Emissions of organic pollutants from traffic and roads: Priority pollutants selection and substance flow analysis

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HIGHLIGHTS

- Methodology to select organic priority pollutants for stormwater treatment
- Sources of organic pollutant emissions in a traffic environment were identified.
- The loads of PAH emitted in the road catchment area were estimated by means of SFA.
- 2–6% of the total PAHs emitted reached road runoff.
- Calculated quantities and measured PAH and oxy-PAH loads show large differences.

GRAPHICAL ABSTRACT
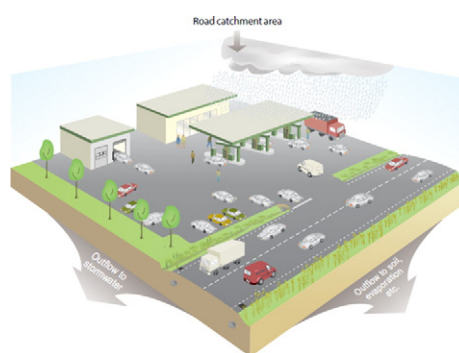
ABSTRACT

A large number of organic pollutants (OPs) emitted from vehicles and traffic-related activities exhibit environmental persistence and a tendency to bioaccumulate, and may have detrimental long-term effects on aquatic life. The aim of the study was to establish a list of significant sources of OPs occurring in road runoff, identify the OPs emitted from these sources, select a number of priority pollutants (PP), and estimate the quantity of PPs emitted in a road environment case study using substance flow analysis (SFA). The priority pollutants included in the SFA were selected from a list of approximately 1100 compounds found after comprehensive screening, including literature and database searches, expert judgments, the Ranking and Identification of Chemical Hazards method, and chemical analysis of sediments. The results showed the following priority order: polycyclic aromatic hydrocarbons (PAHs) > alkanes C_{20}–C_{40} > alkylphenols > phthalates > aldehydes > phenolic antioxidants > bisphenol A > oxygenated-PAHs > naphtha C_5–C_{12} > amides > amines. Among these, PAHs were chosen for a SFA, which was performed for a highway case study area in Gothenburg (Sweden). The SFA showed that the main sources of PAHs emitted in the area were vehicle exhaust gases, followed by tyre wear, motor lubricant oils, road surface wear, and brake linings. Only 2–6% of the total 5.8–29 kg annually emitted PAHs/ha ended up in the stormwater sewer system. The measured PAH loads were found in much smaller amounts than the calculated loads and the outflow to stormwater contained much more of the hazardous PAHs than the total loads emitted in the catchment area.

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1. Introduction

The occurrence of organic pollutants (OPs) in the environment is a result of vigorous global development, including the continuous increase in industrial production, farming and other human activities over the past decades. Organic pollutants are either unintentional by-products released into the environment, for example through combustion processes, or intentionally produced industrial chemicals. Many OPs are persistent to degradation, capable of bioaccumulation in fat tissue, and have the ability to travel long distances through water-air-soil systems and cause negative effects on animal and human health (UN-ECE, 1998a). Although certain OPs are subject to long-range transport, and have been detected in remote areas far from where they are produced and used (Oehme and Manø, 1984; Scheringer, 2010), urban areas are the most important sinks for organic pollutants (Björklund et al., 2011; Eriksson, 2002). Urban areas are affected by pollutants emitted from activities such as transportation, commerce, construction and combustion. Apart from the most volatile substances, OPs emitted from nonpoint sources will settle on urban surfaces and subsequently be washed off by stormwater, into sewer systems and receiving waters. Recently, OPs, including the most hydrophobic compounds, were shown to be present in dissolved and colloidal forms in stormwater, which suggests that the mobility, and consequently the toxicity and associated risks of the compounds, is potentially higher than previously assumed (Kalmykova et al., 2013). It has also been shown that the development of advanced stormwater treatment facilities will be necessary for retention of these forms of OPs (Kalmykova et al., 2014; Nielsen et al., 2015). Indeed, the urban runoff quality is recognized as one of the most significant pressures on aquatic ecosystems worldwide (Ellis, 1991; Malmqvist and Rundle, 2002). For this reason, it is vital that the dominant sources of OPs in urban areas are identified as this will allow actions to be taken to improve surface water quality through control of the sources of emissions and ensure compliance with legal regulations and water quality standards, such as the European Water Framework Directive (EC, 2000) and the American Clean Water Act (EPA, 2002).

Monitoring of urban runoff has revealed that traffic is one of the primary sources of contamination in urban areas (Björklund, 2011; Jonsson et al., 2008; Motelay-Massei et al., 2006; Månsson et al., 2008). There is a myriad of possible traffic-related sources of OPs, including materials used in vehicles, such as tyres, auto body, motor components, and care products; exhaust gases and particles; road surfaces and markings; and road furniture such as traffic signs, sound berms, dividers and barriers (Faure et al., 2000; Rogge et al., 1993; Sartor et al., 1974). The amount of pollutants generated and accumulated on road surfaces is dependent on both traffic-related factors, such as velocity of vehicles and traffic count, and on runoff-related factors including antecedent dry period, duration and intensity of rainfall, and the leaching rate from permanent structures (Barrett et al., 1995; Furumai et al., 2002; Helmreich et al., 2010; Lee et al., 2004). Previous studies show that polycyclic aromatic hydrocarbons (PAHs) (Jensen et al., 2006; Kose et al., 2008), halogenated phenols (Zhang et al., 2008), alkanes, alkenes together with other petroleum derivatives (Rogge et al., 1993), polychlorinated biphenyls (Zgheib et al., 2012), phthalates like diisononyl phthalate (DINP) and diisodecyl phthalate (DIDP), and alkylphenols and their ethoxylates (AP/EOs) (Björklund, 2010), are frequently found in road dust. However, the state of the art overview of traffic-related OPs which risk being emitted to road runoff and could have hazardous effects on aquatic environments and human health is currently missing.

To date, analytical methods have only been developed for a selection of the compounds that may potentially be emitted from road-related sources, and analysis costs are high. Additionally, the fact that the same OPs may originate from different sources makes it difficult to identify specific sources when analysing mixtures of molecules present in road-related environmental matrices. This hinders effective identification of

the major sources of Ops, and makes it difficult to put in place appropriate source-control measures to prevent the emissions. For this reason, it is necessary to prioritize certain OPs and their sources, both to enable efficient monitoring and to provide decision support for measures to decrease emissions. The substance flow analysis (SFA) method is considered suitable for the monitoring of flows and stocks of pollutants. The SFA is an analytical tool developed for quantitative assessment of individual substances through a given system, specified in space and time (Björklund, 2010; Bringezu, 2006; Huang et al., 2012). This method allows monitoring of the amounts of OPs emitted to aquatic environments, as well as estimation of the scale of the potential contamination, based on their occurrence in the surrounding environment and emission factors.

The aim of this research has been to identify the occurrence and sources of OPs in road areas, quantify the emissions of selected OPs – so called priority pollutants (PPs) – from specific sources, and estimate the flows of these OPs in a road case study area. Source identification and emission flows have been researched through literature studies, and estimated PP flows have been compared to monitoring data from the case study area. In this paper, the SFA was conducted on the highest priority OPs to estimate their emitted loads in a case study catchment, as well to illustrate the capability of the method.

2. Method

2.1. Selection of priority pollutants

Fig. 1 illustrates the methodology used to identify sources of OPs in road environments and to select the PPs. Each step in the process is described in detail below.

2.1.1. Identification of organic pollutants present in traffic-related sources

The first step of the study was to identify substances or substance groups that may be released from traffic-related sources, and may therefore be present in road runoff. No pollutant ranking or judgment-based selection was implemented at this stage. The OPs expected to occur in road environments were found through reviews of scientific literature as well as regional and federal reports on monitoring campaigns for OPs. The OPs detected through laboratory measurements of road materials, road-side dust particles, road runoff and sediment were
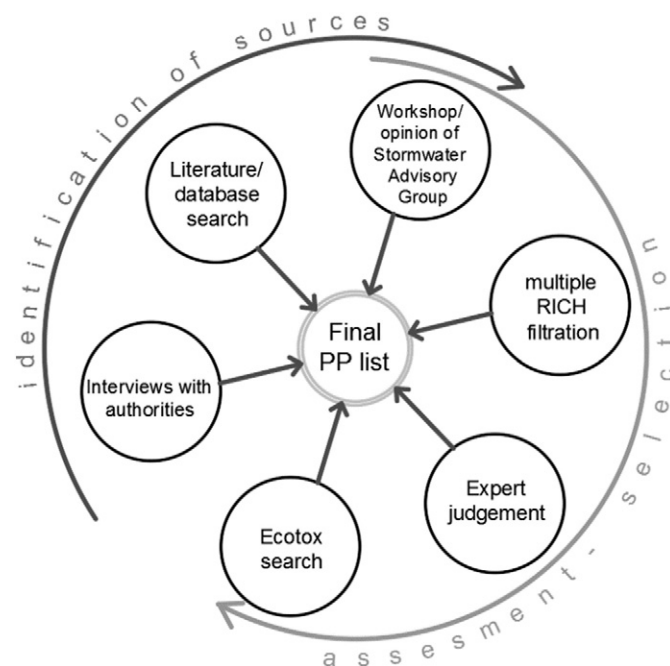

Fig. 1. The iterative selection process used for identifying and selecting organic priority pollutants in urban road environments.

extracted and the list of identified OPs was created. In addition, data on OPs and their sources in road environments were gathered through:

- An extensive literature search in abstract and citation databases as well as common internet search engines, covering both international and national reports and scientific journals.
- Research of the database International Material Data System (IMDS) for substances present in polymeric vehicle components.
- Interviews with experts from car manufacturing companies and companies responsible for the production of road materials in Sweden.

