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Organic pollutants, nano- and microparticles in street sweeping road dust and washwater

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ABSTRACT

Road areas are pollution hotspots where many metals, organic pollutants (OPs) and nano/microparticles accumulate before being transported to receiving waters. Particles on roads originate from e.g. road, tyre and vehicle wear, winter road maintenance, soil erosion, and deposition. Street sweeping has the potential to be an effective and affordable practice to reduce the occurrence of road dust, and thereby the subsequent spreading of pollutants, but there is currently little knowledge regarding its effectiveness. In this paper we investigate the potential of street sweeping to reduce the amounts of OPs and nano/microparticles reaching stormwater, in a case study sampling road dust and washwater from a street sweeping machine, road dust before and after sweeping, and stormwater. The compound groups generally found in the highest concentrations in all matrices were aliphatics C₅–C₃₅ > phthalates > aromatics C₈–C₃₅ > PAH-16. The concentrations of aliphatics C₁₆–C₃₅ and PAHs in washwater were extremely high at ≤ 53,000 µg/L and ≤ 120 µg/L, respectively, and the highest concentrations were found after a 3-month winter break in sweeping. In general, fewer aliphatic and aromatic petroleum hydrocarbons and PAHs were detected in road dust samples than in washwater. The relative composition of the specific PAH-16 suggests tyre wear, vehicle exhausts, brake linings, motor oils and road surface wear as possible sources. The study indicates that many of the hydrophobic compounds quantified in washwater are attached to small particles or truly dissolved. The washwater contains a wide range of small particles, including nanoparticles in sizes from just below 1 nm up to 300 nm, with nanoparticles in the size range 25–300 nm present in the highest concentrations. The results also indicated agglomeration of nanoparticles in the washwater. The street sweeping collected a large amount of fine particles and associated pollutants, leading to the conclusion that washwater from street sweeping needs to be treated before disposal.

1. Introduction

Urban areas are pollution hotspots, where compounds from many diffuse and point sources are emitted, and as cities grow, the negative impact of human activities on the environment becomes more significant and therefore more important to consider. Many water pollutants are transported from their sources to receiving waters via particles in surface runoff (Oliveira and Almeida, 2019; Nielsen et al., 2015). Protecting the environment from small particles containing nano- and microplastics, toxic metals, and organic pollutants (OPs) has a high societal value (Lehner et al., 2019; Rodrigues et al., 2019; Vedolin et al., 2018) in that it helps to achieve national environmental objectives, the

EU Marine Strategy and Water Framework Directives, and the UN Sustainable Development Goal to conserve and sustainably use the oceans, seas and marine resources. Urban runoff, which carries substantial amounts of particles, is recognized as one of the most significant pressures on aquatic ecosystems worldwide (Brudler et al., 2019). The highest concentrations of pollutants, including particles, microplastics, metals and OPs, are often found in runoff from traffic environments.

It is well-known that many OPs are emitted in road environments, from e.g. exhaust gases, fuels, lubricating oils, road construction materials, and vehicle components, such as the body, brakes and tyres (Folkesson et al., 2009; Peikertova and Filip, 2015; Wagner et al., 2018).

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In a literature and database search, 1,100 specific OPs were identified as potentially emitted from the road environment (Markiewicz et al., 2017). Of all OPs, polycyclic aromatic hydrocarbons (PAHs) were pinpointed as having the highest priority for remediation, due to their toxicity and large emitted loads. Other OPs on the priority pollutant list for traffic-related emissions are alkanes C₂₀–C₄₀, alkylphenols, phthalates, aldehydes, phenolic antioxidants, bisphenol A, oxygenated-PAHs, naphtha C₅–C₁₂, amides, and amines. In stormwater, surprisingly high concentrations of PAHs have been found both in the dissolved and colloidal fractions, including nano-sized particles < 10 nm (Nielsen et al., 2015). It has also been shown that a mix of hydrophobic organic pollutants, diesel and colloids has the potential to form nano- and micro-sized emulsions, and that emulsions may act as carriers of OPs in road runoff (Markiewicz et al., 2019). Higher concentrations of nano-particles < 100 nm were found in the mixture of all OPs without colloids added, than in the OP mixture with colloids added.

Particles on roads emerge from e.g. road maintenance, soil erosion, wear of vehicles and roads, as well as deposition. During periods with little or no precipitation, particles build up on road surfaces and may accumulate pollutants such as metals and OPs. The build-up rate of particles on roads, often referred to as road dust, ranges from a few grams to several hundred grams per day and m², depending on traffic intensity and driving conditions (Deletic and Orr, 2005, Morgan et al., 2017). Traffic suspension and turbulence affect the lateral distribution of dust across the road, with low amounts found in wheel tracks, and higher outside them. Normally, the kerb is the most dust-laden surface on a road (Gustafsson et al., 2019). Seasonal variations in the strengths of the dust sources also influence the dust load. In Nordic countries, the sources are stronger in winter due to road wear from studded tyres and winter traction sanding (Kupiainen et al., 2016). Recent studies identified traffic as an important emission source to the environment of nano- and microparticles in the form of plastics: traffic-related sources include wear of tyres, road materials, painting and markers, as well as vehicle parts (Wagner et al., 2018, Horton et al., 2017, Siegfried et al., 2017, da Costa et al., 2016).

Street sweeping techniques have been investigated as potential methods for reducing stormwater pollution close to the emission source multiple times in the past decades (Calvillo et al., 2015). Gustafsson et al. (2019) show that the load of road dust particles < 180 µm varies between 15 g/m² in early autumn and late spring and 200 g/m² in winter and early spring in Stockholm, Sweden. Large amounts of road dust and anti-skid sand are collected every year; the collected amount in Sweden in 2008, estimated by Sweden's municipalities and county councils, was 750,000 tonnes. The collected material by street sweeping contains debris and particles of different sizes; the finest fractions are assumed to be the most polluted, and are composed of minerals, rubber, asphalt, other organic materials and emulsions, which are all efficient carriers of pollutants (Aryal et al., 2017). A strong correlation between smaller particle sizes and high concentrations of metals and PAHs have been observed in street sweeping materials (Lloyd et al., 2018). National regulations on disposal of road dust from street sweeping are currently missing in most countries, why the waste is often deposited on-site or in landfills, where sorbed pollutants risk being released back into the environment. If these urban materials are treated to remove pollutants and reused, landfill deposition and extraction of natural gravel, a limited resource that is often used for anti-skid purposes, could be reduced.

Street sweeping has the potential to be an effective and affordable way to reduce the occurrence of pollutants in road dust, thereby also reducing the spreading of pollutants with particles in road runoff. Furthermore, street sweeping is an “at the source” practice that can limit contamination from traffic-related sources. Currently, there is little research on how street sweeping may improve water quality by catching OPs and nano- and micro-sized particles originating from road traffic. This research aims to increase knowledge about the characteristics of OPs in road dust, sampled with a street sweeping machine, with

focus on particles < 2 mm, including nanoparticles of 0.3–450 nm. The study also aims to evaluate the sustainability and efficiency of street sweeping in reducing the transport of OPs and improving stormwater quality. The hypotheses are: (1) nano- and microparticles and OPs are collected effectively by the street sweeping machine; (2) transport of nano- and microparticles and OPs to receiving waters decreases after street sweeping; (3) the waste material collected in the street sweeping machine contains high concentrations of nano- and microparticles and OPs, hence the material requires treatment before it is disposed or reused. The chemical composition of nano- and micro-particles 1 nm–0.45 µm analysed in this study has not been determined due to limitations in available analysis techniques for such small particles.

In this study, nano- and microparticles are combined with OPs because particles emitted in the road environment, mainly tyre and road wear-associated particles, may contain OPs (Markiewicz et al., 2017), and/or OPs may be sorbed to the surface of the small wear particles (Ivar do Sul and Costa, 2014), and/or OPs can form both nano- and micro-sized emulsions (Markiewicz et al., 2019), and also to better understand the origin and transport routes of both these groups of contaminants.

2. Methods

2.1. Study area

An urban catchment located in Gothenburg, Sweden was selected for studying the removal of pollutants and small particles through street sweeping carried out weekly (Fig. 1). The study area is a mixed catchment adjacent to the Sahlgrenska Hospital and includes hospital and university buildings with large copper roofs, parks, forest, parking lots and roads with annual average daily traffic volumes of between 2,000 and 24,000. The total area of the drained catchment is around 50 ha, made up of 10% parking lots and roads, 13% buildings, 17% other paved areas, and 60% pervious areas. Runoff from impervious surfaces is drained via separate storm sewers, without any treatment, to a natural stream, Vitsippsbäcken.

2.2. Sampling procedure

Street sweeping was performed weekly in 2017, from August 17 to November 26, and on one occasion in December. Prior to August 17, street sweeping was carried out monthly. When the autumn defoliation began at the beginning of October, the amount of collected road dust declined drastically, which could be due to a limited ability of the sweeping machine to collect heterogeneous material, and difficulty in collecting dust covered by leaves. In addition, it was difficult to separate the collected dust from the leaves, which resulted in an altered composition of samples from October (compared to those collected in September). For this reason, OPs were not analysed in samples collected in October. Two additional street sweeping events, on March 26 and April 18, 2018, were performed after the winter. No sweeping was performed during the winter, due to freezing conditions and ice control measures.

