

# A Review of the Combined Threats of Road Salts and Heavy Metals to Freshwater Systems

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*Road salts are altering and mobilizing heavy metals away from roadside soils, potentially into freshwater systems. Despite numerous studies investigating the mobilization of heavy metals by road salts, few studies have investigated (a) the movement rate and fate of heavy metals mobilized by road salts, (b) how road salts alter the bioavailable fraction of heavy metals, and (c) how road salts and heavy metals interact to affect freshwater organisms or human health. In this article, we discuss the consequences of increased concentrations of heavy metals and road salts, examine the mechanisms of heavy-metal mobilization, and highlight areas for future research. Future studies should investigate how metals and road salts alter ecosystem function and ecosystem services in freshwater habitats. Finally, increased research efforts will help assess whether the fate of heavy metals mobilized by road salts increases risks to human health by contaminating drinking water and water used for agriculture.*

*Keywords: deicers, environmental pollution, freshwater contaminants, metalloids*

## **G**lobal urbanization has vastly modified landscapes

and increased the density of impervious land cover, including parking lots and paved roads (Boving et al. 2008). Roads reduce habitat area, fragment habitats, change soil hydrology, reduce water quality, and introduce chemical pollutants (reviewed by Forman and Alexander 1998, Spellerberg 1998, Trombulak and Frissell 2000). Although roads make up a small proportion of the total surface area in a landscape (e.g., 1.2 kilometers per square kilometer in the United States; Forman and Alexander 1998), their ecological, toxicological, and human-health effects can be severe. The magnitude of the effect that a road can have on a surrounding ecosystem depends on the density of other roads, the amount of vehicle traffic, and the slope and aspect of the road within the context of other geographical features (Forman and Alexander 1998, Trombulak and Frissell 2000).

Roads are a major nonpoint source of pollution, especially during precipitation events, because storm-water runoff contains chemicals from road materials, chemicals applied to roads, contaminants from vehicles, and heavy metals from the deterioration of vehicle parts (Forman and Alexander 1998). Many toxic chemicals that are found in runoff from roads threaten the health of ecosystems along roadways, including heavy metals and road salts (i.e., deicers; Bäckström et al. 2004, Cañedo-Argüelles et al. 2016). Unlike road salts, heavy metals accumulate in roadside soils and can then be mobilized by road salts such as sodium

chloride (NaCl), magnesium chloride (MgCl<sub>2</sub>), and calcium chloride (CaCl<sub>2</sub>) into freshwater ecosystems. Salts mobilize heavy metals that are not readily bioavailable but can also transform metals to more bioavailable species (e.g., transformation to free ions, soluble species, and weakly adsorbed species; Violante et al. 2010). An increase in the bioavailable (especially free-ion) fraction of heavy metals could increase the concentrations of heavy metals in food webs (Parker et al. 1995, Peters et al. 2013). The uptake of heavy metals by plants and algae would increase as metals become dissociated from refractory organic soil compounds to become free ions (Parker and Pedler 1997) or weakly bound soil molecules (i.e., easily exchangeable ions; Violante et al. 2010). As the bioavailability fractionation of heavy metals increases (e.g., free ions and easily exchangeable ions; Violante et al. 2010), heavy metals could have greater toxicological effects on organisms. Models such as the free ion activity model (FIAM; Parker and Pedler 1997) and the biotic ligand model (e.g., Campbell et al. 2002) can be used to understand the potential toxicological effects of heavy metals on organisms. However, salts and heavy metals could interact toxicologically. In addition, salts and heavy metals could act beyond the organismal level by disrupting ecosystem functions such as microbial processes and food-web dynamics, thus reducing ecosystem services such as carbon sequestration in wetlands, ponds, and lakes. One of the greatest but poorly known threats that humans face from heavy metals and road

salts is the continuous movement and accumulation of heavy metals and salt in groundwater systems used for human consumption and agriculture (Norrström and Jacks 1998).

We provide a review of the literature regarding the effects of heavy metals, road salts, and their interactions along roads and into aquatic ecosystems. In doing so, we hope to foster discussion and future studies that will lead to predictive models that can identify freshwater systems that are threatened by road salts and heavy metals and to encourage future studies to investigate the threats that these contaminants pose to human health and ecosystem function.

