

Seasonal Variations of Ten Metals in Highway Runoff and their Partition between Dissolved and Particulate Matter

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Abstract Knowledge of differentiation of pollutants in urban runoff between dissolved and particulate matter is of great concern for a successful design of a water treatment process. Seasonal variations in pollutant load are of equal importance. Ten metals (Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn), as dissolved and particulate bound, was studied in the runoff from a major urban highway during a winter season and its following summer. Studded tyres and winter salting were expected to have an impact on the runoff water quality. The dissolved part of Al, Cd, Co, Cr, Mn and Ni was significantly higher in winter in comparison with summer ($p < 0.01$). For Fe, however, the dissolved part was lower during winter. No significant difference was found for Cu, Pb and Zn between the two seasons. The mass concentration (mg kg^{-1}) for all metals was significantly higher over the summer except for Al and Co, which showed a higher mass concentration during the winter. The concentration of selected metals vs. total suspended solids (TSS) showed a linear relationship ($r^2 > 0.95$) during winter runoff events except for Cd. A good correlation ($r^2 > 0.90$) was also found for the summer period for Al, Cu, Fe, Mn, Ni and Zn. It is suggested that the metal

pollutant load during winter could be assessed indirectly by measurement of TSS.

Keywords de-icing salt · Event Mean Concentration · metal pollution · total suspended solids · storm water

1 Introduction

Highway runoff is becoming recognised as the most contaminated of all the different kinds of storm water due to the loading from various sources related to vehicles, road construction material, and road management. A relationship exists between an increasing number of vehicles and a rise in the pollutant load (Barrett, Irish, Malina, & Charbeneau, 1998; Hvitved-Jacobson & Yousef, 1991). Hares and Ward (1999) recognised that a higher level of motorway-derived heavy metal contamination existed in runoff from a road section with an elevated average daily traffic density. Sansalone and Buchberger (1997) concluded that Zn, Cd and Cu were mainly dissolved whilst Pb, Fe and Al were to a greater extent, particulate bound. Other issues include seasonal variations, the impact of colder climates, and the influence of road maintenance such as de-icing, snow ploughing and the use of friction tyres.

Ramakrishna and Viraraghavan (2005) reviewed the environmental impacts of highway salting. Legret and Pagotto (1999) showed that the pollutant load could increase dramatically during the winter season

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when de-icing agents are utilised. Metals can be mobilized from roadside soils due to salty runoff from de-icing operations and transported with storm water (Amrhein, Strong, & Mosher, 1992). It has been recognised that the use of studded tyres is the main contributor to the wear of the asphalt surface (Jacobson, 1994; Jacobson & Hornwall, 1999). Traffic, its related activities and road management have been found to be a major contributor of pollutants in snow (Glenn & Sansalone, 2002; Reinosdotter & Viklander, 2005). The results of the previously mentioned study indicate a relationship between the dissolved fraction of metals and the concentration of suspended solids. Westerlund, Viklander, and Bäckström (2003) concluded that runoff from a snowmelt event displays a higher fraction of particulate bound metals than from a rain event.

Further research on many design parameters is needed to optimise the treatment abilities of the commonly applied sedimentation systems for road runoff. One of the criteria for a successful design of a storm water treatment process depends on the knowledge of the pollutants partition between dissolved and particulate matter. The focus should be furthermore on the occurrence of these contaminants and their seasonal variation as such knowledge could be used in future for optimisation of some treatment processes e.g., assessing the need for chemical flocculation.

The aim of this paper was to study highway runoff during winter and summer conditions with regards to (1) the dissolved fraction of metals ($\mu\text{g l}^{-1}$) and (2) the mass concentration (mg kg^{-1}) of particulate bound metals. We hypothesised that the metal content as a part of the total particulate matter should display significant seasonal variation. This is because the major contributor to the particulate matter is road wear erosion owing to a continuous traffic load and the use of studded tyres on vehicles in winter. We also hypothesised that a good correlation exists between metal and total suspended solids contents as metals are transported predominantly with the particulate matter.

