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Environmental Impacts of Road Salt: Effects of Chloride on Metal Leaching from Lake Erie Sediments

Robert Amsdell[1a] and Hwidong Kim[*2]

[1,2]*Department of Environmental Science and Engineering, Gannon University*
109 University Avenue, Erie, Pennsylvania, USA
[2)] *kim008@gannon.edu*

Abstract. A large amount of road salt, primarily consisting of sodium and chloride, is applied to roads every winter. In addition to increasing salinity in surface water due to the influx of sodium and chloride from melting snow piles, chloride may affect metal solubility by forming metal-anion ligand complexes. To explore the impacts of chloride on metal leaching from sediments, this research tested sediment samples collected from Lake Erie near Presque Isle Bay in Pennsylvania, USA. Metal leaching was assessed using the standard Synthetic Precipitation Leaching Procedure (SPLP) and a modified SPLP solution containing 28 mg/L of chloride, which represents a peak chloride concentration recorded in Great Lake. Among 24 metals analyzed by ICP-MS, the leaching behavior followed the order: Ca > Mo > Ag > Cu under both the presence and absence of chloride in the SPLP condition. However, based on T-test and ANOVA results at the 95% confidence level, most metals showed no statistically significant difference in leachability between the two conditions.

Keywords: Road salts, chloride, heavy metals, Great Lakes

1. INTRODUCTION

Approximately 15-32 million metric tons of road salts are applied as a deicing agent on the roadway in the US every year to improve the road conditions (Kelly, et al. 2019). Sodium chloride (NaCl), commonly known as rock salt, is the most widely used deicing agent due to its low cost and effectiveness; however, its effectiveness is limited to specific temperature ranges. Thus, calcium chloride ($CaCl_2$) and magnesium chloride ($MgCl_2$) are also used as alternatives in colder regions.

Chloride is known as a strong ligand to form complexes with metal ions, increasing the water solubility of metals. Wu and Kim (2017) reported that lead concentrations increased up to 16 mg/L – approximately three orders magnitude higher than the US drinking water action level for lead – when using the synthetic precipitate leaching procedure (SPLP) on lead-contaminated soil samples in the presence of 40% road salt. The strong binding capacity of chloride facilitates the release of lead from exchangeable lead, lead minerals such as $PbCO_3$, and lead oxide, and form lead-chloride complexes ($PbCl^+$, $PbCl_2$, $PbCl^-$, and $PbCl_2^{2-}$) (Wu and Kim, 2017). These phenomena are further supported by Nelson et al (2009), who found that heavy metal transport through soil media can be enhanced by road salts, mainly due to cation exchange. They tested the soil samples collected from Spokane, WA. USA, for Cu, Pb, and Cd in the presence of NaCl and $MgCl_2$. NaCl had a greater effect on metal mobility than

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[a] M.S. Student, E-mail: amsdell007@gannon.edu
[*] Corresponding author, Associate Professor, E-mail: kim008@gannon.edu

$MgCl_2$, as NaCl can promote the dispersion of organic matter and clay, which bind with heavy metals, whereas $MgCl_2$ promotes flocculation and prevents dispersion. The mobility of Cu and Pb was highly impacted by NaCl while Cd mobilization was more greatly affected by $MgCl_2$ under acidic soil conditions. The effects of organic matter and soil properties on metal mobility in the presence of road salts were also highlighted by Nelson et al. (2009).

In addition to the effects of road salt application on water quality, Kolesar, et al. (2018) reported that increased PM2.5 sodium concentrations in northern inland US cities during winter, as well as elevated chloride levels, could be attributed by road salt application.

With the increase of road salt use over the decades, salinity in freshwater has risen. Godwin et al. (2003) investigated changes in ionic concentrations in the Mohawk River, which flows eastward through upstate New York and joins the Hudson River north of Albany, from the 1950s to 1990s. They reported that Na^+ and Cl^- concentrations increased by 130% and 243%, respectively, over the study period. Novotny, et al. (2008) reported that the current chloride concentrations in lakes of the Twin Cities Metropolitan Area (TCMA), Minnesota – measured at both the surface (1 meter below the surface) and bottom (1 meter above the lakebed) – are substantially higher than historical levels. They also found an average annual increase of 1.8% in specific conductivity in these lakes.

