PERFORMANCE EVALUATION OF COLD WEATHER INFILTRATION FACILITIES

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To the Faculty of Washington State University:

The members of the Committee appointed to examine				
the thesis of Stephen Swanson Nelson find it satisfactory and recommend				
that it be accepted.				
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	Chair			
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PERFORMANCE EVALUATION OF COLD WEATHER INFILTRATION

FACILITIES

Abstract

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The effectiveness of stormwater best management practices (BMPs) in

warm climates has been extensively researched. However, little research has been

conducted to determine the effectiveness of these BMPs for flow control and pollutant

removal during cold weather conditions. Freezing conditions and short growing seasons

can adversely affect the pollutant removal efficiency of BMPs by decreasing infiltration

through frozen soils, inhibiting biological uptake, and decreasing residence times. In

addition, deicers may compete for positively charged soil binding sites, thus increasing

metal mobility. The objectives of this study were to determine how well BMPs in cold

climates such as eastern Washington control runoff volumes and flow rates, and how

efficiently they remove heavy metals (Cu, Cd, Pb, and Zn). This will be determined by

integrating the following three subcomponents (1) sampling and monitoring of runoff

during storm events, (2) soil metal concentrations within various BMPs and, (3)

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quantifying metal transport as affected by deicing/anti icing agents in soil cores in a laboratory setting.

Field soil sampling results show a significant decrease in soil metals concentrations with depth in the upper 30 cm of soil. Flow rates and samples have been taken from several runoff events in eastern Washington. These events include two snow melt events, a rain on snow event, and the first flush of a rain event. As expected significantly larger runoff rates are produced by rain on snow events than by isolated snow melt or rain events. Runoff pollutant concentrations yielded some interesting correlations including very high correlations between copper and chloride levels in runoff suggesting that deicing activities may be effecting the concentration and speciation of this metal. The effect of road salts (NaCl, and MgCl₂) on metal mobility in a laboratory soil leaching test showed marked releases of certain metal species and organic matter during exposure to high ionic strength salt solutions followed by low ionic strength deionized water. These results are similar to those found in other literature sources and illustrate some of the concerns faced by designers in regions of the country where deicing practices are necessary.

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Dedication

To My wife Lisa and our families who have provided support to us in all our ventures

Investigation of Eastern Washington Infiltration Ponds During Cold Weather Conditions

S.S. Nelson¹, Dr. D.R. Yonge², Dr. M.E. Barber³, and Dr. M. W. Beutel⁴

ABSTRACT

A large body of research has been conducted to determine the effectiveness of roadside best management practices (BMPs) to reduce flow rates and runoff volumes as well as remove metals and suspended solids from runoff (Sansalone and Buchberger 1995; Sansalone 1999; Deletic 1999; Hathhorn and Yonge 1996). However, the vast majority of these studies have not been focused on winter conditions in areas that receive snow, experience frozen soil, and receive deicer application. Consequently, the effects of cold weather conditions are poorly understood and can potentially effect not only the capacity of a roadside treatment BMP to reduce flow rates, but also the ability of a BMP to retain pollutants. This study investigated the effects of snowfall and deicing salt application on runoff rates as well as the concentrations and speciation of metals in runoff as well as the deposition of metals within the soil column of five infiltration facilities in eastern Washington. A comparison of runoff from rain events to snow melt or rain-on-snow showed that while rainfall generated high peak runoff the runoff from snow melt

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was generally less intense, but had a much longer duration of elevated runoff. Runoff sampling and analysis revealed strong correlations between copper and chloride concentrations in runoff suggesting an association between the application of road salts and the speciation and concentration of some metals. Field soil sampling showed a strong tendency of metals to be removed and retained within the upper 20 cm of the soil column. This would suggest that infiltration of metals is an effective method of removing and sequestering metals from road surface runoff.

Keywords: metals, runoff, deicers

INTRODUCTION

The use of infiltration ponds and other roadside best management practices (BMPs) is a widely accepted method of pollution control and runoff management. However, the performance of BMPs is based primarily on research conducted during temperature conditions above freezing in areas seldom subject to ice formation and the use of deicing compounds. In this study three key areas relating to cold weather runoff treatment were examined: (i) the effects of snow melt on runoff volumes and flow rates, (ii) the concentration and correlation of metals, solids, and chloride in cold weather runoff, and (iii) the effectiveness of infiltration facilities in these regions at removing and sequestering metals from stormwater runoff.

Runoff sequestered in snow banks and road shoulders can contribute significant amounts of runoff to a rain event or release high volumes when temperature conditions rise sufficiently to produce a melt event. The sizing of infiltration ponds, vegetated filter strips, and other BMPs to accommodate snowmelt and adequately reduce pollutant discharges relies primarily on the use of safety factors and equations based loosely on parameters such as air temperature, snow pack depth, and percentage of annual precipitation falling as snow (Washington State Department of Ecology 2004). Little research has been conducted to quantify the effects of snow melt on the volume of runoff generated by a storm event or elevated temperature.

Runoff from road surfaces in areas subject to freezing conditions and deicer use contain many of the same constituents as runoff from other areas. However, the concentration-time profiles ad speciation of these pollutants may be quite unique during the winter months. Suspended solids are generated primarily by traffic activities including abrasion of tires, pavement wear, littering, and vehicular degradation (Sansalone et al. 1998) as well as atmospheric deposition. Snow banks can sequester these solids which may also complex with available metals, creating a significant first flush event when snow accumulations melt (Sansalone et al. 2002). Metals are deposited on road surfaces primarily as a result of vehicular activities. Diesel oil and tire wear are sources of cadmium (Cd). zinc (Zn) and chromium (Cr) which are deposited as a result of vehicle parts wear, and copper (Cu) from bushings, break linings, and bearings are a few sources of metals (Sansalone et al. 2002). Road salts used as deicers are also found in high concentrations during winter months. These salts have been shown to facilitate downward migration of heavy metals deposited in roadside soils (Backstrom et al. 2004). A study by (Backstrom et al. 2004) demonstrated that deicing salts, sodium chloride in particular, can contribute to increased Cd, Pb, and Zn mobilization by various mechanisms including cation exchange and lowered pH. Amrhein (1992) also demonstrated that metals such as copper may be released as a result of road salt application either by direct cation exchange or by mobilization of metal bearing constituents such as organic matter, metal oxides, or clays.

The ability of roadside soils to remove heavy metals from runoff is crucial for the maintenance of receiving water quality. Many metals, depending on their species, will become attached to available negative charge sites within the soil column of an infiltration facility. An accumulation of metals near the upper soil horizons of an infiltration facility would suggest a major sequestering of metals and imply a sufficient treatment of runoff. However, a uniform distribution of metals within the soil column or an increase with depth may indicate downward migration of metals and a loss of treatment capacity within the BMP.

The ability of runoff treatment BMPs in cold weather regions to manage and treat runoff can be crucial to the maintenance of receiving water quality for certain systems. Therefore, the effects of cold weather conditions on hydraulic performance and treatment efficiency of BMPs under winter conditions should be better understood.

METHODS

Washington is home to one of the most diverse climactic ecosystems in the United States. The western portion of the state is well known for its wet mild climate while in the eastern portion of the state climates range from arid desert in the central basin to temperate pine forests in the foothills of the Rocky Mountains in the northeastern corner of the state. While the average precipitation over the entire state is over 100 cm per year, the spatial variability ranges from approximately 380 cm in the Olympic Mountains to less than 20 cm in the desert of eastern Washington (generally defined as the part of the state east of the Cascade Mountain Range) (USGS 1999). Annual

precipitation in eastern Washington ranges from more than 60 cm in the north-eastern part of the state to 20 to 30 cm in the central basin (National Atlas 2006). The Washington State Department of Ecology (Ecology) has divided eastern Washington into four regions (Figure 1). These regions were created based on climate data including annual precipitation and temperature. These regions can be compared to the ecoregions outlined by the U.S. EPA based on criteria including geology, physiography, vegetation, climate, soils, land use, wildlife, and hydrology (Environmental Protection Agency (EPA) 2002).

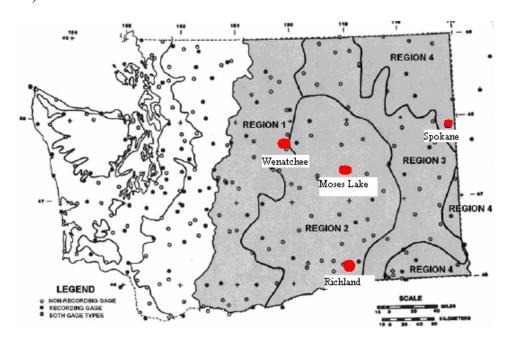


Figure 1. Delineation of the Four Climate Regions in Eastern Washington (Washington State Department of Ecology 2004)

Field Sites.

Five sample sites across eastern Washington were selected based on geographic suitability, high average daily traffic (ADT) counts, hydrologic and soil variability, and BMP site design. Sites were located in Wenatchee (Region 1), the Kennewick/Richland/

Pasco Tri-Cities area (Region 2), Moses Lake (Region 2), and two sites were located in Spokane (Region 3), one north of Spokane and one located in the Spokane Valley. Like the vast majority of the BMPs in eastern Washington utilize infiltration as the primary treatment mechanism, consequently the sites selected were all classified as infiltration BMP's. Highway runoff flow rates were monitored at two of these sites (Spokane valley and Wenatchee). Soil metals concentrations were determined for all five sites. Precipitation data for the Spokane valley and Wenatchee sites was obtained from nearby weather stations. Table 1 lists the locations and properties of each site.

Table 1. Site Characteristics

SITE	ABREVIATION	BMP TYPE	LOCATION	ADT	
Spokane Valley S1		Infiltration pond fed by two culverts (S1A, and S1B) located at the north and south ends of the pond	Intersection if I-90 and Argonne in Spokane valley	80000	
North Spokane	S2	Grassy swale leading to a constructed wetland	Highway 206 north of Spokane	8500	
Richland TC		Large infiltration pond fed by three culverts (TCA, TCB, and TCC)	Interchange of highways 240 and 182	20000	
Moses Lake ML		Small infiltration ditch receiving runoff from a culvert draining a rest stop parking lot	Adjacent to highway I-90 west of Moses Lake	Forth coming	
Wenatchee	W	Treatment train consisting of a biofiltration swale leading to a ditch and finally feeding a large infiltration pond	Adjacent to the Wenatchee river receiving runoff from a main street and urban area north and east	Forth coming	

Hydraulic Evaluation.

The hydraulic component of this study involved the comparison of runoff from natural snow, rain, and rain-on-snow events monitored at the S1 and W sites. Flow rate at the S1 site was monitored using calibrated V-notch weirs placed inside the face of culvert extensions (Figure2). Flow was quantified by converting the voltage produced by a pressure transducer located behind the face of the weir to a flow rate using a voltage-flow rate calibration curve developed in the lab. The W site was outfitted with an automated flow sampling and depth/velocity monitoring device (SIGMA 960 data logger) utilizing a depth-velocity probe. Depth was converted to discharge by determining an area using known channel dimensions and applying velocity to create a discharge using the continuity equation.



Figure 2. Spokane Culvert Pressure Transducer Field Setup.

Runoff and Soil Quality Evaluation

Runoff Sampling.

Discreet field runoff samples were collected during a range of precipitation events including rain-on-snow, snow melt, and rain following freezing conditions. Four runoff events were sampled at the S1 site. The first event was a rain-on-snow event occurring on January 17, 2006. Discreet samples were collected from peak runoff to the end of the runoff period at preselected time intervals, effectively characterizing the tail end of the runoff hydrograph. A second event was monitored and sampled on January 31, 2006.

Discreet samples were taken from the onset of runoff until little runoff was present. A third runoff event, caused entirely by snow melt, occurred on February 13, 2006. This event was also discreetly sampled from the time runoff started to when flow was near zero. A fourth event also caused by snow melt occurred on March 14, 2006 with discreet samples taken throughout the event.

Aqueous samples were filtered using a 0.45 μm filter to separate total and dissolved solids (Standard Methods section 3030 B). Total metals were determined by digestion as specified in EPA Method 200.7. An aliquot of filtered sample was acidified to pH 2 using 1:1 HNO₃ and dissolved metals determined by ICP-MS. Solids concentrations were determined according to section 2540 of Standard Methods. Chloride concentration was determined by ion chromatography.

Metals in Soils.

Soil samples from the five field sites were taken at three discreet distances and depths from the culvert inlets (0-2, 2-4, 4-6 m/0-5, 5-20-20-30 cm). Three background excavations were sampled at the same depths from areas nearby but unexposed to runoff. Soil sampling involved creating an excavation 30-40 cm deep and approximately 0.3 m square. Excavation walls were scraped to expose undisturbed soil horizons and 10-20 g samples were taken at depths of 0-5, 5-20, and 20-30 cm.

Upon return to the laboratory the samples were dried to constant weight at 103° C, crushed using mortar and pestle, and passed through a #5 sieve. The samples were then digested using an aqua reagea digest procedure specified in EPA method 200.7 (U.S.Environmental Protection Agency (1991)). Following digestion and centrifuging, 1 mL of supernatant was removed from the digest tube, diluted to 5 mL in 2% HNO₃ and

analyzed for zinc, copper, lead, and cadmium by ICP-MS using a Hewlett Packard model 4500.

RESULTS/DISCUSSION

Hydraulic Component.

Runoff from several events including rain-on-snow, snow melt, and rain were measured at S1. The data in Figure 3 depicts runoff occurring as a result of rainfall only. Note the rapid rise and decline of flow rate. While each rainfall event exhibited unique runoff characteristics the overall behavior of rainfall generated runoff was similar, with high rapidly occurring runoff peaks followed by a somewhat lower rate of flow rate decrease following the cessation of rainfall.

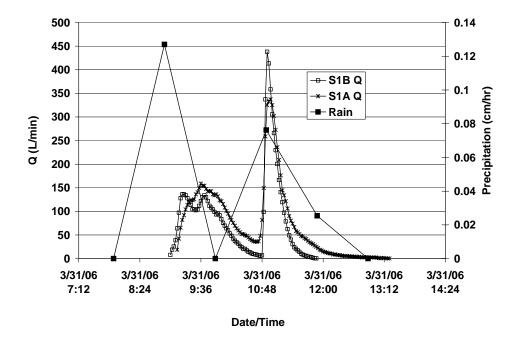


Figure 3. Rainfall Runoff Flow Rates (3/31/2006)

Figure 4 illustrates an example of runoff from a snow event followed by rain recorded from January 16-17, 2006.

