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Review of highway runoff characteristics: Comparative analysis and universal implications

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ABSTRACT

This review interprets highway runoff characterization studies performed on different continents. The results are synthesized to discuss the historical trends, first flush effects of pollutants, pollutant form as dissolved vs. particulate, and to identify surrogate water quality parameters. The information presented in this review showed that: (1) variability has been observed in all quality parameters from each continent and among continents; (2) with a few exceptions the variability seems to be within the expected range; (3) inconsistent monitoring data as well as inconsistent quality assurance and quality control measures were reported among studies, which may be partially responsible for variability of water quality results; (4) compared with historic data, the concentration of total Pb decreased exponentially, which can mostly be credited to leaded gasoline phase-out regulation; (5) first flush effects of pollutants based on concentration have been reported consistently (however, mass first flush effects for pollutants have been reported inconsistently compared with concentration first flush effect); (6) most metal pollutants and phosphorus are present in both the particulate and dissolved forms; and (7) strong correlations were observed between TSS, TDS, TOC and iron (Fe) and 13 other constituents and water quality parameters (turbidity, O&G, TPH, DOC, TKN, EC, Cl, Cd, Cr, Cu, Ni, Pb, Zn).

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Acronyms and symbols

AADT	annual average daily traffic
ADP	antecedent dry periods
ADV	electronic data validation
AF	Africa (continent)
Ag	silver
Aggregate	water quality parameter that is usually comprised of multiple constituents (e.g., TSS shows the overall contribution of all particulate elements)
Al	aluminum
As	arsenic
AS	Asia (continent)
AZ	Australia/New Zealand (continent)
BMP	best management practice
BOD	biological oxygen demand
Cd	cadmium
Cl	chloride
COD	chemical oxygen demand
Cr	chromium
CSR	cumulative seasonal rainfall
Cu	copper
CWA	clean water act
d	dissolved fraction (e.g., dissolved Cu $= \text{Cu}_d$)
DA	drainage area
DO	dissolved oxygen
DOC	dissolved organic carbon
EC	electric conductivity
EMC	event mean concentration
EU	Europe (all countries)
EPA	Environmental Protection Agency
Fe	iron
FIB	fecal indicator bacteria
FHWA	federal highway administration
GIS	geographic information system
HCl	hydrochloric acid
ICP-MS	inductively coupled plasma-mass spectrometry
IF	impervious fraction
LID	low impact development
MFF	mass first flush
MFFn	mass first flush ratio for *n* fraction of volume
mg/L	milligram per liter
MLE	maximum likelihood estimation
MLR	multiple linear regression
MRI	maximum rain intensity
MST	microbial source tracking
na	not analyzed
NA	North America (continent)
NDs	non-detects (e.g. below a measured quantifiable value by analytical instrument)
NH_3–N	ammonia–nitrogen
NH_4–N	ammonium–nitrogen
Ni	nickel
NO_2–N	nitrite–nitrogen
NO_3–N	nitrate–nitrogen
NPDES	national pollution discharge and elimination system
Ns	not significant
NS	not sampled
NURP	national urban research program
O&G	oil and grease
PAHs	polycyclic aromatic hydrocarbons
Pb	lead
PNFF	particle number first flush
PSD	particle size distribution
qPCR	quantitative polymerase chain reaction
QA/QC	quality assurance/quality control
R	correlation coefficient
ROS	regression on order statistics
SA	South America (continent)
SCR	seasonal cumulative rainfall
SD	standard deviation
SE	standard error
t	total fraction (i.e. total Cu $= \text{Cu}_t$)
TDS	total dissolved solids
TER	total event rainfall
TKN	total Kjeldahl nitrogen
TMDL	total maximum daily load
TN	total nitrogen
TOC	total organic carbon
TP	total phosphorus
TPH	total petroleum hydrocarbons
TSS	total suspended solids
UCD	University of California, Davis
WFD	water framework directive (European)
Zn	zinc
µg/L	microgram per liter
µm	micron (1/1000 mm)

1. Introduction

1.1. Background

Urban runoff contains pollutants such as suspended solids, fine particles, heavy metals, nutrients, organic chemicals (including herbicides and pesticides) and fecal indicator bacteria (FIB) that can cause significant degradation of receiving water quality. This is illustrated, for example, by the frequency of coastal beaches being posted for contaminants after storm events due to health risk impacts in the United States (Schiff et al., 2003), and the impact of urban land use on water quality (Arnold and Gibbons, 1996). Water pollution concerns in the U.S. are addressed through the Clean Water Act (CWA) regulation established in 1972 and subsequent national pollution discharge elimination (NPDES) permits established in 1983 (Harrop, 2001). Provisions within the CWA require all states to implement regulations to control the discharge of urban stormwater and reduce the pollutant mass loading prior to discharging into receiving water bodies (Helland, 1998; White and Boswell, 2006). Initially, the discharge regulations were only applicable to communities with a population of 100,000 or more, but starting in 2006 the discharge regulation has been applied to all communities with a population of 10,000 or more (USEPA, 2011). To comply with discharge regulations, all states are also required to compute the pollutant discharge mass loading through the total maximum daily load (TMDL) or the total amount of a pollutant a water body can receive and still meet water quality standards (Ribaudo, 2006).

Up to 2000, most European countries had their own individual surface water quality regulations. To apply regulations across country boundaries, "water framework directives (WFD)" have been established to regulate the discharge of pollutants into surface and groundwater. The WFD was established in June 2000 through an agreement of the conciliation committee, consisting of representatives from the European Parliament and European Council (Kallis and Butler, 2001). Developing WFD in Europe was not an easy task, requiring 12 years to become a comprehensive water regulation. The WFD sets common approaches and goals for the management of water in 27 European countries and created an important overall water policy for management at an international level. Water directives characterize the different phases of environmental policy evolution, from an emphasis on public health protection to environmental protection per se, and from "end-of-pipe" solutions to preventative and integrated management approaches (Kallis and Butler, 2001). Many other developed and developing countries around the world have also recognized the impact of non-point source pollution on their drinking water supplies and developed initiatives to reduce the discharge of pollutant load to their water supplies. The extent and severity of the surface water quality regulations are usually related to the economic status of the countries (WHO/UNISEF, 2006).

Reduction of stormwater pollution can be achieved by source control. However, source control alone may not eliminate the discharged pollutant load. To accomplish pollutant reduction and develop a cost effective treatment, knowledge of runoff water quality characteristics is required. At present, the runoff characterization data base is unevenly distributed among the countries around the world. Since early 2000, the number of publications on this topic has increased in Asia and other communities around the world. At present, no comprehensive literature analysis of highway runoff studies on an international level has been conducted, which is the focus of this review.