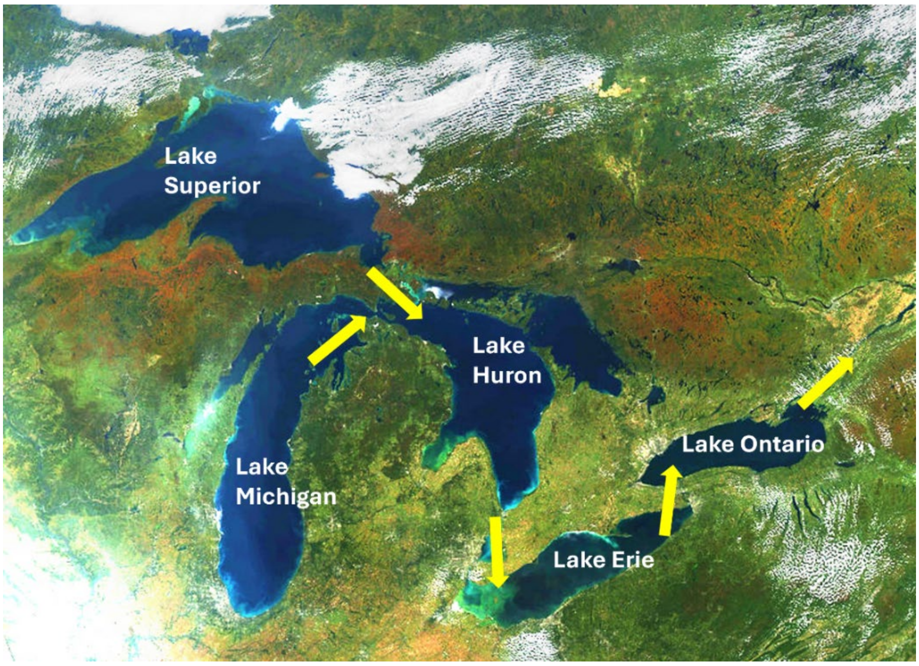

Image credit: USGS/NOAA GLERL, MODIS satellite image of the Great Lakes (modified by the authors)

Fig 1. Great Lakes and water flow directions

Similar trends in chloride level changes have been observed in the Greak Lakes. According to data reported by Chapra, et al. (2012), over a 40-year period, chloride concentrations in Lake Superior were lowest among all five Great Lakes, followed by Lake Huron, Lake Michigan, Lake Erie, and Lake Ontario, in that order. The trends in chloride levels correspond with the directions of water flow, as illustrated in Fig 1. Chloride levels in Lake Ontario peaked at 28.3 mg/L in the year 1975 and decreased over the two decades and began to rise again after 1995. Overall, chloride concentrations in east Lake Erie were lower than those in Lake Ontario but followed similar trends. Chloride levels in Lake Michigan, Lake Huron, and Lake Superior were substantially lower than those in Lake Ontario and Lake Erie but have gradually increased over the last four decades.

Despite the importance of Great Lakes as resources of agricultural production and as a major drinking water resource for more than 30 million people (USEPA 2024), Great Lakes have historically been heavily polluted by over 50,000 chemicals and pollutants (Jeanneret 1989). Although significant efforts over the past several decades have removed many pollutants and contaminants, large amounts of chemicals and heavy metals remain in Great Lake sediments. According to Marvin, et al. (2002) and Mitchell, et al. (2019), measurable concentrations of toxic heavy metals such as As, Cd, Cr, Pb, Ni, and Hg in Lake Erie and Lake Ontario, with lead concentrations reaching up to 108 mg/kg at the 75th percentile of surface sediments in Lake Ontario. With the elevated chloride concentrations, the risk of heavy metals being released from lake sediments may increase.

