

Correlation analysis among highway stormwater pollutants and characteristics

Y.H. Han*, S.L. Lau*, M. Kayhanian** and M.K. Stenstrom*

*Civil and Environmental Engineering Department, University of California, Los Angeles, CA 90095-1593, USA (E-mail: younghan@ucla.edu; simlin@seas.ucla.edu; stenstro@seas.ucla.edu)

**Center for Environmental and Water Resources Engineering, Department of Civil and Environmental Engineering, University of California, Davis, CA 95616, USA (E-mail: mdkayhanian@ucdavis.edu)

Abstract Stormwater runoff from highway land use is a common non-point source of pollutants. A large quantity of highway stormwater runoff characteristics were collected in California during the past three years. Correlations among various water quality parameters and constituents were performed using data sets collected over the 2000–2001, 2001–2002, and 2002–2003 wet seasons for 18, 21 and 23 storm events at three highway sites in west Los Angeles, California. In addition, statistical and graphical correlation analysis of the mass first flush ratio (MFF) with storm characteristics was made to determine if the first flush is related to site or storm characteristics. The results and analyses performed indicate that (1) TSS correlates well with most particulate-bound metals. However, TSS was poorly correlated with most other pollutants. (2) Strong correlations were also observed among dissolved and total metals; DOC, COD, TKN and oil and grease; conductivity and Cl. (3) Total metals, COD and DOC were generally well correlated with mass first flush, suggesting that BMPs that treat the early portion of runoff have an opportunity to remove high concentrations of these pollutants.

Keywords Correlation analysis; highway stormwater runoff; mass first flush; storm characteristics

Introduction

The million miles of highway throughout the United States represent a known but unquantified non-point source (NPS) of pollution (Wu *et al.*, 1998), and have been identified as one of the leading causes of the degradation of the quality of receiving waters. Highway runoff pollution recently has been identified as an important source of micro-pollutants such as heavy metals, hydrocarbons and fuel additives (Barrett *et al.*, 1998; Furumai *et al.*, 2002). These constituents are generated mainly from traffic activities, component wear, fluid leakage, pavement degradation, roadway maintenance and atmospheric deposition (Sansalone and Buchberger, 1997a,b; Shinya *et al.*, 2000).

In an effort to manage a watershed, the impact of highway runoff must be treated as one of the most important components among pollutant sources (Wu *et al.*, 1998). Accurate knowledge of the quantity and quality of runoff is required to assess the impacts of runoff on the environment and to develop appropriate mitigation technologies (Barrett *et al.*, 1998). In addition, knowing the correlation among stormwater pollutants may reduce the huge costs associated with monitoring and hence will assist in evaluating best management practices (BMPs) by using pollutants that may be accompanied by similar correlations in removal rates by BMPs. This paper reports a large quantity of highway stormwater runoff data and characteristics collected in west Los Angeles over the past three years (2000–2003). Correlations among pollutants and storm and site characteristics were examined. The information gained from this correlation analysis may assist transportation agencies to reduce the number of constituents that they are required to monitor, and may be used to evaluate the performance of BMPs.

Methods

Site description

Three monitoring sites in west Los Angeles, California, were selected with catchment areas ranging from 0.39 to 1.69 hectares and annual average daily traffic (AADT) of over 260,000 cars per day. These three sites were chosen as typical small catchment sites with heavy traffic load. All sites were equipped with American Sigma (Loveland, Colorado) Ultrasonic 950 Area-Velocity flow meters, tipping bucket rain gauges and composite auto samplers. Table 1 summarizes the three site descriptions and Figure 1 shows their locations.

Sample collection and analysis procedure

In general, five grab samples were collected every 15 minutes in the first hour after the start of detectable runoff. After the first hour, samples were taken at one-hour intervals for the following 7 hours, providing a total of 12 grab samples. For events lasting longer than 8 hours, one or two additional grab samples were collected. The number of the samples collected depended on the storm duration. Table 2 lists basic statistics of all the sampling events conducted between October 2000 and April 2003. A flow weighted composite sample was collected by the American Sigma equipment, which covered the duration of each storm.

