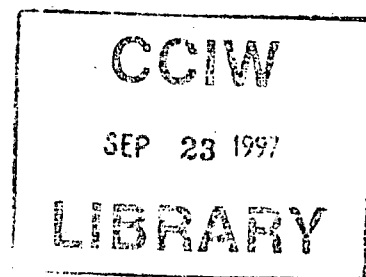


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Heavy Metals and PAHs in Highway Bridge Runoff

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MANAGEMENT PERSPECTIVE

Surface runoff has been long recognized as a significant source of contaminants pertinent to land use activities and other local environmental conditions. The level of runoff pollution is particularly significant in the case of highways, where operation of motor vehicles, and wear and maintenance of highway structures produce such toxic contaminants as heavy metals and hydrocarbons (particularly polycyclic aromatic hydrocarbons, PAHs).

The report that follows provides characterization of highway bridge runoff (average daily traffic 92 000 vehicles) with respect to selected heavy metals and PAHs, and thereby attempts to fill a void in the Canadian environmental literature.

The data presented should be of interest to water managers and environmental planners dealing with receiving waters impacted by highway or road runoff, and environmental scientists interested in sources and pathways of heavy metals and PAHs, and their effects in receiving waters.

Heavy metals and PAHs in highway bridge runoff

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Abstract

Highway bridge runoff from Burlington, Ontario was characterized with respect to five heavy metals (Zn, Pb, Ni, Cu, Cd) and 14 polycyclic aromatic hydrocarbons (PAHs). The highest mean metal concentrations in whole-water samples were observed for zinc, copper, and lead (0.337, 0.136 and 0.072 mg/L), and were comparable to literature values, except for low lead concentrations which reflect the recent use of unleaded gasoline. Mean PAH concentrations in whole-water samples ranged from 0.015 to 0.5 µg/L. Cu, Ni and Zn concentrations in filtered samples accounted for 35–45% of those in whole-water samples. PAHs concentrations in filtered samples represented less than 11% of those in whole-water samples. Runoff sediment contained high mean concentrations of zinc, copper and lead (997, 314, and 402 µg/g) and was “grossly polluted” according to Ontario guidelines for sediment quality. Concentrations of metals in fine-grained sediment (< 45 µm) were greater than in whole sediment samples, but enrichment was insignificant in terms of total loads. The runoff chemistry indicates that uncontrolled discharges of highway bridge runoff could significantly impact receiving water quality, and may require remediation by such stormwater best management practices as water quality inlets, grassed ditches, vegetation strips, detention ponds and wetlands.

Keywords: highway bridge runoff, sediment, heavy metals, polycyclic aromatic hydrocarbons

Introduction

Urban stormwater is a significant source of contaminants which may impact receiving waters in a number of ways. Two of such impacts are particularly important, impacts on aquatic life (physical impacts, thermal enhancement, and toxicity) and potential impacts on human health, including contamination of recreational waters or drinking water (Lijklema et al. 1993; Makepeace et al. 1995). Stormwater generation and contamination is particularly intense in areas which are characterized by high productions of runoff and associated pollutants. Highways and the associated land corridors are primary examples of such areas and hence represent important sources of contaminated stormwater (Kobringer et al. 1982, M. M. Dillon 1990).

Highway runoff usually contains various classes of pollutants, such as solids (including fine particulates), nutrients (e.g. nitrate and ammonia), deicing agents (mostly road salt), heavy metals, and petroleum hydrocarbons (including polycyclic aromatic hydrocarbons (PAHs)) (Thompson et al. 1995). Two of these groups, heavy metals and PAHs, are especially important because of their toxic impacts on receiving waters (Lijklema et al. 1993).

Heavy metals commonly found in highway runoff include zinc, lead, iron, copper, cadmium, chromium, nickel and manganese (Kobringer et al. 1982), and are all related to automobile operation, or abrasion and corrosion of highway structures, or maintenance procedures (Kobringer et al. 1982; M. M. Dillon 1990). The most important source of PAHs in highway runoff is leaking automobile crankcases; tailpipe emissions are a less significant contributor (Hunter et al. 1979). PAHs tend to concentrate in soils and sediments, and only a small fraction of their total load is in the aqueous phase and generally considered bioavailable (Hoffman et al. 1985; Stotz 1987).