1.2. *Objectives of the paper*

This review has the following specific objectives:

1. Summarize and compare the monitoring and chemical characteristics of highway runoff.
2. Synthesize the existing literature to:
    a) Evaluate any historical trends in highway runoff quality,
    b) Examine the first flush effect of pollutants,
    c) Assess the partitioning of pollutants as dissolved vs. particulate, and
    d) Identify surrogate water quality parameters.
3. To discuss future monitoring challenges

2. Referenced papers and their unique identification (ID)

Table 1 summarizes the representative highway runoff characterization studies containing water quality and pertinent monitoring data from various parts of the world. The papers presented in this table may not be the complete compilation of all papers published on stormwater. They were selected based on several criteria including: (i) the paper was published in a peer reviewed journal, (ii) it was easily accessible from an existing open literature data base, (iii) runoff characteristics are restricted to highway and as much as possible snowmelt runoff was not considered, and (iv) the paper included sufficient data on physical site characteristics, hydrologic characteristics, sample collection and analytical methods, and QA/QC procedures. Despite these efforts, it is possible that publications have been missed. The studies are organized based on their original location herewith known as: (1) North America (NA) that includes the United States, Canada and Mexico, (2) Europe (EU) including all European countries, (3) Asia (AS) including the Middle East, (4) South America (SA), and (5) Australia/New Zealand (AZ). Each publication has

Table 1 – Organization of international highway runoff characterization studies.

Location	Year[a]	Reference	Reference ID
Texas	1998	Barrett et al., 1998	NA2
Texas	2006	Barrett et al., 2006	NA3
Washington	1979	Bourcier and Hindin, 1979	NA4
Nationwide	1990	Driscoll et al., 1990	NA9
California	2006b	Han et al., 2006b	NA14
California	2003	Kayhanian et al., 2003	NA24a
California	2003	Kayhanian et al., 2003	NA24b
California	2003	Kayhanian et al., 2003	NA24c
California	2007	Kayhanian et al., 2007	NA25
California	2009	Lau et al., 2009	NA34
Texas	2008	Li and Barrett, 2008	NA35
Nationwide, USA	1983	USEPA 1983	NA46
North Carolina	1998	Wu et al., 1998	NA56
Sweden	2003	Baeckstroem et al., 2003	EU2
Ireland	2007	Desta et al., 2007	EU7
Switzerland	2002	Furumai et al., 2002	EU9
Sweden	2007	Hallberg et al., 2007	EU11
Poland	2007	Klimaszewska et al., 2007	EU12
France	1999	Legret and Pagotto, 1999	EU13
Italy	2005	Mangani et al., 2005	EU14
France	2000	Pagotto et al., 2000	EU15
Germany	1997	Stotz, 1987	EU17
Sweden	2006	Westerlund and Viklander, 2006	EU18
S Korea	1010	Maniquiz et al.	AS4
Japan	2000	Shinya et al.	AS5
Japan	2003	Shinya et al.	AS6
China	2008	Yufen et al.	AS7
Australia	2010	Davis and Birch	AZ2
Australia	2000	Drapper et al.	AZ3

[a] Year that paper was published. The monitoring study was generally performed many years before.

a unique reference identification (ID) number, which is used throughout the text.

3. Summary of monitoring methods

A detailed comparative analysis of monitoring methods employed in runoff characterization studies is beyond the scope of this paper. Here, a summary of observations based on the available data reported from references listed in Table 1 is provided. Physical site characteristics, hydrologic characteristics and sampling characteristics were reviewed. Physical site characteristics include number of highway lanes, pavement type, impervious area (%), annual average daily traffic (AADT), drainage area, and land use.

Review of the hydrologic data showed that the number of storm events and total rainfall was reported frequently, whereas the maximum rain intensity, antecedent dry period, and flow rate were reported less frequently. Runoff sampling characteristics include sampling methods, analytical technique, holding time, detection limits and QA/QC information. Our review showed that automatic flow weighted composite sampling is predominantly used for sample collection; but grab sampling was also used by some studies. Not all studies provided extensive QA/QC procedures when collecting the samples and analyzing the samples chemically. Generally, the U.S. EPA analytical methods and standard method QA/QC procedures were followed. Detection limit was the most variable parameter and values ranging from 0.5 to 50 μg/L have been reported for commonly monitored chemical constituents. The choice of sampling method and analytical technique and the selection of detection limit can significantly influence the number of non-detects in the data and hence data analysis and data interpretation (Helsel and Cohn, 1988; Helsel, 1990; Kayhanian et al., 2002; Shumway et al., 2002).

4. Summary of highway runoff characteristics

Highway runoff quality has been summarized based on: (1) aggregate water quality parameters, (2) metals constituents, (3) nutrient constituents, and (4) other less frequently measured water quality parameters such as fecal indicator bacteria, toxicity, polycyclic aromatic hydrocarbons (PAHs), and herbicides and pesticides.

4.1. *Conventional and aggregate water quality parameters*

Aggregate water quality parameters include: total suspended solids (TSS), total dissolved solids (TDS), dissolved organic carbon (DOC), total organic carbon (TOC), chemical oxygen demand (COD), biochemical oxygen demand (BOD), oil and grease (O&G), hardness as $CaCO_3$, temperature and pH. As shown in Table 2, three aggregate parameters measured frequently were TDS, COD and O&G and the remainders of the aggregate water quality parameters were measured less frequently. As expected, some variations were observed for each aggregate parameter and among different continents (Fig. 1). Similar variability was observed during the California statewide highway runoff characterization studies and, in general, the values reported in Table 2 (with the exception of few reported values) were within the ranges of values obtained in California. The variability could be related to many factors including land use, rain intensity, sample collection and processing, analytical method, and QA/QC procedures. For example, the statistical analysis of data collected from California showed that the concentrations of aggregate constituents from rural highways (with lower traffic volume and with limited commercial and industrial impact) were generally lower than those from urban highways (Kayhanian et al., 2003, 2007). On the contrary, the concentration of TSS in runoff from rural highways was much higher due to the impact of erosion.

4.2. *Metals constituents*

Metal constituents in the literature generally include aluminum (Al), the metalloid arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), nickel (Ni), and zinc (Zn). The majority of studies measured the concentration of Cd, Cr, Cu, Pb, Ni and Zn, and the concentrations of Al, As, and Fe were measured less frequently (Table 3). One important observation is the variability of these metal pollutant concentrations within each continent and between continents (Fig. 2a–f). Median values for Cu, Pb and Zn from European countries are generally 20–30% higher than North America, and the median values for Asia are usually at least 50% higher than the median values from North America. The variation in metal constituents between continents may have occurred for two reasons: (1) much larger data set was available from North America and (2) the majority of highway runoff characterizations from Asia were performed in the highly urbanized areas of Japan, Korea and China. The historic trend related to Pb concentration and the leaded gasoline phase-out regulation, will be discussed in Section 5.

The average concentration of Cu, Pb and Zn was found to have no direct correlation with AADT. A similar conclusion was drawn with data obtained from Caltrans statewide highway runoff characterization data (Kayhanian et al., 2003). Additional statistical analysis could not be performed with the current literature because of differences in reporting. However, statistical analysis of Caltrans data revealed that AADT is one factor among other parameters such as land use, drainage area, total cumulative rainfall, and antecedent dry period that impacts the pollutant concentration. In California, however, the concentration of metals on urban highways with AADT $>$ 60,000 was higher than metal concentrations in rural highways with AADT $<$ 30,000.