In this research, the effects of elevated chloride levels on the mobility of metals potentially present in Lake Erie sediments were explored. To investigate various environmental impacts, two scenarios were examined: 1) low pH due to acid rain and 2) low pH combined with elevated chloride levels. To simulate acid rain conditions, the solution used for the Synthetic Precipitation Leaching Procedure (SPLP) was prepared. The SPLP is designed to determine the mobility of organic and inorganic pollutants under acid rain environments (pH 4.2) (USEPA 1994). The chloride level of 30 mg/L, simulating the peak concentration observed in Lake Ontario in 1975, was utilized in this research.

2. Materials and Methods

2.1 Sample collection and processing

A total of six sediment samples were collected from Lake Erie within 500 meters of the coast. The sampling locations are shown in Fig 2 and listed in Table 1. After excess water is removed, the samples were air-dried at room temperature. The dried samples were then screened using a No. 30 standard sieve (mesh size = 600 micrometers) to remove extraneous organic material such as leaves and mussels.

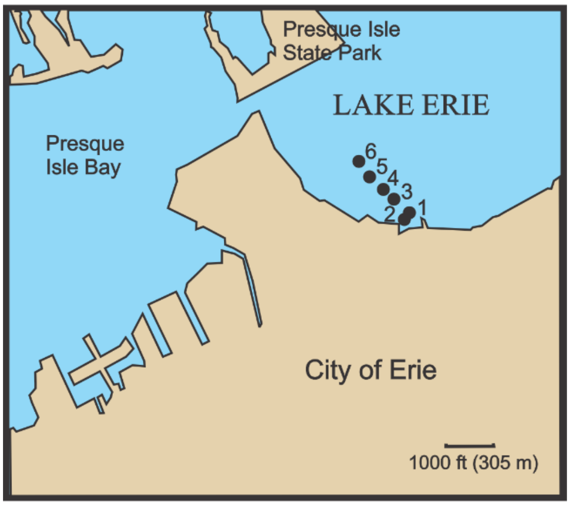

Fig 2. A map of the sampling site

Table 1. Details of the sampling locations (GPS information and water depth)

Waypoints	Latitudes	Longitudes	Depths (m)
1	42° 08.740′	080° 03.986′	2.90
2	42° 08.722′	080° 03.979′	1.89
3	42° 08.808′	080° 04.057′	3.35
4	42° 08.832′	080° 04.101′	3.38
5	42° 08.873′	080° 04.166′	3.47
6	42° 08.921′	080° 04.213′	3.57

2.2 Acid digestion and leaching test

Approximately 1-2 grams of the samples were transferred to digestion tubes and subjected to metal digestion using a hot block digester (QTechCorp, USA) following USEPA Method 3035B (USEPA 1996). To investigate the effects of acid rain, the Synthetic Precipitates Leaching Procedure (SPLP) was conducted according to USEPA SW846 Method 1312, maintaining 1:20 of solid and liquid ratio (USEPA 1994). To assess the impact of elevated chloride concentrations on metal leaching, an additional SPLP solution containing 30 mg/L of chloride was prepared by adding sodium chloride (NaCl) (Fisher, USA). All samples were prepared in triplicate. The leaching test was conducted for 18 +/- 2 hours, after which the solutions were filtered to remove solids from the SPLP solution. The collected SPLP leachates were digested according to USEPA SW846 Method 3020A (USEPA 1992). The digested samples were analyzed for heavy metals using an ICP-MS 7900 (Agilent, USA). The total metal concentrations of each sediment samples (mg/L) were converted to mg/kg as follows (Eq (1)):

$$C_{S,i} = C_{L,i} \times \frac{L}{M_{S,i}}$$

(1)

Where, $C_{S,i}$ = Metal concentration in sediment sample *i* (mg/kg)
$C_{L,i}$ = Metal concentration in digested solution *i* (mg/L)
L = Liquid volume of digested solution
$M_{s,i}$ = Mass of solid (sediment) used for metal digestion (kg)

