INVITED REVIEW PAPER

Urban stormwater quality and treatment

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Abstract−Stormwater runoff from urban and suburban areas generates numerous pollutants. The areas include residential areas, parks, commercial areas, industrial areas and road/highways. Land use and human activities largely determine the nature and level of pollutants. Among the various types of pollutants that can contribute to adverse water quality impact in receiving water bodies, of concern are suspended solids, heavy metals, polycyclic aromatic hydrocarbons and nutrients. This paper provides a critical review of the characteristics and sources of urban stormwater pollutants and the manner in which the pollution occurs. Treatment systems for urban stormwater runoff and for urban stormwater harvesting are discussed.

Key words: Stormwater, Heavy Metals, Suspended Solids, Polycyclic Aromatic Hydrocarbons, Nutrients

INTRODUCTION

It is believed that by 2020, more than half the world's population will reside in urban areas. More urbanization will lead to increasing imperviousness of the earth's surface which in turn leads to i) increases in peak flows, runoff volumes, pollutants in the first flush and their peak load and ii) reductions in the groundwater recharge by blocking infiltration of water into the ground. Reductions of groundwater recharge may cause the decrease in dry weather base flow in water courses and the alteration of the catchment hydrology.

In recent times urban stormwater runoff and its associated non-point pollution sources have been considered as one of the major

Table 1. Possible sources of pollutants in stormwater runoff

	Soil erosions	Vehicles	Human/animal waste	Fertilizers	Household chemicals	Industrial processes	Paints and preservatives
Solids	■	■	■				
Metals	■						
Oil, greese and organics		■			■	■	
Nutrients	■		■	■			

Table 2. Possible sources of pollutants in road runoff

	Brakes	Tyres	Frame and body	Fuel and oil	Pavement	De-icing salt	Litter
Cadmium		■					
Chromium			■				
Copper	■	■					
Iron			■				■
Lead			■				
Nickel			■				
Zinc	■	■	■				
Org. solids					■		■
Inorg. solids					■	■	
PAHs				■	■		

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sources of water pollution [1,2]. Unlike traditional point sources, which are easily identifiable and measurable, non-point pollution sources are difficult to characterize and quantify since they are highly variable [3-5]. The contaminants originate not only from land-use but also from atmospheric deposition. There are many aspects of non-point source pollution that makes it difficult to identify, categorize, control and prevent. The impact of stormwater pollutants on receiving water bodies depend on number of factors such as the nature of pollutants, their concentration, mixture of pollutants, and the total load of the pollutants. There are many pollutants in urban stormwater. The major categories are: (i) total suspended solids (TSS), (ii) heavy metals, (iii) polycyclic aromatic hydrocarbons (PAHs) and (iv) nutrients. These pollutants often go through various physico-chemical processes before they impact on the aquatic community. This paper reviews these four major categories of pollutants.

Table 1 summarizes the pollutants commonly found in stormwater and their possible sources. Table 2 summarizes the pollutants from road runoff. The table shows that most pollutants have diverse sources. Likewise major sources emit most types of pollutants. Atmospheric fallout also contributes a significant amount of pollutants to stormwater runoff.

SEDIMENT AND SUSPENDED SOLIDS

These particulates mainly originates from exhaust gas, traffic, asphalt/building erosion, soil, sand and silts transported by wind and other means and consists of various ranges of particle sizes. Urban runoff contains a wide range of particulate matters ranging in size from smaller than 1 μm to greater than 10,000 μm [6]. Sediment pollutant levels can be measured as Total Suspended Solids (TSS) and/or turbidity. The TSS is a measure of the total mass of suspended sediment particles in water. The particulates (as a dust or as suspended solids in runoff) are enriched with several types of organic and inorganic pollutants and during wet weather they are washed off along with associated pollutants. These particles can play an important role in transporting and partitioning chemicals in aquatic ecosystem.

Once washed off they are known as suspended solids (SS), which increases the turbidity of the receiving water, and reduces the penetration of light, resulting in decreased activity and growth of the photosynthetic organisms. Clogging of fish gills has also been attributed to the presence of SS. The settling of SS in receiving water poses a long-term threat because of their oxygen demand and gradual accumulation of toxic pollutants. Most of the stormwater pollutants are found in particulate form attached to finer sediment fractions and/or affiliated to organic matter. In this way the fate of pollutants are associated with sedimentation or the removal of sediment. Scouring and mixing of sediments during periods of stormwater discharge may remobilize and release those toxic contaminants that have previously settled on the channel floor. During transport, the pollutants may transform from one form to another or may degrade. As a pollutants carrier, SS is considered as an important index for stormwater pollution [7].

1. Sediment Load and Suspended Solid Concentration

The most prevalent form of pollutant in urban runoff is sediment. Conversion of forest areas to agricultural and human habitat release large amount of sediments during stormwater events. Wol-

man and Shick, 1967 [8] discussed post-colonial land-use change in the north-eastern United States and its effect on sediment yield during storm events. They estimated an increase of sediment yields from 35 metric tons km^{-2} to 210 metric tons km^{-2} when forest is converted to cultivated land. Roberts and Pierce, 1976 [9] reported that the Patuxent River (USA) had more than doubled its sediment yield after urbanization of 344 metric tons km^{-2} compared to pre-urbanization values of 143 metric tons km^{-2}.

Pope, [10] reported the maximum sediment concentration in urban catchments was in the order of 15,000-36,000 mgL^{-1} with average concentrations normally in the range between 2,000-3,000 mgL^{-1} in USA. Their urban studies highlighted the very considerable potential of surface and constructional erosion, human usage and dust fall contributions to the SS loading on receiving rivers. Another investigation by Pirner and Harms [11] in a 7 km^2 urban catchment in South Dakota, USA showed discharge of nearly 180 tonnes of sediment into a stream flow of nearly 2,000 Ls^{-1} with over 50% of the particulates discharging in the first half hour of the storm (first flush). An extreme case was presented by Bryan [12] in Durham, North Carolina where a 4.3 km^2 catchment yielded an annual load of 7,657 tonnes. Mance and Harman [13] revealed an annual load of 248 tonnes was discharged into the River Rib from the 2.5 km^2 catchment of Stevenage New Town, USA displaying a typical mass-loading from the urbanised catchments. Trimble [14] studied the contribution of stream channel erosion to sediment yield from an urbanizing catchment. He examined the contribution of channel-bank erosion to sediment yield from the 228 km^2 San Diego Creek catchment, an urbanizing catchment in southern California. The study showed that sediment production from channel enlargement accounted for approximately two-thirds of the measured suspended sediment yield and downstream sediment accumulation. Soil erosion tests to assist in predicting soil loss and subsequent increase in TSS leaving a highway construction site during rain was tested by Liu et al. [15] for one hundred and sixty-two rainfall events. The soil with a higher shear strength resisted soil erosion better than those with a lower strength. Soil loss was nearly independent of shear strength at low rainfall intensity but at high intensities, the shear strength was more important in resisting soil loss. Lower soil loss can be expected for cohesive soils if the compressive strength is high. McPherson et al. [16] compared the pollutants load in wet weather and dry weather periods in the highly urbanized Ballona Creek catchment. They found that wet weather flow is the predominant source of TSS (99%).

Nelson and Booth [17] studied the role of human activity, particularly urban development, on sediment sources in the Issaquah Creek catchment in western Washington USA. They estimated an increase of nearly 50% in the annual sediment yield. The main contributors of sediment in the catchment were landslides (50%), channel-bank erosion (20%), and road-surface erosion (15%). Goodwin et al. [18] also reported higher sediment yields from urban sub-catchments than rural sub-catchments when investigating the temporal and spatial variability of sediment transport and yields in the Bradford Beck catchment (UK) although the annual yields were comparable. Furthermore, for large events, urban sediment transport was dominated by the impact of Combined Sewer Overflow (CSO) discharges. Within these events, peak SS concentration, were generally higher than rural systems. Lee and Heaney [19] used hydrologic analysis from

about 52 years of long-term rainfall data to demonstrate that imperviousness of the surface is an important indicator of the impact of urbanization on stormwater systems and has a direct influence on stormwater quality and quantity. Identification of SS sources in urban stormwater runoff using radioactive tracers ([7]Be and ^{210}Pb) was done by Yin and Li [20]. They found that $60\pm12\%$ of SS at the sewer outlet originated from the drainage system sediments while the rest was from the wash-off of urban ground dust during the rainfall events.

2. Particle Sizes in Sediment and Stormwater Runoff

The size distribution of suspended solids as well as its composition can have a significant influence on the water quality [21,22]. Sartor and Boyd [23] reported that over 50% of metals were found sorbed to sediment particles less than 43 μm. Although the fine solids constituted only 5.9% of the total solids collected, this portion of the sediment contained approximately one-third to one-half of the algal nutrients, nearly three-quarters of the total pesticides and over one-half of the heavy metals. Bradford [24] also found that the fine fraction of street dust accounted for approximately 6% of the total mass of solids and greater than 60% of the trace metals. Ellis et al. [25] investigated properties of stormwater sediment that could be useful as a fingerprint for source identification. They studied the particle size distribution in urban runoff sediment in different runoff events. The particle size distribution in the runoff was correlated to hydraulic conditions and antecedent dry weather conditions.