The sweeping was performed in the morning between 7 and 9 am on streets that were not closed off for traffic while the sweeping took place, which resulted in slight variations in the sweeping performance and covered distances (from 3.5 to 4.5 km, Fig. 1). In most cases, the right lane of the road, nearest the kerb, was swept. The sweeping machine used is a vacuum type produced by Johnston Beam. To suspend the finest particles during sweeping, the pipe that sucks up the road particles is moisturized with water. In dry weather conditions, the machine also sprays water onto the road to prevent fine dust from being emitted into the air. The total water storage volume in the machine is approximately 1.3 m³. The amount of water sprayed during the sweeping events varied depending on weather conditions, which is in line with

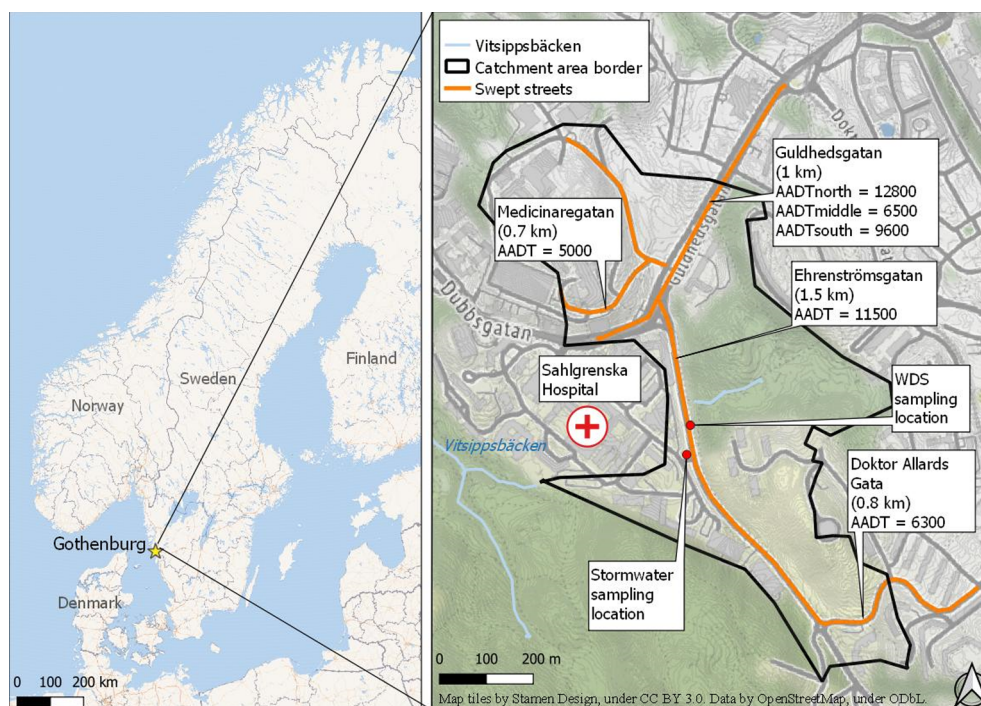


Fig. 1. The studied urban area and catchment of the small stream Vitsippsbäcken, located in Gothenburg, Sweden. The distances presented in parentheses represent an approximated sum of distances covered by the street sweeping machine to clean the street in each direction. AADT is the annual average daily traffic on the specific street.

regular sweeping routines. As the width of the machine is approximately $\frac{3}{4}$ of the width of a lane, the entire road surface was not swept on every occasion.

After each sweeping event, the sweeping machine was driven to a local site for deposition of the collected road dust masses. Here, samples of settled road dust and the washwater were manually collected in glass bottles and stainless-steel buckets. The volume of the collected road dust and washwater was estimated to 10–20 L each. The washwater included stormwater 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 sweeping machine. The machine was used for sweeping other streets during the experimental period, but was washed with water, and in some cases soft soap, before each sweeping of the case study area.

To evaluate the efficiency of the street sweeping, road dust was also sampled before and after sweeping, using the Wet Dust Sampler II (WDSII) technique (Fig. S1, Supplementary data), designed and operated by the Swedish National Road and Transport Research Institute (VTI) (Lundberg et al., 2019; Gustafsson et al., 2019; Jonsson et al., 2008). Samples were collected both before and after street sweeping at Ehrenströmsgatan (Fig. 1) at two occasions. At both occasions, the samples were collected randomly on the kerb, in wheel tracks, and between wheel tracks across the left lane along 5 m of the street, and 10 subsamples were pooled to a composite sample of approximately 10 L, to represent the road pollution level before street sweeping. The procedure was repeated on an adjacent road surface area after street sweeping was performed. More details for the procedure of the WDSII sampling are presented in Supplementary Data, section 1 and 8.

In parallel with the weekly street sweeping, stormwater was collected during five rain events in the autumn of 2017. An automatic sampler (type ISCO 6712) and an area velocity flow meter (Type Triton +) were installed in a stormwater manhole located close to the outlet to Vitsippsbäcken (Fig. 1), allowing flow-weighted sampling of stormwater. One composite sample comprising of flow-weighted subsamples collected during entire rain events was sampled for each rain event. Close to the sampling point, a tipping bucket rain gauge (type MJK) was installed on one of the hospital buildings to monitor rainfall. Precipitation amounts and intensity for sampled storm events are described in the Supplementary data (Table S1).

2.3. Laboratory analysis

Samples of road dust and washwater collected during street sweeping, water from the WDS, and particles in washwater collected on a $0.7 \mu\text{m}$ glass fibre filter, were analysed for general quality parameters, occurrence of nanoparticles and selected OPs. Particle size distribution (PSD) per weight for the road dust, and per volume and number weight for particles dispersed in water, was calculated. A summary of the performed analyses is presented in Table 1.

The particle size distribution (mm) of the collected road dust was determined on triplicate samples (1.8–3.0 kg) by wet sieving, performed according to ISO 13765-5. To characterise the finer fractions $< 0.063 \text{ mm}$, road dust samples from three sweeping events were also analysed using the total sieving curve method, with combined wet and laser sieving techniques (Table 1).

The size distribution of particles in washwater, stormwater, and WDS samples was analysed by dynamic light scattering (DLS) using the Litesizer™ 500 (by Anton Paar). The instrument has a 40 mW semiconductor red laser with a wavelength of 658 nm, and the most preferable detection angle (i.e. back (175°), side (90°) or forward (15°)) was automatically selected by the instrument. The DLS technique measures the speed of the Brownian motion, i.e. random movement of particles suspended in a liquid sample, which speed depends on the particle size. In the DLS measurement applied, red light scatters from particles was registered multiple times; the difference between each record gives the information on the particle speed and the particle average size from which the size distribution is calculated. The first outcome of a DLS measurement is intensity weight distribution by measuring the variation in angles at which the light is scattered from the particles. Here, the small particles scatter light at a larger angle than larger particles. Further, the results can be converted to volume weight, where the system makes several assumptions regarding particle homogeneity, e.g. spherical shape, density and optical properties and where each size interval of PSD shows how large % of the total volume is occupied by the certain particle group size. The results from DLS can also be converted to number weight, where the number of particles is counted in each size interval irrespective of their occupying space. This means that each size interval based on the number of particles in this interval accounts for a certain part (percentage) of the sample and is

Table 1

Methods for the chemical and physical analyses performed on road dust and washwater collected by the street sweeping machine, on stormwater, and on samples from the Wet Dust Sampler.

Analysed parameter	Matrix	Analytical approach	Standardised method, equipment
PSD per weight	road dust	Wet sieving, fractions 20, 16, 11.2, 8, 5.6, 4, 2, 1, 0.5, 0.25, 0.125 and 0.063 mm	ISO 13765-5
PSD per weight	road dust	Wet and laser sieving, using sieving > 2 mm, 1–2, 0.5–1, 0.25–0.5, 0.125–0.25, 0.063–0.125, 0.032–0.063, 0.016–0.032, 0.008–0.016, 0.004–0.008, 0.002–0.004, < 0.002	ISO 11277:2009
PSD per volume weight	washwater, WDS water, stormwater	Dynamic light scattering from 0.3 nm to 10 µm, laser 658 nm (red)	Litesizer™ 500 Anton Paar
PSD per number weight	washwater, WDS water, stormwater	Nanoparticle tracking analysis 10 nm to 2 µm, laser 488 nm (blue)	Nanosight NS300 Malvern Instruments Ltd.
Zeta potential	washwater, WDS water	Electrophoretic light scattering, from 3.8 nm to 100 µm, –600 to +600 mV	Litesizer™ 500 Anton Paar
Concentrations of specific OPs	road dust, particles in washwater	GC-MS was used for analysing PAH-16, oxy-PAHs, aromatics and alkanes, phthalates, alkylphenols and -ethoxylates; HPLC for aldehydes	Dry matter: SS 028113/1 PAH-16, alkane and aromatic substances: commercial lab method modified after SPIMFAB US EPA PAHs (PAH-16): SS EN ISO 18287:2008 Phthalates: E DIN 19742 (2012-04) Phenols: DIN EN ISO 18857-2
Concentrations of specific OPs	washwater, WDS water	GC-MS was used for analysing PAH-16, oxy-PAHs, aromatics, alkanes and alkylphenols and -ethoxylates; HPLC-DAD for aldehydes	PAH-16; alkane and aromatic substances: commercial lab method modified after SPIMFAB US EPA PAHs (PAH-16): US EPA8270, CSN EN ISO 6468 Phthalates: DIN EN ISO 18856 Phenols: ISO 18857-2 ISO 11923:1997
TSS ^a and VSS ^b	washwater, WDS water, stormwater	Performed on triplicates	ISO 11923:1997
OM ^c	road dust	Loss on ignition: loss of dry mass at 550 °C, three replicates.	SS 02 81 13, CSN EN 12879, CSN 72 0103 and CSN 46 5735
DOC ^d	washwater	TOC analyser with IR detection	CSN EN 1484 and CSN 13370
Concentration of specific OPs	stormwater	GC-MS was used for analysing US EPA PAHs (PAH-16) and oil index	PAH-16: commercial lab method; oil index: SS-EN ISO 9377-2 modified.

