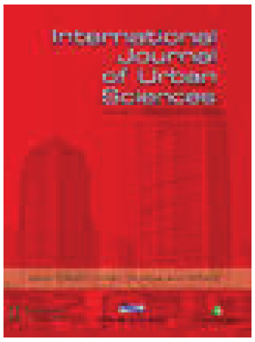

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Review of pollutants in urban road dust: Part II. Organic contaminants from vehicles and road management

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ABSTRACT

Urban road dust is highly enriched with organic pollutants such as polycyclic aromatic hydrocarbons (PAHs), petroleum hydrocarbons, and herbicides that are released primarily from vehicles and/or road management practices. Analysis of sediment cores from urban watersheds clearly demonstrates that increase of pollutant input correlates with traffic volume increase. Pollutants in urban road dust are a significant threat to the health of aquatic organisms. Contaminated urban road dust is mobilized by stormwater runoff and transported into local receiving waterbodies (e.g. streams, rivers, lakes, and estuaries). Concentrations of PAHs in sediments receiving stormwater runoff from densely populated urban areas are high enough to impair the health of aquatic organisms and frequently exceed sediment quality guidelines. To restore streams and rivers impaired by roadway runoff, concentrations of pollutants in road dust need to be reduced through implementing a combination of regulatory policies and management actions. One approach would be to phase out existing pollutant sources such instituting a ban on the use of coal tar-based asphalt road seal coat containing high levels of PAHs. Instituting the use of environment friendly natural herbicides with only spot treatment to target weeds rather than broadcast application and restoration of native vegetation, as integrated roadside vegetation management programs. Adoption of these practices would result in significantly reduced herbicide contamination of roadway stormwater runoff. An alternate option is the removal of contaminants from stormwater runoff before they are delivered to receiving waterbodies using best management practices (BMP) such as retention ponds, detention basins, and grass swales. In densely populated urban areas, however, these BMPs may not be easily adopted due to multiple constraints such as lack of spaces and high land cost. In this case, proprietary BMPs such as media filter, wet vault, and vortex separator, which are also known as manufactured treatment devices, can should be considered.

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1. Introduction

The proliferation of fossil fuel powered motor vehicles is a symbol of economic growth in modern society. The efficient mobility of people and goods from one place to another is

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one the most important components of economic growth and thus the quality and quantity of transportation are very closely related to the level of economic development. Population increase and economic growth have resulted in a significant increase in the number of vehicles and distance driven. For example, the U.S. population increased 1.7 times (181 million to 314 million) from 1960 to 2012 (US Census, 2017). During the same period, the U.S. gross domestic product (GDP) tripled from $543 billion to $16,155 billion (World Bank, 2017) and the total number of registered vehicles increased 3.4 times from 74.4 million to 254 million (USDOT, 2015). The growth of cities and uncontrolled urban sprawl also significantly contributed to the increase of the distance vehicles are driven. Urban land acreage quadrupled from 15 million Acres in 1945 to 61 million Acres in 2007, increasing at about twice the rate of population growth over this period (Nickerson, Ebel, Borchers, & Carriazo, 2011). Total vehicle distance travelled also quadrupled from 1.15 trillion km in 1960 to 4.75 trillion km in 2012 (USDOT, 2015), which was faster than the increase in population and registered vehicles.

While the proliferation of fossil fuel powered motor vehicles created a revolution in physical mobility of population and transport of goods, their operation has resulted in many environmental problems. As presented in the previous companion publication (Hwang, Fiala, Park, & Wade, 2016), daily operation of fossil fuel powered motor vehicles releases many toxic chemicals such as trace metals, fuel combustion byproducts, and petroleum hydrocarbon. Roadway management practices also release toxic chemicals such as herbicides and petroleum hydrocarbons (Hwang, Fiala, Wigart, Townsend, & Edirveerasingam, 2017; Powell, Neal, & Leyva, 1996; Wood, 2001). These pollutants primarily remain on the surface of roads and surrounding areas and can be introduced to local waterways by stormwater runoff.

Road dust is a mixture of particles from multiple sources such as pavement wear, tyre wear, brake pad wear, soil from construction sites, atmospheric deposition, vegetation debris, and others. Abrasion material, most commonly sand, is another dominant source of road dust in areas having snow in winter. Some contaminants, such as engine oil, grease, and pesticides that are released in the form of a liquid, can be adsorbed to dust on road surface. Road dust, especially in urban area, is highly enriched with many organic contaminants from vehicles and roadway management practices (Mahbub, Ayoko, Goonetilleke, & Egodawatta, 2011).

