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Development and application of an innovative approach to predicting pollutant concentrations in highway runoff

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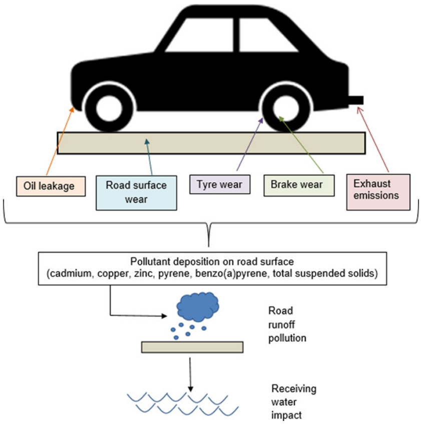
HIGHLIGHTS

- Novel combination of vehicle emission factors to predict deposition loadings
- Site-specific highway runoff concentrations predicted
- Switch to electric vehicles has a limited reduction for metals and suspended solids.
- Outputs can inform decisions on where to install highway runoff treatment systems.

GRAPHICAL ABSTRACT



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ABSTRACT

Recognising the challenges and limitations of current methodologies to predict highway runoff concentrations, this paper presents a novel approach based on the derivation of pollutant emission factors for twelve different types of vehicle. Published emission factor data and properties of differing vehicles types are combined with annual average daily traffic volume (AADT), highway characteristics and rainfall data to determine the pollutant distributions associated with differing highway and traffic types. In this paper, the method is applied to 126 sections of highway in the Greater London Borough of Enfield (United Kingdom; UK) and results are comparable with values reported in the literature. The approach is used to identify the level of AADT predicted to result in an exceedance of environmental quality standards (EQS), with results suggesting that runoff from highways experiencing AADT values as low as 5000 may require treatment prior to discharge to receiving waters. Future scenario analyses indicate that the impact of progressively replacing petrol and diesel vehicles with electric vehicles will have negligible impact on concentrations of zinc (Zn), copper (Cu), cadmium (Cd) and total suspended solids discharging from highway environments. The approach enables identification and ranking of urban highways in terms of their pollution runoff potential and provides an important support to users in prioritising locations for the installation of sustainable drainage options in order to protect receiving water environments.

1. Introduction

Predicting pollutant loads from highway runoff has traditionally involved pollutant probability distribution analysis of monitored storm sampling data, as typified in the US national stormwater database (Kerri et al.,

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1985; Kim et al., 2005; Helmreich et al., 2010). Similar approaches have been applied by many other national organisations and agencies e.g. the UK (Moy et al., 2003) and France (Legret, 2001). Statistical analyses have been used to determine concentrations and loads in terms of assumed independent variables such as traffic volume, rainfall depth and intensity, antecedent dry days, catchment area, runoff coefficients etc. However, this approach is very dependent on the pollutant distributional assumptions (Hall et al., 1990) and is made more uncertain by the limited monitoring data on input contributions resulting from differing highway sub-sources and traffic types (Gardiner and Armstrong, 2007). It is also the case that monitoring programmes are essentially catchment based whilst urban highway discharges lack a representative and viable data collection network (Mayor of London, 2019).

The Environment Agency for England (2018) have stated there is only limited long term data on the type and nature of pollutants associated with highway runoff and their potential impacts on receiving water ecologies. However, at the same time, the report suggests that the transport sector may account for up to 13% of all urban land activities preventing waterbodies from achieving "good" ecological status. This would indicate that highway runoff is capable of generating contaminated runoff that can severely prejudice receiving water quality. This conclusion has been confirmed by many European studies conducted on highway runoff quality e.g. Barbosa and Fernandes, 2021.

Other modelling approaches such as those incorporating the effects of particle size and surface wash off/scour have also been widely developed to determine highway discharge pollutant concentrations and loads (Ellis and Revitt, 1982; Egodawatta and Goonetilleke, 2008; Zhao et al., 2014). Such approaches may be relatively simple and practical, but they also present considerable methodological uncertainties associated with estimation and validation of interfering variables such as hydraulic/surface roughness, particle densities, solubility transforms, rainfall depths etc. In addition, none of these modelling approaches examines the specific sub-source effects resulting from vehicle type and highway dimensions/characteristics. However, whilst there is only a limited database related to the monitoring of highway runoff pollutant discharges, there is a long-standing and well-structured database related to vehicle types and numbers for the majority of UK urban areas. The official UK Department of Transport (DfT, 2021) traffic count website provides this data based on monitored highway lengths within each local authority area. In addition, accurate measurement of road/paving dimensions as well as records of highway barriers and parking areas can be obtained from sources such as Google Earth or Google Vision. Detailed rainfall time series data are also fully available over long time periods for such highly urbanised sites. It is the availability of such detailed site data for the Greater London area that provides the basis for the development of a viable and user-friendly approach to the analysis and quantification of highway traffic pollutant sub-sources.

It has long been recognised that traffic can be a primary source of pollutants within urban areas (Huber and Helmreich, 2016; Markiewicz et al., 2017), but this source typing embraces multiple vehicle associated sub-sources including tyres, brakes and body parts, emissions, oil/fuel leakages and contributions from the highway surface, signage and highway furniture. The analysis of Barbosa and Fernandes (2021) concludes that neither national nor regional typification of highway runoff quality is feasible based only on catchment and rainfall characteristics. This European review reinforces the contention stated by other reviews such as Müller et al. (2020) that traffic related parameters remain critical to the behaviour and determination of highway surface runoff pollutant discharges. More recently such sub-source types have been quantified through substance flow analysis (SFA) in an attempt to determine specific sources of the complex pollutant mixtures presented by highway runoff (Petrucci et al., 2014). Such SFA is essentially based on the derivation of emission factors (EF) for each sub-source and individual pollutants. SFA can thus provide an understanding of the relative pollutant magnitude exerted by each sub-source on the total highway outflow load.

This paper develops a generic substance flow methodology based on the derivation of pollutant EFs for twelve different traffic types to predict highway runoff concentrations within an area of Greater London (UK). The EF is estimated from published data of emission rates and source substance properties associated with differing vehicle types and expressed as substance mass per travelled vehicle-kilometre (e.g. μg/vkm). Total substance flow is then determined by combining the monitored highway area and average daily traffic volume (AADT) with local rainfall data.