All the samples were transported back to the laboratory at UCLA after collection and refrigerated at 4°C until analyzed. This was done to facilitate certain analysis, such as filtration for soluble/total metals and particle size distribution, which must be performed very soon after sample collection. Generally, the samples were transported to UCLA after the first hour and two more times during the following 7 hours. A composite sample was collected after the event.

Correlation analysis

The correlation analysis was conducted using data sets, collected over the 2000–2001, 2001–2002, and 2002–2003 wet seasons for 18, 21 and 23 storm events at three highway sites. Correlations were performed on data from individual sites as well as for the combined data.

In addition, statistical and graphical correlation analysis of the mass first flush ratios (MFF = normalized pollutant mass divided/normalized runoff volume) developed by Ma *et al.* (2002) with storm characteristics was made to determine if the first flush is related to site or storm characteristics. Seven parameters were selected as storm characteristics and MFF was analyzed at five points (10 to 50% by 10% increments). The MFF ratios were also ranked by parameter, and divided into four groups with similar correlation coefficients.

Results

Water quality data collected

The average number of observations for each parameter or constituent was approximately 160 per site and 500 for the combined sites. A total of 44 water quality parameters/

Table 1 Site description summary

Site ID	Location	AADT (Vehicles/day)	Catchment area (hectares)	Number of lanes (each direction)	Approximate impervious (%)
7-201	Hwy 101, Van Nuys	328,000	1.28	6	100
7-202	Hwy 405, Getty Center Exit	260,000	1.69	5	95
7-203	Hwy 405, Santa Monica Blvd.	322,000	0.39	5	100

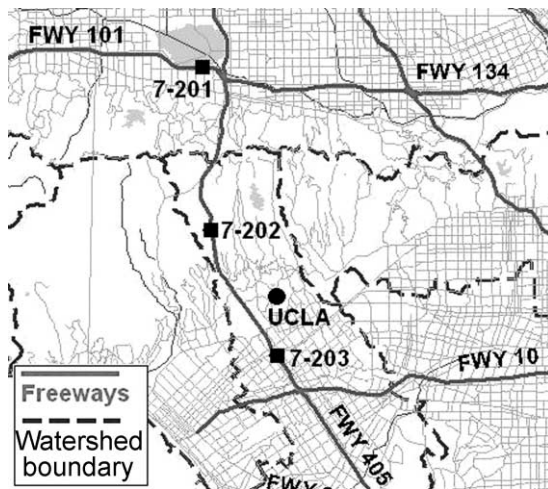


Figure 1 Highway runoff sampling sites

constituents comprising conventional, metals (particulate and dissolved), nutrients, fecal and total coliform, and organics including oil and grease were measured. Table 3 shows the basic statistics of principal constituents among data collected for all sites.

Correlation analysis among constituents

There is a general belief that many stormwater pollutants are sorbed to the surfaces of suspended solids, and previous researchers have correlated metal elements to total suspended solids in highway stormwater runoff (Sansalone and Buchberger, 1997a; 1997b; Shinya *et al.*, 2000). Heavy metals except for nickel showed strong correlation to suspended solids (Shinya *et al.*, 2000).

Table 4a shows correlation among dissolved organic carbon (DOC), total Kjeldahl nitrogen (TKN), chemical oxygen demand (COD) and TSS; Table 4B shows the correlation among metals and TSS. The numbers above the diagonal are Pearson's coefficients, r , and the numbers below the diagonal are probability or P values. TSS was poorly correlated to DOC, COD and TKN, but the DOC, COD and TKN were all well correlated.