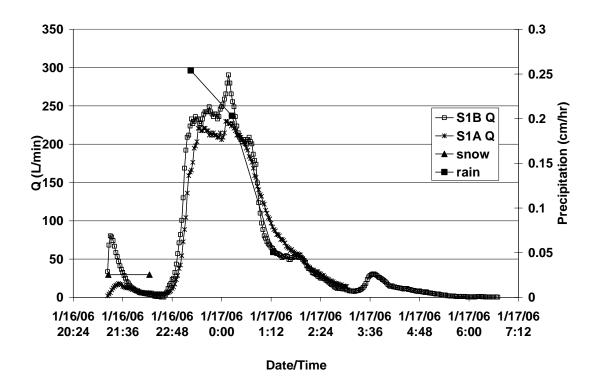


Figure 4. Rain-on-Snow Runoff Flow Rates (S1B 1/16/2006-1/17/2006)

The January 17, 2006 event illustrates the effect snow accumulations can have on runoff volumes. Note the extended period of elevated runoff compared to the rapid peak and decline of a rainfall generated runoff event illustrated in Figure 3. These figures show how different runoff volume and rate characteristics can be created by different precipitation and temperature characteristics. Currently the design criteria for most BMPs are based almost entirely on rainfall generated runoff rates and volumes. Snowmelt runoff is taken into account by the use of correction factors or predictive equations based on scant field data (Washington State Department of Ecology 2004). These results illustrate the differences in runoff characteristics and combined with further research can be utilized to optimize BMP sizing by preventing over/under sizing of facilities and streamlining design processes.

Runoff and Soil Quality Component.

Constituents in Runoff.

Areas affected by low temperatures and deicing practices have unique runoff characteristics as a result of frequent salt application and long periods between runoff events where pollutants can accumulate in roadside snow banks before being flushed off, resulting in potentially high concentrations during a first flush event (Glenn et al. 2002). Constituent concentrations and flush trends for 4 events were monitored. Figure 5 shows an example of concentrations of total lead and copper as well as chloride, solids, and flow values from the January 31, 2006 event taken from S1B. Note the "first flush" of chloride occurring during the first half hour of runoff. Following Figure 5 is a description of each metal studied and trends observed.

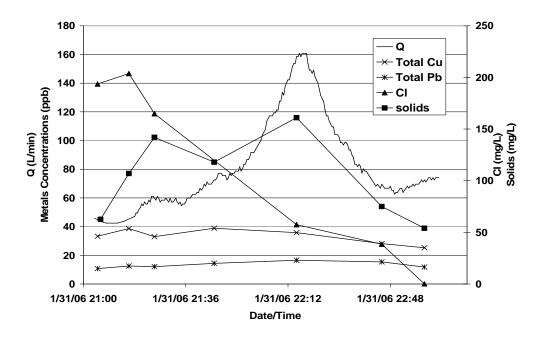


Figure 5. Total Metals, Chloride, Solids Concentrations, and Flow for Event 1/31/06

Copper

Copper showed the strongest correlations and predictable relationships of any metal species studied. Figure 6 illustrates copper's particularly strong association with chloride levels in runoff. This finding is particularly interesting with respect to cold weather runoff and could indicate elevated copper release as a result of road salt application. Copper was found to be overwhelmingly in the dissolved fraction and in some cases was at higher concentrations in the filtered samples than the total digested samples (such samples were not reported here).

S1, and W copper values are much higher than reported values for western Washington (two orders of magnitude in many cases) (Washington State Deptartment of Transportation 2005). These differences in metal concentrations especially between eastern and western Washington, two areas relatively close geographically, illustrate the necessity of assessing the effects of cold weather on runoff characteristics so that more efficient design parameters and deicing practices can be established.

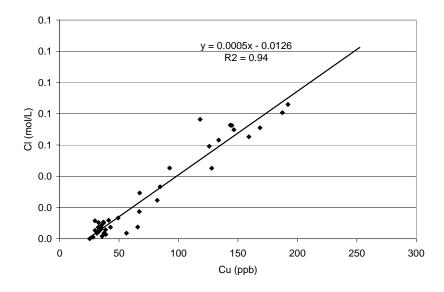


Figure 6 Figure 6. Association of Copper with Chloride in Runoff

Cadmium

Cadmium levels were near or below the method detection limit in all of the samples. Blank digests and filter blanks showed background levels similar to those measured in field samples ($<1 \mu g L^{-1}$). As a result most cadmium values in runoff were deemed insignificant and were not used to imply water quality.

<u>Lead</u>

Little correlation between lead and chloride was noted however, lead showed some association with solids in runoff (Figure 7), following the accepted trend of this metal to be immobile, readily sorbing and, remaining sorbed to available charge sites (McBride 1994). Although the amount of lead in highway runoff has diminished significantly since the ban on leaded gasoline these findings, combined with the soil metals data indicate that lead should be a metal capable of being removed and sequestered for long periods of time.

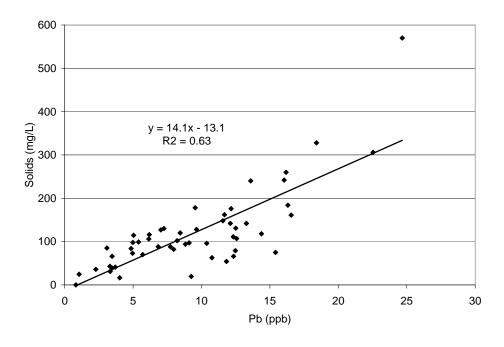


Figure 7. Association of Lead with Solids in Runoff

Zinc

Zinc was found at fairly high levels, comparable in most cases to copper. Sporadic sample contamination was problematic and most of the zinc numbers had to be discarded. However, zinc did show similar behavior trends to lead in terms of associations with solids concentrations in runoff.

Chloride

Chloride values exceeded the Washington state surface water acute criteria of 860 mg L⁻¹ in 44% of samples taken (WAC 1997). As mentioned earlier, previously conducted research, (Backstrom et al. 2004), (Amrhein et al. 1992), and (Novotny et al. 1998) indicate that some deicing salts can facilitate mobilization of some metals, especially copper and cadmium, via various mechanisms including dispersion of organic matter, cation exchange, and chloride complex formation. Our research exhibited copper mobilization that was correlated to chloride concentration.

Metals in Soils.

Many heavy metals have been shown to exhibit strong adsorption tendencies to certain soil constituents: copper tends to associate with organic matter, zinc binds strongly to clay particles, and lead chemisorbs very strongly to organic matter (McBride 1994). The tendency of most metals to adsorb strongly in the upper soil horizons was clearly demonstrated in this study. In nearly all sample sites soil metals concentrations decreased to background levels by a depth of 20-30 cm. Figure 8 illustrates an example of average metal concentration decreases averaged for all five sites. Note the decrease of all metals to background levels.

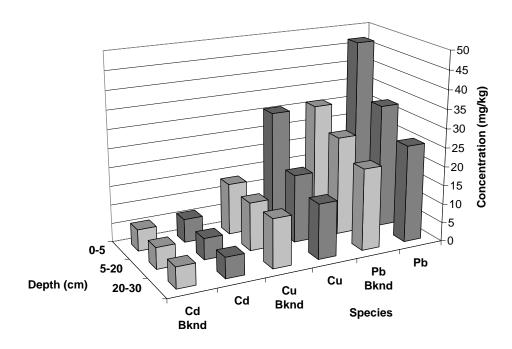


Figure 8. Average Metals Concentration Distribution

As mentioned before each metal has unique characteristics governing its relative solubility. The following is a description of each metal species and its observed characteristics. Table 2 outlines the numerical values for each metal at each site further illustrating the different characteristics of each metal.

<u>Copper</u>

Copper showed significant decreases in concentration with increased depth for nearly all sample sites. This trend was expected and illustrates the ability of copper to associates strongly with organic matter as well as other available charge sites in soils (McBride 1994). All sites except ML showed decreases in copper concentrations to near backgrounds at 20-30 cm. ML and W were atypical in that the soil horizons sampled were composed of sand/gravel deposits left by previous runoff activities and not naturally occurring strata like the S1, S2, and TC sites.

Cadmium

While cadmium showed no decrease in concentration with depth, concentrations were near background at all locations. The absence of elevated cadmium levels may be an indicator of soil leaching in these roadside environments. Cadmium has been shown to form chloride complexes and become replaced by other cations in the presence of road salts (McBride 1994).

Lead

Soil lead concentrations showed the most consistent trends of any metal investigated in this study. Lead showed marked decreases with depth to nearly background concentrations for all sites with the exception of the ML site. As with copper this may be explained by the nature of the soil sampled at this site. These results seem to indicate a significant removal and retention efficiency of infiltration facilities for lead.

Zinc

Zinc concentration profiles were quite similar to lead. Concentrations generally reached or neared background at a depth of 30 cm indicating a sufficient capturing and sequestering in the upper soil horizons. W and ML results were similar to copper and lead in that background levels were not reached by 20-30 cm depth.

CONCLUSIONS

While the results of this study are based on relatively few storm events due to the lack of control of rain-on-snow occurrences, there are a few key observations that can be noted about the treatment efficiency of roadside BMPs in cold weather environments. First, flow characteristics in areas subject to snowfall are unique. Runoff caused by rain events generally generate high peaks of runoff rapidly decreasing upon secession of

precipitation, while snow and rain-on-snow events generate longer periods of runoff albeit at lower peak discharges. Further research regarding specific parameters affecting runoff volumes and flow rates (e.g. temperature, rainfall, existing snow pack, etc.) must be conducted to optimize facility sizing. Second, runoff quality characteristics in cold weather regions are unique. The use of road salts in particular may be affecting the speciation and concentration of metals, especially copper, in runoff. Correlations between copper and chloride suggest a relationship between road salt application and speciation of metals in stormwater runoff. Speciation and mobility of metals, and the effects of deicing salt application, should be further research to investigate the effects of road salts on metal speciation, migration, and bioavailability. Finally, heavy metal adsorption characteristics combined with the results shown in Table 2 and Figure 5 suggest that in many cases infiltration is an effective method of removing and sequestering heavy metals in runoff for long periods of time. While some downward migration of metals may be occurring, the elevated concentrations of metals in the soil when compared to the relatively small concentrations contained in runoff would seem to indicate a buildup of metals within the upper 20 cm of soil. Studies involving sampling of infiltrated stormwater in natural environments, soil core leaching studies investigating the effects of road salts on metal mobility, as well as other research should be conducted to more accurately ascertain the effects of road salts on metal speciation and migration.

Table 2. Metals Concentrations with Distance and Depth from Culvert Inlets (All Values in PPB)

			<u>Cu</u>				<u>Cd</u>				<u>Pb</u>				<u>Zn</u>	
			tance fro ulvert (m				Distance f Culvert]	Distance f Culvert				istance fr Culvert (1	
	Bknd	0-2	2-4	4-6	Bknd	0-2	2-4	4-6	Bknd	0-2	2-4	4-6	Bknd	0-2	2-4	4-6
Depth (cm)								s	pokane [•]	1						
0-5	12.6	56.9	60.7	16.0	4.4	9.4	9.9	3.5	25.7	58.7	92.1	39.8	120.9	511.7	665.8	185.1
5-20	10.6	9.3	12.8	4.7	4.5	7.9	7.8	3.0	26.9	21.9	33.6	21.0	104.0	614.9	94.6	68.7
20-30	14.4	12.2	6.8	10.5	4.4	8.0	3.0	3.1	19.2	16.9	28.8	16.5	89.8	103.3	83.7	139.3
								S	pokane :	2						
0-5	9.3	36.2	32.6	9.8	4.7	5.1	4.8	4.5	10.4	52.1	46.8	13.0	83.7	321.1	258.9	75.0
5-20	9.8	25.9	9.5	9.9	4.6	4.6	4.5	5.6	11.6	48.7	14.0	10.9	83.5	270.3	85.4	150.5
20-30	9.3	11.5	9.9	9.5	4.8	5.2	4.5	5.1	9.4	14.3	13.1	9.3	115.1	72.8	70.0	139.1
								F	Richland							
0-5	18.3	29.4	37.6	42.1	8.0	6.6	5.2	8.7	6.0	42.0	69.3	75.0	176.8	685.2	1021.8	502.5
5-20	18.2	11.9	29.6	28.7	8.3	4.5	5.0	8.1	5.0	14.4	62.9	64.2	187.5	310.9	533.7	374.1
20-30	16.1	12.9	12.6	19.0	7.9	4.5	7.6	8.0	3.8	12.4	26.6	9.0	157.4	299.3	190.8	167.1
								W	enatche	е						
0-5	18.6		25.3	19.2	6.0		5.7	6.1	108.3		72.9	47.4	183.4		239.3	215.3
5-20	17.2		15.1	12.5	5.9		6.0	5.9	83.3		63.1	40.7	165.4		197.7	166.5
20-30	17.0		19.2	21.6	5.9		6.0	6.1	71.9		117.0	33.2	168.9		195.2	195.8
								Me	oses Lak	(e						
0-5	9.4	16.9	30.7	20.2	5.5	5.9	5.6	5.8	5.6	8.0	20.6	20.4	136.3	275.1	462.5	550.3
5-20	8.2	21.8	32.9	27.2	5.2	5.5	5.5	5.6	3.6	14.8	29.7	19.5	71.9	338.0	487.5	442.9
20-30	8.4	16.1	18.7	24.1	5.1	5.4	5.7	5.6	3.8	13.4	21.5	27.7	78.9	293.3	347.9	405.0

ACKNOWLEDGEMENTS

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The Effects of Road Salts on Heavy Metal Mobility in

Two Eastern Washington Soils

S.S. Nelson⁵, Dr. D.R. Yonge⁶, Dr. M.E. Barber⁷

ABSTRACT

Heavy metals deposited on road surfaces and transferred by runoff to roadside environments can have serious impacts on receiving ecosystems. Infiltration is an effective method of controlling runoff volumes as well as removing solids and metal contaminants. Retention of metals within infiltration facilities depends on a number of factors including metal species, soil characteristics, influent concentration and vadose zone detention time. Some influent species, such as road salts, have been shown to mobilize heavy metals putting receiving waters at risk. The aim of this study is to ascertain the effects of two widely used road salt species, (NaCl, and MgCl₂), on heavy metal mobility in two eastern Washington soils. One soil was basic exhibiting a soil pH of 8.3, and taken from an infiltration pond site in Spokane adjacent to I-90, the second soil was an acidic soil, exhibiting a soil pH of 5.9, taken from an infiltration pond site in Richland Washington in the central basin adjacent to Highway 240 and Highway 182. Varying concentrations of each salt were exposed to both soils continuous flow soil columns. Leachate samples were collected and analyzed for metals, organic matter, and

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pH. Experiments were also performed without salt and used as controls. Results indicate that metal mobilization can occur by a number of mechanisms including cation exchange, chloride complex formation, and colloid dispersion (release of organic matter and/or clay that can complex metal species). Sodium chloride resulted in the largest release of copper and lead via indirect mobilization of organic matter. Magnesium had less of an effect on lead and copper but had a much greater effect on the mobilization of cadmium. Releases of metals during, or following, salt application ranged from 50% to 1000% greater than metals releases from the control experiments.

