

Environmental impact of the historical slag pile at Davenport, Iowa, United States: Trace metal contamination in soils and terrestrial vegetation

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Abstract: Metal processing has played a key role in the history of the upper Midwest. In addition to economic prosperity, the legacy of large metal refining operations includes soil and water pollution that is often not immediately apparent. One example is the disposal of iron (Fe) slag on the east bank of Carp Lake, a former sand quarry located adjacent to the Mississippi River in Davenport, Iowa, United States. Iron foundry slag is known to contain high concentrations of Fe as well as trace metals that can include cadmium (Cd), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn), among others. Despite efforts to place soil and revegetate the pile, more than eight decades of erosion have exposed raw slag. To understand the composition of this slag pile and relationships with surrounding environment, samples of slag, soil, and vegetation were collected from sites on the slag pile and along the bank of Carp Lake. Iron was the predominant metal found in slag pile and Carp Lake bank (CLB) soils. Both Pb and Fe concentrations exceeded the US Environmental Protection Agency (USEPA) standards as well as reference site concentrations. Metal concentrations were found to vary across sample sites. This is indicative of the heterogeneous nature of the slag pile. In terrestrial vegetation samples, only Cd was found at actionable levels in the leaves of Amur honeysuckle (*Lonicera maackii*) and dogwood (*Cornus sericea*). Metals in the CLB soils were within USEPA recommended ranges. However, the trace metal concentrations in CLB soils were higher than the reference site. Further, the elevated Pb concentration in the bank soil located closest to the slag pile supports the suspicion that the slag pile is a source of metal contamination. We hope the results of this study will provide reference for environmental quality assessments and will help decision-makers to manage this and similar locations.

Key words: contamination—heavy metal—pollution—slag pile—soil—vegetation

Iron (Fe) smelting produces a large quantity of slag, which can be a source of metal contamination in the soil and water. Generally, the historic slag contains higher concentrations of heavy metals as compared to slag produced using modern extraction processes (Piatak et al. 2012; Vitkova et al. 2010). Previously, slag was frequently considered to be inert and, in the absence of strict regulations, was often dumped without much consideration to the risks of contaminating the surrounding soils, vegetation, or surface water (Evans 1989; Piatak et al.

2015). However, more contemporary studies have confirmed that the weathering of these deposits can mobilize metals from the slag (Manz and Castro 1997; Teng et al. 2003; Vdovic et al. 2006; Douay et al. 2008).

The size and corresponding impacts of slag piles are broad and diverse. For example, some of the largest slag piles include the Coniston slag pile in Sudbury, Ontario, Canada (400,000 m² [~98 ac]), one in Silesia, Poland (150,000 m² [~37 ac]), and the Ely Copper Mine in Vershire, Vermont, United States (7,300,000 m² [1,804 ac]) (Seal II et

al. 2010; Kierczak et al. 2014). In contrast, the number and extent of small and midsize slag piles remains unknown, though their potential environmental significance is likely to be substantial. In the United States, more than 70% of sites with soil pollution are reported to contain heavy metal and/or metalloid contamination (USEPA 2014). Unfortunately, studies focused on the environmental aspect of historical slags are very limited (Piatak and Seal 2012). The identification of such small to midsize slag piles and understanding the potential risk and environmental impact on surrounding ecosystems are critical to the development of future response strategies.

This study was conducted on an Fe foundry slag waste pile located adjacent to Carp Lake, in Davenport, Iowa, United States (figure 1). Carp Lake (3.72 ha [~9 ac]) began as a sand quarry that was established in the 1920s. Based on a review of historic aerial imagery, the general dimensions of the lake have remained approximately the same since its initial construction. Carp Lake is located adjacent to the Mississippi River and is in the 100-year floodplain. The site is known to flood frequently and becomes connected to the Mississippi River during high water events.

Beginning in the 1930s, slag from a local Fe foundry was disposed of on the east shore of Carp Lake. The placement of slag at the site continued through the early 1980s. In the absence of historic records, the exact timeline of slag disposal cannot be known. Likewise, it is not possible to know the exact procedures that were used when placing the waste slag; however, based on the vintage of the deposits and the lack of waste management regulations, it is assumed that waste was simply dumped on the lakeshore on an intermittent basis.