### Heavy metals

*Heavy metals* are metals and metalloids that are either essential or nonessential (Bolan and Duraisamy 2003). Both are toxic to organisms at high concentrations, and their accumulation threatens the health of organisms living along roads (Li et al. 2007, Green et al. 2008) but could also cause harm in freshwater habitats and groundwater. Heavy metals often accumulate in roadside soils, primarily in organic soils (Bolan and Duraisamy 2003), and can contaminate ecosystems and groundwater used for human consumption if heavy metals become disassociated from soil materials (e.g., Norrström and Jacks 1998). The heavy metals that most commonly accumulate in habitats adjacent to roads include aluminum, boron, cadmium, copper, iron, lead, manganese, mercury, nickel, titanium, and zinc (Trombulak and Frissell 2000).

**Heavy-metal accumulation in soils.** Automobile traffic is responsible for the majority of these metals accumulating along roads, mostly because of vehicular degradation (e.g., rust), brake and tire wear, leaking fluids, and the legacy effects of lead used in gasoline (Amrhein et al. 1992, Bauske and Goetz 1993, Davis et al. 2001). However, in urban areas, sources such as the siding of buildings can contribute more metals than vehicles do, including lead, cadmium, and zinc (Davis et al. 2001). Studies reporting heavy-metal concentrations in soils along roads suggest that heavy-metal contamination varies by country, depending on the number of vehicles using the road, the time since road construction, and the types of soil along the road (e.g., Fakayode and Olu-Owolabi 2003, Saedi et al. 2009, Wei and Yang 2010). Although not all contaminated areas present toxic risks to humans and wildlife, heavy-metal contamination of roadside soils is a global problem. Despite metals flowing in runoff or blowing away from roadways, metal concentrations are typically highest within 10 meters of the paved surface and within the top 15 centimeters of soil along the road (Werkenthin et al. 2014). The accumulation or transport of heavy metals away from roads depends on the metal, road surface permeability, and climatic factors such as season and precipitation rates (Göbel et al. 2007, Li et al. 2007). The high concentration of metals surrounding paved roads indicates that the ecological and health threats should be most pronounced around densely traveled roads and in

densely populated areas, because larger and more heavily used roads typically have more contaminants (Fakayode and Olu-Owolabi 2003, Göbel et al. 2007).

**Heavy-metal fractionation and availability in soils.** Dozens of studies have quantified the total concentrations of heavy metals in soils along roads (reviewed by Werkenthin et al. 2014, Huber et al. 2016). Metals can be associated with soils in a variety of ways, including bioavailable species (e.g., free ions, easily exchangeable ions, or adsorbed ions) and poorly available species (e.g., refractory organic complexes or crystalline structures; Violante et al. 2010). However, the total metal content in soil is a poor indicator of the current and potential bioavailability, because metals can exist as various chemical types (i.e., species) in soil and have different potentials for transformation to bioavailable phases (Krishnamurti et al. 1995, Krishnamurti and Naidu 2002). For example, metals present in mineral structures in soil are typically not bioavailable but can be easily transformed by changes to soil chemistry (e.g., Campbell and Stokes 1985). Transformation to a bioavailable state includes changing metals from a mineral state to a soluble, adsorbed, or complexed state. The potential bioavailability depends on the type of metal in the soil, the concentration of free ions in the soil, the concentration of colloiddally bound heavy-metal complexes, and the type of soil (Carvalho et al. 1999, Brown and Markich 2000, Krishnamurti and Naidu 2002, Violante et al. 2010). In addition, the release or transformation of these heavy metals depends on numerous factors, such as the soil type, percentage of soil made up by colloids, pH, water saturation, amount of dissolved organic carbon, and the species of each metal present in the soil (Green et al. 2008). Once these metals become more bioavailable and disassociated from organic soil materials, they can be further transformed into dissolved ions, which are then more easily taken up by organisms (e.g., FIAM; Parker and Pedler 1997, Brown and Markich 2000, Campbell et al. 2002). Heavy metals that enter freshwater ecosystems can present numerous toxicological and ecological effects on organisms and ecosystem function.