2.3 Leachability

Metal concentrations in SPLP leachate may vary depending on the metal concentrations present in each sediment sample. To address the potential for metal release under leaching conditions, the leachability of each metal was determined. Leachability represents the proportion of metal mass released into the aqueous phase relative to the total amount of that metal contained in the soil. In other words, if all metal present in the soil is released into the solution, the leachability would be 100%. In this study, leachability was calculated based on the metal concentration in the leachate and the mass of soil samples used in the leaching test as shown in Eq. (2):

$$Leachability\ (\%) = \frac{Mass\ of\ heavy\ metals\ in\ leaching\ solution}{Mass\ of\ heavy\ metals\ in\ sediment} = \frac{C_{i,L} \times M_i \times 20}{C_{i,S} \times M_i}$$

(2)

Where, $C_{i,L}$ = Metal concentration in leaching solution (mg/L)

M_i = Mass of soil added to each leaching test (kg)
20 = Soil and liquid ratios (L/kg) (e.g., 0.1 L per 0.005 kg)
$C_{i,S}$ = Metal concentration in sediment (mg/kg)

3. Results

3.1 Total metal concentration in sediment samples

Total metal concentrations of sediment samples are present in Figure 3. The concentrations of major cations (Na, Mg, Al, K, Ca, and Fe) and other trace metals (Be, V, Cr, Mn, Ni, Cu, Zn, As, Mo, Ag, Cd, Ba, and Pb) are displayed separately due to differences in their concentration ranges. Among the major cations, iron showed the highest concentration (1.23×10^4 mg/kg $\pm$ 3800 mg/kg), followed by Ca, Al, Mg, K, and Na in that order. These trends are similar to, or slightly lower than, the typical concentrations of major cations found in Lake Erie. In addition to silicon and oxygen, Al, Fe, and Ca are among the most abundant elements found in the Earth's crust.

Trace metal concentrations shown in Figure 3 were also similar to, or slightly lower than, values previously (Marvin, et al. 2002, Yuan 2017). However, it is noteworthy that the samples exhibited relatively high concentrations of barium (Ba) (45.19 mg/kg ± 21.60), which is considered moderately polluted with Ba according to the USEPA guideline (Moffett, et al. 2007). Barium concentrations in the Great Lakes increased dramatically during the 1950s (Yuan 2017) because of high demands of barium and barium compounds in industrial and medical applications (Moffett, et al. 2007). Elevated Ba level have persisted, as barium is relatively immobile in soil once it binds to particles due to its high reactivity (Granmo, et al. 2020). Overall, total metal concentrations found in the sediment samples collected from Lake Erie were comparable to the typical concentrations reported in previous studies.