Keywords: contaminant transport, soil leaching, dissolved metals

INTRODUCTION

Heavy metals are deposited in roadside environments by a number of sources, primarily vehicular parts wear (Sansalone et al. 2002). Runoff from roadways must be treated to reduce flow rates and volumes and remove contaminants such as suspended solids and heavy metals. Infiltration is a highly effective method of pollutant removal and flow control and has been shown to retain most metals from runoff within the upper few cm of soil (Dierkes and Geiger 1999). However, infiltration should not necessarily be thought of as a treatment process. If metals are mobilized by some process and passed into receiving water the facility ceases to treat and actually becomes a source of contamination (Igloria et al. 1996). Some salt species have been shown to increase metal mobility by a number of mechanisms including cation exchange, colloid dispersion, and chloride complex formation (Amrhein et al. 1992; Amrhein et al. 1993; Amrhein and Strong 1990). The studies by Amrhein focused on the use of NaCl and compared it to calcium magnesium acetate (CMA). The studies found that NaCl and CMA can both

mobilize metals by the aforementioned mechanisms. Results such as these and the continued findings regarding the effects of certain salts on the environment have lead many state departments of transportation to change to more environmentally sound deicing practices and deicers. The objective of this study was to evaluate the effects of two commonly used deicing compounds (sodium chloride (NaCl) and Magnesium Chloride (MgCl₂)) on the mobility of heavy metals through soil columns composed of two eastern Washington soils. Both soils were collected from topsoil taken from sites in eastern Washington and had similar organic matter content and cation exchange capacities (physical and chemical characteristics detailed in Table 3). Continuous flow column experiments were used to develop metals release data. Both soils were leached with three concentrations of both NaCl and MgCl₂. Dissolved metals, organic matter, and pH was monitored and mobilization of each metal species in response to salt influent evaluated. In this study mobility or mobilization of a metal is used as an aggregate term referring to the tendency and/or magnitude of a metal released as either a direct or indirect result of salt application. The results of this study indicate that certain metals will me more likely to migrate when exposed to certain salts and in certain soil types.

METHODS

Two best management practice sites in eastern Washington were chosen for soil collection based on average annual daily traffic (AADT), soil metal concentrations, and facility type. The first site was located just east of Spokane Washington and was an infiltration pond receiving runoff from Interstate 90. The second site was located in Richland Washington in the south central region of the state. This site was also an infiltration pond collecting runoff from interstate 84. Both sites are adjacent to highways

with speed limits of 60 mph. The Spokane site soil was exposed to an AADT of 80000, and the Richland site was exposed an AADT of 20000.

Soil from the Spokane site was removed from the upper 0-5 cm of soil using a stainless steel shovel to collect a composite sample from a 50 cm² area just downstream of the culverts. The Richland soil was collected by a backhoe and was taken from the upper 0-20 cm of soil from a 50 cm² patch located at the low point of the pond. Both samples were stored in plastic 5 gallon buckets prior to use. The Spokane soil was classified as a loam and the Richland soil a sandy loam. The Spokane soil also contained 50% more clay than the Richland soil and had a sodium adsorption ratio nearly four times that of the Richland soil. Table shows the characteristics of both soils. Data for this table was obtained from the soils lab at the University of Idaho located in Moscow Idaho.

Table 3. Soil Characteristics

	Cation Exchange Capacity cmol(+)/kg	Exchangeable Sodium Percentage	Sodium Absorption Ratio	pH [*]
Spokane	17	2.90%	2.29	8.1±0.2
Richland	16	1.10%	0.56	5.98±0.3

_	<u>Spokane</u>	Richiand
Sand	47.50%	67.50%
Clay	18.80%	12.50%
Silt	33.80%	20.00%
Texture	Loam	Sandy Loam

pH defined as first portion of leachate from each column.

Averaged from six columns

A total of six soil columns from each site were run. Columns were 6.4 cm in diameter and 20.25 cm high and were constructed of clear plastic (Figure 7). Each column was run using one of the salt species at one of three different concentrations. Laboratory soil columns were constructed by stirring together a known mass of soil to

water in the column. When a thick paste formed more water was added followed by more soil until 550 gm of soil had been added. Pore volume was calculated by assuming a soil specific gravity of 2.65 and measuring the height of soil in the column. The average pore volume was between 225 mL and 275 mL. Columns were attached to a vacuum apparatus and influent applied to the top of the column (Figure 7).



Figure 7. Lab Column Setup

Soil leaching consisted of three phases. In the first phase, 2-3 pore volumes of deinionized water were passed through the columns to remove the most mobile constituents and stabilize flow rates. In the second phase, 3-5 pore volumes of NaCl or MgCl₂ solutions of 0.1, 0.05, or 0.025 M chloride were passed through. These chloride concentrations were based on actual field values where, 0.1 M represents high concentration events, 0.05 M the mid range concentrations, and 0.025 M representing lower salt concentrations. Following salt application, in the third stage, three pore volumes of deionized water were passed through the column to simulate low ionic strength runoff occurring as runoff from a storm event progresses.

Samples were collected in a vacuum apparatus and filtered through a 0.45 μm PTFE syringe filter. A portion of the sample was acidified, by adding 1 mL 1:1 HNO₃ to 4 mL of sample, and then analyzed for dissolved metals, (Cu, Cd, Pb, Zn), by ICP-MS using an HP model 4500. Organic matter was analyzed by measuring absorbance of the sample at 254 nm using an HP 8453 spectrophotometer. A third aliquot of filtered sample was analyzed for chloride by ion chromatography.

Blanks as well as standard additions were performed for quality assurance. All glassware used in sample collection and preparation was washed using sodium hypochlorite bleach solution, followed by a triple rinse of tap water, a triple rinse of deionized water, and a rinse or soak in 1:1 HNO₃. Finally, glassware was triple rinsed in ultrapure deionized water generated by a Barnstead Epure series 1090.

RESULTS/DISCUSSION

The most important factors affecting the strength of a metals' partitioning coefficient and thus its' mobility have been reported to be the valence and ionic radius (McBride 1994). The ratio of these two parameters, referred to as the ionic potential, contributes to their ability to sorb to available charge sites. The more covalent the character of the bond between a metal and its' charge site, the less likely the metal is to be mobilized. Soil characteristics such as pH, the amount and type of available charge sites, and the amount of organic matter present are also important factors to consider when investigating metal mobility (Allen et al. 1993). Higher pH facilitates formation of available charge site in the form of oxide, hydroxide, and carbonate precipitates. Soils high in organic matter and clays will be able to sorb and retain metals more readily than sandier soils with fewer available charge sites (McBride 1994).

In addition to the mobility of metals in soil, the ability of plants and animals to retain metals, or "bioavailability", is also an important factor when investigating heavy metals. Speciation of metals in soils is the major factor controlling bioavailability as stated by McBride (1994) in the following two general rules: 1)"Short term toxicity to plants and microorganisms is most closely related to free metal cation concentration in solution." and 2)" Metal uptake over the long term depends to a large extent on the total metal concentration in solution and on the ability of the soil to maintain this concentration." According to this definition metals such as copper, which is often found associated with organic matter and colloids, may not be as bioavailable as cadmium which is often found in it's more mobile ionic form. However, as this study showed, associations of metals, especially copper to organic matter, can be a strong vector for dissolved copper transport.

Spokane Soil Results.

Lead

Lead (Pb) is generally considered to be one of the least mobile metals in roadside soils as a result of its tendency to chemisorb strongly to available charge sites (McBride 1994). Very little if any release of lead was observed in the presence of MgCl₂ during leaches of the Spokane soil. By comparison NaCl facilitated a more significant release of lead. Lead release occurred in a fashion proportional to the influent salt concentration as illustrated in Figure 8 with lower levels of lead being released in response to lower salt concentrations. The following figures will not include all salt concentrations but trends for nearly all metals and constituents were similar to Figure 8 with lower responses noted for lower influent salt concentrations. The most probable mobilization mechanism for

lead in this scenario is a sodium facilitated dispersion of clays or metal oxides as illustrated in Figure 9. Soils high in sodium when exposed to a low ionic strength solution following a high ionic strength solution (as is the case with the Spokane soil in this scenario) have been shown to disperse organic matter, clays, and metal oxides (McBride 1994). However, the specific mechanisms of lead release are difficult to isolate as there is little correlation between any measured constituents. Despite the lack of correlation between the release of organic matter and lead, clays or metal bearing oxides may be dispersed by the introduction of deionized water following the salt influent (Amrhein et al. 1993). Note also the lower concentrations present during the application of salt solution. This may be due to a formation of insoluble lead chloride.

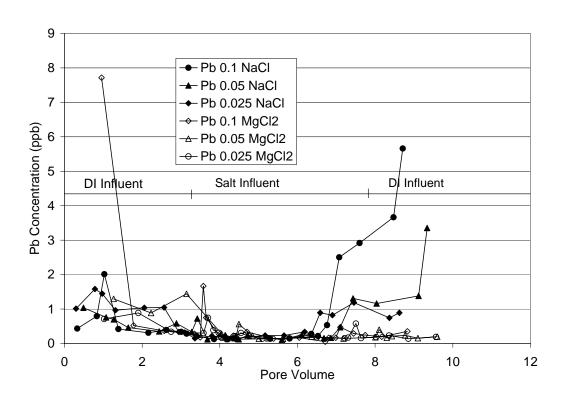


Figure 8. Lead Release in Response to NaCl and MgCl₂ Addition

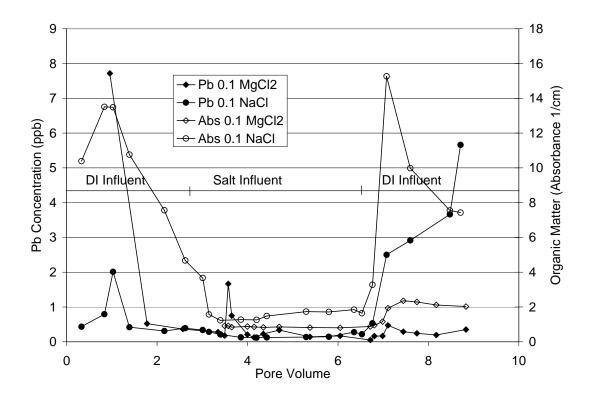


Figure 9. Soluble Lead Concentration in Spokane Soil Leachate

<u>Copper</u>

The data indicate that copper (Cu) is more mobile than lead by virtue of its' tendency to be found in higher concentrations in leachate than the lead, tending to associate with available charge sites less strongly. Copper has been shown to associate with organic matter, oxides, as well as edge sites of clay silicate minerals (McBride 1994) Associations between copper and organic matter in leachate samples were quite strong. While both salt species facilitated the release of organic matter and copper, NaCl had a far larger cumulative effect than MgCl₂ on dispersion of organic matter following the application of salt by deionized water illustrated in Figure 10. This supports findings that magnesium promotes flocculation and soil structure and prevents dispersion while sodium promotes dispersion of organic matter and clays as stated above. As with Pb the release of Cu during the deionized water rinse following application of salt solutions is

likely an indirect release mechanism facilitated by the release of copper bearing constituents, probably organic matter in this case. As with lead the lower salt concentrations had similar but lower intensity trends as the ones shown in Figure 10 11.

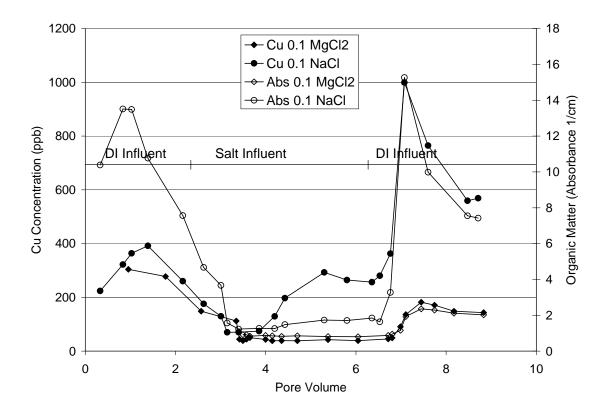


Figure 10. Soluble Copper Concentrations in Spokane Soil Leachate

Cadmium

Cadmium (Cd) is generally considered to be highly mobile in roadside soils (Norrstrom and Jacks 1998). As shown in Figure 11 releases of cadmium occurred in the presence of both NaCl and MgCl₂, and showed significant correlations to the chloride values in both leachate samples. The most probable mechanism for cadmium release is direct cation exchange. Cadmium forms weaker interactions with available charge sites than sodium and magnesium in certain situations and the presence of these cations serves to mobilize this metal (McBride, 1994). The elevated release of cadmium in the presence

of MgCl₂ suggests that cation exchange of Cd²⁺ with Mg²⁺ results in a larger release of cadmium than cation exchange between sodium and cadmium.

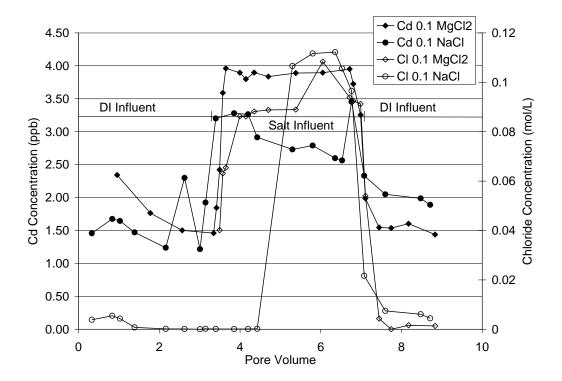


Figure 11. Soluble Cadmium Concentrations in Spokane Soil Leachate

Richland Soil Results.

<u>Lead</u>

Some lead (Pb) release, probably as a result of cation exchange, occurred during the MgCl₂ leach as seen in Figure 12. An indirect release of lead can be noted for the NaCl leach when low ionic strength water was applied following the salt solution. The behavior of lead in the Richland soil showed that lead is mostly immobile and mobilization of lead by cation exchange (MgCl₂) or dispersion of colloids (NaCl), while present, is minimal. Results from this soil were somewhat anomalous in that lower

concentrations of NaCl produced a somewhat greater release of lead during the final deionized leach than the 0.1 M NaCl leach.

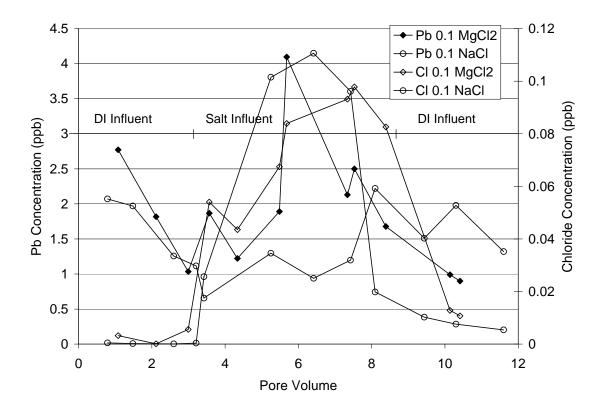


Figure 12. Soluble Lead Concentrations in Richland Soil Leachate

Copper

The release of copper from the Richland soil in the presence of magnesium chloride and sodium chloride was moderate as indicated in Figure 14. Indirect release of organic matter and copper bearing constituents are the most likely mechanisms for copper dispersion in the Richland soil. While NaCl showed a higher release of organic matter than MgCl₂ copper released by both salt species was quite comparable. The 0.1 mol/L concentrations are illustrated in Figure 13 and are representative of the other concentrations and salt species.