**Heavy metals in freshwater systems.** Streams near roads are often contaminated by heavy metals, and the organisms in those streams can be subjected to the toxicological effects of heavy metals. Heavy metals from roadside materials (e.g., mercury, lead, cadmium, and copper) can reduce the diversity of macroinvertebrates, alter food-web structures, and reduce ecosystem services in streams (Maltby et al. 1995, Clements et al. 2000, Hirst et al. 2002, Carlisle and Clements 2005). Clements and colleagues (2000) showed that increased concentrations of a mixture of heavy metals (mostly lead, zinc, copper, and aluminum) reduced the number of organismal species in freshwater streams, and the concentrations of metals directly predicted the abundance of various organismal functional groups (e.g., grazers, scrapers, and predators). Higher metal concentrations led to a reduced number

of individuals for each group. In addition, the richness of mayflies, caddisflies, stoneflies, and chironomids were reduced when metal concentrations were high. In flowing waters, heavy-metal contamination (primarily copper and zinc) reduces litter breakdown rates and reduces microbial respiration (Carlisle and Clements 2005, Ferreira et al. 2016). Finally, heavy metals in streams can negatively affect microbial communities in streams. However, some evidence suggests that algae and diatoms are resilient to heavy-metal contamination (Hirst et al. 2002). Further investigations into the consequence of continued heavy-metal contamination on microbial processes in streams could elucidate the potential for long-term cascading effects that reduce ecosystem function and ecosystem services provided by streams.

Heavy metals accumulate in freshwater lakes and can have toxicological effects on organisms (e.g., Balistrieri et al. 1992, Wenchuan et al. 2001). However, few studies have investigated the effects of heavy metals on entire freshwater food webs (see Altındağ and Yiğit 2005). Field studies of freshwater lakes have mainly focused on the accumulation of heavy metals in single animal species, especially fish (e.g., Malik et al. 2010, Monroy et al. 2014). An important aspect missing from the research of heavy-metal effects in freshwater lakes is the effect they have on ecosystem services of benthic communities. In terrestrial soils, heavy metals can be toxic to microorganisms, thereby reducing ecosystem functions such as nutrient cycling and decomposition of organic materials (Kandeler et al. 1996, Giller et al. 1998, Peters et al. 2013). However, no investigations of heavy-metal toxicity on microbial activity in lake or pond benthic communities have occurred. Investigating the effects of heavy metals on aerobic and anaerobic processes in lakes and ponds would be beneficial to better understand how ecosystem services and ecosystem functions are disrupted in lake systems that vary in depth, soil type, and trophic state.

Wetlands can become toxic, heavy-metal sinks from roadside contamination. However, wetlands can also be used as bioremediation sites (through sludge removal and microbial activity) because of high amounts of organic materials in soils and the ability of microbes and some plant species to tolerate and accumulate high metal concentrations in their tissues (Spellerberg 1998). In addition, the abundant organic material in wetlands has a high binding affinity for heavy metals. However, anaerobic respiration in wetland soils could increase the rate of transformation of bound heavy-metal complexes into more easily mobilized species (e.g., iron and manganese; Kim and Koretsky 2013). One exception is mercury, which can be locked in species that are not bioavailable in anaerobic conditions (Mehrotra and Sedlak 2005). In addition, the abundance of metals such as nickel regulate processes affecting methane and other greenhouse gas production in wetland communities (e.g., Basiliko and Yavitt 2001), because ionic nickel is required for enzyme production in all methanogens (Rothman et al. 2014). Increased mobilization and increased bioavailability of heavy metals into wetlands could provide microbes with an

additional source of ionic nickel. This, along with increased temperatures, would positively affect the overall metabolic rate of these organisms, eventually leading to increased methane production. However, no studies have investigated this potential interaction. Further investigations into the complex effects that heavy metals have in wetlands will help researchers understand whether wetlands act as heavy-metal sinks or whether an increase in heavy metals will change ecosystems services in wetlands, potentially leading to an increased production of greenhouse gases.

Heavy metals enter groundwater through numerous anthropogenic activities, including mining, agriculture, landfills, and urban developments. The transport of roadside heavy metals to groundwater has been demonstrated through observational and experimental studies (Norrström and Jacks 1998, Walraven et al. 2014), although the proportion of heavy metals that enter groundwater because of roadside pollution relative to other forms of pollution is unknown. The increasing prevalence and concentration of heavy metals in groundwater should be alarming, because these metals threaten human health (e.g., lead) and their presence could go undetected for many years, especially in rural communities and in agricultural systems that are infrequently tested.

### Road deicers

Globally, road salts including sodium chloride (NaCl), magnesium chloride (MgCl<sub>2</sub>), and calcium chloride (CaCl<sub>2</sub>) are used as deicers on roadways to melt ice and snow and reduce vehicle accidents (Fay and Shi 2012). Sodium chloride is the most common road salt applied globally (more than 90%), but alternative salts are commonly used in many locations (Fay and Shi 2012). Nonchloride-based alternatives such as calcium magnesium acetate (CMA) and potassium acetate (KAc) are also used but in very small quantities (Fay and Shi 2012).