^a TSS – Total Suspended Solids.

^b VSS – Volatile Suspended Solids.

^c OM – Organic Matter.

^d DOC – Dissolved Organic Matter.

shown as a relative proportion of different particle sizes in the sample. The instrument measures particles ranging from 0.3 nm to 10 µm. However, this instrument cannot identify or measure concentrations of particles in the samples. For this reason, the samples were also analysed with a Nanosight NS 300 track analysis instrument (Malvern Instruments Ltd.), with a blue laser of wavelength 488 nm. The diffraction from the blue laser is stronger for particles containing fluorescent compounds. The method combines actual time visualization, for identifying and tracking the core of each particle using a charge-coupled device camera, with laser light scattering microscopy. The information on scattering intensity of the particles moving under Brownian motion is registered using the NTA 3.2 Dev Build 3.2.16 software and is further converted to particle size using the Stokes-Einstein Equation, and the approximate concentration of the particle size in the sample is calculated. This instrument can quantify particle concentrations and measure particle distribution from 10 nm to 2 µm. A comparison of the NTA and DLS results was hindered because the particle detection principle for these techniques is different. Also, the results extracted from each of these instruments are provided in different physical quantity.

In most cases, the water samples were stored frozen in test tubes before analysis. Prior to analysis, the samples were defrosted, mixed by shaking and filtered through a cellulose acetate 0.45 µm filter. The samples for DLS with the Litesizer were placed in a disposable cuvette (~1 ml per sample). For the Nanosight, a syringe pump allowing analysis at a constant flow rate was used. Samples used for DLS with the Litesizer were analysed three times and a mean value was calculated.

The zeta potential, i.e. stability, of the particles in the washwater

and WDS samples was analysed by electrophoretic light scattering (ELS) using the Litesizer™ 500 (by Anton Paar). Zeta potential was determined to study the particles' ability to form irregular particle clusters, flocs, or aggregates. The instrument can quantify particles from 3.8 nm to 100 µm. The measurement angle was automatically set by the instrument to 15°, and the measuring range was –600 to +600 mV. The water samples were stored frozen in test tubes. Prior to analysis, samples were defrosted and mixed by shaking, then poured into an Omega cuvette with a volume of 350 µm.

Selected washwater, road dust and WDS samples were analysed for the presence of 16 specific US EPA PAHs (PAH-16), nine oxy-PAHs, 13 phthalates, nonylphenols, octylphenols and mono-triethoxylates, aldehydes C₁–C₅, aromatics C₈–C₃₅ five fractions + BTEX (benzene, toluene, ethylbenzenes and xylenes) and aliphatics (alkanes) C₅–C₃₅ six fractions. The analyses for the OPs were performed at commercial laboratories following standardised methods (Table 1). Both dry and wet road dust of different fractions – unsieved (total), < 250 µm, 125–63 µm, and < 63 µm – were analysed for OPs. To simplify the sieving process, the road dust was dried at room temperature or 105 °C before sieving. The stormwater was analysed for the presence of PAH-16 and aliphatic hydrocarbons. In the evaluation of the results, the PAHs have been divided according to their molecular weight as follows: PAH-L(low) = Σ naphthalene + acenaphthylene + acenaphthene; PAH-M(medium) = Σ fluorene + phenanthrene + anthracene + fluoranthene + pyrene; and PAH-H(high) = Σ benzo(a)anthracene + chrysene + benzo(b)fluoranthene + benzo(k)fluoranthene + benzo(a)pyrene + dibenz(a,h)anthracene + benzo(g,h,i)perylene + indeno(1,2,3-c,d)pyrene.

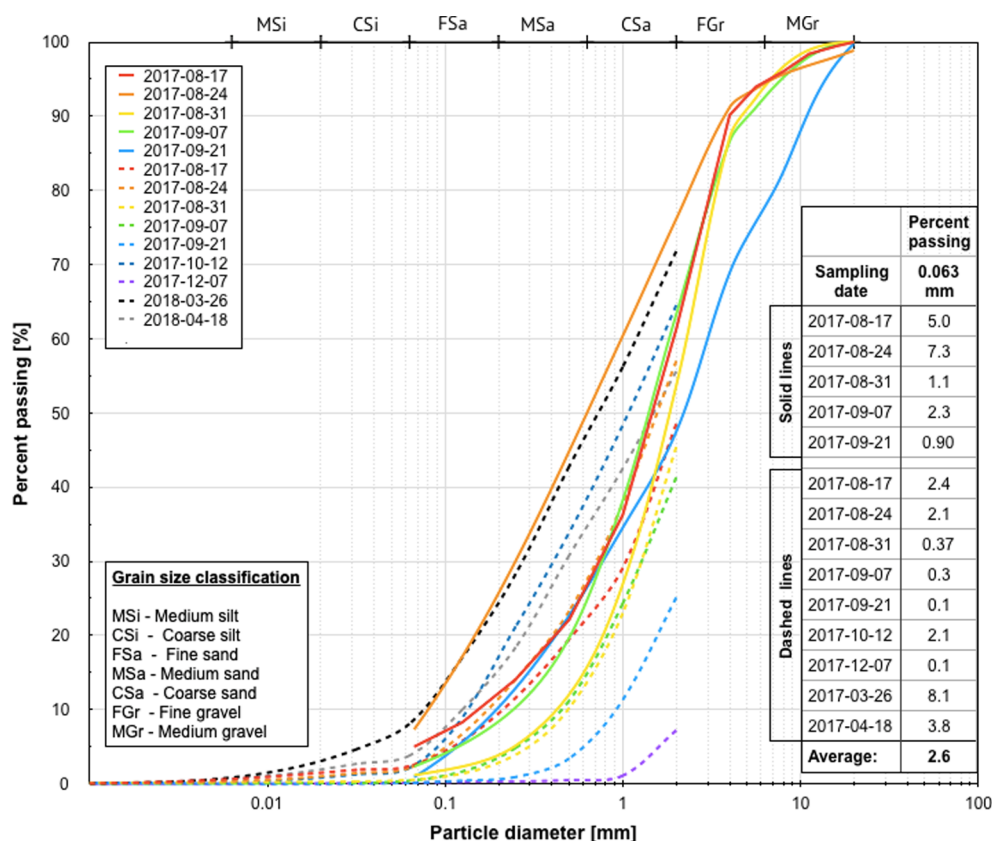


Fig. 2. Sieving curves of road dust sampled by the street sweeping machine during autumn 2017. Grain size classification according to ISO 14688-1:2002. Solid lines represent the size interval 0.063–20 mm. Dashed lines represent the size interval < 0.002–2 mm. Bottom-right: The percentage of fine fractions in analysed road dust samples.

3. Results and discussion

3.1. Particle size distribution in solid and liquid samples

3.1.1. Particle size distribution in road dust

According to the sieving curves (Fig. 2), the most abundant particle fraction in sampled road dust was sand of 600–2000 μm . The composition varied for the road dust samples from August 24, 2017 and September 7 and 21, 2017, which were analysed using two different standardized methods. This illustrates the uneven distribution of the road dust particles in the storage space of the sweeping machine. The road dust particles identified in this study were coarser than those previously reported, e.g. D10 values approximately equal to 170 μm (German, 2003); vacuum cleaner D10 values varying between 45 and 100 μm (Lau and Stenstrom, 2005), and D10 values 41–63 μm reported by Snilsberg and Gryteselv (2016). D10 is the diameter at which 10% of the sample's mass is comprised of particles with a diameter less than this value. The absence of OPs in the total dust samples can be explained by the proportional dominance (7–69%) of the sand fraction in these samples. Indeed, PAHs, aliphatic and aromatic hydrocarbons, as well as phthalates were quantified in higher concentrations in the finer dust fractions (Table S3, Supplementary data).