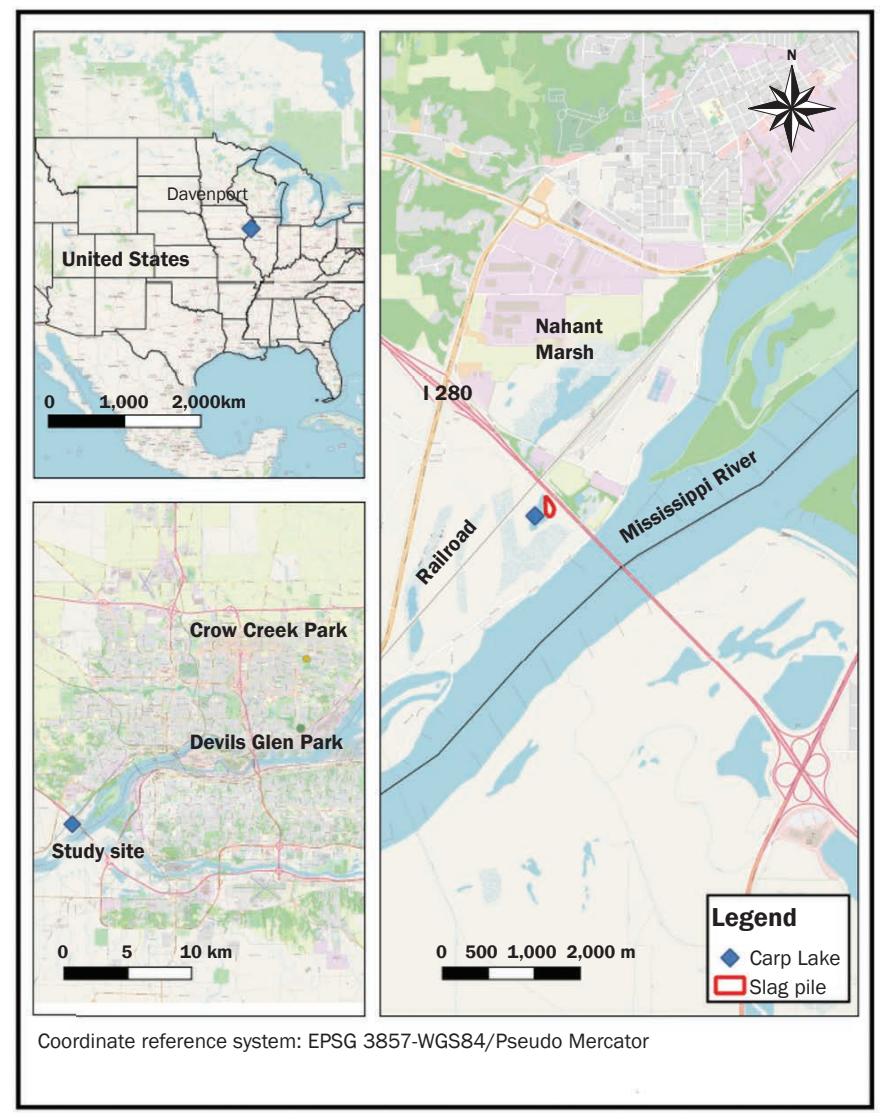
The site (figure 1) consists of three adjacent parcels with a gross area of 16.47 ha (40.69 ac). The property is undeveloped and heavily vegetated. Grey-black colored, glassy, fragmented, porous slag stones are scattered across the slag pile. These slag pieces range from large boulders to sand-sized particles. Currently, a portion of the slag pile is covered with soil and vegetation while others

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Figure 1

Location of the study site in Davenport, Iowa, United States.



remain exposed to the atmosphere. The majority of slag is placed along the east bank of Carp Lake; however, the full extent of slag placement on the site is not known. Likewise, no study has ever been undertaken to determine the composition of this slag deposit and its potential impacts on the surrounding environment.