Table 2 Basic statistics of storm events

		Total rainfall (mm)	Max. intensity (mm/h)	Antecedent dry day	Storm duration (h)	Ave. rainfall intensity (mm/h)
7-201	No. of storm events	18	18	18	18	18
	Min./Max.	2.0/127.0	3.0/51.8	2.0/69.4	2.0/47.5	0.6/10.7
	Median/Mean	18.3/28.3	15.2/20.3	13.7/19.6	8.6/10.7	1.9/3.0
	Standard Dev.	30.5	17.8	17.5	10.3	2.7
7-202	No. of storm events	21	21	21	21	21
	Min./Max.	1.8/156.0	3.0/61.0	1.0/192.2	0.9/46.5	0.2/11.3
	Median/Mean	19.1/26.1	12.2/17.3	19.8/29.0	6.0/9.3	2.0/3.2
	Standard Dev.	34.9	16.3	41.0	10.0	3.3
7-203	No. of storm events	23	23	23	23	23
	Min./Max.	1.5/128.5	6.1/39.6	0.3/192.3	1.4/47.1	0.2/8.9
	Median/Mean	19.8/28.5	21.3/21.1	14.2/26.4	6.9/9.9	2.5/3.3
	Standard Dev.	34.5	11.2	39.9	9.7	2.6
Combined sites	No. of storm events	62	62	62	62	62
	Min./Max.	1.5/156.0	3.0/61.0	0.3/192.3	0.9/47.5	0.2/11.3
	Median/Mean	19.4/27.6	15.2/19.6	16.3/25.3	7.2/9.9	2.2/3.2
	Standard Dev.	33.0	15.0	35.0	9.8	2.8

Table 3 Basic statistics of principal EMCs calculated for combined sites

Parameters	No. of cases ¹	Mean	EMC/Grab ² (Mean, %)	Median	EMC/Grab ³ (Median, %)	Minimum	Maximum	Standard dev.
TSS (mg/L)	62/569	67.7	94.9	57.6	125.4	8.8	466.4	63.0
Turbidity (NTU)	62/569	46.8	89.9	33.1	103.8	10.9	170.5	39.2
COD (mg/L)	62/569	252.5	78.6	119.8	86.5	19.1	2,282.8	372.8
DOC (mg/L)	62/569	66.9	82.8	28.9	97.5	2.9	848.8	127.0
Oil & Grease (mg/L)	62/569	14.0	77.4	8.8	83.2	1.5	80.2	14.6
TKN (mg/L)	62/569	9.6	82.8	4.1	87.8	0.8	111.3	16.4
NH ₃ -N (mg/L)	62/569	4.6	84.1	1.4	105.6	0.0	65.0	9.7
NO ₃ -N (mg/L)	62/569	2.7	83.3	1.3	90.9	0.3	34.7	5.3
Total P (mg/L)	62/566	0.9	98.0	0.4	85.6	0.1	8.2	1.3
Total Cd (μg/L)	46/362	1.8	86.0	1.1	100.2	0.4	20.2	3.0
Total Cr (μg/L)	62/564	9.7	92.8	8.7	105.5	2.3	40.1	6.3
Total Cu (μg/L)	62/564	92.9	83.5	55.7	88.1	15.9	920.8	125.2
Total Ni (μg/L)	62/563	20.0	86.0	11.2	87.7	2.4	253.7	33.9
Total Pb (μg/L)	62/562	25.8	105.8	22.9	121.1	4.6	151.6	20.5
Total Zn (μg/L)	62/564	506.3	90.2	267.9	100.4	83.3	8,881.3	1,137.0
Diss. Cd (μg/L)	43/363	1.4	90.4	0.6	93.6	0.5	17.8	2.7
Diss. Cr (μg/L)	58/566	2.8	89.9	2.0	96.6	0.5	19.3	2.8
Diss. Cu (μg/L)	62/566	65.9	79.0	35.4	91.9	5.2	735.3	103.9
Diss. Ni (μg/L)	62/565	15.7	83.4	7.9	91.0	0.8	229.2	31.3
Diss. Pb (μg/L)	47/564	4.9	110.7	3.6	145.6	0.5	43.4	6.5
Diss. Zn (μg/L)	62/566	415.3	89.5	178.2	97.2	42.3	8,150.0	1,055.7

¹Number of events or EMCs over total number of grab samples²Arithmetic mean of the EMCs divided by mean of grab samples³Arithmetic median of the EMCs divided by median of grab samples

TSS showed a relatively good correlation to particulate metals except for Cd. Among particulate metals, Cu-Ni and Cu-Zn were strongly correlated. The correlation results for all three sites are summarized as follows:

- Very strong correlations ($r \geq 0.90$): TSS-VSS, COD-DOC, total P-diss. P, total Cd-total Zn, total Cd-diss. Cd, total Cd-diss. Zn, total Cu-total Ni, total Cu-diss. Cu, total Cu-diss. Ni, total Ni-diss. Cu, total Ni-diss. Ni, total Pb-particulate Pb, total Zn-diss. Zn, diss. Cd-total Zn, diss. Cd-diss. Zn, and diss. Cu-diss. Ni.