2.1.2. Ranking and selection of priority pollutants using RICH

The Ranking and Identification of Chemical Hazards (RICH) method has been developed to provide transparent ranking for a wide range of substances in stormwater, using the substances' inherent physico-chemical properties and biological effects (Baun et al., 2006). The tool is used for assessing environmental partitioning and chemical hazards and can help identify the most crucial pollutants for stormwater studies, i.e. priority pollutants (Fig. 2). In this study, RICH was applied to classify the compounds identified in the first stage of the study.

The RICH method is based on filtration using the pollutants' physico-chemical properties, which are set according to available international environmental and economic standards, described in detail by Baun et al. (2006). The first filter removes substances of high volatility (Henry's law constant K_H), and divides the remaining compounds into water- or sediment-bound, based on the organic carbon-water partitioning coefficient (K_{oc}). In the subsequent filters, substances that are persistent, based on degradation half-life ($T_{1/2}$) and the water-octanol distribution coefficient (K_{ow}); exhibit risk for bioaccumulation (bioconcentration factor, BCF) and toxicity (EC_{50} or $LC_{50} < 1$ mg/L); and exert long-term adverse effects, including carcinogenicity, teratogenicity, genotoxicity, mutagenicity and endocrine disruption, are "trapped" i.e. selected as possible PPs. It is up to an expert panel to judge whether or not the precautionary principle should be implemented for the remaining OPs that are not "trapped" in the filters.

Data for approximately 300 of the substances identified in the first part of the study was not yet available in RICH (data was available for 171 substances). For these substances, database mining was required to find inherent properties and ecotoxicity responses. This was done using the ECOTOX database (EPA, 1995), the Swedish chemical database KEMI, the online chemical database OCHEM, and the software Estimation Program Interface (EPI) Suite developed by the EPA US. Advanced Database Query was used to extract data, including e.g. BCF and the concentration-based EC_{50} or LC_{50}.

2.1.3. Specialist decision-making

To reduce the work required to find physico-chemical and ecotoxicological data for all substances, the list of identified traffic-related OPs was reviewed by a Stormwater Advisory Group during a workshop with, among others, experts on stormwater quality and chemicals, as well as representatives from the transportation administration and local and regional environmental authorities. The specific objectives of

the workshop were to discuss the overall concept of OPs present in road runoff and to identify substances whose priority is unlikely to be established by e.g. literature or database searches. Subsequent steps in the PP selection engaged the experts who, by applying personal knowledge and experience within the field, identified supplementary substances according to the following criteria:

- Risk of emission/leaching of pollutants from sources to stormwater systems.
- Specific (or groups of) substances emitted from more than one source in the road environment.
- Quantity of OPs present in vehicles, fuels, road construction materials and other sources in Sweden and in the EU.
- Hazardous effects on aquatic environments and human health, determined through physico-chemical properties available in RICH.
- Ability of commercial or research laboratories to analyse the chosen substances in sediment and water.
- No previous SFA study available for the particular pollutant or group of pollutants.

2.1.4. Final PP list

The missing ecotoxicity data for the substances selected by the stormwater advisory group and through expert judgment were retrieved, and added to the RICH method. Additional screening in RICH was consequently possible, and more pollutants were added to the PP list. An additional selection of PPs, based on the experts' judgment criteria, led to a final list of PPs and made it possible to select which PPs to include in the SFA. The final PP list was also confirmed by chemical analysis of suspended sediments trapped in an underground sedimentation treatment facility for stormwater from the Gårda catchment area, which represents a highly-trafficked road environment; see Section 2.2.3 for details.

2.2. Substance flow analysis

2.2.1. System definition

Based on the suggested PP list, and the expert judgment criteria described above, 16 specific PAHs prioritized by the US EPA and released from vehicles, road surfaces and other traffic-related sources, were selected for inclusion in the study (see Section 3.3. for details on the selection), see Fig. 3. The PAHs have been divided according to their molecular weight as follows: PAH-L(low) = $\sum$ naphthalene + acenaphthylene + acenaphthene; PAH-M(medium) = $\sum$ fluorene + phenanthrene + anthracene + fluoranthene + pyrene; and PAH-H(high) = $\sum$ 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. The SFA covers substance flows during one year; the system space boundary is an urban catchment area, located in the Gothenburg municipality (Sweden). According to the Transport Administration in Sweden, approximately 78,500 vehicles/day use the E6/E20 highway passing through the catchment area. The total size of the catchment area is 5.5 ha, of which 2 ha
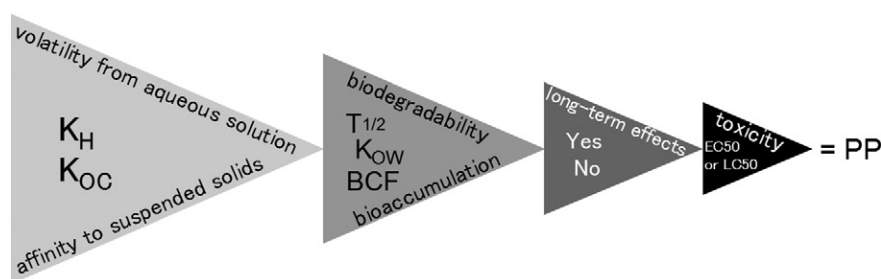

Fig. 2. The RICH method filtration process for Ranking and Identification of Chemical Hazards and priority pollutants. (Adopted from Baun et al., 2006)

of impervious surfaces. 94% of the impervious surfaces consist of roads, footpaths and parking lots (Björklund et al., 2008). It is assumed that the sources of OPs in the Gårda area is 100% road- and traffic-related. Runoff from the area is treated in an underground facility with seven sedimentation chambers (Björklund et al., 2009; Jakobsson et al., 2006; Pettersson et al., 2005). The physico-chemical parameters of stormwater, including PAH concentrations and distribution between particulate, dissolved and colloidal fractions in the inlet and outlet of the facility have been described elsewhere (Nielsen et al., 2015).

2.2.2. SFA procedure

The substance flows in the studied area were calculated using emission factors (EFs) for each source and individual pollutant. The EFs were estimated from literature data on emission rates and substance content in various source materials. Vehicle exhaust-related emissions were based on data found in the air pollutant emission inventory guidebook, published by the European Environment Agency (EEA, 2013). Tyre, road material and break lining emissions were determined using data on wear per driven kilometre and content of substance in tread, asphalt and lining material, respectively. Emissions of motor oil and lubricants were calculated from the substance content in used oils/lubricants and the estimated loss of oil/lubricant by leakage per driven distance. The factors for vehicle-related emissions were expressed in the format [μg/vkm]; mass of substance emitted per travelled vehicle-kilometre. The total substance flows were subsequently calculated using data on the length of the road and the total number of vehicles traveling through the area in one year.

Emitted pollutant loads are either transported out of the system with air (volatilised or bound to small particles); deposited on impervious surfaces in the catchment area, from where it could potentially reach the stormwater sewer system; or deposited on pervious areas, i.e. in sinks other than stormwater. The proportion of the exhaust emissions that end up on impervious surfaces (roads) was based on the findings of Hewitt and Rashed (1990). They estimated, for example, that only 1.3% of the total phenanthrene exhausts emissions is deposited on or near the road surface, and the remaining 98.7% is transported by air away from the road surface. Based on these data, it was assumed that PAH emissions remaining on or near the road represented 0.25% (naphthalene) to 30.8% (benzo[a]anthracene and chrysene) of the exhaust loads. In contrast to exhaust, emissions from tyres, road materials, brake linings and lubricants are assumed to collect entirely on the road surface. Lastly, it was assumed, based on the runoff coefficient for road surfaces, that 80% of the pollutant loads deposited on the road surface reach the nearby stormwater sewer system. The amounts of PAHs

transported by air into the studied system, i.e. the catchment area, were assumed to be negligible as evidenced by snow samples in Gothenburg that exhibit low background concentrations of PAHs (Björklund et al., 2011).

