to receive a representative pooled sample of the study area, and kept at 4 °C before being sent for microplastic analysis. The sweepsand was collected once the masses had been emptied from the machine onto the ground, and the samples were randomly taken directly from the sand heap using a metal shovel. The sweepsand was collected in small portions from different places in the pile of sand and stored in stainless steel containers, from which small sub samples were taken randomly, before being transferred into small glass containers and kept frozen until microplastic analysis.

Table 1
Samples collected for TBMP analysis during the weekly sweeping campaign and after the winter break.

Sampling date	WDS samples[a]	Washwater[c]	Sweepsand[b]	Stormwater[d]
2017-08-17		X	X	
2017-08-24			X	
2017-08-31			X	
2017-09-07			X	X
2017-09-11				X
2017-09-14			X	
2017-09-28			X	
2017-10-12			X	
2017-10-25				X
2017-11-23				X
2017-11-28				X
2017-12-07		X	X	
2018-03-26	X	X	X	
2018-04-19	X	X	X	

[a] Mean values of 14 shots.
[b] Representative samples from ~1 m^3 collected sand.
[c] Representative samples from ~1m^3 collected washwater.
[d] Event mean concentration of a whole rain event.

2.2.3. Stormwater sampling

During the period with weekly street sweeping (Table 1), stormwater was collected during five rain events: 2017-09-07, 2017-09-11, 2017-10-25, 2017-11-23 and 2017-11-28 (Fig. 3). The sampling was carried out using a flow-weighted automatic ISCO sampler, installed upstream of Vitsippsbäcken, in the stormwater well (see Fig. 1) where stormwater from the entire catchment area is collected prior to discharge. The sampling volume collected for analysis of microplastics at the rain events varied between 2.7 and 9 L.

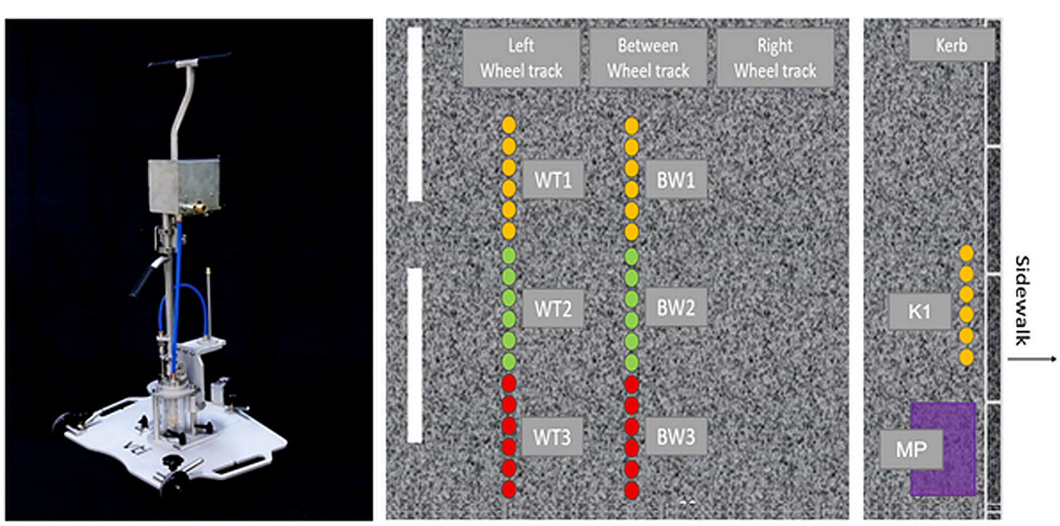

Fig. 2. left: WDS II (Photo Mats Gustafsson, VTI). On the right: Schematic figure over WDS sampling procedure for size distribution, road dust load and loss of ignition (WT = wheel track; BW = between the wheel track; K = adjacent to the kerb; MP = samples for microplastics adjacent to the kerb).

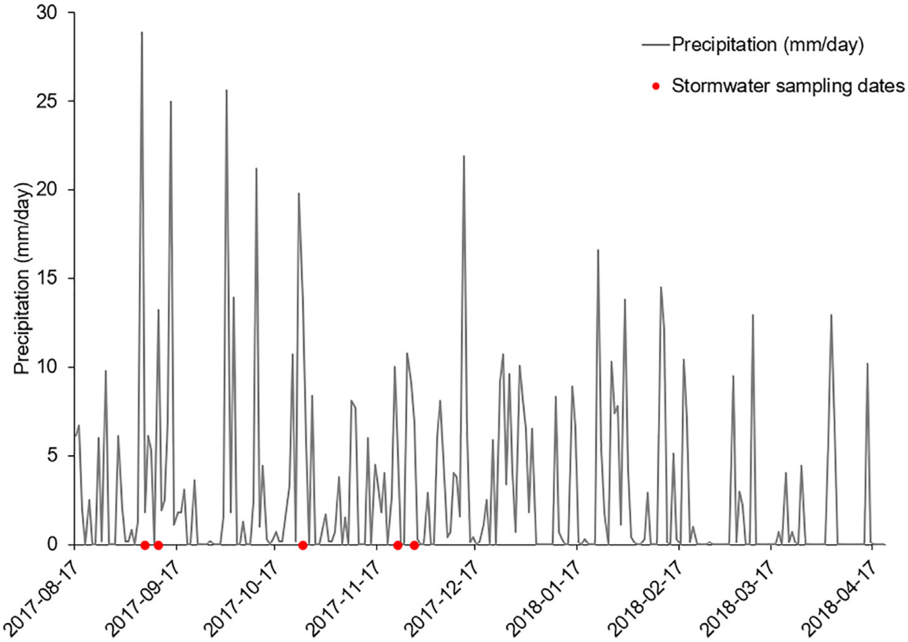

Fig. 3. Precipitation and stormwater sampling occasions. The red dots show the days when stormwater sampling was performed. Data collected from SMHI's open source data base (SMHI, 2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.3. Sample analyses

All water samples collected were stored in glass bottles in fridges to avoid organic growth, and the sweepsand samples were stored in glass containers in the freezer, until the day of analysis.

2.3.1. Size distribution, organic content and road dust

To identify the most common particle sizes on the road, a size distribution analysis on the WDS samples was performed. Two WDS samples before and two after sweeping (2018-03-26 and 2018-04-19) were analysed; one taken at the kerb (K1), and one taken between the wheel tracks (BW2), see Fig. 2. The samples used for size distribution analysis were sieved with a 2 mm sieve prior to the analysis, due to the upper limitation of the laser granulometer. The size distributions of the particles in the WDS samples were determined using laser granulometry (Mastersizer 3000 from Malvern Panalytical), operating on the principle of laser diffraction. For these measurements the Mie scattering model was used. The refraction index used was based on earlier measurement experiences from the research group e.g. (Gustafsson et al., 2019) and engineering judgement. Each laser granulometry measurement was done in triplicate. The analysis was done by summarising the mass of particles up to 2000 μm (corresponding to DL2000). The uncertainty is high for the largest particles due to the low number of large particles in each sample.

The amount of road dust in the WDS samples was measured on sieved samples (mesh size 180 μm) and is presented as DL180 (Dust load <180 μm). The mass-based organic content in the WDS samples (BW1–3, W1–3 and K1) was measured as loss of ignition, in accordance with the standard procedure described by Gustafsson et al. (2019).

2.3.2. Microplastic analysis – sample preparation and analysis using microscopy

Prior to analysis, the microplastics and TBMP in the WDS, sweepsand and washwater samples had to be separated from other particulate matter in the samples by density separation. The samples were mixed with a saturated saline solution and floating particles (particles with a density lower than that of the saline solution) were collected and analysed. All sweepsand samples from 2017 were density separated using a saturated sodium chloride (NaCl) solution with a density of 1.2 g/cm, considered to be dense enough to separate out most TBMP and microplastic particles. Unused tire tread is reported to have a density of 0.8–1.2 g/cm^3 (Kreider et al., 2010; Rhodes et al., 2012; Sofi, 2018). The density of bitumen from the pavement is reported to be 0.9–1.1 g/cm^3 (Nynas, 2017). However, sand particles have a density of ~2.7 g / cm^3 and therefore sand/mineral encrustations in the TBMP can increase the apparent density of these particles (Klöckner et al., 2019). Therefore, to improve the method, sweepsand samples from 2018 to 03-26 and 2018-04-19 were also density separated with a denser solution of saturated sodium iodide (NaI), with a density > 1.8 g/cm^3. No oxidation of any of the samples was needed as they contained only small amounts of organic matter.

Density separation of TBMP and other microplastics in the sweepsand samples was performed in a stainless steel separation tower (approx. 1 m high) slightly modified from a construction by Imhof et al. (2012). Sweepsand samples of 100–500 g wet weight were thoroughly mixed with the saline solution. The slurry was slowly added from the top of the tower while a rotor was turning in the bottom for six hours, after which the rotor was turned off and particles allowed to settle to the bottom or float to the surface, depending on their density. When the saline solution was clear, the top part with floating particles was closed off from the bottom with a ball valve. The solution in the top part was filtrated in sequence over filter with mesh sizes of 300 and 100 μm.