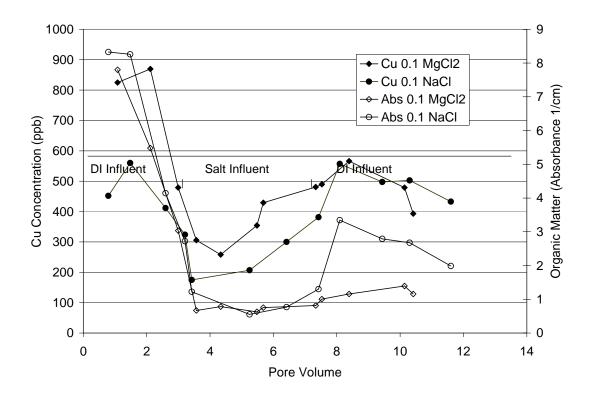


Figure 13. Soluble Copper Concentrations in Richland Soil Leachate

<u>Cadmium</u>

Both NaCl and MgCl₂ solutions facilitated large releases of cadmium. The magnesium salt released much more cadmium than the NaCl solution (Figure 14). The ability of magnesium to better compete with cadium for charge sites over monovalent sodium seems to be the most likely explanation for the higher release of cadmium in response to MgCl₂ over NaCl. Correlations between chloride concentrations and cadmium were quite good especially for the MgCl₂ leach. While only the 0.05 mol/L leaches are depicted trends from all leaches were highly comparable.

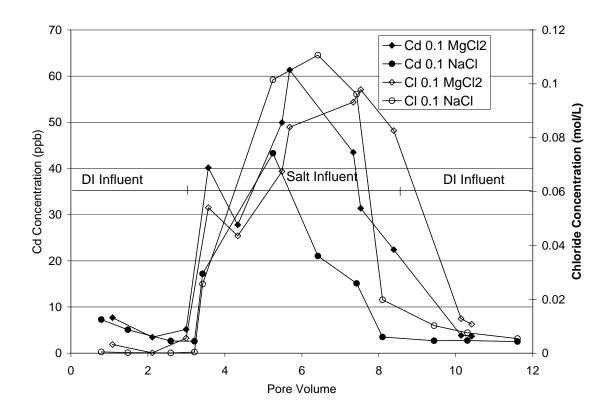


Figure 14. Soluble Cadmium Concentrations in Richland Soil Leachate

<u>Comparison of leaching characteristics between soils.</u> Lead

While both soils were more likely to release lead during exposure to deionized water following NaCl application the Spokane soil released more than the Richland soil. This may be due to the elevated release of organic matter or perhaps a release of clays or some other metal bearing constituent when low ionic strength influent follows higher ionic strength salt solutions. The Spokane soil had a much higher sodium adsorption ratio (SAR) which may have contributed to a greater degree of dispersion of lead bearing constituents. However, the concentrations of lead released were quite low in all trials demonstrating the strong chemisorption characteristics of lead in soils.

Copper

Copper showed strong correlations to organic matter release during the leach of low ionic strength water following application of salt solutions. While both soils responded differently to the salt solutions, Figure 15 was generated by using data from all available samples from both soils and illustrates the close relationship of copper to organic matter especially the in the Spokane soil. The close relationship of copper to the dispersion of organic matter is most likely related to the mobilization of organic matter in the presence of abundant adsorbed Na. The high SAR for the Spokane soil may explain the close relationship between organic matter and copper when compared to the Richland soil.

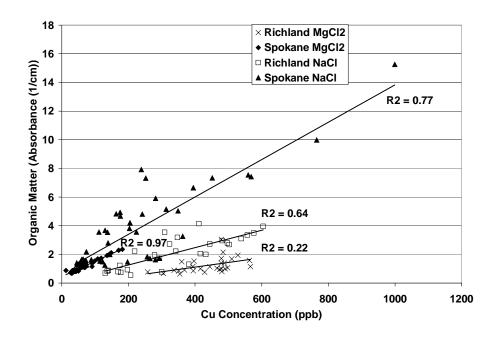


Figure 15. Correlation of Copper to Organic Matter Leachate Concentrations

Both soils exhibited similar trends in response to both road salt species. NaCl dispersed more organic matter in both soils and as a result more copper that may be associated with the organic matter. The higher overall release of copper from the

Richland soil could be attributed to either the lower pH of the soil, a higher concentration of copper in the Richland soil, or fewer available chemisorption sites. Copper concentrations in the Richland soil were on average twice as high as the Spokane soil. As mentioned before low pH soils are less able to retain metals than alkaline soils. (McBride 1994) Also, available charge within the Richland soil may have weaker bonds with copper allowing it to become mobilized more easily.

Cadmium

The release of cadmium from both soils can most likely be attributed to direct cation exchange in the presence of the salt. To illustrate this point cadmium and chloride concentrations following the addition of salt during the MgCl₂ leaches were plotted for both soils. Figure 16 illustrates the close relationship of cadmium to chloride when MgCl₂ is present. Note the weaker relationship of NaCl chloride levels to cadmium compared to MgCl₂.

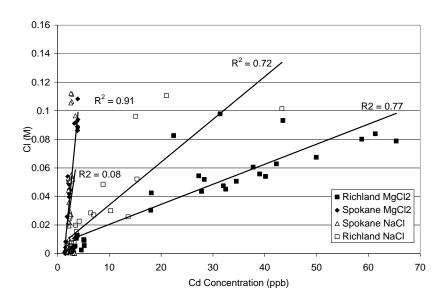


Figure 16. Correlation of Cadmium to Chloride in the Presence of MgCl₂ and NaCl

Table 3: Comparison of Soil Metal Release Characteristics

		Release Mechanisms	Max Release (ppb)	Correlations
<u>Spokane</u>	Pb	Indirect release in response to NaCl application		OM(NaCl)=0.48 OM(MgCl)=0.00
	Си	Indirect release, primarily in response to NaCl application		OM(NaCl)=0.50 OM(MgCl)=0.97
	Cd	Direct cation exchange in response to NaCl and MgCl2		Cl(NaCl)=0.08 Cl(MgCl2)=0.91
<u>Richland</u>	Pb	Indirect release in response to NaCl / cation exchange		OM(NaCl)=0.43 OM(MgCl)=0.31
	Си	Indirect release in response to NaCl application		OM(NaCl)=0.36 OM(MgCl)=0.87
	Cd	Direct cation exchange in response to salt addition		Cl(NaCl)=0.72 Cl(MgCl)=0.77

Both soils exhibited similar release trends for all metals species. However, the magnitude of metals released from the two soils was quite different as illustrated in Table 3. There are numerous factors that may have contributed to the labile nature of cadmium in the Richland soil, the major release of copper in the Spokane soil, and the greater amount of lead released from the Spokane soil in spite of much higher concentrations in the bulk Richland soil. The chemical composition, the amount of organic matter, the nature of charge sites in the two soils, the amount of sodium in the soils, and the pH are

all factors that probably have some effect on the release trends and magnitudes for both soils.

CONCLUSIONS

Road salts can have a significant effect on the mobility of metals in roadside environments. Magnesium, while showing some small effect on copper and little to no effect on lead, was shown to release much higher levels of cadmium from roadside soils than sodium based deicers. NaCl, on the other hand, while having less direct effect on cadmium exhibits a tendency to release much higher levels of metal bearing species such as organic matter and clays. This indirect release mechanism seems to have a much larger effect on moderately to strongly incorporated metals (copper and lead) than direct cation exchange. It should be noted that metals associated with colloidal or dissolved constituents are much less bioavailable than their free counterparts (McBride 1994). Metal mobility is also highly dependent on the soil characteristics. While both soils examined in this study had similar cation exchange capacities the release of some metals, cadmium in particular, was much greater from the Richland soil than the Spokane soil. This could be attributed to the larger fraction of clay in the Spokane soil, some soil management practice such as the addition of topsoil to the BMP, a difference in soil pH, soil organic matter, or some other unquantified parameters.

Eastern Washington soils, for the most part, seem to retain metals within the upper 0-20 cm of soil demonstrating significant retention of metals removed from runoff (see chapter 1). These column leaching experiments showed that while some mobilization of metals by road salts can occur, the majority of these metals are likely in a less bioavailable form as they are complexed with organic matter and/or clay released during

the introduction of deionized water following salt solution application. In addition to the speciation of metals mobilized the overall mass fraction of metals mobilized is quite small when compared to the overall soil metal load. While this may suggest that metals, especially copper, mobilized by deicing salts are less harmful to the environment due to their association with species such as organic matter and clay the fact remains that there is potential for their introduction to the environment due to enhanced mobility.

ACKNOWLEDGEMENTS

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APPENDIX A

DETAILED METHODS

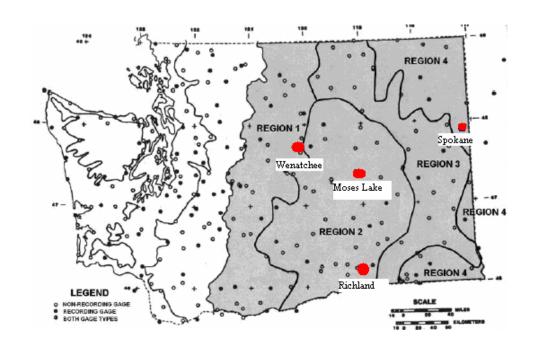


Figure 1A. Delineation of the Four Climate Regions in Eastern Washington
(Washington State Department of Ecology 2004)

Field Sites

A tour of eastern Washington by Washington State University faculty, Washington State Department of Transportation personnel, and myself was conducted at the onset of the study to identify suitable field sites. Five sample sites across eastern Washington were chosen based on geographic suitability, high average daily traffic (ADT) counts, hydrologic variability, and design (Figure 1A). One site was located in Wenatchee (Region 1), one site was located in the Kennewick/Richland/ Pasco Tri-Cities area (Region 2), one site was located in Moses Lake (Region 2), and two sites were located in Spokane (Region 3). All five sites utilized infiltration as the primary treatment mechanism. Runoff rates and stormwater samples were obtained at two of these sites (Spokane and Wenatchee) and soil metals concentrations were determined for all five

sites. Precipitation data was obtained from nearby weather stations operated by the National Weather service or Washington State University.

<u>Spokane</u>

The first site in Spokane (S1) was an infiltration pond located between an off ramp from I-90 to Argonne Street. S1 was fed by two culverts, S1A and S1B (Figure 2A). This site was chosen due to the high ADT of the adjacent highway, large contributing area, easy access, and suitability to runoff sampling and monitoring. Note that a concrete spillway was located at the outlet of both culverts. Both culverts were instrumented to monitor flow as described later in this section. Weather data was obtained from a National Weather Service station located at Felts Field airfield approximately 3.25 Km west. Onsite monitoring proved difficult as snow accumulation on top of tipping bucket rain gauges made accurate measurement of rainfall following snow impossible.



Figure 2A. Spokane 1 (S1) Field Site. Left Photo Shows S1B, Right Depicts S1A

The second Spokane site (S2) was a vegetated filter strip feeding a constructed wetland located north of Spokane adjacent to highway 206 (Figure 3A). This site was relatively new and was exposed to relatively little ADT.



Figure 3A. Spokane Vegetated Ditch Leading to Infiltration Pond

Tri-Cities

The Tri-Cities (TC) site was located at the interchange of highway 240 and interstate 182 in the city of Richland, Washington (Figure 4A). This site was also chosen for its high ADT, easy access, and suitability for monitoring purposes. The site had three inlet culverts forming a triangular infiltration pond.

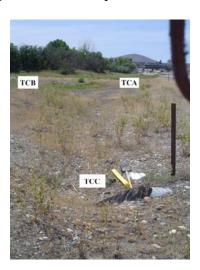


Figure 4A. Tri-Cities Field Site. TCC is in the Foreground While TCA and TCB Are

Roughly 60-90 m Behind TCC

Wenatchee

The Wenatchee site (W) utilizede a treatment train consisting of two ditches followed by an infiltration pond designed to trap and treat runoff from a large urban

watershed preventing release into the Wenatchee River (Figure 5A). High ADT, a large contributing surface area, and the ability to use unmanned monitoring equipment made this an attractive study site. Weather data was collected from a monitoring station located at the Washington State University tree fruit research center located on the watershed for the site approximately 2 kilometers southwest of the site.

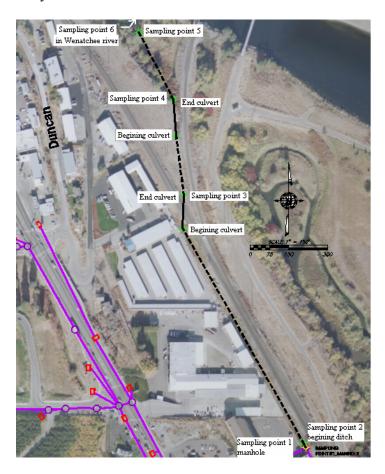


Figure 5A. Plan View Picture of Wenatchee Site Depicting Treatment Train Layout and Sample Collection Points

Moses Lake

The Moses Lake site consisted of an infiltration ditch receiving runoff from a rest area adjacent to I-90. This site was chosen for its unique contributing area potentially high ADT and easy access (Figure 6A).



Figure 6A. Moses Lake Inlet and Infiltration Ditch

Runoff Monitoring

Runoff resulting from snowmelt, rain, and rain-on-snow was continually monitored at two sites (S1 and W). Site S1 was monitored using V-notch weirs placed inside the face of culvert extensions (Figure 7A). Data was recorded by a Telog 3307-A data logger. Flow was quantified by converting the voltage produced by a pressure transducer, located just upstream of the weir, to a flow rate using a voltage-flow rate calibration curve developed in the lab. Calibration of the weirs was carried out in a flume located in the Ahlbrook laboratory and was determined by matching the flow rate passing over the weir to a voltage recorded on a data logger at that time. Flow was quantified using bucket, graduated cylinder, and a stopwatch. The time of the flow was noted, matched to a voltage stored in the data logger and when graphed together a calibration curve shown in Figure 8A created.



Figure 7A. S1 Pressure Transducer Field Flow Monitoring Setup

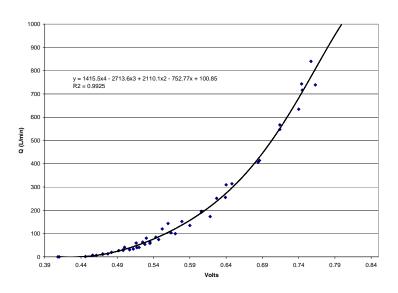


Figure 8A. Calibration Curve for S1 Flow Monitoring

The Wenatchee site was outfitted with an automated flow sampling and depth/velocity monitoring device (SIGMA 960 data logger). The data logger quantified discharge by converting depth to an area using known channel dimensions and multiplying the area by the measured velocity to create a discharge.

The depth/velocity probe was placed in the culvert opening, Figure 9A A. and platform was attached to the service ladder, Figure 9A B. finally the monitoring equipment was inserted, Figure 9A C. and the manhole covered. Power was supplied by a

combination of battery power and on site AC power provided by a nearby outlet. The pressure/velocity probe was not placed directly on the bottom of the culvert as sand and gravel deposition would have covered the sensor. Instead the sensor was placed off center 8 ½ inches above the bottom. Depth and velocity were used to create discharge values as outlined above.

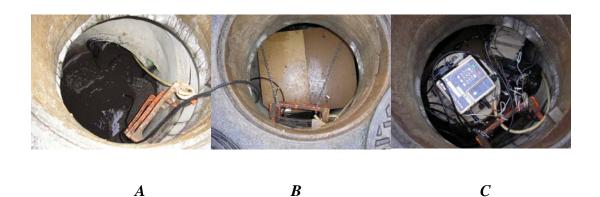


Figure 9A. Installation of Monitoring Equipment at Wenatchee Site

To assure quality control flow was quantified during actual runoff events and compared to recorded data. Flow measurements at the Spokane site were evaluated based on field measurements using stopwatch, bucket and graduated cylinder in much the same fashion as the laboratory calibration procedure.