Table 4 Correlation analysis results among non-metals and TSS and particulate metals

(a) Non-metals							
Para.	TSS	COD	DOC	TKN			
TSS	1	0.40	0.34	0.40			
COD	0	1	0.92	0.84			
DOC	0	0	1	0.81			
TKN	0	0	0	1			
(b) TSS and particulate metals							
Para.	TSS	Part. Cd	Part. Cr	Part. Cu	Part. Ni	Part. Pb	Part. Zn
TSS	1	0.52	0.59	0.60	0.58	0.60	0.61
Part. Cd	0	1	0.72	0.78	0.70	0.68	0.76
Part. Cr	0	0	1	0.78	0.72	0.65	0.70
Part. Cu	0	0	0	1	0.85	0.70	0.83
Part. Ni	0	0	0	0	1	0.70	0.75
Part. Pb	0	0	0	0	0	1	0.74
Part. Zn	0	0	0	0	0	0	1

- above the diagonal: Pearson's coefficient " r "- below the diagonal: Probability values (P -value)

- Strong correlations ($r \geq 0.80$): Conductivity-hardness, COD-oil & grease (O&G), O&G-TKN, DOC-NH₃-N, cond.-Cl, TKN-Cl, cond.-SO₄, hard.-SO₄, total Cd-total Cu, total Cd-total Ni, total Cd-diss. Cu, total Cd-diss. Ni, total Cr-particulate Cr, total Cu-diss. Cd, total Cu-diss. Zn, total Ni-diss. Cd, total Ni-diss. Zn, total Zn-diss. Cu, total Zn-diss. Ni, total Cu-total Zn, total Ni-total Zn, diss. Cd-diss. Cu, diss. Cd-diss. Ni, diss. Cu-diss. Zn, and diss. Ni-diss. Zn.

Table 5 shows the summary statistics for particulate-bound metals. In general, Cr and Pb are in particulate-bound phase and Cu, Ni and Zn are more associated with the dissolved phase.

Mass first flush and correlation analysis

The concept of the first flush phenomenon was first advanced in the early 1970s. Bertrand *et al.* (1998) assumed that there is a significant first flush if at least 80% of the total pollutant mass is transported in the first 30% of the volume discharged during rainfall events. However, such instances are extremely rare and found in only 1% of the events (Lee *et al.*, 2003). Ma *et al.* (2002) previously defined the mass first flush ratio, which describes the fractional mass of pollutants emitted as a function of storm duration. It is defined mathematically as follows:

$$MFF_n = \frac{\int_0^{t_1} C(t)Q(t)dt}{\frac{\int_0^{t_1} Q(t)dt}{V}M} \quad (1)$$

For a mass first flush to exist (e.g. $MFF_n > 1$), the concentration must not only be greater during the early part of the storm, but the mass emissions must also be greater. The ratio allows convenient characterization of the first flush, and allows the first flush to be analyzed statistically. For example, $MFF_{20} = 2.5$ means that 50% of mass load is contained in the first 20% of the runoff. Bertrand *et al.* (1998)'s definition can be described as $MFF_{30} > 2.66$.

Figure 2 shows notched bar plots of the MFF ratios for COD, TSS and six total metals for the combined sites. The ranges of MFF_{10} and MFF_{20} by median are 1.250 to 2.511 and 1.232 to 1.897. This suggests that there is a first flush, although the magnitudes are less than suggested by Bertrand *et al.* (1998). Pb and Cr had the lowest MFF ratios, both of which are shown to be particulate-bound. The value of knowing the first flush is quantifying the additional removal affected by BMPs that can completely capture the early runoff.

In order to assess the runoff characteristics and to compare MFFs for each constituent, the MFFs were ranked by median values. The results are shown in Table 6. In general, TSS, COD, DOC, TKN and NH₃-N had high MFF values, whereas NO₂-N, dissolved Cd, total Cr, dissolved Cr and total Pb had low values. Site 7-203 had higher MFF values than the other sites. Site 7-203 is smaller than the areas of other sites, with shorter rainfall retention time.