In spite of a large volume of data on highway runoff contamination in European and United States literature (Frazer 1990), relatively little information is available from Canada. In fact, in a recent Canadian study of highway stormwater runoff quality, Thompson et al. (1995) listed existing North American databases and not one of them was from Canada, indicating a total lack of such information. A similar finding was made earlier by Frazer (1990) in his survey of about 20 U.S. and European sources of data on highway runoff. The only Canadian data the authors found were those pertaining to several samples collected by M. M. Dillon Ltd. in connection with their literature survey (M. M. Dillon 1990). Yet, such information is of interest because of specific climate, traffic densities, and vehicle specifications and operation typical for Canadian conditions. Consequently, a study of highway bridge runoff was undertaken with the objective of characterizing runoff composition with respect to selected heavy metals and PAHs. The emphasis in this paper was placed on general characterization of runoff composition, which should provide background information for the ongoing studies of highway runoff toxicity (Dutka et al. 1996).

Study Area and Methods

A field study was undertaken at two adjacent sites along the Queen Elizabeth Way (QEW) highway in Burlington, Ontario, over a period of 16 months spanning all seasons. The sampling points were located at the north foot of the James N. Allan Burlington Bay Skyway Bridge (further referred to as the Skyway Bridge), with one set of samples collected for the southbound (up hill) traffic and the other one for the north bound (down hill) traffic.

Samples were collected from the bridge deck drains; discharged runoff normally falls to the ground below the bridge. The QEW highway at this location is paved with asphalt, comprises four lanes in each direction, and the length of the runoff-contributing highway section is about 500 m. Traffic on the bridge is characterized by fairly high volumes, described by the Annual Average Daily Traffic (i.e. the average twenty four hour, two way traffic for the period January 1 to December 31) at 92,000. This volume increases in the summer (July 1 - August 31) to 102,100 and decreases to 83,700 in the winter (December 1 - March 31).

Runoff samples were collected by custom made fluidic devices, comprised of a stainless steel flow splitter, formed by a cylinder fitted with 16 openings ($D = 12$ mm) at the bottom.

Each of these openings conveyed 1/16 of the inflow. Outflow from one of the openings was then directed to the second stage splitter of similar design; outflows from the remaining 15 openings were wasted. Thus, during each event, it was possible to collect a constant fraction of the flow and accumulate it in a large sampling bottle (5 L). These bottles were also fitted with an overflow, which after the first bottle was filled, was collected in the second (overflow) bottle. These samplers were attached to the downspouts of the bridge deck drains and allowed effective, inexpensive, and vandal-proof sampling of highway bridge runoff. Samplers were inspected after individual events, cleaned as required, and the samples were removed and submitted for chemical analyses. Individual events were further documented by the rainfall depth and the duration of the antecedent dry weather period.

To address the treatability of the runoff, it was of interest to determine partitioning of pollutant loads between the dissolved and particulate phases, and for the particulate phase, load fractionation with respect to particle sizes. Selected whole-water samples were filtered ($D_{\text{pore}} = 0.45$ μm), and both filtered and unfiltered samples were submitted for metal and PAH analyses. For sediment samples, the finest fraction ($D < 45$ μm) was extracted from selected samples and analyzed separately. Small quantities of sediment in this fine fraction prevented PAH analyses in all cases but one.

All chemical analyses were performed by the National Laboratory for Environmental Testing (NLET) in Burlington, Ontario. Total concentrations of Cd, Cu, Ni, Pb and Zn in whole-water samples were determined by atomic absorption spectrometry which followed solvent extraction by ammonium pyrrolidine dithiocarbamate and methyl isobutyl ketone. The detection limits were 0.01 mg/L for Cd, Cu, Ni, and Zn; and 0.05 mg/L for Pb. Total metals in sediments were extracted by digestion with a mixture of hydrofluoric, nitric and

perchloric acids in a sealed Teflon bomb at 140 C for 3.5 hours, with concentrations determined by subsequent atomic absorption spectrometry. The detection limits were 10 µg/g for Cd, Cu, Ni and Zn and 50 µg/g for Pb.