2 Material and Methods

2.1 Study area

Runoff data was collected from a section of the European highway system E4 in the central area of Stockholm, Sweden. This section consists of a six-



Figure 1 Two pictures showing the area from that the runoff was collected from the European highway system E4 in Stockholm.

lane road with an asphalt surface (Figure 1). According to data obtained from the existing Motorway Control System (MCS), the total traffic load varied between 4,300 and 144,600 vehicles during time of sampling, while the average speed varied between 52 and 83 km/h, respectively (Table I). The Annual Average Daily Traffic (AADT) at this highway section is 120,000 and the speed limit is 70 km/h. The runoff from this road surface ($13,700 \text{ m}^2$) and also additional insignificant volumes of drainage water from a road tunnel is by gravity transported through a sewer system to a treatment plant at Fredhäll for sedimentation.

2.2 Experimental setup and sampling

The data collection programme was designed to obtain information on the quantity and quality of the highway runoff. Instruments for online measurements of suspended solids, electrical conductivity and flow were located at the end of the grit chamber before a Thompson weir in the Fredhäll treatment plant (Figure 2). An ABB programmable logic controller for process control (PLC, type AC800C), was used for continuous measurements (every 30 s) and data storage at the treatment plant. Suspended solids concentrations were measured using a Cerlic ITX

Table 1 Total number of vehicles and corresponding average speed preceding the runoff event i.e. antecedent dry period and traffic volume, vehicles per hour sampling and average speed during sampling

Date of runoff event	Vehicles during antecedent dry period (n)	Average speed during antecedent dry period (km/h)	Vehicles during sampling (n)	Vehicles per hour of sampling (n)	Average speed during sampling (km/h)
2004-12-14	1,075,600	77	11,400	7,050	52
2004-12-22–23	381,600	74	84,900	4,700	77
2005-01-20–21	380,000	76	144,600	6,200	76
2005-02-10	2,531,600	77	24,900	7,200	76
2005-02-14	235,800	79	55,000	7,750	71
2005-05-03	2,474,000	78	1,400	500	82
2005-06-04	510,200	75	7,700	4,550	77
2005-06-22	1,458,200	76	46,300	7,800	65
2005-08-26	1,305,400	77	4,300	851	83

suspended solids meter at a wavelength of 880 nm. Cleaning of the measuring probe was executed automatically every 30 min for duration of 6 s with compressed air. The Cerlic ITX instrument was calibrated against the received values from the laboratory-analysed TSS concentrations. A total of 99 samples were analysed in the range from 13 to 4,800 mg l⁻¹. The calibration curve had a correlation factor (r^2) of 0.93 (Figure 3).

The electrical conductivity of the water was measured with a Jumo dTransLf01 type 202540. The measuring range of conductivity was 0 to 2,000 mS m⁻¹ and the cell constant, K_c , was 1.00. Flow was measured using the Thompson weir and a Cerlic FLX pressure probe. The measuring range of the Cerlic FLX was 0 to 1 m. The flow range was chosen to be 0 to 60 m³ h⁻¹. A pump (CO/TECH 750) used for the water sampling was located adjacent

to the ITX measuring probe. The sampling pump and the material of the sampling loop were made of plastic (PVC). The water quality sampling was conducted with an ISCO 3700RF equipped with a refrigerator for storage of samples at a temperature of 4°C. Sampling started when the flow exceeded 1 m³ h⁻¹ and was carried out for every volume of 4 m³, except for one sampling event (14 December) when the sampling interval was 1 m³. Rinsing of the tubes connected to the sampler was performed regularly and before every sampling event to avoid mixing of consecutive samples. Sampling was selected for a total of nine runoff events when the treatment facility and its equipment were available. A Swedmeter submersible DS/mA pressure probe (with an operating range of 0 to 5 m and automatic temperature compensation) monitored the water level in the sedimentation basin.

A heated rain gauge with a capacity of 6 mm/min was located 10 m above the ground level in the central part of the study area. Every 0.2 mm of rain or melted snow was recorded and stored by the PLC.