Runoff Sampling/Analysis

Discreet field runoff samples were collected at the S1 site during a range of precipitation events including rain-on-snow, snow melt, and rain. In addition to these events three events were sampled along the treatment train at the W site. Runoff events were predicted by monitoring local weather forecasts and samples obtained by on site grab sample collection. The use of automated samplers, while potentially more convenient, proved to be impractical as freezing conditions would have disabled

sampling devices. Samples at the S1 site were collected by allowing flow over the weir to spill into plastic containers. Samples were typically taken at 5-10 minute intervals at the beginning of runoff to quantify the first flush. Near the end of the runoff period samples were taken on ½ hour to 2 hour intervals until runoff ceased or was minimal. Samples were returned to the laboratory and stored at 4 °C unacidified until preparation and analysis could be performed. Grab samples from the W treatment train were obtained by City of Wenatchee staff at set points illustrated in Figure 5A.

Prior to analysis samples were thoroughly shaken and 200-300 mL of sample was transferred to a mixing chamber. Mixing chamber design consisted of a 500 mL beaker with three baffles affixed to the outer wall. (Figure 10A). A stir bar was added and the sample agitated until solids were adequately suspended. The mixing chambers were constructed to prevent vortex formation and allow homogenous distribution of sample throughout the water column. This configuration allowed samples to include as uniform and representative a sample composition as possible.



Figure 10A. Mixing Chamber

Total solids were determined by filtration of a portion of sample, (ranging from 50-200 mL depending on the solids content), past a 1.2 μ m Whatman GF/C filter. Filters were pre-rinsed by passing 90-100 mL of reagent water through the filter and placing the

filter in an oven set to 110°C for at least one hour. This rinsing served to remove any loose particles that may have been present on the filters. Following drying filters were weighed, their weights and numbers cataloged, and placed in a desiccator until needed. The sample total solids concentration was determined by removing an aliquot of sample from a mixing chamber and passing it through a prepared filter. The filter was removed from the filter holder and placed in its aluminum weighing boat. Filters were then transferred to the oven and dried at 110°C for at least one hour. Dried filters were weighed and the new mass recorded. The added mass divided by the volume of sample passed through the filter yielded the total solids present in the sample. One in ten samples was analyzed in duplicate to check for repeatability.

Samples to be analyzed for total metals were hot acid digested using an aqua reagea digest similar to the soil samples. Approximately 100 mL of mixed sample was transferred to a 250 mL griffin beaker and four mL 1:1 HNO₃ and 4 mL 1:4 HCL was added. Samples were placed on a hot plate set to 85 °C and covered with elevated watch glasses to protect them from contamination and increase temperature to 95 °C. Vigorous boiling was avoided to minimize loss of the aqua reagea azeotrope. Following digestion samples were quantitatively transferred to a 100 mL volumetric flask and made to 100 mL with reagent water. Samples were allowed to stand overnight to settle solids. Following settling 5 mL of sample was transferred to a plastic centrifuge tube and placed in a 4 °C cold room until analysis by ICP-MS. One blank was run with each digest, however due to loss of some samples only two blanks were run over the course of the aqueous sample analysis. Two standard metal additions were run for quality control.

These additions consisted of addition of 1 mL of 5 mg/L metal solution to a sample and monitoring recovery.

Dissolved constituents, (CI⁻, dissolved metals, and organic matter), were determined by analysis of a filtered portion of the aforementioned sample. Samples were filtered using the same procedure as the laboratory samples. A 0.45 µm syringe filter was primed with 50 mL of reagent grade water and 15-20 mL of sample was transferred from the mixing beaker to the syringe. Filters were purged with 1-2 mL of sample and 15 mL of sample deposited in a plastic centrifuge tube. Samples to be analyzed for dissolved metals were acidified to pH <2 and analyzed by ICP-MS. Dissolved organic matter was determined in the same fashion as samples from the soil core leaching study using and HP 8453 spectrophotometer. Approximately ten percent of organic matter samples were run in duplicate to ensure repeatability and blanks were run to ensure adequately low baseline values.

Metals in Soils

Soil samples from the five field sites were taken at three distances and depths from the culvert inlets. Each culvert had three sample excavations taken at distances ranging from 0.3-4.6 m from where the runoff met the soil. Samples from TCA were taken at somewhat further distances from the culvert inlet as a large scour hole filled with cobbles directly below the culvert inlet prevented sampling near the inlet. To obtain soil samples an excavation 30.5-38 cm deep and approximately 0.3 m square was created using a stainless steel shovel. Excavation walls were scraped to expose undisturbed soil horizons and 10-20 g samples were taken at depths of 5, 15, and 25-30 cm using a plastic scoop. The scoop was thoroughly wiped between samples to prevent contamination.

Samples were stored in plastic Ziploc bags and transported to the lab for analysis. Three background excavations were also made at each site. One of the three excavations at each culvert and one of the three background excavations were sampled in triplicate at each depth.

Upon return to the lab the samples were dried to constant weight at 60 °C in a porcelain crucible, crushed using porcelain pestle with the crucible acting as the mortar, and passed through a #5 sieve to remove gravel sized particles. One gram of prepared soil was placed in a digest tube and 4 mL of 1:1 HNO₃ and 10 mL 1:4 HCl were added. The tubes were then placed in a block digester set to 85 °C. Watch glasses were placed over the digest tubes to allow the temperature to rise to 95 °C and prevent contamination. Samples were allowed to digest for half an hour. Following digestion samples were transferred from the tubes to 100 mL volumetric flasks and made to volume with reagent water. Samples were mixed by inversion, (repeated 20 times), and 10-15 mL was transferred to a centrifuge tube. Samples were centrifuged for 10 minutes for solids separation. Following centrifuging 1 mL of supernatant was transferred to a 15 mL plastic centrifuge tube and made to 5 mL using 2% HNO₃ and analyzed for zinc, copper, lead, and cadmium by ICP-MS. Blanks were run with each digest under the same procedures and conditions as samples. Spiked samples were run against unspiked samples in triplicate at least three times over the course of the soil digestions to ensure recovery.

Laboratory Study

Two soils from eastern Washington were used in a laboratory experiment to determine the effects of two road salts, MgCl₂ and NaCl, on heavy metal mobility. The soils used were chosen first for their exposure to high ADT which caused elevated metals

concentrations in the upper soil horizons as well as their contrasting physical characteristics. The first soil, taken from the Spokane site, was a well graded basic soil Figure 11A illustrates size distributions of the two soils) with high pH, and high organic matter, while the second soil from the Tri-Cities was a poorly graded acidic sandy soil.

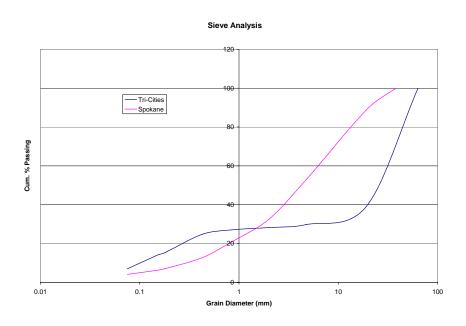


Figure 11A. Soil Size Distributions

Soil was removed from the upper horizons (0-5 cm) of these infiltration facilities where metals concentrations would be highest. Soils were collected using a stainless steel shovel for the Spokane site and a backhoe for the soils removed from the Richland site. After collection soils were stored at room temperature in 5 gallon plastic buckets until column construction. Soils were dried at 110 °C in plastic containers and sieved past a #25 stainless steel sieve. Soil columns were constructed by mixing soil with water until thick slurry was formed; this process was repeated in stages until a total mass of approximately 550 gm of soil was added to each column. Column pore volume was determined by assuming the soil specific gravity to be 2.65 and comparing the theoretical

volume of each column to the measured soil volume. Pore volumes for all columns was assumed to be 225 mL as that was the average for all columns.

An average pore volume of 225 mL was assumed for each column. Following construction columns were allowed to sit 48 hours to settle. Columns were attached to a vacuum apparatus consisting of vacuum flasks attached to a vacuum pump (Figure 29), and a vacuum of 5 inHg or less was applied to facilitate leaching.



Figure 12A. Laboratory Soil Column Setup

Flow through some of the columns was variable especially the Richland soil, which was very sandy and had much higher hydraulic conductivity than the Spokane soil. In an effort to control the total number of pore volumes infiltrated columns with higher hydraulic conductivity were periodically removed from the vacuum to allow accumulation of leachate from the lower hydraulic conductivity columns. All experiments were conducted at 4 °C to simulate actual temperature conditions in the field during cold weather runoff. Each core was exposed to a series of leachates containing a range of salt concentrations. The first three pore volumes through every column consisted of an application of deionized water and were intended to stabilize flows and remove unstable constituents. The following 4-5 pore volumes were MgCl₂ or NaCl solutions with Cl² concentrations of 0.1 M, 0.05 M or 0.025 M respectively. These chloride values

were based on actual chloride concentrations determined from field sample analysis during winter runoff events. The 0.1 M concentration represents the peak chloride concentrations observed during field sampling, while the 0.05 M and 0.025 M concentrations are representative of more dilute runoff sampled at later stages of runoff. The final 2-3 pore volumes of leachate consisted of deionized water. This flush of low ionic strength leachate was intended to simulate later stages of a runoff event when less salty runoff reaches the infiltration facility. Leachate passing through the soil core was collected on timed intervals, filtered, and analyzed for metals, chloride, organic matter, and pH.

Leachate preparation consisted of filtration past a 0.45 μm PTFE filter. Each filter was purged with 50 mL of reagent water to remove any contamination and to prime the filter. Four mL of filtered sample was acidified to pH <2 by addition of HNO₃ and analyzed by ICP-MS for dissolved metals (Cu, Cd, Pb, and Zn). A second portion of the sample was set aside and diluted for chloride and organic matter determination. Portions of sample diluted to 1/10 concentration were placed in 2 mL vials and analyzed for chloride by ion chromatography. A third aliquot of filtered sample was diluted to either 1/10 or 1/50 concentration and analyzed for dissolved organic matter using absorption of uv at 254 nm. Analysis of absorption consisted of powering up the HP 8453 spectrophotometer and allowing 10-20 min for warm up of a deuterium lamp. A quartz cuvette was rinsed twice with deionized water and once with alcohol to remove any internal contamination. The outside of the cuvette was then wiped down using a chimwipe. A baseline values was obtained by purging the cuvette twice with reagent water and finally filled with reagent water and taking a reading. The procedure for

analyzing samples was analogous to the blank preparation: rinsing, wiping, and purging with sample twice before the actual measurement was taken. Blanks were run every 10-15 samples to ensure that contamination was minimal and background levels were low. Duplicate samples were also analyzed to ensure analysis precision. Absorbance was reported in units of cm⁻¹.

pH was measured immediately following sample collection by an Accumet AR 10 pH meter.

One column from Richland and one column from Spokane were leached using only deionized water. This provided baseline organic matter and metals release data to compare with salt leach data.

Standard/Glassware Preparation

All standards were created using reagent water with resistance of >18 M Ω cm generated by a Barnstead Epure series 1090. Acids used were Fisher scientific trace metal grade HCl and HNO₃. All metals standards were prepared using dilutions of 1000 μ g/L solutions and were suspended in a 2% HNO₃ matrix. All glassware used in metals digestion and analysis were washed with sodium hypochlorite bleach, triple rinsed with tap water, triple rinsed with deionized water, and soaked in a 1:1 HNO₃ acid bath for 1-14 hours (in most cases overnight). Following soaking glassware was triple rinsed using reagent water and dried.

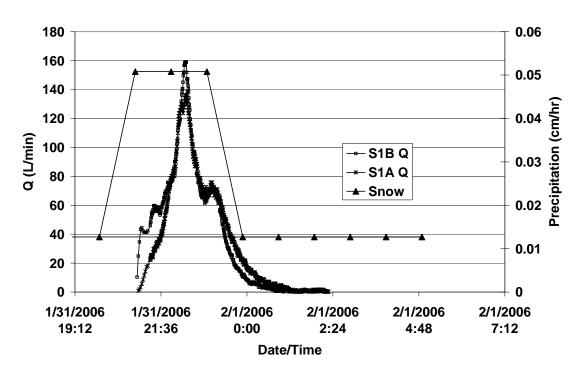
APPENDIX B

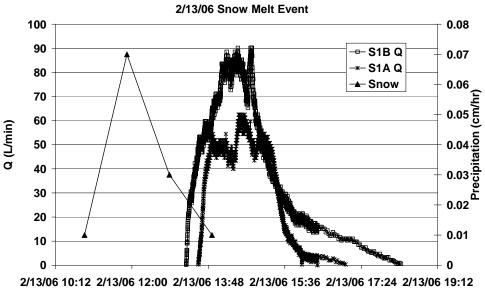
PRECIPITATION AND RUNOFF CHARTS

The following figures were taken from reduced data from the S1 and W field sites. The first ten figures represent runoff monitored at S1 and are arranged by order of precipitation event: snow melt, rain, and rain-on-snow. S1AQ and S1BQ represent flow rates monitored at both culverts at the S1 site as described in Appendix A *field sites*. (Site selection).

The final two figures of this appendix depict runoff at the W site. The precipitation causing these runoff values is undetermined due to the lack of instrumentation capable of differentiating precipitation type. However, it can be assumed that precipitation is rain due to temperatures above 0° C.