Whole-water samples were analyzed for a suite of 14 PAHs (listed in next Section) by liquid/liquid extraction followed by gas chromatography. A 1 litre water sample was extracted with methylene chloride (CH₂Cl₂), and the extract dried, concentrated, cleaned up, and fractionated on a 3% (w/v) water-deactivated silica gel column. The residue was then reconcentrated to a final volume of 1 mL prior to analysis. Gas-liquid chromatography with a mass selective detector was used to quantify the PAH compounds utilizing a 30m x 0.25 mm fused silica RTX-5 column. The detection limits for individual PAH compounds ranged between 0.19 ng/L for fluorene and acenaphthylene and 0.5 ng/L for benzo(b)fluoranthene. For PAHs in sediments, 10 grams of sediment was extracted by sonification using 1:1 acetone / hexane. The extracts were then back extracted to CH₂Cl₂ and concentrated, cleaned up and quantified by the same procedures as for whole-water samples. Detection limits ranged from 10 ng/g for indene and other low molecular weight PAHs to 30 ng/g for benzo(b)fluoranthene and other high molecular weight PAHs.

Results and Discussion

The runoff was collected as flow proportional composite samples and the concentrations determined represent Event Mean Concentrations (EMCs), which are commonly used in runoff quality characterization and control planning (U.S. EPA 1983). The EMCs were further processed by determining means of the whole data set (C_{mean}), and 10 and 90 percentile concentrations ($C_{10\%}$, $C_{90\%}$). The latter two concentrations describe the data variation about the mean in the format established in the U.S. Nationwide Urban Runoff Program (U.S. EPA 1983).

Some of the EMC data were censored (i.e. below the analytical detection limits) and this had to be accounted for in calculating the means of such data sets (El-Shaarawi 1989). For this purpose, it was assumed that the distribution of EMCs was lognormal throughout the full range of values. The distribution fitted to the detected data was then used to estimate the values of censored observations (using a plotting position formula), which were combined with uncensored data to estimate the mean. The assumption of lognormal distribution of EMCs was checked graphically by plotting probabilities of non-exceedance of EMCs. In these plots, the Bloom's (1958) plotting position formula was used in the form:

$$P_{(m+i)} = (m + i - 0.375) / (n + 0.25)$$

where n = total number of observations, m = number of censored observations, and i is the rank among uncensored observations in ascending order.

Examples of EMC distributions are given in Figs. 1 and 2. In Fig. 1, the three most ubiquitous urban metals are shown, Cu, Pb and Zn; with some of the lead concentrations falling below the detection limit. As demonstrated in Fig. 2, lognormal distributions also fit the PAH concentrations well, with a possible exception of pyrene.

Finally, the graphical assessment of lognormal distributions was complemented by probability correlation tests proposed by Stephens (1986) and Verill and Johnson (1988). In these procedures, the hypothesis that metal and PAH data follow the lognormal distribution could not be rejected at 0.05 level of significance in all cases except for copper, where the corresponding level was 0.01 (Lawal 1996). The robust lognormal probability regression method was used to estimate basic statistical parameters of the EMC sets, including the mean, and the 10th and 90th percentiles.

For assessing the treatability of highway runoff, it was of interest to estimate the dissolved and suspended components of metals and PAHs in runoff. This issue was addressed in two ways: firstly by examining correlation between suspended solids (SS), determined as total suspended solids concentrations, and the major constituents studied, and secondly by filtration of selected samples. Correlation coefficients determined through this analysis are presented in Table 1.

The data in Table 1 indicate that metals are poorly correlated with suspended solids, which can be explained by a significant dissolved load component, as further confirmed by tests on filtered samples. High correlations between SS and PAHs reflect the hydrophobic nature of PAHs and consistency of PAH concentrations in suspended particulates.

Concentrations of five heavy metals studied in the Skyway Bridge runoff are presented in Table 2 and Fig. 3.

The EMC data show fairly large dispersion about the mean; differences between 10 and 90 percentiles are an order of magnitude or more. The Skyway Bridge data are in good agreement with the U.S. and European data provided for comparison, with the exception of much lower Pb concentrations, which are indicative of environmental conditions after phasing lead out of gasoline. The U.S. and European lead data were of an older date and reflected much greater use of leaded gasoline.