The quantity of most deicing salts used on roadways has dramatically increased around the world. For example, in 1950, less than 1 million tonnes of road salt (NaCl) was applied to roads in the United States. Road-salt applications increased to approximately 10 million tonnes by 1990 and nearly 20 million tonnes by 2014 (Bolen 2014). In addition, some European countries, such as Sweden and Denmark, have reported greater than 33% increases in NaCl application rates since the 1970s (e.g., Löfgren 2001).

This rapid surge in road-salt application has increased awareness about the potential ecological effects of salts (Corsi et al. 2010, Cañedo-Argüelles et al. 2016). These ecological effects include increased toxicity for organisms in roadside soil and nearby freshwater communities (Corsi et al. 2010). Probably the most widely observed effect near roads is the toxicity to roadside plants, whose leaves and needles turn brown because of a buildup up chloride, eventually leading to plant death (Devitt et al. 2014). In addition to toxic effects on plants, road salts degrade soil structure (e.g., weakening the binding of organic material

to mineral structures), alter soil chemistry (e.g., increasing pH), and increase ammonification through the displacement of  $H^+$  ions (Green et al. 2008). Increasing salt concentrations can also reduce microbial abundance and function and reduce the survival and species richness of soil invertebrates (Černohlávková et al. 2008, Ke et al. 2013).

Numerous laboratory studies have shown that road salts are highly toxic to individual species of aquatic organisms (Blasius and Merritt 2002, Linton et al. 2008, Gillis 2011, Tyree et al. 2016, Hintz and Relyea 2017). However, researchers have recently shifted from lab-based studies to experimental and observational studies of food webs and the ecosystem-level effects of salt in terrestrial and freshwater communities (e.g., Hintz et al. 2017, Schuler et al. 2017). The shift in focus from individual species to communities has indicated that the acute and chronic effects of salt in freshwater systems are more complex than laboratory-based toxicity tests indicate and can include shifts in the composition of aquatic organisms, trophic cascades, and effects on ecosystem services such as denitrification (e.g., Lancaster et al. 2016, Hintz et al. 2017, Schuler et al. 2017). In addition, these community-level effects can be observed at concentrations below the chronic (230 milligrams, mg,  $Cl^-$  per liter, L) and acute (860 mg  $Cl^-$  per L) thresholds established by the US Environmental Protection Agency (US EPA 1988).

Human health risks also exist because of the increase in road-salt application. Groundwater contamination from road salt has increased, with some municipalities reporting concentrations above the EPA secondary-drinking-water threshold of 250 mg  $Cl^-$  per L (Corsi et al. 2010). Chloride concentrations as high as 7730 mg per L have been recorded in contaminated freshwater systems in the United States (Corsi et al. 2010). Researchers from other countries have also reported toxic chloride concentrations. For example, researchers in Denmark have reported concentrations of chloride in soil water as high as 1400 mg  $Cl^-$  per L during winter (Pedersen et al. 1999), and researchers from Sweden have reported dramatic increases in soil and water chloride concentrations because of the increased application of road salts (Thunqvist 2004). Increased salinity in drinking water most often causes foul-tasting water, but is not generally toxic to humans, except for those with medical issues such as high blood pressure, heart failure, or some kidney diseases. The majority of threats to water used for human consumption does not come directly from the salt but instead comes from the chemicals associated with road salts and the effects that the increased salinization of drinking water has on lead pipes (Kaushal 2016, Stets et al. 2017).

### The mobilization and alteration of heavy metals by road salts

Heavy metals readily bind to organic roadside soils; however, road salts can release heavy metals that are associated with organic soils through several interrelated mechanisms (Amrhein and Strong 1990, Amrhein et al. 1992, Bauske and Goetz 1993, Löfgren 2001, Bäckström et al. 2004). Road salts

can increase the distribution and bioavailability of heavy metals by disrupting soil structure, changing soil chemistry, and altering ion exchange (Norrström and Jacks 1998, Norrström and Bergstedt 2001). Once mobilized, heavy metals and salts move from soil along roadways, potentially accumulating in freshwater ecosystems.