The fraction < 0.063 mm made up on average 2.6% of the total road dust samples (Fig. 2), similar to results by Kim et al. (2016) but lower than reported by the Norwegian Roads Administration 2016 (Snilsberg and Gryteselv, 2016), where the cumulative percentages of particles < 0.063 mm were 7.4–35%. The higher ratio of the larger particles collected by the sweeping machine in this study may be due to higher volumes of sand being used for winter street maintenance, lower efficiency of the sweeping machine, as well as its velocity, operational settings and design due to the Norwegian Public Roads Administration 2018 and the Swedish National Road and transport Research Institute 2016. Furthermore, the roughness and wetness of the pavement (Snilsberg et al., 2018, Abdel-Wahab et al., 2011), rotation frequency,

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.

3.1.2. Particle size distribution in washwater and stormwater

In this study, the volume weighting model was selected in terms of desired representation of the results (see also section 3.4 “strengths, limitations and uncertainties” for further discussion). The particle size, intensity weight distribution is presented in Fig. S3, Supplementary data. The results show that both washwater and stormwater contained nanoparticles 1–100 nm (Figs. 3, 4 and S3, Supplementary data). As nanoparticles are likely to pass through and affect biological membranes (Rossi et al., 2014), they have been demonstrated to exert toxicological effects (Forte et al., 2016), and have a proven ability to transport toxic substances, the risk of these particles should not be underestimated (Velzeboer et al., 2014).

There are differences in the analytical techniques used, where the Nanosight instrument measures particles from 10 nm up to 2 μm quantitatively, using a blue laser with stronger refraction from particles which contain molecules with fluorescence, such as organic molecules like PAHs. The Litesizer measures smaller particles, from 0.3 nm up to 10 μm , using a red laser and its measurements are only qualitative. This means that the heights of the peaks in Fig. 4 are relative only to the content in the analysed sample, and not comparable to concentrations or numbers in other samples. This also explains the differences between the PSD curves presented in Figs. 3 and 4. The results from the methods complement each other but are not directly comparable as described in chapter 2.3.

The finest fractions of the particles expected to be present in the washwater after collection by the street sweeping machine were in the same size range as those found in the sampled stormwater, and mainly in the size range of 100–300 nm (Fig. 3). Similar PSD have been observed in other traffic-related matrixes, e.g. stormwater (Nielsen et al., 2015) and water from car washing (Markiewicz et al., 2020), tyre and

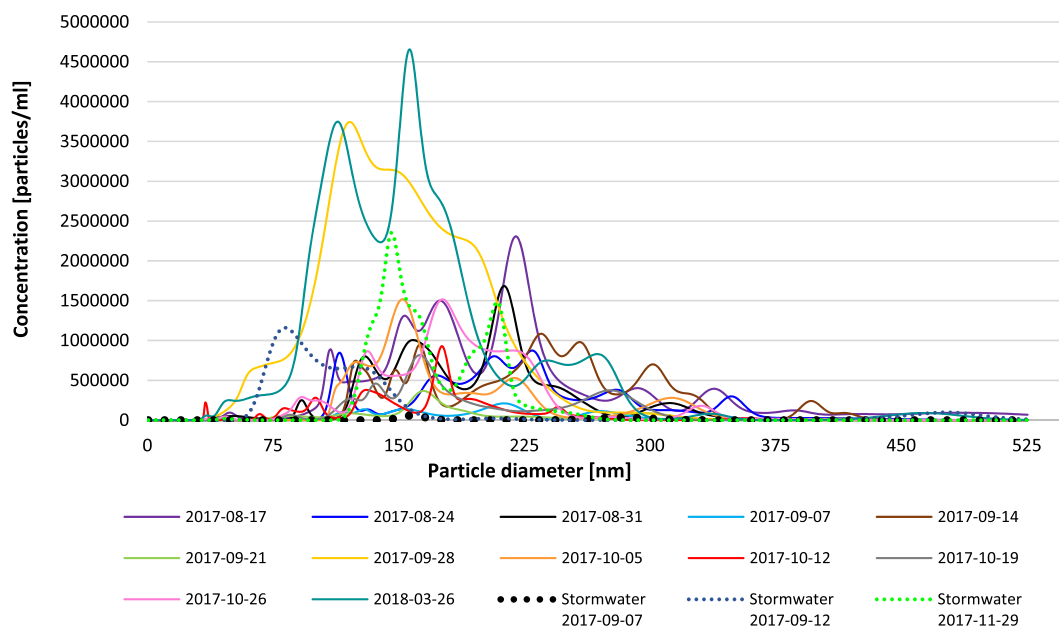


Fig. 3. Particle size distribution in wastewater sampled by the street sweeping machine on 12 occasions and in stormwater sampled on three occasions in the case study area. Analysed by nanoparticle tracking analysis (Nanosight).

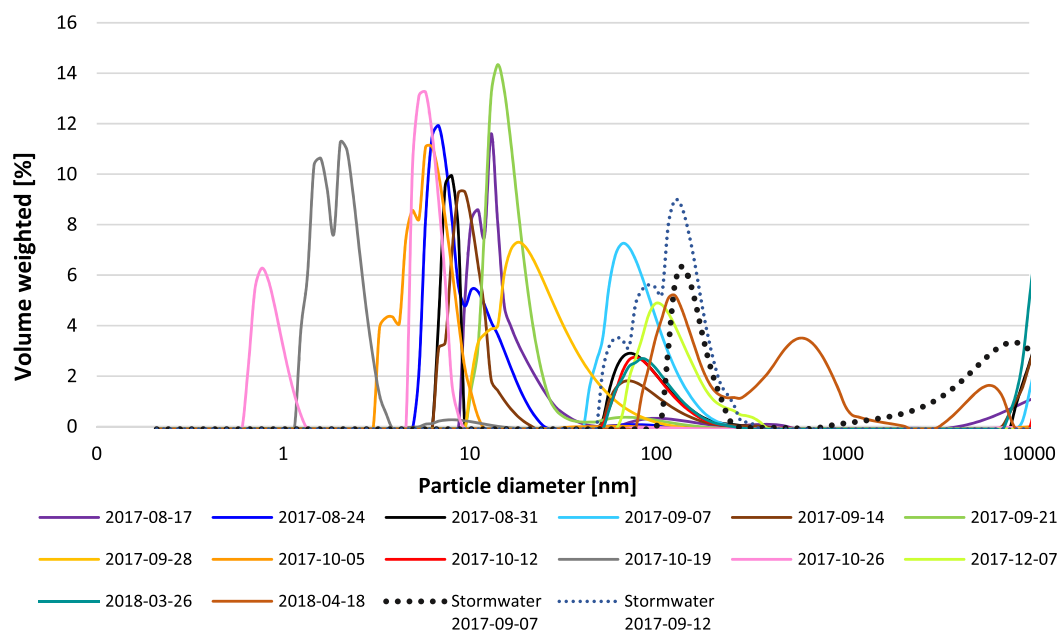


Fig. 4. Particle size distribution in wastewater sampled by the street sweeping machine on 14 occasions, and in stormwater sampled on two occasions in the case study area. Analysed by dynamic light scattering (Litesizer™500). The distribution is presented as volume weight % and the curves show the relative composition of the different fractions and not the amount of particles in the samples. (Notice that the x-axis has a logarithmic scale).

pavement wear (Gustafsson and Eriksson, 2015, Grigoratos et al., 2018, Mathissen et al., 2011), and brake wear (Puisney et al., 2018, Kukutschová et al., 2011, Wahlström et al., 2010).

Many of the samples (Fig. 4) contained relatively large percentages of nanoparticles smaller than 30 nm, and in further research studies it will be of high interest to try to identify the content of these extremely small particles. Particles larger than 0.45 μm were also detected in both wastewater and stormwater, although samples were passed through a 0.45 μm filter. This could be explained by particles e.g. fibers > 0.45 μm passing through the filter or by the instability of the samples, as their zeta-potentials varied between -1.5 and -23 mV (Table S6, Supplementary data), potentially causing particle agglomeration (Clogston and Patri, 2011), a process during which particles dispersed

in the liquid phase stick to each other and spontaneously form irregular particle clusters, flocs, or aggregates.

Several studies have investigated in detail the effect of precipitation on transport of particles of different sizes from road surfaces (Murakami et al., 2004, Zhao and Li, 2013, Egodawatta et al., 2007). The general conclusion is that more intense rain events have a greater ability to mobilize coarser road dust particles than less intense rain. However, Egodawatta et al. (2007) suggested that mobilization of fine particles also increases with more intense rainfall because less intense rain events cannot mobilize high-density fine particles. In this study too few stormwater samples were studied to confirm this tendency, however Fig. 3 shows that low intensity precipitation of 14.3 L/(s*ha) on 2017-09-07 (Table S1) resulted in a low concentration of particles in

the stormwater sample. A few days later, on 2017-09-12 an intense rainfall of 160 L/(s*ha) resulted in a considerable increase in the particle concentration. Moreover, Fig. 4 shows occurrence of slightly smaller particles down to ~70 nm after the intense precipitation on 2017-09-12 than after the light rain on 2017-09-07 down to ~110 nm, which suggest higher mobilization of fine particles of high density.