1/31/06 Snow Melt Event

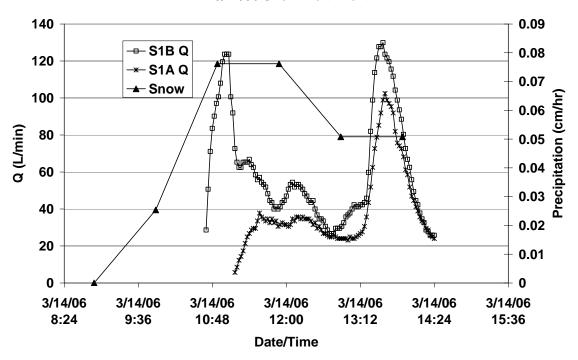




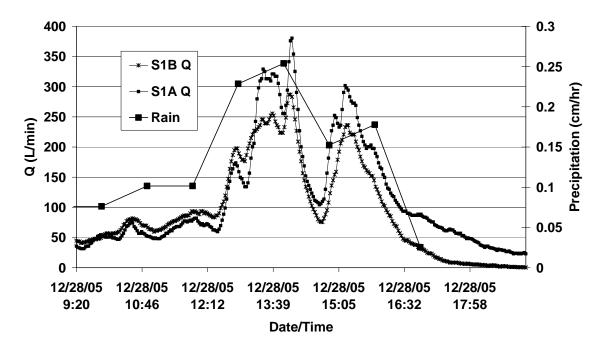
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Date/Time

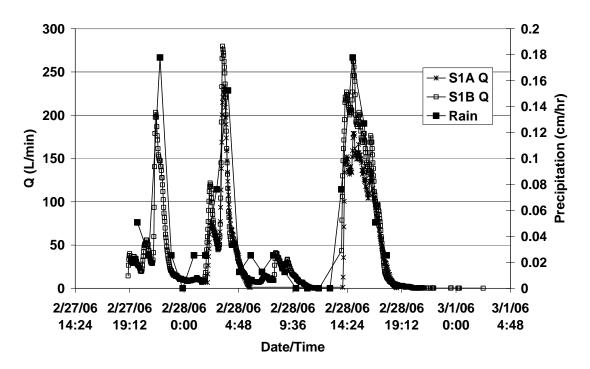
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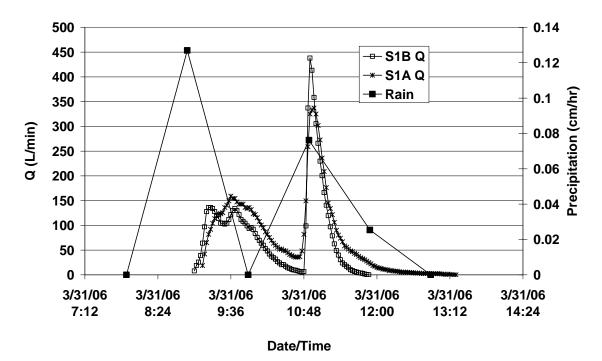
12/28/06 Rain Event



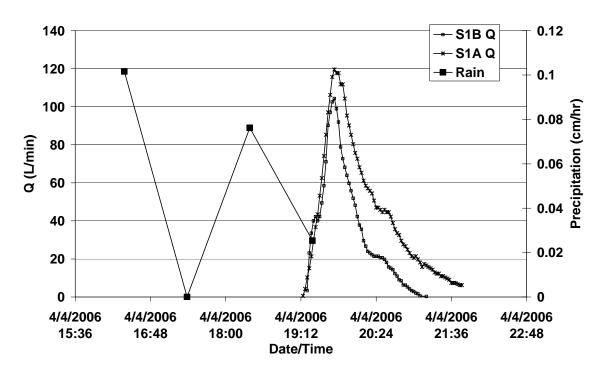
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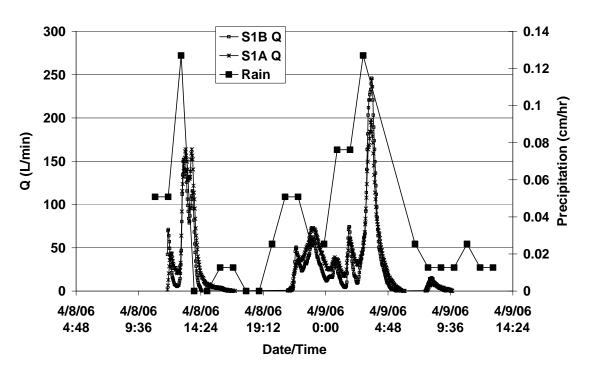
3/31/06 Rain Event



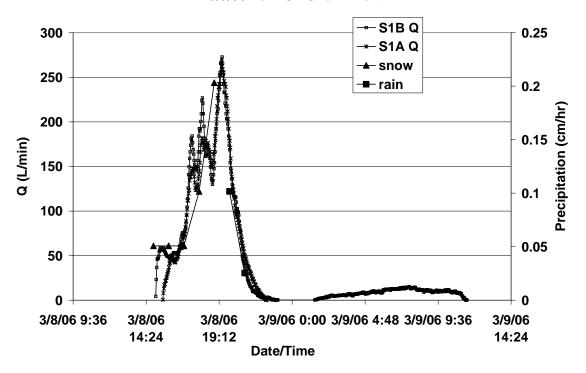
4/4/06 Rain Event



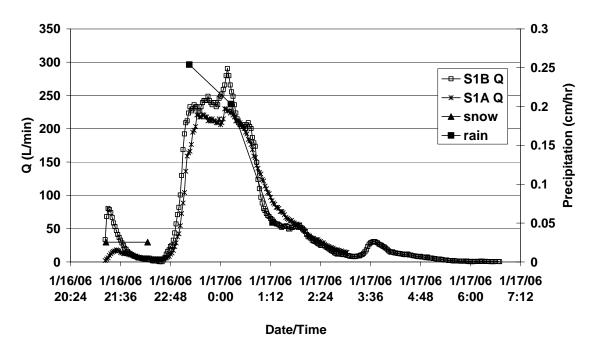
4/8/06 Rain Event



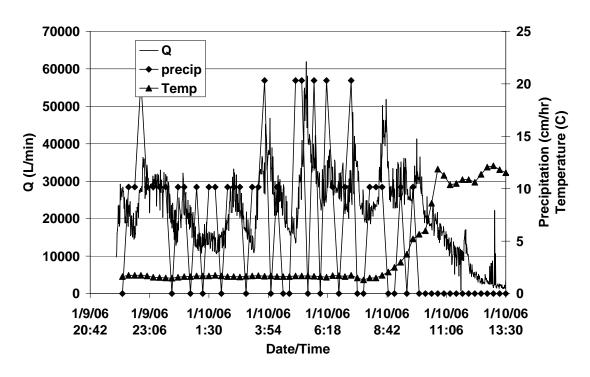
3/9/06 Rain-On-Snow Event



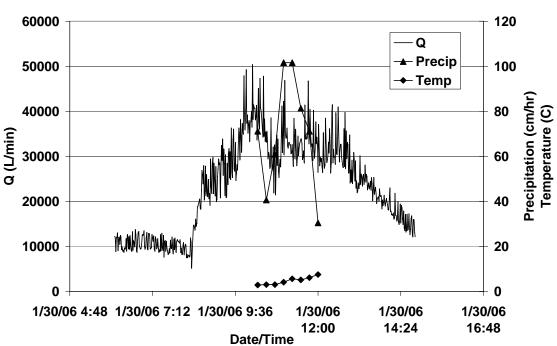
1/17/2006 Rain-On-Snow Event



1/10/06 W





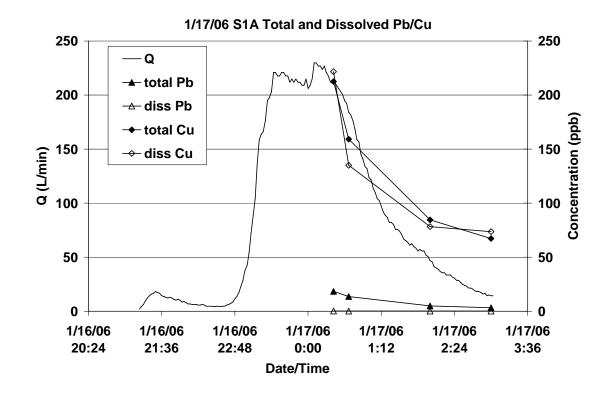


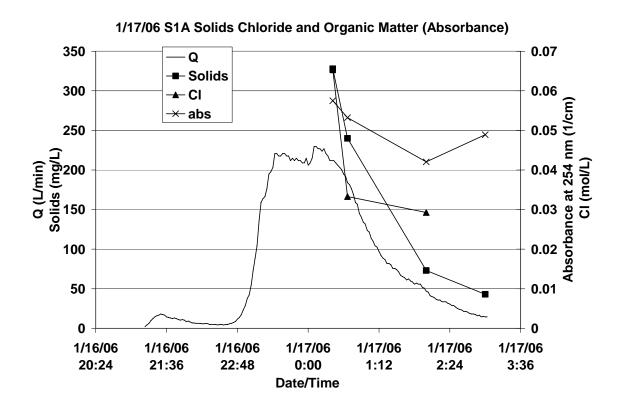
APPENDIX C

FIELD RUNOF TOTAL AND DISSOLVED METALS, ORGANIC MATTER, CHLORIDE, AND SOLIDS

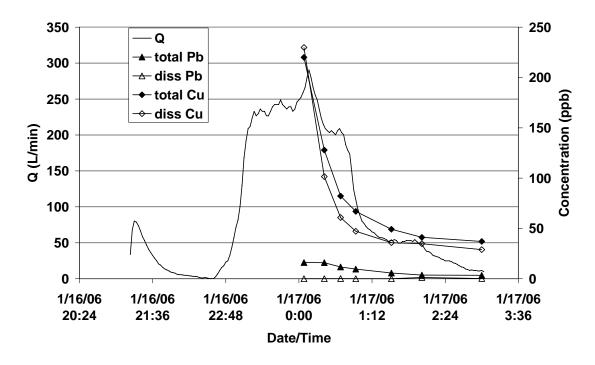
The following figures illustrate concentrations of constituents quantified from samples taken at the S1 and W sites over the winter and spring of 2006. The first 20 figures are from the S1 site and are broken down by event and culvert inlet. Results from each event and culvert sampled are further broken down with total and dissolved Cu and Pb in the first figure, and solids, chloride, and organic matter (measured as absorbance) in the second figure.

The final 3 figures of the appendix represent concentrations of copper, lead, and solids measured from samples taken at the W site. Samples were obtained along the treatment train described in Appendix A *Site Selection*.

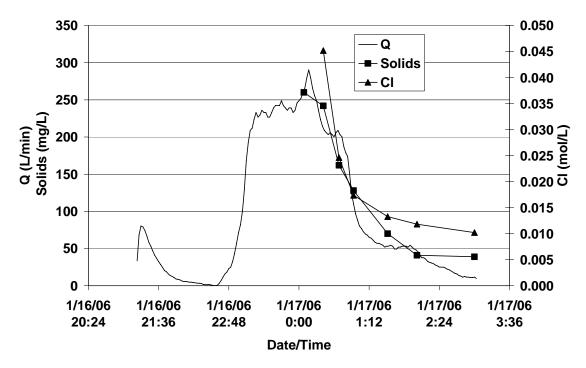




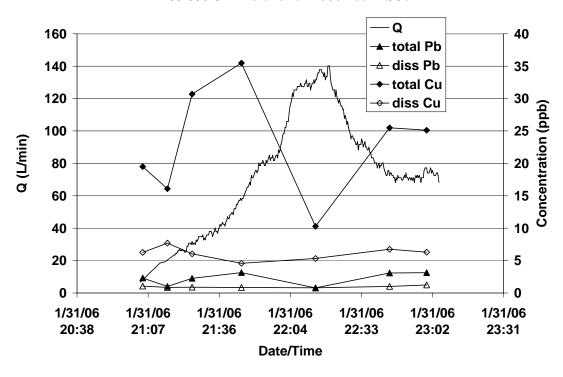
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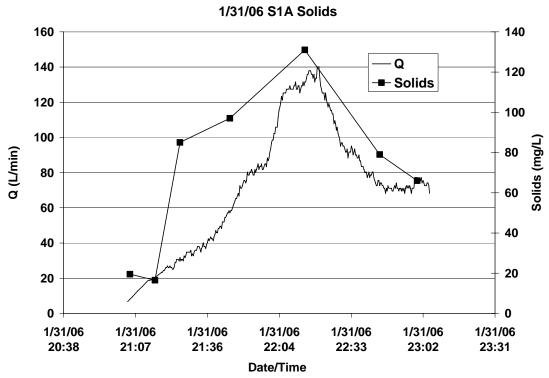


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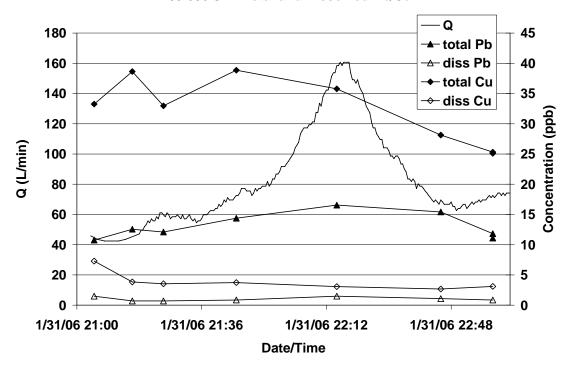


1/31/06 S1A Total and Dissolved Pb/Cu

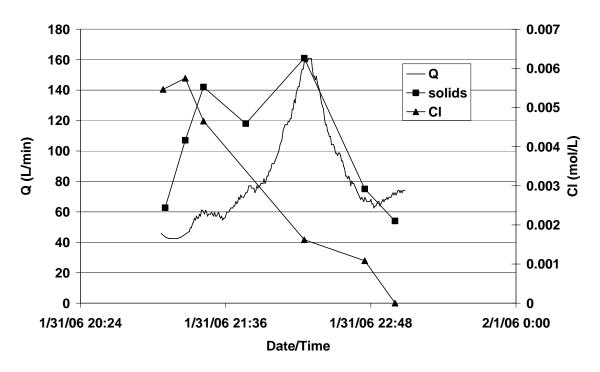




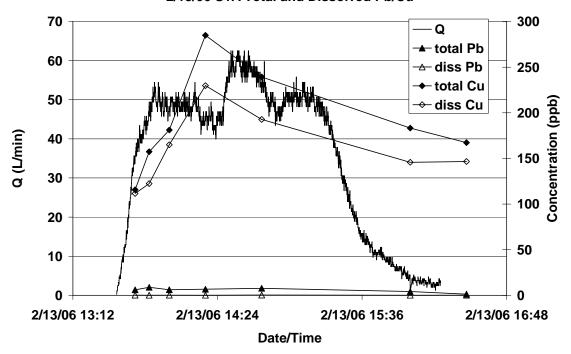
1/31/06 S1B Total and Dissolved Pb/Cu



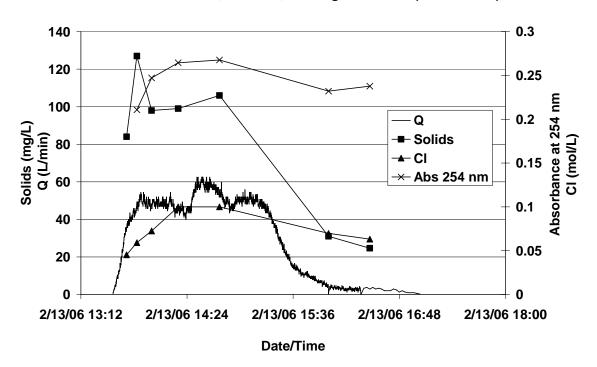
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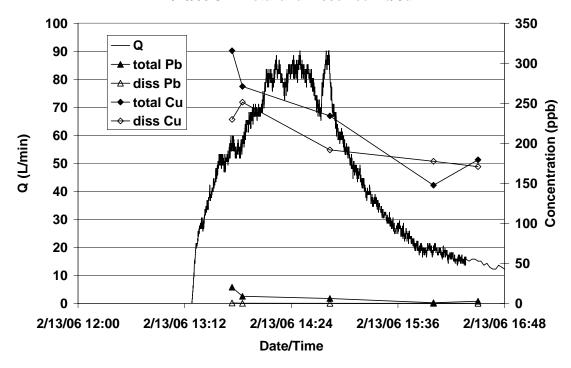