The washwater and WDS samples contained only small amounts of mineral particles, and instead of using the large separation tower a separation funnel made of glass (500 mL) was used. Density separation was carried out with NaI on all washwater and WDS samples, except the washwater samples collected on the 2017-08-17 and 2017-12-07, which were directly analysed without any pre-treatment. Before density separation, the water samples were filtrated (mesh size 20 μm) and the particulate matter on the filters was then rinsed off into the separation funnel, using a saturated solution of NaI. The funnel was shaken vigorously for a few minutes to make sure all particles in the sample came into contact with the saline solution, then left to stand to allow the particles to settle to the bottom or float to the surface depending on their density. The bottom heavier mineral particles were removed by opening a valve at the bottom of the funnel, and the overlying saline

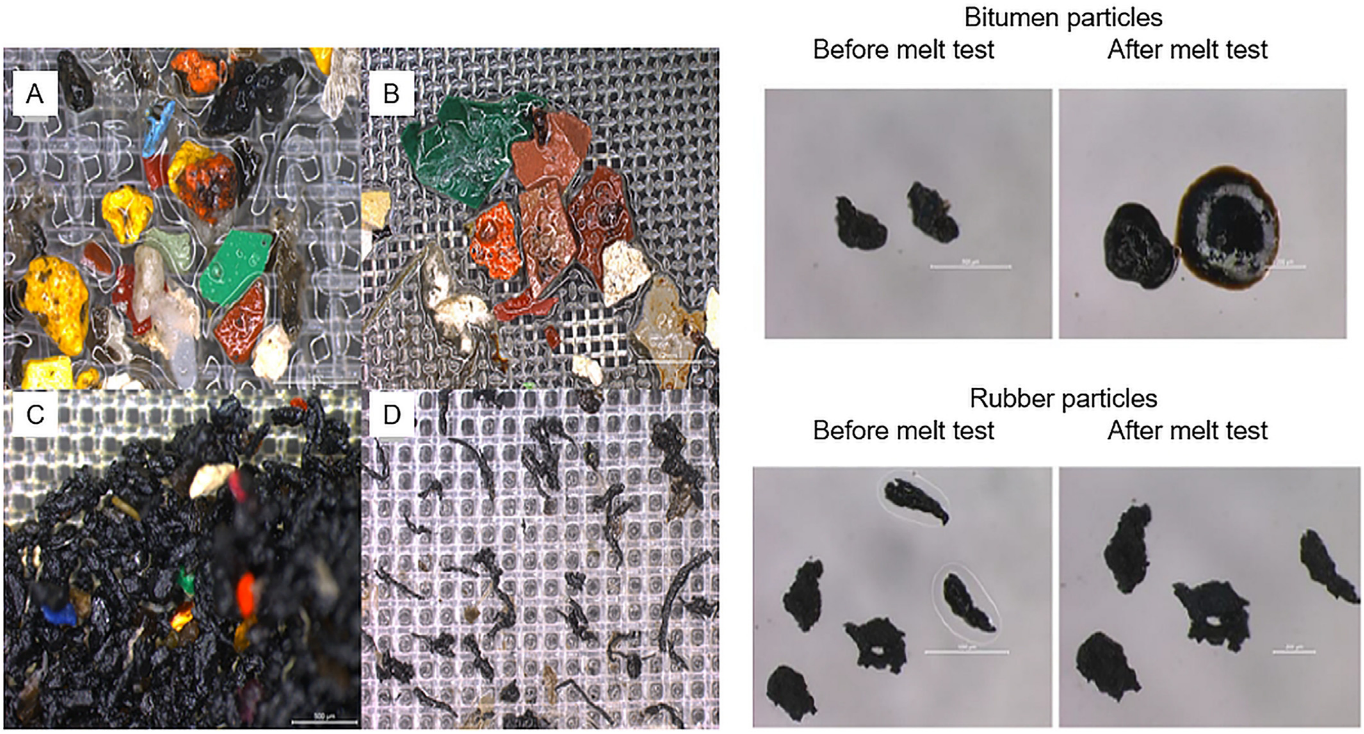

Fig. 4. to the left: Pictures A and B show paint particles from sweepsand. C shows rubber particles from sweepsand, and D shows rubber particles from washwater. Picture top right: bitumen particles before and after heating. Bottom picture: rubber particles before and after heating. Photos by Kerstin Magnusson, IVL Swedish Research Institute.

solution with the floating particles was filtered in sequence over filters with mesh sizes of 300 and 100 μm. All stormwater samples could be analysed without any pre-treatment and were in sequence filtered with mesh sizes of 300, 100, and 20 μm.

The filters were analysed with a stereo microscope (Leica M205 C 80-160×) and the detected plastics were categorised into five morphological particle groups: plastic fibres, plastic fragments, plastic flakes, plastic films, and road particles further identified and classified as tire wear and bitumen (Fig. 4). Classification was carried out through a combination of tactile and visual identification. When touching the particles with a tweezer, it was possible to distinguish the rubber structure from the sticky structure of the tar-like bitumen, as rubber particles are elastic and regain their shape after being squeezed, whereas bitumen particles remain compressed. To further test the reliability of the visual and tactile analysis, melting tests were carried out by placing a selection of analysed particles on an object glass, which was heated from below using an alcohol burner. Rubber particles are morphologically unaffected by heating, whereas bitumen particles melt and expand in size (Fig. 4).

Procedural duplicate blanks for each category were prepared by running the protocols for sweepsand, washwater and stormwater (density separation and filtration) with just saturated NaI or milli-Q water.

A summary of the size ranges of the microplastic analyses conducted and density separation liquids used for the different samples is provided in Table 2.

2.4. Estimated amounts of tire wear on the road surface and in stormwater

To estimate the potential amount of tire wear particles (TWP), or rather tire tread wear particles, emitted from traffic, two approaches are commonly used: 1) the emission factors per vehicle km (vkm)

Table 2
Analysed size ranges of particles and density separation liquid used for each sample type.

	Samples			
	WDS[a]	Sweepsand[b]	Washwater[c]	Stormwater[d]
No. of samples in total	4 (2 before sweeping and 2 after sweeping)	10	4	5
Particle sizes	≥100 μm	≥100 μm	≥100 μm	≥100 μm and ≥ 20 μm
Additional[e]	NaI for all samples	NaCl for all samples. NaI for 2 samples from spring 2018	No density separation for samples from 2017. NaI for samples from 2018	No density separation

[a] Mean values of 14 shots.
[b] Representative samples from ~1 m^3 collected sand.
[c] Representative samples from ~1 m^3 collected washwater.
[d] Event mean concentration of a whole rain event.
[e] Density separation for microplastic analysis was performed using saturated NaCl (1.2 g/cm^3). For sweepsand and washwater samples from 2018 the density was carried out using saturated NaI (>1.8 g/cm^3).

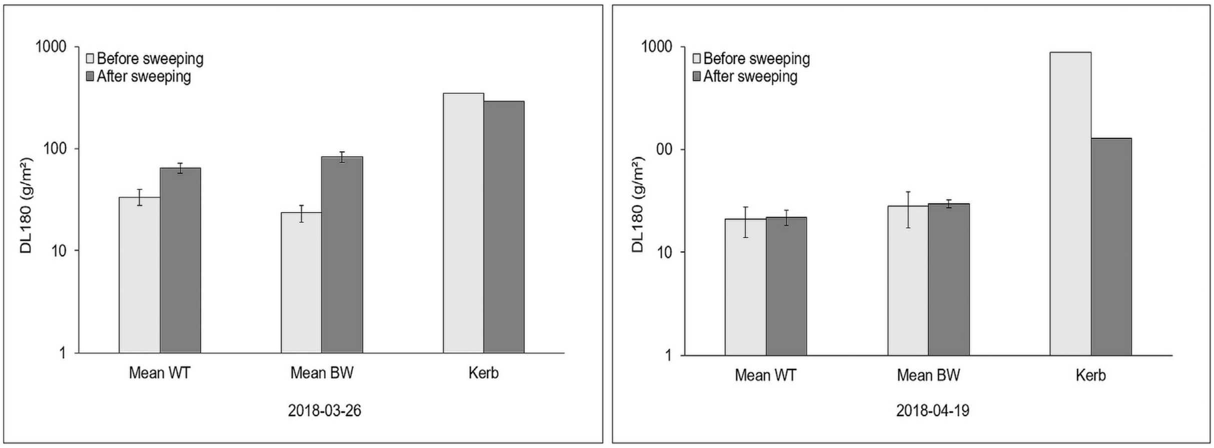

Fig. 5. Road dust load (DL180 (g/m^2) content before and after sweeping.The bars show the mean values with error bars from the bulk triplicate samples taken in wheel tracks (WT) and triplicate between wheel tracks (BW). The kerb samples relate to single samples.

multiplied by the total mileage; and 2) the number of tires multiplied with the weight loss during use. The majority of the tires in Europe are collected and recycled when no longer in use (ETRMA, 2017), which means that the potential wear can be measured and calculated for almost all tire types. AQEG (2019) concluded that there is no standardised, robust, reliable and repeatable method for determination of emission factors. The emission factors vary because of the great variety in the type of rubber, road surface material and driving conditions around the globe and at different times; uncertainties in the measurements also contribute to the variation. An average value for European cars during normal driving conditions is 0.1 g/vkm (Boulter, 2005). Councell et al. (2004) have compiled and compared the wear factors from 35 different studies. The median value was approximately 0.1 g/vkm for passenger cars, and several times higher for heavy duty vehicles. In order to confirm the emission factors and evaluate the results from the measurements carried out in this study, simple calculations of the potential theoretical tire concentrations in the stormwater have been performed and compared to the measured concentrations.