Metals that are often associated with Fe slag include cadmium (Cd), copper (Cu), Fe, lead (Pb), nickel (Ni), and zinc (Zn), among others (Hybska et al. 2017; Eberhardt and Benson 2008). Once released into the environment, metals can become sorbed onto soil particles, enter the water column through leaching, and/or accumulate in plant tissue (Chopin et al. 2003). Metals are of special

relevance due to the potential threat they pose to human and environmental receptors (Parson 2001; Granero and Domingo 2002; Pruvot et al. 2006; Zhao et al. 2009). For example, elevated levels of Pb can cause damage to the central nervous system and kidneys (ATSDR 2007). Likewise, Pb and Cd rank second and seventh, respectively, based on their frequency, toxicity, and potential for human exposure as per the Priority List of Hazardous Substances, US Department of Health and Human Services (ATSDR 2007).

The exact impacts of any slag pile are difficult to know without extensive lab studies. This is a function of the heterogeneity of the deposits as well as the unique environmental conditions in which they are found. As

a result, the potential environmental impacts of each deposit must be assessed based on the composition of the slag material, land use, and environmental conditions (Lottermoser 2002; Navarro et al. 2008).

Based on the proximity of this slag pile to Carp Lake, the Nahant Marsh wetland complex, and the Mississippi River, as well as the frequency of flooding at the site, this study was conducted to determine the concentrations of metals in the slag pile, surrounding soils, and in vegetation. Additional work has been done on the extent of aquatic impacts, which will be reported separately.

Materials and Methods

Between June of 2017 and August of 2018, samples of soil, slag, and vegetation were collected at 10 sites located on the slag pile and at three sites situated around the bank of Carp Lake. Sample locations were established using a survey-grade global positioning system (GPS) and were flagged for future reference (figure 2). A 1 m² (10.7 ft²) frame was used to standardize the sampling of vegetation, slag, and soils.

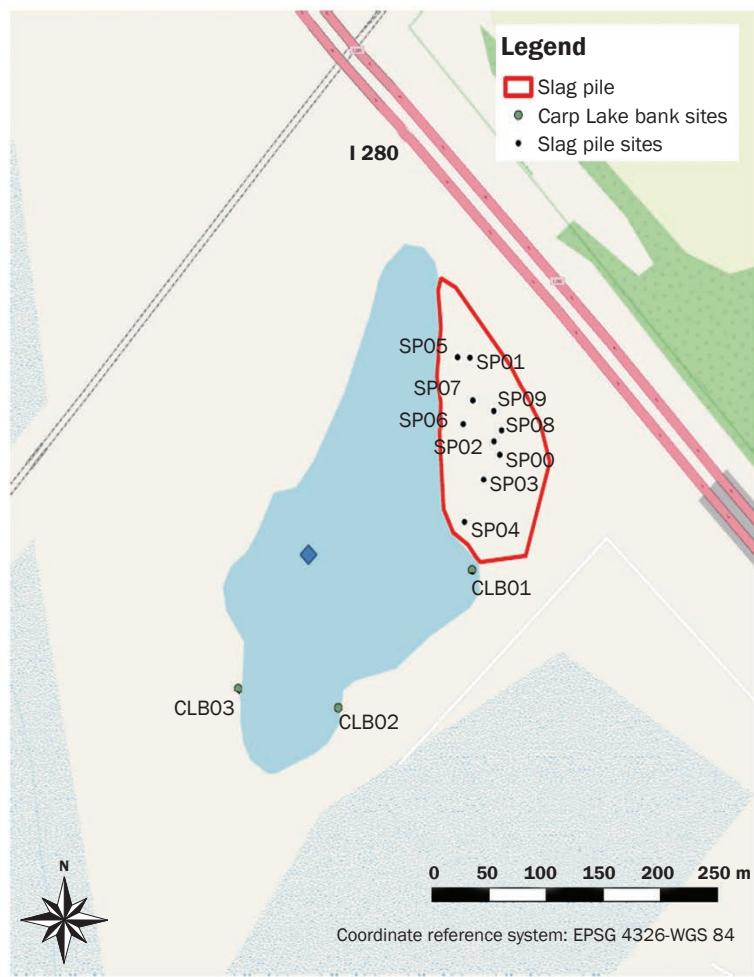
Samples of surface soil (top 7.6 cm [\sim 3 in]) were collected from 10 randomly selected locations on the slag pile. At sites SP00, SP07, SP08, and SP09, slag was visible on the surface. At these four sites, samples of slag were broken off using a shovel. Leaf samples were collected from vegetation at SP04, SP05, SP06, and SP07. Additionally, soil was collected at three sites on the bank of Carp Lake (CLB). CLB sampling sites were selected based on their distance from the slag pile.