4.3. *Nutrients*

Selective nutrients monitored from different studies include nitrates (NO_3^-), nitrites (NO_2^-), ammonium (NH_4^+), total Kjehldal nitrogen (TKN), total nitrogen (TN), phosphate (PO_4^{3-}) and total phosphorus (TP) (Table 4). Nitrogen and phosphorus constituents can be transformed in the environment from dissolved to particulate forms or from one dissolved form to another, with an overall impact that can be substantial. As shown, most studies did not measure all

Table 2 – Conventional water quality parameters in highway runoff.

Reference ID	Average aggregate and conventional water quality parameter EMC or value								
	TSS (mg/L)	TDS (mg/L)	DOC (mg/L)	TOC (mg/L)	COD (mg/L)	BOD (mg/L)	O&G (mg/L)	Hardness as $CaCO_3$ (mg/L)	pH
NA2	129	–	25	46	130	12	4.2	–	–
NA3	117.8	–	–	–	64	–	–	–	–
NA9	142	–	–	25	114	–	–	–	–
NA14	67.7	–	66.9	–	252.5	–	14	78.9	6.7
NA24a[c]	476.3	127.3	–	–	146.7	–	4.3	–	–
NA24b[d]	144.7	135.6	–	–	119	–	10.9	52.5	7.4
NA24c[e]	168	107.2	–	–	145.5	–	2.5	30.3	7
NA25	112.7	87.3	18.7	21.8		–	6.6	36.5	7.1
NA35	137.8	–	–	–	80.1	–	–	–	–
NA46	180	–	–	–	82	–	–	–	–
NA56	283	157	–	–	70	–	4.4	–	–
EU2	–	–	–	11.8	–	–	–	–	6.4
EU7	270[a]	–	–	–	–	–	–	–	–
EU9	128	–	–	–	–	–	–	–	–
EU12	–	–	–	–	–	–	–	–	7.1[a]
EU13	71	–	–	–	103	–	–	–	7.3
EU14	–	–	–	–		–	–	–	7.7[a]
EU15	46	–	–	–	80	–	–	–	7.4
EU17	–	190[a]	–	–	103.7[a]	–	4.9[a]	–	–
EU18	64.5	–	–	–	–	–	–	–	–
AS4	76	–	15.9		33.3	16.6	1.3	–	
AS5	62.5[a]	–	–	40.5[a]	–	–	–	–	–
AS6	54.3	–	–	48.3	–	–	–	–	–
AS7	176.1	–	–	–	278.6[b]	14.3[b]	–	–	–
AZ3	336[a]	–	–	–	–	–	–	–	6.43[a]

For names of the conventional water quality parameters refer to symbols and acronyms.
EMC = event mean concentration.
– = not reported.
For references listed under reference ID refer to Table 1.
a Calculated in this study.
b Reported as median value.
c Calculated based on prior 1998 monitoring data.
d Urban highways: AADT > 60000 cars/d.
e Rural highways: AADT < 30000 cars/d.

forms of nitrogen and phosphorus constituents. The nutrient forms most frequently measured were TKN or total N and total P. The sources of nitrogen and phosphorus species measured in highway runoff may be related to both traffic and non-traffic sources. However, the contribution of N and P from traffic-related sources in runoff appears to be less significant than that from to natural sources such as soil and vegetation from surrounding land uses (Kayhanian and Paytan, 2011). The variability of data within North American studies is relatively low (Fig. 3). The concentration of reported nitrate from two European studies, on average, is about four times larger than the average value from North American studies. Similarly, the average concentration of total nitrogen (or TKN) and TP from two Asian Studies is about four times higher than the average concentration reported from North American studies. This large variation may be related to the proximity of the farm land to the highways.

4.4. *Infrequently measured water quality parameters*

Water quality parameters measured less frequently in highway runoff characterization studies include fecal indicator bacteria (FIB), toxicity, polycyclic aromatic hydrocarbons (PAHs), herbicides and pesticides. Due to limited data availability and space limitation, these additional analyses are not presented in this review. However, the lack of a larger data set does not automatically indicate that these pollutants are less important. Insufficient published data at present time may be reflective of limited water quality standards and/or less regulatory compliance requirements. In the future, as more surface water regulations are enforced, more research will be conducted and consequently more data related to these constituents will be collected. As an example, an extended list of primary pollutants including herbicides, PAHs and industry derived organic compounds (e.g. PCB-28) has been proposed by Eriksson et al. (2007) for European countries. The challenges associated with monitoring of pathogenic organisms and future directions of stormwater runoff management are further discussed in Section 6.

5. Discussion

This section of the paper is prepared to synthesize the review results to discuss: (1) historical water quality trends, (2) the

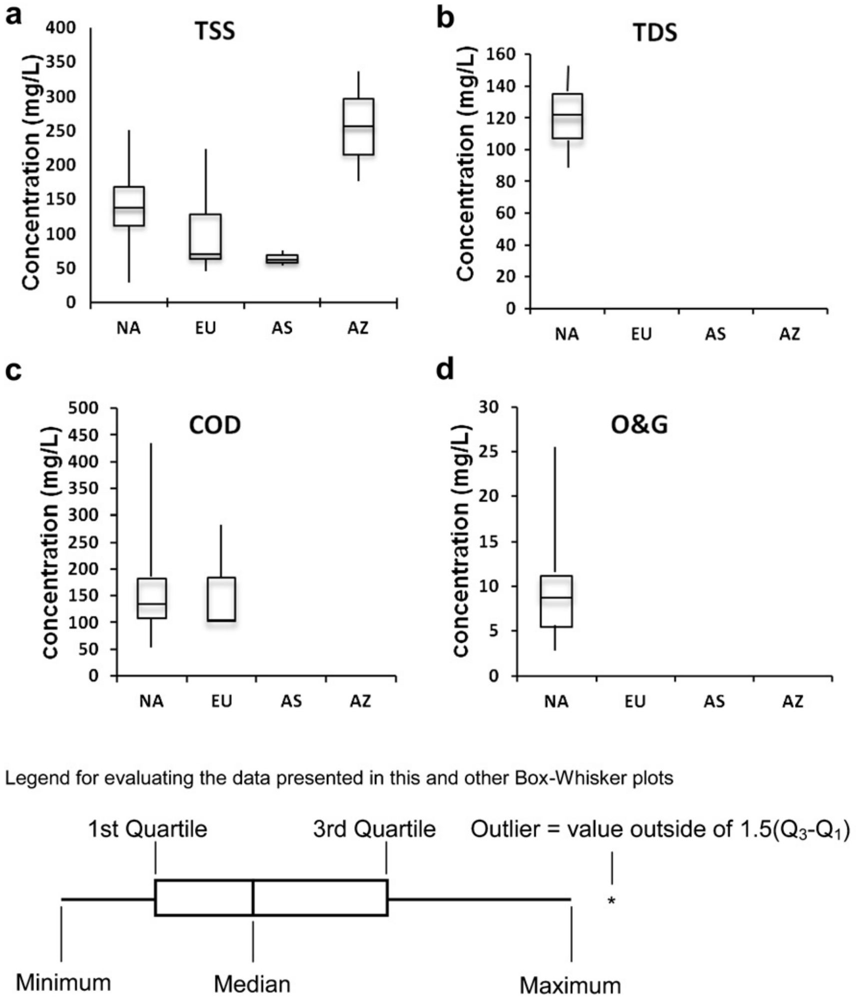

Fig. 1 – Range of mean values for selected aggregate constituents in highway runoff from different continents: (a) TSS, (b) TDS, (c) COD, and (d) Oil and grease.

first flush effect of pollutants, (3) the partition of pollutants (dissolved vs. particulate), (4) the use of certain water quality parameters as surrogates, and (5) the potential application of modeling approach for universal application.