3. Results and discussion

3.1. Road dust load (DL180), particle size distribution, and organic content in samples collected with WDS

The total mass of road dust particles smaller than 180 μm (DL180) collected with WDS (Fig. 5) is of a similar magnitude as the total mass found in earlier studies of several city streets in Stockholm (Gustafsson et al., 2019). The particle size distributions in the WDS samples indicate that most particles measured in the micro-range are found in the size range of 10–100 μm (Fig. 6). Both the total mass (DL180, Fig. 5 and Supplementary Information Table S1) and the number of microparticles (illustrated by the particle size distribution, Fig. 6) between and in the wheel tracks were of similar magnitude, while values were higher adjacent to the kerb than in and between the wheel tracks on both sampling occasions. This could be explained by swirling dust or precipitation moving the particles towards the kerb, where they are caught and more sheltered than on other parts of the road (Gustafsson and Johansson, 2012).

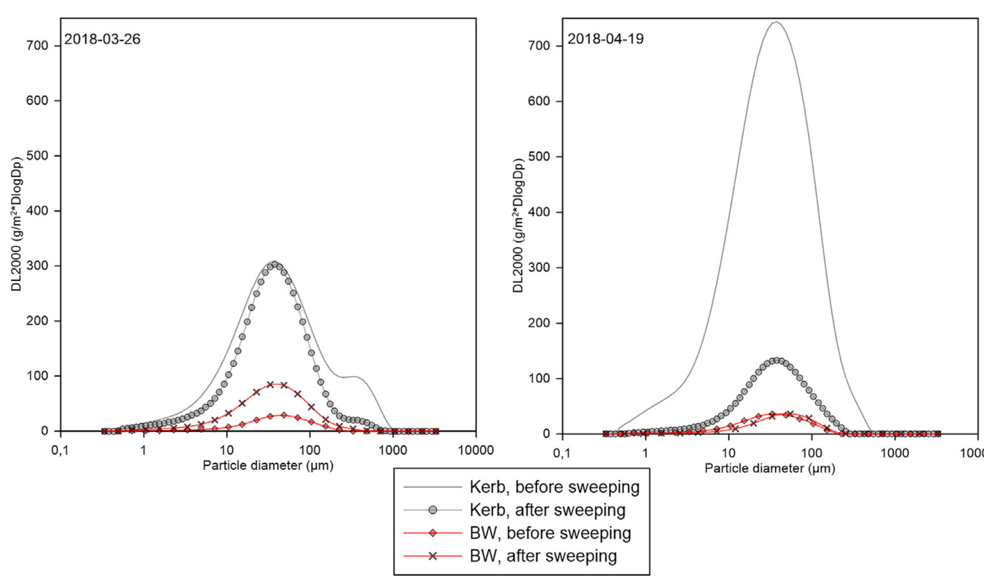

Fig. 6. Size distribution of road dust particles in samples taken adjacent to the kerb (grey lines) and between wheel tracks (red lines) from the sampling on the 2018-03-26 (left) and 2018-04-19 (right). The unit on the y-axis is based on the size distribution presented as the proportion of total particle volume and has been summed for particles up to 2000 μm. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Both the number of particles in the particle size distribution analysis and the DL180 (mass) showed higher concentrations for the 2018-04-19 than for the 2018-03-26 in samples taken adjacent to the kerb (Fig. 5, Fig. 6, and Supplementary Information Table S1). A possible explanation for this is that the less frequent maintenance and snow removal during this period may have resulted in a build-up of road dust on the road between the two WDS sampling occasions. Another explanation may be the meteorological variations between the two occasions, as meteorological factors, like wind and precipitation, are of importance for the observed DL180 concentrations on roads (Egodawatta et al., 2007; Gustafsson et al., 2019; Murakami et al., 2004; Zhao and Li, 2013).

Adjacent to the kerb, both the DL180 (Fig. 5) and the number of particles (Fig. 6) decreased after cleaning on both occasions, whereas both the mass and the number of particles were similar in and between the wheel tracks (2018-04-19), and on one occasion (2018-03-26) even higher than before the sweeping. A possible explanation is that the particle concentration on the street (in and between wheel tracks) was very low on the measurement occasion, i.e. the heterogeneity of the distribution on the street is as high as the total concentration, and therefore the effect of the street sweeping is difficult to assess. Another possible explanation is that the rotating kerb brushes of the sweeper redistributes road dust from the kerb out into the lanes. Similar road dust patterns have been reported before, e.g. in (Gjerstad et al., 2019; Järlskog et al., 2017). Water is often sprayed on the dust in front of the brush which results in a redistribution of dust from the kerb out to the driving lane, where the coarser fractions are sucked up by the sweeper, while finer fractions will be harder to collect due to the added water as previously hypothesized by (Gustafsson et al., 2019). The result can be increased concentrations of fine road dust in the driving lane. Additionally, the roughness and wetness of the pavement (Abdel-Wahab et al., 2011; Snilsberg et al., 2018), the rotation frequency, the penetration and tilt angle, as well as the design of the brush (Vanegas-Useche et al., 2015; Vanegas Useche et al., 2010), may affect pollutant removal by the street sweeper. This study has not evaluated a specific sweeping machine/technique, however, the sweeper used here may not be the most efficient machine for fine particles due to the use of water. However, a possible solution might be to sweep the driving lanes an additional time before sampling (only the driving lanes, not the kerb) to be able to remove the redistributed road dust and to ensure a cleaner street after sweeping.

All the above-mentioned factors, as well as the sampling and analysis methodology, may also influence the size of the particles found in the sweepsand and washwater. For example, a recent study conducted in the same area and using the same sweeper found that the majority of the particles present in the sweepsand, analysed by sieving, were in the size range 0.600–2000 μm, and the majority of the road dust particles in washwater in the nano size, analysed by light scattering, were in the range 100–300 nm in samples filtrated on 0.45 μm (Polukarova et al., 2020). However, previous studies analysing material collected by sweeping or vacuum cleaning have found that the most abundant particle fraction in road dust is approximately equal to 250–500 μm, as reported by Snilsberg and Gryteselv (2016), 550 μm (German, 2003), and between 200 and 400 μm (Lau and Stenstrom, 2005). These results illustrate the importance of developing a standard analysis methodology for characterising the size of evaluated road dust particles.

The estimated organic content was about 3–6 w % of the total mass in and between wheel tracks in all WDS samples, and slightly higher adjacent to the kerb (9–10 w %), see Table S2. The organic content ratio of the WDS samples was found to be independent of sweeping and sweeping frequency, and to be similar on both sampling occasions, indicating similar types of sources in all samples, as also found recently by (Polukarova et al., 2020).

3.2. Microplastics and TBMP

Both tire and bitumen microplastic particles (TBMP) and other microplastics were analysed in all samples, but the focus of this study was TBMP (hereafter TBMP relates to rubber and bitumen, rubber is classified as tire wear particles and henceforth called TWP).

The estimated amounts of plastic, paint and TBMP identified in the microplastic analysis of particles ≥100 μm are presented as averages for all sample occasions in Fig. 7. As can be seen, the number of microplastics ≥100 μm defined as plastic and paint particles (1–100 particles/L) was appreciably lower than the number of TBMP (1000–5000 particles/L) in all samples apart from the microplastics in stormwater 1000–5000 particles/L versus <10 TBMP particles/L. The concentrations of the identified types of microplastics and bitumen in the stormwater were, however, very low (1–15 particles per/L and type). However, it must be considered that the paint, plastic and TBMP particles in the NaCl density separated samples may have been underestimated as some may have encrusted with heavier mineral particles and may therefore been excluded from the analysis after the density separation (Klöckner et al., 2019).