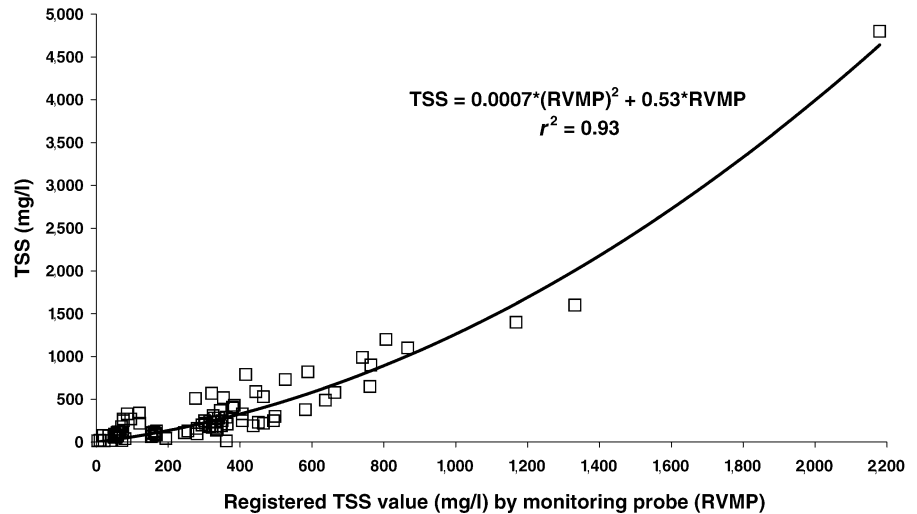
2.3 Analysis and data treatment

The water samples were made into a homogenous mixture by vigorous shaking of the ISCO sample bottle. Volumes of 250 ml were poured into acid-washed polyethylene (PE) bottles for the metal analyses and into 1,000 ml PE bottles for the analysis of TSS, conductivity and chloride. The samples were brought without delay for analysis at an accredited



Figure 2 The treatment plant for road runoff (1) is located under a bridge (2) (cf. Figure 1). The southern entrance of the Fredhäll tunnel is visible (3).

Figure 3 Calibration curve for continuous monitoring TSS.



laboratory (SWEDAC ISO/IEC 17025, Reg. number 1087). Metals were analysed by means of ICP-SFMS. Filtered samples (Sartorius 0.45 μm) were used for the analysis of soluble metals. Analyses of TSS, conductivity and chloride were carried out according to Swedish Standard SS-EN 872, SS-EN 27888 and SS-EN ISO 10304, respectively.

The difference level in the sedimentation basin was used for calculating the volume during each runoff event. The total mass load of TSS was calculated based on the calculated volume and measured TSS concentrations from the online measurement. Thus the EMC for TSS was found according to Eq. 1:

$$\text{EMC} = \frac{M}{V} \quad (1)$$

where M is the total mass of TSS during the runoff event and V is the total volume over the runoff event.

The metal concentration in the particulate matter Me_{part} was calculated according to Eq. 2 using a calculation base of 1 l volume:

$$\text{Me}_{\text{part}} = \frac{\text{Me}_{\text{total}} - \text{Me}_{\text{dissolved}}}{\text{TSS}_{\text{analysed}}} \quad (2)$$

where Me_{total} is the total concentration of metal, $\text{Me}_{\text{dissolved}}$ is the dissolved metal concentration and $\text{TSS}_{\text{analysed}}$ is the analysed concentration of total suspended solids in the sample.

Statistical evaluation was performed by the use of the program SPSS, version 11.05. The fitting of TSS and turbidity was performed using the program Microsoft Excel for linear regression.