The metal concentrations in the QEW runoff often exceeded those reported by MOEE for stormwater discharges along the Toronto Waterfront, and also far exceeded Ontario Provincial Water Quality Objectives, even at the 10 percentile levels for Zn and Cu. These findings indicate that highway runoff is contaminated by heavy metals to a greater degree than runoff from large urban areas.

Mean runoff PAH concentrations, as well as 10 and 90 percentile concentrations, are presented in Table 3. For comparison, this table also contains data from the MOEE studies

(Paul Theil Associates and Beak Consultants 1995, Maunder et al. 1995). PAH concentrations in Skyway Bridge runoff and the storm sewer outfalls along the Toronto Waterfront are comparable (Table 3; Fig. 4), with three possible exceptions, acenaphthene, benzo(b)fluoranthene and benzo(k)fluoranthene.

In the assessment of treatability of highway runoff, the partitioning of samples into the aqueous and solid phases using filtration ($D_{\text{pore}} = 0.45 \mu\text{m}$), brought the concentrations of Cd and Pb below the detection limit; average concentrations of Cu, Ni, and Zn ($N = 11$) were 0.047, 0.031 and 0.148 mg/L, respectively (Fig. 5). Thus, a significant fraction (35 - 45%) of whole-water Cu, Ni and Zn load is in the dissolved form and its removal from runoff would require a more complex treatment than settling alone. Suitable treatment facilities could include filters (particularly biofilters with metal uptake by biofilm) and subsurface flow wetlands (Anderson et al. 1997, Rochfort et al. 1997).

For PAHs, the concentrations in filtered samples dropped below the detection limits with three exceptions - phenanthrene ($C_{\text{mean}} = 22.2 \text{ ng/L}$, $N = 3$), pyrene ($C_{\text{mean}} = 49.8 \text{ ng/L}$, $N = 3$), and fluoranthene ($C_{\text{mean}} = 49.8 \text{ ng/L}$, $N = 3$) (Fig. 6). These levels represented less than 11% of those in whole-water samples, and were three to four orders of magnitude below the solubility levels of these compounds (1.18, 0.12, 0.24 mg/L, respectively) (Mackay et al. 1992; NWRI 1995). These limited observations clearly indicate the significance of the particulate phase in PAH transport and its implications for remediation of runoff pollution. Effective removal of suspended solids with adsorbed contaminants can be achieved by relatively common best management practices (BMPs) such as inlet control devices, settling (Schueler 1987), vegetation strips and grassed ditches (Ellis et al. 1994).

Sediments transported by runoff were also analyzed for metals and PAHs. Selected sediment samples were sieved and the finest fractions ($D_{\text{fine}} < 45 \mu\text{m}$) analyzed. Small quantities of sediment obtained this way allowed only analysis for metals. The results of metal analyses of the sediment are summarized in Table 4 and Fig. 7, together with the Ministry of the Environment and Energy Guidelines for the Protection and Management of Aquatic Sediment in Ontario (Ontario MOE 1992).

With the exception of Ni, the mean heavy metal concentrations in the runoff sediments are higher than the severe effect level (SEL) of the MOEE Guidelines (Ontario MOE 1992), indicating "gross pollution" of sediment that "will significantly affect" its use by benthic organisms. The metal concentrations were also substantially higher in fine sediment fraction ($<45 \mu\text{m}$) than in the unsieved sediments. This observation was statistically significant for Zn, Ni and Cu, at the 95% confidence level and was in agreement with the expected greater adsorption of heavy metals on finer particles with a larger surface area to volume ratio than coarser particles.

The concentrations of PAHs in sediment samples are presented for unsieved samples in Table 5. Small quantities of sediment in the fine fraction class did not allow for their

analysis for PAHs (except for one sample). The assessment of sediment PAH data against the MOEE guidelines was complicated by the fact that some of the compounds on the MOEE list (the guidelines refer to the sum of 16 EPA priority PAHs) were missing in the analytical protocol available for this study. Even with this limitation, it is obvious that the PAH data in Table 5 (the sum of 14 PAHs = 21 µg/g) would exceed the LEL guideline value (2 µg/g), but not the SEL value expressed as 11,000 µg of the total of 16 PAHs per g of organic carbon. In view of the high organic content of the runoff sediment (volatile suspended solids represented 40% of the total suspended solids), it is probable that the SEL value would not be exceeded.

