

THE SOURCES, IMPACT AND MANAGEMENT OF CAR PARK RUNOFF POLLUTION: A REVIEW

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Abstract

Traffic emissions contribute significantly to the build-up of diffuse pollution loads on urban surfaces with their subsequent mobilisation and direct discharge posing problems for receiving water quality. This review focuses on the impact and mitigation of solids, metals, nutrients and organic pollutants in the runoff deriving from car parks. Variabilities in the discharged pollutant levels and in the potentials for pollutant mitigation complicate an impact assessment of car park runoff. The different available stormwater best management practices and proprietary devices are reported to be capable of reductions of between 20% and almost 100% for both suspended solids and a range of metals. This review contributes to prioritising the treatment options which can achieve the appropriate pollutant reductions whilst conforming to the site requirements of a typical car park. By applying different treatment scenarios to the runoff from a hypothetical car park, it is shown that optimal performance, in terms of ecological benefits for the receiving water, can be achieved using a treatment train incorporating permeable paving and bioretention systems. The review identifies existing research gaps and emphasises the pertinent management practices as well as design issues which are relevant to the mitigation of car park pollution.

Highlights

- Impervious car park surfaces represent a major source of urban water pollution.
- Surface deposition processes promote the build-up of solids, metals and organics.
- Rainfall characteristics influence the pollutant levels in car park runoff.
- Car park runoff management and design issues are addressed.
- An impact assessment approach identifies optimal pollutant mitigation treatments.

Keywords: car park runoff quality, surface deposition, stormwater wash-off, pollutant mitigation, ecological benefits, surface water management.

GLOSSARY

AA: annual average
BMP: best management practice
BOD: biochemical oxygen demand
COD: chemical oxygen demand
DEHP: di(2-ethylhexyl)phthalate

DIDP: diisodecyl phthalate
DIN: dissolved inorganic nitrogen
DINP: diisononyl phthalate
EA: Environment Agency for England and Wales
EMC: event mean concentration
EQS: environmental quality standard
EU: European Union
FIO: faecal indicator organism
GIS: geographic information system
HC: hydrocarbon
HMWB: heavily modified water body
IUPAC: International Union of Pure and Applied Chemistry
LUPI: land use area pollution index
MAC: maximum allowable concentration
PAH: polycyclic aromatic hydrocarbon
PCB: polychlorinated biphenyl
PI: pollution index
PM₁₀: particulate matter finer than 10 µm
PMI: pollution mitigation index
PSD: particle size distributions
RE: river ecosystem
SPI: site pollution index
SUDS: sustainable drainage systems
SWMM: Stormwater Management Model
TKN: total Kjeldahl nitrogen
TN: total nitrogen
TOC: total organic carbon
TP: total phosphorus
TPH: total petroleum hydrocarbons
TS: total solids
TSS: total suspended solids
US EPA: United States Environmental Protection Agency
WFD: Water Framework Directive

1. INTRODUCTION

The provision of car parking spaces continues to expand as the number of cars increases together with their associated use for work and leisure activities. Current global estimates are that there are 600,000,000 passenger cars and this number continues to grow daily (Ben-Joseph, 2012). Car parks (also referred to as parking lots) have become a key aspect of both transport and land use planning in connection with the development of shopping centres and supermarkets, cinemas, sporting arenas, factories and office complexes. In the USA, it has been estimated that a land area of approximately 9104 km² is occupied by car parks (Ben-Joseph, 2012) with 67% being classified as shopping and retail centres (National Parking Association, 2011). European projections indicate the existence of over 28 million parking spaces (Surinach et al., 2011), whilst in the UK, estimates range up to 11.3 million spaces in regulated facilities (British Parking Association, 2013) which equates to a land take of 214 km². Environmental and human impacts associated with car parks include excessive traffic movements, inefficient use of land, risks to pedestrians, air pollution and water pollution through the generation of polluted runoff (Mugavin, 1995).

Car parking surfaces are typically impervious (e.g. asphalt, concrete) but may have a gravel base or in more recent cases be constructed using permeable paving materials. Where an impervious car park surface exists, virtually all the incident rainfall, except for that removed by evaporative losses, produces surface runoff which needs to be removed efficiently to prevent localised flooding and to ensure safe driving conditions (Kalantari and Folkesson, 2012). These requirements have driven the installation of effective drainage systems which rapidly transport the surplus water to outlets, often discharging directly to receiving water systems where increased peak flow discharges may result in accelerated receiving stream erosion and down stream flooding. Car parks, like roads, represent a major source of water pollution in urban areas with the range of pollutants typically associated with car park runoff including total suspended solids (TSS), metals, anthropogenic organic compounds, nutrients and microbial contaminants (Gobel et al., 2007). Davis et al. (2010) estimated the amount of runoff which would be generated by the area occupied by car parks in Tippecanoe County, Indiana, USA was 900% higher than before the land was converted. Associated increases in pollutant loads were calculated to be by factors of 6.2 for TSS, 3.5 for total nitrogen (TN), 3.0 for total phosphorus (TP) and much greater for metals with a 137-fold enhancement for Zn.

Car park runoff quality, like all diffuse urban runoff quality, can be expected to be highly variable (Freni et al., 2008) and is subject to factors such as catchment surface type, storm intensity and frequency and the antecedent conditions (Greenstein et al., 2004). The latter is considered by many investigators to be an important variable and has consistently been incorporated into modelling relationships to describe the build-up of solids, and their associated pollutants, on urban surfaces. The subsequent wash-off of surface accumulated pollutants contributes significantly to diffuse pollution loads which are subject to mitigation in line with the ambitious targets established by the EU Water Framework Directive (EU WFD, 2000).

To protect the quality of receiving waters, it is evident that changes to traditional drainage practices will be necessary particularly in the face of increasing urbanisation and associated car ownership and use. Traffic is a major contributor to diffuse pollutant loads in many urban and suburban environments. Whereas the management of road runoff quantity and quality typically comes under the remit of highways agencies and municipalities, the large car parks associated with the growing numbers of commercial/retail parks are often owned by private companies, many of whom are as yet unaware of the emerging need to control the quality of water discharging from their sites. In order to contribute to a widening of the knowledge which exists for car park derived pollution, this review initially considers the processes leading to the build-up of surface sediments and associated pollutants and how these aspects influence the quality of car park runoff. The focus is specifically on the behaviours of solids, metals, organic pollutants and nutrients for which the specific sources to car park surfaces are identified in Figure 1. Subsequently, the available treatment processes and techniques for reducing the polluting potential of stormwater deriving from car parks are discussed and an impact assessment procedure is described for determining the ecological benefits for receiving waters which can be achieved by treating car park runoff.

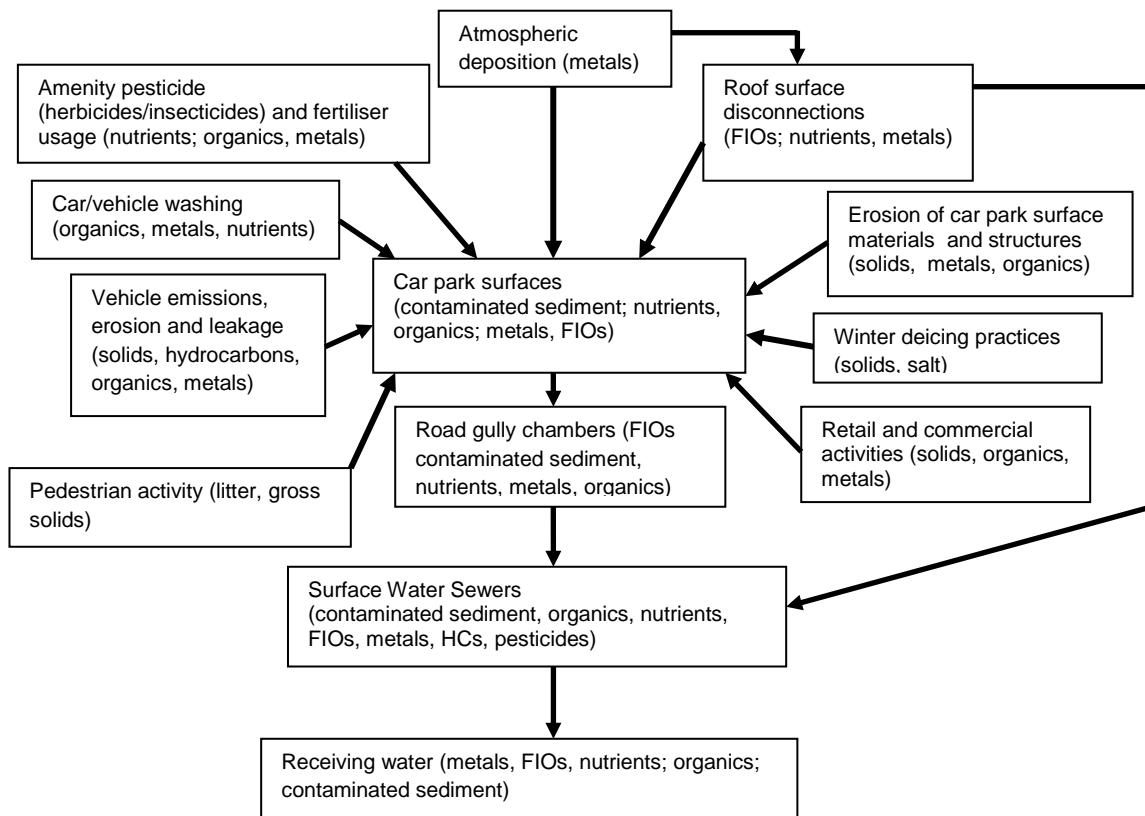


Figure 1. Principal sources and pathways of pollutants contributing to car park runoff.

2. SURFACE DEPOSITION AND BUILD UP

To date there has been little reported work on the accumulation processes which influence the behaviours of solids and their associated pollutants on car park surfaces with the focus being on road surfaces (e.g. motorway, urban, residential). Busy residential roads are considered to behave similarly to car parks in relation to controlling influences such as traffic densities and speeds. The range of contributing pollutant sources which can potentially be found on car park surfaces are identified in Figure 1. Total solids (TS), which represent particles smaller than 6 mm, have been traditionally taken as the reference parameter for the consideration of accumulation rates for road, and by inference car park, surfaces.

2.1 Accumulation modelling processes

Most surface pollutant accumulation models are based on semi-empirical formulations in which the antecedent dry period (t) is considered to be an important independent variable. A wide range of mathematical models including linear, power, exponential and Michaelis-Menton functions have been used to describe the temporal build-up process (Huber and Dickinson 1988). However, the most widely employed predictive relationships for urban surfaces are the exponential (Bertrand-Krajewski et al., 1993; Charbeneau and Barrett, 1998; Deletic et al., 1997; Shaheen, 1975) and power functions (Ball et al., 1998; Egodawatta et al., 2013).