Table II Data on climatic conditions (temperature and precipitation)

Month/year	Average temperature ^a (°C)	Average temperature ^b (°C)	Precipitation ^a (mm)	Precipitation ^b (mm)
12/2004	-1.0	1.6	46	31.8
01/2005	-2.8	1.3	39	31.8
02/2005	-3.0	-1.7	27	13.2
03/2005	0.1	-1.4	26	10.8
04/2005	4.6	6.9	30	14.2
05/2005	10.7	11.0	30	44.2
06/2005	15.6	15.1	45	64.6
07/2005	17.2	19.4	72	95.6
08/2005	16.2	17.1	66	77.5

^a Air temperatures and precipitation for an average year for the Stockholm Area (Station Observatorielunden, Swedish Meteorological and Hydrological Institute, SMHI)

^b Measured precipitation from rain gauge at the study site

Table III Antecedent dry periods and sampling conditions

Date of runoff event	Antecedent dry period (h)	Start of sampling	End of sampling	Total time of sampling (h)	Duration of runoff (h)	Water samples (n)
2004-12-14	190	07:03	08:40	1.6	4	6
2004-12-22–23	86	13:09	07:14	18.1	21	12
2005-01-20–21	66	06:14	05:29	23.3	53	10
2005-02-10	438	09:16	12:43	3.5	22	19
2005-02-14	56	08:40	15:46	7.1	34	1
2005-05-03	440	01:28	04:20	2.9	5	12
2005-06-04	84	08:21	10:02	1.7	3	12
2005-06-22	232	03:53	09:48	5.9	8	5
2005-08-26	213	00:34	05:37	5.1	7	23

3 Results and Discussion

3.1 Climatic conditions, electrical conductivity of water and EMC of suspended solids

Data on climatic conditions that are of relevance in the study, i.e., temperature and precipitation, are presented in Table II. Air temperature was above the normal average temperature, with the exception of March 2005, and precipitation was elevated compared to an average year. Antecedent dry periods varied between 56 and 440 h (Table III).

During the winter period the electrical conductivity was considerably higher than during the summer period as shown in Table IV. The elevated conductivity was, as expected, directly proportional to the chloride concentration. The chloride concentration in the runoff varied between 885 and 4,160 mg/l. For the

winter period the linear relationship between conductivity and chloride concentration ($r^2=0.99$) was found to be according to Eq. 3:

$$[\text{Cl}^-] = \phi \times 3.43 - 188 \quad (3)$$

where $[\text{Cl}^-]$ is the concentration in milligrams per liter of chloride ions and ϕ is the conductivity measured in millisiemens per meter.

The continuous use of sodium chloride as de-icing agent on the studied highway section was likely to be the reason for the elevated conductivity during the winter. Moreover, a study by the UK Environmental Agency (2003) suggested that increased concentrations of metals in storm water were related to the use of de-icing salts.

The EMC was higher during winter than during summer, thus the pollutant load was elevated in

Table IV Conductivity, runoff volume, EMC of TSS and load of de-icing salt (total amount of NaCl applied before sampling and, in parentheses, the amount applied during sampling)

Runoff event	Conductivity of start of run-off (mS/m)	Conductivity of end of runoff (mS/m)	Max conductivity (mS/m)	Runoff volume (m ³)	EMC (mg l ⁻¹ TSS)	Salting (g/m ²)
2004-12-14	226	344	347	27	220	24
2004-12-22–23	380	370	990	91	950	20 (25)
2005-01-20–21	234	340	995	105	4,400	20 (25)
2005-02-10	360	560	983	118	1,380	(35)
2005-02-14	283	1,270	1,280	26	170	15 (5)
2005-05-03	251	27	251	215	250	–
2005-06-04	297	58	297	125	90	–
2005-06-22	161	38	161	97	230	–
2005-08-26	134	17	134	191	50	–

winter as shown in Table IV. Our findings agree with the results presented by Legret and Pagotto (1999).

3.2 Dissolved and particulate metals

The dissolved part of Al, Cd, Co, Cr, Mn and Ni was higher during winter than during summer ($p < 0.01$, see Table V). There was no significant difference between the sampling periods with regards to the dissolved part of Cu, Pb and Zn. The only metal that showed a higher dissolved part during summer was Fe ($p < 0.01$). Throughout the summer, Cd, Cr, Cu, Mn, Ni, Pb and Zn displayed elevated concentrations in particulate matter ($p < 0.01$) as well as Fe ($p < 0.05$). The particulate part for Al and Co during summer was lower than during winter ($p < 0.01$) when compared to the other studied metals (Table IV).