A large fraction of dust on road surface is washed off by stormwater runoff that is mostly discharged into lakes, streams, rivers, and estuaries (Hwang & Foster, 2006; Menzie, Hoeppner, Cura, Freshman, & LaFrey, 2002) and poses significant threats to fish and other aquatic species (Bay, Jones, Schiff, & Washburn, 2003; Beasley & Kneale, 2002; Carr et al., 2000; Logan, 2007; Maltby, Forrow, Boxall, Calow, & Betton, 1995). Road dust can be suspended by vehicles and winds and dispersed to the air (Abu-Allaban, Gillies, Gertler, Clayton, & Proffitt, 2003; Amato et al., 2009; Thorpe, Harrison, Boulter, & McCrae, 2007). Fiala (2017) estimated that approximately 20% of trace metals in road dust is suspended and is redeposited to surfaces within 400 m of the highways.

A national rivers and streams assessment (USEPA, 2013) found that 55% of the assessed rivers and streams (by length) in the U.S. did not support healthy biological communities when compared to the least disturbed sites in similar ecological regions. In addition, the contribution of urban stormwater runoff to the river water quality degradation was found to increased continuously (USEPA, 2013). Urban waterbodies that receive stormwater runoff can experience dramatic water quality impairment, habitat degradation, biological diversity decline, and cause adverse effects to public health and economic activities through exposure via drinking water, fish consumption, and contact recreation (Calvillo, Williams, & Brooks, 2015; Dong & Lee, 2009; Gaffield, Goo, Richards, & Jackson, 2003; Huang, Pedersen, Fischer, White, & Young, 2004; Lee, Maruya, & Bulski, 2004; Meland, Salbu, & Rosseland, 2010; Wu, Jiang, Zhang, Chen, & Zhang, 2014). Benthic species diversity in streams was found to decline with increasing of stormwater runoff input (Klein, 1979; Novotny, Bartosova, O'Reilly, & Ehlinger, 2005; Pitt, Field, Lalor, & Brown, 1995). Elevated prevalence of skin and liver tumours were observed in rivers highly contaminated by PAHs and other carcinogenic contaminants delivered by stormwater runoff (Hwang & Foster, 2016; Pinkney, Harshbarger, May, & Reichert, 2004). Federal, state and local governments invest tremendous amounts of efforts and funds to manage stormwater runoff because it is the prerequisite for the restoration of degraded rivers and streams. Estimated costs of controlling highway runoff range from $2.9 billion to $15.6 billion per year over 20 years requiring 1.6–8.3% of annualized highway transportation expenditures in the U.S. (Nixon & Saphores, 2007). Therefore, it is very important to better understand sources and transport of traffic-related pollutants in order to develop innovative, efficient, practical and cost-effective solutions for the control and reduction of contaminants in stormwater runoff.

The present review covers sources and occurrence of organic pollutants, including polycyclic aromatic hydrocarbons (PAHs), petroleum hydrocarbons, and herbicides, in road dust and roadway stormwater runoff primarily in the U.S. Regulatory policies and best management practices (BMPs) designed to remove pollutants in roadway stormwater runoff are also briefly summarized. Review on trace metals in urban road dust was published in the previous companion article (Hwang et al., 2016).

1.1. Polycyclic aromatic hydrocarbons (PAHs)

Polycyclic aromatic hydrocarbons (PAHs) are a group of hundreds of multiple compounds with two or more fused benzene rings with or without alkyl substitutes that arise mainly from pyrogenic and petrogenic sources (NRC, 1983). Pyrogenic PAHs are produced during incomplete combustion of fossil fuels, organic materials, and wood (Edwards, 1983). Vehicular emission is highly enriched with nonalkylated pyrogenic high molecular weight PAHs (Rogge, Hildemann, Mazurek, Cass, & Simoneit, 1993a; Schauer, Kleeman, Cass, & Simoneit, 1999). Many pyrogenic PAHs such as benzo[a]pyrene and dibenzo[ah]anthracene can be transformed to carcinogen when metabolized by monooxygenase like cytochrome P450. Polycyclic aromatic hydrocarbons are also found in petrogenic sources such as gasoline, diesel, and asphalt (NRC, 1983; Overton et al., 2016; Zielinska, Sagebiel, McDonald, Whitney, & Lawson, 2004). PAH patterns found in road dust and stormwater runoff indicate that pyrogenic inputs are dominate PAHs. The recent increase of PAHs found in urban sediment cores are linked to increase of traffic volumes (Andersson, Klug, Eggen, & Ottesen, 2014; Van Metre & Mahler, 2005; Van Metre, Mahler, & Furlong, 2000).

Incomplete combustion of gasoline and diesel is a significant source of PAHs in urban road dust (Brown & Peake, 2006; Hwang & Foster, 2006; Majumdar, Rajaram, Meshram, & Rao, 2012; Wang, Yang, Jia, Zhou, & Li, 2009). Reported driving distance-based vehicular PAH emission rates have a wide range from 0.1 to 14 mg/km (Lim, Ayoko, Morawska, Ristovski, & Jayaratne, 2005; Schauer et al., 1999; Zielinska et al., 2004) that are highly dependent on various factors such as fuels, vehicle types and age, maintenance status, and driving cycles (Hesterberg, Lapin, & Bunn, 2008; Ropkins et al., 2009;). Rogge et al. (1993a) found much lower tail pipe emission rate of PAHs from vehicles equipped with catalytic converter (52.5 µg/km) than those from vehicles not equipped with catalytic converters (1406 µg/km).