All samples were dried at 103°C to 105°C (\sim 217°F to 221°F) for two hours and allowed to cool in a desiccator before being homogenized and separated into 1.00 g (\sim 0.03 oz) subsamples. Each subsample was then digested through the serial addition of 70% nitric acid (HNO₃) and 30% hydrogen peroxide (H₂O₂) under heat as per US Environmental Protection Agency Method 3050B (USEPA 1996). After digestion, the samples were diluted to 100 mL (\sim 3.4 fl oz) using Type 2 laboratory water. The extractable concentrations of Cd, Cu, Fe, Pb, Ni, and Zn were then determined according to USEPA Method 7000B (USEPA 1996).

Additionally, samples of slag were examined using a Leica T490-PL 40X to 1,000X compound trinocular microscope (Leica Microsystems, Buffalo Grove, Illinois). The system was equipped with a 16 MP digital

Figure 2

Map of sampling sites located in the vicinity of Carp Lake, Davenport, Iowa, United States.



camera with a 0.5X reduction lens to allow for a wider field of view. An indirect light source was used to minimize the appearance of shadows on the faceted surfaces of slag samples. A ground glass calibration slide (AmScope, Irvine, California) was used to calibrate measurements made with the microscope. Once images were acquired, ImageJ (Abramoff et al. 2004) was used for all image postprocessing.

Data reduction, statistical analyses, and quality assurance/quality control procedures were performed according to USEPA guidelines (USEPA 2000). Analytic quality assurance/quality control procedures included the collection and analysis of triplicate samples, the analysis of matrix blanks, and standard additions. The data analysis for this paper was generated using the Real Statistics Resource Pack Software. The significance of the relationships between metal concentrations were determined by using Pearson's correlation test. For comparison purposes, data were collected at reference sites located at Crow Creek Park and Devils Glen Park in Bettendorf, Iowa (figure 1).

Results and Discussion

Slag Pile Soils. The concentrations of extractable metals in slag pile and reference soils are presented in table 1. Samples from the slag pile had measurable concentrations of Fe, Ni, Pb, and Zn. Iron and Zn were most abundant among the metals that were measured. Copper was not observed at concentrations above method detection levels, and Cd was measurable only at SP00. The heterogeneity of metal distributions in the slag pile soils was consistent with findings from other studies in which varied concentrations of metals were found in soils that were contaminated by anthropogenic activities (Chopin et al. 2003; Tam and Wong 2000; Horvath and Gruiz 1996).

Among these metals, the concentrations of Pb and Zn appeared to follow the same trends with notably high concentrations at SP00 and SP07 as presented in figure 3. Based on Pearson correlation analysis for each metal-metal combination in the slag pile soils, Pb was positively correlated ($P < 0.05$) with Zn. Since the occurrence of these metals was strongly linked, they are likely to have originated from the same or similar sources.

The concentration of Fe at the reference site was greater than the concentration at six

Table 1

Extractable metal concentrations in slag pile and reference soils.

Site	Cd (g kg ⁻¹)	Cu (g kg ⁻¹)	Fe (g kg ⁻¹)	Ni (g kg ⁻¹)	Pb (g kg ⁻¹)	Zn (g kg ⁻¹)
SP00†	0.020	*	4.81	0.130	1.730	2.980
SP01	*	*	4.32	0.100	0.270	0.740
SP02	*	*	4.14	*	*	0.050
SP03	*	*	4.07	0.050	*	0.060
SP04	*	*	25.06	0.020	*	0.100
SP05	*	*	378.21	0.140	0.180	0.370
SP06	*	*	14.58	*	*	0.060
SP07†	*	*	26.42	0.040	1.010	2.090
SP08†	*	*	6.64	0.020	*	0.090
SP09†	*	*	5.37	0.020	*	0.040
Reference soil	*	*	10.95	*	*	0.060