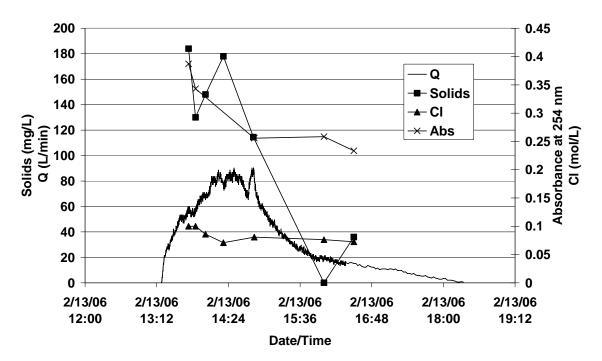
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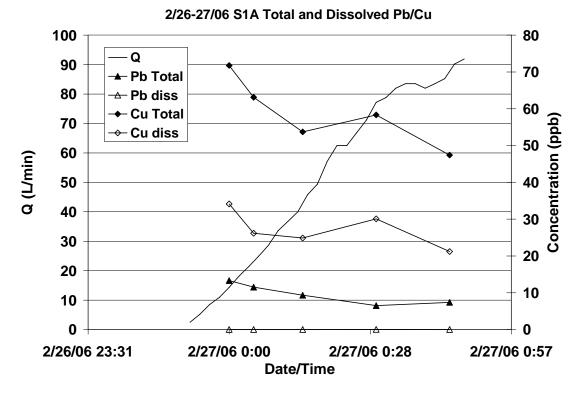


2/13/06 S1B Total and Dissolved Pb/Cu



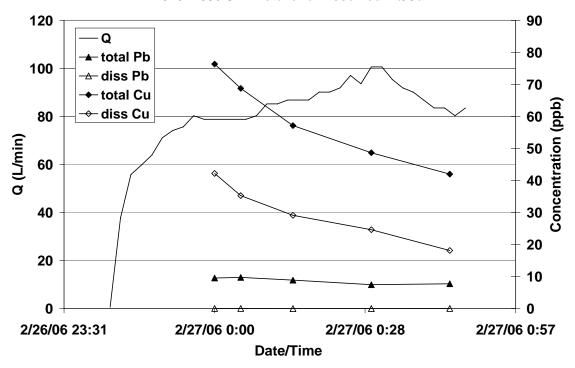
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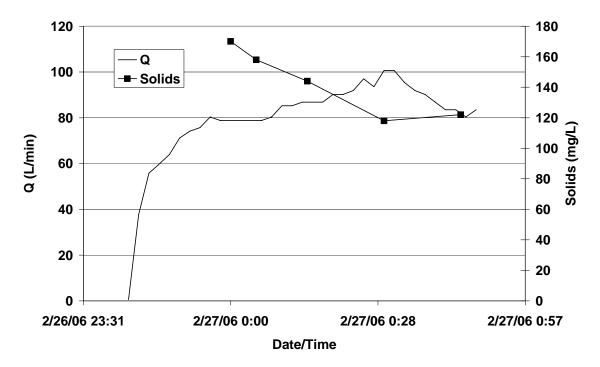




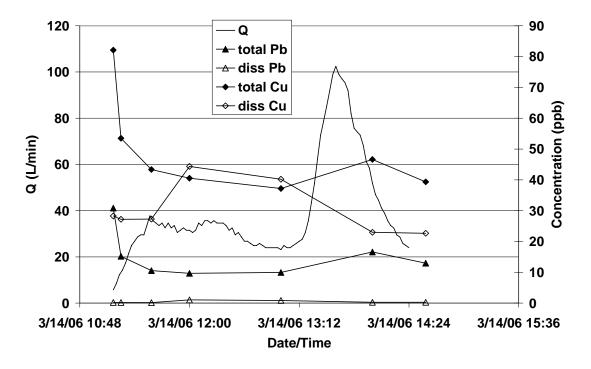
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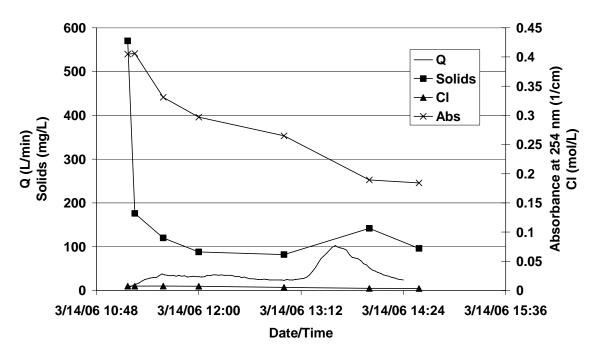
2/26-27/06 S1B Solids



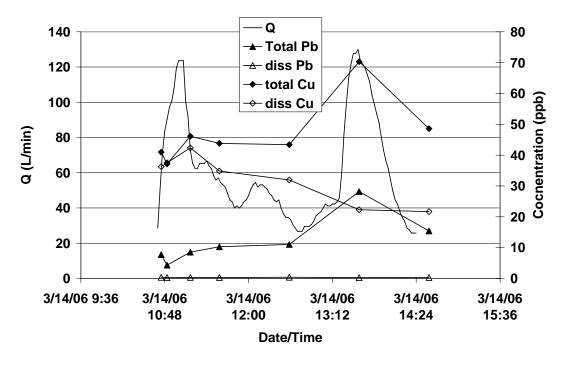
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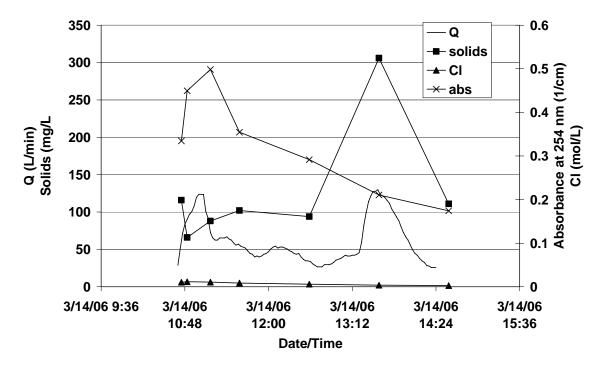
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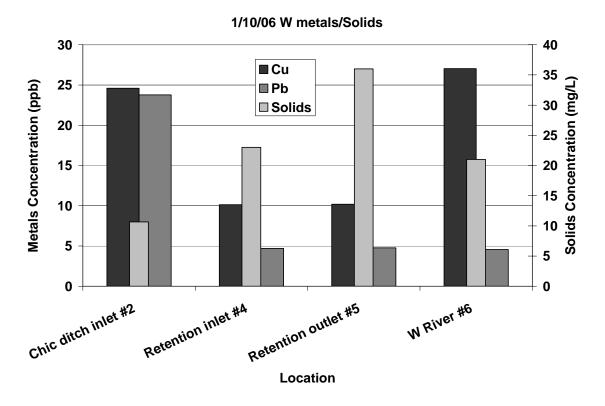


3/14/06 S1B Total and Dissolved Pb/Cu

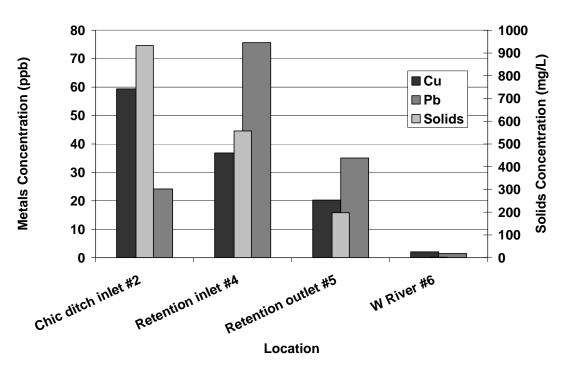


3/14/06 S1B Solids, Chloride, and Organic Matter (Absorbance)

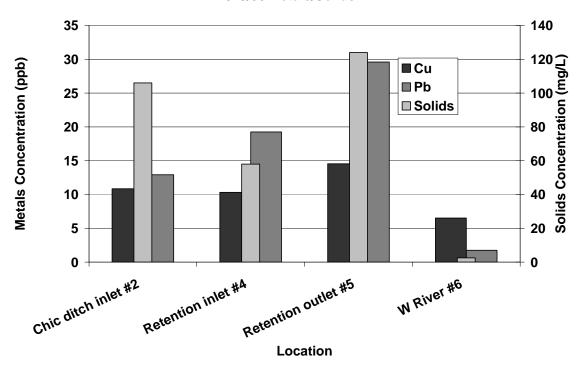








2/28/06 Metals/Solids



APPENDIX D

SOIL METALS CONCENTRATIONS

The following figures represent soil metals concentrations at all five field sites. Figures contain sample location names composed of multiple letters and numbers. The ordering scheme of these descriptors is as follows. The first series of letters and numbers, (S1, S2, TC, W, and ML) represent site locations as described earlier. The following letter designates a culvert inlet if there are multiple culverts sampled at a site. The number following the culvert designator indicates the distance the excavation was made from the culvert. 1, 2, and 3 indicating distances of 1-2 m, 3-4 m, and 4-6 m respectively. The final letter/number designation (D1, D2, D3) represent depths samples were taken from. Figure 1 illustrates the above explanation. S1A1D1 represents a sample taken at the Spokane Valley site, from culvert "A", at excavation 1 (1-2 m from the culvert), at D1 (a depth of 0-5 cm).

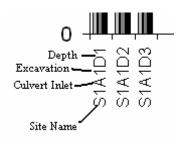
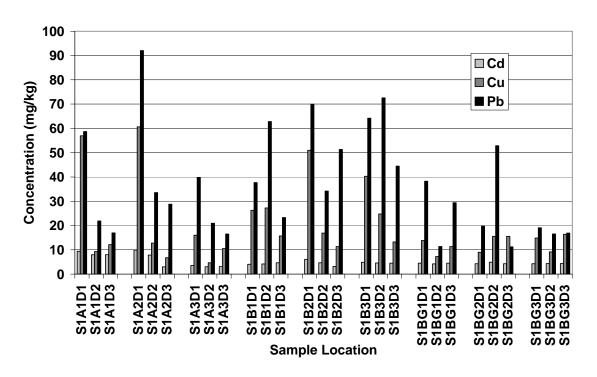
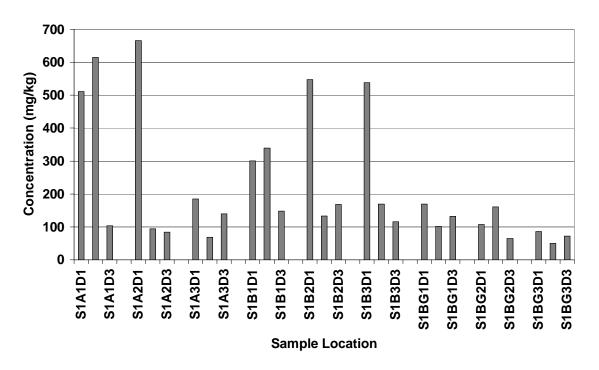