The primary objective of the paper is to demonstrate the basis and approach adopted for the development and application of the SFA methodology. The modelling approach enables EFs to be worked up from monitored observations and associated datasets for the complex suite of vehicular sub-sources utilising a simple Excel recoding template. The screening tool has been developed with the specific intention of identifying and ranking urban highways in terms of their pollution runoff potential and to determine the pollutant distributions associated with differing highway and traffic types. The methodological predictions also enable the identification of pollutant "hotspot" distributions that can help support better targeted decision-making management for highway runoff treatment and control, particularly in terms of identifying critical surface water outflow discharges which are likely to prejudice receiving water and ecological standards. The paper objectives therefore support longer-term sustainability for highway runoff management.

2. Methodology

The three stages of the developed methodology for the estimation of highway pollutant loads and runoff concentrations are illustrated in Fig. 1. Stage 1 derives the daily number of vehicle kilometres travelled (vkm) using the road characteristics (length, vehicle types and annual average daily traffic flows; AADT) and combines this with pollutant source strengths (defined by different traffic source based emission factors for specific pollutants; Stage 2) to obtain a highway surface pollutant loading. This enables the derivation of pollutant emission loads for 12 different vehicle types, which after adjustment for deposition capability are converted into discharged runoff concentrations using the predicted drainage volumes derived from local rainfall data and the impervious area of the road section under consideration (Stage 3). The different vehicle types which have been included in this analysis are petrol, diesel and electric passenger cars, petrol, diesel and electric light duty vehicles, diesel rigid axle and articulated heavy goods vehicles, motorcycles, taxis, buses and coaches.

In addition to discharged runoff, deposited vehicle derived pollutants can be removed from road surfaces by splash/spray and dispersion (Folkeson et al., 2009). Studies of the distribution of pollutants associated with the different removal pathways have determined values for transport in road runoff of 36% (Steiner et al., 2006), 35% (POLMIT, 2002) and 8–54% (Best et al., 2002). Based on these reported values a default figure of 35% has been selected to describe the proportion of pollutants removed from the road surface in runoff during rainfall events.

The monthly average road runoff pollutant concentrations are computed using an Excel spreadsheet designed to facilitate the input of data relating to the highway characteristics, vehicle types and volumes, pollutant emission factors and monthly rainfall. The pollutant generating components considered are exhaust emissions, brake wear, tyre wear, road surface wear and oil leakage and for each one established emission factors have been converted to pollutant emission factors. In addition to particulate emissions, the pollutants described in this paper are three metals (Cd, Cu and Zn) and two polyaromatic hydrocarbons (PAHs; pyrene and benzo(*a*)pyrene). The determinations of these emission factors for the 12 different vehicle categories are fully described in Section 2.1 (for exhaust emissions), Section 2.2 (for brake wear emissions), Section 2.3 (for tyre wear emissions), Section 2.4 (for road surface wear) and Section 2.5 (for oil leakage emissions).

The selection of one month as the time period for pollutant deposition on the road surface and the calculation of pollutant runoff concentration is based on an established pattern of pollutant accumulation on road surfaces. Typically, mathematical models for predicting the temporal pollutant build-up processes on urban surfaces identify an initial high accumulation

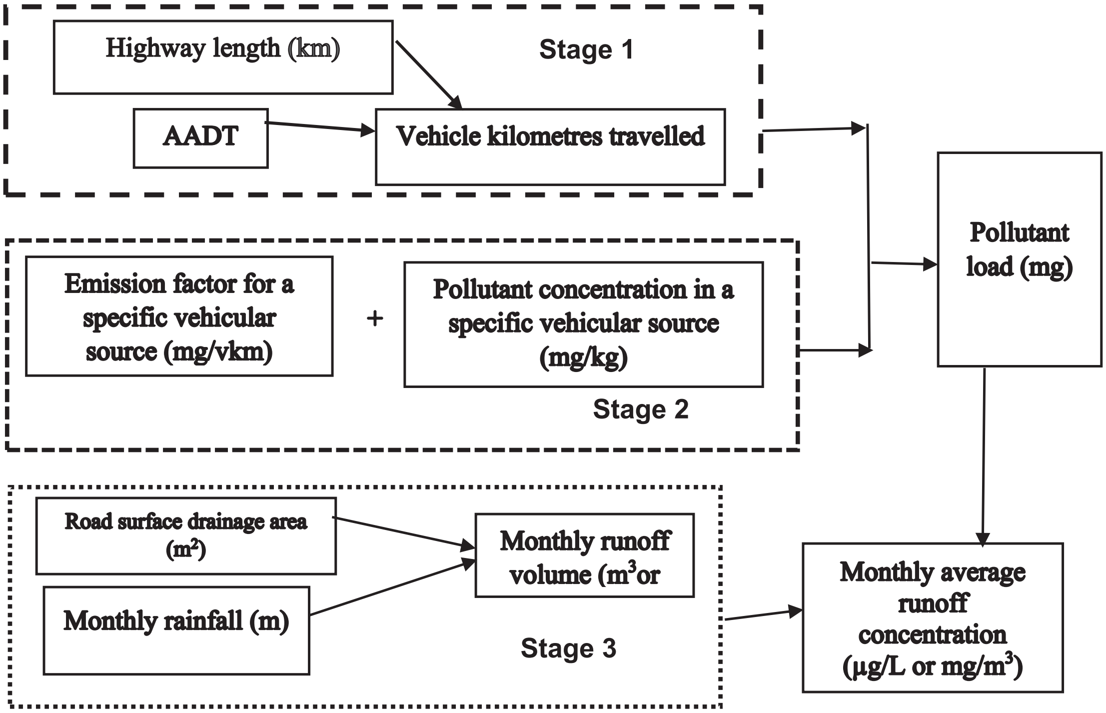

Fig. 1. Outline of the methodology for the estimation of highway runoff pollutant concentrations.

rate which then decreases gradually and eventually reaches an almost constant value after a time in excess of 14 days (e.g. Deletic et al., 1997; Egodawatta et al., 2013). In the UK, a prolonged dry period is unlikely to exceed one month and therefore this has been chosen as the time period during which the maximum accumulation of pollutants on a road surface occurs giving rise to typical maximum monthly average runoff concentration. This concentration will also be dependent on the applied runoff coefficient, which for asphalt surfaced roads is 0.75–0.95 with values towards the higher end of the range for good quality surfaces (e.g. Thomas, 2017). A value of 0.90 has been selected to represent the 'worst case scenario' in terms of the volume of runoff produced.