3.2. Concentrations of OPs in street sweeping road dust, washwater and stormwater

Data on the organic pollutants analysed in washwater and road dust samples are presented in Table 2, and concentrations of all the ~70 specific compounds are available in Tables S2 and S3, Supplementary data. The compound groups generally found in the highest concentrations in all matrices were aliphatics C_5 – C_{35} > phthalates > aromatics C_8 – C_{35} > PAH-16. Aldehydes C_1 – C_5 , oxy-PAHs, alkylphenols and their mono-triethoxylates were only quantified in a few samples. The highest quantification frequency (QF), as well as the highest concentrations of PAHs, aliphatic and aromatic petroleum hydrocarbons in washwater, were found in the samples collected in March and April 2018, after a period of more than three months when no street sweeping was carried out, see Table S2, Supplementary data. In these samples, the concentrations of OPs were at least one magnitude higher than in the other samples, and the concentrations of PAHs and aliphatics in the washwater were extremely high. The lowest QF and concentrations of OPs were detected in the two washwater samples collected midway through the sweeping campaign in September and October. The QF and concentrations of OPs did not appear to be related to DOC or TSS; the sample from April 2018 with extreme OP concentrations exhibited comparably low DOC and TSS concentrations (Fig. S2, Supplementary data).

It is evident that higher concentrations of PAH-16 and aliphatic and aromatic petroleum hydrocarbons were quantified in washwater samples from the first sweeping event in the spring (2018-03-26), compared to the first sampling occasion of the study (2017-08-17) (Table S2). However, it is not evident which factor(s) that would have the largest effect on pollutant abundance in collected matrices. Time between sweeping events (one month for August 2017, three months for March 2018) is only one of the factors affecting the build-up of particles and pollutants on road surfaces. Others are precipitation, winter road maintenance actions including snow removal, deicing and sanding, and seasonal emission variations. Increased emissions of vehicle-related pollutants during winter conditions could be due to increased road wear (including bitumen) caused by studded tyres (Gustafsson et al., 2019) or lower ambient air temperatures and cold-start conditions, which have been shown to cause higher PAH emissions for diesel vehicles (Alves et al., 2015), as well as higher PAH and oxy-PAH emissions for ethanol/gasoline fueled vehicles (Ahmed et al., 2018).

In general, more aliphatic and aromatic petroleum hydrocarbons and PAHs were quantified in washwater than in road dust samples (Table 2). The only exception was certain phthalates (DnBP, DnOP, DCHP and DINP), which were sporadically quantified in sieved road dust samples (Table S3, Supplementary data). In the non-sieved road dust samples, only aliphatics > C_{16} – C_{35} and DEHP were quantified. The only alkylphenol quantified, i.e. 4-*tert*-octylphenol, was found in one of the road dust samples. The organic pollutants analysed in the road dust samples did not show a strong time trend, unlike in the washwater samples, where OP concentrations were exceptionally high after the winter sweeping break.

Non-sieved samples contained less organic matter (OM) than sieved samples (< 2.2% vs. > 3.3%), see Loss on ignition Table S3, Supplementary data. The particles (> 0.7 μ m) in washwater collected on the 26 March 2018 contained 13% OM; more than the corresponding sieved road dust sample with size 63–125 μ m and 3.3% OM. In addition, concentrations of total PAHs were almost three times higher in the particles in washwater than in the corresponding road dust particles.

These findings were expected, as hydrophobic organic compounds are preferentially sorbed to particles with organic content, and smaller particles can sorb more pollutants per weight unit than large particles (Herngren et al., 2010, Dong and Lee, 2009, Sansalone and Buchberger, 1997).

Our results for the washwater samples (Table S2, Supplementary data) suggest that many of the hydrophobic compounds quantified, e.g. aliphatic hydrocarbons > C_{16} – C_{35} $\log K_{ow}$ > 8.9, DEHP $\log K_{ow}$ = 7.5; PAH-16 $\log K_{ow}$ 4.3–6.8, aromatic hydrocarbons > C_{16} – C_{35} $\log K_{ow}$ > 5.9, are either attached to nano- or microparticles or truly dissolved. Previous findings have shown that phthalates, PAHs and alkylphenols that all are hydrophobic by definition can be found in both the colloidal (< 1 μ m) and the truly dissolved phases of urban runoff (Kalmykova et al., 2013, Nielsen et al., 2015). In addition, research has shown that under high turbulence, a mix of hydrophobic organic pollutants, diesel and colloids has the potential to form nano- and micro-sized emulsions (Markiewicz et al., 2019), and the highest concentrations of nanoparticles < 100 nm were found in the mixture of all OPs without colloids added. Street sweeping produces the same conditions, which suggests that emulsions can form in the washwater of the sweeping machine.

The concentrations of PAHs and DEHP in washwater repeatedly exceeded the Canadian Environmental Quality Standards (CEQS) for the protection of Aquatic Life and the Environmental Quality Standards (EQS) formulated by the European Commission. No standards are available for the rest of the detected OPs in this study. In general, PAHs and DEHP water quality guidelines were exceeded in all samples where they were quantified, i.e. not only in the severely polluted washwater samples collected in March and April. As an example, benzo[a]pyrene exceeded the CEQS in all samples; in one sample the concentration was 400 times higher than the CEQS. The Swedish guidelines for less sensitive land-use (e.g. offices, roads, industries) were exceeded for aliphatics > C_{16} – C_{35} in all road dust samples. (Swedish EPA, 2009). In addition, the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life were exceeded for several PAHs (e.g. fluoranthene and pyrene) in three of the road dust samples. The findings call for proper management of street sweeping waste. For regions where sweeping cannot be performed during winter, which may result in highly polluted washwater in spring, the road administrations could prioritize treatment of washwater from the first sweeping events in spring. As the larger dust fractions contained lower concentrations of OPs, these may be most suitable for reuse. It is imperative to develop innovative and sustainable methods to treat the polluted waste from the street sweeping machine, as further research under publication from our group has shown that these materials besides OPs also contain metals and microplastics.

3.2.1. Aliphatic and aromatic petroleum hydrocarbons

Short-chain aliphatic hydrocarbons < C_{10} and aromatic BTEX compounds were generally not quantified in the washwater samples (Table S2, Supplementary data). Among the aliphatics, the long-chain (> C_{16} – C_{35}) compounds were measured in high concentrations compared to other compounds; 190–53,000 μ g/L (Table S2). These compounds are not often measured in stormwater, but it is not unusual to find total petroleum hydrocarbon concentrations exceeding 1,000 μ g/L in road runoff (Flanagan et al., 2018, Snilsberg and Gryteselv, 2016, Moilleron et al., 2002). The concentrations of aliphatic petroleum hydrocarbons in the four stormwater samples taken during the weekly street sweeping autumn 2017 did not vary much and were in the range from 200 till 300 μ g/L (Table S4). Similar to the long-chain aliphatics, the largest aromatic compounds (> C_{16} – C_{35}) were found in higher concentrations than aromatic compounds of lower molecular weight. Generally, the shorter aliphatics and aromatics are found in gasoline (petrol), whereas diesel contains more of the larger aliphatic compounds (Yang et al., 2017; Heath et al., 1993). The comparably low concentrations of the shorter aliphatics and aromatics in both the

Table 2
Summary of analysed organic pollutants (quantification frequency; minimum, median and maximum concentration of analysed compounds) in wastewater, filtered particles (> 0.7 µm) in wastewater, and in road dust collected by the street sweeping machine.

Organic pollutants	Wastewater						Particles in wastewater retained on 0.7 µm filter						Road dust					
	Number of analysed samples	Quantified substances/fractions	Conc. for a specific substance/fraction µg/L			Number of analysed samples	Quantified substances/fractions	Conc. for a specific substance/fraction mg/kg			Number of analysed samples	Quantified substances/fractions	Conc. for a specific substance/fraction mg/kg					
			Min	Median	Max.			Min.	Median	Max.			Min.	Median	Max.			
Polycyclic aromatic hydrocarbons PAH-16	5	8-16	0.010	0.42	24	1	11	0.083	0.14	0.83	14	0-12	0.012	0.14	0.35			
Oxygenated-PAHs 9 compounds	2	5	0.11	0.17	0.30	n.a. ^b	-	-	-	-	2	0	-	-	-			
Phthalates 13 compounds	5	0-2	1.2	16	150	1	0	-	-	-	11	0-4	0.052	0.60	12			
Aliphatics C ₈ - C ₃₅ 6 fractions	5	4-5	1.2	96	53,000	1	1	950	950	950	11	0-1	68	200	780			
Aromatics C ₈ - C ₃₅ 5 fractions + BTEX ^d	5	1-5	0.060	10	82	1	3	1.1	1.3	1.8	11	0	-	-	-			
Aldehydes C ₁ - C ₅ five compounds	1	0	-	-	-	n.a. ^b	-	-	-	-	2	4	0.26	1.2	7.6			
Nonylphenols, octylphenols and -(mono-tri)ethoxylates 8 compounds	1	0	-	-	-	n.a. ^b	-	-	-	-	3	1	0.011	0.013	0.15			

^a PYR = pyrene, the PAH in highest concentration.
^b n.a. = not analysed.
^c DINP = di-iso-nonylphthalate.
^d BTEX = benzene, toluene, ethylbenzenes, xylenes.
^e 4-tert-octylphenol.

washwater and road dust causes them to evaporate and become an air pollution problem. The $> C_{16}$ – C_{35} aliphatics were quantified at a considerably higher concentration (950 mg/kg) in the particulate phase of washwater than $> C_{16}$ – C_{35} aromatics (1.8 mg/kg) and were also the only petroleum hydrocarbons quantified in road dust (Table S3). The aliphatics have lower water solubility and volatility than the aromatics with the same number of carbons, which explains the aliphatics' more frequent occurrence in the solid phases (Heath et al., 1993). Several hydrocarbons were quantified in the particulate phase of washwater, which suggests that these compounds are principally attached to particles smaller than 63 μm with a high OM content.