Sartor et al. [26] measured the particle size distribution in dry deposited road pavement particles. Their result showed that 15% of particulate matter were less than 104 μm, 28% were between 104 and 246 μm, 25% between 246 and 840 mm, 8% between 846 and 2,000 μm, and 24% larger than 2,000 μm. Shaheen [27] found that about 10% of the particles were less than 75 μm, 32% between 75 and 250 μm, 24% between 250 and 420 μm, 19% between 420 and 850 μm and 15% between 850 and 3,350 μm accounting more than 58% of the particles greater than 250 μm. However Pitt et al. [28] under the US Environment Protection Agency's (USEPA) National Urban Runoff Program (NURP) found that most of the particles (60-100%) were finer than 100 μm. Ball et al. [29] measured the particle size distributions in a broad range and found the mean median diameter of particulates was approximately 70 μm (for particles smaller than 200 μm) while the 10 and 90 percentile diameters were approximately 16 and 189 μm, respectively. On average, 34% of the fine sediment collected was less than 43 μm. Since the average fine sediment constituted 16.8% of the total sediment load, approximately 6% of the total sediment collected from the road surface was less than 43 μm.

Yang et al. [30] investigated the distribution of particle sizes of fine sediment (<20 μm) contained in collected road dust samples using a 10-stage cascade (available particle cut-size diameters are 0.056, 0.166, 0.31, 0.52, 1.0, 1.8, 3.2, 5.6, 10 and 18 μm) at Kaohsiung city in Taiwan. The cumulative percent (%) of particle size distribution for total particle mass at the particle size <1.0 μm was (4.26%, ~2.21%), <2.5 μm was (7.01-10.7%), <10 μm was (33.0-55.6%) for the urban, steel plant, cement plant and seashore areas, respectively. Kobriger and Geinopolos [31] reported the distribution of particles from vehicle-related deposition processes. 37% arose from pavement wear; 37% from tire wear, and 18.5% from abrasion of vehicle parts, such as brakes and engines. Deposition from

settleable exhaust accounts for 7.5% of the total particulate mass.

Randall et al. [32] found that over 80% of the total particles in stormwater runoff were less than 25 μm. Due to different hydraulic conditions in runoff, the washed off particles show a different nature compared to those deposited on the surface. Kobriger [33] found the particle size in stormwater from highways was less than 88 μm.

Roger et al. [34] studied the particle size distribution in motorway runoff water at Herault, France and found 90% of the solid matter by weight was less than 100 μm. Almost 78% of the solid matter by weight was less than 50 μm. The particles less than 50 μm composed of clay (56%), quartz (15%), chalk (12%), organic matter (9%), feldspars (5%) and dolomite (2%). Sansalone et al. [35] considered larger particles in runoff from a freeway and found that approximately 10% of the solids by mass were less than 100 μm, 25% to 60% of the mass were between 100 and 400 μm, and 40% to 70% of the mass were larger than 400 μm. Their result showed that urban stormwater runoff from paved surfaces transports a wide gradation of solids ranging in size from smaller than 1 μm to greater than 10,000 μm. Particles between 425 to 850 μm in size contributed the greatest total surface area and results indicated that 20% of particulate matter was from 600 to 1,000 μm and 30% was from 1,000 to 10,000 μm. Particle counts from the pavement exhibited a first flush. Andral et al. [36] investigated the particle size distribution and hydrodynamic characteristics of solid matter carried by runoff from motorways. They found that the total SS concentration ranged between 16-58 mgL[−1] in runoff samples. They compared the sediment particle size in the runoff samples with sediment deposited in the channel during rainfall. Their study revealed that the runoff contained approximately three-quarters of fine particles (<50 μm). On average runoff contained 78% of fine particles (<50 μm) and 9% of particles between 50 to 100 μm. Channel sediment composed of large particles, with 53% being between 500 and 1,000 μm in diameter. Herngren et al. [37] used simulated rainfall to study particle size distribution in SS in urban stormwater. Their study showed that the majority of the road dust particles were below 76 μm, and 0.45-75 μm size class dominated in all runoff samples (up to 85%). They also observed approximately 22% of the particles in runoff samples were larger than 300 μm. However, Furumai et al. [38] measured the particle size distribution in road runoff in Switzerland. The finding showed that the particles larger than 250 μm were rare. They observed a different wash off behavior of particles in runoff.

Aryal et al. [39] highlighted the dynamic behavior of SS. Their results showed that fine (<45 μm) and coarse particles (>45 μm) have different wash off behavior. A majority of the particles were less than 106 μm. The concentration of the fine fractions (<45 μm) was found to be less dependent on rainfall characteristics as compared to the coarse fractions (>45 μm). The fine fraction attained almost a similar concentration (saturation state) after crossing a certain TSS concentration threshold whereas the coarse fraction was more influenced by TSS concentration. Similar results were reported by Kim and Sansalone [40] who studied event-based size distribution of particulate matter transported in eight urban runoff events. Their study revealed that on an event basis, fine particulate matter (<75 μm; the suspended and settleable fractions) accounted for 25-80% of the gradation on a mass basis, and gravel-size particulate matter (42,000 μm) ranged from 0.5% to 30%. The mean of d_{50} (size with 50% of total particulate matter) for all eight events was 136

µm corresponding to a Φ_{50} size (corresponding to 50% of the cumulative frequency) of 3.34 µm. They concluded that the particulate matter in rainfall-runoff were similar in size to very fine sand. The particle size distribution (PSD) results showed that the mass fraction of particulate matter (PM) larger than 1,000 µm (a typical upper size limit measurable by various optical particle size analyzers) accounted for up to 35% of the total mass load of PM with the median of 22%. One important finding of this study was that the medium to coarse sand sizes (200-2,000 µm) could comprise a significant portion of the total PM load in source area runoff, and therefore should be accounted for during sampling and analytical PSD procedures. Murakami et al. [41] reported that the wash off behaviour of SS depends on particle size as well as their density. Li et al. [42] studied the particle size distribution (2 and 1,000 µm) in highway runoff during a rainy season at three highway sites in west Los Angeles. They observed particle aggregation with time and the concentration of small particles decreased with a corresponding increase in the concentration of larger particles in stored samples. The particle concentration decreased as the storm progressed and the number of large particles decreased more rapidly than the total number of particles. Particles demonstrated a strong first flush. On average, 40% of the particles were discharged in the first 20% of the runoff volume. Faram et al. [43] found that sediment characteristics largely varied from site to site with particle D_{50} values ranging from 7-112 µm. The construction site sediment was the finest overall (D_{50} from 7 to 8 µm), followed by park and ride and urban road sediments (D_{50} from 11 to 15 µm) in samples collected from a hydrodynamic vortex separator (Downstream Defender (DSD)).

Westerlund and Viklander [44] analysed road runoff in northern Sweden with respect to the concentrations of and the loads of particles in different size fractions (4-6, 6-9, 9-15, 15-25, 25-40, and 40-120 µm) between a snowmelt period and a rainfall period, as well as during events within each period. The results showed that there was on average, eight times higher concentrations and five times higher loads of particles during the snowmelt period compared to the rain period for all particle size intervals. The difference was explained by the lower runoff volume during snowmelt compared to rainfall runoff and the long residence time of a snow-bank, where the snow becomes continuously polluted. Sansalone and Buchberger [45] also produced higher SS concentration in snow melt induced runoff compared to rainfall runoff. Cristina et al. [46] studied traffic generated particles in urban highway snowmelt runoff from 10 highway shoulder sites in urban Cincinnati, Ohio, USA. They observed very high concentration of particulate matter in highway snowmelt exceeding 10^5 mgL^{-1}. They focused on the finer fraction of particles (15-75 µm) and coarser fraction of particles (75-4,705 µm). Their result showed that heavy metals were mostly associated with the coarser fraction of particles. Their result also revealed significant difference in densities for coarse (2.86 g cm^{-3}) and fine particle (2.75 g cm^{-3}).

Krishnappan et al. [47] measured the SS size distribution in stormwater ponds in Canada during different seasons using a submersible laser particle size analyser. They found that then SS matrix was complex and contained mostly flocs of various sizes. The sizes and density were at minimum levels during winter (30 µm) and at maximum levels during summer (212 µm). They concluded that flocs in the size range between 5-15 µm would settle faster than the smaller

particles of higher density and larger flocs of lower density. The larger flocs were also found to be susceptible to break up by turbulence.

3. Summary

- A change in land use and human activities are major causes in the increase of sediment loads to the nearby aquatic environments such as rivers and lakes.
- Impervious surfaces such as roads and highways cause an increase of sediment load to aquatic environments.
- A wide range of total suspended solids concentration between 1-3,600 mgL^{-1} has been reported. The size of the particles reported in stormwater runoff and range from a few micrometers to millimetres. Most of the observed cases have particles less than 75 µm. The nature of sediment particles could be useful in identifying possible sources.
- Seasons have large influence on sediment load. Results show much higher sediment concentrations as well as loads during snowmelt compared to rainfall periods.

HEAVY METALS

Many different definitions exist for heavy metals. Some are based on density, some on atomic weight while others are based on their chemical properties and their toxicity. In urban environments, heavy metal usually refers to toxic metals that originate from human activities. Human and living organisms require these toxic elements for their body functions but only in trace amounts. Excess levels of these heavy metals can damage human health and ecosystems. The most common heavy metals of concern are Pb, Cr, Zn, Ni, Cu and Cd.

Heavy metals are the most prevalent substances in urban dust and runoff. They are of particular interest in stormwater runoff due to their toxicity, ubiquitousness, and the fact that they are not easily chemically transformed and remain in the system for long time threatening the environment. The heavy metals of most concern in the environment are chromium (Cr), nickel (Ni), zinc (Zn), copper (Cu), lead (Pb), vanadium (V), cobalt (Co), cadmium (Cd) and mercury (Hg). Due to their toxicity, heavy metal discharge to the environment has been regulated by laws throughout the world. The restriction level varies with the type of heavy metal and from country to country. Heavy metals level has been investigated in road and soil particles, river, lake and coastal sediments and in urban runoff.

Davis et al. [48] estimated the contribution of heavy metals from different specific sources. Their estimation showed that about 75 µg of Cu, 3 µg of Pb and 89 µg of Zn per km-vehicle are released from the brakes only. They reported brake was as an important source of Cu while tire wear was the major source for Zn. The major source of Cr is the body surface of vehicles, which is coated with hexavalent Cr for corrosion prevention.