2.1. Exhaust emissions

The majority of vehicles currently in the UK national vehicle fleet burn hydrocarbon based fuels (e.g. petrol, diesel, compressed natural gas and liquid petroleum gas) resulting in polluted exhaust emissions. These are subject to air quality emission standards particularly targeting gaseous emissions (carbon monoxide, nitrogen oxides, hydrocarbons, sulphur oxides) and particulate matter (PM_{10}). There is less information available of direct relevance to stormwater quality issues, through the emissions of metals and complex organic compounds (such as PAHs) (Kennedy et al., 2002).

2.1.1. Total particulate emission factors

Emissions from highway vehicles depend on a number of influencing factors including the age and composition of the fleet, the size and weight of the vehicle, the fitted abatement technologies to reduce emissions and the type and quality of fuel used. The available data on the emissions of particulates from exhaust systems addresses the different sizes of particulate matter from <10 μm (PM_{10}) down to <2.5 μm ($PM_{2.5}$) (e.g. Thorpe et al., 2007). The larger PM_{10} fraction has been selected for use in this study and the emission factors representative of the UK fleet in 2017 under urban driving conditions (NAEI, 2019) are reported in Table 1. They refer to tailpipe emissions from engines at their normal operating temperatures for the identified vehicles over a range of different driving cycles. Particles in the PM_{10} range are only expected to remain airborne for short periods of time and to be predominantly deposited some distance downwind of the vehicle emission source. Given the predicted preference for vehicle emitted particles to remain airborne, a conservative value of 10% has been selected for the amount deposited in the immediate road surface environment. This is consistent with the results from an investigation during the widespread use of leaded petrol which found that 90% of the exhaust Pb emissions from fast moving vehicles were deposited away from the immediate vicinity of a highway (Harrison et al., 1985).

2.1.2. Metal emission factors

Pulles et al. (2012) have published recommended metal emission factors associated with the combustion of transport fuels within Europe based on both their own tailpipe measurements as well as a comprehensive review of previously reported values. The reported results, expressed as μg per kg of petrol burnt, are 0.29 for Cd, 4.0 for Cu and 36.0 for Zn. The corresponding values for diesel fuel are <0.05 for Cd, 7.3 for Cu and 19.0 for Zn. These values can be converted into emissions per vehicle-kilometre (vkm) using the density of the fuel (petrol, 0.74 kg/L; diesel, 0.83 kg/L) and the fuel consumption for each category of vehicle according to the mode of driving. Average fuel consumption data for different types of vehicles under urban driving conditions are presented in Table 1 together with the calculated emission factors for Zn, Cu and Cd.

2.1.3. PAH emission factors

The emission factors for both total and individual PAHs have been widely reported (e.g. Ravindra et al., 2008; Shen et al., 2011; EMEP/EEA, 2020) for petrol (cars and light duty vehicles) and diesel engine vehicles (cars, light duty vehicles and heavy goods vehicles). The PAH exhaust emission factors in the European Environment Agency 2019 emission inventory guidebook (EMEP/EEA, 2020) are most relevant as the methodology uses vehicle flow data relating to 2019. For petrol engines the guidebook reports PAH emission factors for the use of both conventional and closed - loop catalysts. However, the vehicle flow data does not provide this distinction and therefore average values have been applied for pyrene and benzo(*a*)pyrene are shown in Table 1. For diesel engine light duty vehicles, the 2019 EMEP/EEA emission inventory provides values for direct and indirect injection but it has been necessary to determine mid-point values which are presented in

Table 1
Fuel consumption and exhaust emission factors for different types of vehicles.

Vehicle type	Average fuel consumption (L/km)	Emission factors					
		PM_{10}	Zn	Cu	Cd	Pyrene	Benzo(*a*)pyrene
		(mg/vkm)	(μg/vkm)		(ng/vkm)	(μg/vkm)	
Petrol passenger cars	0.074	1.000	1.971	0.219	15.88	3.79	0.40
Diesel passenger cars	0.057	10.00	0.899	0.345	2.37	25.63	1.74
Petrol light duty vehicles	0.128	1.000	3.410	0.379	27.47	3.79	0.40
Diesel light duty vehicles	0.102	13.00	1.609	0.618	4.233	25.63	1.74
Heavy goods vehicles (rigid axle)	0.309	25.00	4.873	1.872	12.82	31.59	0.90
Heavy goods vehicles (articulated)	0.317	20.00	4.999	1.921	13.16	31.59	0.90
Motorcycles	0.0355	8.000	0.946	0.105	7.62	3.79	0.40
Taxis	0.0633	10.00	0.998	0.384	2.63	25.63	1.74
Buses	0.475	43.00	7.491	2.878	19.72	31.59	0.90
Coaches	0.260	43.00	4.100	1.575	10.79	31.59	0.90

Table 1 for pyrene and benzo(*a*)pyrene. Heavy goods vehicles (rigid axle and articulated) are grouped with buses and coaches in terms of their diesel engine emission factors (Table 1).

2.2. Brake wear emissions

The different types of brake lining materials include non-asbestos organic, low-metallic, semi-metallic and metallic with the first three types having the most common applications (Kennedy et al., 2002). Braking is considered to be a major source of non-exhaust traffic-related emissions particularly in intra-urban locations where forced deceleration regularly occurs. Because of their size distributions, brake wear particles are either released to the atmosphere or deposited on the road surface with only small amounts being retained on the vehicle. It is estimated that approximately 50% of the generated brake wear particles become airborne (Garg et al., 2000; Kukutschová et al., 2011) with the majority of the remainder being deposited on the road surface.

2.2.1. Total particulate emission factors

The majority of published research work has considered the impact of brake wear on atmospheric pollution and therefore has concentrated on PM_{10} emissions and associated emission factors (e.g. Garg et al., 2000; Abu-Allaban et al., 2003; Grigoratos and Martini, 2015). However, when assessing the amount of brake wear material which can potentially be deposited on the road surface it is necessary to consider the full particle size range. During the lifetime usage of a braking system, typically around 80% of the friction material can be expected to have worn away, enabling total wear amounts to be estimated. The total particulate brake wear emission factors used in this study (Table 2) are derived from those reported by EMEP/EEA (2020) for a range of different vehicle types and values reported by Klein et al. (2017) for urban roads in the Netherlands. Where there is variability in the data, mid-point values have been selected.

Table 2
Brake wear emission factors for different types of vehicles.