5.1. *Historical trends of highway runoff quality*

Water quality trends based on monitoring data collected in the 1980s vs. more recent monitoring data are analyzed. Lead concentration was a focus (Table 5) to see if the amount of Pb discharged into the environment is reduced due to leaded gasoline phase-out regulations. One of the earliest studies to report Pb concentration in highway runoff is the USEPA study under the Nation Urban Runoff Program (USEPA, 1983). A few years later, the Federal Highway Administration (FHWA) performed a highway runoff characterization study throughout the United States (Driscoll et al., 1990). An additional four and nine studies were performed to measure Pb concentration in North America during the 1990s and 2000s, respectively. During the same period one study each was performed in 1987 and 1999, with an additional seven studies reporting Pb concentrations from highway runoff in Europe. Because of the availability of the historic data from these two continents, the average Pb concentration from NA and EU during 1980s, 1990s and 2000s can be compared (Fig. 4a). Clearly, the current average Pb concentration is substantially lower compared to early historic data. In addition, since both NURP and FHWA measured Pb at two to three highway sites in California, and later the same location was monitored under statewide highway runoff characterization study, the results made it possible to perform a historical comparison. The comparative trend of the average median total Pb event mean concentration (EMC) from highways in California during the 1980s, 1990s and 2000s is shown in Fig. 4b. The current average total Pb EMCs in highway runoff in California have been reduced exponentially compared to the FHWA highway runoff Pb median EMC measured in the late 1980s. The reduction in Pb EMC from highway runoff can potentially be credited to the leaded gasoline phase-out regulation (Kayhanian, 2012). This successful environmental regulation may be used as model for reducing pollution by source reduction.

Review of additional data presented in Table 3 showed no clear reduction or increase in the concentration of other metal pollutants. This may be understandable, particularly when

Table 3 – Metals in highway runoff.

Reference ID	AADT	Average metal EMC[a]								
		Al (mg/L)	As (μg/L)	Cd (μg/L)	Cr (μg/L)	Cu (μg/L)	Fe (mg/L)	Pb (μg/L)	Ni (μg/L)	Zn (μg/L)
NA2	58150[f]	–	–	37	–	–	2.824	53	–	222
NA3	43000[f]	–	–	26.8 (5.9)	–	–	–	12.6 (0)	–	167.4 (47.1)
NA4	–	–	–		–	140	89	1860	–	19100
NA9	30000[g]	–	–	54	–	–	–	400	–	329
NA14	303000[f]	–	–	92.9 (65.9)	1.8 (1.4)	9.7 (2.8)	–	25.8 (4.9)	20 (15.7)	506.3 (415.3)
NA24a[c]	–	–	–	42.4 (16.3)	–	–	–	89.1 (10.2)	–	279.3 (96.8)
NA24a[d]	328000[f]	–	11.6 (1.2)	59 (14.7)	1.1 (0.3)	9.4 (2.6)	–	92.5 (6.1)	10.4 (3.6)	228.8 (79.1)
NA24b[e]	328000[f]	–	0.7 (0.2)	9.4 (6.5)	0.2 (ND)	5.5 (1.7)	–	8.2 (1.2)	8.6 (3.6)	63.4 (35.3)
NA34	–	–	–	93.1 (67)	1.8 (1.3)	10.1 (2.8)	–	33 (4.9)	20 (15.7)	507 (416)
NA35[b]	50000[f]	–	–	21 (5.8)			–	9.8 (<1)	–	138.1 (48.8)
NA45	–	–	2.4 (1.4)	34.7 (10.9)	1 (0.68)	8.3 (2.3)	–	25 (1.8)	9 (4)	200(51)
NA46	–	–	–	43	–	–	–	182	–	202
NA56	25000[g]		–	24.2	2.5	8.1		21	8.1	–
EU2	15600[g]	0.15	–	18.2	0.056	–	0.334	5.64	–	130
EU7[b]	25600[g]	–	–	63.3 (ND)	–	–	–	35	–	250(22.5)
EU9[a]	52000[f]	–	–	57	–	–	–	26	–	354
EU11	120000[f]	(0.02)	–	(20.3)	(0.04)	(3.36)	(0.037)	(0.14)	(3.08)	(100)
EU12[b]	46800[f]	–	–	–	16.6	–	–	319	–	334
EU13	12000[g]	–	–	45	1	–	–	58	–	356
EU14[b]	18000[g]	4.9 (0.16)	–	350.6 (24.4)	–	–	3.1 (0.07)	56.54 (2.8)	–	1784 (107.7)
EU15	24000[g]	–	–	30 (19)	0.88 (0.32)	–	–	40 (3.3)	–	228 (140)
EU17[b]	43000[f]	–	–	90.6	4.86	11.7	3.58	203	–	433
EU18[b]	74000[f]	–	–	38.3 (7.2)	0.18 (0.03)	–	–	16.6 (0.3)	9 (1.2)	149.6 (40.6)
AS4	–	–	–	–	–	–	–	1200		300
AS5[b]	75000[f]	1.98	–	66	1.75	6.5	3.715	33.5	5.5	647
AS6	62000[f]	1.35	–	68.3	–	–	2.48	30.5	–	718.4
AZ2	84500[f]	–	–	97.3	–	–	–	44.5	–	347
AZ3	50000[f]	–	–	115[b]	–	–	–	215[b]	–	600

Values reported in () are dissolved fraction.
EMC = event mean concentration.
– = not reported.
For references listed under reference ID refer to Table 1.
a Calculated in this study.
b Reported as median value.
c Calculated based on prior 1998 monitoring data.
d Urban highways: AADT > 60000 cars/d.
e Rural highways: AADT < 30000 cars/d.
f Upper bound AADT value for urban highways.
g Upper bound AADT value for rural highways.

the metal pollutant generated directly related to transportation activities.

5.2. *First flush effect of pollutants*

Pollutants that collect during a dry period on highways may tend to be carried off with the early portion of storm runoff, causing a “first flush” of pollutants. The first flush is a phenomenon in which a larger pollutant concentration or mass is discharged during the first portion of the stormwater runoff compared with the rest of the runoff. The first flush of highway runoff has been reported by numerous researchers (Bertrand-Krajewski et al., 1998; Barrett et al., 1998; Deletic, 1998; Gupta and Saul, 1996; Saget et al., 1996; Sansalone and Buchberger, 1997a; Flint and Davis, 2007; Mangani et al., 2005; Furumai et al., 2002; Barco et al., 2008), and has been found over seasons or based upon individual storm events. Although first flush can be reported based on concentration or mass, concentration first flush is more straight-forward and is usually reported by a pollutograph in which the pollutant concentration is plotted against time. In most cases, the rain intensity or hydrograph is also reported on the same plot. The concentration first flush effect has been universally reported in the literature for most conventional water quality and chemical constituents. In addition, the first flush effect was extended to particle size distribution and toxicity. For instance Li et al. (2005) found in a particle first flush that over 90% of fine particles (<20 μm) were associated with 30% of runoff volume. In a separate study, Kayhanian et al. (2008a) showed the impact of first flush on toxicity (based on microorganism mortality rate and reproduction inhibition) by observing that the greatest degree of toxicity was related to the early stages of a storm event.