2.2.3. Sediment sampling procedure and calculation of stormwater loads

To validate the pollutant loads estimated with the SFA method, the concentrations and loads of PAHs and oxy-PAHs (degradation products of PAHs) were analytically determined on sediment samples collected from the Gårda stormwater treatment facility in May 2015. To confirm the importance of the compounds in the PP list, alkanes C_5–C_{35}, aromatics C_8–C_{35}, aldehydes C_1–C_5, twelve phthalates, octyl- and nonylphenols and their mono–tri ethoxylates were additionally analysed in the sediments; for results see Table A in Supporting information. Stormwater from the entire catchment area pass through the sedimentation system or an overflow bypass at times when the sedimentation chambers are filled up with stormwater, e.g. during heavy rainfalls.

One sediment sample was collected from each of the chambers 1, 3, 5 and 7, using an Ekman bottom grab sampler, stored in glass containers, and immediately sent to an accredited, commercial laboratory to be analysed for selected OPs. Analyses of PAHs and oxy-PAHs were performed on GC–MS according to methods based on SPIMFAB's (the Swedish Petroleum and Biofuel Institute) quality manual. The pollutant concentrations in chambers 2, 4 and 6 were estimated by interpolation of the measured quantities in chambers 1, 3, 5 and 7.

The sediment pollutant loads were estimated based on the analysed PAH and oxy-PAH concentrations in the sediment, the total dry weight of sediment in the facility, and the accumulation period of sediment (28 months). The total dry weight was calculated from known chamber dimensions, measured sediment height, density, and water content (Jakobsson et al., 2006). In Jakobsson's study, detailed physical and chemical characteristics of the Gårda sediment, e.g. metal concentrations, organic content, particle size distribution, are reported. Pettersson et al. (2005) estimated that approximately 30% of the PAHs found in stormwater in the Gårda catchment area is retained in the sediments; the remaining 70% is transported with the effluent or overflow water to the receiving water. The observed PAHs loads in the sediments were adjusted using Pettersson's data, after which the total annual stormwater load (reported in grams) of each PAH pollutant in the catchment area was determined.

3. Results and discussion

3.1. Sources of OPs in road environments

Possible sources of OPs in the road environment are summarized in Table 1, which contains information collected through literature reviews, national Swedish screening reports and interviews with experts.

3.1.1. Tyres

The composition of tyre rubber is complex, and often depends on quality demands as well as the vehicle for which it is designed. Tyres are believed to be a major source of OPs in road runoff, as a broad spectrum of toxic and harmful substances are released as they wear (Rogge et al., 1993). Several studies have confirmed the content of toxic additives used in the production process to enhance tyre performance and stability (Table 1). Examples include PAHs-containing highly aromatic (HA) oils used as softeners, and plasticizing phthalates or alkylphenols (APs) and their ethoxylates (AP/EOs) for aging protection (Ahlbom and Duus, 1994; Källqvist, 2005; Marcus, 2006). Since 2010, European legislation restricts the use of HA oils in rubber processing, which may lead to future reductions in PAHs emissions (EC, 2006). Despite this, there is still a relatively high number of tyres containing HA-oils on Swedish roads, due both to a delay in tyre
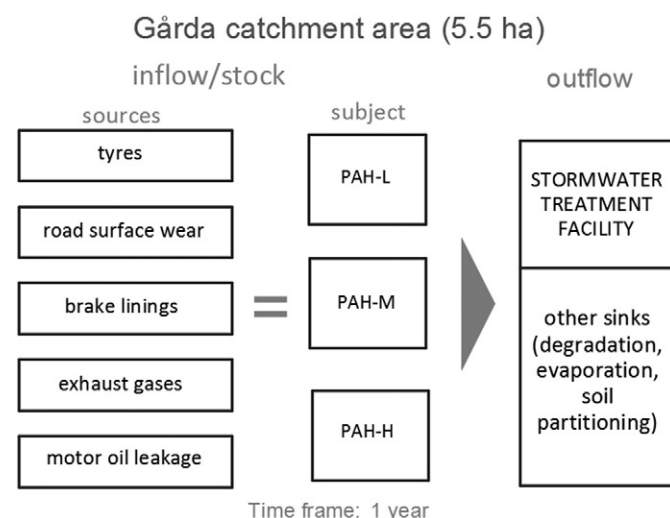
Gårda catchment area (5.5 ha)

Fig. 3. Flow chart of the SFA for the Gårda catchment area, with main sources of inflow, outflow to the stormwater facility and other sinks of PAH-L, PAH-M and PAH-H.

Table 1
Sources of organic pollutants identified in the road and traffic environment.

Type of source		Potential emitted substances	References
Vehicles			
Tyre wear		Phthalates, APs/EOs[a], n-alkanoic acids, n-alkanes, oleic acid, stearanes, hopanes, natural resins, PAHs[b]	Ahlbom and Duus, 1994; Kose et al., 2008; Källqvist, 2005; Marcus, 2006; Norin and Strömvall, 2004; Rogge et al., 1993
Brake lining		PAHs, oxy-PAHs[c], polyglycol ethers, benzaldehydes, benzoic acids, oleic acids, n-alkanoic acids	Rogge et al., 1993
Integrated vehicle components	Undercoating	Phthalates, PAHs	APME, 1999; Hewlett, 2013; Hoffmann, 1996; Johansson and Zimerson, 2000; Nielsen et al., 2000; Štrumberger et al., 2005
	Plastic components	Phthalates, APs/EOs, BPA[d], amines	
	Paint/lacquer	Phthalates, APs/EOs	
Car care products		Phthalates, APs/EOs, naphtha	Andersson and Sörme, 2004; Björklund et al., 2007; Johansson and Zimerson, 2000
Fuels, oils and lubricants	Lubricant oil (gear box, engine) hydraulic oil, chemical spillage	PAHs, phthalates, APs/EOs, naphtha, BPA, antioxidants[e]	Ahlbom and Duus, 1992; Björklund et al., 2007; Johansson and Zimerson, 2000; Remberger et al., 2003
	Exhaust gases/combustion of fuels (gasoline/diesel/LPG/CNG)	PAHs and related substances, aldehydes, alkanes, phenols, fatty acids, ethers	Brorström-Lundén E. et al., 2010; Ichiki et al., 2008; Kose et al., 2008; Schauer et al., 2002
Road construction materials			
Bitumen/asphalt		Phthalates, APs/EOs, PAHs, amides, amines, BPA, antioxidants	Kose et al., 2008; Norin and Strömvall, 2004; Wess et al., 2004; Zhang et al., 2011
Concrete		APs/EOs	Togerö, 2006
Road paint		Phthalates, APs/EOs, BPA, alkyd resins	Andersson and Sörme, 2004; Aznar et al., 1997; Bauer, 1980; Norin, 2004; Staples et al., 1998; Vikelsøe et al., 1999

[a] Alkylphenols and alkylphenol ethoxylates.
[b] Polycyclic aromatic hydrocarbons.
[c] Oxygenated PAHs.
[d] Bisphenol A.
[e] Alkylphenols.

replacement and to rubber recycling and international transport (ETRMA, 2010; ETRMA, 2011).

3.1.2. Brake linings

The brake linings of a vehicle are subjected to high friction heat during forced deceleration, which leads to lining wear. The wear, composed of particles containing various chemicals, is released into the environment (Rogge et al., 1993; Warner et al., 2000). Approximately 35% of the brake component mass loss is emitted as airborne particulate matter (Garg et al., 2000). The most abundant organic compounds identified in brake lining dust are polyglycol ethers (Table 1), used mainly in hydraulic fluids. Other compounds found in trace amounts in the lining dust include alkanes, alkanoic acids, PAHs, oxy-PAHs, benzaldehydes, benzoic acids, and oleic acids (Rogge et al., 1993).