Even though the concentrations of metals and PAHs in fine-grained sediment (< 45 µm) were greater than those in whole sediment samples, this enrichment was insignificant in terms of total loads, because the fine fraction represented less than 1% of the total sediment mass.

Overall, concentrations of metals and PAHs in runoff from the QEW highway bridge indicate an intermediate degree of contamination, with a significant presence of dissolved metals and severe contamination of sediment by metals.

Further assessment of the toxicity of highway runoff was done by comparing measured concentrations of individual PAHs with their acute toxicities in the Microtox™ or 48-hour Daphnia magna tests from the COMPUTOX™ database (NWRI 1995). At least on an individual basis, the concentrations of PAHs in Table 3 are two orders of magnitude or more lower than their acute toxicities, e.g., phenanthrene mean concentration of 0.4 µg/L, Microtox™ EC₅₀ 50 µg/L and 48-hour Daphnia magna acute toxicity of 690 µg/L. However, chronic effects are of concern since many PAHs and their alkyl homologs are mutagenic and several are carcinogenic such as the known, potent carcinogen, benzo(a)pyrene. The chemical characterization of highway runoff indicates potential chronic effects, with a low probability of acute effects. This observation was confirmed by preliminary results of the ongoing study of highway runoff ecotoxicity determined by a battery of bioassays (Dutka et al. 1996).

Summary and Conclusions

Runoff from the James N. Allan Burlington Bay Skyway Bridge, with the annual average daily traffic of 92,000, was characterized with respect to five heavy metals (Zn, Pb, Ni, Cu, Cd) and 14 PAHs. The highest mean metal concentrations in whole-water samples were observed for zinc, copper, and lead (0.337, 0.136 and 0.072 mg/L, respectively) and compared well with the older literature data from Europe and the USA, except for lead. Lower lead concentrations in Skyway Bridge runoff likely reflect the use of unleaded gasoline, whereas the older literature data from European countries and the USA are indicative of the conditions prior to phasing lead out of gasoline. The mean runoff

concentrations of all five metals also substantially exceeded the Ontario Provincial Water Quality Objectives. PAH concentrations in whole-water samples ranged from 0.015 to 0.5 µg/L, and were similar to those found in urban runoff discharged along the Toronto Waterfront. The presence of these two groups of pollutants (heavy metals and PAHs) in the bridge runoff is significant because of their potential toxic impacts on receiving waters.

The runoff treatability was addressed by partitioning the metals and PAHs into dissolved and particulate loads ($D_{\text{pore}} = 0.45 \mu\text{m}$). Concentrations of Cu, Ni and Zn in filtered samples accounted for 35 - 45% of those in whole-water samples. PAH concentrations in filtered samples were less than 11% of those in whole-water samples and fell below the detection limits with three exceptions, phenanthrene, pyrene and fluoranthene. The majority of the heavy metal and PAH pollutant load from the Skyway bridge runoff could therefore be removed by settling of suspended solids in common BMPs. Removal of dissolved metals would require a more complex treatment than settling alone.

The sediment in runoff samples contained high mean concentrations of zinc, copper and lead (997, 314, and 402 µg/g, respectively) and was classified as "grossly polluted" according to the Ontario MOEE guidelines for sediment quality. Concentrations of metals in fine-grained sediment ($< 45 \mu\text{m}$) were appreciably greater than those in whole sediment samples, but this enrichment was insignificant in terms of total loads, because the fine fraction represented less than 1% of the total sediment mass.

The results of this study are of major significance, as they fill the gap in data characterizing highway runoff in Canada, and provide important information necessary for selection of appropriate treatment. The runoff chemistry indicates that uncontrolled discharges of highway bridge runoff could significantly impact the receiving water quality, and may require remediation by stormwater best management practices where protection of sensitive receiving waters is needed.