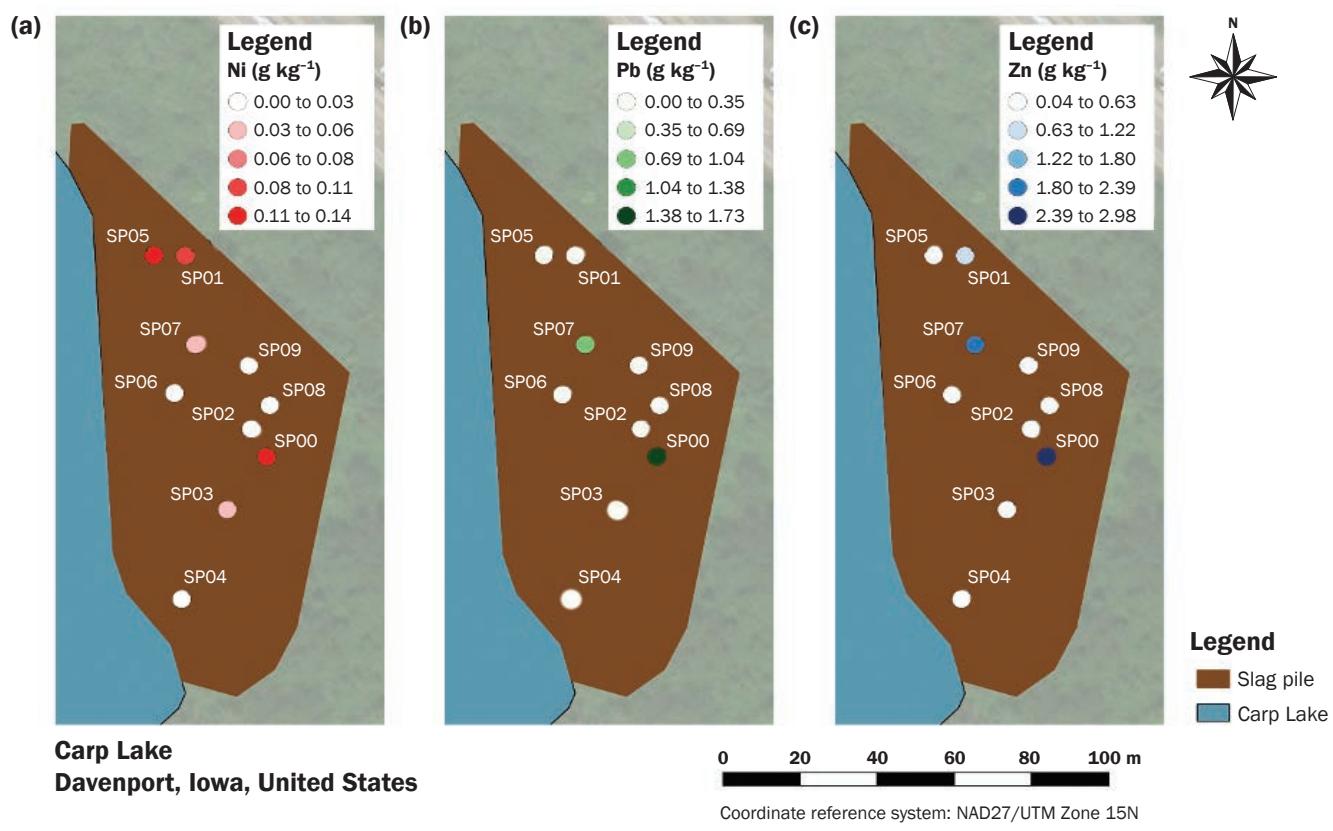
Notes: Cd = cadmium. Cu = copper. Fe = iron. Ni = nickel. Pb = lead. Zn = zinc.

*Below method detection level.

†Surface slag sample also collected.

Figure 3

Concentrations of (a) nickel (Ni), (b) lead (Pb), and (c) zinc (Zn) in spoil pile soils.



of the study sites. Likewise, the extractable Zn concentrations at the reference site were greater than the concentrations measured at two sites. However, the concentrations of extractable metals measured at the reference sites were consistent with background ranges reported for shallow soil samples in Scott County, Iowa (0.020 to 0.023 g kg^{-1} Ni and 0.053 to 0.080 g kg^{-1} Zn) (IA DNR 2010). No data on Fe concentrations in soils at these sites were provided.