1. Heavy Metals in Soil and Sediment

Ellis and Revitt [25] studied several heavy metals in the sediment on street surfaces as a function of sediment grain size. The distribution pattern of Pb, Cd, Cu and Zn were related to level and type of traffic densities. Their study showed that concrete motorways had particles less than 250 µm whereas other locations having asphalt surfaces exhibited varying degree of wear, which provided considerable amount of free coarse materials. Heavy metals occurred in the ranges reported in Table 3.

Table 3. Heavy metals range (Ellis and Revitt [25])

	Size (μm)	Pb (μg g^{-1})	Cd (μg g^{-1})	Mn (μg g^{-1})	Zn (μg g^{-1})	Cu (μg g^{-1})	Fe (μg g^{-1})
Sediment	<250	40-1690	0.72-4.2	766-855	119-2133	42.6-640	6780-22700
	>250	111-2296	1.3-6.8	694-1244	91.6-1760	27.2-212	4195-22850

Byrne and DeLeon [49] conducted a study on heavy metals in 8 sediment samples along the Lake Pontchartrain, Lousiana, USA to determine the distribution and sources of heavy metal pollutants in the sediments. The heavy metals measured were barium (4-207.4 μg g^{-1}), copper (0.5-46.2 μg g^{-1}), nickel (2.3-18 μg g^{-1}), lead (0.7-231 μg g^{-1}), and zinc (8.4-169 μg g^{-1}). They linked the high values of heavy metals concentration with urban stormwater runoff and municipal discharges. The lowest level were found in the sediment where human activities were minimum and the amount of heavy metals in the sediment were mainly associated with stormwater runoff, in combination with municipal and industrial discharge.

Harrop et al. [50] studied heavy metal levels in sediments collected from the near vicinity of roads in a North London Borough. They found the maximum levels of Pb, Cu, Zn and Ni were 56-20,535 μg g^{-1}, 16-43,470 μg g^{-1}, 48-13,740 μg g^{-1} and 49-443 μg g^{-1} respectively. They concluded that the elevated levels were due to the presence of scrap metal yards. They measured Pb in background soil and found the concentration to be 407 μg g^{-1}.

Bubb et al. [51] studied the distribution of heavy metals (Pb and Zn) in the river Yare, Norfolk, UK. They observed sediments within the River Yare contained mean concentrations of 180.5 μmg^{-1} for Pb and 327.6 μmg^{-1} for Zn, with corresponding enrichment ratios of 2-17 for lead and 3-13 for zinc compared with catchment background levels. They pointed out that the major lead contamination source appeared to be the urban runoff which contains coarse particulates (250-1,000 μm). Zinc, by contrast, showed a more widespread distribution. They also found from sediments with a low volatile solids content (<5%), such as in coarser sand and gravel deposits, that organic matter may have effectively controlled the accumulation of lead and zinc, limiting the availability of active binding sites.

Zanders [52] reported heavy metals in road sediment of different particle sizes in New Zealand. Particles collected in a 2-days interval from road side gutters by vacuum suction contained predominantly fine particles (52% <250 μm) and heavy metal contents of Cu, Zn, and Cu were 181-212 mgkg^{-1}, 1,073-2,080 mgkg^{-1} and 251-334 mgkg^{-1} respectively. A high percentage of the heavy metal load was associated with particles smaller than 125-250 μm (64% of Zn, 57% of Cu and 46% of Pb).

Birch and Scollen [53] studied heavy metals in road dust, gully pits and parkland soils in a highly urbanized sub-catchment of Port Jackson, Australia. The mean concentrations of Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn in road dust were 6, 34, 164, 28,000, 284, 27, 487, and 523 μgg^{-1} respectively, for all road types, and concentrations in gully pits were 7, 20, 112, 24,000, 316, 20, 199, and 257 μgg^{-1}, respectively. They found a large proportion of these metals associated with the mobile fine fraction (<62.5 μm). The majority (54-100%) of these elements were extracted with weak leaching agents (EDTA and 1 M hydrochloric acid).

Snowdon and Birch [54] studied the nature and distribution of copper, lead and zinc in soil in a highly urbanized sub-catchment of Port Jackson, Sydney, Australia. They found those elements at

elevated level across the whole catchment with mean total concentrations of Cu, Pb and Zn of 62, 410 and 343 mgkg^{-1} whereas mean enrichment over background levels were 2, 5.5 and 4.6 times respectively.

Yongming et al. [55] studied heavy metals in urban dust in central China. They found Cr at concentration levels in the range of 28-853 mgkg^{-1}; Cu at 20-1,071 mgkg^{-1}; Hg at 0.11-5.21 mgkg^{-1}; Mn at 414-1,318 mgkg^{-1}; Pb at 29-3,060; Sb at 0.63-59.85 mgkg^{-1} and Zn at 80-2,112 mgkg^{-1}. Among them Ag and Hg had commercial and domestic sources; Cr, Cu, Pb, Sb and Zn were mainly from industrial sources, combined with traffic sources; As and Mn were from soil sources, and As were from industrial sources. Another heavy metals investigation in a regional city of Urumqi, China by Wei et al. [56] found the heavy metal concentration range to be much lower compared to central China: Cd 0.11-14.57 mgkg^{-1}; Cr 20.23-54.28 mgkg^{-1}; Cu 33.63-94.54 mgkg^{-1}; Ni 19.40-43.28 mgkg^{-1}; Pb 13.87-53.53 mgkg^{-1}; Mn 535-926 mgkg^{-1}; Be 1.55-2.75 mgkg^{-1}; Co 6.69-10.97 mgkg^{-1}; Zn 69.26-294.47 mgkg^{-1}; and U 1.54-2.13 mgkg^{-1}. Source of Cu, Pb, Cr and Zn were mainly high traffic roads. Ni and Mn were mainly from the industrial areas; Co and U were from the soil material, Be was from the railway activity and Cd was from industrial activities, organic manures and phosphate fertilizer in the park.

2. Heavy Metals in Runoff

Laxen and Harrison [57] reported the lead contribution from highways which come primarily from the combustion of leaded petrol. Lead concentrations in highway runoff were 10^3-10^4 times the background concentrations in surface waters. They reported that smaller lead particles emitted from the exhaust system form a relatively stable aerosol and large part of it escapes from the highway environment. The coarser lead particles on the other hand rapidly deposited to the local ground and were confined to a strip of land roughly 30 m wide to either side of the highway.

Yousef et al. [58] reported the total and dissolved heavy metals in stormwater runoff from highway bridges in Central Florida, USA. The range of heavy metal concentrations (μgL^{-1}) in runoff was as follows: Zn (498 μgL^{-1} total, 336 μgL^{-1} dissolved), Pb (1,558 μgL^{-1} total, 187 μgL^{-1} dissolved), Cr (11 μgL^{-1} total, 2 μgL^{-1} dissolved), Ni (53 μgL^{-1} total, 49 μgL^{-1} dissolved), Cu (52 μgL^{-1} total, 27 μgL^{-1} dissolved), Fe (2,429 μgL^{-1} total, 287 μgL^{-1} dissolved) and Cd (5 μgL^{-1} total, 1 μgL^{-1} dissolved). Hewitt and Rashed [59] studied heavy metals in the runoff waters from a major rural highway in northwest England during a number of storm runoff events. The particulate phase (>0.45 μm) contained more than 90% of the inorganic Pb, approximately 70% of the Cu and approximately 56% of the Cd. The heavy metal concentration profile of the particulate-phase largely followed those of the suspended sediments. Buffleben et al. [60] investigated the heavy metal pollutants associated with the aqueous and SS phases entering Santa Monica Bay from the Ballona Creek catchment, a mixture of residential, commercial, and light industrial land uses. Their study showed that the SS was a major

carrier for the heavy metals containing cadmium (61-89%), chromium (51-93%), copper (40-89%), lead (77-99%), and nickel (61-100%). Arsenic was found primarily in the aqueous phase (60-70%).

Granier et al. [61] observed the following mean heavy metal concentrations in the stormwater runoff in France: 142 µgL^{-1} for Pb, 733 µgL^{-1} Zn and 15 µgL^{-1} for Cu. They considered the observed pollutants concentration as high and described the significance of heavy metal transport in urban runoff.

Barbosa and Jacobsen [62] measured the heavy metals concentration in highway stormwater in Portugal. They found that Cd and Cr were below the detection limit (<1 µgL^{-1}) whereas Cu, Pb and Zn concentrations were 1-54 µgL^{-1}, 1-200 µgL^{-1} and 50-1,460 µgL^{-1} respectively. They observed 61-69% of the total SS, Zn, Cu and Pb load in the first flush (50% of the runoff volume). Gan et al. [63] observed very high concentration of heavy metals in highway runoff in urban and rural area of Guangzhou, China. The event mean concentration (mgL^{-1}) of heavy metals were 0.14 for Cu, 1.76 for Zn, 118.2 for Pb, 1.6 for Cd, 22.6 for Ni and 40.4 for Cr. Their report showed that heavy metal concentrations in urban sites were 6-73% higher than that of rural sites except for pH, TOC and Oxidation Potential (OP). They considered the surrounding land use as the main contributing factor to this difference.

3. Seasonal Variation of Heavy Metals

Westerlund et al. [64] reviewed the seasonal variations of SS and heavy metals in road runoff in Lulea, Sweden. The results showed that the concentrations of SS, lead, copper and cadmium were higher for the snow melt period, compared to the rain (without snow) generated runoff on the catchment, and the highest concentrations were found during the rain-on-snow events. Metals during a snow melt period were more in particulate bound compared to a rain period characterized by a higher percentage of the dissolved fraction. The following table (Table 4) is the total heavy metals during a snow melt period and a rain period.