A review of the literature for mass first flush, however, showed that no one method has been applied consistently. One specific definition offered by Bertrand-Krajewski et al.

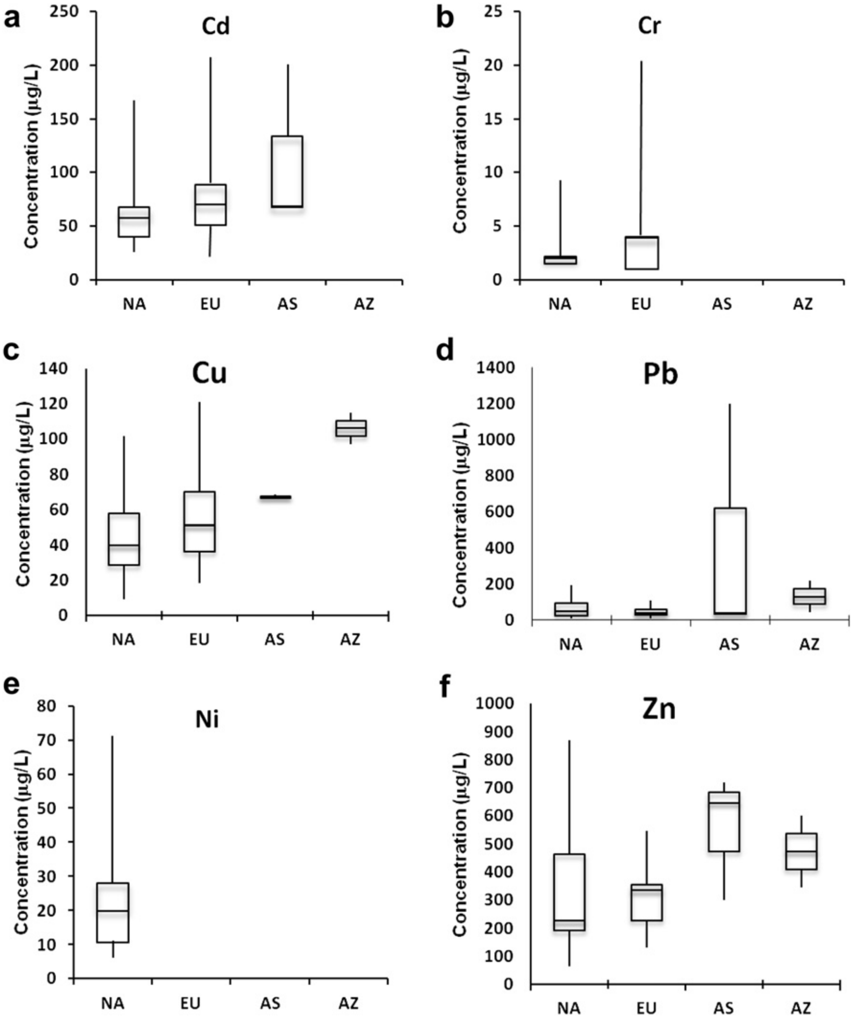

Fig. 2 – Range of mean values for selected metal constituents in highway runoff from different continents: (a) Cd, (b) Cr, (c) Cu, (d) Pb, (e) Ni, and (f) Zn.

(1998) defined the mass first flush when 80% of the pollutant mass is emitted in the first 30% of the runoff. Other definitions with minor deviation have been offered by other researchers (Vorreiter and Hickey, 1994; Saget et al., 1996; Gupta and Saul, 1996; Sansalone and Buchberger, 1997b; Sansalone et al., 1998; Deletic, 1998). While they all suggested mass first flush as a higher pollutant mass emission rate in the early part of the storm than in the later part, no uniform definition has been developed. Recently, Kayhanian and Stenstrom (2005, 2008) proposed the concept of mass first flush (MFF) ratio. The MFF ratio quantitatively describes the mass first flush and is sufficiently broad to be applicable to any initial portion of the storm. Based on this definition, the end user can specify the mass first flush without arbitrarily selecting a proportion of mass with respect to runoff volume. This definition was proposed since not all watersheds will produce the same mass first flush nor will the mass first flush for all pollutants be the same. Based on the MFF definition one can determine the mass first flush for any constituent at any runoff volume. For example, the mass first flush ratio for 10% of total runoff volume (MFF_{10}) is determined by dividing the normalized mass fraction (0.45) by normalized runoff volume (0.1) to equal 4.5. The mass first flush only exists when the MFF is greater than 1. The higher MFF ratio represents a larger mass first flush effect. The pollutants that consistently ranked high with respect to mass first flush ratio were COD, TKN, and DOC (Lau et al., 2009) as well as total Cu, Ni and Zn (Han et al., 2006b). To take advantage of first flush treatment, most conventional treatment systems can be divided into two compartments in which the first compartment can be devoted to first flush treatment and the overall BMP treatment size can be reduced by half (Li et al., 2006 and Abrishamchi et al., 2010).

5.3. *Partitioning of pollutants*

The primary unit processes or mechanisms used to treat highway runoff are settling and filtration. The design, operation and maintenance of these stormwater treatment practices is highly dependent on particle size, and the effectiveness of a process to remove pollutants would depend upon the size of particles with which the pollutant is associated. To address this issue, some researchers evaluated the size-resolved particulate metal concentrations, nutrients and PAHs (Ellis et al., 1987; Sansalone and Buchberger, 1997b;

Table 4 – Nutrients in highway runoff.

Reference ID	Average nutrients EMC						
	NO_3 (mg/L)	NO_2 (mg/L)	NH_4 (mg/L)	NH_3 (mg/L)	TKN (mg/L)	PO_4–P (mg/L)	Total P (mg/L)
NA2	1.07	–	–	–	–	–	0.33
NA3	0.43[e]	–	–	–	1.13	–	0.13 (0.06)
NA9	0.76[e]	–	–	–	1.83	0.4	–
NA14	2.7	0.3	–	4.6	9.6	0.2	0.9
NA16	<0.05	–	–	–	2.4		0.2 (<0.05)
NA24a[b]	–	–	–	–	5.2	0.2	0.7
NA24b[c]	1.1	0.1	1	–	2.1	0.12	0.3
NA24c[d]	0.6	ND	2.3	–	2	0.1	0.2
NA25	1.07	–	–	–	2.06	0.11	0.29
NA35[a]	0.554[e]	–	–	–	1.71	–	0.2 (0.04)
NA45	0.28[e]	–	–	1.07	2	–	0.25 (0.2)
NA46	0.86[e]	–	–	–	1.9	0.15	0.42
NA56	2.25[e]	–	–	0.83	1.42	–	0.43
EU12[a]	2.87	–	0.27	–	–	–	–
EU13	–	–	–	–	2.3	–	–
EU14[a]	4.8	0.725	0.43	–	–	–	–
EU15	6.7	–	1	–	2.1	–	–
EU17[a]	–	–	0.72	–	–	–	0.91
AS4	–	–	–	–	4.3[f]	–	0.8
AS7	–	–	–	–	13.62[f]	–	0.46
AZ3	–	–	–	–	4.1[f]	–	0.81