Other sources contributing to the accumulation of PAHs on road surfaces include tyre and pavement wear particles and leaked engine oil. Tyre treads commonly include 20-30% of carbon black and 10–20% of extender oils (Thorpe & Harrison, 2008; Kreider, Panko, McAtee, Sweet, & Finley, 2010). Industrially manufactured carbon black is produced by pyrolysis of organic matter at high temperature and thus formation of PAHs as impurities is unavoidable. Tsai, Shieh, Hsieh, and Lee (2001) found 30–125 µg/g of total PAHs in carbon black. Highly aromatic distilled extract utilized as extender oil in tyre manufacturing process is a byproduct of petrochemical lubricant production and known to contain high levels of PAHs (SCA, 2003). PAHs can be generated during distillation of high-boiling petroleum products as a result of thermal cracking. The European Commission adopted a Directive in 2005 to restrict the use of certain extender oils, which are used to soften tyre rubber, containing more than either 1 µg/g of benzo[a]pyrene and/or 10 µg/g of total PAHs (sum of 8 listed PAHs) for tyre production (European Commission, 2005).

Total PAHs detected in tyres range widely from 10 to 230 µg/g with pyrene as the most abundant (20–50% of total measured PAHs) and followed by benzo[*ghi*]perylene and coronene (Aatmeeyata & Sharma, 2010; Boonyatumanond, Murakami, Wattayakorn, Togo, & Takada, 2007; Rogge, Hildemann, Mazurek, Cass, & Simoneit, 1993b; Sadiktsist, Bergvall, Johansson, & Westerholm, 2012). Aatmeeyata and Sharma (2010) collected tyre wear particles (greater than 10 µm) produced from simulated runs on a concrete road interaction and identified 378 ng of PAHs can be released from a tyre during 1 km of driving of small sedans (378 ng/tyre/km), which is equivalent to 1512 ng/vehicle/km. Driving distance weighted average release of PAHs from tyres of all vehicle fleets, including buses, trucks, and small and big passenger vehicles, would likely be much greater than the release from small sedans because wear rates of each tyre per unit distance are much higher in sport utility vehicles, minivans, buses, and trucks. Buses and trucks have 2 to 4 times more tyres per vehicle than small cars.

Other sources contributing to the accumulation of PAHs on road surfaces include leaked engine oil and pavement wear particles. A fraction of PAHs produced during combustion of fuels can accumulate in engine oil and thus concentrations of PAHs in used engine oils are greatly elevated compared to those in unused engine oils. Benzo[a]pyrene in virgin lubricating oils is much lower (< 1 µg/g) than in used oil from gasoline cars (86 µg/g) (USEPA, 1996). Boonyatumanond et al. (2007) found 97 ± 65 µg/g of PAHs in used engine oils but Clonfero, Nardini, Marchioro, Bordin, and Gabbani (1996) and Brinkman and Dickson (1995) detected PAH concentrations as high as 4600 µg/g and 10,000 µg/mL, respectively. Concentrations of PAHs in engine oils are dependent on various factors such as fuels, vehicle types and age, maintenance status, and driving cycles. Pruell and Quinn (1988) and Wong and Wang (2001) showed a rapid increase of PAHs in vehicle engine oils with miles driven. Engine oils leaked from vehicles are absorbed onto road dust causing elevate concentrations of PAHs.

Coal-tar-based asphalt coating agents contain very high levels of PAHs and are responsible for increased PAH concentrations in many urban streams and lake sediments (Van Metre & Mahler, 2010; Van Metre, Mahler, & Wilson, 2009). The City of Austin, Texas, USA (2005) reported a median concentration of 50,000 μg/g of the sum of 16 PAHs for coal-tar based sealcoat products that is about 1000 times higher than that for asphalt-based sealcoat products. Particles in runoff from parking lots with coal-tar emulsion sealcoat had mean PAH concentrations of 3500 μg/g, 65 times higher than the mean concentrations from unsealed asphalt and cement parking lots (Mahler, Van Metre, Bashara, Wilson, & Johns, 2005). Watts, Ballestero, Roseen, and Houle (2010) also found two to three orders of magnitude higher concentrations of PAHs in stormwater runoff from coal-tar-sealed parking lots compared to stormwater runoff from unsealed parking lots.

Many studies reported highly elevated concentrations of PAHs (0.08–149 μg/g) in urban road dust (Table 1) that are up to four orders of magnitude higher than those found in rural soils. In order to compare data reported in different studies cautions are needed to avoid confusions because total PAH concentrations in various studies are the sum of different sets of PAHs. In addition, some studies have serious analytical errors that have to be taken into account for more accurate data comparison. Unlike to the

Table 1. Concentrations (μg/g) of polycyclic aromatic hydrocarbons (PAHs) in road dust.