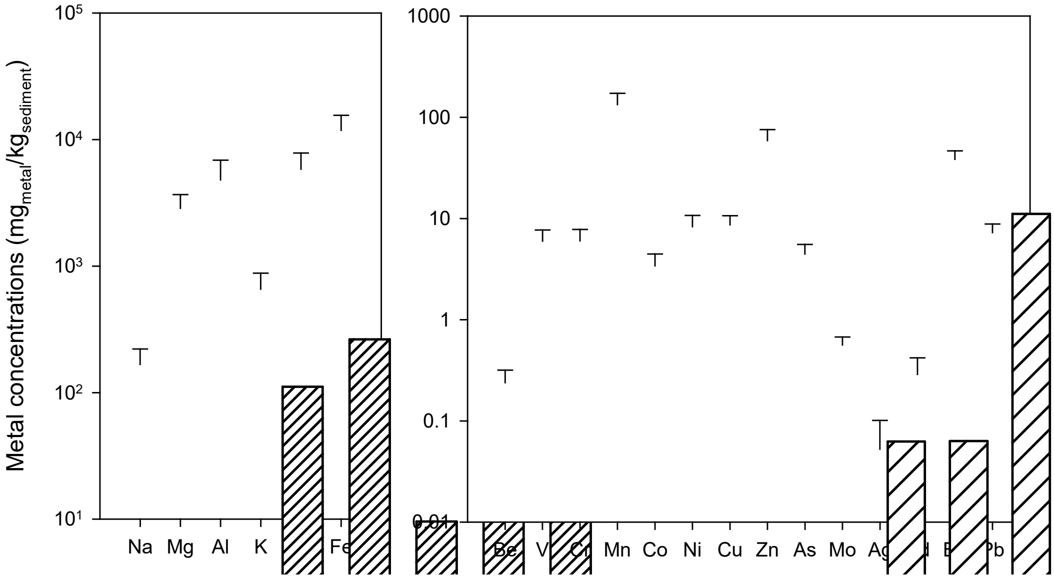

Fig 3. Total metal concentrations in sediment samples; (left) major cations and (right) trace metals.

3.2 Metal leaching behaviors

3.2.1 Metal concentrations in leaching solutions

Figure 4 presents the metal concentrations in leaching solutions (mg/L). The average concentrations of major cationic components ranged from 0.5 to 34.0 mg/L, with relatively large standard deviations observed among sediment samples. Error bars in Figure 4 represent the standard deviations of all samples, including triplicates of each sample. On average, metal concentrations in the SPLP solution containing chloride were higher than those in the SPLP solution without added chloride.

It is important to note that leaching test conditions differ from natural lake water conditions; the SPLP leaching solution is set at 4.22 ± 0.05 and is highly oxidized, whereas the pH of natural lake water is slightly acidic to near neutral (6.50 – 6.80), and the environments near the lake bottom can be less oxidized or even reduced.

A similar trend in the leaching behavior of major cations was also observed for trace metals as shown in the graph on the right side of Figure 4. However, the average concentrations of certain metals, such as Cu and Ag, were slightly higher in the SPLP solution without added chloride than in the solution with chloride. This is likely because Cu and Ag are known to form insoluble precipitates with chloride ions, such as Nantokite (CuCl(s)) and Cerargyrite (AgCl(s)), in the presence of chloride. Thus, the leaching behaviors of Cu and Ag were likely influenced by complexation with chloride.

Except for Manganese (Mn), the average concentrations of all other trace metals in both leaching solutions were below 0.1 mg/L. Among these, barium (Ba) exhibited the highest concentrations (0.035 mg/L), which corresponds to its relatively high levels in the sediment samples, as shown in the total metal concentration results.

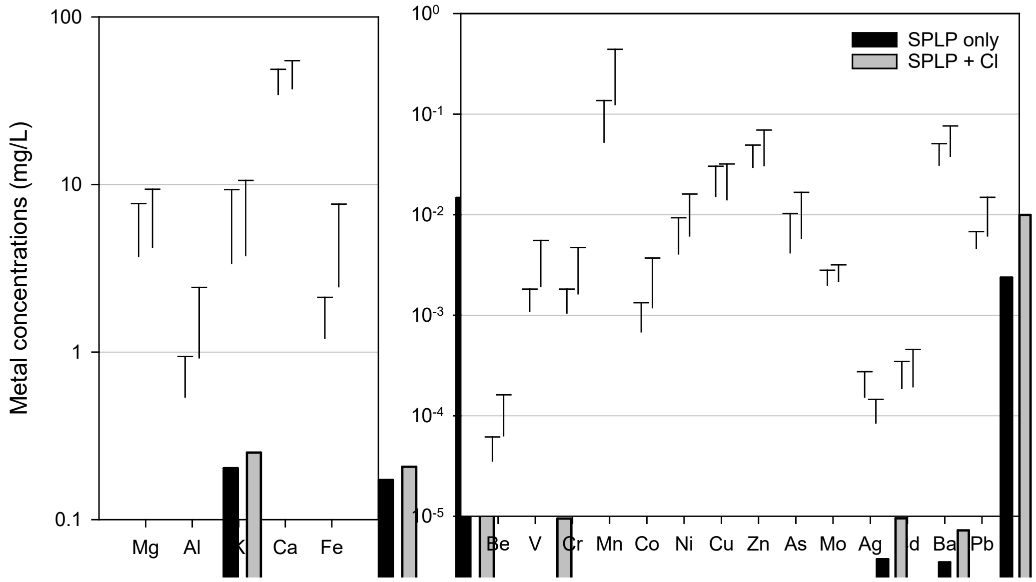

Fig 4. Metal concentrations in two types of the leaching solutions; SPLP only and SPLP with chloride.