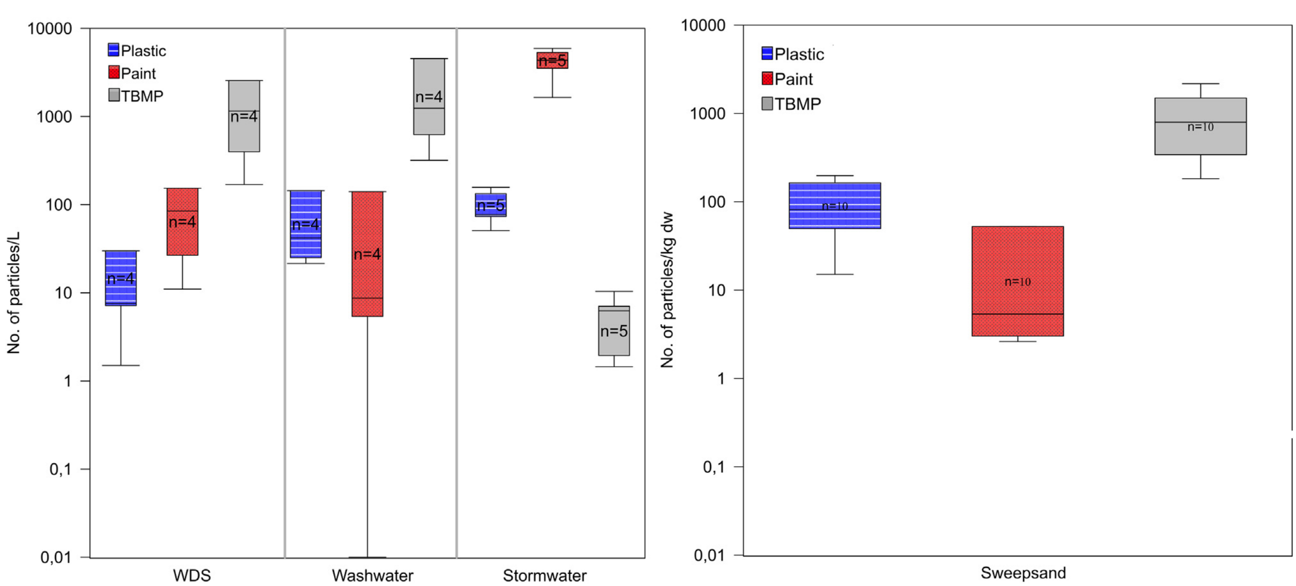

Fig. 7. The different groups of microplastics ≥100 μm identified in each type of sample; were density separated with NaCl (sweepsand, autumn 2017) or NaI (WDS, washwater and sweepsand, spring 2018), washwater (autumn 2017) and all stormwater was analysed without any density separation. NB! the y-axis is logarithmic.

Eisentraut et al. (2018) measured microplastics in road runoff and stormwater sediment and the results showed a dominance of TWP particles, by a factor of 13–49, compared to the number of thermoplastic microplastics, which is well in line with the results from our study. However, the results of this study show the opposite of a study of Abbasi et al. (2019), where road dust samples from 11 cities in Iran were analysed. The authors found that the average road dust concentration of microplastics was approximately four times higher than the concentration of micro rubber (tire wear). The highest plastic concentrations were found in industrial areas, where the ratio between microplastics/rubber was even higher, and varied depending on the location's position in relation to the sources (Abbasi et al., 2019).

3.2.1. WDS samples

The analysis of TBMP in samples collected with the WDS (Fig. 8) differ from the patterns of the DL180 analyses (Fig. 5), because the number of TBMP particles found was more than ten times higher in samples collected on the 2018-03-26 than in those collected on the 2019-04-19. Most of the density separated particles in the WDS-samples, 97% and 92%, respectively, were classified as bitumen before and after street sweeping in the samples from 2018 to 03-26, compared to 46% and 95% before and after street sweeping in the samples from the 2018-04-19. The observed 46% (2018-04-19) consisting of bitumen particles is well in agreement with a previous study (Unice et al., 2012) which found that approximately 50% of TBMP is TWP. The higher amount of bitumen microparticles in the samples from the 2018-03-26 can be explained by the higher road wear caused by studded tires, which are used in Sweden during the winter (Gustafsson et al., 2019). As the microscopic analysis was only performed on particles ≥100 μm, there is a possibility that both the total concentrations and the ratio between rubber and bitumen were different in the smaller size fractions.

The TWP content of the TBMP in the WDS samples was at its highest before the sweeping on the 2019-04-19 (Fig. 8), when the TWP made up 54% of the total number of TBMP particles (compared to 3–8% in the other three samples). The TBMP decreased after both sweeping events (Fig. 8). This was in contrast to the observed number of TWP prior to and after sweeping, which instead followed the same pattern as the DL180 (Fig. 5) and the total number of particles (Fig. 6) found on the road, i.e. increased after the sweeping event on the 2018-03-26, and decreased after the sweeping event on the 2019-04-19. However, the number of TWP, as well as DL180, in the samples from the 2018-03-26 was very low both before and after sweeping. On this occasion, the TBMP mainly contained bitumen, and the TWP behaviour may be a result of a natural heterogeneity. The result agrees with Gustafsson (2001) that road wear mainly occurs, and dominates over tire wear, during the winter months, when studded tires are in use (Nov 1 - March 31 in Sweden). During the rest of the year, tire wear becomes more important in relation to road wear. Further, during the period immediately after the studded tire season, the tire wear may be fairly high, due to the rougher road surfaces caused by the road wear from the studs.

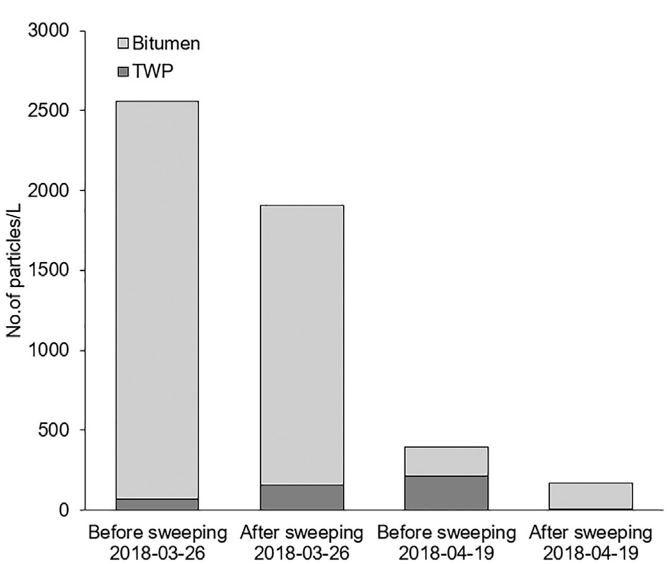

Fig. 8. Concentrations (number/L) of TWP and bitumen microparticles ≥100 μm) in samples collected with the WDS before and after sweeping on the 2018-03-26 and 2018-04-19. (All samples density separated with NaI).

3.2.2. Results sweepsand and washwater

The concentration of TBMP in the sweepsand varied over the measurement period (Fig. 9). However, for the two sweepsand samples collected in 2018, a separation of rubber and bitumen was made. The TWP content found in the NaCl density separated samples from 2017 were 6% and 15% of the total TBMP in the samples collected on the 2018-03-26 and 2018-04-19, respectively (see Supplementary Information Fig. S2). The change in composition can, again, be explained by the use of studded tires during the winter season (2018), and the increased road wear they cause compared to the tires mainly used during other parts of the year, when the majority of the samples from 2017 (7 out of 8) were collected. A decrease in TBMP, and a simultaneous increase in rubber particles, was noted in April when most vehicles had changed from studded tires to summer tires. The lowest concentration of particles was found in August. This can be explained by less traffic during the summer season, smoother road surfaces, and variations in meteorological factors (Egodawatta et al., 2007; Gustafsson et al., 2019; Murakami et al., 2004; Zhao and Li, 2013).

Samples collected in 2018 were first density separated using a saturated NaCl solution (1.2 g/cm^3), however subsamples of the same samples were later reanalysed after being density separated using saturated NaI solution (>1.8 g/cm^3). As shown in Fig. 9, the number of TWP were more than two, and up to nine, times higher in samples where NaI was used instead of NaCl. This is not in agreement with the reported densities for tire wear are 0.8–1.2 g/cm^3, and 0.9–1.1 g/cm^3 for bitumen, but in line with road TWP (Kreider et al., 2010; Nynas, 2017; Rhodes et al., 2012; Sofi, 2018). Sand particles have a density of ~2.7 g / cm^3 and therefore sand/mineral encrustations in the TBMP can increase the apparent density of these particles to ~1.7 or 1.8 g/cm^3 (Klöckner et al., 2019). If field sampled TBMP had the same densities as the reported ones, the saturated NaCl solution would capture all but the heaviest tire particles. However, the results support previous suggestions that these particles incorporate minerogenic and chemical components from other traffic related sources, which increase their density and may therefore reduce the efficiency of density separation, also heavier particles as PET and PVC are not captured in NaCl (Kreider et al., 2010) The results in this study further demonstrate that NaI is more relevant to use than NaCl for density separation of TBMP and other microplastics from roads. Furthermore, the number of TWP is higher in April (no studded tires) than in March (studded tires in use), the pattern was similar in the WDS samples (Fig. 8 above), which is well in line with the hypothesis that the number of TWP increase during the summer.