TKN = total Kjeldahl nitrogen and PO_4–P is phosphate given as phosphorus concentration.
EMC = event mean concentration.
– = not reported.
For references listed under reference ID refer to Table 1.
a Calculated in this study.
b Calculated based on prior 1998 monitoring data.
c Urban Highway with AADT > 60000 cars/d.
d Rural Highway with AADT < 30000 cars/d.
e Nitrate + Nitrite.
f Total N

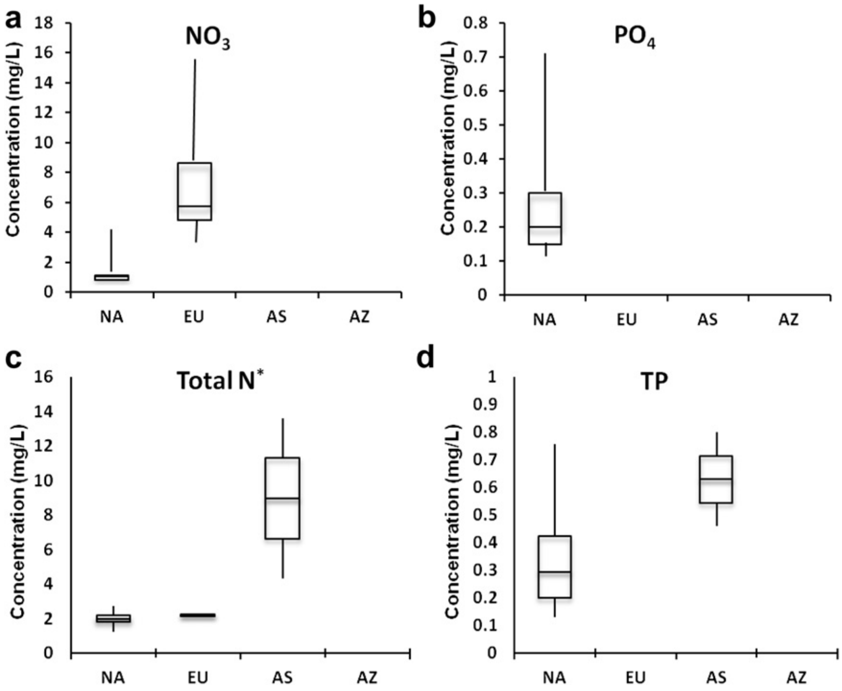

Fig. 3 – Range of mean values for selected nutrient constituents in highway runoff from different continents: (a) NO_3, (b) PO_4, (c) total N, and (d) total P. (* in Fig 3c indicate that most total N was measured and reported as TKN).

Table 5 – Summary of total Pb concentration from highway runoff from 1979 to present.

Continent[‡]	Reference ID	Data source	Year	Average total Pb concentration (μg/L)
North America	NA4	Bourcier and Hindin	1979	1860
	NA46	USEPA	1983	182
	NA9	Driscoll et al.	1990	400
	NA50	Sansalone and Buchberger	1997a	64
	NA2	Barrett et al.	1998	53
	NA56	Wu et al.	1998	21
	NA24a*	Kayhanian and Borroum	2000	89
	NA24b**	Kayhanian et al.	2003	93
	NA24c***	Kayhanian et al.	2003	8
	NA3	Barrett et al.	2006	13
	NA14	Han et al.	2006b	26
	NA25	Kayhanian et al.	2007	48
	NA35	Li and Barrett	2008	10
	NA34	Lau et al.	2009	33
Europe	EU17	Stotz,	1987	203
	EU13	Legret and Pagotto	1999	58
	EU15	Pagotto et al.	2000	40
	EU9	Furumai et al.	2002	26
	EU2	Baeckstroem et al.	2003	6
	EU19	Westerlund et al.	2003	17
	EU14	Mangani et al.	2005	57
	EU7	Desta et al.	2007	35
	EU12	Klimaszewska et al.	2007	319[a]
Asia	AS5	Shinya et al.	2000	34
	AS6	Shinya et al.	2003	31
	AS4	Maniquiz et al.	2010	1200[b]
Australia/New Zealand	AZ3	Drapper et al.	2000	215[c]
	AZ2	Davis and Birch	2010	45

[‡] Complete phase out of leaded gasoline in the United States and Canada was 1996 and 1999, respectively. Most European countries phased out leaded gasoline in 2000. In Asian countries such as South Korea, Japan and China the leaded gasoline phase out occurred in 1999, 1999, and 2000, respectively. The complete phase pot of leaded gasoline in Australia and New Zealand were 2005 and 1999, respectively.

*Include highway runoff data prior to 1998.

*Include only highway runoff data during 2000 and 2001.

a This high Pb concentration may be explained by possible contribution from other nearby sources or analytical error since the concentration of Zn reported as lower than Pb which usually reverse is true for highway runoff concentration.

b The site is multiple lane road with intense traffic acting like a highway. This high Pb concentration may be explained by possible contribution from other nearby sources or analytical error.

c This high Pb concentration may be explained by possible contribution from other nearby sources or analytical error.

Roger et al., 1998; German and Svensson, 2002; Zhao et al., 2010; Kayhanian et al., 2004; Aryal et al., 2005; McKenzie eta al., 2008). As an example, representative studies that reported size resolved metal (Cu, Pb, and Zn) concentration are summarized in Table 6. As shown, higher metal concentrations are generally associated with smaller particles. Also, it is important to note that in all studies, the reported size fractionation was restricted to particles larger than 25 μm. With an improved particle processing method, McKinzie et al. (2008) and Kayhanian and Givens (2011) investigated particles less than 20 μm. The results of these two recent studies showed that even though very fine particles (e.g., less than 20 μm) represent only a small mass fraction of the suspended solids, they can carry a disproportionately large metal load. Therefore, having advance knowledge of particle size distribution and size-resolved pollutant concentration is advantageous for designing and evaluating the performance of BMPs.

In addition, the partitioning of organic and inorganic pollutants between dissolved and particulate forms is currently of special interest. The dissolved forms are the most bio-available, and will result in a quicker response by the aquatic biota. Unfortunately, not all studies reported both dissolved and particulate fractions and hence comprehensive analysis across all studies is not possible. A review of the existing data in Table 3 reveals that lead, aluminum, iron, arsenic, cadmium and chromium are mostly associated with particles; whereas zinc, copper and nickel are mostly found in a dissolved form. Based on the limited data available from Table 4, the majority of phosphorus measured was in particulate form. However, this may not always be true for all locations and highway sites. For instance, nearly 90% of phosphorus measured on three highways in Lake Tahoe, California was in the dissolved form (Kayhanian et al., 2004). While not discussed here, some of the lower molecular weight PAH compounds were also present in dissolved form (Aryal et al., 2005).