Correlation analysis among storm characteristics and MFF_n

Previous researchers have noted interactions between storm characteristics and first flush. Furumai *et al.* (2002) found a stepwise washoff phenomenon, and the wash-off process did not appear to be linear with the runoff rate and to the mass of pollutant. Gupta and Saul (1996) noted that the maximum rainfall intensity, maximum inflow, rainfall duration, and the antecedent dry weather period were the most important parameters influencing first flush.

Table 5 Summary statistics for particulate-bound metals

		7-201	7-202	7-203	Combined			7-201	7-202	7-203	Combined
No. of cases	Cd	85	147	130	362	Standard dev.	Cd	19.1	31.6	33.7	30.0
	Cr	172	192	200	564		Cr	17.8	15.3	15.5	16.3
	Cu	172	192	200	564		Cu	15.8	20.6	22.8	20.8
	Ni	171	192	200	563		Ni	17.0	22.4	24.6	22.2
	Pb	170	192	200	562		Pb	18.2	15.1	18.7	17.8
Minimum	Zn	172	192	200	564	Maximum	Zn	20.6	26.1	19.1	22.6
	Cd	6.9	0.0	5.0	0.0		Cd	100.0	100.0	100.0	100.0
	Cr	0.2	16.0	30.2	0.2		Cr	100.0	100.0	100.0	100.0
	Cu	0.2	2.3	2.1	0.2		Cu	91.5	84.0	85.3	91.5
	Ni	0.3	0.9	1.4	0.3		Ni	97.5	87.5	90.6	97.5
Median	Pb	23.2	25.3	7.5	7.5	Mean	Pb	100.0	100.0	99.5	100.0
	Zn	0.8	1.2	0.3	0.3		Zn	92.1	93.4	78.7	93.4
	Cd	50.0	50.0	50.0	50.0		Cd	53.4	48.3	52.3	50.9
	Cr	72.5	74.8	75.0	74.3		Cr	68.8	73.8	74.5	72.5
	Cu	26.4	42.4	25.7	31.3		Cu	29.6	42.0	32.7	34.9
	Ni	22.0	31.9	28.8	27.6		Ni	25.7	35.4	35.3	32.4
	Pb	91.7	92.7	81.7	88.1		Pb	84.4	86.6	77.1	82.6
	Zn	22.0	33.6	19.6	24.1		Zn	27.7	36.5	25.3	29.8