The exponential equation defining pollutant build-up can be expressed as:

$$dP/dt=k_0 - k_r P \quad (1)$$

where P = amount of pollutant per unit area on the catchment surface (kg m^{-2}), k_0 = constant rate of pollutant deposition ($\text{kg m}^{-2} \text{h}^{-1}$); k_r = constant pollutant removal rate (h^{-1}) and t = antecedent or inter-event time (h).

Due to the inability of a storm event to completely remove all solids and associated pollutants, modifications to Equation 1 have been introduced to account for the residual or initial amount of pollutant available on the catchment surface after the previous runoff or street sweeping event (Chen and Adams, 2006; Osuch-Pajdzinska and Zawilski, 1998; Zhang and Yamada, 1996). Additionally, Alley and Smith (1981) observed that the combination of wind and vehicle effects as well as the chemical and biological decay of some constituents resulted in the surface pollutant accumulation rate being greatest in the time immediately following the previous runoff or cleaning event after which it decreases and eventually approaches a constant rate. On this basis, Equation 1 can be modified as follows:

$$P = P_m(1 - e^{-k_r t}) + P_0 e^{-k_r t} \quad (2)$$

where $P_m (=k_0/k_r)$ represents the maximum amount of pollutant build-up (kg m^{-2}) and P_0 = residual amount of pollutant after the previous runoff or street sweeping event (kg m^{-2}).

Using Equation 2, Chen and Adams (2006) predicted the build-up parameters for a 54.6 ha catchment composed mainly of low to medium density residential properties combined with some commercial land use located in Toronto, Canada. The values established for the maximum pollutant build-up (P_m), the residual amount of pollutant (P_0) and the pollution deposition rate (k_0) vary significantly for different pollutants with the average maximum build-up parameters tending to be low for metals and highest for TS and chemical oxygen demand (COD) (Table S1 of the supplementary material). The high variability in the predicted pollutant deposition rates is not necessarily matched by the pollutant removal rates (k_r) which remain substantially constant for all the considered pollutants (Table S1).

The power equation which has been shown to perform well in describing pollutant build-up on urban surfaces is:

$$B = a D^b \quad (3)$$

where B is the build-up rate ($\text{g m}^{-2} \text{d}^{-1}$), D is the number of antecedent dry days, a is a multiplication build-up factor related to the build-up load and b is a power build-up coefficient which explains the build-up process. Typically the values of b are negative (Egodawatta et al., 2013) indicating that during dry conditions the build-up process decreases gradually after an initial high accumulation and eventually reaches an almost constant value after 14 days. This predicted enhanced early build-up combined with the amounts of material which can potentially be deposited on surfaces similar to those found in car parks indicate the need for regular cleaning to ensure the effective removal of pollutants, particularly those associated with coarse solids.

2.2 Accumulation of particulates/solids

Sources of particulate matter deposited on car park surfaces can include traffic, atmospheric deposition, roofing materials and street furniture (e.g. lighting, barriers and signage), general litter, accidental spills and erosion of soils from surrounding areas via both direct and indirect deposition routes. Traffic has been identified as an important contributor of particulate materials in urban areas with specific sources including the wear and tear of car bodies and engines, abrasion of brakes and tyres, exhaust emissions, friction-assisted break-up of paving and road materials and de-icing/anti-skid materials (Gunawardana et al., 2012; Hengren et al., 2006; Sorme, 2003; Thorpe and Harrison, 2008; Westerlund, 2005). Thorpe and Harrison (2008) distinguished between particulate matter emitted from exhaust emissions and that derived from non-exhaust emissions and concluded that they contributed similar loads to local traffic related emissions. Hence, the imposition of increasingly stringent emission control legislation will not completely reduce the loads of particulates deriving from vehicular activities and supplementary treatment options will continue to be required.

In addition to urban land-use type and the prevailing weather conditions, the mass of particulate matter accumulated on an urban surface is influenced by surface texture depth. For example, a residential area with a surface texture depth of 0.76-0.92 mm produced an average solids loading of 0.81-1.79 g m⁻² which increased to 1.75-2.22 g m⁻² for a commercial area with a surface texture depth of 0.63-1.11 mm (Gunawardana et al., 2012). These values are considerably lower than the model predictions for TS obtained by Chen and Adams (2006) (Table S1) which are more compatible with the surface loading values reported by Carraz et al. (2006) for urban roads (7.3 to 740 g m⁻²) and by Vaze and Chiew (2002) for a series of car parking bays (5-70 g m⁻²). Within the non-exhaust emissions category, re-suspension and emissions from brake and tyre wear have been identified as the largest sources of PM₁₀ with the particle mass, size and composition at any particular site being determined by factors such as material composition of the surface and vehicle components, their state of wear and the mode of driving (Thorpe and Harrison, 2008). The mode of driving associated with car parks together with low speeds combined with repeated turning and wheel movement, will contribute to the substantial surface loadings observed. However, direct comparisons between different investigations are complicated by the variations in the sampling methodologies employed and future research investigations would benefit from a consistent approach.

Urban particulate matter is composed of materials which characteristically reflect a wide range of size fractions and compositions with regard to their mineralogy, organic content and associated pollutant load. Both the pollutant load and size distribution of particulates influences their behaviour and subsequent environmental impact with physicochemical mechanisms such as dissolution, entrainment and settling rates raising particular concerns for the finer fractions of particulate matter (McKenzie et al., 2008; Thorpe and Harrison, 2008; Wei and Morison, 1994). Various researchers have defined the fine fraction of surface sediments using different cut-off points, but typically the descriptor 'fine fraction' relates to the <63 µm, <75 µm or <150 µm fractions. Particle size distributions (PSD) for road dust samples collected from a range of land-use activities, with similarities to car parks, are presented in Table 1. For deposited dusts, the relationship between mass and particle size fractions is not always consistent although there is a tendency for the majority of the mass to reside in the finer particle fractions which is the opposite of the usually reported trend for urban surface sediments (Li et al., 2006; Vaze and Chiew, 2002, 2004). Hengren et al. (2006) compared 3 different urban sites and found that the largest amount of sediment collected was from a large supermarket car park (570 car parking spaces) with the finer than 75 µm size fraction contributing 60.9% (Table 1). In contrast, for a similarly sized university parking lot, Kayhanian et al. (2012) found that only around 5% of the collected particulate mass was less

than 38 μm in diameter. There is, however, agreement on particle numbers with car park surface dusts containing predominantly fine particles with reports of 97% being less than 38 μm (Kayhanian et al., 2012) or over 90% being below 150 μm (Herngren et al., 2006). Since street sweeping practices only efficiently remove coarser particles (German and Svensson, 2002), the problems posed by the finer, more contaminated particles will remain a significant problem in runoff where they tend to dominate on a mass distribution basis (Kayhanian et al., 2012). Therefore, additional treatment practices are required to deal with the levels of fine sediments which are expected to be washed off car park surfaces.

Table 1. The percentage masses of sediments in each size fraction reported for deposited urban dust for a range of land-use activities with similar traffic usage to car parks.

	<0.4 5 μm	0.45-75 μm	76-150 μm	151-300 μm	>300 μm
Residential ^a	8.6	58.1	23.3	5.8	4.2
Residential ^b	10.9-12.0 ^c		10.1-18.9	46.4-56.8	13.5-32.6
Supermarket car park ^a	7.1	53.8	28.9	8.0	2.2

^a Herngren et al. (2006)

^b Dong and Lee (2009)

^c percentage mass range covers the <0.4 – 75 μm size fraction

2.3 Accumulation of metals

Vehicles are frequently identified as an important source of metals in the urban environment (Lundy et al., 2012a; Revitt, 2004; Sorme, 2003; Thorpe and Harrison, 2008), particularly at locations heavily influenced by traffic through both direct deposition and subsequent re-suspension processes. A range of particulate associated metals are produced by friction or abrasion within braking systems and between tyres and surface materials. The wide range of metals reported to be present in brake linings and tyre treads are identified in Table S2 of the supplementary material indicating that their contents may cover several orders of magnitude leading to highly variable erosion rates which will also be influenced by mode of use (Thorpe and Harrison, 2008). Metals such as V, Ni, Fe, Mg and Ca have been detected in road bitumen samples although they may originate from sources other than the bitumen itself (Kennedy and Gadd, 2003). In a separate review, Sorme (2003) reported metal concentrations of 13 mg Cu kg⁻¹, 52 mg Zn kg⁻¹, 24 mg Pb kg⁻¹, 4 mg Cr kg⁻¹, 0.5 mg Ni kg⁻¹ and 0.09 mg Cd kg⁻¹ in asphalt.

The concentrations of a range of metals reported in road dusts from a supermarket car park and from urban areas with similar characteristics to car parks are presented in Table 2. The studies were conducted in the same geographical region but represent diverse ranges of metal concentrations deriving from similar land-use types with the results reported by Herngren et al. (2006) being two to three orders of magnitude lower than those of Gunawardana et al. (2012). There is no obvious explanation for these large variations although it is accepted that differing metal concentrations will occur in these environments due to factors such as traffic density, weather conditions and neighbouring land-use activities. However, the metal concentrations reported by Gunawardana et al. (2012) do appear to be more consistent with other comparable studies (e.g. Wei and Yang, 2012) and with the levels to be expected given the identified metal contents of the source contributions.

Table 2. The concentrations (mg kg^{-1}) of a range of metals reported for urban road surface dusts from different land-uses, including car parks.

Land use	Cr	Ni	Cu	Zn	Cd	Pb
Residential ^a	14.5±8.7	7.92±5.00	131.4±26.0	296.6±78.0	0.51±0.10	32.5±12.2
Mixed ^a	9.37±2.30	7.01±1.6	98.4±24.0	236.5±42.0	0.35±0.10	29.1±4.70
Commercial ^a	3.16±0.40	4.53±0.07	70.8±20.0	90.4±23.0	0.54±0.10	38.4±9.50
Residential ^b	0.012		0.50	1.27	0.002	0.03
Supermarket car park ^b	0.023		0.27	0.38	0.004	0.25

^a Gunawardana et al. (2012)

^b Hergren et al. (2006)

The concentrations of metals associated with the various particle size fractions have been determined by several researchers. In an early study, Wei and Morrison (1994) found the expected decreasing concentrations of Pb and Pt with increasing particle size over the size ranges <63 μm , 63-125 μm , 125-1000 μm and an associated increasing toxicity with decreasing particle size. Although reporting low metal contamination, Hergren et al., (2006) found that more than half of the total metal concentrations in a car park dust were in the particles finer than 75 μm . However, when compared to a residential site there was a metal increase in the larger particles which was explained by the relatively higher traffic flows in the busy commercial area. The metal loadings determined for the car park site paralleled the sediment loadings and were consistently highest in the 0.45-75 μm fraction with the highest value being recorded for Fe (52 $\mu\text{g m}^{-2}$). These fine particles will also be the most easily washed off the surface highlighting the need for effective treatment to eliminate metal pollution in runoff discharged from car parks.