3.2.2. PAHs and their relative composition

The PAHs were quantified in all total (unsieved) road dust samples ($n = 3$), four of the sieved road dust samples ($n = 8$, $< 250 \mu\text{m}$, 63–125 μm , and $< 63 \mu\text{m}$) and in all washwater ($n = 5$), stormwater ($n = 5$) and WDS ($n = 4$) samples (Tables S2–S4 and S7 Supplementary data). In the washwater samples collected in March and April, the PAH-16 concentrations were very high, reaching 90 and 120 $\mu\text{g/L}$, respectively. In the WDS samples from March and April, the concentrations of PAH-16 were much lower and varied between 1.3 and 3.0 $\mu\text{g/L}$. These lower concentrations may be explained by the smaller amount of road dust collected with the WDS than with the sweeping machine, which collected road dust from a larger road surface in the study area. The event mean concentrations (EMCs) of PAH-16 analysed in stormwater during the street sweeping campaign during the autumn were 0.33–0.77 $\mu\text{g/L}$ (Table S4), and in the same order of magnitude as the EMCs reported for stormwater in other studies (Hou and Li, 2018, Zgheib et al., 2012, Pettersson et al., 2005).

In road dust, PAH-M and PAH-H were quantified in the total fractions and in the samples with particle fractions $< 250 \mu\text{m}$. The PAH concentrations in road dust (Table S3) were lower than the concentrations previously detected in a road sedimentation facility for highway runoff (AADT approximately 100,000 vehicles), located in Gothenburg (maximum PAH-16 = 9.6 mg/kg (Markiewicz et al., 2017)); dust collected from roads with similar traffic as those in the current study (PAH-16 = 16–23 mg/kg (Lorenzi et al., 2011)); and several times lower than concentrations measured in road dust collected in Ulsan, Korea (e.g. residential area PAH-16 = 46 mg/kg (Dong and Lee, 2009)). The comparatively low concentrations of PAH in road dust in the current study is likely due to a combination of limited traffic impact (AADT $< 24,000$), low OM content and coarse particles in the collected road dust, but also related to the finest fraction of particles from the collected road dust that is suspended in the street sweeping machine washwater. The highest PAH-16 concentration (1.3 mg/kg) was found for particles in washwater, which also had a high OM content (Table S3).

The relative composition of PAHs varied in the road dust samples (Fig. 5); these differences may be explained by sample pre-treatment (analysis of dry/wet sample), seasonal variations and differences in content of OM and particle size. In the samples from March 2018, total fraction and 63–125 μm (both with high concentrations), the PAHs occurred in the following order of concentration: pyrene $>$ fluoranthene $>$ benzo(b)fluoranthene \approx phenanthrene \approx benzo(ghi)perylene. This PAH composition indicates tyre wear, vehicle exhausts, brake linings, motor lubricant oils, and road surface wear as possible sources, based on the calculated emission factors for these specific PAHs from different traffic and road sources (Zhang et al., 2019, Markiewicz et al., 2017). The relative composition of the PAHs in road dust for the autumn samples 2017 is slightly different from previous studies of sediment from highway runoff, collected in Gothenburg in 2003 and 2015, because of relatively lower percentage of phenanthrene and relatively higher of PAH-H. Phenanthrene is however, in relatively higher percentage in the samples 2018 after the winter sweeping break. The dried fraction of collected road dust, 63–125 μm March 2018 and the $< 63 \mu\text{m}$ April 2018, contained a much lower percentage of

chrysene, compared to the samples with more coarser fractions (Fig. 5). Chrysene $>$ pyrene $>$ fluoranthene $>$ benzo(ghi)perylene are PAHs that may be an indication of PAHs from tyres (Markiewicz et al., 2017), which suggests that tyre particles are found in all road dust particle fractions. Tyres are expected to become a less important source of PAHs over time, as the use of HA oils in tyres is now restricted in Europe.

In the five washwater samples, which also contained the smallest particles from the road dust and a high VSS content (Table S2 Supplementary data), the relative composition of PAHs is approximately the same as in road dust: pyrene $>$ fluoranthene $>$ benzo(b)fluoranthene $>$ phenanthrene $>$ benzo(ghi)perylene (Fig. 5). In the autumn samples, collected during the weekly street sweeping (on September 28 and October 26, 2017), the relative percentage of PAH-L and some PAH-M was higher. In samples collected after the winter sweeping break, a clear trend of higher PAH-H percentage was observed, which correlates well with K_{ow} of PAHs, the high concentrations of DOC, TSS and VSS found in the sample collected on March 26, 2018 (Table S2). Surprisingly, the separated washwater particles ($> 0.7 \mu\text{m}$) from March 26, 2018 contained relatively higher percentage of PAH-L than the corresponding washwater sample. In the stormwater samples, the relative composition of PAH-L was high, which was to be expected, as these are the most water-soluble PAHs and therefore can be expected to be the most easily transported by road runoff. The relative composition of PAH-16 in the WDS samples was uniform, and no differences in the composition before and after sweeping of the street surface could be identified. These samples contained relatively low percentage of PAH-L which may be explained a possible evaporation during the high-pressure sampling procedure.

3.2.3. Oxygenated PAHs in washwater

Oxygenated polycyclic aromatic hydrocarbons (oxy-PAHs) may be directly emitted from vehicle exhausts (Keyte et al., 2016, Bandowe et al., 2014) or found as a degradation product of PAHs (Zhu et al., 2018, Lundstedt et al., 2007). Oxy-PAHs were quantified in one washwater sample (Table S2). In Table S5 in the Supplementary data, the concentrations of the quantified oxy-PAHs, i.e. 7H-benz(de)anthracene-7-one, benzo(a)anthracene-7,12-dione, 6H-benzo(cd)pyren-6-one, benzo(a)fluorenone and naphthacene-5,12-dione, are presented together with the corresponding PAH. The calculated ratio of quantified oxy-PAH/PAH-16 is very high compared to previous results from contaminated soil (Lundstedt et al., 2007), which indicates that oxy-PAH concentrations in water samples may be high even where the concentrations of PAH-16 are low. No other study analysing oxy-PAHs in urban water has been found in the literature. The occurrence of oxy-PAHs in washwater is alarming, as the toxicity of oxy-PAHs is much higher than that of corresponding PAHs; oxy-PAHs also have a higher water solubility, and thereby a higher mobility in the environment (Lundstedt et al., 2007). As only a few samples were analysed for oxy-PAHs in this study, more studies are needed on their occurrence in road dust and runoff.

3.2.4. Phthalates, aldehydes and alkylphenols

Only three phthalates were detected in the washwater: DBP ($n = 1$; 1.2 $\mu\text{g/L}$); DEHP ($n = 4$; 1.7–22 $\mu\text{g/L}$); DINP ($n = 1$; 150 $\mu\text{g/L}$) (Table S2, Supplementary data). The quantification limit for DINP fluctuated between 11 and 230 $\mu\text{g/L}$, and it cannot be ruled out that DINP was present in other samples although not quantified. The same three phthalates were detected in road dust samples: DBP ($n = 3$; 0.052–0.17 mg/kg); DEHP ($n = 5$; 0.21–1.4 mg/kg); DINP ($n = 2$; 3.3–12 mg/kg) (Table S3). In the past century, large amounts of DBP and DEHP were used as plasticizers in e.g. PVC, paint, sealant and car care products, and the compounds are commonly found in urban runoff and road dust (Flanagan et al., 2019, Škrbić et al., 2016, Björklund et al., 2009). The detected concentrations of DEHP and DINP in the road dust samples were generally lower than those previously reported in road dust and street-side dust by Björklund et al. (2009) (DEHP:

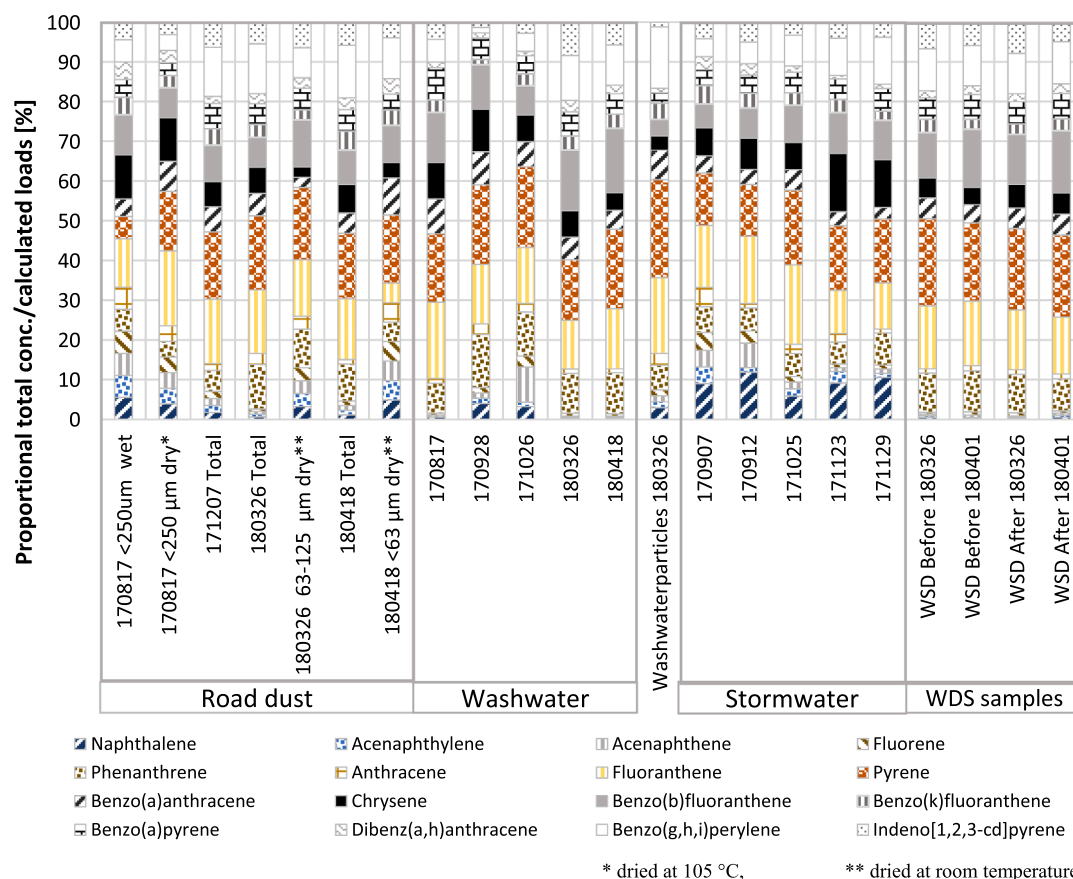


Fig. 5. Proportion (%) of specific PAHs to total PAH concentrations measured in road dust and washwater sampled by the street sweeping machine, in stormwater, and in Wet Dust Sampler (WDS) samples.

2.8–48 mg/kg; DINP: < 5.0–200 mg/kg) and Škrbić et al. (2016) (DEHP: 1.22 mg/kg). The DBP concentrations were in the same order of magnitude as reported by Björklund et al. (< 0.10–0.40 mg/kg) and Škrbić et al. (2016) (0.071 mg/kg).

Aldehydes and alkylphenols were only analysed in a few washwater and road dust samples (Tables S2 and S3 in Supplementary data). Among the alkylphenols, 4-*tert*-octylphenol was quantified in road dust. This compound is used in the manufacture of tyres, paint and coatings (Månsson et al., 2008). Among the aldehydes, formaldehyde and acetaldehyde were most frequently quantified and had the highest concentrations in road dust samples (1.2–5.1 and 1.1–7.6 mg/kg, respectively). Formaldehyde, acetaldehyde and benzaldehyde are compounds known to be emitted from gasoline vehicles (Sigsby et al., 1987). Aldehydes may also be emitted from ethanol and methanol driven vehicles, or may be photochemical degradation products from vehicle hydrocarbon emissions (Gramsch et al., 2018).

3.3. Evaluation of street sweeping by WDS and stormwater sampling

To investigate the effects of street sweeping on recipient water quality, stormwater was collected during two sampling campaigns in the catchment area: one where no measures were in place (autumn 2016, five storm events) and another when sweeping was performed every two weeks (autumn 2017, five storm events). Generally, the mean concentration of PAH-16 in stormwater decreased from 1.6 µg/L for storm events when no measures were in place, to 0.51 µg/L for events with sweeping (Table S4, Supplementary data). However, a Mann-Whitney test indicates that there is no statistical difference in PAH concentrations ($U = 73.5$; $p = 0.106$) and oil ($U = 10.0$; $p = 0.589$) in stormwater collected during sweeping and no-sweeping periods. The measured stormwater quality can only give an indication of the effect of

street sweeping, as the two stormwater sampling periods differ in additional factors that influence pollutant supply and mobility, e.g. traffic count, road maintenance, wind and precipitation. In this case, precipitation depth and intensity (Table S1) could not be used to explain differences in PAH concentrations in stormwater as no statistically significant correlations were found (mean PAH concentration and rain depth: Spearman's $\rho = 0.642$, $P = 0.120$; mean PAH concentration and rain depth: Spearman's $\rho = 0.157$, $P = 0.711$). However, as not all storm events were sampled and measured during fall 2016 (no sweeping) and 2017 (sweeping), the true effect of precipitation on stormwater quality is not known. For example, in 2017 the daily precipitation frequency (70%) as well as the total (342 mm over 92 days) and daily average (3.72 mm) precipitation were higher than during the no-sweeping period (50% daily precipitation frequency; total 311 mm over 138 days; daily average 2.25 mm) in 2016. It could be speculated that more rain in the fall of 2017, together with street sweeping, lead to lower PAH concentrations in stormwater compared to the fall of 2016. Previous studies (Selbig and Bannerman, 2007, Rochfort et al., 2009) on street sweeping have established the positive effects of street sweeping in reducing the amount of road dust on road surfaces, but due to the variability in stormwater characteristics and the difficulty in achieving a control set of data, the effects of sweeping on stormwater quality is not as evident.

The WDS was used on two occasions, to compare the concentrations of OPs on the road surface before and after sweeping took place (on each occasion, one composite “before” and one “after” sample was analysed). On 26 March 2018, concentrations of TSS, DEHP, aliphatics > C₁₆–C₃₅ and all detected PAHs were higher in the WDS water collected after street sweeping (Table S7). On the contrary, street sweeping led to lower concentrations of all OPs and TSS in WDS samples collected on the 18 April 2018. When the OP concentrations were

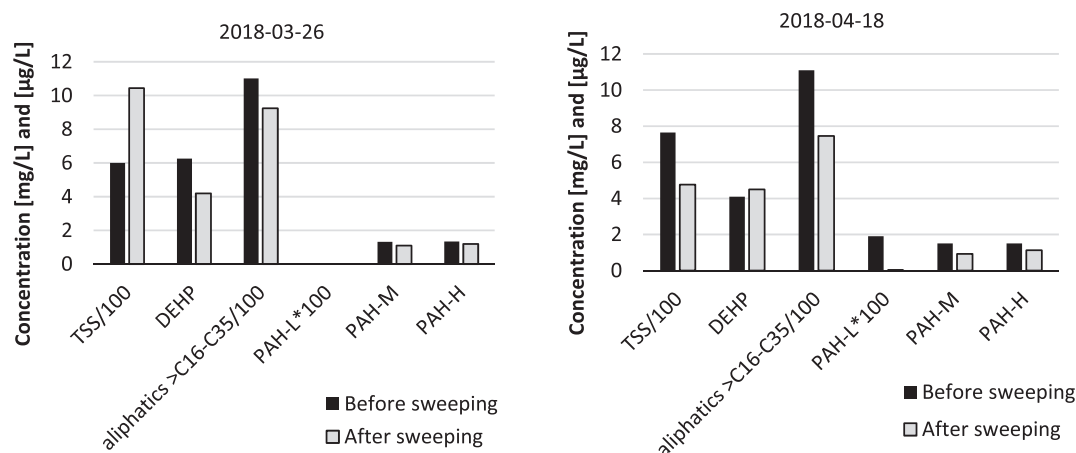


Fig. 6. The concentrations of TSS [mg/L] and OPs [$\mu\text{g/L}$] in WDS samples collected before and after street sweeping 2018-03-26 (left) and 2018-04-18 (right). Concentrations of OPs have been normalized towards the TSS concentrations in the samples.

normalized for the TSS concentrations – assuming that OPs are attached to particles in the WDS samples – the concentrations after street sweeping were lower on both occasions (Fig. 6), although this difference is not statistically significant (see [Supplementary Material](#) for details). These data suggest that street sweeping has a positive effect on the OP loads on road surfaces because all OPs were then lower after sweeping. However, the results also suggest that the brushing motion of street sweeping enables particles fixed to the road surface to become more mobile, as the concentrations of TSS and OPs were still high after sweeping. This effect of street sweeping has been observed by others (Snilsberg et al., 2018, Järleskog et al., 2017, German, 2003, Vaze and Chiew, 2002). It is expected to be considerable in spring: a long break in sweeping combined with winter road conditions, including application of de-icing salt, which is also dust-binding (Denby et al., 2016), and humidity may lead to accumulation of a large fixed load on the road surface. This fixed material, which is greatest near the kerb (Gustafsson et al., 2019) is disturbed by the sweeper's brushes and particles and attached pollutants are then mobilised through suspension or with road runoff.