Study area	Study sites	Concentration	No. of PAHs	Reference
Sydney, Australia	Urban	8.7 105	16	Aryal, Baral, Vigneswaran, Naidu, and Loganathan (2011)
Sydney, Australia	Urban	1.65 4.0	16	Nguyen et al. (2014)
Niteroi, Brazil	Business	0.43 1.25	21	Netto, Krauss, Cunha, and Rego (2006)
Shanghai, China	Urban	6.88 32.6	16	Liu et al. (2007)
Xincheng, China	Residential	1.63 8.99	16	Hongtao et al. (2009)
Dalian, China	Residential	1.97 10.1	25	Wang et al. (2009)
Dalian, China	Industrial	5.07 17.1	25	Wang et al. (2009)
Guangzhou, China	Residential/ Business	0.84 12.3	16	Wang et al. (2011)
Chang Zhu Tan, China	Urban	3.5 24.5	16	Long, Chi, Qing, Dai, and Wu (2011)
Yangtze River Delta, China	Urban	2.24 58.2	16	Shi et al. (2013)
Lanzhou, China	Residential/ Business	1.47 13.7	20	Jiang, Hu, Yves, Zhan, and Wu (2014)
Tianjin, China	Urban	0.54 34.3	16	Yu, Xie, Ma, Kan, and Zhou (2014)
Xi'an, China	Urban	10.4 26.3	29	Wei et al. (2015)
Cairo, Egypt	Urban	0.045 2.6	16	Hassanien and Abdel Latif (2008)
Banglore, India	Urban	0.048 1.80	17	Tuyen et al. (2014a)
New Delhi, India	Urban	0.65 1.70	17	Tuyen et al. (2014a)
Isfahan, Iran	Urban	0.18 3.22	13	Soltani et al. (2015)
Teheran, Iran	Urban	0.13 1.41	16	Saeedi, Li, and Salmanzadeh (2012)
Japan	Residential	0.062 6.33	16	Ozaki, Akagi, Kindaichi, and Ohashi (2015)
Ulsan, Korea	Residential	45.8	16	Dong and Lee (2009)
Ulsan, Korea	Business	149	16	Dong and Lee (2009)
Ulsan, Korea	Industrial	55.3 112	16	Dong and Lee (2009)
Bangkok, Thailand	Urban	0.15 2.68	20	Boonyatumanond et al. (2007)
Newcastle, UK	Residential/ Business	0.60 46.0	16	Lorenzi, Entwistle, Cave, and Dean (2011)
Santa Monica, CA, USA	Urban	2.2 14.3	15	Lau and Stenstrom (2005)
Hanoi, Vietnam	Urban	0.53 4.70	18	Tuyen et al. (2014b)
Hanoi, Vietnam	Urban	0.13 5.54	25	Tuyen et al. (2014a)

case of lead in road dust (companion publication: Hwang et al., 2016), no clear temporal trends are apparent for PAHs in road dust.

High levels of PAHs in urban road and parking lot dust result in high levels of PAHs in stormwater runoff (Table 2). Like trace metals, PAHs emitted from vehicles deposit on impervious surfaces and are washed off by stormwater runoff. PAHs in the stream and river waters during storm events are two to three orders of magnitude higher than those in base flow (Foster, Roberts, Gruessner, & Velinsky, 2000; Hwang & Foster, 2006). PAHs are highly hydrophobic and quickly adsorbed on the surface of suspended particles when deposited in water. Solid-water partitioning coefficients (K_D) of PAHs vary from 10^3 to 10^7 (Bucheli & Gustafsson, 2000; Hwang & Foster, 2006; Schwarzenbach, Gschwend, & Imboden, 2003), indicating that particles in stormwater runoff scavenge PAHs and act as efficient conveyer transporting PAHs from impervious surfaces to adjacent streams, rivers, lakes, and estuaries. A significant fraction of PAHs bound to particles in stormwater runoff settles and accumulates in sediments close to discharge points. Sedimentary PAHs in rivers, estuaries, and marshes receiving stormwater runoff from adjacent urban areas showed a gradient decreasing sharply downstream of inputs (Hwang, Green, & Young, 2006; Maltby et al., 1995).

There is no practical way to regulate PAHs in vehicle emissions like lead and MTBE because PAHs are not been intentionally added to petroleum products such as gasoline, diesel, and crank case oil, but are produced during incomplete combustion and heating of fuels. Although improved fuel combustion systems, cleaner fuels such as ultra-low sulphur diesel, and devices such as catalytic converters contribute to the significant reduction of PAH emission (Lim et al., 2005; Rogge et al., 1993a), release of PAHs from vehicles is inevitable unless we replace or lower the use of fossil fuel powered vehicles. Due to high PAH content, the use of coal-tar-based asphalt coating agents have been banned in some states (e.g. Maryland, Minnesota, Texas, and Washington) in the U.S. (Coal Tar Free America, 2017). Proper vehicle maintenance to minimize the engine oil leak can also significantly reduce the release of PAHs from vehicles. PAH levels in ingredients such as black carbon and extender oil for tyre manufacturing also need to be further reduced to minimize urban watershed impairments.