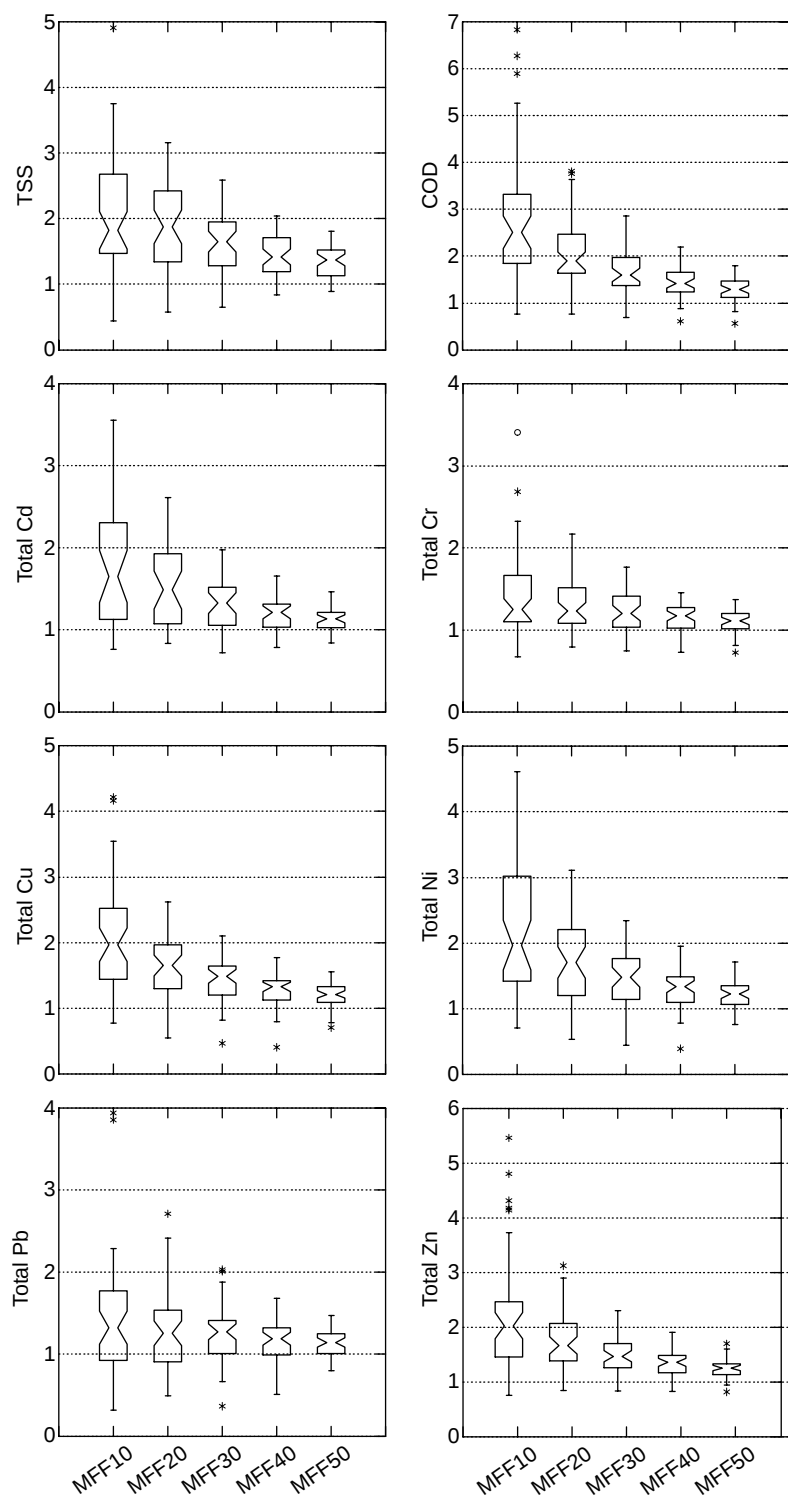


Figure 2 MFF plots for combined sites

Table 6 Ranking of MFF₂₀ by median value for all sites

Rank	7-201		7-202		7-203		Combined sites	
	Parameters	Median	Parameters	Median	Parameters	Median	Parameters	Median
1	COD	1.84	PO ₄ -P	2.28	DOC	2.74	DOC	1.95
2	PO ₄ -P	1.77	Total Cd	2.04	Dissolved Ni	2.48	TKN	1.94
3	Total P	1.77	DOC	2.01	COD	2.41	COD	1.90
4	TSS	1.71	TKN	2.01	TKN	2.39	NH ₃ -N	1.88
5	Oil & Grease	1.60	Dissolved Zn	1.99	NH ₃ -N	2.26	TSS	1.87
6	Dissolved Ni	1.57	Dissolved P	1.98	Total Ni	2.17	Dissolved Ni	1.79
7	NH ₃ -N	1.55	Total Zn	1.97	Dissolved Cu	2.12	PO ₄ -P	1.77
8	Dissolved P	1.55	NH ₃ -N	1.96	TSS	1.98	Dissolved P	1.76
9	TKN	1.52	Dissolved Ni	1.94	Total Cu	1.96	Total P	1.75
10	Hardness	1.51	Dissolved Cu	1.91	Oil & Grease	1.91	Total Ni	1.70
11	Conductivity	1.48	Dissolved Pb	1.90	Total Zn	1.80	Oil & Grease	1.69
12	Total Zn	1.46	Dissolved Cd	1.90	Conductivity	1.79	Dissolved Cu	1.69
13	Dissolved Pb	1.46	COD	1.86	Dissolved P	1.79	Conductivity	1.68
14	DOC	1.43	Total Ni	1.84	Total P	1.76	Total Zn	1.67
15	Total Ni	1.41	TSS	1.69	Hardness	1.71	Total Cu	1.65
16	Total Pb	1.40	Oil & Grease	1.69	Dissolved Zn	1.66	Dissolved Zn	1.65
17	Total Cu	1.40	Total Cu	1.68	NO ₃ -N	1.63	Hardness	1.54
18	NO ₂ -N	1.36	Total P	1.62	PO ₄ -P	1.53	Dissolved Pb	1.50
19	Dissolved Cu	1.34	Turbidity	1.43	Total Cd	1.49	Total Cd	1.49
20	Dissolved Zn	1.31	NO ₃ -N	1.42	Dissolved Cd	1.39	NO ₃ -N	1.39
21	Turbidity	1.29	Hardness	1.40	NO ₂ -N	1.37	Dissolved Cd	1.37
22	Total Cr	1.27	Total Pb	1.39	Dissolved Pb	1.35	NO ₂ -N	1.36
23	Total Cd	1.19	Conductivity	1.37	Total Cr	1.26	Turbidity	1.29
24	Dissolved Cr	1.15	Dissolved Cr	1.35	Total Pb	1.24	Total Pb	1.26
25	Dissolved Cd	1.06	NO ₂ -N	1.25	Turbidity	1.22	Total Cr	1.23
26	NO ₃ -N	0.94	Total Cr	1.19	Dissolved Cr	1.16	Dissolved Cr	1.19