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Table 1. Correlation between suspended solids and selected constituents

	Correlation coefficient					
	N = 53			N = 29		
	Cu	Pb	Zn	Phenanthrene	Pyrene	Fluoranthene
TSS	0.33	0.42	0.38	0.83	0.93	0.93

Table 2. Concentration of five heavy metals in highway bridge runoff and urban runoff (mg/L)

Metal	C_{10%}	C_{mean}^a	C_{90%}	U.S. data^b (mean)	European data^c (mean)	MOEE^d	MOEE^e	PWQO^f
Zn	0.059	0.337	0.775	0.644	0.507	0.19	0.15	0.030
Pb	0.001	0.072	0.166	0.670	0.286	0.046	0.057	0.005
Ni	0.004	0.069	0.177	0.040	0.041	0.012	0.01	0.025
Cu	0.023	0.136	0.277	0.065	0.114	0.16	0.045	0.005
Cd	0.0001	0.015	0.039	0.015	0.006	0.005	0.00094	0.0002

^aN = 53

^b Mean of mean concentrations reported in the U.S. literature, N varied from 8 to 29 for various metals; reported by Frazer (1990)

^c Mean of mean concentrations reported in the European literature, N varied from 4 to 12 for various metals; reported by Frazer (1990)

^d MOEE, Storm sewer outfalls, Etobicoke and Scarborough (Paul Theil Associates and Beak Consultants 1995)

^e MOEE, Storm sewer outfalls, The City of Toronto (Maunder et al. 1995)

^f Ontario Provincial Water Quality Objectives (MOEE 1996)

Table 3. Concentration of 14 PAHs in highway bridge runoff and urban runoff (ng/L)

PAH	C _{10%}	C _{mean} ^a	C _{90%}	MOEE ^b	MOEE ^c
Indene	1.3	16.9	42.1		
2-Methylnaphthalene	3.8	74.3	217.5		112
1-Methylnaphthalene	3.0	39.1	101.2		58
Acenaphthylene	1.5	14.7	35.9	18.0	12
Acenaphthene	1.5	24.8	62.5	96.0	
Fluorene	3.4	56.2	141	108	
Phenanthrene	8.3	397	955	550	
Pyrene	31.8	454	1020	615	186
Fluoranthene	14.2	504	1320	782	500
Benzo(b)fluoranthene	3.7	23.9	55.8	553.0	119
Benzo(k)fluoranthene	2.3	135	344	570.0	
Benzo(a)pyrene	5.2	186	481	320.0	48
Indenopyrene	0.3	143	322	274.0	
Benzo(ghi)perylene	0.8	222	546	335.0	194

^a Skyway Bridge runoff (N = 29)

^b MOEE, Storm sewer outfalls, Etobicoke and Scarborough (Paul Theil Associates and Beak Consultants 1995)

^c MOEE, Storm sewer outfalls, The City of Toronto (Maunder et al. 1995)

Table 4. Concentration of metals in highway bridge runoff sediment ($\mu\text{g/g}$ dry weight)

Metal ^a	C_{mean} ^b	Standard Deviation ^b	$C_{\text{fine, mean}}$ ^c	MOEE Guidelines	
				LEL ^d	SEL ^e
Zn	997	528	1634	120	820
Pb	402	285	527	31	250
Ni	56	24	126	16	75
Cu	314	268	737	16	110