1.2. Total petroleum hydrocarbon (TPH)

Major traffic-related sources of petroleum compounds in road dust are leaked engine oil and asphalt pavement wear. Engine oil is a complex mixture of aliphatic and aromatic hydrocarbons, lubrication additives, metals, and various other organic and inorganic compounds. Approximately 80% of engine oil is aliphatic hydrocarbons (primarily alkanes and

Table 2. Concentrations (μg/L) of polycyclic aromatic hydrocarbons (PAHs) in urban road and highway stormwater runoff.

Study area	Study sites	Concentration	No. of PAHs	Reference
Beijing, China	Urban	1 37	15	Zhang et al. (2008)
Heraklion, Greece	Highway	2.66 22.2	16	Terzakis et al. (2008)
Dunedin, New Zealand	Urban	1.20 11.6	16	Brown and Peake (2006)
Winterthur, Switzerland	Highway	1.3 13.1	16	Aryal, Furumai, Nakajima, and Boller (2005)
Bangkok, Thailand	Urban	38 128	16	Boonyatumanond et al. (2007)

cycloalkanes with l-6 rings) and aromatic hydrocarbons (Vazquez-Duhalt, 1989). The lubrication additives, which are approximately 20% of engine oil, consist primarily of zinc diaryl, molybdenum disulfide, zinc dithiophosphate, metal soaps, detergents and dispersants (Vazquez-Duhalt, 1989).

Engine oil can be leaked from poorly maintained vehicles due to various reasons such as improperly installed oil filters, damaged oil drain plug gaskets, and degraded engine gaskets. Leaked engine oil deposits onto roads, driveways, and parking lots and flows into lakes, streams, rivers, and estuaries in association with stormwater runoff. When leaked engine oil comes in contact with road dust, they are strongly adsorbed to the dust. Approximately 80% to 95% of total hydrocarbon load in roadway stormwater runoff is attached to particulates, indicating road dust is a primary vector of transport of petroleum hydrocarbon to receiving waterbodies (Moilleron, Gonzalez, Chebbo, & Thévenot, 2002; Stenstrom, Silverman, & Bursztynsky, 1984). Elevated levels of oil and grease in urban watershed sediments can be used as a tracer for inputs of roadway stormwater runoff.

Engine oil leaks from vehicles, called as silent oil spill, are the predominant anthropogenic source of petroleum hydrocarbon entering lakes, streams, rivers, and oceans (COS and NRC, 2003). Estimated amounts of engine oil released from vehicles onto traffic road surfaces are highly variable because it is extremely difficult to measure them directly. The estimated loss of engine oils during use in vehicles is approximately 16% and 21% in passenger and heavy-duty vehicles, respectively (Kuczenski, Geyer, Zink, & Henderson, 2014). Annual average amount of petroleum entering the marine environment in North America was estimated approximately 65 million litres in the 1990s (COS and NRC, 2003) that is much greater than the amount spilled by Exxon Valdez (41 million litres) in Alaska. Approximately 700 million litres of used motor oil were not properly collected in the U.S. in the early 1990s, and a significant portion of them entered soil and waters in urban areas (USEPA, 1996). Approximately 567 million litres of lubricating oil were sold in California in 2004, while only about 329 million litres were recycled in the same year (CIWMB, 2006).

Asphalt pavement wear particles also significantly contribute to petroleum hydrocarbons in road dust and stormwater runoff (Faure, Landais, Schlepp, & Michels, 2000; Kreider et al., 2010; Unice, Kreider, & Panko, 2013). About 95% of the paved roads in the U.S. are surfaced with asphalt that is mostly produced using hot-mix constructions (Anderson, Youtcheff, & Zupanick, 2000). Hot-mix asphalt pavements consist of about 80% mineral aggregates (e.g. small rocks and pebbles), 15% processed petroleum polymers as binders, and 5% air voids (Anderson et al., 2000). The most common form of petroleum polymer is asphalt, which is a highly viscous residue produced by the non-destructive distillation of crude oil during petroleum refining. Asphalt binders are waterproof and thermoplastic glues that hold mineral aggregate materials such as small rocks and gravels together.