Similarly, Hallberg et al. [65] discussed winter and summer seasonal variation of ten metals in highway runoff in Stockholm, Sweden and their partition between dissolved and particulate matter (Table 5). They found that the dissolved part of Al, Cd, Co, Cr, Mn and Ni was significantly higher in winter compared to summer. For Fe, however, the dissolved part was lower during winter. They observed no significant difference for Cu, Pb and Zn between the two seasons. They found a good linear relationship between TSS and selected metals (r^2>0.95) during winter runoff events except for Cd. During summer, they also found a good correlation (r^2>0.90) for Al, Cu, Fe, Mn, Ni and Zn.

Westerlund and Viklander [44] reported the particles and associated metals present in road runoff during snowmelt and rainfall periods. Important factors influencing the concentrations and loads were the availability of material, the intensity of the lateral flow, and, additionally, the antecedent dry period. During the snow melt period, particle sizes and TSS were highly correlated with total concentrations of Cd, Cu, Ni, Pb, and Zn. During the rain period, the correlations between total metal concentrations and the different particle sizes were not as significant as during snow melt periods.

4. Summary

- The most investigated heavy metals in literatures were Cu, Zn, Pb, Cd, Cr, V, Ni and Hg.
- The sources of heavy metals in soil, sediment and runoff vary from location to location and are catchment specific.
- The heavy metals concentration in solid and aqueous phases in stormwater runoff vary widely.
- Most of the heavy metals are associated with finer particles.
- Seasonal variations have a strong influence on heavy metals concentration in runoff.
- Partition of heavy metals in dissolved form and particulate bound form depends on various factors such as location, seasons, temperature and pH.

HYDROCARBONS

Several types of hydrocarbons have been found in urban dust and runoff [66,67]. Their origins were found to be both natural as well as anthropogenic although were produced mainly from incom-

Table 4. Total Heavy metals during melt and rain period (West-erlund et al. [64])

	Melt period (µgL^{-1})	Rain period (µgL^{-1})
Cd	0.1-1.9	0.07-0.35
Cu	29-465	14-85
Ni	14-177	1.9-23
Pb	8.5-168	5.1-30
Zn	83-1680	42-358

Table 5. Partition of heavy metals concentration in dissolved and particulate phase in summer and winter (Hallberg et al. [65])

	Dissolved (µgL^{-1})		Particulate (µgL^{-1})	
	Summer	Winter	Summer	Winter
Al	15.57-20.02	31.13-40.25	23129-27957	28705-32420
Cd	0.034-0.048	0.154-0.217	0.85-1.43	0.69-1.03
Co	0.66-1.42	13.53-17.35	37-53	52-70
Cr	2.38-4.34	5.21-9.74	100-120	79-92
Cu	17.41-23.21	14.74-23.06	529-654	211-253
Fe	16.57-57.83	9.76-16.52	47909-57325	45571-51504
Mn	12.01-23.69	117.31-150.46	807-1,071	560-691
Ni	2.74-3.42	4.02-5.06	34-43	30-35
Pb	0.09-0.19	0.090.17	114-142	57-67
Zn	79.4-120	74.0-119	1,531-1,746	1131-1,374

plete combustion of fuel. The hydrocarbons include simple aliphatic hydrocarbons to polycyclic aromatic hydrocarbons (PAHs). The most prevalent hydrocarbons are PAHs. They constitute a large and diverse range of organic molecules. PAHs are synthetic organic compounds that contain two or more fused aromatic rings made up of carbon and hydrogen atoms. They are considered as hazardous pollutants in the group of non-halogenated organic compounds along with benzene, phenols, aldehydes etc. Much of the PAHs are typical of road surface material, fuel combustion products, lubrication system losses, degradation of automobile tires, paint and corrosion products [68]. Besides this, airborne particles, and stormwater pollution sourced from soil erosion, industry and vegetation may be transported by wind and rainfall from other sites and deposited in highway catchments [30,69,70]. Sharma et al. [71] reported that in Sault Ste. Marie, Ontario vehicles and coke ovens are the major contributors to PAH in street sediments. Lim et al. [72] reported that the major contribution of PAHs was traffic sources (59-71%). According to Benner et al. [73], nearly 35% of the total PAHs input was from road related activities. These PAHs have low degradability and possible carcinogenicity or mutagenicity. Consequently, the USEPA has identified 16 species of PAHs as priority pollutants in the Clean Water Act [74]. Fig, 1 shows the 16 USEPA PAHs.

Latimer et al. [75] studied sources of petroleum hydrocarbons in urban runoff with different landuse type. Their result indicated that in all landuse types, hydrocarbons were primarily from used crankcase oil, with a small amount of fuel oil detected at industrial sites. They further reported that only a small amount came from the natural source surveyed, and the majority of this oil probably would have come from: (1) oil drops within the driving lanes on the road surface or deposits in parking areas and/or (2) direct dumping of waste crankcase oil into storm drains.

1. PAHs in Soil, Road Dust and SS from Impervious Surfaces

Munch [76] studied PAHs along with heavy metals in forest soils next to urban roads at the town of Dortmund, Germany. The traffic volume along road was about 3200 vehicles per day since 1961. They found that the PAHs concentration in the soil along the road edge was much higher than the forest soil by a factor of upto 100 with the highest concentration of pyrene at 58.2 ppm. They observed an exponential decrease of PAHs with distance from the road.

Pathirana et al. [77] studied the distribution of PAHs in an urban roadway system in Australia. They found PAHs widely distributed in leaf litter exhibiting the highest concentration of 1,254 ngg^{-1} total wet weight which was several fold higher than the soil (471 ngg^{-1}). They found that PAH content in the soil, leaf litter and vegetation declined exponentially with distance from the roadway, soil depth and vegetation height respectively.

Weiss et al. [78] investigated PAH and many other contaminants in soil in an industrialised urban area of Linz, Austria. They analysed 18 PAHs in soil and found the sum of PAHs in the range of 0.28-79.0 mgkg^{-1}. The pattern of distribution of PAHs around the site investigated showed a high degree of similarity irrespective of location and soil pollution. Walker et al. [79] discussed the potential contribution of urban runoff to surface sediment of the Pasaic River, NJ USA. Among PAHs, phenanthrene and fluoranthene had the highest concentration level. Based upon the particle sizes of the materials, the authors hypothesized that the two sources of PAHs in urban runoff were from particles of asphalt abrasion (in larger particles) and a fossil fuel source. They expected annual PAHs load was around 681 kgyr^{-1} in Narragansett Bay, USA.

An interesting relationship between PAHs and particle sizes were
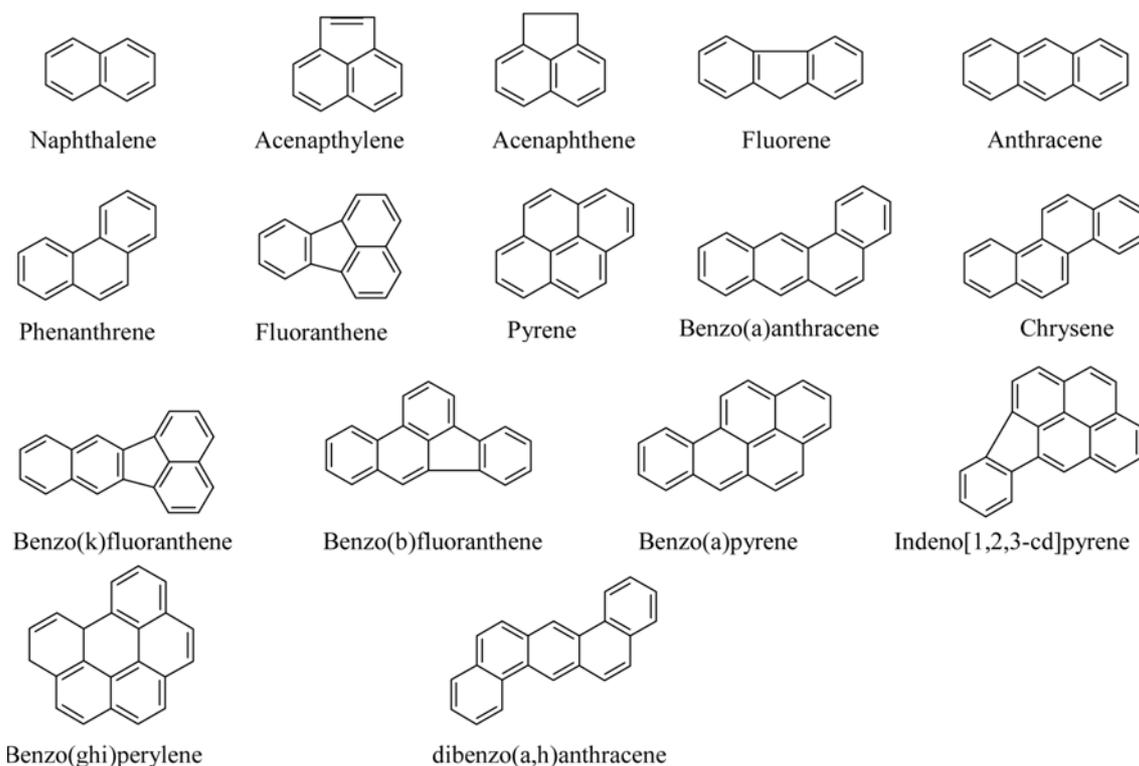
Fig. 1. 16-USEPA PAHs.

observed by Krein and Schorer [80] in a study of road runoff and its contribution to river sediment in Hunsruck Mountains Germany. Their study of PAHs in different particle sizes showed a relationship between molecular size and grain size. In street runoff, they found small 3-ring molecules enriched the fine sand fraction. They also found distinct bimodal distribution for 4- and 5-ring PAHs. Aryal et al. [81] discussed the particle bound PAHs (12 species) in highway runoff in Winterthur Switzerland. They observed that the particle bound PAHs concentration range was from 1.3-13.1 μgL^{-1} in the first 3 mm of runoff. In a particle size study, Aryal et al. [82] observed the weighted average PAH content in each event ranged from 17 to 62 μgg^{-1} in TSS, 23 to 54 μgg^{-1} in the fine fraction and 16 to 84 μgg^{-1} in the coarse fraction in Winterthur, Switzerland. They also calculated the PAHs load ranged from 0.06 to 0.22 gha^{-1} for a total of 12 PAH species.