^a All Cd concentrations were below the detection limit $10 \mu\text{g/g}$

^b Unsieved samples, $N = 16$

^c Fine sediment fraction, $N = 5$

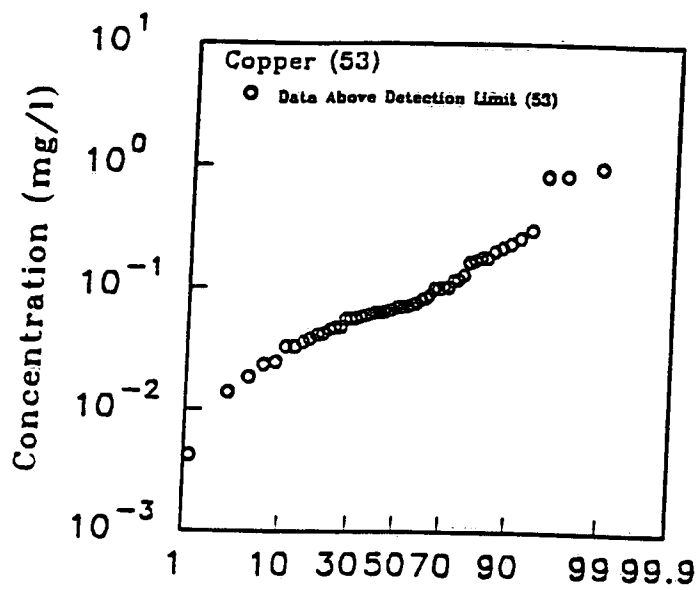
^d LEL = lowest effect level

^e SEL = severe effect level

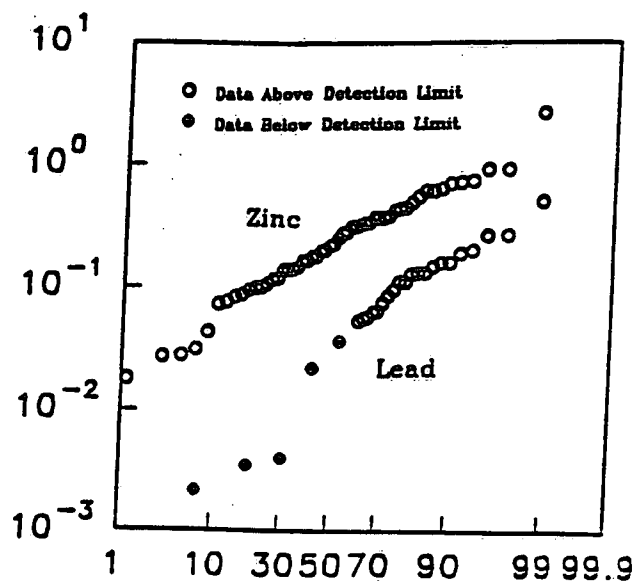
Table 5. Concentration of PAHs in highway bridge runoff sediment (ng/g)