The concentrations of TBMP in the washwater increased over the measurement period (Fig. 10), by up to 4500 particles/L ≥ 100 μm, which indicate that street sweeping might be an efficient method to reduce the load to and the ambient concentrations in the stormwater, see below. However, different sample preparation methods were applied, 2017 no density separation was done, 2018 NaI solution was used to separate particles. This might have affected the results and more analyses are required.

Approximately 20% of the TBMP in the washwater consisted of rubber in the samples from 2018 to 03-26 and 2018-04-19 (see Supplementary Information Fig. S1). A potential explanation is more intense road management in winter, which leads to higher amounts of coarser and inorganic materials, such as sand and salt.

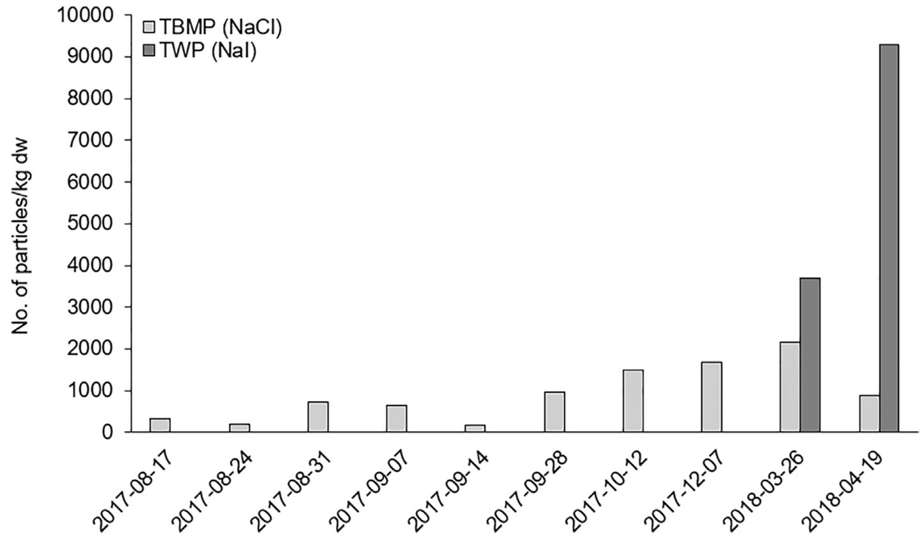

Fig. 9. TBMP and TWP ≥100 μm in samples from sweepsand collected between 2017--08-17 and 2018-04-19. Light bars depict No. of particles/kg dw when separated with NaCl Dark bars depict No. of particles/kg dw when density separated with NaI.

3.2.3. Microplastics in stormwater

Most of the microplastics and TBMP in the stormwater was found in the size range 20–100 μm, see Fig. 11. The smaller fraction (≥20 μm) TBMP are ten to several hundred times higher than for the larger fraction ≥100 μm. These results indicate the importance of measuring a broad particle size spectrum, and to also include particles ≤100 μm, which is currently not always done in microplastic and TBMP analysis. Also, in this study TBMP ≤20 μm are not measured, why their occurrence in the stormwater has not been analysed. In the other medias, TBMP ≤100 μm are not measured at all, which preferably should be done to better understand the occurrence, further fate, and risks associated with microplastics and TBMP. Due to excluding the smaller particles, likely the ambient TBMP number concentrations on roads and the particles' potential transport routes via air, stormwater, and freshwater are underestimated.

The measured stormwater concentrations varied between the measurement occasions, from 1500 to 6000 particles/L for ≥20 μm, and ≤ 1–2 particles/L for ≥100 μm (Fig. 11). The concentrations of ≥100 μm TBMP were in the same range as previously found in stormwater in the San Francisco bay, i.e. 1–30 microplastic particles/L in total, of which approximately half had TWP properties (Sutton et al., 2016). The samples collected on the 2017-09-07 captured stormwater from a 30 mm rainfall after nearly one week of dry weather. The measured TBMP concentration was therefore a result of a one week build-up, followed by well diluted runoff caused by the heavy rain, and measured as the flow-weighted event mean concentration, i.e.1.5 particles/L classified as bitumen and rubber in the size range ≥ 100 μm, and almost 3500 particles in the smaller fraction, ≥20 μm. The highest stormwater event average concentrations (particles ≥20 μm) were found in the samples collected late in November, with >5000 TBMP/L (2017-11-29) and close to 6000 particles/L (2017-11-23). The high concentrations can be explained by a long period without sweeping, no winter maintenance, and low precipitation in the weeks prior to the sampling occasions. The impact of precipitation is also seen in the lowest TBMP microparticle concentrations, observed in samples from 2017 to 10-25 (Fig. 11), which were collected on a day with high precipitation after several rainy days (Fig. 3).

The highest concentrations of TBMP in the collected stormwater were found in the size range 20–100 μm, which indicates that the larger

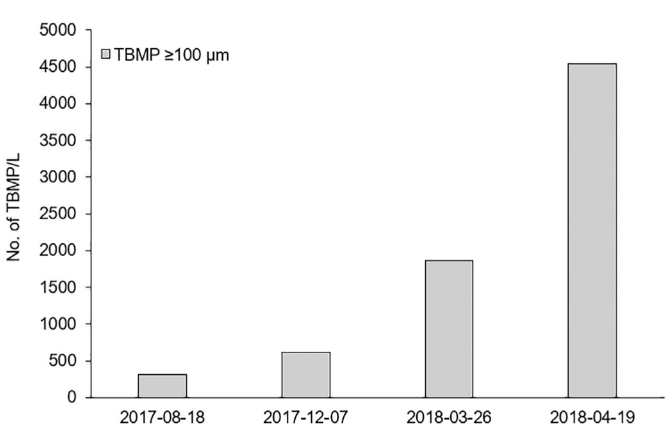

Fig. 10. Concentrations (number/L) of TBMP ≥100 μm in washwater collected between the 2017-08-17 and the 2018-04-19. No distinction between TWP and bitumen was made. (Samples from 2017 was filtered without density separation and samples from 2018 with NaI).

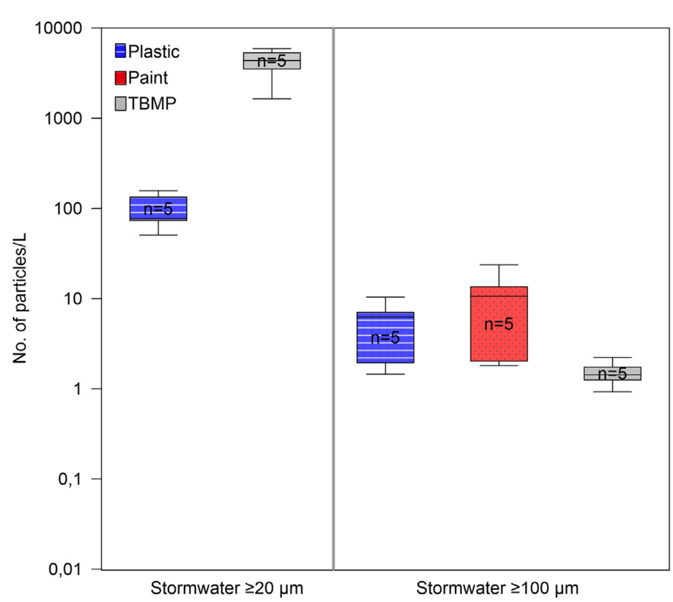

Fig. 11. Plastic, paint and TBMP in stormwater, measured as flow-weighted rain event mean concentration. Boxes to the left relate to particles ≥20 μm and boxes to the right show particles ≥100 μm. (No density separation of the samples).

fraction (≥100 μm), consists of heavier particles. These are less mobile than the smaller ones, and are therefore more likely to settle than smaller particles (Vogelsang et al., 2018).The dominance of particles 20–100 μm in stormwater could also be the result of a higher abundance of TBMP in this size range. This was something that also was found in a study by (Kreider et al., 2010).The results confirm that in order to better understand the transport routes and recipient loads it is important to analyse a wider range of microparticle sizes in the different media, and combine this with measurements of meteorological conditions.

3.3. Estimates and comparisons of theoretical and measured amounts of TWP in the stormwater

Applying the tire tread wear emission factor 0.1 g per vehicle kilometre (vkm) (Boulter, 2005; Councell et al., 2004), an estimated road length of 5.5 km, and an AADT of 5500–13,000 vehicles/day on the roads within the catchment area, the average tire wear per vehicle in the catchment area was calculated to 3,0–7,1 kg rubber emitted due to tire wear per day. This corresponds to an annual emission of 1.1–2.6 tons.