Surface Slag. A digital image of a representative sample of surface slag is presented in figure 4. Surface slag samples had a rough texture. When cleaved to expose unweathered surfaces, the slag was black and glassy. Samples also contained vesicles that resulted from gas entrainment in the slag (figure 4b). This appearance was consistent with the findings reported by Cardoso et al. (2018).

The extractable concentrations of Cd, Cu, Fe, Pb, Ni, and Zn in surface slag samples are presented in table 2. The metal concentrations in the slag pile soils were generally higher than those measured in the slag. Other investigators have reported that the dissolution of metals from slag through envi-

ronmental weathering can lead to elevated metal concentrations in surrounding soils (Chopin et al. 2003; Piatak and Seal 2012; Sloto and Reif 2011; Bril et al. 2008).

Since the complete history of the site cannot be known, it is not possible to determine the specific length of time surface slag has been present in its current location. However, based on discussions with the current land-owner, neighbors, and others who are familiar with the property, slag disposal at the site stopped in the early 1980s. Consequently, it is reasonable to consider that surface slag samples have been exposed to at least four decades of environmental weathering. The effects of weathering were also notable upon the visual comparison of surface slag with samples that were freshly cleaved to expose material that had been sequestered from the atmosphere and surface water. In this case, the weathered surface slag was a dull grey color that is commonly associated with the formation of tenorite ($\text{CuO}[s]$) (Anthony et al. 1997).

It is notable that there were measurable extractable Cu concentrations in the surface slag, while the concentration of Cu in

the slag pile soil was below detection levels. This observation is best explained by examining the solubility of Cu^{2+} , Ni^{2+} , and Zn^{2+} . Consider a solution at standard environmental temperature and pressure that contains 10^{-3} M total of Cu, Ni, and Zn. If the solution is in equilibrium with solid phase Cu, Ni, and Zn oxy/hydroxide species, the percentage of free ions in solution is given in figure 5.

In this case, Cu^{2+} readily forms $\text{CuO}(s)$, which is insoluble at $\text{pH} > 6.5$. At $\text{pH} 7$, 81% of Ni is soluble and 100% of Zn is soluble. However, only 0.52% of Cu is dissolved. Consequently, at typical environmental pHs, Cu is substantially less mobile than the other metals examined in this study. Seal et al. (2010) reported a similar low percentage of extractable Cu in a slag that also contained Cd, Cu, Ni, and Zn.

Slag Pile Vegetation. Vegetation samples consisted of Amur honeysuckle (*Lonicera maackii*) at SP04, SP05, and SP06, and dogwood (*Cornus sericea*) and green ash (*Fraxinus pennsylvanica*) at SP07. The concentrations of extractable Cd, Cu, Fe, Pb, and Zn in slag pile vegetation are presented in table 3. Among

Figure 4

(a) Digital image of slag from SP07. (b) Vesicles resulting from entrained gases are visible in the detail.

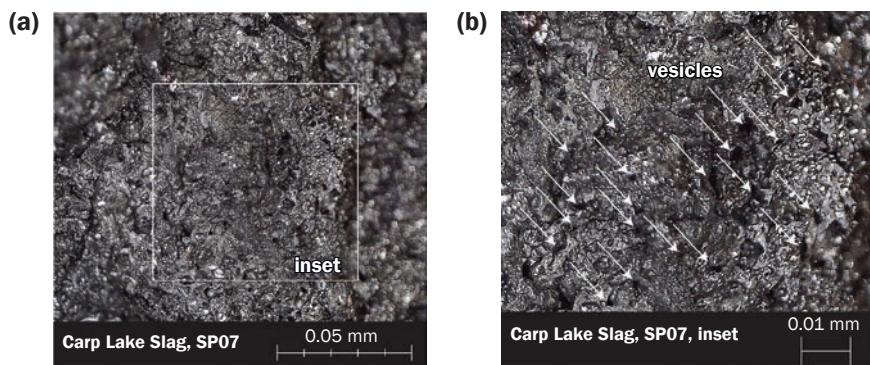


Table 2

Metal concentrations in surface slag samples.