As asphalt surface is aged, asphalt binder become hardened and asphalt wear particles can be generated during mechanical interaction between pavement materials and tyres with much higher wear rates in damaged roads. Damages (e.g. rutting, fatigues, and cracks) to the asphalt pavement surfaces can be caused primarily by traffic, weather, and ageing. To improve winter time traffic road safety, almost all countries having a considerable amount of snowfall use some road maintenance equipment such as snow plow to remove snow from traffic roads. During this practice, snow plows equipped with steel blades grind road surfaces and generate large amounts of pavement wear particles (Hwang et al., 2017). Driving vehicles equipped with snow chains or studded tyres also increase wear rates of asphalt pavement (Lindgren, 1996). Fiala (2017) reported asphalt wear rate of 11 g/km per vehicle during winter. About 10–20% of fine solids (< 20 µm) in highway stormwater runoff collected in the Lake Tahoe Basin in the State of California, USA was attributed to asphalt pavement wear (Hwang et al., 2017).

The existence of petroleum compounds in environmental samples can be represented by analyses of total petroleum hydrocarbon (TPH) or oil and grease (O&G). Total petroleum hydrocarbons are typically quantified using organic solvent extraction and instrumental analysis with gas chromatograph-flame ion detector (Overton et al., 2016). O&G are quantified using organic solvent extraction and gravimetric measurement. Concentrations of petroleum hydrocarbons in road dust and stormwater runoff are highly variable and dependent on many factors such as traffic volume, pavement type, season, and precipitation frequency, amount and rate.

Concentrations of TPH were much higher in highway road dust (1680 µg/g) than in road dust collected from commercial (157 µg/g) and residential (353 µg/g) areas (Latimer, Hoffman, Hoffman, Fasching, & Quinn, 1990). Concentrations of unresolved complex mixture and n-alkanes in road dust collected from the central part of the City of Kuala Lumpur, Malaysia were 450 ± 281 µg/g and 7.36 ± 4.77 µg/g, respectively, that is much higher than those in road dust from rural areas (Omar et al., 2007). Mean TPH levels in dust from high traffic roads in the Tshwane Metropolitan area, South Africa ranged from 404 to 852 µg/g (Okonkwo, Awofolu, Moja, Forbes, & Senwo, 2006). Median TPH in the first flush of road runoff collected from 21 sites in Brisbane, Australia ranged from 46 to 1850 µg/L (Drapper, Tomlinson, & Williams, 2000). Total aliphatic hydrocarbon in road runoff in the central part of Paris, France ranged from 393 to 1359 µg/L. Concentrations of O&G typically found in urban stormwater runoff range from 1000 to 480,000 µg/L (Chiew, Mudgway, Duncan, & McMahon, 1997; Lopes & Dionne, 1998). Concentrations of O&G in highway stormwater runoff in California, USA ranged from 2000 to 80,000 µg/L with a mean of 14,000 µg/L (Han, Lau, Kayhanian, & Stenstrom, 2006). O&G are typically much higher at the beginning of storm events, which is known as first flush, compared to later phase in the same event (Irish et al., 1995).

Suspended solids in stormwater runoff have been found to be consistently enriched with petroleum hydrocarbons (24,800 µg/g) compared to street dust (1680 µg/g), which may be attributed to that smaller particles contain higher levels of petroleum hydrocarbons (Barrett, Irish, Malina, & Charbeneau, 1998) and are more easily suspended by stormwater runoff and carried into storm drains (Latimer et al., 1990). Total hydrocarbon extracted from sediments from urban waterways on the East and West U.S. coasts ranged from 8 to 2350 µg/g, with a mean and median concentration of 415 and 244 µg/g, respectively (Stout, Uhler, & Emsbo-Mattingly, 2004).

In the U.S., used engine oil is classified as a hazardous waste due to severe impacts on soil and water quality. It is known that one litre of used oil can contaminate one million litre of water (USEPA, 1996). The Clean Water Act states that discharge of used engine oil into the waters of the U.S. within 12 miles of the coast is illegal. Used engine oil must be properly collected and recycled in accordance with the Resource Conservation Recovery Act. Collected engine oil can be re-refined into a base stock for new lubrication oils, asphalt products, or a fuel for large industrial boilers (Boughton & Horvath, 2004). Petroleum hydrocarbon concentrations in sediments and in water supplies declined after the enactment of the Resource Conservation Recovery Act through improved regulations and management of hazardous waste. More careful maintenance of vehicles can further reduce petroleum hydrocarbons in road dust that can eventually improve the quality of urban watersheds.