3.2.2 Leachability of metals

The concentrations of the metal leached from the soil in given conditions are often insufficient for evaluating the metal leaching behavior due to variations of metal content of the soil. High metal content may result in greater leaching into the aqueous phase, depending on mineral types and binding capacity of the soil. Consequently, the leaching behavior may not be accurately characterized if the metal content in soil is relatively low. Therefore, as described in the Material and Method section, the leachability of each metal was estimated to more effectively examine leaching behaviors.

Figure 5 presents the leachabilities of each metal under SPLP and SPLP with chloride conditions. Calcium (Ca) exhibited the highest leachability (> 10%) among all metals, indicating that more than 10% of the Ca in the soil may be dissolved under leaching conditions. Among trace metals, molybdenum (Mo) showed the highest leachability (6.58%), followed by Ag, Cu, Ba, Pb, Cd, and As, in that order. The remaining trace metals were generally lower than 1.00%.

When comparing two leaching conditions (SPLP and SPLP with added chloride), the average leachability of most metals was higher in the presence of chloride; except for Cu and Ag. The trends are consistent with the pattens of metals dissolved in the aqueous phase, as shown in Figure 4.

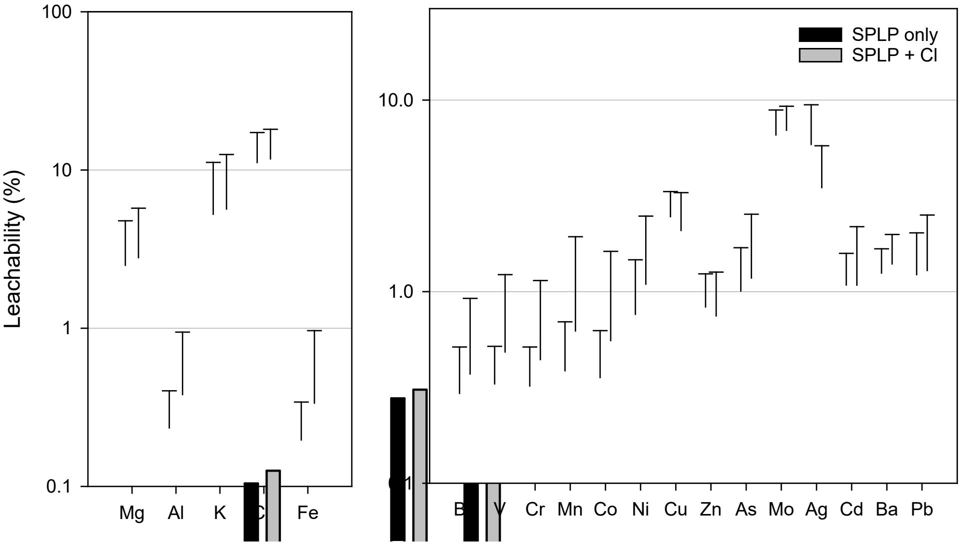

Fig 5. Leachabilities of metals observed in SPLP and SPLP with chloride conditions

To address variations in metal leachabilities among sample, ANOVA (Analysis of Variance) was performed at a 95% confidence level. The resulting p-values are presented in Table 2. Despite visual differences in metal leachabilities illustrated in Figure 5, the ANOVA results showed that all p-values exceeded 0.05, indicating that none of the differences in metal leachability between the two conditions were statistically significant. These statistical results suggest that, at the tested chloride concentration, the impact of chloride on metal leaching under SPLP conditions is not significant.

Table 2. Statistical test (ANOVA) results comparing metal leachability in two leaching conditions.