Site	Cd (g kg ⁻¹)	Cu (g kg ⁻¹)	Fe (g kg ⁻¹)	Ni (g kg ⁻¹)	Pb (g kg ⁻¹)	Zn (g kg ⁻¹)
SP01	0.020	0.010	3.12	0.010	0.040	0.020
SP07	*	0.010	1.89	0.010	*	0.020
SP08	*	0.770	52.35	0.170	0.020	0.070
SP09	*	0.040	13.91	0.030	*	0.020

Notes: Cd = cadmium. Cu = copper. Fe = iron. Ni = nickel. Pb = lead. Zn = zinc.

*Below method detection level.

the metals examined in this study, Fe was present at the highest concentration, which was consistent with its ubiquitous distribution across the region (IA DNR 2010). Zinc and Cu were also present at concentrations that remained below or within typical values, while Pb was measured at concentrations lower than those reported by Kabata-Pendias and Pendias (2011). In contrast, the concentrations of Cd were notable in all slag pile vegetation samples except in the leaves of the green ash sample collected at SP07.

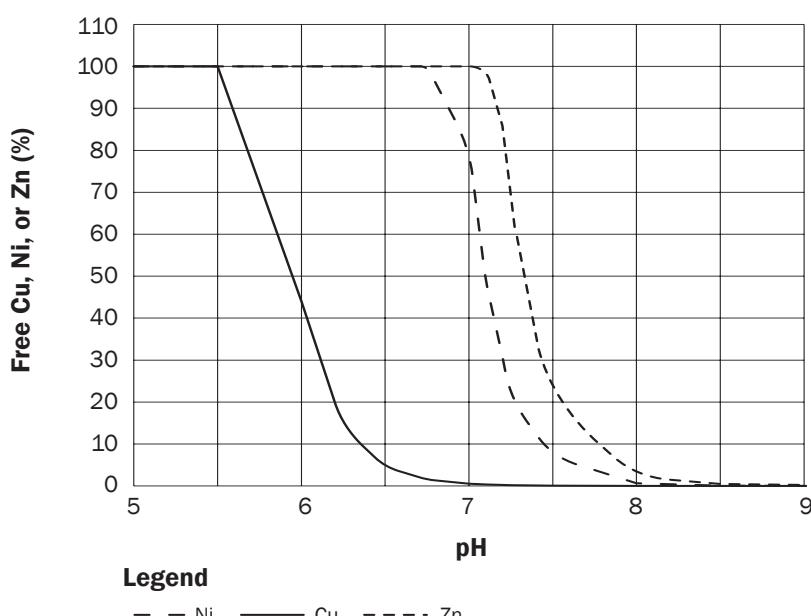
There were no clearly discernable relationships between the soil metal concentrations and their corresponding concentrations in the vegetation. This was consistent with other reports where the lack of any correlation was attributed to the differences in the physical and chemical properties of soils that directly impact the mobility of metals (e.g., the role of soil organic matter on metal sorption, cation exchange capacities, etc.). Determining these parameters is difficult in highly heterogeneous systems such as the slag pile at Carp Lake (USEPA 2007; Adriano 2003; Rosselli et al. 2003; Weng et al. 2001, 2002).

Unterbrunner (2007) suggested the use of a bioaccumulation factor (BAF) as an indicator of a species' potential to accumulate metals from the surrounding soil. The BAF is the ratio of the concentration of a metal in plant leaves (mg kg⁻¹ dry weight) to the concentration in the surrounding soil (mg kg⁻¹ dry weight) (USEPA 2007; Scragg 2005). When metal concentrations in soil and leaves are measurable (above detection limits) and the BAF > 1, it can be concluded that a particular plant species has the capability to accumulate metals in their leaves. In this study, Amur honeysuckle at SP06 was found to have a BAF of 1.61 for Zn. The only other BAF values of note were for Amur honeysuckle at SP04 with a BAF of 1.00 for Cd and a BAF for Zn of 0.88. The BAF values for other metals and tree species were all less than 0.34.