3.1.3. Vehicle components

External polymeric materials used in e.g. bumpers, mudguards or bodywork parts of vehicles are susceptible to extensive wear. Plastic elements of vehicle body parts may contain phthalates, phenols, PAHs or amines (Nielsen et al., 2000), and lacquers and paints may contain phthalates or phenols (Hoffmann, 1996; Johansson and Zimerson, 2000), see Table 1. Emissions may increase in summer as the release of e.g. phthalates from plastic components strongly depends on temperature. Polymeric materials are widely used in the automobile industry to enhance appearance and increase cost effectiveness due to lower fuel consumption, and there has been a significant increase in their use over the past decades. The Global Automotive Declarable Substance List (GASDL) was set up to facilitate collaboration between automotive industries in relation to declaration and restriction of the use of certain chemicals in automotive materials (Group, 2016). The IMDS database (International Material Data System) provides information on substances found in vehicle parts, which according to GASDL may pose a risk to humans and the environment. Because the composition of various vehicle parts depends on their suppliers, searches in the IMDS database were performed with regard to suppliers to car manufacturers in Sweden. Based on information on polymeric elements used in vehicles, gathered from Štrumberger et al. (2005), around 70 organic substances which may be released from car parts were identified.

3.1.4. Car care products

Products intended for protection and preservation of lacquered vehicle surfaces are sources of AP/EOs, an abundant group of detergent chemicals (Table 1). In addition, cleaning agents, grease or wax are sources of substances such as phthalates, APs/EOs and naphtha residues (Andersson and Sörme, 2004; Björklund et al., 2007; Johansson and Zimerson, 2000).

3.1.5. Fuels, oils and lubricants

Fuel emissions make up a sizeable proportion of the on-road emissions of organic pollutants (Table 1). In the US EPA studies on mobile exhaust and evaporative emissions, over 1000 OPs were recorded (EPA, 2006). Identified organic compounds in diesel and gasoline exhausts are e.g. (n-, branched, cyclo-) alkanes and alkenes; aromatic hydrocarbons including PAHs and oxy-PAHs; biomarkers such as steranes, hopanes, terpanes; aldehydes and ketones; and alkanoic, alkanedioic, aromatic and benzoic acids (Rogge et al., 1993; Schauer et al., 1999; Schauer et al., 2002). The emission rates of individual organic pollutants depend on factors such as vehicle and fuel type, driving pattern, and whether a catalytic converter is used.

Lubricant motor oil is used to protect the engine from corrosion and friction, and contains a variety of protective chemicals (Denton, 2007). In addition to petroleum hydrocarbons, oil and lubricant spills are also sources of phenols, e.g. bisphenol A (BPA) and AP/EOs used as fuel additives or in hydraulic fluids (Ahlbom and Duus, 1992; Johansson and Zimerson, 2000). Antioxidants including 2,6- and 2,4-di-*tert*-butylphenols are used as lubricants in the petroleum industry or as additives in hydraulic oil (Remberger et al., 2003).

3.1.6. Asphalt and concrete

Abrasion of road materials is an important source of emissions of organic pollutants in road environments (Table 1). Road wear depends strongly on factors such as traffic characteristics, use of studded tyres, the material's susceptibility to abrasion, and weather conditions (Klint, 2001). Asphalt, a mixture of bitumen and mineral matter, can vary in composition depending on applicability, manufacturing process,

and the chemical formation of the crude oil from which bitumen is produced (Wess et al., 2004). The production of asphalt involves supplementation with additives; in recent years the proportion of these additives has increased, to meet higher demands for resistance and durability (Andersson et al., 2006). The most common additives include liquid and mineral adhesives, such as amines and amides; polymers like elastomers and plastomers; synthetic wax; and fibres (Huntsman, 2007; Morgan et al., 2013; NVF, 2006), but also recycled tyre granules, which are a source of phthalates and PAHs (Burstyn et al., 2000; Chauhan et al., 2012; Klint, 2001; Källqvist, 2005).

Concrete pavement materials contain various additives and admixtures (Table 1) used to improve their performance in areas such as water reduction, faster hardening, greater strength and durability, and general resistance to abrasion, consequently reducing leachability to the runoff (Ramachandran, 1996; Rixom and Mailvaganam, 1986; Tiruta-Barna et al., 2006; Togerö, 2006). The types of admixtures used in concrete include water reducers and accelerators, as well as air-entraining, water-proofing and de-foaming agents (Rixom and Mailvaganam, 1986). Some of these admixtures are based on organic substances like sodium thiocyanate, resin acids and nonylphenol ethoxylate, of which all have been proven to be susceptible to leaching (Togerö, 2006).

3.1.7. Road paint
Road paint is heavily exposed to abrasion and needs to be renewed every 5–10 years. Alkylphenol ethoxylates, phthalates, and alkyd resins are present in road paint components, such as emulsifiers, gradation agents and hardeners (Andersson and Sörme, 2004; Aznar et al., 1997). According to ECB (2002) typical paint mixtures can contain up to 3% of nonylphenol ethoxylates.

3.2. Results from the RICH screening process

For the first RICH filtration, the intention was to assess 306 compounds identified from the literature search. However, due to limitations in the available ecotoxicity data in RICH, only 171 of these substances were screened. As a result, 93 of the compounds were "trapped" in all RICH filters (defined in Fig. 2), i.e. classified as compounds with high toxicity and of high priority for further studies. Of these 93 compounds, 18 occur predominantly in the water phase and 75 in the sediment phase (Table 2). For the remaining 79 substances, which were removed by the respective filters of the RICH method, no further assessment was required. These substances are not associated with the water or sediment phase, exhibit low potential for bioaccumulation and toxicity, and/or do not cause long term-effects. An additional literature search of potential PPs to be screened in RICH provided a list of another 741 compounds, which was considered too many for a manual search for the required ecotoxicity data in RICH. After the workshop and expert meetings, the number of organic compounds was reduced to 61, and included amides, amines, aldehydes, alkanes and naphtha. It was decided that a manual search for ecotoxicity data would be performed for these compounds. Sufficient data was found for 53 of the compounds. An additional screening of RICH was then performed, after which 16 sediment-bound compounds were added to the total results (Table 2). Despite the fact that not all the data were available for RICH filtration, the obtained results indicate the level of hazard for specific groups of organic compounds.

3.3. The PP list and the highest PPs for SFA

From the RICH evaluation, a list of 109 compounds with high impact on the environment and/or humans was identified. This list was further evaluated at an expert meeting with four researchers and experts in the area of OPs. After the expert meeting, ten groups of compounds (Table 3) were chosen to be on the PP list of OPs emitted from the road and traffic environment. In Table 3, the compounds are presented in order from the highest priority to the lowest. With the exception of phenolic antioxidants, amides and amines (analytical methods not available), all these compounds were measured in high concentrations in sediment from Gårda (Table A in Supporting information). This list was also in line with the outcomes of the Stormwater Advisory Group workshop (14 participants), where compounds were deemed high priority depending on their sources and potentially high emitted volumes. The following groups of compounds met all the selection criteria ("no" in the column for availability of SFA means that the criterion is met): PAHs, alkanes and aldehydes. In this paper, a SFA of PAHs is presented. The reasons why PAHs were given the highest priority for SFA were that all the selection criteria were met, that PAHs are often the most frequently detected OPs in road environments (Eriksson et al., 2007; Gasperi et al., 2008), and that several of the PAHs are included in the priority pollutant list from the European Water Framework Directive. SFAs have already been performed for phthalates and nonylphenols and their ethoxylates (NP/EOs) in the Gårda catchment area, and the result showed that approximately 4.1 kg of four selected phthalates and 400 g of NP/EOs were emitted annually (Björklund, 2010). It was concluded that vehicles are the main source of these compounds in Gårda. The sum concentration of octyl- and nonylphenols and their corresponding mono, di and tri ethoxylates was 4.0 mg/kg DS in sediment from Gårda. Four of 12 analysed phthalates were measured in high concentrations (mg/kg DS): di-isononylphthalate (160) > di-(2-ethylhexyl)phthalate (17) ≫ di-n-butylphthalate (0.19) ~ di-isobutylphthalate (0.15).

3.3.1. PAHs and oxy-PAHs
As indicated in Section 3.1, PAHs arise from many sources and are among the most abundant pollutants emitted in road environments (Ravindra et al., 2008). PAHs are released from several traffic related sources, mainly exhausts and tyre wear, and represent a high risk of leaching to surface waters (for more details, see Section 3.4). In addition, PAHs deserve special attention due to their severe toxicity, including carcinogenicity. In the Gårda sediment, the concentrations of PAH-M and PAH-H reached 5.2 and 3.9 mg/kg DS respectively, which exceeds the Swedish EPA guidelines for sensitive land use of 3.0 mg/kg DS for PAH-M and 1.0 mg/kg DS for PAH-H (Naturvårdsverket, 2009).