Vehicle type	Emission factors					
	Total particulates	Cu	Zn	Cd	Pyrene	Benzo(*a*) pyrene
	(mg/vkm)	(μg/vkm)		(ng/vkm)		
Passenger cars (petrol, diesel, electric); taxis	14	140	105	36.4	15.4	10.4
Light duty vehicles (petrol, diesel, electric)	26	260	195	67.6	28.6	19.2
Heavy goods vehicles (rigid axle, articulated)	55	550	413	143	60.5	40.7
Coaches	52	520	390	135	57.2	38.5
Buses	75	750	563	195	82.5	55.5
Motorcycles	8	80	60	20.8	8.8	5.9

2.2.2. Metal emission factors

The emission factors for metals have been calculated from the total particulate emission factors and the metal concentrations in brake linings and brake dusts. However, the metallic compositions of brake linings can vary markedly (depending on manufacturer and lining type) with ranges often covering several orders of magnitude (Thorpe and Harrison, 2008; Boulter, 2006). Similar variations have been reported for metals in brake dust samples (Gadd and Kennedy, 2000; Garg et al., 2000). From these values, Cd, Cu and Zn concentrations in brake wear emissions of 2.6 mg/kg, 10,000 mg/kg and 7500 mg/kg, respectively, have been deduced for all vehicles and used to derive the metal emission factors shown in Table 2.

2.2.3. PAH emission factors

There is limited information available on the composition of brake linings with respect to particular organic compounds (Kennedy et al., 2002) but it is believed that the organic composition of brake wear debris differs significantly from that of the original lining material (Kukutschová et al., 2011). The build-up of heat during brake application is considered to be responsible for the generation of trace amounts of PAHs. Rogge et al. (1993) have detected n-alkanes and a number of PAHs and substituted PAHs in brake lining dusts with a total PAH concentration of 16.16 mg/kg. Although there are no differentiating data for the brake wear produced by different vehicle types, the range of values reported for pyrene and benzo(*a*)pyrene are 1.1 to 3.5 and 0.74 to 3.7 mg/kg (Rogge et al., 1993; Chlopek et al., 2016). The concentration values at the lower end of these ranges are considered appropriate for the calculation of the emission factors shown in Table 2 as these are consistent with those reported by Siopi (2015) for pyrene (9.7–92.4 ng/vkm) and benzo(*a*)pyrene (6.5–62.2 ng/vkm).

2.3. Tyre wear emissions

The amount of tyre wear is dependent on tyre characteristics, vehicle characteristics, road surface characteristics and vehicle operation. The consequences are that heavy goods vehicles have been reported to emit up to ten times higher amounts of tyre wear particles compared to light duty vehicles and passenger cars and that concrete surfaces produce lower wear emissions compared to other surface types (Amato et al., 2011; Denier van der Gon et al., 2012). Smolders and Degryse (2002) have determined the mean diameter of roadside tyre debris to be 65 μm for cars and 80 μm for heavy goods vehicles indicating the preference for this material to be deposited on the road surface rather than remaining airborne. Hence most of the material emitted during the tyre wear process can be expected to be deposited fairly rapidly with the amounts varying between 80% and 95% (e.g. Panko et al., 2013; Klein et al., 2017; EMEP/EEA, 2020). In this study, a default value of 85% has been used for the amount of tyre wear deposited on the road surface.

Table 3

Tyre wear emission factors for different types of vehicles.

Vehicle type	Emission factors					
	Total particulates	Zn	Cu	Cd	Pyrene	Benzo(a) pyrene
	(mg/vkm)	(μg/vkm)	(ng/vkm)			
Passenger cars (petrol, diesel, electric); taxis	100	550	280	130	1390	235
Light duty vehicles (petrol, diesel, electric)	159	382	318	95.4	2210	374
Heavy goods vehicles (rigid axle, articulated)	850	9350	1530	357	4165	1233
Coaches	250	1375	500	150	1225	363
Buses	415	4565	747	174	2034	602
Motorcycles	60	330	168	78	834	141

2.3.1. Total particulate emission factors

A new tyre fitted to an average European car typically loses between 1.0 and 1.5 kg of its original weight to the roadside environment during its service lifetime of around 3 years and 50,000–60,000 km (Environment Agency, 1998). Therefore, a typical 'average' tyre wear factor would be of the order of 100 mg/vkm for a four wheel passenger car (Boulter, 2006). This value has been selected for use in this study for all types of passenger cars and taxis because of its comparability to other reported passenger car tyre wear values of 74 mg/vkm (Luhana et al., 2004), 132 mg/vkm (Klein et al., 2017) and a range of 4 to 500 mg/vkm (EMEP/EEA, 2020).

The higher expected wear values for heavy goods vehicle tyres are typified by reported values in the range of 136–1403 mg/vkm (EMEP/EEA, 2020) and a value of 850 mg/vkm (Klein et al., 2017). Careful consideration of the distribution in this reported range, which exemplifies the dependence of tyre wear on the vehicle configuration, such as the number of axles and the load, has led to the choice of 850 mg/vkm for both rigid axle and articulated heavy goods vehicles (Table 3). Klein et al. (2017) also report tyre wear emission factors for buses (415 mg/vkm), light duty vehicles (159 mg/vkm) and motorcycles (60 mg/vkm). These values have been selected for use in this study and are shown in Table 3. Coaches are considered to be interim in size between light duty vehicle and buses and therefore a value of 250 mg/vkm has been assigned for tyre wear.

2.3.2. Metal emission factors

The composition of tyres is dominated by organic compounds and although formulations vary according to required performance standards (e.g. Environment Agency, 1998; Gadd and Kennedy, 2000). The use of zinc oxide as an accelerator in the vulcanisation process leads to significant quantities of Zn in tyre tread and tyre wear debris. The ranges and averages for the Zn content of tyres have been reported for passenger cars (430–15,500; 7400 mg/kg), light duty vehicles (1190–3640; 2400 mg/kg) and heavy goods vehicles (13,800–18,300; 16,050 mg/kg) (Boulter, 2006; Thorpe and Harrison, 2008). However, Panko et al. (2013) have noted that the heat and friction generated during the interaction of tyres with the road surface alters the chemical composition and therefore, when considering road surface deposits, it is important to consider the zinc levels in tyre debris. Kreider et al. (2010) report Zn levels for both road collected and laboratory generated tyre wear particles of between 3000 and 4000 mg/kg suggesting that there is a reduction in Zn concentrations in tyre wear debris compared to tyres themselves. This has been taken into account in assessing the Zn concentration (5500 mg/kg) used in this study for car tyre wear debris. This value has also been used for taxis, coaches and motorcycles. Similarly, the Zn concentrations in tyre wear debris from heavy goods vehicles (and also buses) have been reduced to 11,000 mg/kg whereas the lower value for light duty vehicles (and coaches) (2400 mg/kg) has been left unchanged.