From the above findings and to protect the integrity of receiving waters, it is important to note that treating dissolved pollutants may be as important as treating particulate pollutants, a fact that is being ignored or not acknowledged with existing treatment practices. While laboratory research

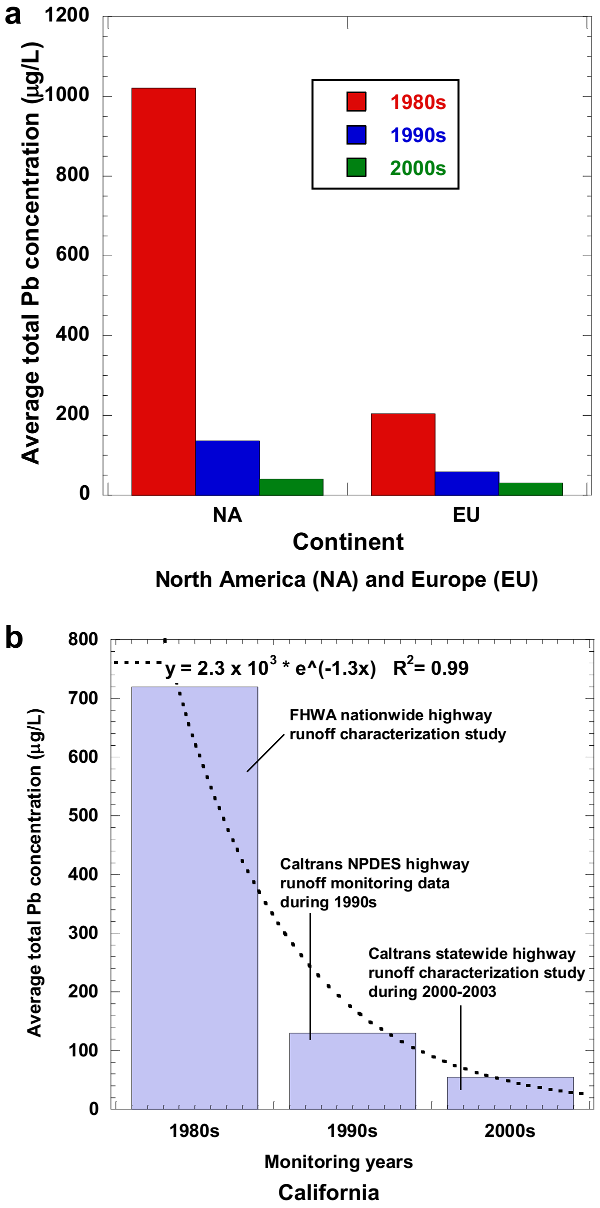

Fig. 4 – Trend of average total Pb EMC in highways runoff from 1980s to present: (a) North America and Europe, (b) California.

utilizing the adsorption and precipitation unit processes is beginning to be conducted in the field (Erickson et al., 2012), there is much to be done in this area.

5.4. *Potential use of specific constituents as surrogate water quality parameters*

Finding strong correlations among chemical constituents and water quality parameters would reduce the number of monitoring constituents, save analytical costs, and provide an opportunity for pollutant source identification and modeling efforts. An overview of the applicability of specific surrogate water quality parameters is given in Table 7. The suitability of pairs or multiple surrogate water quality parameters for given monitoring sites was determined by a correlation factor (R^2 values) of 0.8 or higher.

While there is general agreement for the correlation of TSS and particulate pollutants, in some situations a lower correlation (e.g., $R^2 < 0.8$) between TSS and particle-bound metal pollutants is observed. This lower correlation may be due to the particle size distribution in runoff and the method used for TSS measurement (Kayhanian et al., 2008b). For example, if the majority of particles contributing to TSS measurement are >75 μm and particles while mainly fine particles (<20 μm) contributed to particulate metal concentration, a weaker correlation is expected (Kayhanian and Givens, 2011). However, when the particles in a runoff sample are <75 μm and similar contributing particles were included in both particulate metal analysis and TSS measurement, then a stronger correlation between TSS and particle-bound metal pollutants is expected. Similar conclusions can be made with respect to correlations between TSS and turbidity (Table 7).

The number of correlations with an $R^2 > 0.8$ suggest that assessing trends in runoff characteristics, developing a strategic monitoring plan, and estimating mass loading or performance evaluation of treatment practices could be accomplished based on measurements of relatively few surrogate parameters (Table 7). A relationship between the aggregate water quality parameters and metal constituents reported in Tables 2 and 4 was attempted, but no correlation was observed. This may be an indication that there are no globally and universally applicable correlations with the current available data. To substitute one surrogate water quality parameter for another, the required mathematical relationships may need to be developed from local measurements. Under this specific condition, a shorter list of water quality parameters may serve as a potential substitute for a larger list when a comprehensive monitoring program is desired. Table 7 indicates that four constituents (TSS, TDS, TOC, Fe) may be monitored to serve as surrogates for 13 other constituents and water quality parameters (turbidity, O&G, TPH, DOC, TKN, EC, Cl, Cd, Cr, Cu, Ni, Pb, Zn) for many of the monitoring sites listed. For certain stormwater management programs, this shorter list of constituents can potentially reduce the monitoring effort and save substantial analytical costs.

6. Future monitoring challenges

A large amount of data has been collected internationally, and great progress has been made on a wide range of monitoring issues and chemical characteristics of highway runoff. Yet, there are some remaining challenges that once resolved, can universally benefit all global communities. They are briefly discussed below.

6.1. *Establishing standard effluent numeric water quality*

The establishment of effluent discharge quality based on simple water quality parameter measurements such as BOD and TSS helped the wastewater treatment industry develop

Table 6 – Metal concentrations for different particle size ranges.

Reference	Sample type	Size range (μm)	Metal concentration (μg/g)		
			Cu	Pb	Zn
Sansalone and Buchberger, 1997b	Highway runoff sediment	25–38	364	265	1189
		38–45	353	236	996
		45–63	364	266	1027
		63–75	333	258	1057
		75–150	312	248	1014
		150–250	204	195	574
		250–425	78	65	325
		425–850	48	53	314
		850–2000	45	37	259
Roger et al., 1998	Highway runoff sediment	<50	420	1570	4370
		50–100	250	1480	1700
		100–200	200	1550	1100
		200–500	100	850	930
		500–1000	50	460	930
German and Svensson, 2002	Street sweeping	<75	470	–	410
		75–125	270	–	230
		125–250	340	–	190
		250–500	200	–	120
		500–1000	50	–	70
Lau and Stenstrom, 2005	Street sweeping	<43	220	350	960
		43–100	230	300	805
		100–250	230	210	500
		250–841	240	44	150

– Not reported.

adequate treatment systems. It also allowed the treatment plants to assess their effluent water quality by comparison with known numerical discharge criteria. In contrast, no specific numerical limits have been established for stormwater runoff. Without such standards, compliance with regulation is difficult. For instance, in the United States when a water body is identified as impaired with respect to a pollutant or list of pollutants, it is regulated by a loading calculation mechanism called total maximum daily load (TMDL). Calculation of a TMDL is generally complicated and difficult to perform, particularly when there is no effluent numerical standard.