Oxygenated PAHs (oxy-PAHs), i.e. oxidation products of PAHs, are often found at contaminated sites (Lundstedt et al., 2007). According to the RICH method, all identified oxy-PAHs met all the hazardous criteria provided by the method. Oxy-PAHs are not emitted in large quantities, but their high levels of toxicity assign them to the PP list. A proportion of the PAHs emitted from the road and traffic environment to stormwater may degrade to oxy-PAHs. In Gårda, oxy-PAH concentrations in the sediment reached 1.1 mg/kg DS, which is high compared to previous studies (Lundstedt et al., 2007; Bandowe and Wilcke, 2010). Eight of the nine analysed oxy-PAHs were identified in the sediment: benz[a]anthracene-7,12-dion, a degradation product of

Table 2
Number of organic pollutants classified as hazardous by the RICH method, and of high priority for further studies.

	First RICH screening (171[a])	Second RICH screening (53[a])	Final list
Hazardous in water phase	18	0	18
Hazardous in sediment phase	75	16	91
Total hazardous compounds	93	16	109

[a] Total number of compounds screened.

Table 3
Identified (groups of) priority pollutants according to the selection criteria method used.

PP group/compound	Criteria						
	Risk of transport from source to storm-water	Emission from more than one source	Significant amounts in the sources estimated by the Expert group[d]	Hazardous effects according to RICH[e]	Chemical analysis available	SFA available for road environment	High concentrations in the sediment in the Gårda case study area
PAH 16	Yes	Yes	Yes	Yes	Yes	No	Yes[j]
Alkanes C_{20}–C_{40}	Yes	Yes	Yes	Yes	Yes	No	Yes[j]
APs[a]	Yes	Yes	Yes	Yes	Yes	Yes[f]	Yes[j]
Phthalates	Yes	Yes	Yes	Yes	Yes	Yes[f]	Yes[k]
Aldehydes C_1–C_{40}	Yes	Yes	Yes	Yes	Yes	No	Yes C_1–C_5
Phenolic antioxidants	Yes	Yes	Yes	Yes	Yes	Yes[g]	n.m.
BPA[b]	Yes	Yes	Yes	Yes	Yes	Yes[h]	n.m.
Oxy-PAHs[c]	Yes	Yes	No	Yes	Yes	No	Yes[l]
Naphtha C_5–C_{12} (petroleum)	Yes	Yes	Yes	Yes	Yes	No	n.d.
Amides	Yes	Yes	No	Yes	No	No	n.m.
Amines	Yes	Yes	No	Yes	No	Yes[i]	n.m.

n.d. – not detected.
n.m. – not measured.
n.a. – detected, but soil quality guideline not available.

[a] Alkylphenols.
[b] Bisphenol A.
[c] Oxygenated PAHs.
[d] High volumes of organic pollutants occurring in the road environment established on the basis of knowledge of the Expert group.
[e] Ranking and Identification of Chemical Hazards.
[f] Björklund, 2010.
[g] KEMI, 2010.
[h] KEMI, 2012.
[i] KEMI, 2008.
[j] Classification according to Swedish EPA general guidance for contaminated soil (Naturvårdsverket, 2009).
[k] Classification according to Polish regulation of the Minister of the Environment on soil and ground quality standards (Żelichowski, 2002).
[l] Compared to previous studies (Lundstedt et al., 2007; Bandowe and Wilcke, 2010).

benz[*a*]anthracene, and 9,10-anthraquinone, a degradation product of anthracene, occurred in the highest concentrations.

3.3.2. Alkanes, aldehydes, naphtha

Alkanes and aldehydes are present in the largest quantities in the traffic and road environment (Schauer et al., 1999; Schauer et al., 2002). Based on the RICH method, mainly C_{20}–C_{40} alkanes and C_1–C_{20} aldehydes were identified as toxic. In the Gårda sediment, alkanes C_{10}–C_{35} were measured in concentrations ranging between 1.0 and 10 g/kg DS. Several studies confirm that *n*-alkanes and aldehydes, such as formaldehyde and acetaldehyde, are among the most abundant organic compounds identified in vehicle exhausts, and that crotonaldehyde is also found in significant amounts (Davis et al., 2007; Perrone et al., 2014; Schauer et al., 2002). In sediments from Gårda, the presence of both formaldehyde (5.7 mg/kg DS) and acetalde-hyde (1.7 mg/kg DS) was confirmed. Moreover, in the current study the source apportionment of alkanes and aldehydes showed on-road emis-sions, such as exhausts and fuel spillage, to be the most common sources (EPA, 2006; Schauer et al., 1999; Schauer et al., 2002). Brake linings and tyre wear were also important sources of alkanes and aldehydes (Rogge et al., 1993).

Naphtha is derived from crude oil and includes molecules ranging from C_5 to C_{12} (Devold, 2013; Ramadhan et al., 1997). Naphtha can be used for a broad spectrum of applications, including as feedstock for producing additives for (high octane) gasoline, a solvent in cleaning liq-uids such as glass cleaners, corrosion removers or degreasers, and in the production of lubrication agents (Johansson and Zimerson, 2000). Data on hazardous effects of complex mixtures such as naphtha were not available in the RICH method. In addition, naphtha can be found under different technical names and its chemical composition varies depend-ing on its use and crude oil quality. For this reason, it is difficult to define its impact on aquatic organisms. Despite this, the Stormwater Advisory Group workshop recommended that naphtha be included on the PP list, mainly due to the large volumes used. The concentrations of naphtha were below the detection limits of the analytical method used in Gårda. The low sediment concentrations are explained by the high

volatility of the C_5–C_{12} alkanes; these hydrocarbons should preferably be measured in the water and vapor phases.

3.3.3. Phenolic compounds, including antioxidants, bisphenol A

The most commonly detected phenols in stormwater or stormwater sediments are alkylphenols and their ethoxylates, as well as methylphenols, bisphenol A and antioxidants such as tertiary butylphenols (Björklund et al., 2007; Boyd et al., 2004; Flint et al., 2012; Remberger et al., 2003; Rosqvist, 2004).

In this study several antioxidants in the form of tertiary butylphenols were assigned to the PP list because of their abundance in road-related sources. In Sweden, 400 tons of butylphenols are used annually in a va-riety of products and materials, e.g. asphalt, lubricants, stabilisers, fuels and rubber or plastic components (Björklund et al., 2007; Remberger et al., 2003; Rosqvist, 2004). Antioxidants such as 4-*t*-butylfenol, 2,4-di-*t*-butylfenol, 2,6-di-*t*-butylfenol and *t*-butylhydroquinone have been found in elevated concentration in surface waters (Remberger et al., 2003; Rosqvist, 2004) and the ranking performed in RICH con-firmed the compounds' toxicity and tendency to bioaccumulate in aquatic organisms as well as their resistance to degradation and possi-ble endocrine disrupting effects.

Bisphenol A was identified as a PP based on its high production vol-ume (approximately 3.8 million tons worldwide), many application areas and continuous release to the environment from final products (Oehlmann et al., 2009). BPA is used mainly as a monomer in the pro-duction of polycarbonate and epoxy resins (Flint et al., 2012; Hoekstra and Simoneau, 2013; Michałowicz, 2014; Staples et al., 1998), as an an-tioxidants in products used in the plastics and rubber industry (KEMI, 2012) and as an inhibitor of end polymerisation in PVC manufacturing (EFSA, 2010). Uses of BPA in road-related components include asphalt modifiers (Zhang et al., 2011), road paints (Bauer, 1980; Staples et al., 1998) and vehicle parts such as automotive lenses, dashboards and bumpers (APME, 1999). The RICH method classified BPA as a chemical likely to be found in the sediment phase, resistant to degradation and exhibiting endocrine disrupting properties. BPA meets all the selection

criteria (Table 3), although an SFA has previously been performed for Sweden (KEMI, 2012).

3.3.4. Amines and amides

Amines are common products of road wear as they are often used as liquid adhesives in asphalt and admixtures, and as hardening accelerators for concrete and cement (Andersson et al., 2006; Rixom and Mailvaganam, 1986). Several amines and amides have been identified in vehicle parts (Hewlett, 2013), and amides are also used as blocking resistant agents in asphalt modifiers (Nakanishi et al., 2010). However, as indicated by the expert panel during the selection process, both groups are only found in trace amounts in the identified sources. Water and sediment analysis for these groups are currently not available, why sediment concentrations in the Gårda treatment facility were not measured. The RICH screening identified several toxic amines and a few amides, supporting the decision to grant both groups priority over other stormwater related pollutants.

3.4. Substance flow analysis of PAHs

3.4.1. Emission factors of PAH-16 from road-related sources

Emission factors (Table 4) for each specific EPA-priority PAH were determined to enable the calculation of compound flows in the Gårda area. The main sources are vehicle exhausts, tyre wear (mainly from HA oils), motor lubricant oils, road surface wear and brake linings. The total amounts of PAHs emitted from traffic and roads are calculated to be in the range of 990–3900 μg per vehicle kilometres (vkm).