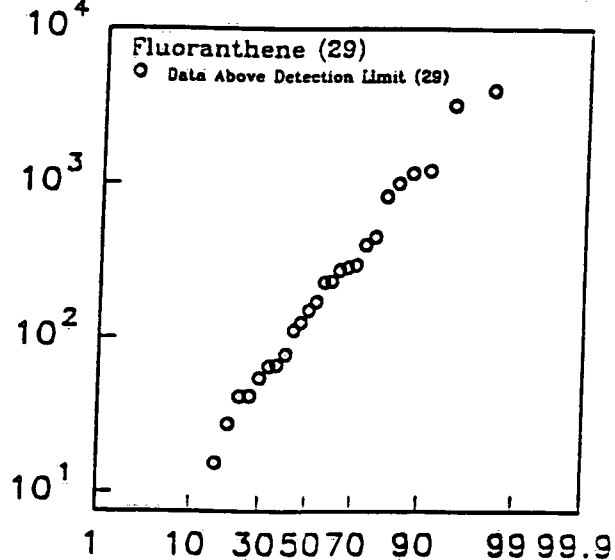
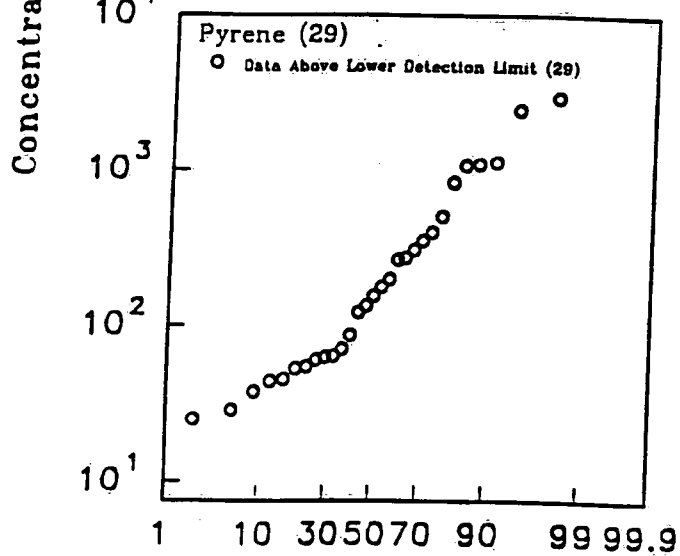
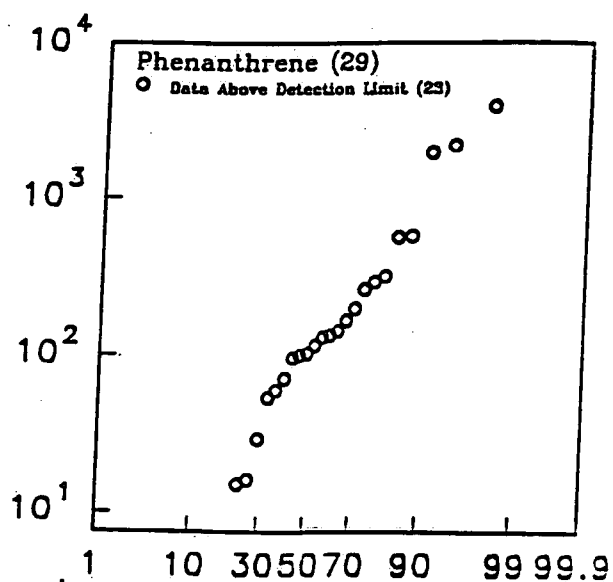
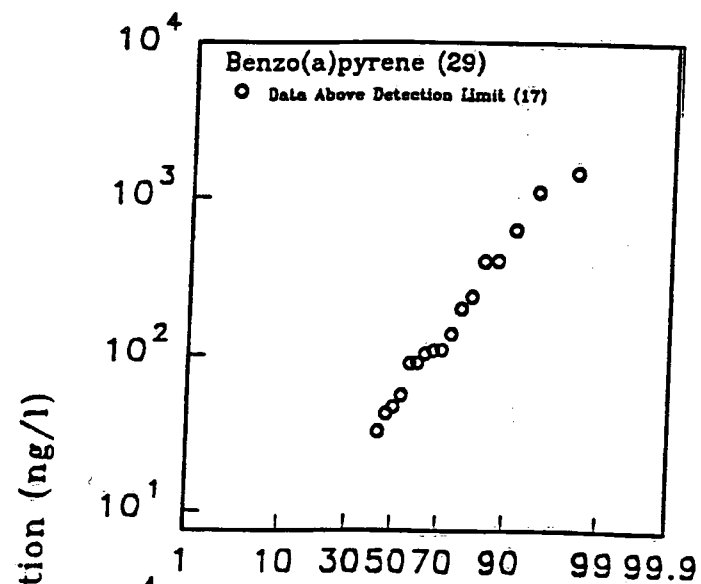
PAH	Mean concentration	Standard deviation
Indene	<10	
2-Methylnaphthalene	226	377
1-Methylnaphthalene	123	203
Acenaphthylene	71	130
Acenaphthene	251	302
Fluorene	517	738
Phenanthrene	4382	5032
Pyrene	3100	2889
Fluoranthene	4183	3839
Benzo(b)fluoranthene	1226	994
Benzo(k)fluoranthene	1596	1295
Benzo(a)pyrene	2106	1501
Indenopyrene	1248	927
Benzo(ghi)perylene	1647	1611

- Figure 1. Lognormal probability plot of three heavy metals in Skyway Bridge runoff**
- Figure 2. Lognormal probability plot of four PAHs in Skyway Bridge runoff**
- Figure 3. Concentration of metals in highway and urban runoff**
- Figure 4. Concentration of PAHs in highway and urban runoff**
- Figure 5. Effect of filtration on metal concentrations in QEW highway bridge runoff**
- Figure 6. Effect of filtration on PAH concentrations in QEW highway bridge runoff**
- Figure 7. Metal concentrations in QEW highway bridge runoff sediments**



Probability of Nonexceedance (%)





Probability of Nonexceedance (%)