The concentrations of Cu and Cd deriving from tyre wear are generally several orders of magnitude lower than those for Zn. However the reported concentration ranges are rather wide, reflecting the different manufacturing techniques. The reported values for different vehicle types are limited but include passenger cars (Cd: <0.05–2.6 mg/kg, average 1.3 mg/kg; Cu: 0.4 to 5.5 mg/kg, average 2.8 mg/kg), light duty vehicles (Cd: <0.05–0.1 mg/kg, average 0.6 mg/kg; Cu: 1 to 3 mg/kg, average 2.0 mg/kg) and heavy goods vehicles (Cd: 0.28–0.56 mg/kg, average 0.42 mg/kg; Cu: 1–2.5 mg/kg, average 1.8 mg/kg) (Boulter, 2006; Thorpe and Harrison, 2008). The average values quoted here have been used to determine the emission factors for different vehicle types as identified in Table 3.

2.3.3. PAH emission factors

PAHs in tyres are found in the highly aromatic oils (extender oils) that are used during the manufacturing phase to make the rubber easier to work, to soften the tread and to improve tyre grip on wet roads. The concentrations of PAHs in tyre wear particles, derived from passenger cars and heavy goods vehicles, have been identified in several reviews (e.g. ten Broeke et al., 2008; Boulter, 2006; Kennedy et al., 2002). The data for benzo(*a*)pyrene and pyrene has been extracted from these reviews with the emphasis being placed on the most recent data available and neglecting elevated values. This is necessary because since 2010 the use of extender oils has been banned in all new tyres produced in Europe as well as those imported (EU, 2005). The selected values for the concentrations of benzo(*a*)pyrene and pyrene in tyre wear for cars (2.35 mg/kg and 13.9 mg/kg, respectively) and heavy goods vehicles (1.45 mg/kg and 4.9 mg/kg, respectively) have been used to determine the emission factors shown in Table 3. All types of passenger cars and light duty vehicles are grouped together and heavy goods vehicles also incorporate buses and coaches. Comparisons of the calculated emission factors with the ranges reported by Siopi (2015) for benzo(*a*)pyrene (38–12,000 ng/vkm) and pyrene (317–132,000 ng/vkm) show that they are consistently at the lower end of these ranges indicating consistency with the recently introduced legislation regarding the banning of highly aromatic extender oils.

2.4. Road surface wear emissions

The majority of road surfaces in the UK are asphalt based and constitute mixtures of mineral aggregates (stone material), filler and bitumen binder (5% composition). Very little of the road surface wear particles can be expected to become airborne and EMEP/EEA (2020) and Klein et al. (2017) report that 90% and 95%, respectively of emitted material is available for immediate deposition. The lower value of 90% has been applied in this study.

2.4.1. Total particulate emission factors

Although there are only a limited number of reported studies, a wide range of road surface wear factors have been reported varying from 3.8 mg/vkm (Muschack, 1990) to 440 mg/vkm (Kennedy et al., 2002). Only two reports (EMEP/EEA, 2020; Klein et al., 2017) discriminate between the impact of the different types of vehicles giving values of 150 mg/vkm and 180 mg/vkm for passenger cars, taxis and light duty vehicles, 760 mg/vkm and 922 mg/vkm for heavy goods vehicles and 840 mg/vkm for

Table 4

Road surface wear emission factors for different vehicle types.

Vehicle type	Emission factors					
	Total particulates	Zn	Cu	Cd	Pyrene	Benzo(a) pyrene
	(mg/vkm)	(μg/vkm)		(ng/vkm)		
Passenger cars (petrol, diesel, electric); taxis	165	14.65	7.36	21.45	29.95	14.93
Light duty vehicles (petrol, diesel, electric)	165	14.65	7.36	21.45	29.95	14.93
Heavy goods vehicles (rigid axle, articulated)	840	74.59	37.46	109.2	152.46	76.02
Coaches	550	48.84	24.53	71.5	99.83	49.78
Buses	840	74.59	37.46	109.2	152.46	76.02
Motorcycles	74	6.57	3.30	9.62	13.43	6.70

buses. The averages of these values, where relevant, have been used together with specifically assigned values for coaches (550 mg/vkm) and motorcycles (74 mg/vkm) (see Table 4).

2.4.2. Metal emission factors

Lindgren (1996) has reported the metal concentrations in the stone aggregate (gabbro and porphyry) and bitumen constituents of asphalt. Average stone aggregate concentrations have been calculated for Cd (0.132 mg/kg), Cu (46.2 mg/kg) and Zn (92.7 mg/kg) and combined with the low metal levels associated with the 5% bitumen in stone aggregates to produce overall values for Cd, Cu and Zn of 0.13 mg/kg, 44.6 mg/kg and 88.8 mg/kg. These values have been combined with the total particulate emission factors to produce the metal emission factors shown in Table 4.

2.4.3. PAH emission factors

The vacuum distillation processing of crude oil to produce bitumen results in considerable reductions in PAH composition. Reported total PAH concentrations in bitumen are typically within the range of 10 to 100 mg/kg (Gadd and Kennedy, 2000; Ifenna and Osuji, 2013). Within this total PAH composition, the reported pyrene and benzo(*a*)pyrene concentrations have been shown to be highly variable with ranges of 0.73–6.68 mg/kg (average 3.63 mg/kg) and 0.15–6.63 mg/kg (average 1.81 mg/kg), respectively (Gadd and Kennedy, 2000; Pinheiro et al., 2009; Ifenna and Osuji, 2013). Since bitumen only represents 5% of the composition of asphalt aggregates, the average pyrene and benzo(*a*)pyrene concentrations in road surface wear particles are predicted to be 1.815×10^{-4} μg/mg and 0.905×10^{-4} μg/mg. These values have been used to calculate the road surface wear emission factors shown in Table 4 for different vehicle classes. Previously published emission factor ranges arising from asphalt surface wear are 8.7–57.6 ng/vkm for pyrene and 15.0–64.0 ng/vkm for benzo(*a*)pyrene (Siopi, 2015). These are in good agreement with the values reported in Table 4.

2.5. Oil leakage emissions

The principle contaminants in used motor lubricating oil are monocyclic aromatics, PAHs and chlorinated organics with metallic salts added to prevent the build-up of engine deposits, to maintain engine cleanliness and to act as anti-wear agents (Zieba-Palus and Kocielniak, 2000). The majority of oil leaking from the engine sump is expected to be deposited directly on to the road surface. A small amount may be retained on the lower parts of the engine with some being transferred to the chassis through turbulence effects beneath vehicles. In addition, there may be some volatilisation of the lost oil due to the high operating temperatures in the engine compartment. However, only in the region of 10% of leaked oil is expected to be vaporised or retained on the vehicle with 90% being deposited on the road surface.