The PAHs emitted in the largest quantities are 2–4 rings (Table 4), in the following order [μg/vkm]: naphthalene > chrysene > pyrene > phenanthrene > fluoranthene. The main sources of these PAHs are diesel passenger cars (PC) and light commercial vehicles (LCV, weight < 3.5 tons), which emit a total of 740–2300 μg/vkm, followed by PCs and LCVs fuelled by gasoline/E85 (ethanol)/compressed natural gas (CNG), tyre wear, heavy-duty vehicles (HDV, weight > 3.5 tons), motor oils and lubricants, road surface wear and brake linings.

PAHs are emitted in high quantities in exhaust gases (Table 4) from all types of vehicles and fuels (Ntziachristos and Samaras, 2013). The PAHs most commonly found in vehicle exhausts are 2–4 rings: naphthalene > phenanthrene > fluoranthene > pyrene. The PAHs occurring in the highest concentrations in tyres are 4–6 rings: chrysene > pyrene > fluoranthene > benzo[g,h,i]perylene, mostly found in HA oils. As mentioned, the use of HA oils in tyres is now restricted in Europe, but since the lifetime of tyres is 50,000–60,000 km (Ntziachristos and Boulter, 2009; UKEA, 1998) or 4 years, and wear out refers to approximately 1 kg per tyre (Baumann and Ismeier, 1998), HA oils still occur in the road and traffic environment. Specific PAHs dominating in the lubricant motor oils are 3–6 rings: phenanthrene > benzo[a]pyrene > benzo[g,h,i]perylene ~ indeno[1,2,3-c,d]pyrene ~ anthracene = pyrene. The content of PAHs in the oil depends on the age of the oil and is due to the pyrolysis and pyrosynthesis of the oil at the high temperature in the engine, and the contamination by fuel combustion products (Wong and Wang, 2001). Used engine oil from gasoline cars contain considerably more PAHs than that from diesel cars and trucks (Clonfero et al., 1996).

PAHs also occur in the bituminous binder in asphalt (Brandt and De Groot, 2001). The PAHs dominating in emissions from asphalt are 4–6 rings: benzo[a]anthracene > benzo[g,h,i]perylene = chrysene > fluoranthene (Table 4). The amount of brake wear emitted depends on the brake pad composition, the vehicle type and the driver behaviour. The EF was estimated to 8.8–84 mg/vkm, based on data from Rogge et al. (1993), Westerlund (2001), Ntziachristos and Boulter (2009) and Luhana et al. (2004). Benzo[g,h,i]perylene followed by chrysene and benzo[a]anthracene (4–6 rings) are the PAHs mostly emitted from brake linings.

3.4.2. Estimated loads of PAHs in case study area

The results of the SFA (Fig. 4) indicate that the total amount of PAHs emitted annually in the Gårda catchment area (5.5 ha) is 32–160 kg. Through recalculation, this gives an annual emission load of 5.8–29 kg/ha for the urban road environment. The results suggest that only 2–6%, i.e. 500–9700 g/year, of the total amount of PAHs emitted in Gårda end up in the stormwater treatment facility. The majority of the PAHs are distributed to other sinks, through emissions to air, and immobilized in surrounding soils and on urban surfaces. Unaccountable losses of water and PAHs, e.g. through degradation, in the underground pipe system is also a possible sink. The results are surprising, as chemical analysis of PAHs in sediments show substantially lower emissions to the stormwater in Gårda than the loads calculated using emission factors. Possible uncertainties in the load estimations, sediment sampling and analysis are further discussed in Section 3.4.3.

The largest sources of PAHs in the Gårda catchment area, i.e. the inflow (Fig. 4), are exhaust gases ≫ tyres > motor oil leakage > road surface wear = brake linings. The PAHs that dominate in the emissions are PAH-L and PAH-M – naphthalene ≫ phenanthrene > fluoranthene = pyrene – all deriving from emissions from vehicles run on diesel, gasoline, E85 or CNG, and from both cars and LCVs (see Table 3). In the outflow, i.e. the flow to stormwater (Fig. 4), the composition of the PAHs is very different from the inflow, and here the PAH-M and PAH-H dominate, specifically chrysene > pyrene ≫ indeno[1,2,3-c,d]pyrene = fluoranthene = phenanthrene. This implies that PAHs present in the stormwater are found in much smaller amounts compared to the total loads, but contain much more of the hazardous PAHs as both chrysene and indeno[1,2,3-c,d]pyrene are classified as probable human carcinogen agents (ATSDR, 2009). The main source of the PAHs found in stormwater is tyres, followed by motor oil leakage, exhaust gases, road surface wear and brake linings. For this reason, it is of high importance to prioritize the environmental work to remove all tyres containing HA oils, or remainders of HA oils, from the market. It is also important to focus on developing innovative engine combustion techniques, to minimise the use of motor oils that contain PAHs, but also to develop techniques that prevent motor oil spills, whether from the engine or the exhaust system, from reaching the surrounding environment.

3.4.3. Comparison of SFA results and quantitative sediment-derived loads

The total PAH loads in the Gårda catchment area, estimated using EFs, are approximately 6 to 52 times (min/max EF loads, respectively) higher than the loads estimated from sediment concentrations and masses (Fig. 5). The measured concentrations of oxy-PAHs, which occur as a product of postemission oxidation of PAHs in sediment (Lundstedt et al., 2007), were added to the total PAH loads. Measurement uncertainties relating to sediment masses and PAH concentrations have been accounted for in the sediment load calculations. However, assumptions and estimations of EFs and the proportions of pollutants that reach the stormwater system also include uncertainties. The percentage of PAHs deposited on the road, as presented by Hewitt and Rashed (1990), were used in this study to calculate the proportion of PAHs reaching the stormwater system. The PAHs deposited on the nearby soil and grass, i.e. not on the road surface, are much less likely to reach the stormwater system. The partitioning of PAHs between road surface and soil/grass was not reported for all studied PAHs (Hewitt and Rashed, 1990). Accordingly, assumptions were made for certain compounds, which adds to the uncertainty of calculated PAHs loads. In addition, load calculations do not take into account the proportion of PAHs sorbed to stationary road-side sediments (not transported with stormwater) or to sediments which are removed through street cleaning. As mentioned previously, a considerable share of the PAHs found in stormwater is in the dissolved and colloidal phases, which prevents sorption and precipitation of the PAHs and facilitates their transport through the sedimentation facility (Kalmykova et al., 2013;

Table 4
Minimum and maximum emission factors of polycyclic aromatic hydrocarbons from traffic and roads.

16 US EPA-priority PAH	Vehicle exhaust[a]			Tyre (with HA-oils) wear[b] μg/vkm × 10^{-3}	Motor lubricant oils[c] μg/vkm × 10^{-3}	Road surface wear[d] μg/vkm[e] × 10^{-3}	Brake linings[e] μg/vkm × 10^{-3}	Sum min–max μg/vkm × 10^{-3}
	μg/vkm × 10^{-3} gasoline, E85, CNG[g] PC[h] and LCV[i]	μg/vkm × 10^{-3} diesel PC and LCV	μg/vkm × 10^{-3} HDV[j]					
Naphthalene (2-ring)	$(11–610) × 10^3$	$(650–2100) × 10^3$	$57 × 10^3$	$38–18 × 10^3$	0–830	3.6–80	nd	$(720–2800) × 10^3$
Acenaphthylene (3-ring)	nd	$26 × 10^3$	nd	nd	0–48	0.90–2.4	nd	$26 × 10^3$
Acenaphthalene (3-ring)	nd	nd	nd	$29–22 × 10^3$	0–190	0.54–3.4	nd	$30–22 × 10^3$
Fluorene (3-ring)	nd	nd	$40 × 10^3$	$9.6–18 × 10^3$	0–840	1.3–5.6	nd	$(40–59) × 10^3$
Phenanthrene (3-ring)	$4700–62 × 10^3$	$(28–86) × 10^3$	$23 × 10^3$	$220–22 × 10^3$	0–3400	8.7–30	8.5–81	$(56–200) × 10^3$
Anthracene (3-ring)	800–7700	1400–3400	8600	$9.6–8000$	0–960	2.5–15	nd	$(11–29) × 10^3$
Fluoranthene (4-ring)	$2800–18 × 10^3$	$(18–38) × 10^3$	$21 × 10^3$	$130–62 × 10^3$	0–800	12–88	6.1–58	$(42–140) × 10^3$
Pyrene (4-ring)	1800–5800	$(12–39) × 10^3$	$32 × 10^3$	$320–130 × 10^3$	0–960	8.7–58	9.7–92	$(46–210) × 10^3$
Benzo[a] anthracene (4-ring)	430–840	2700–3300	2400	$67–34 × 10^3$	0–640	21–180	13–130	$5600–41 × 10^3$
Chrysene (4-ring)	430–530	2400–7500	$16 × 10^3$	$210–210 × 10^3$	6.0–270	23–96	15–143	$(19–230) × 10^3$
Benzo[b]fluoranthene (5-ring)	360–880	600–3300	5400	$230–26 × 10^3$	9.0–480	9.0–62	3.7–35	$6600–36 × 10^3$
Benzo[k]fluoranthene (5-ring)	260–300	190–2900	6100	nd	11–270	2.3–9.6	5.5–52	$6600–10 × 10^3$
Benzo[a]pyrene (5-ring)	320–480	630–2900	900	$38–12 × 10^3$	0–1400	15–62	6.5–62	$1900–18 × 10^3$
Dibenz[a,h]anthracene (5-ring)	10–30	240–560	340	$9.6–4800$	0–90	9.0–72	nd	$610–5.9 × 10^3$
Benzo[g,h,i]perylene (6-ring)	560–2900	950–6000	770	$48–52 × 10^3$	0–1000	23–96	23–220	$2400–63 × 10^3$
Indeno[1,2,3-c,d]pyrene (6-ring)	390–1000	700–2500	1400	$9.6–9200$	0–980	2.8–8.8	nd	$2500–15 × 10^3$
Total PAH 16	$(24–710) × 10^3$	$(740–2300) × 10^3$	$220 × 10^3$	$(1.5–630) × 10^3$	$26–13 × 10^3$	140–880	91–870	$(990–3900) × 10^3$