Figure 1. Soil Sample Descriptor

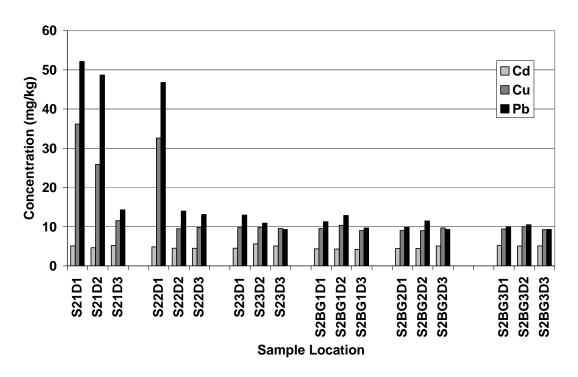
S1 Metals Concentrations



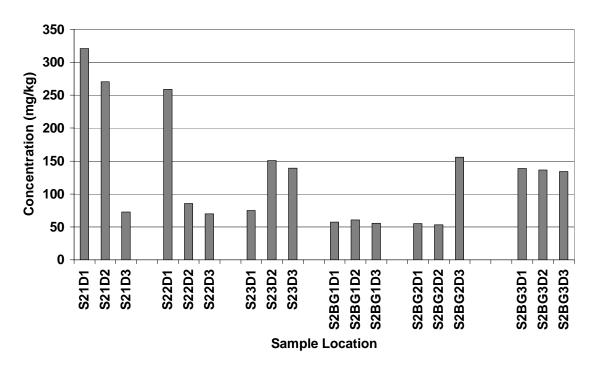
S1 Zn Concentrations



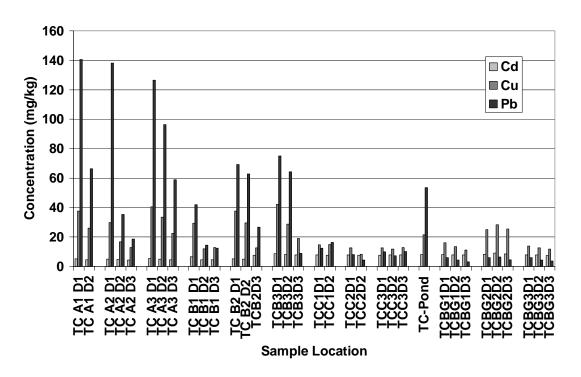
S2 Metals Concentrations



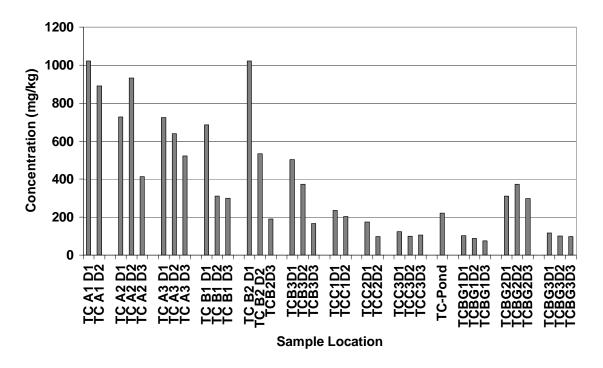
S2 Zn Concentrations



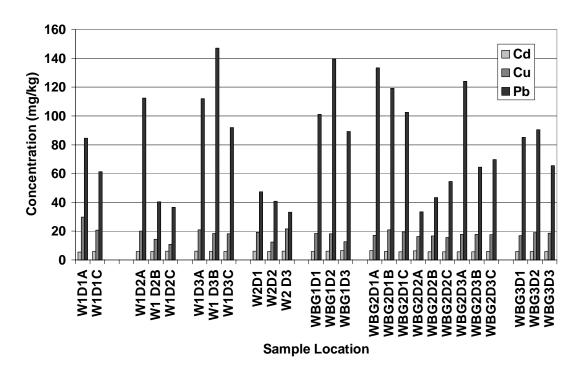
TC Metals Concentrations



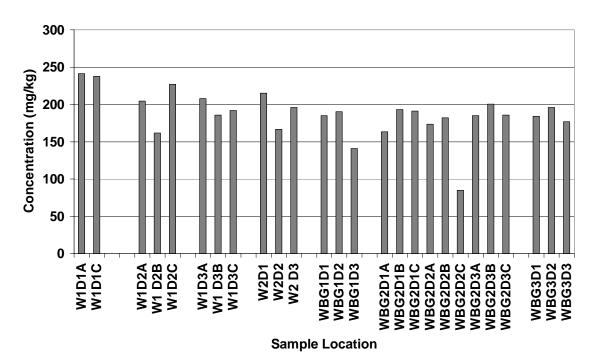
TC Zn Concentrations



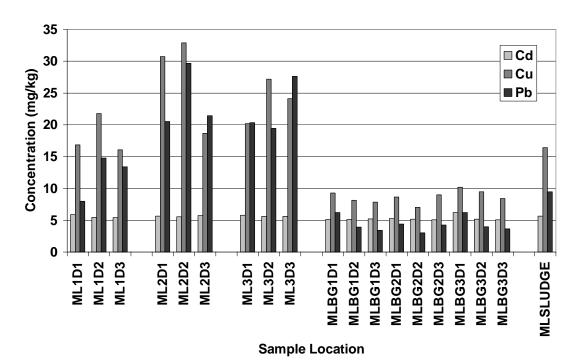
W Metals Concentrations



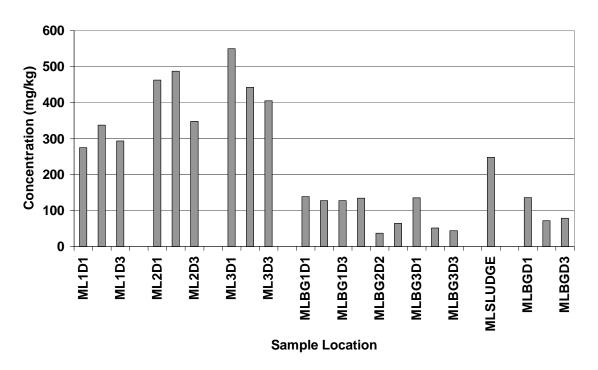
W Zn Concentrations



ML Metals Concentrations



ML Zn Concentrations

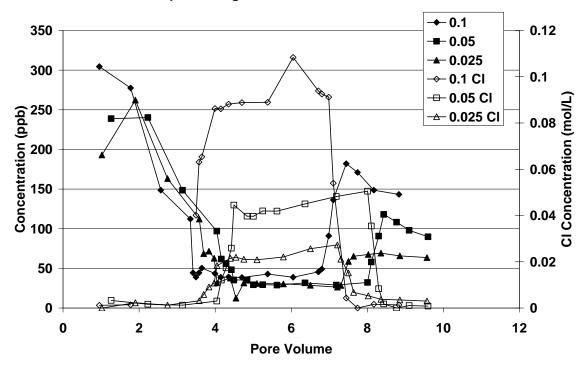


APPENDIX E

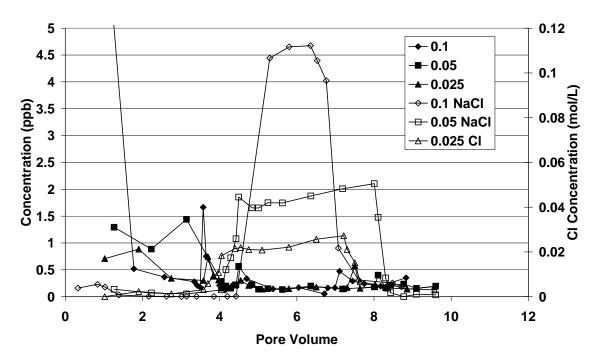
LABORATORY STUDY FIGURES

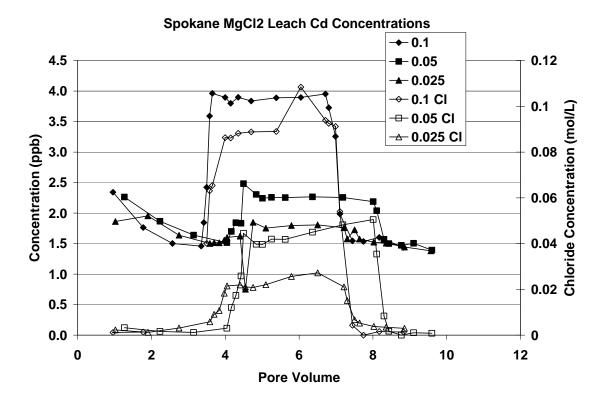
The following figures illustrate results taken from the laboratory leach study. The figures are broken down by core leach, with the three metal species (Cu, Pb, and Cd) and absorbance all plotted versus pore volume. Chloride concentrations have also been included to illustrate the effects of salt solutions. The final two figures of this appendix are the results of background leaches performed on both soil types.

Spokane MgCl2 Leach Cu Concentrations

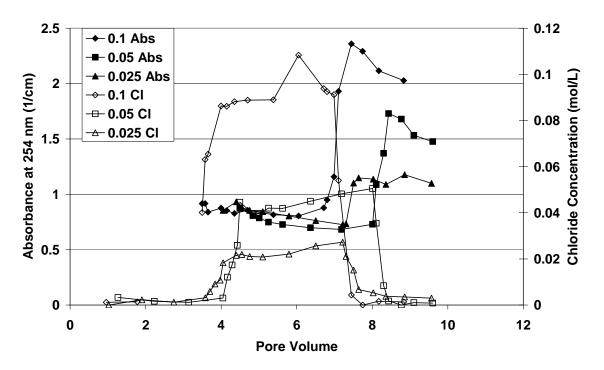


Spokane MgCl2 Leach Pb Concentrations

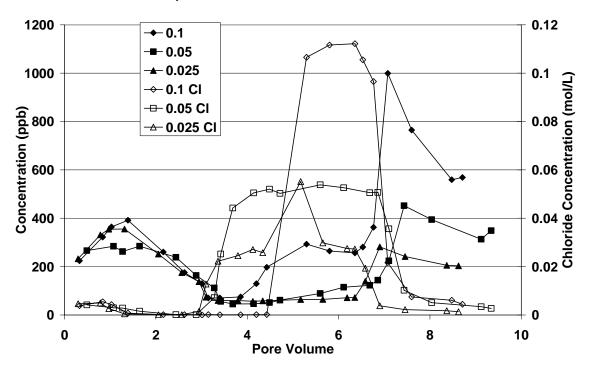




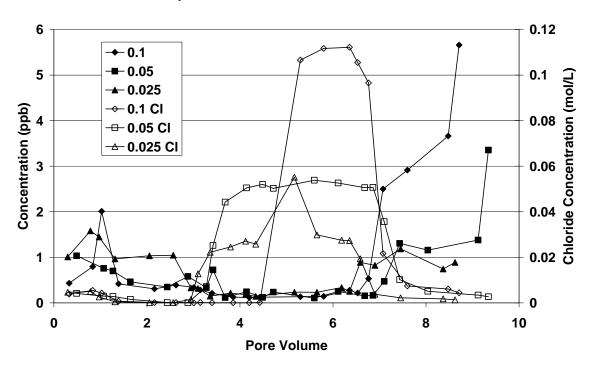
Spokane MgCl2 Leach Absorbance



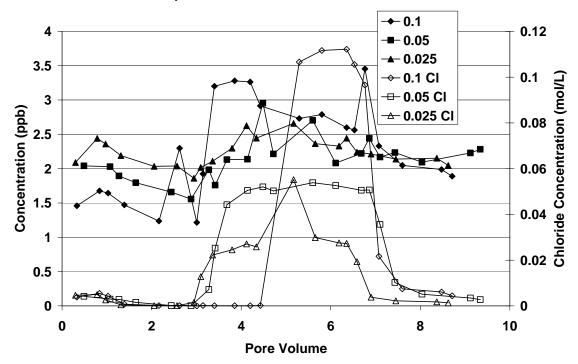
Spokane NaCl Leach Cu Concentrations



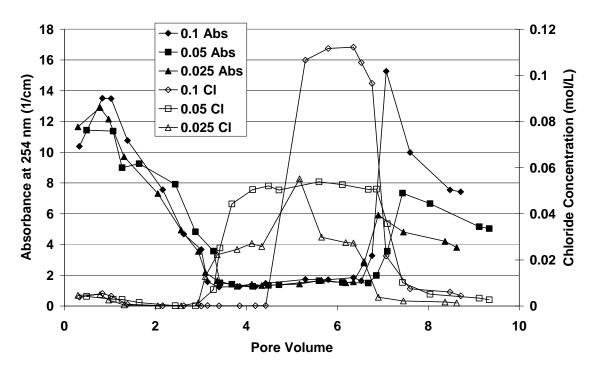
Spokane NaCl Leach Pb Concentrations



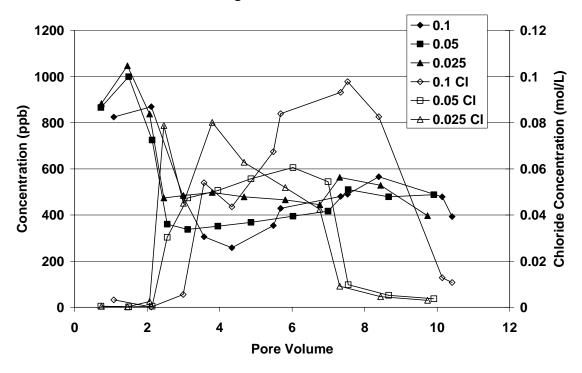
Spokane NaCl Leach Cd Concentrations



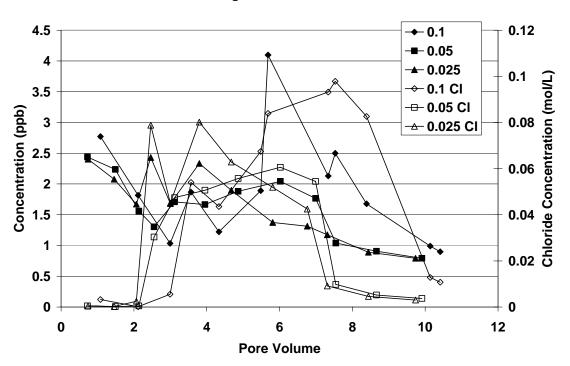
Spokane NaCl Leach Absorbance



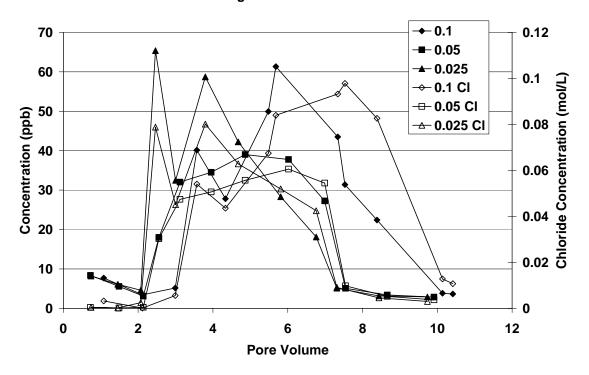
Richland MgCl2 Leach Cu Concentrations



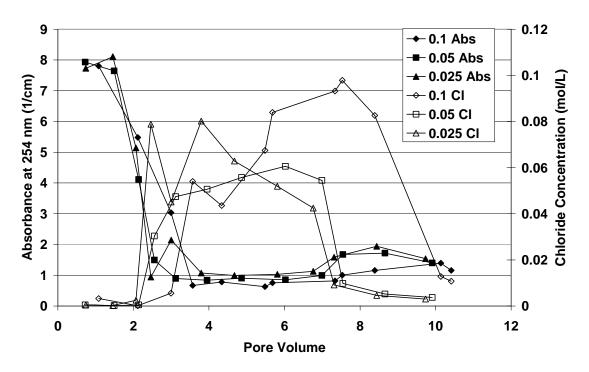
Richland MgCl2 Leach Pb Concentrations

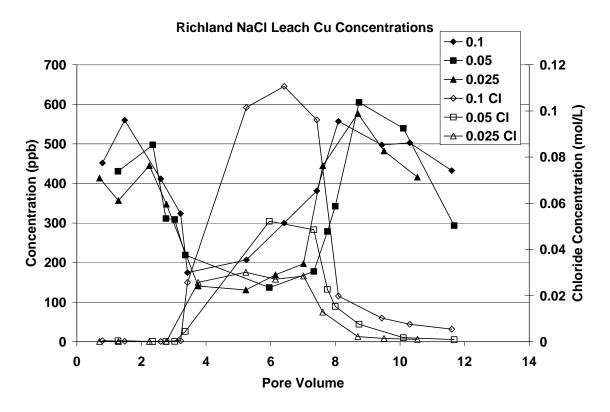


Richland MgCl2 Leach Cd Concentrations

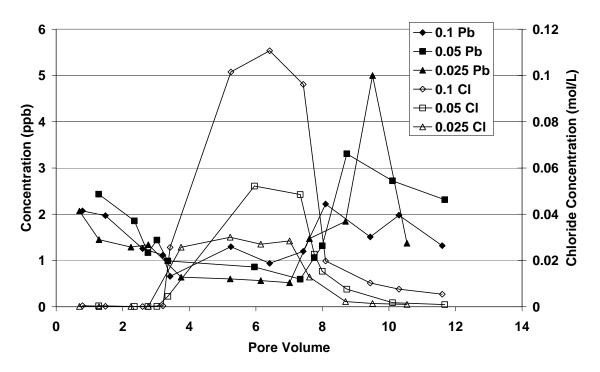


Richland MgCl2 Leach Absorbance

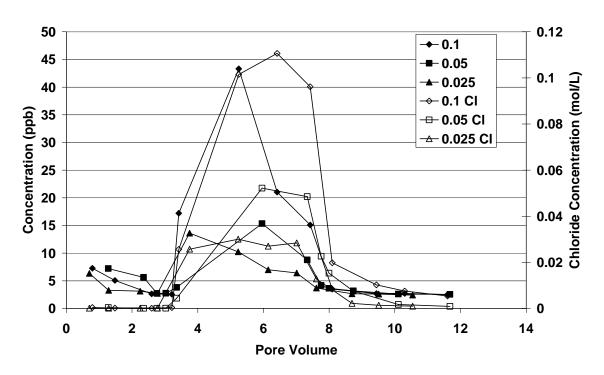




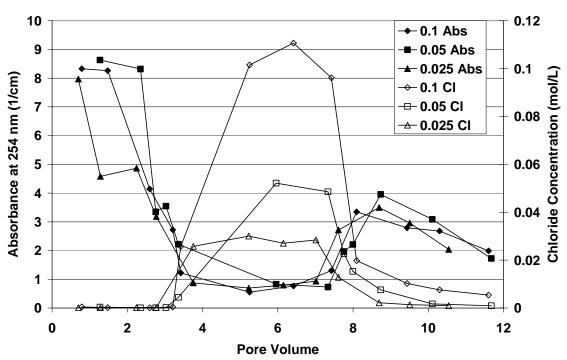
Richland NaCl Leach Pb Concentrations



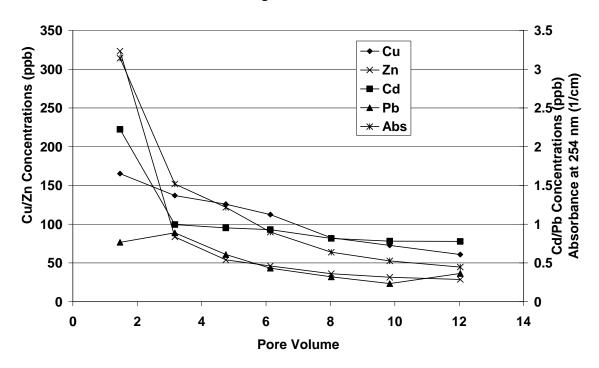
Richland NaCl Leach Cd Concentrations



Richland NaCl Absorbance



Richland Background Metals/Absorbance



Spokane Background Metals/Absorbance