2.5.1. Total particulate emission factors

Oil loss rates due to leakage of 2.8 mL/1000 km for cars and light duty vehicles and 2.1 mL/1000 km for heavy goods vehicles and buses have been reported (Kennedy et al., 2002). Given an average value for the specific gravity of motor oil of 0.89 kg/L, these correspond to oil leakage values of 2.49 mg/vkm for cars and light duty vehicles and 1.87 mg/vkm for heavy goods vehicles, buses and coaches. The allocated value for motorcycles is half that of cars at 1.25 mg/vkm. The total particulate oil leakage emission factors for different vehicle types (except electric vehicles) are summarised in Table 5.

2.5.2. Metal emission factors

Zieba-Palus (1998) has reported the concentrations of Zn, Cu and Cd in new and used lubricating oils (5000–6000 km). Although there are differences between different oil grades produced by different manufacturers, the objective here is to derive an average metal concentration which is representative of the total vehicle fleet. There is evidence that the concentrations of Cu and Cd increase with usage (most probably due to wear processes) whereas the higher initial Zn concentrations decrease only marginally possibly due to the breakdown of the originally introduced zinc additives. As oil leakage will occur regardless of the age of the oil, the average of the reported values for new and used oils has been derived in order to be representative of the different ages of oil in use at any particular time. The selected values for Zn, Cu and Cd which have been used to determine their emission factors (Table 5a) are 1618 mg/kg, 1.45 mg/kg and 0.72 mg/kg, respectively (Zieba-Palus, 1998; Roslan et al., 2016). The derived emission factors reported in Table 5a are comparable with the ranges reported for different types of vehicles by Kennedy et al. (2002) for Zn (2.1–2.9 μg/vkm), Cu (1.9–2.5 ng/vkm) and Cd (2.4–3.2 ng/vkm).

Table 5
Oil leakage emission factors for different vehicle types a) for particulates, Zn, Cu and Cd and b) for pyrene and benzo(*a*)pyrene.

a)

Vehicle type	Emission factors			
	Total particulates	Zn	Cu	Cd
	(mg/vkm)	(μg/vkm)	(ng/vkm)	
Passenger cars (petrol, diesel); taxis; light duty vehicles (petrol, diesel)	2.49	4.03	3.61	1.79
Heavy goods vehicles (rigid axle, articulated); coaches; buses	1.87	3.03	2.71	1.35
Motorcycles	1.25	2.02	1.81	0.90

b)

Vehicle type	Emission factors	
	Pyrene	Benzo(*a*)pyrene
	(ng/vkm)	(ng/vkm)
Petrol passenger cars and light duty vehicles	138.2	13.7
Diesel passenger cars and light duty vehicles	129.5	10.0
Heavy goods vehicles, coaches and buses (diesel)	97.2	7.5
Motorcycles (petrol)	69.4	6.3

2.5.3. PAH emission factors

Kingett Mitchell (1994) has reported the concentrations of pyrene and benzo(*a*)pyrene in used engine oil. Pyrene concentrations were 55.5 mg/kg (for petrol engines) and 52 mg/kg (for diesel engines) with comparable values for benzo(*a*)pyrene of 5.5 mg/kg and 4 mg/kg. These concentrations have been used to differentiate between the emission factors for diesel and petrol vehicles for pyrene and benzo(*a*)pyrene as shown in Table 5b. Despite used engine oils being expected to have higher PAH concentrations than newer oils, the derived values are highly comparable with the emission factor ranges for diesel and petrol vehicles reported by Kennedy et al. (2002) for pyrene (97–140 ng/vkm) and benzo(*a*)pyrene (7.0–14.0 ng/vkm).

3. Application of methodology

3.1. Site testing of predictive methodology

The developed methodology has been applied to 126 sections of highways of varying types (including motorways, major highways, link highways and local trafficked highways) in the London Borough of Enfield (northern edge of the greater London Metropolitan area, UK). Details of vehicle flows (AADT) by direction for each section of highway are available in the 2019 traffic statistics published annually by the UK Department for Transport (DfT, 2019). Data are provided for motorcycles, passenger cars and taxis, light duty vehicles, buses and coaches, and heavy goods vehicles constituting a total of 1.27 billion vehicle miles travelled on highways in Enfield in 2019. These have been disaggregated using national statistics (e.g. UK Government, 2021a; UK Government, 2021b) and local knowledge into the 12 different vehicle categories included in the methodology

Table 6

Comparison of means and ranges of predicted and measured total concentrations of selected pollutants in highway runoff.

	Predicted values		Measured values [1]		Water quality standard/ guideline value
	Mean ± SD	Range (minimum–maximum	Mean	Range (25th percentile – 75th percentile	
Total suspended solids (mg/L)	98 ± 79	2–422	148.7	67.7–327.0	25[2]
Zn (μg/L)	284 ± 279	4–1629	314.9	90.4–1096	96[3]
Cu (μg/L)	31 ± 23	1–115	66.1	22.7–195.7	28[3]
Cd (μg/L)	0.05 ± 0.04	0.001–0.183	1.61	0.88–2.92	0.25[4]
Benzo(*a*)pyrene (μg/L)	0.12 ± 0.1	0.002–0.463	0.031	2.5×10^{-5}–0.69	1×10^{-4}[4,5]
Pyrene (μg/L)	0.93 ± 0.69	0.02–3.26	1.03	<0.01–12.5	

Key: [1] Mitchell (2001); [2] European Union (EU) Freshwater Fisheries Directive (repealed; surrogate guideline value as the EU Water Framework Directive (2000) does not include an EQS for TSS; [3] total concentration derived from bioavailable national EQS; [4] total concentration derived from dissolved EQS AA (class 5); [5] EQS AA.

description (see Section 2.1). AADT ranges from 69,311–76,355 for sections of motorway, to 191–7014 for local highways.