[a] Ntziachristos and Samaras, 2013.
[b] ETRMA, 2011; Björklund, 2010; Denier van der Gon et al., 2008.
[c] Wong and Wang, 2001; Klein et al., 2013; EEA, 2013.
[d] Lindgren, 1998; Denier van der Gon et al., 2008.
[e] Warner et al., 2000; Ntziachristos and Boulter, 2009; Rogge et al., 1993.
[f] Vehicle kilometres.
[g] Compressed natural gas.
[h] Passenger cars.
[i] Light commercial vehicles (<3.5 tons).
[j] Heavy-duty vehicle (>3.5 tons).

Nielsen et al., 2015). Previous studies also showed that the calculated K_{OW} values for certain organic pollutants were lower than the theoretical values (Kalmykova et al., 2013). For this study, it should be noted that the distribution of PAHs to the sediment, and the amount transported out from the Gårda facility, is determined on measured concentrations in a few sediment samples. It is also possible that some of the underground sewer pipes are leaching or incorrectly connected, and thus contribute to the loss of stormwater transported to the sedimentation facility. Taken together, it is assumed that the PAH loads actually reaching the sedimentation system in Gårda, i.e. the calculated sediment loads, are underestimated.

The highest discrepancy between EF and sediment loads was found for chrysene, followed by naphthalene and benzo[a]anthracene (26–260, 20–90 and 15–79 times higher loads estimated from EFs, respectively). This trend is visualized in Fig. 6 (the proportion of individual PAHs is the same for sediment concentrations and sediment loads). Naphthalene is volatile and more water soluble (approximately 20 mg/L) than the other PAHs, and has been found predominantly in the dissolved phase in contaminated waters (Kalmykova et al., 2013), why monitored concentrations in sediments may not reflect the true loads emitted to the stormwater system. Chrysene was previously found in high concentrations in tyres, and low sediment concentrations may be due to the decreased use of HA oils in rubber, as a result of the European restrictions that came into force in 2010. Emission patterns and loads of PAHs in traffic areas tend to fluctuate over time and follow changes in source occurrence, such as preferred type of fuel, efficiency of combustion engines, and restrictions imposed on for example the

use of HA oils in rubber tyres and coal-tar-based pavement sealcoats (Ravindra et al., 2008; Shen et al., 2011; Van Metre and Mahler, 2014). For this reason, EFs based on outdated references may lead to incorrect PAH loads. Furthermore, reported EFs (Table 4 and Fig. 6) vary by several orders of magnitude, illustrating how difficult and uncertain estimations of PAH emissions indeed are. Consequently, this leads to large uncertainties when PAH loads are estimated using EFs.

A comparison of the proportions of PAHs in sediments collected in Gårda in 2003 (Jakobsson et al., 2006) and 2015 (same sampling procedure) revealed that the PAH concentrations were on average 1.5 times higher in samples collected in 2015. These results indicate that PAH emissions accumulate and vary over time. Samples collected in 2003 contained a significantly (independent samples t-test, $p > 0.005$) higher proportion of phenanthrene, anthracene, chrysene and dibenz[a,h]anthracene, i.e. mostly PAH-M (Fig. 6). Samples collected in 2015 contained a higher proportion of fluoranthene, benzo[b,k]fluoranthene, benzo[a]pyrene and benzo[g,h,i]perylene, i.e. mostly PAH-L (Fig. 6). To investigate whether the source apportionment has changed over time, the proportions of PAHs in 2003 and 2015 were compared with calculated loads using EFs. No strong trends in source apportionment were seen between the years; exhaust gases and tyres are major sources of the most abundant PAHs in samples collected in both 2003 and 2015. Tyres were expected to be less important in the more recent samples due to the reduced use of HA oils in rubber. One indication of this is that chrysene, with high EFs for tyres, was much more abundant in the 2003 samples. However, benzo[b,k]fluoranthene, benzo[a]pyrene and benzo[g,h,i]perylene, which were more abundant in the 2015 samples, are also mainly emitted from
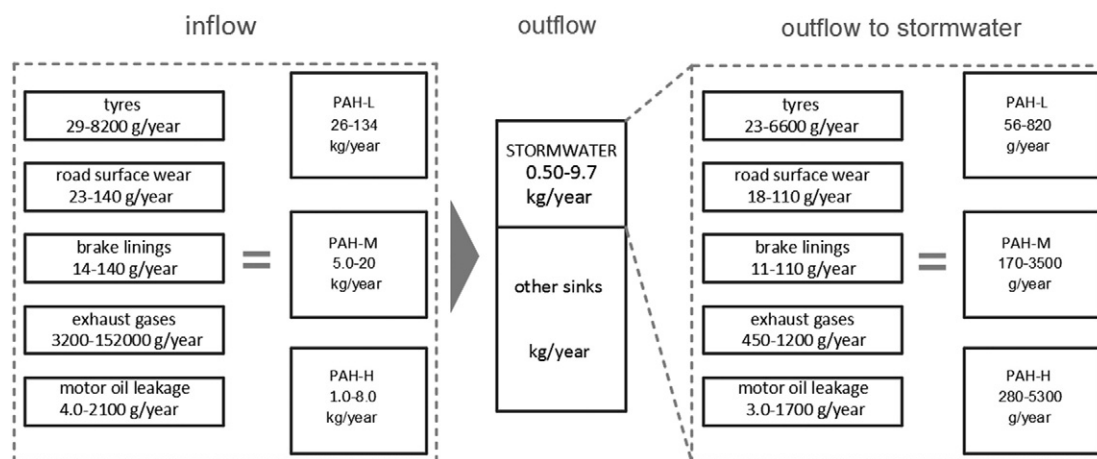
Fig. 4. Estimated fluxes of polycyclic aromatic hydrocarbons in the Gårda case study area.

tyres, although the tyre EFs for these PAH-H are one to two magnitudes lower than for chrysene (Table 4).

In sediment measurements from 2003 and previous studies, the concentration of pollutants were highest in chamber 7 of the Gårda facility (Fig. 6 and Pettersson et al., 2005). In contrast, the sampling performed in 2015 (Supporting information Table A) revealed that concentrations of all parameters in chamber 7 are lower than in chambers 3 and 5. For this reason, sample 7 may not be representative. If that is the case, the PAH loads estimated from sediment concentrations are likely to be underestimated.

4. Impact of this study

The results from this study will be used to propose methods to counteract the emissions of OPs in road runoff to the aquatic environment, using a multi-criteria decision analysis (MCDA) approach. MCDA has frequently been used for assessment of environmental problems, although it has been sparsely used for stormwater management (Ellis et al., 2004; Jia et al., 2013). Development of an MCDA method will contribute to the implementation of Best Management Practices including e.g. road sweeping, inlet control, infiltration areas and investments in local treatment facilities in the urban area. Identification of PP sources should call on relevant stakeholders within the vehicle and oil industry, as well as planning departments to formulate strategies for the upstream pollution control at the local, national and international scale.