1.3. Herbicides

A practice known as integrated roadside vegetation management uses one or more techniques such as mowing, herbicide application, prescribed burning, and biocontrol to restrict the growth of a variety of weeds and brushes that may cause physical road surface damages and fires, interfere with the function of stationary object (e.g. guardrails, signposts), impair drivers' vision, and produce unpleasant views. Large amounts of herbicide application for roadside vegetation management can be traced to an influential book – *Roadside, the front yard of the nation* by Jessie Bennett published in 1936 (Quarles, 2003). Costs of labour-intensive mowing and weeding significantly increased after the World War II that resulted in considering herbicide application as a less expensive alternative (Quarles, 2003). Roadside herbicide application has also increased to control invasive plants and noxious weeds (e.g. poison ivy, musk thistle, ragweed). Henderson (2000) surveyed 14 states in the U.S. and found that budget they spent for herbicide application was at least equivalent to what they spent on mowing. The California Department of Transportations and road agencies of counties in the State of California applied approximately 500,000 L of liquid herbicides and 42 metric tons of dry herbicides annually on roadsides (Clary, 1999). Due to public concerns regarding the environmental risk of herbicide application on highway right-of-way, the California Department of Transportation reduced the use of herbicides 51.6% by 2000 and set a goal of 80% reduction compared to a baseline usage in 1992–1993 by 2012 (Shields, 2001). However, the California Department of Transportation was not able to meet this goal due to an increase in shoulder distance, fire protection, and noxious weed control mandates, and requests from local cities and counties (Caltrans, 2013).

Contamination of road dust and roadside soils by herbicides has been less recognized compared to trace metal and PAH contamination. Pre-emergence (e.g. diuron, oxadiazon, and simazine) and post-emergence (e.g. 2,4-D, clopyralid, glyphosate, picloram) herbicides have been widely applied along the right-of-way of traffic roads (Fleischer, Gaylor, Dickens, & Turner, 1989; Powell et al., 1996; Quarles, 2003; Ramwell, Heather, & Shepherd, 2002; Wood, 2001). Overuse of herbicides can also weaken non-target species, making them more vulnerable to weed invasions (Brandt, Henderson, & Uthe, 2011). Herbicides deposited on the surface of roads and roadside soils can be washed off by stormwater runoff and delivered to local waterbodies and can pose adverse effects to aquatic organisms. Concentrations of herbicides in roadway stormwater runoff are highly variable depending on herbicide application rates, precipitation rates, time gaps between application and precipitation, seasons, and other physical and chemical factors such as hydrophobicity and photodegradation (Huang et al., 2004; Wood, 2001). Maximum concentrations reported for glyphosate, atrazine, diuron, oxadiazon and oryzalin in roadway stormwater runoff collected in the City of Derby, UK were 650, 2210, 1810, 390 and 70 µg/L, respectively (Ramwell et al., 2002). Powell et al. (1996) reported concentrations of 29–337 µg/L of simazine and 46–2849 µg/L of diuron in highway shoulder stormwater runoff in the State of California in the early 1990s. Huang et al. (2004) and Wood (2001) found approximately two to three orders magnitude lower concentrations of herbicides in highway stormwater runoff collected in the late 1990s in the State of California and the State of Oregon, respectively.

Among herbicides commonly applied to the roadside, glyphosate has a strong affinity to solids, whereas other herbicides are much more hydrophilic. Organic carbon normalized solid-water partitioning coefficient of glyphosate is 1 to 2 orders of magnitude higher than those of other commonly applied herbicides such as atrazine, diuron, and oxadiazon (Gerritse, Beltran, & Hernandez, 1996; Oliveira, Koskinen, & Ferreira, 2001), indicating that wash-off rates of glyphosate are much lower compared to other more hydrophilic herbicides that pose greater health risk to aquatic organisms.

Due to adverse impacts on aquatic organisms, the frequencies and amounts of artificial herbicide application have declined as in the State of California. Application of environment friendly natural herbicides such as Finale (glufosinate ammonium) can reduce the use more harmful artificial herbicides (Nowak, 2013). Spot treatment to target weeds, rather than broadcast application, has been also adopted to reduce artificial herbicide application amounts. Restoration of native vegetation, which requires less herbicide application, as a part of integrated roadside vegetation management programmes also significantly contribute to less herbicide application.

2. Best management practices

The proliferation of fossil fuel powered motor vehicles and the increase of total distance travelled by them are closely linked to a significant increase of pollutants in almost all urban watersheds. The growth of cities and uncontrolled urban sprawl boosted land development and greatly increased transportation-related impervious covers such as paved roads and parking lots that account for at least one-third of the developed urban land. As impervious covers increase, volume and velocity of stormwater runoff also increase and results in greater erosion power that washes off the dust from road surfaces vigorously and carries more pollutants to receiving waterbodies (Blansett & Hamlett, 2013). These processes are enhanced by heavy rains, tropical storms, and hurricane, and these episodic events can transport road dust to receiving waters over a short period.

To reduce water quality impairment by roadway stormwater runoff, appropriate source and/or treatment control best management practices (BMPs) need to be adopted. BMPs refer to any procedures designed to control or abate stormwater runoff pollution. The optimal goal of all source and treatment control BMPs is the removal of pollutants in stormwater runoff prior to discharge into receiving water. Once toxic constituents in stormwater are dispersed in receiving waterbodies, it is extremely difficult to avoid exposure of living organisms to these pollutants.