The 2019 traffic statistics (DfT, 2019) divide the highways into identified sections with reported lengths of between 0.2 km and 8.5 km depending on highway type and traffic usage patterns. The widths of impermeable highway surfaces (including, where appropriate, pavements, hatched central reservations, lay-bys, parking bays, cycle lanes and hard shoulders) have been determined using Google maps supported by ground truthing where considered necessary. The calculated total impermeable drainage areas vary from 1150 to 142,750 m^2, which has been used to determine the average monthly rainfall volume discharged from each highway surface area. The annual rainfall depth for 2019 monitored in the grounds of Enfield Civic Centre was 687.0 mm (personal communication) giving a monthly average of 57.25 mm. The variations in the calculated runoff pollutant concentrations are discussed in the following sections together with their significance for the highway environments in the London Borough of Enfield.

Table 6 gives an overview of the mean and range of total concentrations predicted per pollutant for the 126 highways. The magnitude of the standard deviation and range indicate considerable variation between data sets as could be anticipated from the range in highway types and drainage areas involved in the study. In all cases the minimum concentrations correspond to the predictions for minor roads (AADT < 2000) and these also contribute to a slight lowering of the predicted mean value. The predicted means and ranges for runoff are compared in Table 6 with the results obtained from twelve major monitoring studies of UK main roads (Mitchell, 2001). There is good agreement between predicted and measured data for total suspended solids (TSS), Zn, Cu and pyrene. Although the predicted runoff concentrations for benzo(*a*)pyrene are within the reported measured range they tend to overestimate the mean value. However, the predicted benzo(*a*)pyrene concentration is consistent with a mean value of 0.15 μg/L reported by Crabtree et al. (2006) for the runoff from six UK highways (AADT > 15,000). Comparison of the predicted and measured data in Table 6 indicates an under-prediction in the case of Cd although runoff concentrations as low as 0.05 μg/L have been reported in a comprehensive review of the runoff from urban highway sites across six continents (Huber et al., 2016).

3.2. Receiving water quality standards

Receiving water environmental quality standards (EQS) are available for total Cd and benzo(*a*)pyrene only under the EU Priority Substances Directive (EU PSD, 2013). EQS are available for Zn and Cu at a national level in certain countries (e.g. the UK and Sweden) which relate to the bioavailable fraction ($EQS_{bioavailable}$). To enable comparison with total concentrations (the fraction predicted in this study), $EQS_{bioavailable}$ have been converted to an equivalent total concentration by referring to the literature to predict the fraction of the total concentration likely to be present in the dissolved form. The predicted dissolved concentration (together with catchment-specific data on pH, dissolved organic carbon and calcium levels) were then entered into the bioavailable prediction model BIOmet to back-calculate the total concentration that equated to the $EQS_{bioavailable}$ for the Thames river basin catchment (in which Enfield is located) assuming the receiving water provided at least an eight-fold dilution. Currently there is no EQS for suspended solids and therefore the guideline value from the EU Freshwater Fisheries Directive (2006; repealed) is used as surrogate. The results are shown in Table 6 except for pyrene for which an EQS is not available.

Comparison with EQS/guideline values (established for or converted to total concentrations) indicate that mean predicted TSS concentrations would require a further four-fold dilution (increasing to a 17 fold dilution at its maximum predicted concentration) to avoid exceedance of the identified guideline value in receiving waters. Predicted total mean concentrations of Zn and Cu exceed the identified guideline values, with maximum predicted concentrations requiring 17- and 4-fold dilutions, respectively. With regard to benzo(*a*)pyrene, runoff from even the least trafficked highways would require at least a 10-fold dilution on discharge to avoid exceedance of the EQS. In contrast, even maximum predicted Cd concentrations in runoff (i.e. without receiving water dilution) would not prejudice receiving water guidelines. The high dilutions which have been shown to be necessary for TSS, Zn, Cu and benzo(*a*)pyrene to conform to receiving water quality standards clearly identify those locations where effective treatment of the highway runoff is recommended prior to discharge.

3.3. Regression analysis

Regression analysis of the relationship between transformed log pollutant and log AADT (see Fig. 2a and b for TSS and Zn, respectively, as examples) indicates r-squared values ranging from a minimum of 89.8% for Zn to a maximum of 90.7% for both Cu and TSS. This bivariate linear model based on AADT derives a very high level of statistical confidence in the traffic factor being the fundamental driver in pollutant generation. However, residual probability analysis demonstrates systematic under and over estimation of the data values with the residual versus fits regression data showing the existence of a degree of scatter. The indicated inherent bias in the dataset reflects that in addition to vehicle numbers the identified correlation is also influenced by factors such as traffic type and driving mode as well as aerial deposition.

The regression equations enable identification of the predicted background concentrations (i.e. the concentrations predicted to discharge from highways in the absence of traffic) as well as the AADT above which EQS values, identified in Table 6, would be exceeded. The derived ranges and mean values for TSS (0.14–0.41 mg/L; 0.24 mg/L), Zn (0.21–0.73 μg/L; 0.39 μg/L), Cu (0.05–0.15 μg/L; 0.09 μg/L), Cd (0.09–0.27 ng/L; 0.16 ng/L), pyrene (1.6–4.7 ng/L; 2.8 ng/L) and benzo(*a*)pyrene (0.21–0.60 ng/L; 0.35 ng/L) indicate that, even in the absence of traffic, runoff derived from highway surfaces will contain each of the assessed pollutants, although at comparatively much lower concentrations than those predicted for trafficked highways (Table 6). Of particular note is that the concentration of B(*a*)P predicted in runoff from highways without traffic would still exceed the EQS. Pollutant sources, which all contribute to the minimum background concentration level of these substances in a 'no-traffic' highway environment include highway materials (e.g. bitumen), highway marking paints, street furniture (e.g. crash barriers) and aerial deposition.

In terms of the AADT numbers predicted to result in an EQS exceedance, the ranges for TSS (1025–7239), Zn (1591–12,126), Cu (4926–35,460) and