The build-up of the tire wear on the road depends on several factors, including maintenance and meteorological conditions (Kole et al., 2017; Wagner et al., 2018). The less precipitation, but wetter the street surface, and the less windy, the higher the build-up and the lower the spread through swirling, air transportation and runoff. A fairly small amount of runoff was expected during the period when no street sweeping or other maintenance was carried out, i.e. between the measurement after street sweeping 2018-03-26 and the measurements 2018-04-19, as it only rained on a few occasions (April 2–5, 7 and 15). The total precipitation for the period was ~40 mm (Fig. 3). Assuming the TWP runoff was 50% (Hann et al., 2018; Unice et al., 2019a; Wagner et al., 2018), this resulted in a concentration of 1.7 to 4.0 TWP/m^3 in the stormwater if the AADT was 5500–13,000 v/day (Supplementary Information calculations). Assuming an average particle size of 50 μm (as found in the particle size distribution in this study, Fig. 6), and a density of 1–1.8 kg/m^3 (Kreider et al., 2010; Rhodes et al., 2012; Sofi, 2018) this would result in 14,200–60,400 particles/L in the stormwater if the AADT was 5500 to 13,000 v/day. This is ten times higher than found in the stormwater analysis (1500 to 6000 particles/L, Fig. 11).Of the particle fractions analysed in stormwater in this project (i.e. ≥20 μm), most of the particles were in the lower size range 20–100 μm, but most of the TBMP can be expected to be found in the sediment in agreement with previous estimates that have indicated that 1–25% will reach surface waters (Hann et al., 2018; Unice et al., 2019a; Unice et al., 2019b; Wagner et al., 2018). However, it should be noted that the amount of TBMP in the fractions ≤20 μm is still unknown. Recent particle size distribution analysis indicates that washwater, stormwater and WDS samples contain a wide range of small particles, including nanoparticles in sizes from just below 1 nm (Polukarova et al., 2020), indicating that the number of particles found in this study likely is underestimated.

The results of this study indicate that the theoretical tire wear emission factor estimates are realistic, and that most of the tire wear is found as microparticles 20–100 μm, a high amount of which may also enter the stormwater. Our results is in line with previous estimates that have indicated that 1–25% will reach surface waters (Hann et al., 2018; Unice et al., 2019a; Unice et al., 2019b; Wagner et al., 2018), while the rest can be found in and transported via sediments. However, it should be noted that the amount of TBMP in the fractions <20 μm is still unknown. Furthermore, these assumptions have a high rate of uncertainty regarding the emission factors, as well as the average TWP density and diameter which have shown great variations in previous studies. Wagner et al. (2018) further illustrated the need of more research in the lab and in several additional field studies to better predict and understand the fate of TBMP in the environment. Recent particle size distribution analysis indicates that washwater, stormwater and WDS samples contain a wide range of small particles, including nanoparticles in sizes from just below 1 nm (Polukarova et al., 2020), indicating that the number of particles found in the stormwater this study likely are underestimated. All the runoff from impervious surfaces in the case study catchment area goes, without treatment, straight into the small creek Vitsippsbäcken, from which it continues via underground pipes into the river Göta älv, close to its outlet to the Kattegat in the North Sea. This means that large amounts of the emitted TWP and TBMP may also reach the Göta älv estuary and the marine environment. The results of the study therefore indicate that the contribution of microplastics from TBMP, from the catchment area to the Göta älv estuary may be significant. The national load of microplastics from TRWP is at least 8200 tons per year, as estimated by (Magnusson et al., 2016) based on the available emission factors that have also been applied in this study. Accordingly, stormwater transportation of TWP and TBMP may contribute significantly to the microplastic loads in the Swedish surface waters, sediments and marine environments. In addition other transport routes such as snow collected from urban streets and dumped directly into coastal waters and deposition from TBMP via air may contribute to the load of TBMP in surface waters, sediments and marine environments (HELCOM, 2016; Valotto et al., 2015; Wijesiri et al., 2016). Despite removing TBMP from roads, street sweeping (sweepsand and washwater) may become a source of TBMP and other pollutants elsewhere in the environment. To what extent and where depends on the subsequent treatment.

3.4. General discussion

The results presented in this paper are based on field samples analysed in the case study area. The samples were collected during different seasons with and without winter tires on the vehicles, before and after street sweeping and also after periods allowing TBMP build-up without sweeping. An advantage with the WDS method is that the WDS can be used under wet and moist conditions, such as on wet pavements or during rain events, which is not possible with other measurement techniques e.g. vacuum cleaners. Even if the number of TBMP increased after sweeping in some of the WDS samples, a considerable amount of TBMP was detected in the sweepsand as well as in the washwater during the same sampling occasion indicating that street sweeping can be an effective measure. The street sweeping thereby reduced the TBMP that could have been potentially transported via stormwater out in the environment.

A limitation of the study is the limitation of collected samples in the different media. However, the stormwater samples were taken by flow-proportional samples meaning that one stormwater sample corresponds to the mean value of a whole rain event and thus equals to many single grab samples. The sweepsand samples was systematically taken in small portions from different places in the pile of approximately 1 m^3 of sweepsand from the machine to receive a representative pooled sample for the whole study area. The same technique was used for the total volume of approximately 1m^3 of well mixed washwater from which the samples for analysis was taken. For this reason, it is worth to notice that "just one" sample correspond to a mean value and a representative sample from of a huge amount of stormwater, sweepsand and washwater.

A strength of the present findings can be the fact that all matrices studied are field samples. A related weakness, however, is that the sampled media is presumably very heterogenous and varies on the street due to ambient conditions like, traffic, precipitation and wind characteristics. The wind speed, and wind direction, was similar during the measurement period, but may vary considerably with time. For future studies a larger number of samples in all media, taken under the same periods and during periods with higher variations in meteorological conditions is recommended, to be able to further evaluate the distribution and transport pathways of TBMP in different media. Another limitation of the present study is the restricted particle size fraction analysed and thereby the likely underestimation of the ambient number of TBMP. Only the fraction ≥100 μm was measured in the WDS, sweepsand and washwater samples. In the stormwater, particles ≥20 μm were analysed and the number of particles in the particle fraction 20–100 μm dominated over the larger fraction. This indicates that the number of particles in the other media may be significantly underestimated. Further the selection of solution for density separation was shown to be of importance. The stormwater was not at all in need of such separation, while the WDS and the washwater samples were separated with NaI which was found to be preferable over NaCl. Unfortunately, only the last sweepsand samples were density separated with NaI, likely underestimating TBMP particles with higher density as also shown in comparison to the NaCl-separation. In summary, the results indicate the need of a broad particle size spectra and the application of NaI or solutes with higher density than NaCl for sample separation.

4. Conclusions

To be able to estimate the total amounts of TBMP and TWP in stormwater and road particle matrices, a broad size spectrum of particles should be analysed, which includes particles at least ≤100 μm and preferably smaller. The media used for density separation of TBMP is of great importance, and the results in this study shows that NaI, or another high dense solute, is preferable over NaCl. Here the number of TWP in the NaI separated samples were 2–10 times higher than the number of TWP in the NaCl separated samples. This may be explained by the fact that TWP collected in the field are considerably heavier due to mineral encrustations compared to "pure rubber" particles that have not been exposed to other wear materials. The underestimation due to sample separation techniques can be likely applicable to other types of microplastic particles in the environment as well (primary and secondary).

The largest proportion of anthropogenic microparticles detected in the samples consisted of rubber and bitumen, which confirms that TBMP is one of the major sources of microplastics. The ratio may, however, vary depending on several factors including method for separation, particle size fraction analysed, sampling techniques, nearby emission sources and meteorological conditions. The results indicate that the measured emissions of TWP are well in line with previous theoretical estimations on tire wear particles on road surfaces. During winter, studded tires contribute to high concentrations of road (bitumen) wear particles which were found both on the road, in the stormwater and in the sweepsand. Outside the studded tire season, TWP makes up approximately 50% of the TBMP.

Road sweeping collects considerable amounts of TBMP, thereby preventing particles from reaching various recipients in the environment for example vi stormwater and wind.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.138950.

References

Abbasi, S., Keshavarzi, B., Moore, F., Turner, A., Kelly, F.J., Dominguez, A.O., Jaafarzadeh, N., 2019. Distribution and potential health impacts of microplastics and microrubbers in air and street dusts from Asaluyeh County, Iran. Environ. Pollut. 244, 153–164. https://doi.org/10.1016/j.envpol.2018.10.039.

Abdel-Wahab, M.M., Wang, C., Vanegas-Useche, L.V., Parker, G.A., 2011. Experimental determination of optimum gutter brush parameters and road sweeping criteria for different types of waste. Waste Manag. 31, 1109–1120. https://doi.org/10.1016/j.wasman.2010.12.014.