Unterbrunner et al. (2007) also reported significant accumulation of Cd and Zn by the leaves of willow, poplar, and birch trees. A high BAF for Zn was also observed by Vandecasteele et al. (2005) when comparing the phytoremediation efficiency of two willow species. This suggests that Amur honeysuckle could be an efficient Zn accumulator species that might be of relevance when considering opportunities for phytoremediation at this and similar sites. Likewise,

Figure 5

Percentage free copper (Cu), nickel (Ni), and zinc (Zn) in a solution with 10^{-3} M total of Cu, Ni, and Zn. Calculations were made at standard environmental temperature and pressure and the presence of relevant oxy/hydroxide solids was included.



regardless of any particular mechanisms at work in this system, the concentrations of Cd in the leaves of honeysuckle and dogwood are notable as they represent a potential pathway for Cd to enter the food chain.

Carp Lake Bank Soils. The average extractable metal concentrations measured in the CLB and reference soils are presented in table 4. Similar to the slag pile soils, the predominant extractable metal in the CLB soils was Fe. Other than Fe, the concentrations of all other metals were highest in samples at CLB01, the site closest to the spoil pile.

To test the relationship between metal concentrations and distance from the slag pile, a one-way analysis of variance was performed using the Real Statistics Resource Pack Software's data analysis tool. From this assessment, it could not be concluded that the data were significantly different across the

three sites ($\alpha = 0.05$, $p = 0.978$, $F = 0.022$, and $F_{crit} = 3.682$). When averaged across all CLB sites, the concentrations of Cd, Cu, Ni, Pb, and Zn in CLB soils were greater than those measured at the reference site as presented in table 4. Regardless of statistical significance, the fact that the highest Pb concentration in CLB soils was measured closest to the slag pile supports the suspicion that the slag pile is a source of metal contamination.

Summary and Conclusions

The slag pile at Carp Lake is an important example of site at which there are no meaningful records of the placement of contaminated material. This factor presents substantial challenges when attempting to assess the exact nature and extent of contamination. When considering the type and extent of contamination and Carp Lake's location

adjacent to the Mississippi River, the need to study this and similar sites is high. At Carp Lake, the distribution of metals in slag deposits was found to be highly heterogeneous. This is likely a reflection of the differences in Fe ore that was smelted over a period of approximately 80 years. The slag pile was a source of metals in the adjacent soils and vegetation. In this case, metals were most likely mobilized through weathering of the porous slag material. To adequately assess the potential for further off-site contamination, ground and surface water, lake sediment, and metal uptake in aquatic vegetation will be assessed.

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Table 3

Average extractable metal concentrations (mg kg^{-1}) in leaves of Amur honeysuckle, dogwood, and green ash collected from slag pile sites.

Site/common name of leaves	Element				
	Cd (mg kg^{-1})	Cu (mg kg^{-1})	Fe (mg kg^{-1})	Pb (mg kg^{-1})	Zn (mg kg^{-1})
Typical range*	0.05 to 0.20	5 to 30	†	5 to 10	27 to 150
SP04/honeysuckle	2.30	2.40	177.5	2.30	89.0
SP05/honeysuckle	0.40	4.40	163.2	1.60	85.4
SP06/honeysuckle	1.10	4.30	16.8	1.70	91.3
SP07/dogwood	2.60	14.70	72.05	1.50	27.6
SP07/green ash	‡	15.40	101.4	2.90	51.6

Notes: Cd = cadmium. Cu = copper. Fe = iron. Ni = nickel. Pb = lead. Zn = zinc.

*Kabata-Pendias and Pendias (2011).

†No data provided.

‡ Below method detection level.

Table 4

Average extractable metal concentrations in Carp Lake bank (CLB) and reference soils.

Metal	Average concentration (range) (g kg^{-1})			CLB site average	Reference site
	CLB01	CLB02	CLB03		
Cd	0.003	0.001	0.002	0.003	0.001
Cu	0.019	0.017	0.015	0.017	0.009
Fe	6.27	7.57	8.55	7.47	8.21
Ni	0.014	0.014	0.011	0.013	0.012
Pb	0.023	0.011	0.015	0.016	0.012
Zn	0.076	0.062	0.059	0.065	0.044

Notes: Cd = cadmium. Cu = copper. Fe = iron. Ni = nickel. Pb = lead. Zn = zinc.

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