Nagbe et al. [67] discussed the PAHs in stormwater runoff from urban and coastal areas of South Carolina. They observed the concentration of ΣPAH (for 14 compounds) ranged from 282-5,590 ngL^{-1}. The PAHs profiles they observed in runoff were more similar to those of atmospheric particulate PAHs and unlike those in used crankcase oil. In Louisiana, USA, Mielke et al. [83] found a total PAHs content of 2,927 ngg^{-1} (median) in the inner-city and 731 ngg^{-1} (median) in a suburban area. They observed a strong correlation between PAHs and heavy metals. In terms of specific site accumulation, the PAHs and metals concentration in dust showed similar patterns in dust i.e. the concentration of dust was highest in a busy street, followed by foundations, then residential streets and then open areas.

Murakami et al. [84] discussed the runoff behaviour of particle-bound PAHs from road and roofs in particles of size more than and less than 45 μm particles. They found that the PAHs found in the runoff were similar to road dust than the roof dust. The total PAHs content (12 PAHs) in particles smaller than 45 μm was 3.3-15 μgg^{-1} compared to the content in soil of 0.84-0.85 μgg^{-1}. PAHs in particles larger than 106 im were as low as 0.14-0.37 μgg^{-1}. Murakami et al. [85] discussed the effect of size and density of particles on PAHs distribution in road dust in Tokyo, Japan. Their road dust fractionation of light density particles (<1.7 gcm^{-3}) and heavy density particles (>1.7 gcm^{-3}) showed that the lighter particles contained 28-44% of PAHs despite having a contribution of less than 4%. PAHs contribution in lighter fraction was 1-2 orders of magnitude higher than those in the heavy fraction.

Yang et al. [86] also observed that the finest fraction (<63 μm) contained almost half of the total PAHs mass in soil. The PAHs distribution in each fraction showed a similar pattern indicating a common PAH source. The PAH concentrations varied from 545.5 to 1,039.3 $mgkg^{-1}$. The coarse soil (>500 μm) contributed only 5.1% of the total PAHs mass although concentration was very high in this fraction. They observed a good correlation between PAHs and organic carbon.

Liu et al. [87] studied concentrations, spatial distribution and sources of 17 polycyclic aromatic hydrocarbons (PAHs) and methylnaphthalene in surface sediments of rivers and an estuary in Shanghai, China. The total PAH concentrations, excluding perylene, ranged from 107 to 1,707 ngg^{-1} (dry weight) with an average of 823 ngg^{-1}. They observed good positive correlation between total organic carbon and PAHs (r^2=0.81). They found that the sources of PAHs at

locations far away from cities were mainly from petrogenic (where the ratio of low molecular weight PAHs/high molecular weight PAHs >1). While in industrial and residential areas, both petrogenic and pyrogenic (low molecular weight PAHs/high molecular weight PAHs <1) inputs were significant.

Agrawal et al. [88] studied surface soil (0-5 cm) from various agricultural sites in Delhi for Σ_{16}PAHs. They reported Σ_{16}PAHs ranged from 830 to 3,880 μgkg^{-1} (dry wt.) with an arithmetic mean of 1,910 ±1,020 μgkg^{-1}. They observed 2-5 times higher Σ_{16}PAHs values at urban sites compared to rural sites. In general, low molecular weight PAHs was predominant which was correlated to petrogenic sources and/or to biomass burning in rural areas which is a low temperature combustion process and/or petrogenic sources. They observed good positive correlation between organic carbon and low molecular weight PAHs (2, 3 and 4-ring PAHs).

Kafi et al. [89] discussed the spatial variability of the characteristics of combined wet weather pollutant loads in Paris. Their study in wet weather flow found PAHs concentration varied from catchment to catchment and in the range of 1.0-5.2 μgL^{-1}. They found higher PAHs concentration during wet weather flow compared to dry weather flow. The pollutant load per unit of active surface area had 36-523 $mgha^{-1}$. Sun et al. [90] discussed the distribution and source of 16 PAHs in the Henan Reach of the Yellow River, Middle China. ΣPAHs levels ranged from 144.3 to 2,361 ngL^{-1} in water, from 506.6 to 10,510 ngg^{-1} in suspended particulate matters (SPMs) and from 16.4 to 1,358 ngg^{-1} in sediment, respectively. A high concentration of PAHs in water indicated that water in the area was more heavily contaminated by PAHs, which was possibly due to industrial wastewater, urban runoff and emission from atmospheric particles at these sites. They observed a domination of 2-3-ring PAHs in water and 2-4-ring PAHs in suspended particulate matters. PAHs in sediment comprised mainly of 3-5-ring. The observed ratios of phenanthrene/anthracene (<10) and fluoranthene/pyrene (>1) reflected a pattern of pyrogenic input.

2. Sources of PAHs

Numerous investigations have been carried out to understand the sources of PAHs in urban areas. The source characterization have relied on PAHs homologs. The first attempt to quantify the proportion of pyrogenic to petrogenic PAHs was by Boehm and Farrington [91]. Colombo et al. [92] used n-alkanes ratio to identify pyrogenic PAHs sources and n-C16 ratio (sum of all alkanes/n-C16) to identify petrogenic PAHs sources. This ratio is usually large (i.e. 50) in biogenic samples and small (i.e. 15) in oily samples. Gogou et. al. [93] studied the aliphatic and aromatic hydrocarbons in the Eastern Mediterranean. The total concentration of aliphetic and PAH were 562-5697 and 14.6-158.5 ngg^{-1} respectively. They confirmed the petroleum hydrocarbons input by studying the specific α- and β-hopanes. Yin et al. [94] investigated soil PAHs in the vicinity of Nanjing, China. They observed the ratio of fluoranthene to the sum of fluoranthene and pyrene concentrations (Flt/(Flt+Pyr)) was more than 0.5 in 99% of vegetable soil samples. They concluded that the PAHs in soils were generally derived from straw and coal combustion sources.

Stout et al. [95] studied the PAHs in urban sediment by conventional hydrocarbon finger-printing and isotopic ^{210}Pb, ^{137}Cs and principle component analysis. They found that urban runoff in the post-industrial (after 1930) sediment was dominated by low but consis-

tent concentration (10-20 mgkg^{-1} dry) of pyrogenic PAHs derived primarily from combustion of fossil fuel(s).

Bixian et al. [96] used lead isotope (^{210}Pb) to understand prolonged deposition of PAHs in sediments from the Pearl River and Estuary, China. A different parent compound distribution at the lower reach compared to the upper reach indicated another possible input of PAHs in the lower reach. Total PAH values ranged from 4,837 to 7,302 ngg^{-1} at upper reach region, but decreased to 1,460-2,789 ngg^{-1} at lower reach. The decrease of PAH contents in sediments from stations in the lower reach was believed to be due to biodegradation and desorption of low molecular weight PAHs (especially naphathalene, acenaphthylene and acenaphthene) during transport from the upper reach and deposition in the lower reach. They found a predominance of parent PAHs over alkylated PAHs in sediment at one sampling site and alkyl PAHs at the other two sites in upper reach that reflected the presence of a combustion source and significant petrogenic sources respectively.

Pengchai et al. [68] applied the principle component analysis to identify the sources of PAHs. They found that light duty vehicles produced higher molecular weight PAHs whereas heavy duty vehicles (diesel) produce low molecular weight PAHs.

Kim et al. [97] used carbon isotope (^{13}C) to characterise sources of PAHs in sediment from lakes, harbour, and shipping waterway. The sediment from shipping waterways showed pyrogenic origins for PAHs. Kim et al. [97] found that increasing environmental exposure systematically changed the molecular ratio of isomeric PAHs in favour of thermodynamically stable isomers. Chandru et al. [98] characterised alkanes, hopanes and PAHs in tar-balls collected from the East Coast of Peninsular Malaysia. They found Σalkane, Σhopanes and ΣPAHs 4.1-26.9 mgg^{-1}, 24-32 μgg^{-1} and 6-117 μgg^{-1} respectively. They concluded that the primary source of tar-balls was from the South East Asia Crude Oil, most likely from oil platforms, ballast water and tanker washing. Kose et al. [99] carried out source analysis of PAHs in road dust and urban runoff using the marker compound tri-terpene. They found that tyre was the major source of PAHs in road dust followed by pavement asphalt. Contribution of automobile exhaust gases to the road dust was not large. In runoff, road dust was the major source of PAHs. Pies et al. [100] ccharacterized and identified sources of PAHs in river bank soils in Mosel and Saar Rivers in Germany by considering the distribution patterns of 45 PAHs (including 16 EPA PAHs and some alkyl PAHs), specific PAH ratios, distribution patterns of n-alkanes and principal component analysis (PCA). Their result showed that pyrogenic PAHs dominated soil samples collected upstream of the confluence of the Mosel and Saar Rivers, and petrogenic and pyrogenic PAHs dominated samples downstream of the confluence. Based on an analysis of reference materials and organic petrography, Pies et al. [100] concluded that coal particles dominated the petrogenic input. The study concluded that to accurately determine the origin of PAHs, several identification methods must be applied.