Amato, F., Querol, X., Johansson, C., Nagl, C., Alastuey, A., 2010. A review on the effectiveness of street sweeping, washing and dust suppressants as urban PM control methods. Sci. Total Environ. 408, 3070–3084. https://doi.org/10.1016/j.scitotenv.2010.04.025.

Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62, 1596–1605. https://doi.org/10.1016/j.marpolbul.2011.05.030.

AQEG, 2019. Non-Exhaust Emissions from Road Traffic. https://uk-air.defra.gov.uk/assets/documents/reports/cat09/1907101151_20190709_Non_Exhaust_Emissions_typeset_Final.pdf.

Auta, H.S., Emenike, C.U., Fauziah, S.H., 2017. Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ. Int. 102, 165–176. https://doi.org/10.1016/j.envint.2017.02.013.

Barboza, L.G.A., Gimenez, B.C.G., 2015. Microplastics in the marine environment: current trends and future perspectives. Mar. Pollut. Bull. 97, 5–12. http://10.1016/j.marpolbul.2015.06.008.

Bogacki, M., Oleniacz, R., Rzeszutek, M., Szulecka, A., Mazur, M., 2018. The impact of street cleaning on particulate matter air concentrations: a case study of a street canyon in Krakow (Poland). E3S Web Conf 45, 00009. https://doi.org/10.1051/e3sconf/20184500009.

Bondelind, M., Nguyen, A., Sokolova, E., Björklund, K., 2019. Transport of Traffic-Related Microplastic Particles in Receiving Water. Mannina G. Springer International Publishing, pp. 317–321. https://doi.org/10.1007/978-3-319-99867-1_53.

Boulter, P.G., 2005. A Review of Emission Factors and Models for Road Vehicle Non-exhaust Particulate Matter. https://trl.co.uk/sites/default/files/PPR065.pdf.

Councell, T.B., Duckenfield, K.U., Landa, E.R., Callender, E., 2004. Tire-Wear particles as a source of zinc to the environment. Environmental Science & Technology 38, 4206–4214. https://doi.org/10.1021/es034631f.

Egodawatta, P., Thomas, E., Goonetilleke, A., 2007. Mathematical interpretation of pollutant wash-off from urban road surfaces using simulated rainfall. Water Res. 41, 3025–3031. https://doi.org/10.1016/j.watres.2007.03.037.

Eisentraut, P., Dümichen, E., Ruhl, A.S., Jekel, M., Albrecht, M., Gehde, M., Braun, U., 2018. Two birds with one stone—fast and simultaneous analysis of microplastics: microparticles derived from thermoplastics and tire wear. Environmental Science & Technology Letters 5, 608–613. https://doi.org/10.1021/acs.estlett.8b00446.

ETRMA, 2017. Annual Report 2016–2017. https://www.etrma.org/wp-content/uploads/2019/09/20170905-etrma-annual-report-2016-17-final.pdf.

European-Commission, 2019. Environmental Health Risks of Microplastic Pollution. https://doi.org/10.2777/65378.

German, J., 2003. Reducing Stormwater Pollution - Performance of Retention Ponds and Street Sweeping. 2003. Chalmers University of Technology.

GESAMP, 2019. Microplastics in the Ocean. http://www.gesamp.org/site/assets/files/1720/24472_gesamp_leaflet_pq.pdf.

Gjerstad, K.-I., Gustafsson, M., Blomqvist, G., Denby, B., Elmgren, M., Grythe, H., Janhäll, S., Järlskog, I., Johansson, C., Kulovuori, S., Kupiainen, K., Lundberg, J., Malinen, A., Norman, M., Ritola, R., Silvergren, S., Stojiljkovic, A., Sundvor, I., Þorsteinsson, Þ., Stefani, M., Vogt, M., 2019. NORDUST- Nordic Road Dust Project. http://www.nordfou.org/Documents/NorDUST/NorDust%20Final%20report.pdf.

Gustafsson, M., 2001. Icke-avgasrelaterade partiklar i vägmiljön : literature review. http://urn.kb.se/resolve?urn=urn:nbn:se:vti:diva-4538.

Gustafsson, M., Johansson, C., 2012. Road Pavements and PM10. Summary of the Results of Research Funded by the Swedish Transport Administration on How the Properties of Road Pavements Influence Emissions and the Properties of Wear Particles. Trafikverket, Report 2012:241, 2012.

Gustafsson, M., Blomqvist, G., Järlskog, I., Lundberg, J., Janhäll, S., Elmgren, M., Johansson, C., Norman, M., Silvergren, S., 2019. Road dust load dynamics and influencing factors for six winter seasons in Stockholm, Sweden. Atmospheric Environment: X 2, 100014. https://doi.org/10.1016/j.aeaoa.2019.100014.

Hann, S., Sherrington, C., Jamieson, O., Hickman, M., Kershaw, P., Bapasola, A., Cole, G., 2018. Investigating Options for Reducing Releases in the Aquatic Environment of Microplastics Emitted by (but Not Intentionally Added in) Products. http://ec.europa.eu/environment/marine/good-environmental-status/descriptor-10/pdf/microplastics_final_report_v5_full.pdf.

HELCOM, 2016. Report of the HELCOM Stakeholder Conference on Marine Litter. https://helcom.fi/wp-content/uploads/2019/08/Report-of-the-Stakeholder-Conference.pdf.

Imhof, H.K., Schmid, J., Niessner, R., Ivleva, N.P., Laforsch, C., 2012. A novel, highly efficient method for the separation and quantification of plastic particles in sediments of aquatic environments. Limnol. Oceanogr. Methods 10, 524–537. https://doi.org/10.4319/lom.2012.10.524.