6.2. *Advances in evaluating fecal pollution*

At present, most of the regulatory compliances pertinent to discharge of pollutants from highway runoff in the U.S. and other parts of the world are related to metals and nutrients. In the future, however, the regulation may include emerging pollutants and pathogenic organisms. As stated before, in California alone over 7200 water bodies are listed as impaired water bodies and about 17% of these water bodies have pathogens as the cause of impairment. Fecal indicator bacteria (FIB) such as fecal (thermo tolerant) and total coliforms or *E. coli* are used as presumptive indicators of the presence of pathogenic organisms in urban runoff. Most studies treat FIB in highway runoff as just one point source among others, and so there are few investigations in the peer-reviewed literature that deal specifically with FIB in highway runoff. A major constraint is the sample holding time, which must not exceed 6 h. In addition, samples have to be kept on ice. There is an ongoing debate in the scientific community on the validity of FIB as a metric to predict the health risk and the presence of human pathogens in the absence of point sources of fecal contamination (see e.g., Bae and Wuertz, 2012), and the U.S. EPA is set to issue revised criteria for recreational water quality in 2012 (U.S. EPA, 2007). As with other discharges, FIB values in highway runoff tend to be highly variable. In general, when considering all monitoring studies where pathogens and FIB were measured, a major conclusion is that many studies had insufficient pathogen data for assessing correlations (Wu et al., 2011). For example, Rajal et al. (2007a) found no relationship between coliforms and human viruses in stormwater.

In the absence of sufficient data on pathogens in highway runoff, there has been increased interest in tracing sources of microbial pollution directly. Great strides have been made in the development of microbial source tracking (MST) methodology to detect host-associated bacteria or viruses/bacteriophages (Scott et al., 2002; Simpson et al., 2002 and Hagedorn et al., 2011). Methods are either cultivation-dependent (growth of target bacteria or viruses is required) or cultivation-independent (no growth is required). The latter methods tend to be more geographically independent (the same genetic assay can be used in different locations), but this assumption must still be validated before use in field studies is possible.

In recent years, the bacterial order *Bacteroidales* has emerged both as an alternative to the standard FIB and as an MST identifier due to its prevalence in the gastrointestinal tract and the availability of genetic assays to distinguish human feces from that of specific animal hosts like cattle and

Table 7 – Principal and related surrogate water quality parameters.

Reference	Principal WQ parameter	Surrogate WQ parameter	R^2
Andrews, 2004	Fe	Cu, Pb	0.8
Deletic and Maksimovic, 1998	TSS	Turbidity	0.8
Driscoll et al., 1990	TSS	Particulate metals	0.8
Gnecco et al. (2005)	TSS	COD	0.95
Han et al., 2006a	TSS	Particulate metals	0.8
Han et al., 2006a	DOC	COD, TKN, O&G	0.8
Kayhanian et al., 2007	TDS	Cl, EC	0.8
Kayhanian et al., 2007	TOC	DOC	0.96
Kayhanian et al., 2007	TPH	O&G	0.86
Kayhanian et al., 2007	TSS	Turbidity	0.8
Kayhanian et al., 2007	Fe	Total Cr, Pb, Cu, Pb, Zn	0.8–0.9
Kayhanian et al., 2007	TDS	Cl, EC	0.9, 0.8
Kayhanian et al., 2003	DOC	Total PAHs, herbicides, pesticides	0.8
Khan et al., 2006	O&G	COD, DOC	0.8
Lau et al., 2009	TSS	Total PAHs, particulate metals	0.8
Thomson et al. (1997)	TSS	Particulate Cu, Pb Ni, Zn	0.85

Note: Slopes and intercepts are not available since they were not reported by most studies. However, the high R^2 value is an indicative of a good correlation and validity of their use as surrogate parameters. The exact relationship for specific monitoring areas could be determined independently.

dogs (Santo Domingo et al., 2007; Wuertz et al., 2011). Assays have been used successfully to assess components of highway runoff and can also be employed to achieve quantitative source apportionment in a water sample; measured concentrations of specific DNA sequences that are characteristic of certain animal or human signatures are fed into a statistical model developed to correct for errors in measured data due to less than perfect specificity and sensitivity of assays (Kildare et al., 2007; Wang et al., 2010). Methods for the detection of human pathogens in ambient waters and other environmental matrices have been improving over the years, largely due to the application of molecular methods like quantitative polymerase chain reaction (qPCR) to detect nucleic acids. Future applications may prove to be reliable and sensitive enough – when coupled with validated water filtration methods (Rajal et al., 2007b) – to eventually replace FIB for the assessment of microbial water quality in discharges like highway runoff.

6.3. *Reducing monitoring need with low impact development*

Managing urban stormwater runoff and pollution prevention efforts in urban areas is often accomplished by including low impact development (LID) practices, which are decentralized methods for managing stormwater that include infiltration basins and trenches, porous pavements, rain and roof gardens, vegetative swales, and filter strips, among others. The principles of LID focus on prevention rather than mitigation by managing rainfall as close to the source as possible. When designed and practiced properly, some LID methods will eliminate the need for construction of conventional treatment practices and will also eliminate much of the monitoring costs.

7. Conclusions

The following conclusions are the result of this review:

- ***Consistency in monitoring data***: Most studies reported selected metal constituents and conventional water quality parameters, while limited data are available on nutrients, PAHs, toxicity, indicator bacteria, and herbicides/pesticides. Inconsistent monitoring data reporting made it difficult to perform valid statistical analysis.
- ***Variability of chemical characteristics***: Large variability in most water quality parameters was observed within each continent and between continents. The median concentration of most reported pollutants in Europe was generally 20–30% higher than the values from North America. The median concentration of pollutants from Asia was much higher and in some cases the concentration was four times larger compared with North America. One reason for these differences may be greater available space in North America and less intense highway usage.
- ***Historical trend with respect to lead concentration***: Historical data indicate an exponential decrease in concentration of total Pb. Current mean total Pb concentration is less than 45 μg/L compared to values of 1000 μg/L or greater in the 1970s. The decrease in total Pb concentration can be credited to the lead phase-out regulation.
- ***First flush effect of pollutants***: Most studies reported a concentration first flush effect in which higher concentration of pollutants are observed at the beginning of the storm events. Mass first flush effect for TKN, COD and DOC as well as Cu, Ni and Zn occurred more frequently than the other conventional and metal pollutants. Mass first flush concept can be used for treatment strategies to reduce treatment practice size and improve treatment efficiency.
- ***Partitioning of pollutants***: Existing monitoring data showed that most metal pollutants and phosphorous are primarily associated with particulates. There is some inconsistency in reported data related to Cu, Ni, Zn and P in which shown to be mostly in dissolved form. Some lower molecular weight poly aromatic hydrocarbons (PAHs) were also reported as the dissolved form. This finding clearly indicates the importance of treating dissolved pollutants that are generally being ignored or not treated by the existing treatment processes.
- ***Surrogate water quality parameters***: Strong correlations were found for three aggregate water quality parameters (TSS, TDS, and TOC) and iron (Fe) with 13 other constituents and water quality parameters (turbidity, O&G, TPH, DOC, TKN, EC, Cl, Cd, Cr, Cu, Ni, Pb, Zn). For certain stormwater management programs, this shorter list of constituents may

be used as surrogate parameters and hence this shorter list could potentially reduce the overall monitoring effort and substantially reduce annual analytical costs.

Acknowledgments

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