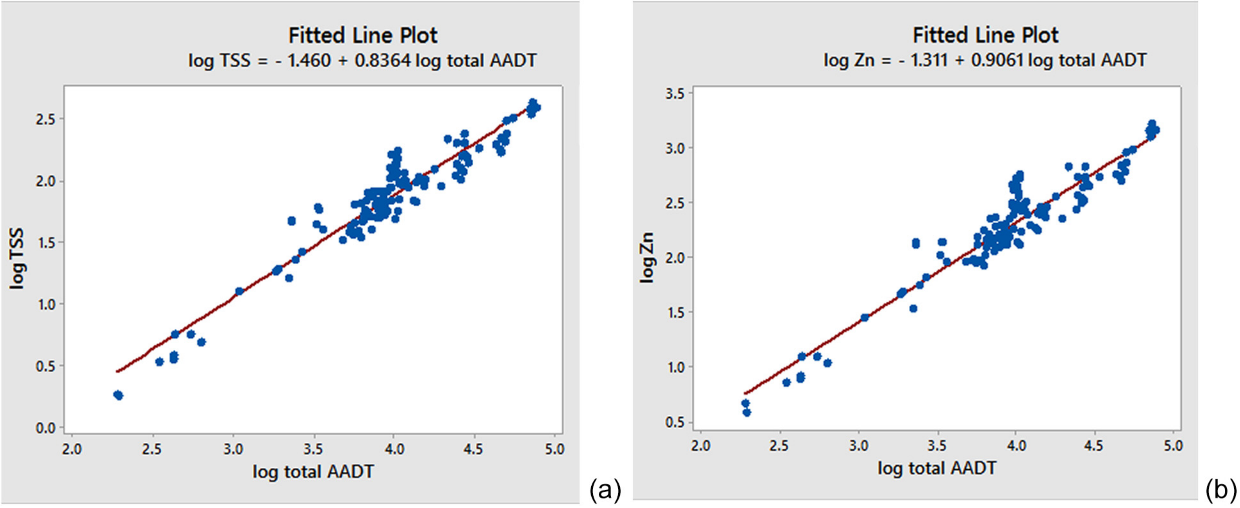

Fig. 2. Fitted line plots for log TSS (a) and log Zn (b) versus log AADT.

Cd (32,131–347,421) are broadly within the ranges of those cited in the literature identifying when runoff from a highway should be treated prior to discharge to a receiving waterbody. For example, National Road Authorities in Austria and Italy recommend treatment of runoff from highways with an AADT > 15,000, with this being a mandatory requirement in Germany (Meland, 2016). However, the mean AADT values predicted to lead to an exceedance of the respective guideline values/EQS for TSS (2582) and Zn (4291) suggest that lower AADT values may be relevant for triggering the need for treatment. Clearly, the mitigation of TSS and Zn should be a priority with runoff from highways experiencing AADT values as low as 5000 requiring treatment prior to discharge to receiving waters if EQS are not to be consistently exceeded.

3.4. Future scenarios

The current composition of the vehicle fleet in the UK is heavily influenced by petrol and diesel engined vehicles with only of the order of 2% being powered by electricity. However, as part of measures to improve air quality in urban areas, the UK Government has committed to phase-out the sale of petrol and diesel cars and vans by 2030, with tail pipe emissions to be ‘fully zero’ from 2035 (DfT, 2020). The impact that such a change would have on runoff pollutant concentrations has been assessed by considering the scenario in which all cars and light duty vehicles are replaced by electric vehicles (i.e. tail pipe emissions set to zero and no oil leakages). In addition, a second scenario has been considered in which a 50% reduction in the total number of heavy goods vehicles is achieved. Both scenarios have been evaluated for a section of four-lane carriage way (AADT 69,311; heavy goods vehicles compose 15.7%) and a section of single carriage way (AADT 15,286; heavy goods vehicles contribute 2.4%) within the London Borough of Enfield.

The results for the complete introduction of electric cars and light duty vehicles indicate only marginal percentage reductions in pollutant runoff concentrations would be achieved for the evaluated metals and TSS for both highway types (i.e. from 0.06–1.12%). This is consistent with exhaust emissions representing a minor source of these pollutants compared to brake and tyre wear. This is not the case for pyrene and benzo(*a*)pyrene for which much higher percentage reductions are predicted (from 19.9–47.8%). A different pollutant reduction pattern is observed for the scenario involving a 50% lowering of the number of heavy goods vehicles. This equates to reductions of 18.8–37.3% for metals and TSS, 15.5% for pyrene and 19.2% for benzo(*a*)pyrene on the four-lane carriage way and reductions of 2.1–13.1% (metals and TSS), 3.4% (pyrene and benzo(*a*)pyrene) from the single lane carriage way and is indicative of the major contributions made by non-exhaust emissions for heavy goods vehicles, particularly on highly trafficked roads.

Although this scenario analysis is a theoretical exercise, the results clearly indicate that, whilst the planned move to electric vehicles is predicted to achieve substantial decreases in runoff concentrations of pyrene and benzo(*a*)pyrene, minimal reductions in metals and TSS would occur. Hence, even after the implementation of phase two of the UK Government plan (i.e. tail pipe emissions set to zero) the discharge of highway runoff to receiving waters are likely to continue to prejudice its ecological status. Further, this analysis also demonstrates the disproportionate contribution of heavy goods vehicles to runoff pollutant concentrations, whereby reductions of 50% in total heavy goods vehicles (which contribute 15.7% on the most trafficked highways) correspond to predicted reductions ranging from 18.8% (for Cd) to 37.3% (Zn) for the four-lane carriage way.

4. Conclusions

This study describes a novel approach to predicting highway runoff concentrations on a site-by-site basis involving the combination of data on vehicle characteristics, emission factors and local rainfall. The approach has been applied to 126 sections of highways in a region of North London (UK) with differing AADTs and contributing drainage areas. Predicted concentrations fall within the range of empirically determined concentrations reported in the literature, with total AADT and the number of heavy goods vehicles emerging as principal drivers of runoff quality. The impact of the recently agreed UK Government commitment to phase-out the sale of petrol and diesel cars and vans by 2030 on runoff pollutant concentrations was evaluated on two sections of highway with differing levels of AADT numbers and compositions, together with a second hypothetical scenario in which the number of heavy goods vehicles were reduced by 50%. Results indicate that whilst concentrations of pyrene and benzo(*a*)pyrene in highway runoff fall under both scenarios, replacement of petrol/diesel cars by electric vehicles will have only a marginal impact on highway runoff concentrations of metals and TSS. These results have clear implications for future receiving water protection policies. For example, the continued need for strong highway discharge controls (particularly maintenance of minimum dilution levels in receiving waters) is identified even after the adoption of ‘cleaner emission technologies’, as well as the need for continued research into product and/or substance replacement within all elements of highway environments. In terms of implications for practice, the methodology and obtained results can be used to inform a site assessment of the suitability for direct discharge to a receiving water based on its dilution capacity, with the approach offering value to practitioners in terms of supporting decisions on prioritising when and where highway runoff should be treated.

CRediT authorship contribution statement

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J Bryan Ellis methodology, writing - original draft preparation; writing- reviewing and editing;

Nathalie Gilbert writing- reviewing and editing;

John Bryden writing- reviewing and editing;

Lian Lundy methodology, writing - original draft preparation, writing- reviewing and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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