The winter period displays elevated pollutant levels thus reaffirming the impact of snow and use of de-icing agents described in previous studies (e.g., Hvitved-Jacobson & Yousef, 1991; Legret & Pagotto, 1999; Viklander, 1997). Highway salting increases the corrosion of motor vehicles. An

extensive study performed in Sweden (Hedlund, 1995) showed that cars driven on roads where de-icing salts were applied displayed two to three times more corrosion damage than those cars driven on unsalted roads. De-icing salt is thus an important contributing factor to a higher metal load, both particulate and dissolved, originating from traffic. Another issue to consider is the longer wet exposure times, which may have an adverse effect on corrosion, causing increased metal loadings in the runoff. Bertling (2005) pointed out that the metal runoff rate was higher for lower rain intensities for a given total rain volume than for higher intensities, and that this because of the longer contact time at lower rain intensities. In our study, the duration of the runoff events varied between 3 and 53 h during winter, as opposed to 3 to 8 h during summer (Table III). Studded tyres will increase the wear of the asphalt surface and add to the metal pollutant load depending on the asphalt composition (Jacobson, 1994; Jacobson & Hornwall, 1999). Another factor is the use of snowploughs and the wear of the asphalt surface from road maintenance. In addition, a temporal study by Glenn

Table V Dissolved metals and metals bound to particulate matter during winter (period 2004-12-05–2005-02-14) and summer (period 2005-05-03–2005-08-26)

Metal	Dissolved matter ($\mu\text{g l}^{-1}$)	Particulate matter (mg kg^{-1})	Period	Analyses* (n)
Al	15.57≤17.80≤20.02	23,129≤25,543≤27,957	Summer	51
Al	31.13≤35.69≤40.25	28,705≤30,562≤32,420	Winter	47
Cd	0.034≤0.041≤0.048	0.85≤1.14≤1.43	Summer	43**
Cd	0.154≤0.185≤0.217	0.69≤0.86≤1.03	Winter	47
Co	0.66≤1.04≤1.42	37≤45≤53	Summer	50
Co	13.53≤15.44≤17.35	52≤61≤70	Winter	48
Cr	2.38≤3.36≤4.34	100≤110≤120	Summer	50
Cr	5.21≤7.47≤9.74	79≤86≤92	Winter	47
Cu	17.41≤20.31≤23.21	529≤591≤654	Summer	51
Cu	14.74≤18.90≤23.06	211≤232≤253	Winter	47
Fe	16.57≤37.20≤57.83	47,909≤52,618≤57,327	Summer	52
Fe	9.76≤13.14≤16.52	45,571≤48,537≤51,504	Winter	48
Mn	12.01≤17.85≤23.69	807≤939≤1,071	Summer	48
Mn	117.31≤133.88≤150.46	560≤626≤691	Winter	48
Ni	2.74≤3.08≤3.42	34≤39≤43	Summer	52
Ni	4.02≤4.54≤5.06	30≤32≤35	Winter	47
Pb	0.09≤0.14≤0.19	114≤128≤142	Summer	51
Pb	0.09≤0.13≤0.17	57≤62≤67	Winter	47
Zn	79.4≤100≤120	1,531≤1,639≤1,746	Summer	49
Zn	74.0≤96.5≤119	1,131≤1,252≤1,374	Winter	47

Confidence intervals are given ($p < 0.01$).

*The total number of water samples collected and analysed during the summer and winter period was 52 and 48, respectively.