2.4 Accumulation of organics

Both gaseous and particulate emissions of total petroleum hydrocarbons (TPH) are produced by petrol and diesel cars as a consequence of incomplete combustion processes (Bjorkland, 2011) with their dispersion being influenced by factors such as exit velocity, exit angle and cross-wind intensity (Ning et al., 2005). However, it is likely that the deposition location will be closer to the source in car parks due to the slow moving traffic. Table S3 of the supplementary material identifies the range of vehicle and surface related sources which can give rise to polycyclic aromatic hydrocarbons (PAHs) and the resulting concentrations which have been found in traffic related sediments (Mostafa et al., 2009). Although not expected to be comparable to the situation in car parks, heavily trafficked streets are included to illustrate the increased levels which can occur relative to residential streets.

In a study of the total concentrations of the 16 US EPA PAHs in street dusts collected from a range of urban land use areas, Dong and Lee (2009) found a dependence on factors including traffic density, vehicular speed and the age of the asphalt road surfacing materials. In particular, PAH levels were almost five times higher (245 mg kg^{-1}) on a slow one-way street compared to a busy, wider and faster moving road. This was explained by a combination of the lower traffic speed combined and a reduced frequency of street sweeping on the former. A similar situation should not be allowed to occur on car parking areas. As well as determining the PAH concentrations in total dust samples, Dong and Lee (2009) also reported size range distributions (<75 μm , 75-180 μm , 180-850 μm and 850-2000 μm particle sizes) with the PAH concentration paralleling the general metal trend by increasing with decreasing particle size.

Phenolic resins are commonly used as binders in braking systems and may constitute 20-40% of the total brake lining system (Thorpe and Harrison, 2008). Tyres typically consist of a rubber matrix reinforced with steel and textiles, with rubber hydrocarbons reported to contribute 45-55%, carbon black 22-30% and hydrocarbon oils and resins 5% each to the overall composition. A further key source of organic materials is asphalt road material, which is typically composed of mineral aggregate (95%) supplemented by a combination of bituminous binders and a range of modifiers and fillers such as coal fly ash and rubber tyres to improve the road performance properties (remaining 5%).

3. STORMWATER WASH-OFF

As it travels over surfaces, stormwater can mobilise and transport pollutants deposited on urban surfaces to varying degrees depending on the use and nature of the catchment surface, rainfall (intensity, frequency and duration) and runoff volume. A generic overview of the pathways adopted by car park generated pollutants is provided in Figure 1. The identified exit routes concentrate on separate stormwater discharges with the only treatment stage being the use of gully pots, which themselves can act as pollutant sources. Pollutant transformations may occur due to natural and anthropogenic changes within a particular catchment and this may complicate the identification of the specific sources of pollutants reaching receiving waters.

3.1 Wash-off processes

The physically-based simulation modelling procedures available for describing wash-off processes and for predicting runoff quality arising from urban surfaces have been discussed by Huber (1986). Most models use suspended solids as the primary indicator pollutant with the assumption that other pollutants, such as heavy metals and hydrocarbons are adsorbed to the particulate surfaces (Herngren et al, 2005; Sartor et al, 1974). The complex removal processes contributing to pollutant wash-off from impervious surfaces have relied on semi-empirical descriptions (e.g. Sartor et al, 1974) which mirror the functional formulations used for pollutant build-up. The model that has been most widely adopted is an exponential relationship where the rate of wash-off is assumed to be directly proportional to the amount of material remaining on a surface:

$$dP/dt = -k I P \quad (4)$$

Integration of this equation yields the relationship:

$$P = P_0 (1 - e^{-kIt}) \quad (5)$$

where P is the amount of material/pollutant mobilised after time t, P₀ is the initial amount of material/pollutant on the surface, k is the wash-off rate constant or coefficient and I is the rainfall intensity.

Variations on this exponential wash-off model have been used in the US EPA's Stormwater Management Model (SWMM) (Huber and Dickinson, 1988) and in the US Army Corp's STORM model (USACE, 1977). From an investigation of the reliability of the exponential wash-off model using simulated rainfall events, Egodawatta et al (2007) found that a storm event only has the capacity to remove a fraction of the available pollutants. This was

accounted for by incorporating a ‘capacity factor’ (C_F ; values between 0 and 1) which is influenced by the kinetic energy of the rainfall, the characteristics of the pollutants, and the surface slope and condition. A modified exponential wash-off equation was suggested as follows:

$$P = C_F P_0 (1 - e^{-kt}) \quad (6)$$

The wash-off rate coefficient, k (mm^{-1}), is an empirical parameter which has no direct physical meaning but has been related to the ease of pollutant release with higher values corresponding to soluble street surface contaminants e.g. de-icing salts (Ellis and Revitt, 1989). Generally, to reduce the complexity of the wash-off equation whilst still providing reliable estimations, water quality models use a constant value for k (Huber and Dickinson, 1988). Although there is currently no data relating directly to modelling rainfall induced pollutant wash-off from car park surfaces, Egodawatta et al (2007) report k values ranging from 5.6×10^{-4} to $8.0 \times 10^{-4} \text{ mm}^{-1}$ for solids wash-off from 3 residential roads with the higher value being considered to be most representative. This compares with a value of $1.7 \times 10^{-3} \text{ mm}^{-1}$ for the wash-off of solids from low trafficked residential streets which was proposed by Ellis and Revitt (1991) based on an earlier review of the literature. Chen and Adams (2006) have predicted the wash-off rate constants for a range of pollutants from a residential area in Toronto, Canada and obtained highest values for TSS, TKN and TP (1.27×10^{-2} to $1.78 \times 10^{-2} \text{ mm}^{-1}$) with the lowest values for COD ($3.0 \times 10^{-3} \text{ mm}^{-1}$).

High intensity rainfall events can mobilise coarser particles due to the creation of high turbulence. Egodawatta et al. (2007) found that around 50% of the solid associated pollutants were mobilised by intensities of between 40 and 90 mm h^{-1} . However, these results were derived from constant intensity simulated rainfall events, whereas rainfall temporal patterns during actual storms are typically highly variable. Brodie and Egodawatta (2011) have extended the constant-intensity wash-off concept to actual storm event runoff and tested the outcome using measured suspended particle load data from a road site. The best agreement was found to be for intermediate-duration storms (duration >1 h but <5 h) as particle loads in the storm event wash-off increased linearly with average rainfall intensity. However, above a threshold intensity, there was evidence of the particle load reaching a plateau level.

Soonthornnonda et al. (2008) have presented an exponential wash-off rate equation for a range of stormwater pollutants based on a linear build-up of pollutant mass and applied this to residential and open land areas. They define a transport coefficient (c) and use this to predict that the ability of metals to be removed from a surface is ranked in the order $\text{Pb} > \text{Ag} > \text{Zn} > \text{Cu} > \text{Ni} > \text{Hg} > \text{Cd}$. This is interpreted in terms of a decreasing degree of particle association with Pb having the highest, and Hg and Cd the least particle affinity which is comparable to the order of total removal rates ($\mu\text{g m}^{-2} \text{ cm}^{-1}$ runoff) from a highway surface reported by Ellis et al. (1987) of $\text{Pb} (370) > \text{Zn} (348) > \text{Cu} (92.2) > \text{Cd} (4.80)$. TSS were predicted by Soonthornnonda et al. (2008) to be preferentially removed from all drainage areas with TP being least efficient due to limited accumulation as a consequence of solubility considerations.

Critics of the exponential wash-off model have advocated the need for a more detailed interpretation of the physical/hydrological processes which influence the removal of pollutants from urban surfaces. This has led to the development of deterministic algorithms based on sediment transport theory (Price and Mance, 1978; Sonnen, 1980) and their incorporation into models such as MOSQUITO (Payne et al, 1990), MouseTrap (Garsdal et al,

1995) and HydroWorks(TM) (David and Matos, 2002), where surface solids are removed by a combination of raindrop impact and overland flow. The removal potentials of other pollutants are then estimated as a fraction of the sediment mass discharged through the application of "potency factors". Although this approach may lend itself to easier calibration than the exponential wash-off formulations, which require the establishment of reliable wash-off rate constants, the deterministic methodology is limited by the ability of the modeller to define appropriate values for the threshold shear stresses (Deletic et al, 1997) as well as other parameters in relation to the complex surface micro-topography which comprises even the smallest of drainage inlet conditions.

3.2. Pollutants in run-off

Pollutant concentrations (expressed as event mean concentrations; EMC) and loadings in runoff from urban areas with similar traffic uses to those experienced by car parks are shown in Table 3. The values quoted are for total pollutants with no distinctions between those pollutants associated with the particulate and soluble phases. Car parks have been grouped together with commercial areas as the two are often synonymous in terms of usage and are frequently exposed to similar vehicle activities. Concentrations of TSS are lower than those deriving from residential and urban road surfaces but the ranges of solids loadings indicate the potential for enhanced runoff processes from car park/commercial surfaces. Comparisons for the metals are dependent on type with some metals (e.g. copper and zinc) showing comparable car park runoff concentrations to those found from suburban roads. There is no hydrocarbon data available specifically for car parks but runoff from commercial areas matches the ranges of concentrations which have been observed being washed off urban roads. The TSS and metal concentration data for gully pot liquors have also been included in Table 3 because, although intended to be treatment devices, in the absence of regular cleaning they have the potential to become pollutant sources.

Although many of the pollutants identified in Table 3 may be present as either the dissolved or particulate phases, it is widely reported that the majority of the urban pollutant load is associated with particles which act as mobile substrates (Bjorkland, 2011; Herngren et al., 2006; Lee et al., 1997; McKenzie et al., 2008; Vaze and Chiew 2002, 2004). The finer particles are more readily mobilised during rainfall as evidenced by reported particle size distributions for urban surface runoff (Table 4). Particle size distributions reported on a volume, number or mass basis all identify the predominance of finer particle sizes (often finer than 10 μm) in the runoff from heavily used roads with a tendency to a slightly coarser distribution being observed for car parks and residential roads. The increased surface area associated with finer particles, combined with a higher cation exchange capacity, provides them with a greater affinity for pollutants, such as metals (Dong and Lee, 2009; Liebens, 2001; Ujevic et al., 2000). Hence, the finer fractions contribute more pollutants per unit mass and following wash-off will remain in suspension longer providing the opportunity to travel further (Herngren et al., 2006; McKenzie et al., 2008). Knowledge of the particle size distribution of the pollutant load and the environmental behaviour of associated size fractions influences the preferential removal mechanism and hence the selection of surface runoff pollutant mitigation measures. These studies further emphasise that street cleaning alone (with its preferential removal of coarser particles) will not be successful in mitigating the pollutant loads associated with car park runoff, and that systems capable of addressing the fine particulate fraction such as Sustainable Drainage Systems (SUDS) or stormwater Best Management Practices (BMPs) are also required.