Metals	p-value	Metals	p-value
Mg	0.7856	Ni	0.4642
Al	0.3962	Cu	0.3850
K	0.8737	Zn	0.6591
Ca	0.8138	As	0.7018
Fe	0.4623	Mo	0.6904
Be	0.4099	Ag	0.0663
V	0.4924	Cd	0.9935
Cr	0.5723	Ba	0.4998
Mn	0.5467	Pb	0.8840
Co	0.5372		

3.3 Impacts of pH and chloride on water quality

Although the experimental conditions were not sufficient to statistically prove the hypothesis that chloride (road salt) enhances metal leaching, the results from this research provide meaningful insights into potential future water quality. The two conditions used in this study simulate increasing surface water acidity (influenced by acid rain and climate change) and elevated chloride levels due to the influx of road salt into freshwater systems. These environmental impacts were demonstrated by comparing reported trace metal concentrations in lake water with those obtained in this research (Table 3). As shown in Table 3, average metal concentrations in both leaching conditions were substantially higher than those typically found in lake water; for example, the average Fe concentrations in leaching tests were 880 times (SPLP only) and 1800 times (SPLP + Cl) than those in lake water. Among toxic metals, Pb concentrations reached as high as 6,160 ng/L under SPLP + Cl condition, compared to just 6.1 ng/L in lake water. Although these leaching conditions represent 'worst' scenarios, the results suggest that metals in sediments – potentially deposited by unregulated waste discharge to the Great Lakes over the past centuries – have significant potential to be released into the environment as the conditions gradually change.

Table 3. Comparison of selective trace metal concentrations (ng/L) in Lake Erie and those in leaching solutions. The values in parentheses represent standard deviations.

	Zn	Cd	Pb	Cu	Cr	Fe	Ni
Lake Erie water (pH = 8.1 ± 0.4) (Nriagu, et al. 1995)	86.2 (88.5)	3.1 (2)	6.1 (4.2)	869.9 (99.2)	137 (39.8)	1372.5 (1,241.9)	872.3 (213.1)
SPLP* (this study)	29,660 (19,590)	190 (160)	4,650 (2,130)	15,260 (15,060)	1,060 (770)	1,209,770 (913,740)	4,080 (5240)
SPLP + Cl* (this study)	30,800 (38,730)	190 (260)	6,160 (8,760)	14,090 (17,840)	1,640 (3,080)	2,460,270 (5,184,770)	6,150 (9,920)

*Initial pH = 4.22 ± 0.05

4. Conclusions

In this research, the effects of chloride on metal leaching under SPLP leaching conditions were examined to explore the potential impacts of road salts—heavily applied to roads in northern U.S. states during the snow season—on water quality in Lake Erie. To assess these impacts, sediment samples were collected near Presque Isle Bay in Lake Erie, analyzed for total metal content, and tested for leachable metals under two conditions: SPLP and SPLP with 30 mg/L of chloride. Total metal concentrations in the sediment samples were slightly lower than average values reported elsewhere but were still representative of typical Lake Erie sediments. Results for metal concentrations in leaching solutions, as well as leachability (the ratio of the mass of metal leached to the total mass of metal in the sediment), showed that substantial amounts of metals were released from sediment samples under leaching conditions. When comparing average leachability values between the two conditions, most metals exhibited higher leachability in the presence of chloride, except for Cu and Ag, which are known to form complexes with chloride, reducing their leachability. However, statistical comparison of the two conditions showed no significant differences at the 95% confidence level, likely due to the large standard deviations observed for each metal.

Although these tests were conducted under laboratory-scale leaching conditions, the findings highlight the environmental factors—elevated chloride levels and acidic pH (mainly due to acid rain and increasing atmospheric CO_2)—that may impact future water quality. While substantial progress has been made in improving water quality in the Great Lakes over recent decades, these lakes, which are critical water resources for millions of people in the region, still have the potential for adverse water quality impacts triggered by environmental changes, especially given the historical accumulation of pollutants. Since this study only examined metal release under specific leaching conditions, further research is needed to better understand the effects of chloride on water quality under various lake water scenarios.

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