Zhang et al. [101] applied low molecular weight/high molecular weight ratio of PAH to identify the sources in a one year monitoring of stormwater runoff, road dust, rain and canopy through-fall sampling in Beijing, China to identify the source of PAHs in urban environments. Their PAHs composition result indicated a dominance of pyrogenic sources for PAHs in runoff, rain and canopy through-fall. Road dust had both petrogenic and pyrogenic sources. The main

PAHs sources in rain were coal/oil combustion (54%), vehicular emission (34%) and coking (12%). Peng et al. [102] studied the temporal trends of hydrocarbons in sediment cores from the PRE and the northern SCS. Petrogenic PAHs were confined to the PRE, and PAHs in sediments in the northern SCS originated predominantly from combustion of coal and biomass. They concluded from the stable isotopic fingerprinting technique that the petrogenic hydrocarbons in the PRE resulted mainly from local accidental spills/leakages of lube oil and crude oils. They found that accumulation rates of PAHs in the northern SCS have significantly increased after 1978 due to the implemention of policy reform in China and the country's subsequent growth.

Mostafa et al. [103] carried out source characterization and the environmental impact of urban street dusts in different cities in Egypt based on hydrocarbon distributions based on n-alkane profile differences. The mean total PAH concentration of the dust in selected streets ranged between 27 and 379 ngg^{-1} with a geometric mean of 130.5 ngg^{-1}. They found a domination of 3- and 4-ring PAHs in street dust (68-98%). They observed a predominant amount of fluoranthene, pyrene, chrysene and benzo[ghi]perylene in heavily traffic street. The presence of the fossil markers (hopanes and steranes) was strong evidence for a petrogenic input, probably from unburned lubricating oil. They also found diesel exhaust as a significant contributor to street dust from the heavily trafficked streets

3. Summary

- A wide range of PAH in sediment, soil, road dust and urban runoff has been studied and observed.
- Seasonal and climatic factors may change the availability of PAHs.
- Sources of PAHs can be predicted using different techniques such as low molecular weight to higher molecular weight ratio, isotopes, fossil markers, etc.

NUTRIENTS

Nutrients (nitrogen and phosphorus) are essential elements in all aquatic ecosystems. However, when these nutrients are found at excessive levels, they can have negative impact on aquatic system by promoting the growth of plants and algae. Such nutrients contribute to the eutrophication of water bodies. Common sources of these nutrients are chemical fertilizers that applied to agriculture land, lawns, gardens, golf courses, landscape areas, and garden. Residential areas and turf areas are major contributors of the nutrients in urban runoff [104-107]. Nutrients are found in various forms. Phosphorus can be found in inorganic phosphate (phosphate, orthophosphates, polyphosphates) or organic bound phosphate. Specific measurements are done in the form of total phosphorus, soluble reactive phosphorus, and biologically available phosphorus. Similarly nitrogen is also found in inorganic and organic forms. It is measured as total nitrogen, Kjeldal nitrogen, nitrate and nitrite nitrogen, and ammonia nitrogen.

Among the phosphorus compounds, phosphate is a key form in stormwater runoff. Phosphate in runoff exists in soluble reactive phosphorus or orthophosphates, polyphosphate and organic bound phosphates. Among them orthophosphates are found in sewage and natural water sources whereas polyphosphates are found in synthetic substances such as detergents. Organic phosphates are found

in aminoacids, peptides and proteins.

Among the nitrogen compounds, nitrate is of most concern. Nitrates are easily adsorbed onto soil due to its anionic nature. Accumulation of nitrate in soil helps in its leaching to groundwater. Nitrates sources are fertilizers, human wastewater, and animal wastewater. Many studies have reported that urban stormwater is one of the major contributors of nutrient contamination of groundwater systems [108,109].

1. Nitrogen and Phosphorus in Urban Runoff and Sediment

Lee [110] pointed out the role of phosphorus in eutrophication in many lakes and reservoirs. He also pointed out the importance in the development of mathematical models to obtain relationship between algal growth and nutrients. Colston [111] considered urban land runoff as a significant source of pollution when he found the yield of COD, BOD was equal to 91% and 67% of the raw sewage respectively in their study at Durham, North Carolina.

Cherkauer [112] studied the impact of urbanization on water quality during a flood in small catchments in the urban areas of Milwaukee, USA. The paper reported the significant response of pollutants to the rain in urban basins compared to rural catchments. Boyd [113] analysed centrifuged particulates from the Don River, Canada and confirmed that a significant proportion of the phosphorus and metals delivered to the harbour were associated with suspended sediment. He estimated the phosphorus partitioning ranged from about 20% to 90% particulate, with a median at approximately 40%. Juracek [114] studied lake-bottom sediment to estimate historical non-point-source phosphorus loads. His finding showed that the contribution to the total mean annual phosphorus load in Hillsdale Lake is about 7% from point sources and about 93% from non-point sources. Meals and Budd [115] showed that Lake Champlain, NY, USA received 18% of the phosphorus load from urban sources. Basnyat et al. [116] found urban areas as a major contributing source for nitrate followed by active agriculture. Okubo et al. [117] found that adsorption and desorption equilibrium of nutrients between suspended solids and ambient water was important for estimating the amount of nitrogen and phosphorus released from suspended solids. They observed the release of nitrogen from 5-30% and 0-40% in 30 days.

Waschbusch et al. [118] investigated sources of phosphorus in stormwater and street dirt from two urban residential catchments in Madison, Wisconsin. They collected numerous runoff samples from the test catchments and used an urban stormwater quality model (SLAMM l) to quantify the significance of the different phosphorus sources. Lawns and streets were found to be the most significant sources of phosphorus in the test basins, contributing about 80% of the total annual loading.

The ecological effect of urban stormwater to benthic community was reported by Pratt et al. [119]. They correlated a high degree of correspondence between the known sources of urban runoff and concluded disturbance caused in the benthic community was due to urban runoff.

Zanoni [120] reported the characteristics of nutrients in urban runoff. His report indicated most of the nutrients in urban runoff are particulate bound and different from municipal sludge. Jacobsen et al. [121] investigated road runoff pollutants characteristics and found 60-80% of phosphorus and 50-60% of nitrogen in road runoff to be associated with particles. Vaze and Chiew [122] reported

more than 90% of phosphorus and nitrogen associated with particulate form in urban stormwater runoff. Their results showed that most of the TN and TP were attached to the particles between 53 and 300 μm. Ball and Abustan [123] report that up to 85% of phosphorous and 70-80% of the nitrogen was associated with particulate matter. They also showed that on average about 80% of phosphorous was in particulate form during the peak of the storm events that were monitored.

Prasad et al. [124] analysed autumn leaf litter from five tree species in Toronto, Canada and reported the total phosphorus (TP) and total nitrogen (TN) composite range was from 0.07-0.26% and 0.7-1.2% respectively (dry weight) and of which 0.006-0.007% and 0.05-0.24% could leach into water. Dorney [125] reported TP composition of leaf litter ranged from 0.06-0.44% for residential street trees in Milwaukee and Shorewood, USA. Riley and Abood [126] studied the nutrients load from rubbish sample from a shopping centre in Sydney Australia and treated it with aerated and non-aerated stormwater before recording the leaching potential of TP and TN over the subsequent period of days. They found that 0.03 and 0.05% of dry leaf weight of TP and TN leached into deionised water respectively. Alison et al. [127] also did similar type of test for nutrient contribution of leaf litter in urban stormwater. He reported that about 80% of stormwater gross pollutants were organic and pointed out that 5-20% of TN and TP could leach into stormwater.

Railey and Banks [128] found some role of phosphorus and heavy metals in the spread of weed in urban bushland in NSW, Australia. Natural and anthropogenic sources contribute pollutants to road runoff at various points when passing through the stormwater system.

Ball and Abustan [129] investigated the phosphorus export from an urban catchment in Sydney, Australia. They derived a relationship between inorganic suspended solids and particulate phosphorus for this catchment. The derived relationship was applied to the evaluation of the performance of detention pond and/or wetlands for treating stormwater runoff.

Seitzinger et al. [130] studied the bioavailability of dissolved organic nitrogen (DON) from natural and anthropogenic sources to estuarine plankton. Overall, urban and suburban stormwater runoff had a higher proportion of bioavailable dissolved organic nitrogen (59%±11) compared to agricultural pastures (30%±14) and forests (23%±19). DON bioavailability varied seasonally although the seasonal pattern differed for the three sources. Seitzinger et al. [130] further suggested that seasonal budgets of bioavailable nitrogen (dissolved inorganic nitrogen plus bioavailable dissolved organic nitrogen) as a function of land use and concluded that 80% of the total dissolved nitrogen (TDN) from urban and suburban runoff is bioavailable which was relatively higher than forests and pastures (20-60%).

Lewis and Grimm [131] obtained a positive correlation between rainfall intensity and the total load of TN and TP. They found no difference between the type of climate and nutrients load. They further pointed out the relationship between urban design and material export from catchments to water bodies.

Strynchuk et al. [132] studied the decomposition of grass and plant leaves and their contribution in nutrient load to receiving water bodies in Brevard County, FL USA. Their study showed how the release rate of nutrients could be calculated which was useful to determine the maintenance frequencies of stormwater treatment prac-

tice that treat nutrient.

Taylor et al. [133] found 80% of nitrogen in dissolved form in urban stormwater when analyzing runoff in Melbourne, Australia. Out of the 80%, 11% was in the form of ammonia nitrogen. They did not find variations in concentration of nitrogen species during the baseflow period and the storm periods but noticed a higher concentration in particulate form during storm events.

2. Summary

• Urban runoff contributes both nitrogen and phosphorus in particulate and soluble form.

• Suspended solids and ambient water play a crucial role in sorption and desorption of nitrogen and phosphorus in sediment.

• Urban and suburban stormwater runoff contained a higher proportion of bioavailable dissolved organic nitrogen compared to agricultural catchments.