Table 3. Ranges of pollutant concentrations (expressed as event mean concentrations) and loadings found in runoff from car parks and commercial areas and from urban surfaces with similar traffic uses to car parks (adapted from Lundy et al., 2012a).

Pollutant type	Source	Event mean concentrations	Loadings (kg/ha/yr)
Total suspended solids (mg L ⁻¹)	Car parks and commercial areas	7.8-270	12-2340
	Residential		
	High density	55-1568	130-840
	Low density	10-300	50-230
	Urban roads	11-5400	815-6289
	Roadside gully chambers	15-1840	
Metals (µg L ⁻¹)	Car parks and commercial areas	Cd: < 1; Zn: <1-700; Pb: <1-10; Cu: <1-205; Ni: 2-493	Cu: 0.03-0.04; Pb: 0.01-1.9; Zn: 0.10-0.17
	Residential roads	Cd: 0-5; Cu: 6-17; Zn: 87-150; Pb: 6-140	Pb: 0.001-0.03
	Suburban roads	Pb: 10-440; Zn: 20-1900; Cu: 10-120	Pb: 0.01-1.91; Zn: 1.9-19.0; Cu: 0.4-3.7
	Gully liquors	Pb: 100-850	
	Hydrocarbons (µg L ⁻¹)	Car parks and commercial areas	Total HC: 3.3-2,000; Total PAH: 0.35-3,000
	Residential		
	High density	Total HC: 0.67-25.0	PAH: 0.002
	Low density	Total HC: 0.89-4.5	Total HC: 1.8
	Urban roads	Total HC: 2.8-31;	Total HC: 0.01-43.3
Nutrients (mg L ⁻¹)	Car parks and commercial areas	Total N: 0.41-2.54; NH ₄ -N: 0.2-4.6; TP: 0.04-0.53; orthoPO ₄ : 0.001-0.03	NH ₄ -N: 0.38-0.81; TP: 0.20-0.34; orthoPO ₄ : 0.11-0.19
	Motorways and roads	TN:<4	NH ₄ -N: 7.2-25.1
	Residential	TN:<0-6; NH ₄ -N: 0.18-3.8; TP:0-1.16	NH ₄ -N: <0.65; TP:<0.81
	Gully Liquors	TN: 0.7-1.39	

Table 4. Comparison of the particle size distributions reported in the runoff from car park surfaces with those observed from road surfaces exposed to differing traffic densities.

Runoff type	Particle size distribution	Reference
Car park (shopping centre)	85% < 75 μm^{a}	Herngren et al. (2005)
	74% < 150 μm^{a}	Goonetilleke et al. (2009)
Car park (lorry)	91% < 150 μm^{a}	Goonetilleke et al. (2009)
Residential road	57% < 150 μm^{a}	Goonetilleke et al. (2009)
Highway/motorway	50% < 12.6 μm^{a}	Andral et al. (1999)
	90% < 10 μm^{b}	Li et al. (2006)
	96% < 25 μm^{b}	Westerlund and Viklander (2006)
	99% < 38 μm^{b}	Kayhanian et al. (2012)
	70% < 38 μm^{a}	
	67% < 38 μm^{c}	

^a based on distribution by volume

^b based on distribution by number

^c based on distribution by mass

Although there is a generally accepted correlation between increasing metal concentrations and decreasing particle sizes in stormwater (MacKenzie et al., 2008), Herngren et al. (2005) found that this distribution was more random for particles larger than 0.45 μm in the runoff from a commercial car park. In connection with this, it was also noted that the car park surface had a relatively coarse texture and repeated erosion caused by the increased stopping and starting of vehicles would result in larger sized particles capable of irreversibly binding the finer particles initially released from vehicles.

The range of fuel related organic compounds, which are frequently determined in urban runoff, includes alkanes, alkenes and PAHs all of which are produced as a result of petrol and diesel emissions, and spills and leaks of lubricants (Dong and Lee, 2009). Hoffman et al (1982) monitored the petroleum hydrocarbons contained in storm runoff from a 12.5 ha shopping mall parking area and found that between 83 and 93% was associated with particulate material and constituted between 1.7 and 3.3% of the solid material mass. Lopes et al. (2000) applied a mass balance approach to volatile organic compounds in stormwater and concluded that the more hydrophobic compounds, such as benzene, were derived from the car park surface itself following concentration in accumulated oil, grease and soot particles whereas oxygenated compounds, such as methyl tertiary-butyl ether, arose from atmospheric deposition. Asphalt has been identified as a source of PAHs in stormwater with a high-traffic volume car park yielding 0.056 $\text{g year}^{-1} \text{m}^{-2}$ of a total of 16 PAHs compared to 0.032 $\text{g year}^{-1} \text{m}^{-2}$ from a low-traffic-volume car park (Smith et al., 2000). More recently the effect of using coal tar emulsion as a sealant has been highlighted with mean PAH concentrations in runoff being elevated by a factor of 65 in runoff particles compared to unsealed asphalt or concrete based car park surfaces (Mahler et al., 2005).

There have been few reports on the wash-off of nutrients from car parking areas although a comprehensive study by Passeport and Hunt (2009) found that an asphalt surface yielded similar levels of phosphorus species, but lower levels of nitrogen species, to those found in highway runoff. The mean EMCs for TN, TKN, $\text{NH}_3\text{-N}$, $\text{NO}_3\text{-N}$, TP and orthophosphate were 1.57, 1.19, 0.32, 0.36, 0.19 and 0.07 mg L^{-1} , respectively. These values are consistent

with the range of EMCs reported for TN (0.98-2.54 mg L⁻¹) and TP (0.08-0.53 mg L⁻¹) for a Korean car park (Kim et al., 2007a) for which the nature of the impermeable surface was not identified. Rushton (2002) calculated the maximum nutrient loadings (kg ha⁻¹.year) released from an asphalt car park surface to be 2.04, 0.57, 0.81, 0.34 and 0.19 for TN, NH₃-N, NO₃-N, TP and orthophosphate, respectively.

4. CAR PARK RUNOFF TREATMENT PROCESSES

As for the majority of urban surfaces, the traditional approach to draining car park surfaces has been based on the removal of surface water as quickly as possible using effective drainage channel systems. The presence within the drainage system of gully pots represents a possible contribution to water quality enhancement but these devices can also contribute to the outgoing pollution load under certain meteorological conditions. More recently there has been increased emphasis on keeping the stormwater above the surface for treatment and for using natural processes, where possible, to remove pollutants as well as attenuating flow intensities. Such systems, commonly referred to as either SUDS or BMPs, may need to be used as part of a treatment train to achieve the level of improvement required in car park runoff quality. Proprietary systems, combining natural and manufactured pollutant removal capabilities have also been designed for the treatment of car park discharges.

4.1 Traditional drainage approaches

4.1.1 Drainage channels

Conventional drainage channels, although not specifically installed in car parks as water quality enhancement devices, have the ability to retain sediments, and associated metal and hydrocarbon pollutants, despite being exposed to persistent periods of heavy rainfall (Lundy et al., 2012b). A clear trend of decreasing sediment loads towards the drainage outlet was observed with the finer, more contaminated materials being preferentially transported identifying the need for an entrapment/treatment stage before discharge of the runoff to a receiving water. Newman et al (2013) have described a 'macro-pervious pavement system', consisting of oil separating collector channels and floating mat interceptors, for the retention and reduction of oils and suspended solids in car park runoff. The discharged effluent was found to be acceptable for release to a surface water receptor due to removal efficiencies consistently in excess of 95% for TSS, total hydrocarbons and a range of metals as result of sedimentation, sorption, filtration and possible biodegradation processes in the channels.

4.1.2 Gully pots

A prime function of gully pots is to retain the solids which are washed-off contributing surfaces during rainfall events hence reducing sediment deposition in the downstream drainage system and so protecting the environmental quality of receiving waters and, in some instances, safeguarding treatment plant efficiencies (Butler and Karunaratne, 1995; Butler and Memon, 1999; Deletic et al., 2000; Memon and Butler, 2002). However, without regular gully pot cleaning, the accumulation of sediments can lead to clogging problems with associated reductions in hydraulic efficiency and an increased possibility of urban flooding problems during storm events. There is also evidence to show that gully pots serve as poor sedimentation basins with the majority of the most heavily contaminated particulate fractions remaining in suspension (Morrison et al., 1988; Sartor and Boyd, 1972). Based on laboratory experiments, Ciccarello et al. (2012) have confirmed that solid removal efficiencies are

inversely proportional to flow rate and directly proportional to particle size and specific gravity but are not influenced by the presence of a sediment layer inside the gully pot.

In addition to operating under wet weather conditions, gully pots are also exposed to prolonged dry weather conditions during which the release of oxygen demanding contaminants can lead to the development of anaerobic conditions (Memon and Butler, 2002; Morrison et al, 1995). Morrison et al. (1995) monitored the biochemical changes occurring in the sediment and supernatant liquor between storms in a gully pot draining a small 390 m² catchment composed of a car parking area, a road and a roadside kerb. Sediment maturation and acidic dissolution processes were shown to release pollutants from the contaminated chamber sediments and interstitial pore waters into the initially relatively clean gully pot liquor which subsequently appeared as early contributions in gully pot outflows during wet weather conditions.

Particulate metal concentrations have been found to be concentrated in the finest fractions of both gully pot water samples (Karlsson and Wiklander, 2008a) and gully pot sediments (Birch and Scollen, 2003). In a comparison of the particle size distributions and metal concentrations in surface sediments and gully pots, Poletto et al (2009) found that the more polluted finer fractions were not retained by gully pots but transported to the nearest local receiving watercourses. Metal speciation schemes have been applied to gully pot sediments (Birch and Scollen, 2003; Striebel and Gruber, 1997) and to gully pot liquors and suspended solids (Morrison and Revitt, 1987) and enable the processes which influence metal mobilisation and transport within a gully pot system to be identified (Morrison et al., 1988). These include the release of free metal ions, weakly complexed metal forms and weakly bound metals to exert a potentially toxic effect coinciding with the rising limb of a storm. The increasing use of Pt and Pd in catalytic convertors has resulted in concentrations of up to 450 ng g⁻¹ in gully pot sediments with evidence for the preferential mobilisation of Pd (Jackson et al., 2007).

There have been few reports of the behaviour of specific organic pollutants within the gully pot system although Karlsson and Viklander (2008b) found the PAHs within a gully pot mixture to be predominantly particulate associated achieving combined concentrations for 16 PAHs of 10 µg g⁻¹ in the sediment phase. The most abundant individual PAHs were naphthalene, phenanthrene, fluoranthene and pyrene. Jartun et al. (2008) measured the combined 16 PAH concentrations in sediments collected from stormwater traps and detected a mean value of 7.6 µg g⁻¹ which was comparable with that reported by Karlsson and Viklander (2008b). This study also analysed a combined total of 7 PCB congeners in gully pot sediments and found much lower levels represented by a mean concentration of 0.08 µg g⁻¹ and a range of <0.0004 µg g⁻¹ to 0.704 µg g⁻¹.