Järlskog, I., Blomqvist, G., Gustafsson, M., Janhäll, S., 2017. Utvärdering av städmaskiners förmåga att reducera vägdammsförrådet i gatu- och tunnelmiljöer : En fältstudie i Trondheim. 2016. http://urn.kb.se/resolve?urn=urn:nbn:se:vti:diva-12559.
Jiang, J.Q., 2018. Occurrence of microplastics and its pollution in the environment: a review. Sustainable Production and Consumption 13, 16–23. https://doi.org/10.1016/j.spc.2017.11.003.
Jonsson, P., Blomqvist, G., Gustafsson, M., 2008. Wet Dust Sampler: Technological Innovation for Sampling Particles and Salt on Road Surface. Transportation Research Board, TRB, pp. 102–111. http://urn.kb.se/resolve?urn=urn:nbn:se:vti:diva-5213.
Klöckner, P., Reemtsma, T., Eisentraut, P., Braun, U., Ruhl, A.S., Wagner, S., 2019. Tire and road wear particles in road environment - quantification and assessment of particle dynamics by Zn determination after density separation. Chemosphere 222, 714–721. https://doi.org/10.1016/j.chemosphere.2019.01.176.
Kole, P.J., Löhr, A.J., Van Belleghem, F.G.A.J., Ragas, A.M.J., 2017. Wear and tear of tyres: a stealthy source of microplastics in the environment. Int. J. Environ. Res. Public Health 14. https://doi.org/10.3390/ijerph14101265.
Kreider, M.L., Panko, J.M., McAtee, B.L., Sweet, L.I., Finley, B.L., 2010. Physical and chemical characterization of tire-related particles: comparison of particles generated using different methodologies. Sci. Total Environ. 408, 652–659. https://doi.org/10.1016/j.scitotenv.2009.10.016.
Lau, S.-L., Stenstrom, M.K., 2005. Metals and PAHs adsorbed to street particles. Water Res. 39, 4083–4092. https://doi.org/10.1016/j.watres.2005.08.002.
Liu, F., Olesen, K.B., Borregaard, A.R., Vollertsen, J., 2019. Microplastics in urban and highway stormwater retention ponds. Sci. Total Environ. 671, 992–1000. https://doi.org/10.1016/j.scitotenv.2019.03.416.
Lundberg, J., Blomqvist, G., Gustafsson, M., Janhäll, S., Järlskog, I., 2019. Wet dust sampler—a sampling method for road dust quantification and analyses. Water Air Soil Pollut. 230, 180. https://doi.org/10.1007/s11270-019-4226-6.
Magnusson, K., Eliasson, K., Fråne, A., Haikonen, K., Hultén, J., Olshammar, M., Stadmark, J., Voisin, A., 2016. Swedish Sources and Pathways for Microplastics to the Marine Environment- a Review of Existing Data. https://www.naturvardsverket.se/upload/miljoarbete-i-samhallet/miljoarbete-i-sverige/regeringsuppdrag/2016/mikroplaster/swedish-sources-and-pathways-for-microplastics-to-marine%20environment-ivl-c183.pdf.
Murakami, M., Nakajima, F., Furumai, H., 2004. Modelling of runoff behaviour of particle-bound polycyclic aromatic hydrocarbons (PAHs) from roads and roofs. Water Res. 38, 4475–4483. https://doi.org/10.1016/j.watres.2004.07.023.
Norman, M., Johansson, C., 2006. Studies of some measures to reduce road dust emissions from paved roads in Scandinavia. Atmos. Environ. 40, 6154–6164. https://doi.org/10.1016/j.atmosenv.2006.05.022.
Nynas, 2017. Nybit E 190 Safety Data Sheet. https://notes.nynas.com/Apps/1112.nsf/wsds/SE_SV_Nybit_E_190/$File/Nybit_E_190_SE_SV_SDS.pdf.
Panko, J.M., Chu, J., Kreider, M.L., Unice, K.M., 2013. Measurement of airborne concentrations of tire and road wear particles in urban and rural areas of France, Japan, and the United States. Atmos. Environ. 72, 192. https://doi.org/10.1016/j.atmosenv.2013.01.040.
Polukarova, M., Markiewicz, A., Björklund, K., Strömvall, A.-M., Galfi, H., Andersson Sköld, Y., Gustafsson, M., Järlskog, I., Aronsson, M., 2020. Organic pollutants, nano- and microparticles in street sweeping road dust and washwater. Environ. Int. 135, 105337. https://doi.org/10.1016/j.envint.2019.105337.
Rhodes, E.P., Ren, Z., Mays, D.C., 2012. Zinc leaching from tire crumb rubber. Environmental Science & Technology 46, 12856–12863. https://doi.org/10.1021/es3024379.
SMHI, 2019. Ladda nermeteorologiska observationer Nederbördsmängd (dygn) Göteborg A. SMHI. http://www.smhi.se/data/meteorologi/ladda-ner-meteorologiska-observationer/#param=precipitation24HourSum,stations=all,stationid=71420.
Snilsberg, B., Gryteselv, D., 2016. Renholdsforsøk i tunnel og gate i Trondheim våren 2015. https://www.vegvesen.no/fag/teknologi/tunneler/publikasjoner/_attachment/1431625?_ts=155b568f6f8&fast_title=Renholdsfors%C3%B8k+i+tunnel+og+gate+i+Trondheim+v%C3%A5ren+2015%3A+Strindheimtunnelen+og+Haakon+VII+gate%C2%A0.
Snilsberg, B., Gryteselv, D., 2017. Road Cleaning in Tunnel and Street. 2016. https://www.vegvesen.no/fag/publikasjoner/publikasjoner/Statens+vegvesens+rapporter.
Snilsberg, B., Gryteselv, D., Sætermo Veivåg, I.-L., 2017. Road Cleaning in Tunnel and Street. 2017. https://www.vegvesen.no/fag/publikasjoner/publikasjoner/Statens+vegvesens+rapporter/_attachment/2105462?_ts=16069741358&download=true&fast_title=Renholdsfors%C3%B8k+2017%3A+Uttesting+av+ny+spylebom+i+tunnel+og+gate+i+Kristiansund.
Snilsberg, B., Gryteselv, D., Sætermo Veivåg, I.-L., Lamo Hauan, T., Dalseth Austigard, Å., 2018. Renholdsforsøk 2017. https://www.vegvesen.no/fag/publikasjoner/publikasjoner/Statens+vegvesens+rapporter.
Sofi, A., 2018. Effect of waste tyre rubber on mechanical and durability properties of concrete – a review. Ain Shams Engineering Journal 9, 2691–2700. https://doi.org/10.1016/j.asej.2017.08.007.
Sommer, F., Dietze, V., Baum, A., Sauer, J., Gilge, S., Maschowski, C., Gieré, R., 2018. Tire abrasion as a major source of microplastics in the environment. Aerosol Air Qual. Res. 18, 2014–2028. https://doi.org/10.4209/aaqr.2018.03.0099.
Sundt, P., Schulze, P.-E., Syversen, F., 2014. Sources of Microplastic Pollution to the Marine Environment. https://www.miljodirektoratet.no/globalassets/publikasjoner/M321/M321.pdf.
Sutton, R., Mason, S.A., Stanek, S.K., Willis-Norton, E., Wren, I.F., Box, C., 2016. Microplastic contamination in the San Francisco Bay, California, USA. Mar. Pollut. Bull. 109, 230–235. https://doi.org/10.1016/j.marpolbul.2016.05.077.
U.S.EPA, 1983. Results of the Nationwide Urban Runoff Program. Volume I- Final Report. Water Planning Division, Washington, p. 1983.
Unice, K.M., Kreider, M.L., Panko, J.M., 2012. Use of a deuterated internal standard with pyrolysis-GC/MS dimeric marker analysis to quantify tire tread particles in the environment. Int. J. Environ. Res. Public Health 9, 4033–4055. https://doi.org/10.3390/ijerph9114033.
Unice, K.M., Kreider, M.L., Panko, J.M., 2013. Comparison of tire and road wear particle concentrations in sediment for watersheds in France, Japan, and the United States by quantitative pyrolysis GC/MS analysis. Environmental Science & Technology 47, 8138–8147. https://doi.org/10.1021/es400871j.
Unice, K.M., Weeber, M.P., Abramson, M.M., Reid, R.C.D., van Gils, J.A.G., Markus, A.A., Vethaak, A.D., Panko, J.M., 2019a. Characterizing export of land-based microplastics to the estuary - part I: application of integrated geospatial microplastic transport models to assess tire and road wear particles in the seine watershed. Sci. Total Environ. 646, 1639–1649. https://doi.org/10.1016/j.scitotenv.2018.07.368.
Unice, K.M., Weeber, M.P., Abramson, M.M., Reid, R.C.D., van Gils, J.A.G., Markus, A.A., Vethaak, A.D., Panko, J.M., 2019b. Characterizing export of land-based microplastics to the estuary - part II: sensitivity analysis of an integrated geospatial microplastic transport modeling assessment of tire and road wear particles. Sci. Total Environ. 646, 1650–1659. https://doi.org/10.1016/j.scitotenv.2018.08.301.
Valotto, G., Rampazzo, G., Visin, F., Gonella, F., Cattaruzza, E., Glisenti, A., Formenton, G., Tieppo, P., 2015. Environmental and traffic-related parameters affecting road dust composition: a multi-technique approach applied to Venice area (Italy). Atmos. Environ. 122, 596–608. https://doi.org/10.1016/j.atmosenv.2015.10.006.
Vanegas Useche, L.V., Wahab, M.M.A., Parker, G.A., 2010. Effectiveness of gutter brushes in removing street sweeping waste. Waste Manag. 30, 174–184. https://doi.org/10.1016/j.wasman.2009.09.036.
Vanegas-Useche, L.V., Abdel-Wahab, M.M., Parker, G.A., 2015. Effectiveness of oscillatory gutter brushes in removing street sweeping waste. Waste Manag. 43, 28–36. https://doi.org/10.1016/j.wasman.2015.05.014.
Vanegas-Useche, L.-V., Abdel-Wahab, M.-M., Parker, G.-A., 2018. Determination of the normal contact stiffness and integration time step for the finite element modeling of bristle-surface interaction. Computers, Materials & Continua 56, 169–184. http://www.techscience.com/cmc/v56n1/27799.
Vanegas-Useche, L.-V., Abdel-Wahab, M.-M., Parker, G.-A., 2019. Dynamic analysis of a horizontal oscillatory cutting brush. Computers, Materials & Continua 60, 871–893. http://www.techscience.com/cmc/v60n3/23069.
Verschoor, A.J., Valk, E.d., 2018. Potential Measures against Microplastic Emissions to Water. https://doi.org/10.21945/RIVM-2017-0193.
Verschoor, A., de Poorter, L., Dröge, R., Kuenen, J., de Valk, E., 2016. Emission of Microplastics and Potential Mitigation Measures. https://ia802807.us.archive.org/4/items/blg-777944/blg-777944.pdf.
Vogelsang, C., Lusher, A., E Dadkhah, M., Sundvor, I., Umar, M., B Ranneklev, S., Pettersen Eidsvoll, D., Meland, S., 2018. Microplastics in Road Dust - Characteristics, Pathways and Measures: Norwegian Institute for Water Research (NIVA), 2018.
Wagner, S., Hüffer, T., Klöckner, P., Wehrhahn, M., Hofmann, T., Reemtsma, T., 2018. Tire wear particles in the aquatic environment - a review on generation, analysis, occurrence, fate and effects. Water Res. 139, 83–100. https://doi.org/10.1016/j.watres.2018.03.051.
Wijesiri, B., Egodawatta, P., McGree, J., Goonetilleke, A., 2016. Understanding the uncertainty associated with particle-bound pollutant build-up and wash-off: a critical review. Water Res. 101, 582–596. https://doi.org/10.1016/j.watres.2016.06.013.
Zhao, H., Li, X., 2013. Understanding the relationship between heavy metals in road-deposited sediments and washoff particles in urban stormwater using simulated rainfall. J. Hazard. Mater. 246-247, 267–276. https://doi.org/10.1016/j.jhazmat.2012.12.035.