To determine the effect of storm and site characteristics on first flush, statistical and graphical correlation analyses of the MFF with storm characteristics and other constituents were made. Seven parameters were selected as storm characteristics: total event rainfall; rainfall intensities (max. 5 min. and 15 min., and average); antecedent dry days (ADD); antecedent rainfall, and rainfall duration. Almost no correlations existed between MFF and storm characteristics, with the largest values of R being about 0.5. Table 7 shows the correlation among TSS, COD, DOC and the total forms of the six previously discussed metals. The table shows that the constituents are generally well correlated, suggesting that BMPs that can treat the first flush have an opportunity to remove all six metals and TSS, COD and DOC.

Table 7 Correlation among MFF₂₀ Ratios for TSS, COD, DOC and the total metals

	TSS	COD	DOC	Tot. Cd	Tot. Cr	Tot. Cu	Tot. Ni	Tot. Pb	Tot. Zn
TSS	1	0.23	0.14	0.13	0.45	0.18	0.20	0.57	0.16
COD	0.12	1	0.87	0.53	0.47	0.79	0.73	0.08	0.70
DOC	0.36	0.00	1	0.53	0.38	0.85	0.77	0.02	0.73
Tot. Cd	0.45	0.00	0.00	1	0.37	0.67	0.68	0.28	0.79
Tot. Cr	0.00	0.00	0.01	0.03	1	0.65	0.45	0.60	0.43
Tot. Cu	0.24	0.00	0.00	0.00	0.00	1	0.93	0.20	0.84
Tot. Ni	0.20	0.00	0.00	0.00	0.00	0.00	1	0.23	0.87
Tot. Pb	0.00	0.62	0.91	0.11	0.00	0.19	0.13	1	0.24
Tot. Zn	0.31	0.00	0.00	0.00	0.00	0.00	0.00	0.12	1

- above the diagonal: Pearson's coefficient "*r*"

- below the diagonal: Probability values (*P*-value)

Conclusions

Three years of highway runoff data were examined to determine the event mean concentrations of the pollutants normally associated with highway runoff. Strong correlations were found among many pollutants. TSS was poorly correlated to DOC, COD and TKN, but the DOC, COD and TKN were all well correlated. TSS showed a relatively good correlation to particulate metals. Among particulate metals, Cu-Ni and Cu-Zn were strongly correlated. Cr and Pb were primarily particulate-bound (72.5% and 82.6% particulate) and Cu, Ni and Zn were more associated with the dissolved phase (34.9%, 32.4% and 29.8% particulate).

Highway runoff also showed a first flush, with median MFF₁₀ and MFF₂₀ ratios ranging from 1.3 to 2.5 and 1.2 to 1.9, respectively. TSS, COD, DOC, TKN and NH₃-N had the greatest MFF ratios, whereas NO₂-N, dissolved Cd, total Cr, dissolved Cr and total Pb had the lowest values. The MFF ratios for total metals, COD and DOC were highly correlated except for Pb, suggesting that a BMP that treats the early runoff has an opportunity to treat high concentrations of all eight pollutants.

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