4.2 Sustainable drainage/best management practice systems

In an attempt to categorise these treatment systems they have been divided into three types: infiltration/filter systems, storage facilities and alternative paving structures. It is necessary to recognise that although these names characterise a mechanism/function associated with a particular treatment system they may not fully describe all the inherent pollutant removal processes. Thus, although constructed wetlands and bioretention cells are included within the storage facility category they also provide pollutant removal through filtration within both the substrate and the vegetation components.

4.2.1 Infiltration and filter systems

Treatment systems included in this category are filter strips, swales, infiltration trenches, filter drains and soakways. However, to date the last two types have not been used to treat car park runoff. Yu and Benelmouffok (1990) identified the density and height of the grass as important parameters in filter strips with reported pollutant removal efficiencies for TSS (71%), Zn (51%), Pb (25%), total P (38%) and nitrate N (10%) being observed for runoff from a shopping mall car park. Heal et al. (2009) demonstrated that filter strips were highly effective as the first component of a treatment train receiving runoff from a lorry car park due to in-situ remediation of organic contaminants and nutrients. Although there is the potential for sediment and metal accumulation in filter strips, regeneration through replacement of the top 10 cm of soil is relatively inexpensive and presents limited disruption. Infiltration swales of different ages have been assessed for their hydraulic performance and pollutant removal potential for car park runoff over a 15 year operational lifetime (Achleitner et al., 2007) with mass balance calculations indicating efficient removal performances for hydrocarbons, Cu, Zn, Pb and Cd.

4.2.2 Storage facilities

Retention ponds, detention basins, constructed wetlands and bioretention cells have all been used to treat car park runoff. Veenhuis et al. (1989) monitored the performance of a dry detention basin, with a gravel base, receiving runoff from a 19 ha impermeable area associated with a shopping mall. The average removal efficiencies for suspended solids, BOD, COD, total organic carbon (TOC), TP and dissolved Zn were 60-80% but dissolved solids and nitrite/nitrate demonstrated negative removal. Important conclusions from this study were the need to incorporate a front-end sediment trap to reduce clogging of the filter media and the use of a liner when the protection of aquifers is important.

Constructed wetlands have been shown to be effective in removing a range of pollutants from highway runoff (e.g. Bulc and Slak, 2003; Revitt et al., 2004; Terzakis et al., 2008) but there is less information available on the use of these systems for the treatment of car park runoff. However, Ellis et al. (1994) have reported the use of a detention pond planted with five species of macrophyte for the control of pollutants discharging from a combined transit operating base/car park. The emphasis was on the vegetative removal of pollutants with *Typha latifolia* and *Sparganium* species being shown to be the most suitable for the uptake, storage and metabolism of TPH, Pb and Zn. Constructed wetlands receiving stormwater from a shopping mall car park were able to facilitate nutrient cycling and water quality maintenance functions through a significant and active microbial community and a thriving plant community (Duncan and Groffman, 1994).

Bioretention cells, also referred to as bioinfiltration systems and rain gardens, generally consist of a sand/soil/organic media substrate overlain with a mulch layer and various forms of vegetation and possess the ability to reduce runoff volumes/peak flows and pollutant (TSS, nutrients, hydrocarbons, metals) discharges through removal mechanisms such as filtration, adsorption, sedimentation and biological activity/uptake (Davis et al., 2009). Hunt et al. (2008) have monitored a 229 m² bioretention cell receiving runoff from a 3,700 m² asphalt parking area and observed that peak inflows were reduced by over 96.5% for storms with less than 42 mm of rainfall. The corresponding decreases in the concentrations of TN, TKN, NH₄-N, TP, BOD, TSS, Cu, Zn and Pb were 32%, 44%, 73%, 31%, 63%, 60%, 54%, 77% and 31%, respectively. Glass and Bissouma (2005) examined the influent and effluent from a

bioretention cell receiving parking lot runoff and found extremely high TSS removal (98%) with Cu (81%) being the most efficiently removed metal and then decreasing in the order Cu>Zn>Pb>Cd>Fe>Cr>Al. Due to efficient flow attenuation, mass removals are often higher than those based on event mean concentrations as illustrated by the average TSS performances (57% and 47%) exhibited by two bioretention cells receiving parking lot runoff (Davis, 2007). The same study reported efficient removals of TP (76%), Cu (57%), Pb (83%) and Zn (62%).

4.2.3 Alternative surface structures

Permeable surface structures have been widely used in car parking areas as a sustainable drainage technique to reduce runoff volume and discharged pollutants through storage and infiltration processes, combined with adsorption and microbial degradation, occurring within the base and sub-base. Various types of surface materials have been used including porous asphalt and concrete surfaces, concrete pavers (permeable interlocking paving systems which may also be porous), and concrete- or polymer-based grass or gravel grids and geocells. The clogging problems associated with porous materials have limited their use compared to permeable pavements but under simulated experimental conditions Yong et al. (2013) predicted the clogging potentials of different types of porous paving system to be between 12 and 26 years. Boving et al. (2008) highlighted that sand imported by cars to parking areas during winter conditions is a principal cause of clogging. The contamination of underlying soil and groundwater is a potential problem, particularly in car parks where high levels of soluble pollutants may occur (Beecham et al., 2012) and the installation of an impermeable membrane is recommended in such situations (Scholz and Grabowiecki, 2007). Concerns about the effectiveness of porous pavements in controlling stormwater runoff on clay soils was specifically tested by Dreelin et al. (2006) who demonstrated a 93% runoff reduction for a plastic matrix filled with sand and planted with grass over a gravel base compared with a conventional asphalt surface. This result was achieved for small low-intensity rain events and it may be that additional SUDS/BMPs are needed to control the peak runoff associated with large but infrequent storms. The use of plastic cellular structures with high void ratios has been proposed as a way of increasing hydraulic storage below permeable pavements (Wilson, 2008).

Hogland et al. (1990) have claimed that the use of porous concrete asphalt laid on a free-draining, crushed stone aggregate sub-base has the ability to produce significant peak flow and discharge volume reductions when used for car parking areas as well as providing a 25% cost saving. The removal of particulate associated pollutants by porous concrete occurs by a filtering mechanism within the core of the material (Chocat et al., 2013; Raimbault et al., 1999) with the added potential for metals to be adsorbed by cement containing sites (Serclerat et al., 2000). The use of grass-concrete has been compared with impermeable asphalt in dealing with car park runoff and shown to be able to reduce the discharged volume by between 65 and 100% (Smith, 1984). This study also noted the importance of the antecedent conditions with an increasing number of dry days reducing both the runoff volumes and the peak runoff rates in the case of the grass-concrete surface.

As part of an experimental study, a permeable concrete block surfaced car park laid on a sub-base constructed using four different stone types on a geotextile layer, was monitored over a 3 year period for both water volume and quality improvements in drained waters (Bond et al., 1999; Pratt et al., 1989). Compared to traditional impermeable surfaces, the volume of stormwater discharged during a rainfall event was reduced by an average of around 60%

depending on the sub-base content. The discharged concentrations of suspended solids were limited to between zero and 50 mg L^{-1} compared to up to 300 mg L^{-1} for fully impervious surfaces which is comparable to the 75-81% reduction of suspended solids observed by Pagotto et al. (2000) and Rushton (2001). Brown et al. (2009) reported that permeable pavements constructed as either porous asphalt or open-jointed paving blocks achieved an excellent removal of suspended solids (90-96%) as well as performing a 'sieving action' in which coarser solids were preferentially retained at the geotextile interface. Given these efficient removal performances it is surprising that Beecham et al. (2012) only recorded a 38% reduction in the average concentration of TSS for a permeable pavement structure receiving car park drainage.

Pratt et al. (1999) investigated the potential for the underground free draining structure to act as an aerobic digester supporting the microbial degradation of oils removed from the car park surface. Following seeding with an appropriate bacterial inoculum and nutrient mixture, 98% of an applied input oil concentration of 1800 mg L^{-1} was retained or biodegraded (Bond et al., 1999). Newman et al. (2002) studied the complexity of the developing microbial population in terms of the replacement of the original inoculums as well as demonstrating the importance of the protozoan population. Puehmeier and Newman (2008) have identified the potential role that can be played by the presence of a purpose designed geotextile in enhancing oil biodegradation.

The use of porous concrete blocks as a component of permeable paving systems has been widely used in car parks. Macdonald and Jefferies (2001) monitored water quality improvements represented by discharge reductions for TSS (32%), BOD (49%), Ni (63%) and TPH (69%) compared to an impermeable tarmac surface but increases were observed for Cu (25%) and Zn (42%). The reduction in percentage runoff was somewhat disappointing at only 22%. For a different car park, Schluter and Jefferies (2002) found that discharged metal concentrations were restricted by the use of porous concrete block surfaces (e.g. $\text{Cd} < 0.01 \mu\text{g l}^{-1}$; $\text{Zn} < 22.2 \mu\text{g l}^{-1}$) and for 16 storms the average outflow discharge volume was 46.5% of the incident rainfall. Abbott and Comino-Mateos (2003) assessed the performance of a porous block surfaced car park associated with a motorway service station and found that the average amount of water draining from the sub-base for 20 storm events was 22.5% of the incident rainfall volume with a hydraulic retention time of 2-3 days. Surface infiltration occurred both through the blocks ($250\text{-}14,000 \text{ mm h}^{-1}$) and the gaps between the blocks ($11,000\text{-}229,000 \text{ mm h}^{-1}$) but the former was considerably more susceptible to blocking. A combination of high pressure jetting and vacuum suction has been advocated for regenerating the required infiltration rates (Dierkes et al., 2002).

Legret and Colandini (1999) have specifically focussed on the behaviour of metals and by applying a mass balance scenario to a porous asphalt structure draining a street surface have determined that pollutant retention was mainly in the porous surface accounting for 89% of the incoming Pb but only 43%, 15% and 34% for Cu, Cd and Zn respectively. However, apart from Zn, these metal removal performances were considerably better than Beecham et al. (2012) have reported for a car park draining through a permeable pavement structure (Cu, 3%; Ni, 18%; Pb, 9%; Zn, 38%).