TREATMENT MEASURES FOR STORMWATER RUNOFF

Treatment measures to reduce pollution in stormwater runoff can be classified into three subsets based on the type of pollutants which are removed or reduced. Primary treatment removes gross pollutants and coarse sediments and any nutrients, or hydrocarbons attached on particulates larger than 1.25 mm. The removal method is through entrapment of litter/debris and sedimentation of coarser particles. Typical devices include, *litter racks, sediment traps, gross pollutant trap (GPT), litter booms, and oil/grit separators*. Another form of primary treatment system is *permeable pavements* which are commonly used as a source control measure in which the runoff infiltrates into the ground rather than increasing stormwater runoff. In this manner the pollution loads transported downstream are reduced. Source control of stormwater is generally regarded as the most effective strategy for achieving the long-term goals of storm-water management.

Secondary treatment measures attempt to remove finer particles and attached pollutants by using filtration techniques and/or by inducing sedimentation. Secondary treatment consists of sedimentation of finer particles or filtration techniques to remove fine particles and attached pollutants. This can be achieved with pre-entrance treatments which uses filtration techniques to separate out entrained sediment prior to its entry into the drainage network. Some common examples of pre-entrance treatments are filter or *buffer strips and grass swales*. Infiltration is also promoted in one of two ways; either by stormwater flowing over vegetated land enhancing infiltration, or through the use of purpose built structures such as porous pavements and infiltration trenches. In-transit secondary treatment systems entrain pollutants flowing through the stormwater system. A typical example is a *bio-retention system*. These systems remove particulate and soluble contaminants by passing stormwater through vegetation and a filter medium. The vegetation can consume (biological uptake) nutrients detained in the filter.

Tertiary treatment includes a range of physico-chemical measures such as enhanced sedimentation, filtration and adsorption, and biological uptake. An example is *Constructed wetland* systems which are shallow, extensively vegetated water bodies that use processes of extended detention, fine filtration and biological pollutant uptake to remove pollutants from stormwater.

Table 6 list a range of stormwater treatment measures and summaries the target pollutant for each device and its applicable catchment areas. The treatment measure target different types of pollutants and is designed or is more suited to treat runoff from catchments of sizes. Any one treatment measure will not be able to treat a suite of pollutants. A vegetated swale, for example, may be able to remove coarse sediment, but it will not be effective for removing nutrients and dissolved material. By contrast the later are more effectively removed by a wetland or biofilteration. The swale may then become

Table 6. Summary of target treatment and catchment areas for a range of stormwater treatment measures and actual removal rates and indicative levels of pollutants in the outflow [134,136]

Stormwater treatment measure	Target pollutant	Target catchment area (ha)	Measured Removal Rates and Levels of Pollutants				
			Suspended solids[a]	Total phosphorus[a]	Total nitrogen[a]	Turbidity[a]	E. coli[a]
Permeable pavement	Reduced runoff[b], suspended solids and attached pollutants	<2	NA	NA	NA	NA	NA
GPT	Coarse Sediment, litter	8-15	0-70% 42-140	0-30% 0.18-0.25	0-15% 1.7-2.0	0-70% 18-60	Negligible
Swale	Coarse sediment, Suspended solids	<2	55-75% 35-63	25-35% 0.16-0.18	5-10% 1.8-1.9	44-77% 14-34	Negligible
Sand filter	Suspended solids and attached pollutants	1-6	60-90% 14-56	40-70% 0.08-0.15	30-50% 1.0-1.4	55-90% 6-93	-25-90% 500-11000
Bio-retention	Suspended solids and attached pollutants, some dissolved organics	<2	70-90% 14-42	50-80% 0.05-0.13	30-50% 1.0-1.4	55-90% 6-93	-58-90% 900-15000
Wetland	Turbidity, dissolved pollutants, inorganic and organic matter	>6	50-90% 11-67	35-65% 0.09-0.16	15-30% 1.4-1.7	10-70% 19-53	-5-99% 100-9000

Values in % are removal rates while the others are indicative levels of pollutants in the outflow for a range of stormwater treatment measures
[a]Concentrations in mgL^{-1} except for turbidity (NTU) and *E. coli* (cfu/100 mL)
[b]By reducing runoff porous pavement reduce the pollutants transported downstream

Table 7. Pollutant Removal Efficiency of different combination (50 : 50) of media types used in filtration [135]

Parameter	Carbon-sand	Peat-sand	Zeolite-sand	Sand
Colour	26			
Nitrate	97	100		
Sulfate		5		
Potassium	15		39	
Dissolved solids		45		
COD	85-96			
Hydrocarbons	43-90	36-68	82-79	94-100
Zn	48	58	62	88

the pre-treatment measure for the wetland, creating a treatment train. Stormwater pollution is effectively managed through a treatment train which is a sequence of treatment measures formulated to remove a range of pollutants at appropriate timescales, based on the available foot print areas to site treatment measures.

There are a number of types of media that are recognised as being effective in removing pollutants in secondary and tertiary treatment systems such as bioretention systems, filtration and infiltration systems. Filtration media can be used to biochemically confine a stormwater treatment system using a soil ameliorative to enhance stormwater treatment. Table 7 provides a summary of the average pollutant removal efficiencies associated with different media types and combinations investigated in a laboratory scale filter [135]. The combinations were investigated using a 50:50 ratio of the two materials involved.

The performance of the range of stormwater treatment measures are summarized in Table 6, [134]. It gives the indicative pollution removed by the treatment system and levels of pollutants in the effluents from these treatment systems. In general these systems are not effective in the removal of dissolved nutrients (mainly nitrogen) and pathogens. The turbidity is also not completely removed. Further, although not shown in Table 6, these systems are not suitable for removing heavy metals, and organic matter as they may be found in dissolved form.

The key drawback of the current range of stormwater treatment measures can be listed as follows:

• Treatment systems (as listed in Table 6) are less reliable, as indicated by the wide band in treatment rates, and requires ongoing

maintenance to ensure the performance rates indicated in Table 6.

• They do not have high rate treatment systems and requires a large storage component, eg wetland and bioretention. They require a large land footprint required for adequate levels of storage and treatment which is often not available in inner-city urban areas.

• They are not effective for removal of heavy metals, turbidity, dissolved nutrients (mainly nitrogen) and pathogens.

TREATMENT FOR STORMWATER HARVESTING AND RECYCLING

Stormwater harvesting and reuse offers a potential alternative water supply for at least non-potable uses. It complements other approaches to sustainable urban water management such as rainwater tanks, the reuse of wastewater and greywater and demand management. Collectively these areas form the basis of developing sustainable water technologies.

Urban stormwater is perceived by the community to be of better quality than grey water and wastewater and its reuse has a better public acceptance. The benefits of a successful stormwater harvesting scheme are reductions in (i) demand for town (potable) water (ii) stormwater pollution loads to downstream waterways and estuaries (iii) stormwater volumes and discharges. Stormwater pollution is a major source of pollution in Sydney Harbour and Melbourne's Port Phillips Bay. Stormwater contributed 94% of sediments and 50-60% of nutrients to Sydney Harbour [137].

1. Stormwater Treatment Targets and Objectives

The three aspects of stormwater quality of particular relevance to stormwater harvesting and reuse schemes are [134]:

• Pathogens, including faecal coliforms and *E. coli* - for public health implications.

• Chemical constituents - for public health and environmental considerations, and some end-use requirements (e.g. irrigation).

• Suspended solids and turbidity - for their potential impact on both the effectiveness of disinfection and the function of irrigation schemes.

To date the analysis and evaluation of stormwater quality has been on the basis of surrogate components. Further the assessments have been predominantly targeted for the impacts on receiving water and not for reuse purposes. Table 8 provides indicative values for a range of non-potable uses including residential non-potable uses such as toilet flushing, irrigation, construction applications, and fire fighting.

Table 8. Stormwater indicative targets criteria for reuse application [134]

Application	Typical Indicative Targets
Level 1: Reticulated non-potable residential uses (e.g. garden watering, toilet flushing, car washing)	*E. coli* <1 cfu/100 mL, Turbidity≤2 NTU, pH 6.5-8.5, pathogen reduction[b]
Level 2 (with human exposure). Spray or drip irrigation of open spaces, parks and sportsgrounds, dust suppression, construction site, ornamental water-bodies, fire-fighting.	*E. coli* <10 cfu/100 mL, Turbidity≤2 NTU, pH 6.5-8.5, pathogen reduction[b], TP<0.05 mg/L[a], TN<5 mg/L[a]
Level 3 (no human exposure) Spray or drip irrigation or subsurface irrigation of open spaces, parks and sportsgrounds, Industrial uses-dust suppression, construction site.	*E. coli* <1,000 cfu/100 mL, pH 6.5-8.5, TP<0.05 mg/L[a], TN<5 mg/L[a]

[a]Indicative values for long term irrigation

[b]1 mgL^{-1} Cl$_2$ residual after 30 minutes or equivalent

Additional criteria may be applicable for more specific application eg industrial reuse purposes.

Stormwater for harvesting and reuse purposes should be assessed primarily for nutrients, physical properties (suspended solids, turbidity), bacteriological properties (total and faecal coliform), heavy metals (such as iron, manganese and lead), organic matter since it is more unlikely to meet these water parameters. Traditional stormwater management measures were reviewed in section 6. The key drawback of the current range of stormwater management treatment systems if used for stormwater harvesting and reuse are that they are less reliable and requires ongoing maintenance to ensure the performance rates indicated in Table 6. Further They are not effective for removal of heavy metals, turbidity, dissolved nutrients (mainly nitrogen) and pathogens. Coincidentally, it is primarily these parameters that make stormwater unsuitable for harvesting and reuse. They also require a large land footprint required for adequate levels of storage and treatment which is often not available in inner urban areas.