**Five missing values due to a total concentration of Cd below the detection limit for the analysis method.

Table VI Constants K and M from the regression analysis of metal ($\mu\text{g/l}$) vs. TSS (mg/l) and the relationship, $Me = K \times TSS + M$

Metal	K	M	r^2	Min ($\mu\text{g l}^{-1}$)	Max ($\mu\text{g l}^{-1}$)	Period
Al	19	520	0.96	300	10,672	Summer
Al	25	2,300	0.99	241	122,900	Winter
Cd	0.0005	0.08	0.82	0.036	0.242	Summer
Cd	0.0004	0.30	0.92	0.055	2.24	Winter
Co	0.02	2.4	0.77	1.01	12.52	Summer
Co	0.04	20.6	0.98	2.57	231.1	Winter
Cr	0.09	4.3	0.84	1.62	44.72	Summer
Cr	0.08	9.4	0.99	0.57	366.2	Winter
Cu	0.44	29	0.92	6.26	221.4	Summer
Cu	0.25	14	0.99	5.4	1,215.7	Winter
Fe	36.5	1,100	0.93	1,027.6	19,297	Summer
Fe	46.4	1,570	0.99	406.5	225,993	Winter
Mn	0.50	51	0.92	19.6	251.3	Summer
Mn	0.60	134	0.99	20.3	2,906	Winter
Ni	0.03	3.8	0.93	0.13	14.46	Summer
Ni	0.03	5.3	0.99	0.66	150.7	Winter
Pb	0.10	2.6	0.89	1.37	47.52	Summer
Pb	0.06	1.0	0.99	1.06	295.75	Winter
Zn	1.76	72	0.94	22.80	643.0	Summer
Zn	1.15	132	0.99	20.80	5,703	Winter

The minimum and maximum value of total metal concentrations during summer and winter are given.

and Sansalone (2002) suggests strong indications that the traffic and road maintenance was, by far, the major pollutant of roadside snow.

During summer, the levels of metals in the particulate matter differed significantly with Al and Co being the exceptions. This would indicate that the sources for the metals are different during summer. In summer, corrosion will decrease in importance in the

absence of de-icing agents and the runoff events will be shorter in length. Possible traffic sources of the studied metals could be from tyres and brake linings as evaluated in studies by the Swedish National Road and Transport Research Institute (VTI, 2005) and Westerlund (1998), respectively. Sources of metals are unclear in the summer. Among the pollutant sources for runoff described by Hvitved-Jacobson and Yousef

Figure 4 Concentration of dissolved (*open triangle*) and particulate bound Zn (*open square*) versus total suspended solids during summer runoff conditions.

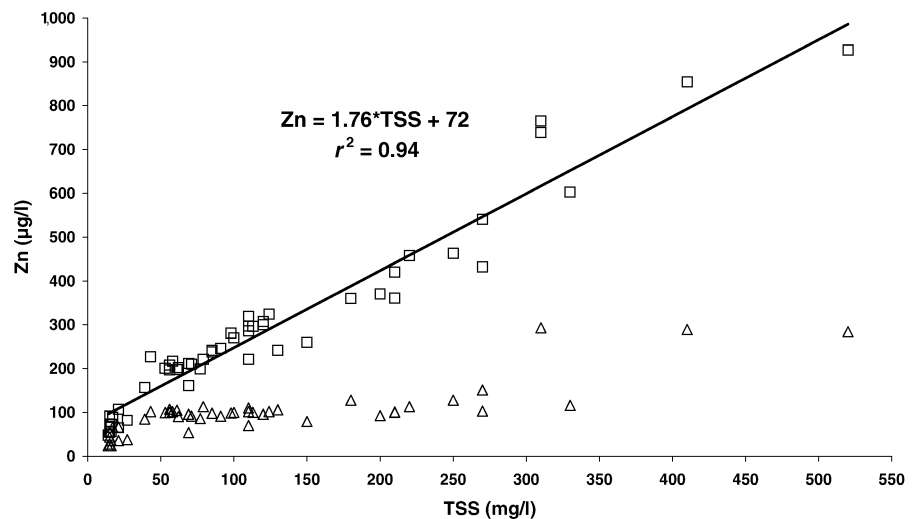
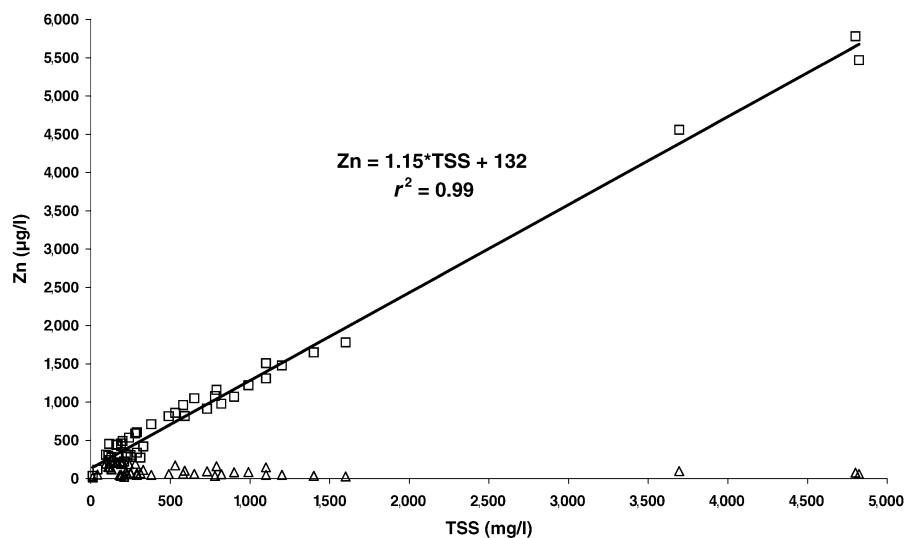