Collins et al. (2008, 2010) compared the hydrologic and nitrogen species removal of four types of permeable pavement (pervious concrete; interlocking pavers with small sized aggregate in the joints [12.9% and 8.5% open surface area]; concrete grid pavers filled with sand) with a standard asphalt surface in a 1 year old parking lot. All permeable pavements

significantly reduced surface runoff volumes and peak flow rates with the concrete grid pavers being the most efficient. Permeable pavement subsurface drainage consistently contained lower concentrations of $\text{NH}_4\text{-N}$ and TKN but only the concrete grid paver cell significantly removed $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$, probably due to the efficient filtering action of the sand in the filling and the bedding layers. Beecham et al. (2012) observed 33% and 58% reductions in TP and TN concentrations of drainage waters passing through a permeable pavement structure compared with a conventional pavement. In another comparative study, four different permeable pavement types (flexible plastic grid filled with sand and planted with grass; plastic grid filled with gravel; concrete block lattice filled with soil and planted with grass; concrete blocks with gravel in inter-block spaces) were assessed against asphalt in a car park at intervals of one year (Booth and Leavitt, 1999) and six years after construction (Brattebo and Booth, 2003). No major signs of wear were observed and incident rainfall continued to efficiently infiltrate through the permeable pavements. The infiltrated water contained significantly lower concentrations of dissolved Cu and Zn than in the surface runoff from the asphalt area and no motor oil was detected in the infiltrate.

4.3 Treatment trains.

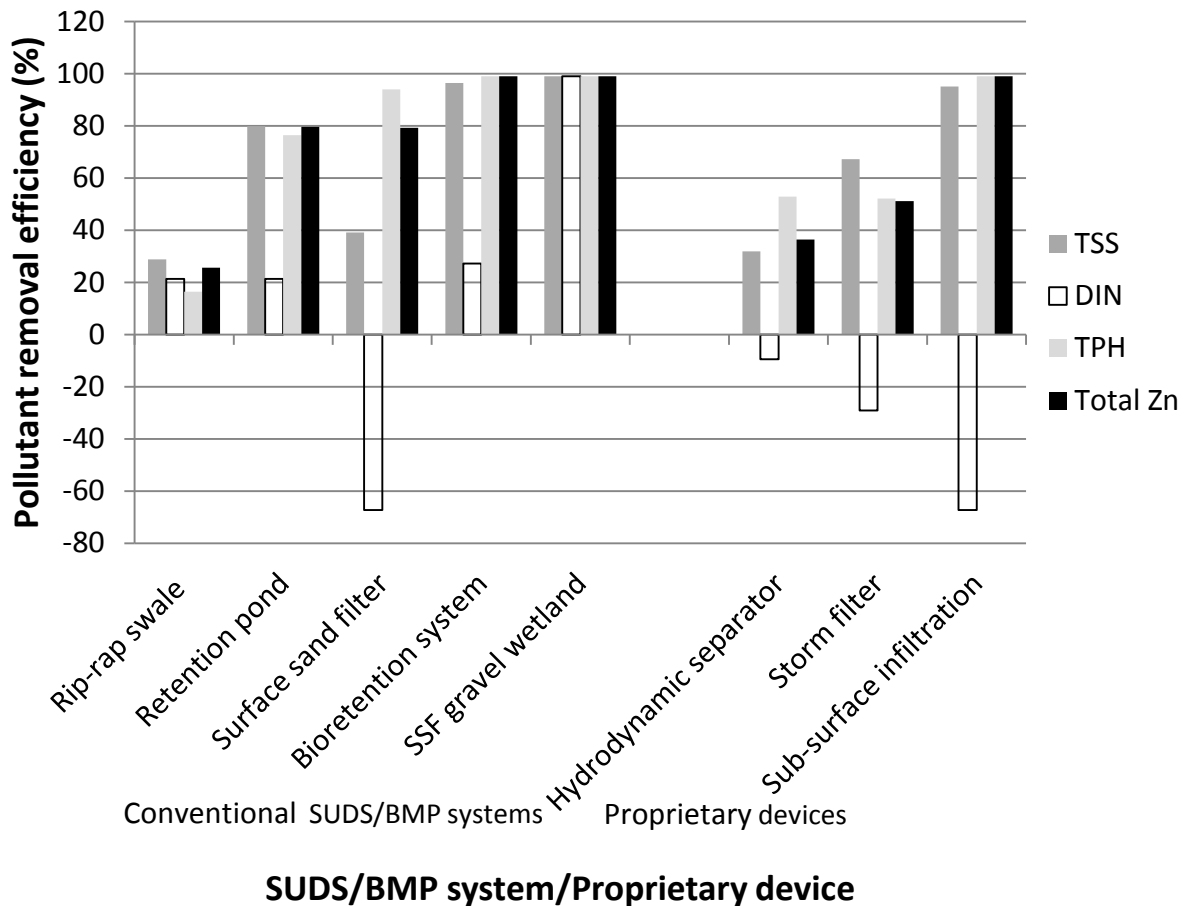
A treatment train consists of two or more treatment systems used in combination to maximise the availability of different pollutant removal processes (e.g. settlement, adsorption, filtration, microbial degradation, plant uptake). Rushton (2002) has compared the drainage from four different types of car park surface (asphalt with no swale; asphalt with a swale, concrete with a swale, and permeable paving with a swale) with final treatment in a small pond. The results confirmed that the most effective method for reducing pollutant loads is to retain the runoff on site and allow time for infiltration as well as for chemical, biological and hydrological processes to take place. Permeable pavement surfaces in conjunction with swales demonstrated the best overall pollutant removal efficiencies of up to 90% although phosphorus loads (orthophosphate and TP) were enhanced by the presence of vegetated swales. A similar comparison of surface types was conducted by Gilbert and Clausen (2006) for residential driveways and for these relatively small drainage areas the order of decreasing runoff volume was found to be asphalt>permeable paving>crushed stone. Runoff from permeable paving driveways contained significantly lower concentrations of all monitored pollutants (TSS, metals, nutrients) except for TP which was lowest from the crushed stone surface.

Heal et al. (2009) have advocated the use of a treatment train approach compared to the use of individual SUDS/BMPs and have illustrated this point through the benefits achievable with respect to flow attenuation, water treatment, spillage containment and maintenance for the treatment of runoff from motorway service station car parks. The highly contaminated runoff from the heavy goods vehicle parking area was initially treated using a 10 m wide grass filter strip, followed by a stone filled and lined infiltration trench, a spillage basin and a final attenuation pond. Based on concentration values, the average removal efficiencies within the treatment system were 84% for $\text{NH}_4\text{-N}$, 94% for BOD, 97% for TSS, 96% for total Cu and 97% for total Zn. The discharge from the coach park was collected by a conventional gully and pipe system and passed through a proprietary silt and oil interceptor prior to entering a wetland/pond/wet swale treatment train. Effluent quality leaving the pond system was assessed as not posing a problem for minimally impaired ponds except in the case of 32% of the monitored BOD levels. The car park runoff was transported by sub-surface, gravel-filled collector trenches, which provided an efficient clean-up, to a balancing pond and was subsequently identified as not posing an ecological risk. The mean sedimentation rate across

all ponds was 1.7 cm year^{-1} which is consistent with results from other urban ponds (Heal et al., 2006).

4.4 Proprietary treatment devices

The suppliers of manufactured devices may make exaggerated claims for their runoff treatment capabilities and therefore it is important to only consider impartial and independently published data and to compare, where possible, with the performances of conventional SUDS/BMP systems. Roseen et al. (2006) have conducted a comprehensive comparison of three different proprietary devices (hydrodynamic separator; storm filter; subsurface infiltration device) with five conventional treatment systems (rip-rap swale; retention pond; surface sand filter, bioretention system; subsurface flow gravel wetland) for the treatment of TSS, dissolved inorganic nitrogen (DIN), TPH and total Zn deriving from a 3.6 ha asphalt surface car park (Figure 2). Where operational problems were not encountered, the wetland was consistently the best performer (>95% removal of all four pollutants based on loadings at the inlet and outlet to the treatment system) followed by the bioretention system and retention pond which both achieved a lower removal efficiency for DIN. The most efficient proprietary device was the subsurface infiltration system which demonstrated high removals of TSS, TPH and total Zn (>95%) but increased the effluent levels of DIN. This was also the case for the storm filter which otherwise exhibited a moderate performance but was generally less efficient than the conventional sand filter. The hydrodynamic separator performed poorly across the range of four monitored pollutants which may indicate the presence of re-suspension processes. Kim et al. (2007b) identified a lack of cleaning maintenance as a cause of this and Tran and Kang (2013) noted that hydraulic residence time was a critical parameter influencing the large variability (-31% to 98%; mean 58%) in the TSS removal efficiencies achieved for the runoff from a 2,500 m² road surface.



Key: TSS = total suspended solids; DIN = dissolved inorganic nitrogen; TPH = total petroleum hydrocarbons

Figure 2. Comparison of percentage pollutant removal efficiencies achieved by conventional SUDS/BMP systems and proprietary devices.

Fuerhacker et al. (2011) have assessed a filtration system consisting of three filter chambers filled with layers of reactive media (composite, zeolite, vermiculite and granular activated carbon) and preceded by sedimentation and oil separator tanks for the treatment of car park runoff. The monitored pollutants and their mean removal efficiencies were TSS (85%), mineral oil (93%), TOC (52%), NH₄-N (71%), total Cu (75%), total Zn (73%) and Σ16 PAH (83%). An investigation of the individual treatment components showed >60% of the Cu was removed within the filter chambers, but >60% of Zn and TSS loads were removed in the sedimentation tank, oil separator and the geotextile filter, which separated each of the filter layers.

The Storm Treat system incorporates a small constructed wetland as part of the overall treatment system. The overall design allows a 5-10 day hydraulic residence time and in addition to the biochemical/plant uptake treatment provided by the wetland also provides sedimentation, oil/grease separation and filtration. In a 2 year investigation treating commercial parking lot runoff, this system was found to remove 49% TSS, 74% TP, 44% TKN, 45% total Zn and 29% Cu but only 2% total Pb on a mass basis (Stonstrom et al., 2002).

4.5 Operation and maintenance of car park runoff treatment systems

All runoff treatment options require effective maintenance to ensure continued functioning at an optimal level. It is therefore essential that an operation and maintenance strategy is developed at the initial site design and is ready for immediate implementation on completion. The nature and frequency of the required strategy will vary according to the type of system but may range from routine seasonal grass-cutting and weeding (e.g. swales and filter strips) to more specialist de-silting of gully pots (bi-annually) and pressure washing of porous surface materials (annually). More long term maintenance requirements will include the removal of accumulated sediments in facilities such as retention ponds on 20-25 year basis.