Source control BMPs have been implemented to remove pollutants before they enter the storm sewer system. Generally, proactive source control BMPs are easier and much more effective than treatment control BMPs to prevent environmental impacts of pollutants from traffic activities. Although non-fossil fuel powered vehicles such as electric and fuel-cell vehicles account for less than 5% of the total passenger and cargo vehicles, their market share will grow continuously that will result in significant reduction of emission from fossil fuel combustion. Road dust from various sources typically accumulate at the edge of roads so road sweeping and road vacuuming have been implemented to remove road dust (Amato, Querol, Johansson, Nagl, & Alastuey, 2010; Calvillo et al., 2015; Walker, Wong, & Wootton, 1999). Road sweeping can remove large amounts of road dust but is inefficient in reducing pollutants in roadway stormwater runoff because a vast majority of pollutants are associated with smaller (< 250 um) particles not efficiently remove by sweepers (Calvillo et al., 2015). Road vacuuming can remove a substantial amount of smaller road dust along with associated pollutants (Calvillo et al., 2015 and references there in).

If source control BMPs cannot be adopted, treatment control BMPs need to be considered to retain pollutants in stormwater runoff before they enter receiving water. Treatment control BMPs can be classified as structural BMPs and proprietary BMPs. Structural treatment BMPs such as detention ponds, retention ponds, and grassed swales, infiltration trenches, and porous pavement can be used to physically trap contaminants in stormwater runoff before discharged into receiving water. In structural BMPs, contaminants in stormwater runoff settle out or are filtered through the underlying soil. Structural treatment BMPs showed effectiveness in removal of both trace metals, nutrients, and organic contaminants in roadway stormwater runoff (Bäckström, 2003; Carpenter, Vallet, Pelletier, Lessard, & Vanrolleghem, 2014; Collins et al., 2010; Ellis, Revitt, Shutes, & Langley, 1994; Hogan & Walbridge, 2007; Leroy et al., 2016; Scholes, Shutes, Revitt, Forshaw, & Purchase, 1998; Stagge, Davis, Jamil, & Kim, 2012). Detention and retention ponds can also be used for flood control (Braune & Wood, 1999; Bressy et al., 2014; Nehrke & Roesner, 2004). However, in dense urban areas, some structural treatment BMPs such as detention and retention ponds may not be easily adopted due to multiple constraints such as lack of spaces and high land cost to install these BMPs. In this case, proprietary BMPs such as media filter, wet vault, and vortex separator, which are also known as manufactured treatment devices, can be considered (Cates, Westphal, Cox, Calabria, & Patch, 2009; Faram, Iwugo, & Andoh, 2007; Tran & Kang, 2013; Wilson, Gulliver, Mohseni, & Hozalskio, 2007).

3. Conclusions

Motor vehicles release tremendous amounts of chemicals of concern into the surrounding environment. Analysis of sediment cores from urban watersheds clearly demonstrates that an increase of pollutant input correlates with traffic volume increase. Incomplete combustion of fossil fuels generates carcinogenic compounds, such as PAHs. Pavement wear and pavement management practices also contribute a significant amount of toxic chemicals, including PAHs, petroleum hydrocarbons, and herbicides.

Almost all urban waterways have experienced severe degradation of water and sediment quality. Many ecological and toxicological evidence shows organic pollutants, especially PAHs, caused severe habitat degradation for aquatic organisms and adverse effects on them. To restore streams and rivers impaired by roadway runoff, a combination of regulatory policies and management actions must be implemented. The best option is phasing out existing pollutant sources. Phase out of coal tar-based pavement sealing agent is a good example demonstrating the effectiveness of regulations on the significant reduction of PAH loadings that has led to the improvement of water and sediment quality in urban watersheds. However, some sources (e.g. tail pipe emission, tyres, pavement wear) cannot be phased out because cost-effective alternate techniques or products are not available in the market.

Unlike lead and copper that were intentionally added to gasoline and brake pads, respectively, to improve their performance, PAHs are produced from incomplete combustion of gasoline and diesel. Thus it is difficult to control the emission of PAHs through limiting levels of PAHs in fuels. Some devices such as catalytic converters and diesel particulate matter filters significantly reduce PAH emission, however, it is very hard to further reduce PAH emission. Although market shares of non-traditional fossil fuel powered vehicles (e.g. hybrid, electric, and fuel cell powered vehicles) are still low, they will replace fossil powered passenger and cargo vehicles continuously, and PAH emission from vehicle engines will hopefully decline in the future. Application of environment-friendly natural herbicides, spot treatment to target weeds, rather than broadcast application, and restoration of native vegetation, as parts of integrated roadside vegetation management programmes, can be adopted to significantly reduce herbicide in roadway stormwater runoff.

If emission of pollutants from associated sources cannot be managed properly, single or sequential BMPs should be implemented to avoid delivery of pollutants into local waterways by roadway stormwater runoff. In highly developed urban areas, more efficient and cost-effective proprietary BMPs need to be developed to overcome roadblocks (e.g. lack of space, cost, treatment capacity) in implementing BMPs.

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