Figure 5 Concentration of dissolved (*open triangle*) and particulate bound Zn (*open square*) versus total suspended solids during winter runoff conditions.



(1991) was atmospheric fallout. Atmospheric fallout could be an important source given the location of the watershed in heavily industrialised and metropolitan areas.

3.3 Correlation of particulate bound metals and TSS

In winter the linear correlation for the total metal concentration and TSS showed a correlation factor greater than 0.95 for all metals except Cd as presented in Table VI. The correlation factors found throughout the summer for Al, Cu, Fe, Mn, Ni and Zn were 0.96, 0.92, 0.93, 0.92, 0.93 and 0.94, respectively. The lowest correlation factors were observed for Co (0.77), Pb (0.89), Cd (0.82) and Cr (0.84, Table VI). The analysed TSS range for the winter period was 13 to 4,800 mg l⁻¹, and 14 to 520 mg l⁻¹ for the summer period. The relation between TSS and metals are exemplified by Zn in Figures 4 and 5. The strong correlation of metals and TSS between and during individual events suggests that the composition of TSS is similar and independent of the variations of runoff from this particular type of area. These results concur with Hvitved-Jacobson and Yousef (1991) and also with Sansalone and Buchberger (1997), who concluded that Fe and Al were particulate bound to a greater extent. Moreover, Thomson, McBean, Snodgrass, and Monstrenko (1997) used databases comprising of data from rain events, snowmelt events and combined snowmelt and rain events (“mixed events”) for establishing correlations between TSS and metals. The findings

by Thomson et al. (1997) and our results indicate similarities for Cr, Cu, Fe, Ni and Zn. However, for Al, Pb and Cd there is a difference in magnitude between their findings and this study. It is also important to recognise that Thomson et al. (1997) also included total organic carbon (TOC) and total dissolved solids (TDS) as surrogate parameters for metals. Characklis and Wiesner (1997) showed that the presence of Zn was correlated very well with organic carbon and that Fe existed almost exclusively in the macrocolloidal fraction (0.45–20 µm).

However, it would be of interest to perform additional studies to evaluate the affects of TOC and TDS for the use of TSS as a surrogate parameter for metals.

4 Conclusion

The studied metals were mainly related to the particulate matter and the concentration of metals displayed significant seasonal variation. In winter, the metal load was higher than during summer. A good correlation was found between the total metal concentration and TSS for all metals in winter, with the exception of Cd. During summer, a good correlation was found between the total Al and Fe concentrations and TSS. The findings of this study suggest that the total metal load during winter could be assessed indirectly by the measurement of TSS. Furthermore, measuring TSS can, also assess the

metal load in summer of Al, Cu, Fe, Mn, Ni, Zn and possibly Pb.

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