5. IMPACT ASSESSMENT APPROACH FOR CAR PARK RUNOFF

The runoff from car parking areas has the potential to make an important contribution to urban diffuse pollution and hence to jeopardise the ability of receiving water bodies to conform to the requirements of Article 5 of the EU WFD (Defra, 2012). There is a need to consider a suitable impact assessment for evaluating the role of such discharges and the mitigation which can be achieved through the use of appropriate treatment technologies. SUDS/BMP devices are attractive because of their ability to attenuate flow volumes and to remove pollutants whilst also having the potential to provide ecological/amenity benefits (Revitt et al., 2008). The selection and design criteria for SUDS/BMPs, as set out in UK guidance manuals, are essentially based on effective drainage area, site characteristics such as gradient, soil type and hydraulic infiltration rate as well as design storm event properties (Woods-Ballard et al., 2007). Risk assessment procedures for surface water flooding are now becoming well developed and tested in the UK (Environment Agency, 2010) but stormwater runoff quality has received less consideration. Recently, Ellis et al. (2012) have proposed an impact assessment procedure which identifies the principal drivers to surface water quality risk exposure as being the varied impermeable surface types and activities associated with urban land use which influence the sources and types of pollutants flushed to the drainage network. Subsequently, an assessment of the relative treatability of the runoff pollutants enables their impact on a receiving water body to be assessed. The focus in this review is on the potential residual risk posed by car park runoff to receiving waters and how this can be managed to ensure that this can be minimised.

5.1 Pollution Index (PI) assessment.

The polluting potential of an urban land use surface type can be represented by a pollutant index based on the interquartile range of EMC values for runoff generated from different rainfall events and by referencing this against regulatory EU environmental quality standards (EQS) (Ellis et al., 2012). A pollution index (PI), with values between 0 and 1, is then derived from reported EMC distributions for a given pollutant or pollutant group and the likelihood that the 50th percentile EMC values will exceed receiving water body environmental quality standards, specified either as a maximum allowable concentration (MAC) or annual average (AA) values. The PI values reported in Table 5 for three important car park runoff pollutants (TSS, TPH and Zn) have been modified from those published in Ellis et al. (2012) based on the additional evidence for industrial/commercial car parks provided in this review.

Table 5. Pollution index (PI) values derived for TSS, TPH and Zn in car park runoff and pollution mitigation index (PMI) values for the same pollutants for specific treatment systems.

		Pollutant		
		TSS	TPH	Zn
PI value for car park runoff		0.7	0.75	0.45
PMI values for specific treatment system	Filter strip	0.5	0.8	0.7
	Swale	0.7	0.4	0.4
	Bioretention cell	0.1	0.2	0.2
	Retention pond	0.4	0.6	0.6
Permeable paving		0.2	0.3	0.3

5.2. SUDS/BMPs pollution mitigation index (PMI) assessment.

Pollution mitigation indices (PMIs) are derived from the reported pollutant removal efficiencies for different SUDS/BMP treatment systems supplemented by an alternative more theoretical approach based on a consideration of the inherent unit operating processes (Scholes et al., 2008). This enables the generation of a ranked preference listing of SUDS/BMPs in terms of their relative performances and facilitates the generation of the PMI values given in Table 5 when integrated with the monitored removal efficiencies for car park runoff. The adopted scaling range (between 0 and 1) is qualitative with a lower index value indicating a better treatment performance. The selected treatment systems identified in Table 5 have all been used to treat car park runoff with the PMI values indicating that bioretention cells have the highest relative capability to remove all three pollutants.

5.3 Overall site pollution index (SPI).

An overall site water quality impact assessment can be determined by combining the PI and PMI indices whilst taking into account the flow paths followed by pollutants through the individual SUDS/BMP devices, arranged in series, to derive an individual land use area pollution index (LUPI):

$$\text{LUPI} = \text{LUST} \times \text{PI} \times [\text{PMI}_{\text{SUDS1}} \times \text{PMI}_{\text{SUDS2}} \times \text{PMI}_{\text{SUDS3}} \dots \dots \dots \text{n}] \quad (7)$$

where LUST is the land use surface type area, PI is a specific pollutant index for that surface type and PMI_{SUDS} refers to the pollutant mitigation index for each SUDS/BMP device proposed either individually or as part of a treatment train approach. Where different land use areas exist, the overall site pollution index (SPI) can be derived by summing the GIS-area weighted LUPI values and dividing by the total site area. Although there are a number of working assumptions (Ellis et al., 2012) it is considered that the proposed methodological approach provides screening guidance on the residual water quality risk following the selection and installation of SUDS/BMPs facilities. The extent of risk exposure can be derived by comparing the calculated SPI index with a recognised value for receiving water quality and ecological status (Ellis et al., 2012) as shown in Table S4 of the supplementary material. Comparison of the SPI categories in Table S4 with the PI values in Table 5 indicate that highly polluted discharges ($\text{SPI} \geq 0.7$; RE5) would be expected for TSS and TPH discharged directly, without treatment, from car park surfaces with Zn being capable of contributing substantially to the poor quality of a receiving water.

5.4. Application of the SPI approach.

Two scenarios are described to illustrate how the SPI approach can be applied to assessing the potential impact of car park runoff. The first scenario compares the benefits which can be gained by controlling the discharges of TSS, TPH and Zn from a car park surface using SUDS/BMPs either independently or as part of different treatment trains. The second scenario considers the different land uses which may be associated with and contribute drainage to a car park site (e.g. ground level impermeable surfaces, roof surfaces and surrounding green spaces) and assesses how the receiving water may be protected for the same three pollutants.

5.4.1 Car park surface runoff treatment

The predicted differences in the receiving water benefits achievable by the application of different treatment options to car park runoff are illustrated in Table 6. Without treatment, runoff quality for all three pollutants approaches the lowest ecological quality as expressed by the river ecosystem (RE) classification. Of the three individual treatment systems assessed, bioretention cells are predicted to provide the highest level of treatment for TSS, TPH and Zn followed by permeable paving and filter strips. However, the latter only provides a modest relative improvement in ecological status with respect to all three pollutants and would result in the discharge of low quality water. TPH can be seen to be the most resistant pollutant to removal for all three types of SUDS/BMPs.

The performances of two different examples of treatment trains are also assessed in Table 6. A three component system in which the car park runoff is initially drained across a filter strip into a grassed swale and then delivered to a retention pond is predicted to provide good treatment for TSS and Zn but to be less effective for TPH. A two component treatment train composed of permeable paving linked to bioretention cell(s) is capable of providing good removal of all three pollutants consistently demonstrating the ability to produce an effluent equivalent to the most desirable RE classification.

5.4.2. Hypothetical car park site

The hypothetical car park serves a shopping centre and covers an area of 5.55 ha subdivided into the following drainage influencing components:

- Car park surface (3.5 ha)
- Building roof surfaces (1.5 ha)
- Goods delivery area (0.2 ha)
- Petrol filling station (0.15 ha)
- Surrounding green space (0.2 ha)

The PI values quoted in Table 7 for the car park and the filling station are identical for TSS and Zn but have been increased for TPH in the latter to represent the increased possibility of fuel spillage occurring in this area. The PI values for all three pollutants for the delivery area have been slightly elevated in comparison with those for the car park to account for the heavier vehicles using this area and the different mode of activity. The calculated LUPI values are for a basic treatment scenario in which the only installed runoff treatment systems are a permeable pavement structure throughout the car parking area, green roofs for all buildings and a petrol interceptor specifically for the drainage deriving from the filling station forecourt. No treatment is provided for the goods delivery area or for the green spaces which surround the car park site. The relative magnitudes of the LUPI values indicate that the major contributors to the discharges of all three pollutants are the car park surface and the roof areas

followed by the delivery area with the filling station forecourt and the surrounding green spaces posing a much decreased impact (Table 7).

Table 6. Predictions and comparisons of the potential receiving water quality benefits achievable as a result of treating car park runoff with either individual SUDS or with SUDS treatment trains.

Treatment type	TSS		TPH		Zn	
	SPI value	RE equivalent	SPI value	RE equivalent	SPI value	RE equivalent
None	0.7	4/5	0.75	5	0.45	4
Filter strip	0.35 ^a	3	0.6	4	0.32	3
Bioretention cell	0.07	1	0.15	2	0.09	1
Permeable paving	0.14	2	0.23	3	0.27	3
Filter strip + swale + retention pond	0.10 ^b	1/2	0.14	2	0.08	1
Permeable paving + bioretention cell	0.01	1	0.02	1	0.05	1

^a Product of PI value for TSS and PMI value for TSS when treated using filter strip

^b Product of PI value for TSS and PMI values for TSS when treated using a combination of filter strip, swale and retention pond

In order to further reduce the overall SPI values for TSS, TPH and Zn and improve the quality of the site runoff it would be appropriate to target the car park and roof areas for further treatment. The impact of doing this is illustrated in Figure 3 through the following modifications to the basic treatment scenario:

- Treatment scenario A: Car park runoff leaving the permeable paving system is collected using an impermeable membrane and passed to a bioretention cell for further treatment; the data in Table 6 indicate that this represents an effective treatment train combination for runoff from car park surfaces .
- Treatment scenario B: The effluent leaving the green roof is disconnected from the separate sewer system and is also directed to a bioretention cell, for further treatment, before being discharged off-site.
- Treatment scenario C: Both of the described additional treatments for car park and roof surface runoff are put into place.

The basic treatment scenario is predicted to achieve an allocated RE3 category for all three pollutants in the discharged water from the hypothetical car park site which, without further dilution by clean water, would pose a threat to the continued existence of many sensitive

Table 7. Calculation of the SPI values for TSS, TPH and Zn for the five separate areas of a hypothetical car park for which the basic treatment scenario consists of a permeable pavement structure for the car parking area, a green roof for all buildings and a petrol interceptor for the filling station forecourt.

	PI values			PMI values			LUPI/SPI values		
	TSS	TPH	Zn	TSS	TPH	Zn	TSS	TPH	Zn
Car park surface ^a	0.7	0.75	0.45	0.2	0.3	0.6	0.49	0.79	0.95
Roof areas ^b	0.3	0.2	0.5	0.85	0.9	0.8	0.38	0.27	0.6
Delivery area	0.8	0.8	0.5	-	-	-	0.16	0.16	0.10
Filling station forecourt ^c	0.7	0.9	0.45	0.9	0.1	0.9	0.09	0.01	0.06
Green space	0.2	0.05	0.05	-	-	-	0.05	0.01	0.01
LUPI sum							1.17	1.24	1.72
SPI for site							0.21	0.22	0.31

^a PMI values for car park surface are as discussed in Section 5.4.1

^b PMI values for roof areas are as recommended by Ellis et al. (2012)

^c PMI values for filling station forecourt are based on the ability of petrol interceptors to target the removal of petrol, oil and diesel