Stormwater discharge is relatively high and therefore needs to be treated at a high rate. The alternative is to store the stormwater before treatment in a manner similar to current stormwater management treatment systems. Raw stormwater in storage has low value and will degrade under anaerobic and anoxic condition while pretreatment of stormwater adds value to the stored water which can be beneficially reused.

High rate treatment systems can be used to create a sustainable urban development with a low demand on town water, low stormwater pollution export and reduced stormwater discharges. These treatment systems have been used successfully in water and wastewater treatment and include fibre filters, deep bed filters and biofilters. By products of the treatment process (concentrated pollutants and sludge) can be discharged to the sewer alleviating sludge disposal problems and is attractive in creating a low maintenance system. The use of these treatment systems for water reuse can significantly reduce the stormwater pollution export from a site that is transported downstream into the receiving water.

2. High Rate Treatment Technology

Physio-chemical treatment systems can achieve a relatively high pollutant removal at a high rate. These high rate systems are targeted to enhance water quality to at least level 2 (Table 8).

2-1. Fibre Filter

High rate fibre filters were developed by Lee et al. [138] and its high efficacy for the tertiary treatment of wastewater was proved in terms of high filtration velocity and good removal of particulate matter. In place of the sand, fibre media consisting of bundles of U-shaped fine polyamide fibers with a packing density of 105-125 kg/m^3 are used. Compared with the conventional rapid sand filter, the filtration velocity of a fibre filter is more than 5-20 times (at 20-60 m/h) and the specific surface is more than twice Lee et al. [139]. The fibre packing combines the two advantages of a large specific surface area and very large porosity (more than 90%) which results in high removal efficiency and low pressure drop despite the high filtration velocity, [138]. In-line additions of flocculants enhance the pollutant removal capacity for both dissolved organics and trace metals.

The performance of a high rate fibre filter in treating stormwater in conjunction with in-line flocculation ferric chloride ($FeCl_3 \cdot 6H_2O$)

was assessed, [140]. The performance of the fibre filter was good as measured by turbidity removal (95%), total suspended solids reduction (98%), colour removal efficiency (99%), TOC removal (reduced by 30-40%) and total coliform removal (93%). Similar results were obtained for nutrients such as nitrogen and phosphorous where the removal efficiency was significant. The removal efficiency of heavy metal was variable as the concentration in the raw stormwater was small. Eventhough the concentration of some of these metals such as iron, aluminium, copper, and zinc were significantly reduced, others like nickel, chromium and cadmium showed lower removal rates.

2-2. Deep Bed Filter

Deep bed filtration is often referred to as media filtration or rapid filtration and has been widely used for water treatment as a final clarification unit to remove particles. Deep bed filtration is an effective process in removing particles of various nature and sizes that are present in water and wastewater. Deep bed filtration finds its greatest application in the clarification of dilute suspensions (less than 500 mg/l) of particles ranging in size from about 0.1 to about 50 µm, [141]. Deep bed filter is usually single medium, dual media or mixed media operating under pressure or gravity, [140]. The filtration velocity of deep bed filter is usually 5-20 m/h. The other characteristics are summarised in Table 9. The use of in-line flocculation in conjunction with deep bed filtration has some advantages. This process produces smaller flocs that can be intercepted by the filter media. Further, it is simple, easy to operate and cheap.

The performance of dual media and single media deep bed filters in treating stormwater in conjunction with in-line flocculation of ferric chloride ($FeCl_3 \cdot 6H_2O$) was assessed, [143]. The single filter media (80 cm) consisted of either anthracite or sand, and the dual media filter consisted of sand (40 cm at the bottom) and anthracite (40 cm on top). Filtration velocities of 5 m/h, 10 m/h and 15 m/h were examined. The removal efficiency for turbidity, suspended solids and TOC was found to be 95%, 99% and 30-45% respectively at a flocculant dose of $FeCl_3$ of 15 mg/L. The removal efficiency for nitrogen was lower than phosphorus which was relatively good (up to 50%). After filtration the amount of total coliform reduced significantly (up to 80%). The removal efficiency for heavy metals such as Cd, Pb, Cr and Ni was found to be already low for all filter systems partly because the concentrations of these metals in the influent were low. Molecular weight distribution (MWD) analysis was used to assess the removal of organic matter present in storm water. The molecular weight of organic matter present in raw stormwater was in the range of 27,500 to 700 dalton (Da), with highest fraction of 1,500 to 700 Da. It was found that deep bed filters was effective in removing organic matter (especially larger molecular

Table 9. Characteristics of rapid filters

Characteristics	Sand filter	Anthracite filter
Filtration rate, m/h	10-20	10-20
Depth of bed, cm	80	80
Particle size, mm	0.35-0.5	0.7-0.8
Max headloss (gravity filter), m	5	5
Max headloss (pressure filter), kPa	200-400	200-400
Backwash rate, m/h	40-50	40-50

Table 10. Comparison of MF and UF membrane processes, adapted from [146]

	MF	UF
Membrane	Symmetrical, Asymmetrical	Asymmetrical
Thickness (μm) Thin film (μm)	10-150	150-250 1
Pore size (μm)	4-0.2	0.2-0.02
Rejection of	Particles, clay, bacteria	Macro molecules, proteins, polysaccharides, virus
Membrane materials	Polysulfone (PSO), Polyvinylidenedifluoride (PVDF), Polypropylene (PP)	Polysulfone (PSO), Cellulose acetate (CA), Polyvinylidenedifluoride (PVDF), Polypropylene (PP)
Membrane module	Tubular, hollow fiber	Tubular, hollow fiber, spiral wound, plate-and-frame
Operating pressure (kPa)	100-1000	<200

weight of 27,500 Da).

2-3. Membrane Filtration and Membrane Hybrid Systems

Advances in low pressure driven membrane technologies such as microfiltration (MF) and ultrafiltration (UF) have permitted their use in stormwater due to their high efficiency, ease of operation and small footprint, [144]. MF that is generally used has a pore size of 0.1-0.2 μm, although there are exceptions, as MF membranes with pores sizes of up to 10 μm are available. For UF, pore sizes generally range from 0.01-0.05 μm or less, [145]. In addition, in terms of a pore size, the lower cutoff for a UF membrane is approximately 0.005 μm, [145]. A basic comparison between MF and UF membrane is given on Table 10.

Membrane filtration can reduce the turbidity of stormwater from 25-35 NTU to less than 0.29 (i.e 98% removal), [143]. The average TOC removal by submerged membrane itself was only about 7%. Microfiltration alone cannot remove the TOC due to its large pore size (0.3 μm). The TOC removal is due to adsorption of organics on membrane.

Membrane filtration is usually coupled with a pre-treatment of fibre filter or deep-bed filter and such systems are called membrane hydrid systems. This significantly improved the removal efficiency of the system yielding high quality reuse water. An in-line flocculation-filtration followed by hollow fibre membrane system improved the TOC removal efficiency from 7% to 40% while achieving almost complete removal of turbidity.

2-4. Bio-Filter

Biofilter typically comprises a column of support media (typically granular activated carbon, (GAC)) onto which microbes grow. In the initial stage of operating the biofilter, adsorption of substances including micro-organisms is the dominant process, while in the later stages organic degradation by microbial activities becomes more important. The activities of microbes determine the performance of biological filtration in removing pollutants.

Mohammad et al. [147] presented the results of the long term biofilter experiments conducted with raw stormwater collected from a stormwater canal in Sydney. GAC were used as a single filter media in biofilter columns. Media heights of 75 and 40 cm were used. The filter columns were operated at filtration velocities of 0.12 and 0.25 m/h. The removal efficiency for turbidity and DOC for the GAC filter media were found to be 75% and almost 100% respectively.

Based on a limited sample of stormwater, the removal efficiency for phosphorus was upto 74% and that of nitrogen was up to 30%. The removal of zinc, iron, lead and nickel were good (upto 90%) although the concentration of heavy metal in the raw surface water sample was already low. The highest DOC removal efficiency of up to 99% was achieved using the GAC media filter. MWD showed that the GAC biofilter removed a large range of organic compounds present in the stormwater. The colour removal was 95% using GAC filter.

Biofilter is an environmentally friendly pre-treatment which can remove the majority of organic matter in stormwater. This biofilter (1) removes the majority of dissolved organic matter and reduces/eliminates the biofouling on the subsequent membrane process; (2) requires only a small amount of energy; (3) does not require regeneration of GAC; (4) does not involve any chemical requirement; (5) achieves significant removal of dissolved organics for a long period of time with little operation and maintenance and (6) is simple to construct and operate.

CONCLUSION

Non-point pollution is a major source of pollutants in urban environments. A wide range of pollutants deposited on urban catchment surfaces are transported with stormwater during the wet weather periods and ultimately enter aquatic receiving waters. These pollutants are mainly due to human activities in urban and suburban areas. Among the various pollutants, suspended solids, heavy metals, polycyclic aromatic hydrocarbons and nutrients were the focus of previous studies. A wide variation in concentration of pollutants has been reported. Heavy metals, polycyclic aromatic hydrocarbons and nutrients are found both in dissolved form and in particulate bound form. The partition of the pollutants varies from place to place and also depends on the season. In particulate form, most of the pollutants are bound to the finer fractions, especially that less than 100 μm. The types of land-use activities present in a drainage catchment are important in determining the stormwater quality. Treatment systems are available for stormwater runoff and are predominantly natural treatment system. Treatment systems for stormwater harvesting have been used successfully in water and wastewater treatment and include fibre filters, deep bed filters and biofilters. They can

achieve a relatively high pollutant removal at a high rate which is necessary to treat stormwater without the need for pre-treatment storage.

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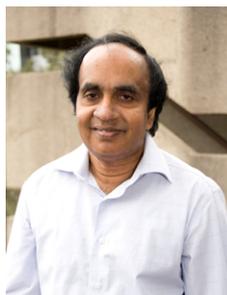
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