There are a few limitations in the implementation of the findings of this study. For the SFA of PAHs, the information available is quite extensive yet plenty is missing or outdated. For other compounds on the PP-list, reliable emission factors are missing. This imply that identifying the most important sources of OPs is complex, which means that countermeasures to reduce OP sources and emissions are difficult to outline. Therefore further studies on emissions and determination of emission factors of OPs are in demand. Additionally, in this research a case study of a small road catchment was presented; thus it was possible to monitor the fluxes of pollutants in stormwater. Other urban areas may present a mix of different activities and constructions, and more difficult to monitor, hence comparing SFA results with measured loads may not be possible.

To successfully implement reduction measures, it is of high importance to identify the dominant sources of OPs in the catchment. However, it is challenging to estimate accurately the sources of OPs from roads and vehicles to road runoff. In the source identification stage of the SFA, quantities of the released OPs were neglected. Data found on emissions or occurrences of specific OPs, from different entities found in road and traffic environments, served to establish them as possible sources of OPs. Studies that were used to establish the sources of OPs to road runoff may have limitations related to outdated emission factors or agglomeration of multiple source deposits that hinders accurate source recognition. Additionally, long-range transport of low molecular weight pollutants means that the location of a source is not clear. Also, finding literature covering the most important sources is time consuming and
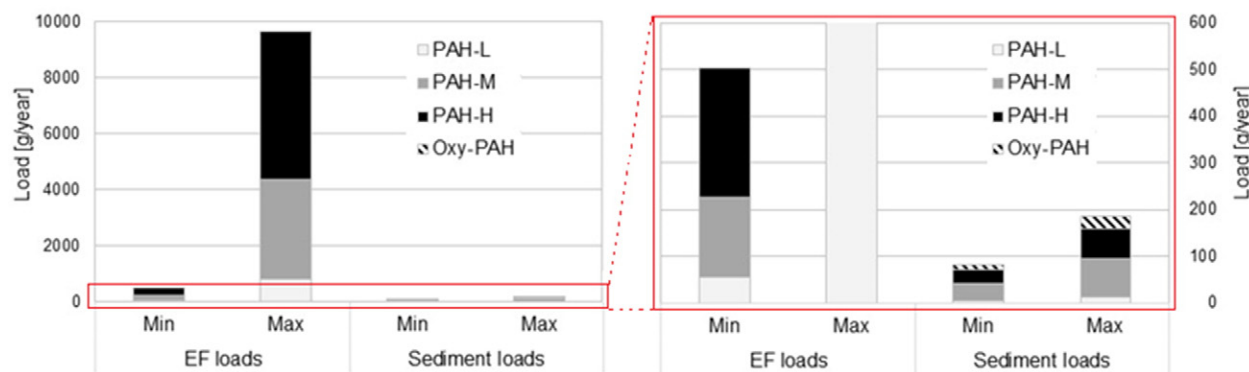

Fig. 5. Estimated loads of total polycyclic aromatic hydrocarbons (PAHs) in stormwater from the Gårda catchment area using emission factors (minimum and maximum reported EFs) and calculated from quantified PAH and oxy-PAH concentrations in the Gårda sedimentation facility. Minimum/maximum sediment loads are estimated based on a ±20% PAH/oxy-PAH analytical error and a ±20% measurement error in the sediment mass. The data is presented in two panes as the values are of different magnitude.
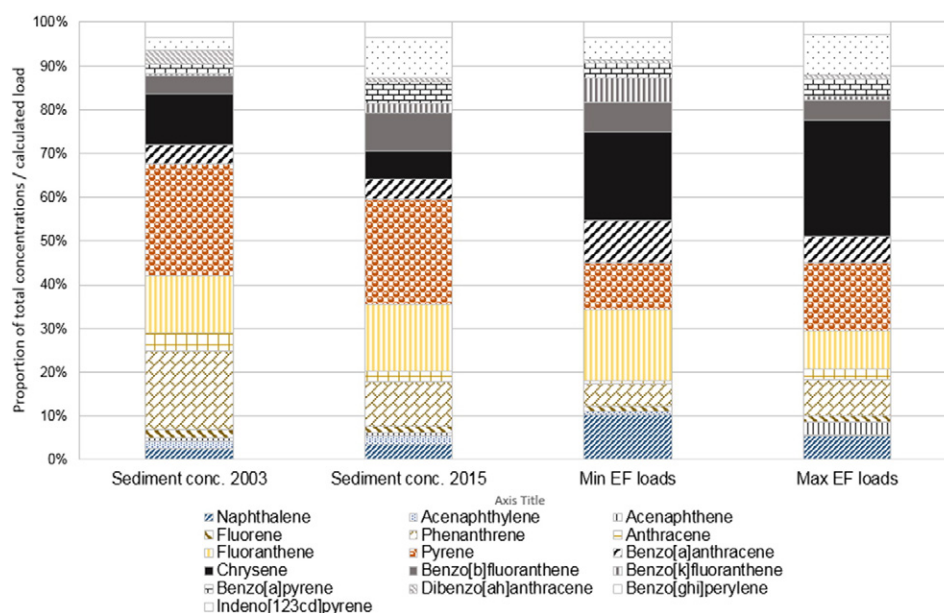
Fig. 6. The proportion (%) of individual polycyclic aromatic hydrocarbons (PAHs) to total PAH concentrations measured in the Gårda sedimentation system in 2003 and 2015, and to minimum/maximum loads calculated from emission factors.

performing SFAs for all the groups of selected PPs would be difficult owing to the lack of data for many OPs.

For precise estimation of source abundance, we recommend using geographical information systems to measure and identify surface areas of different types of land use (e.g. asphalt, roofs and green areas) and statistical data for traffic counts. Important is also to make more measurements in the field to determine loads, under varying times of the year; in addition to sediment samples, water samples may allow more efficient tracking of the pollutant depositions. As discussed, EF vary by orders of magnitudes depending on the literature source and also vary over time, because of legislation and improved processes (e.g. vehicle engines are constantly improved to emit less). Because the EFs in this study are derived from literature sources worldwide, it is our opinion that the EFs could be used in many other parts of the world. Adjustment to EFs are probably not needed in regions where the vehicle fleet is similar to that of Sweden, for example the rest of the EU (similar regulations for tyres oil, diesel and gasoline products and vehicle emissions).

5. Conclusions

The literature study in this paper identified 1100 organic pollutants as potentially emitted to the environment from road-related sources. This comprehensive list of OPs was systematically reduced to a priority pollutant list using literature and database searches, advice from practitioners and experts, as well as the RICH method. The RICH method is very useful for prioritizing persistent and toxic OPs, however, only a third of the priority OPs identified in this study could be found in the RICH database. Therefore, data for the missing substances were collected from different databases and added to RICH.

The following list of priority pollutants was obtained through the screening: PAHs > alkanes C_{20}–C_{40} > alkylphenols > phthalates > aldehydes > phenolic antioxidants > bisphenol A > oxy-PAHs > naphtha C_5–C_{12} > amides > amines. A substance flow analysis was performed for the highest priority pollutants – PAHs – in a highway case study area (5.5 ha). The total amounts of PAHs emitted from traffic and roads were calculated to 990–3900 µg per driven vehicle kilometre. The total amount of PAHs emitted annually from the studied area may be as high as 32–160 kg. The main sources of PAHs include exhaust gases ≫ tyres > motor oil leakage > road surface wear = brake linings.

Although only 2–6% of the total amount of PAHs are transported to stormwater in the catchment area, this load contain a higher proportion of the hazardous PAHs, originating mainly from tyres, motor oil leakage and exhaust gases. A sensitivity analysis of the suggested sources was conducted using sediment concentrations for 2003 and 2015. The same sources – exhaust gases and tyres – appeared to be dominant in both years.

The SFA results were compared to measurements of PAHs and oxy-PAHs in accumulated sediments in the case study area. The PAH loads in sediments were considerably lower, however, they were also deemed to be underestimated. The large variation in the estimated PAH emissions from specific sources found in the literature illustrates how difficult and uncertain estimations of PAH emissions are, and how the importance of different sources may change over time.

The suggested list of priority pollutants provides guidance for how to select pollutants that ought to be subjected to further study, either by the means of an SFA or by monitoring. The SFA results can complement or substitute empirical monitoring of the pollutant emissions. The procedure and results of this study can be used for predicting the load of OPs in road environments and to propose ways to prevent their emission into the aquatic environment. For example, for the case study area the SFA results suggest that tyres, followed by motor oil leakage, vehicle exhaust gases, road surface wear and brake linings are the main contributors of PAHs to road runoff. For this reason it is of high importance to prioritize the efforts to remove all tyres containing HA oils from the market. In addition, knowledge of the type of pollutants that may reach the stormwater system is useful for the planning of stormwater management and treatment facilities.

The supporting data provides Table A, which shows the results of the laboratory analysis of selected organic pollutants in sediment from the Gårda case study. Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.scitotenv.2016.12.074.

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