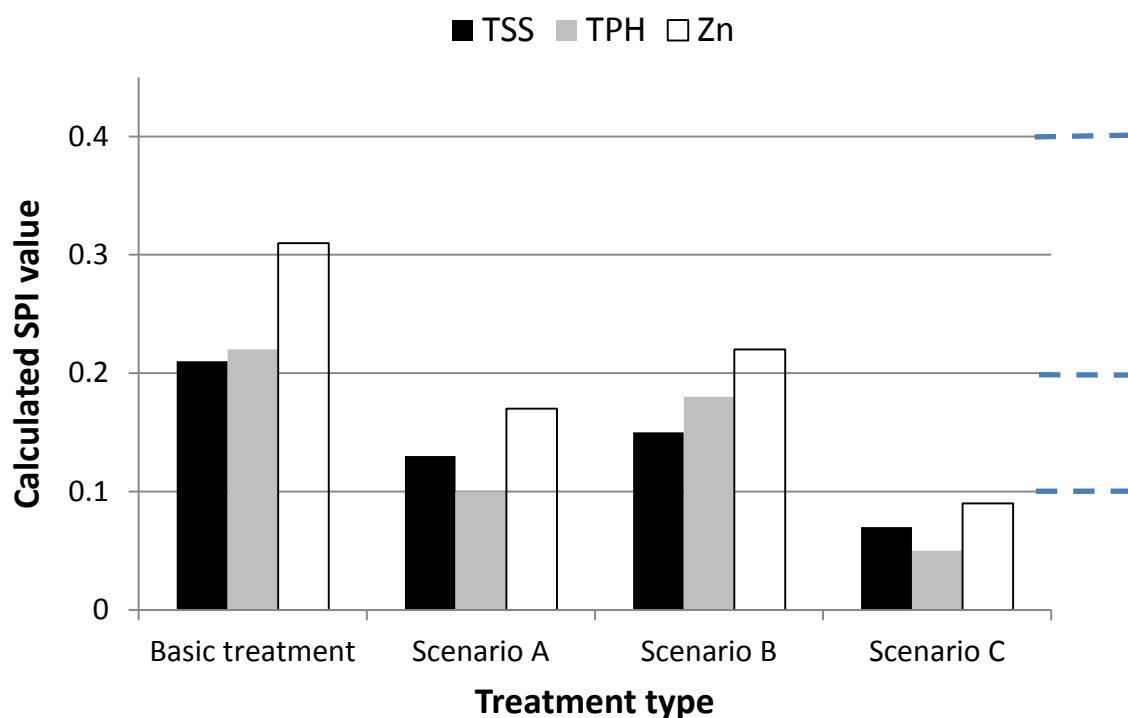


Figure 3. Predicted drainage water quality (defined by the River Ecosystem Classification) deriving from a hypothetical car park site for four different treatment scenarios.

species in a receiving water. Further treatment of the roof runoff (Treatment scenario B) would improve the situation regarding TSS and TPH (RE2) but still leave concerns over the polluting potential of Zn. The predictions indicate that vegetative treatment in addition to permeable paving (Treatment scenario A) would considerably improve the quality of car park runoff for all three pollutants with TPH now bordering on the highest water quality. The preferential option for the hypothetical car park site is shown to be double treatment train options for both car park and roof runoff (Treatment scenario C) with the highest biological water quality (RE1) being attainable for all three pollutants. The evidence is less decisive for Zn and therefore for this pollutant it may be desirable to introduce treatment for the goods delivery area. However, the use of permeable paving would not be appropriate given the repeated usage of this area by heavy goods vehicles. An alternative would be to direct the runoff to treatment in a bioretention cell which would lead to a prediction of an overall SPI for Zn of 0.05. There would also be a further reduction in the effluent levels of TSS and TPH and the overall result would be comparable with discharged water of a high quality which is able to support a high biological diversity even in the absence of any further dilution on reaching a receiving water.

CONCLUSIONS

This review has highlighted the existence of a number of research gaps which need to be addressed as part of a multi-disciplinary research programme to further establish our understanding of the processes of deposition, mobilisation and fate of diffuse pollutant loads generated within car park environments. Although several models have been developed to predict pollutant build-up and wash-off processes from impermeable surfaces, there is only limited monitoring data identifying how these processes apply to car park surfaces. Further knowledge of wash-off processes is particularly important given the potential influence of climate change induced shifts in rainfall intensity, frequency and duration patterns. Traffic patterns and driving modes within car parks are unique and the roles of frequent stopping, turning and reduced speeds on the quantity and behaviour of emitted particles and associated pollutants needs to be fully evaluated. Ideally, it would be beneficial if a car park runoff database could be developed based on standardised approaches for field sampling and analysis to enable reliable comparisons to be made between different car park sites. In addition to extending the current availability of pollutant runoff data, the database would incorporate, and ideally extend the information on the performances of stormwater SUDS/BMPs and proprietary products with regard to the mitigation of pollutant loads derived from car park surfaces.

Since it is primarily the rainfall characteristics which influence surface pollutant mobilisation, one solution for reducing the runoff from car parks would appear to be the use of on-site interception facilities, such as green roofs and/or rainwater harvesting to retain a rainfall depth (e.g. the first 5 mm). However, this will only be effective for the more frequent (e.g. 1 in 1 year) storms and will not ameliorate a prolonged build-up of surface contamination which is subsequently efficiently removed by more intense storm events. This is particularly true of the contaminated fine particles which will not have been removed by surface sweeping practices. Therefore, even if interception is practiced, there will be a need for additional treatment using SUDS/BMPs to ensure mitigation of the polluted discharges arising from car parks.

The reviewed data indicates that a range of both stormwater SUDS/BMPs and proprietary products are capable of removing the pollutants associated with car park runoff, although

with differing degrees of efficiency. To ensure that optimal performance is retained it is essential that an appropriate operation and maintenance strategy is implemented and effectively resourced. Regular surface cleaning has been shown to be important for car parks but needs to be supplemented by the use of SUDS/BMPs to ensure that the highly contaminated fine particles are efficiently removed. The site constraints associated with car parks are an important consideration as well as the economic requirements of the operator in terms of balancing the need to maximise the number of parking spaces against the space that can be allocated to treatment. Although some SUDS/BMPs such as permeable paving do not impose an additional space burden others such as swales, filter strips, bioretention cells and ponds will require extra space. Therefore, it is essential that when used in car park situations, these SUDS/BMPs are carefully selected and their size optimised to achieve the required level of treatment. An added advantage of these systems is the benefits that they can bring to the design of a car park site in terms aesthetic and possibly amenity characteristics.

The application of an impact assessment procedure to the data associated with a hypothetical car park scenario allows an evaluation of the potential impacts of surface derived diffuse pollutant loads on receiving waters. The developed screening level tool, which is based on integrated scientific considerations, demonstrates the ability of SUDS/BMPs to mitigate the environmental impacts of car park runoff and represents a means of prioritising individual or combined treatment options for this purpose. However, whilst the value of such a conceptual linked-modelling approach has a clear merit, it will need to be subjected to a full field assessment before being fully utilised as a planning tool by enforcement and guidance bodies.

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SUPPLEMENTARY MATERIAL

Table S1. The calibrated pollutant build-up parameters determined by modelling a predominantly residential urban surface in Toronto, Canada.

Pollutant type	Build-up parameters			
	Maximum pollutant build-up (P_m ; g/m^2)	Residual pollutant amount (P_0 ; g/m^2)	Pollutant deposition rate (k_0 ; $g/m^2 \cdot h$)	Pollutant removal rate (k_r ; h^{-1})
TSS	27.5	2.5	0.326	0.0178
TS	197.5	30	3.34	0.0064
TKN	0.59	0.04	0.0057	0.0094
TP	0.098	0.025	0.0023	0.0127
COD	240	14.9	4.78	0.0199
Al	0.4	0.021	0.0075	0.0174
Cu	0.018	0.0015	0.0019	0.0104
Fe	1.23	0.06	0.0247	0.0196
Zn	0.041	0.0025	0.00046	0.0115

Adapted from Chen and Adams, 2006.

Table S2. An overview of the metal concentrations ($mg\ kg^{-1}$) which have been found in brake linings, brake dust and passenger car tyre treads indicating their potential contributions to car park surface dusts.

Metal	Car brake linings	Car brake dust	Passenger car tyre treads
As ^a	<2-18	<2-11	-
Cd ^a	<1-41.4	<0.06-2.6	<0.05-2.6
Cr ^a	<10-411	135-1320	<1-30
Cu ^a	11-234,000	70-39,400	1-490
Ni ^a	3.6-660	80-730	<1-50
Pb ^a	1.3-119,000	4-1,290	1-160
Sb ^a	0.07-201	4-16,900	<0.2-0.9
Zn ^a	25-188,000	120-27,300	430-9640
Cu ^b	52,100-119,000		1.8
Zn ^b	7200-28,800		10,000
Pb ^b	9,050-18700		6.3
Cr ^b	73-151		
Ni ^b	70-182		
Cd ^b			2.6

^a Thorpe and Harrison (2008)

^b Sorme (2003)

Table S3. The concentrations of selected PAHs in urban street dusts (with different traffic densities), lubricating oils, tyres, asphalt and exhaust emissions.

Source	Σ PAHs (ng g ⁻¹) ^a	Σ selected PAHs (ng g ⁻¹) ^b
Residential street	27-76	3.0-11
Heavily trafficked street	283-379	124-205
Fresh lube oil	2926	63
Used lube oil	1428	467
Asphalt	1596	420
Auto exhaust	1476	564
Tyre particles	364	225

Adapted from Mostafa et al. (2009)

^a sum of the concentrations of phenanthrene, C1-fluoranthene-pyrenes, anthracene, benz[a]anthracene, 3-methylphenanthrene, chrysene, 2-methylphenanthrene, C1- chrysenes, 9-methylphenanthrene, C2- chrysenes, 1-methylphenanthrene, C3- chrysenes, C2- phenanthrenes–anthracenes, C4- chrysenes, C3- phenanthrenes–anthracenes, benzo[b]fluoranthene, C4- phenanthrenes–anthracenes, benzo[k]fluoranthene, dibenzothiophene, benzo[e]pyrene, C1- dibenzothiophenes, benzo[a]pyrene, C2 -dibenzothiophenes, perylene, C3- dibenzothiophenes, indeno[1,2,3-cd]pyrene, fluoranthene, dibenz[ah]anthracene, pyrene and benzo[ghi]perylene

^b sum of the concentrations of pyrene, fluoranthene, benz[a]anthracene, chrysene, benzofluoranthenes, benzopyrenes, indeno[1,2,3-cd]pyrene, and benzo[ghi]perylene

Table S4. Relationships between site pollution index (SPI) and receiving water quality characteristics expressed by impact level, biological quality, ecological potential and river ecosystem classification.

Site Pollution Index (SPI)	Impact Level	Biological Quality	EU HMWB ^a ecological potential	EA ^b RE ^c class
<0.1	Negligible	High biological diversity; several species in taxa.	Very good	RE1
0.1 – 0.2	Minimal	Small reduction in pollution tolerant taxa.	Good	RE2
0.2 – 0.4	Moderate	Many sensitive species absent; rise in pollution tolerant taxa.	Moderate	RE3
0.4 – 0.7	Substantial	Sensitive taxa scarce; some pollution tolerant species in large numbers.	Poor	RE4
>0.7	Severe	Restricted to pollution tolerant species with a few taxa dominant.	Bad	RE5

^a Heavily modified water body

^b Environment Agency for England and Wales

^c River Ecosystem classification.