Furthermore, the before and after samples, collected with the WDS, were tested for significant differences in mean (based on five replicates) particle concentration (particles/mL), D10, D50 and D90 (nm) using the Mann Whitney U test. Although particle concentrations and D50 were higher in the “before” samples, no significant differences in concentrations or sizes were found for the samples collected in 2018-03-26 ($U_{concentration} = 7.0$, $p_{concentration} = 0.251$; $U_{D10} = 11.0$, $p_{D10} = 0.754$; $U_{D50} = 9.0$, $p_{D50} = 0.465$; $U_{D90} = 8.0$, $p_{D90} = 0.347$) (Fig. 7). However, as seen in Fig. 7, the “before” sample collected in 2018-04-18 contained significantly higher concentrations, D50 and D90 than the “after” sample ($U_{concentration} = 0.0$, $p_{concentration} = 0.014$; $U_{D10} = 4.0$, $p_{D10} = 0.142$; $U_{D50} = 0.0$, $p_{D50} = 0.014$; $U_{D90} = 1.0$, $p_{D90} = 0.027$). These results indicate that street sweeping results in fewer and smaller particles remaining on the road surface.

However, the long-term benefits of frequent street sweeping on the quality of stormwater should not only be determined by examining the efficiency of the sweeping machine and the quality of stormwater samples. As Bartolozzi et al. (2018) note, research of the overall environmental impact of street sweeping is lacking and no authors have studied the service using a life cycle assessment (LCA) approach or cost-benefit analysis. This study has demonstrated both the occurrence of nanoparticles in the material collected by the sweeping machine and high concentrations of OPs associated with fine road dust particles. Hence, future studies are recommended to investigate the quantity of nanoparticles in the total road dust load collected by the sweeping machine and the proportion of nanoparticle in the washwater. This data is crucial since an LCA of street sweeping should include information

about the treatment of the waste material collected in the sweeping machine, including extraction and treatment of nanoparticles.

3.4. Strengths, limitations and uncertainties

The occurrence of nanoparticle in road dust in this study is confirmed by two different instruments using DLS and NTA. Both techniques are valuable and commonly used tools that differ in the detection principle, which results in differences in reported size distribution; intensity and number weight for DLS and NTA, respectively (Kim et al., 2019), see also Chapter 2.3. In addition, the volume of the analyzed sample is only known for NTA and can give approximate information about the particle concentration, which can be considered an advantage over the DLS technique, if quantitative data is needed. The strength of the DLS technique is that it is less prone to errors due to the large amount of data obtained from each measurement, and it requires less system configurations than NTA (e.g. screen gain, focus, detection threshold) that can alter the results. The DLS technique is on the other hand sensitive to the presence of large particles, which can hinder an accurate size determination when a majority of particles are of distinctly smaller sizes (Filipe et al., 2010). Since the NTA technique measures the movement of individual particles, it is less sensitive to the presence of larger particles and the results are likely to be more reliable than from DLS. However, this study shows that DLS was more efficient than NTA in the detection of the smallest particles, which could be related to different detection limits and laser wavelength used. Therefore, these techniques complement each other and using both techniques is a strength of this study.

The uncertainty with using the results from DLS in the weighing model other than intensity i.e. volume and number is that converting the data from intensity weight to volume or number weight requires assumptions about physical properties of the particles (e.g. shape and density). Moreover, the results converted to number weight require only good quality data and any noise during the measurement can give unreliable results (Nobmann, 2017). Data presented as volume weighted can easily be affected by larger particles, fibers or dust occurring in the sample (Filipe et al., 2010). For the purpose of this study, it was sufficient to show the occurrence of the particles in the smallest sizes (0.3–450 nm) and denote the presence and quantity of agglomeration; therefore, volume weight was considered suitable for presenting the results.

Characterization of the larger particles collected by the street sweeping machine also exhibited uncertainties. Since precipitation and wind pattern can vary, the size and quantity of road dust particles collected by the sweeping machine may differ. Data from more sweeping occasions would be needed to establish the characteristics of

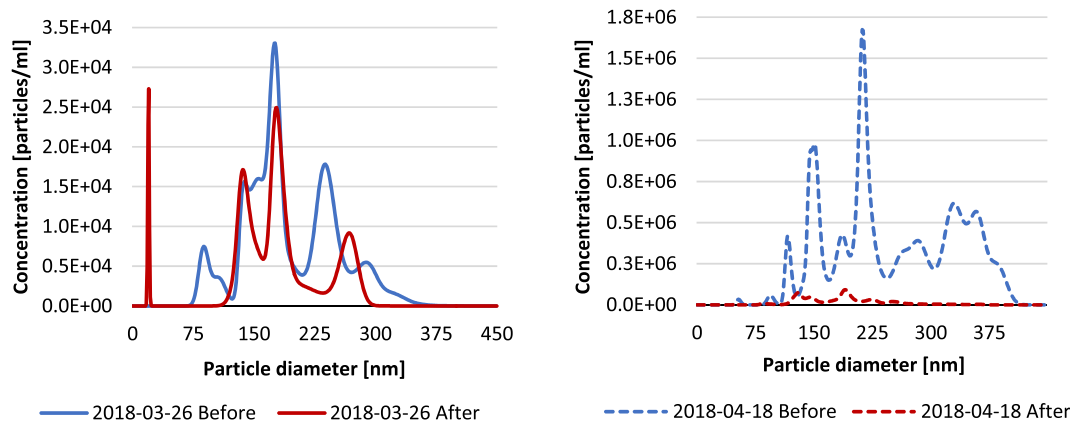


Fig. 7. Particle size distribution, measured using Nanosight in water samples collected with the WDS.

material collected by street sweeping during different seasons. Additionally, characterizing wash-off of particles and pollutants during precipitation and build-up in dry periods, with applied watershed/mass balance modeling, may potentially give information about material available for removal by street sweeping.

The limitations of the analyzed stormwater samples are presented and discussed in 3.3. The small number of subsamples collected with the WDS and pooled together is an important limitation since these results only describe an average dust load on the kerb, in wheel tracks, and between wheel tracks across the left lane. Since the number of sampling occasions for the WDS was limited the presented results should, as pointed out in 3.3, only be considered as an indication of the positive effect of street sweeping on the OP load on road surfaces.

4. Conclusions

Based on the hypotheses of this study, the following conclusions were drawn:

(1) **nano- and microparticles and OPs are collected effectively by the street sweeping machine**

The street sweeping machine collected a considerable amount of fine particles and associated pollutants. The measured order of concentrations for the organic pollutants found in all matrices was: aliphatics > phthalates > aromatics > PAH-16. The concentrations of long chain C₁₆–C₃₅ aliphatics and PAH-16 in washwater were extremely high, $\leq 53\,000\ \mu\text{g/L}$ and $\leq 120\ \mu\text{g/L}$ respectively, compared to results from other studies and water quality guidelines. Phthalates, such as DBP, DEHP and DINP, were detected in both washwater and road dust, and toxic oxy-PAHs were identified in one washwater sample. The highest concentrations of aliphatics and PAHs were found in March and April, i.e. after the 3-month long break in sweeping during winter.

This is the first study where nanoparticles in sizes down to 1 nm have been identified in washwater from street sweeping. The washwater contained nanoparticles, with the highest presence in the size range 25–300 nm. This indicates that OPs and nano- and microparticles are collected effectively by the street sweeping machine, thereby confirming hypothesis 1.

(2) **transport of nano- and microparticles and OPs to receiving water decreases after street sweeping**

For all measured pollutants (aliphatic petroleum hydrocarbons and PAHs), the mean concentrations in stormwater were lower during the sweeping campaign. The study also indicates that many of the hydrophobic compounds quantified in washwater are attached to small

particles or truly dissolved. Therefore, transport of nano- and micro-particles and OPs to receiving waters may be prevented by street sweeping, which partly confirms hypothesis 2. However, as nano-particles in washwater exhibited agglomeration behaviour, further investigation of the stability of stormwater and washwater particles is needed.

(3) **the waste material collected inside the street sweeping machine contains high concentrations of nano- and microparticles and OPs, hence the material requires treatment before it is disposed or reused.**

The waste material collected inside the street sweeping machine contained high concentrations of OPs and nanoparticles in sizes from below 1 nm up to 300 nm, hence the material requires treatment before it is disposed or reused, confirming hypothesis 3. Treatment of the contaminated materials is a complex issue as washwater may contain extreme concentrations of OPs and fine particles, and the pollutant abundance in the solid waste is often dependent on particle size. It is currently not well investigated how these materials can be decontaminated and prepared for reuse. Treatment methods require additional research, hence are beyond the scope of this study.

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 material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2019.105337>.

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