High concentrations of sodium can displace calcium and magnesium in the soil, thereby disrupting the structure of the soil, especially following large flows of low-electrolyte water (e.g., few dissolved ions) into sodium-rich soils (Amrhein et al. 1992, Norrström and Bergstedt 2001, Bäckström et al. 2004, Norrström 2005). This occurs when large quantities of salt are applied to roads to remove ice and snow and then a heavy rain event occurs, leading to low-electrolyte runoff. This disruption of soil structure leads to the increased porosity of the soil and increases the mobilization of metals and their associated compounds (Norrström and Jacks 1998). For example, Tang and Weisbrod (2009) showed that lead is more mobile when associated with colloidal structures in soil, which is likely because of the breakdown of colloidal structures and the disassociation of organic compounds caused by road salt (Norrström and Bergstedt 2001). This allows chloride complexes to form and mobilize heavy metals (e.g., lead) away from organic rich topsoils and into groundwater (Howard and Sova 1993, Bäckström et al. 2004). Heavy-metal mobilization by the complexation of chlorides depends on whether soils are organic or mineral based. In organic soils, aqueous ligand complexes will reduce the adsorption of some metals, such as cadmium, lead, zinc, and mercury, which increases the potential for mobilization by chloride complexes (Hahne and Kroontje 1973, Lumsdon et al. 1995). As metals are desorbed and transformed into a soluble phase, chloride complexes can increase the solubility of the metals, increasing their persistence in soil and freshwater ecosystems (Warren and Zimmerman 1994). The breakdown of surface-soil structure due to road salts is important for understanding the potential rate of mobility of heavy metals, because soils below the upper horizons typically have little organic material and are primarily mineral based. Once metals become disassociated from organic surface soils, they can move quickly into freshwater systems and can persist, associated with chloride complexes (Warren and Zimmerman 1994).

Finally, salts can directly displace metals without affecting soil structure or pH. The binding affinity of many organic compounds is higher for sodium, calcium, and magnesium than it is for most heavy metals (reviewed by Bäckström et al. 2004). High concentrations of cations from salts displace soil-bound metals such as cadmium, copper, and zinc (Granato et al. 1995, Bäckström et al. 2004). Soils with high concentrations of NaCl can result in high ionic strength, reducing the binding affinities of heavy metals to soils and increasing their potential mobility (Naidu et al. 1994, Norrström 2005). Although NaCl is the most commonly used road salt, other road salts (e.g.,  $MgCl_2$  and  $CaCl_2$ ) have a greater effect on heavy-metal mobilization through

ion displacement than NaCl (Acosta et al. 2011). Therefore, calcium and magnesium are more likely to mobilize metals because of ion exchange compared with sodium because of their higher exchangeability (i.e., higher binding affinity) in organic soils.

The majority of mobilization of heavy metals by salts is likely due to ion exchange rather than changes to soil structure or pH (Bäckström et al. 2004). However, the importance of ion exchange depends on the type of metal, the type of salt, soil structure, and soil composition. Cadmium, cobalt, and copper are more easily mobilized by ion exchange than lead and zinc are (Bäckström et al. 2004, Heier et al. 2013), but chromium is likely less mobilizable through ion exchange compared with other metals (Pagotto et al. 2001), possibly because of strong associations with iron and magnesium oxides in soil (Guo et al. 1997). Dozens of experimental and field investigations show that road salts increase the mobility of heavy metals through interrelated mechanisms. However, very little information exists on the mobility rates of heavy metals, the long-term trends of heavy metals in ground- and surface-water concentrations, the proportion of heavy metals that are bioavailable, and the rates of transformation from unavailable to bioavailable metals in roadside soils. Walraven and colleagues (2014) showed that after lead reached depths of 15 centimeters, it moved quickly (72–324 mg per square meter per year) into groundwater because of low soil pH and low amounts of organic material below those depths. Despite relatively low concentrations of lead in the groundwater, Walraven and colleagues (2014) demonstrated that 100% of the lead in the groundwater had anthropogenic origins. To understand the risks associated with heavy-metal contamination and road-salt use, more comprehensive studies are needed that trace the origins (e.g., anthropogenic or natural) of heavy metals in roadside soils and in freshwater ecosystems. Furthermore, these studies should investigate how road salts change the bioavailable fraction of heavy metals in roadside soils and whether road salts increase the rate that heavy metals move into freshwater ecosystems (relative to salt-free areas).

Nonchloride salt alternatives (e.g., CMA) also increase the mobilization of heavy metals in soil (Granato et al. 1995), but the mechanism of increased transport varies. CMA mobilization of heavy metals is more likely to occur through ligand complexation and with competitive exchange of ions (Amrhein et al. 1992). However, some evidence suggests that the overall effect of CMA on the mobilization of heavy metals is likely less effective than that of NaCl, with specific exceptions being cadmium and lead (Amrhein et al. 1992). Further research is needed to understand the underlying mechanisms explaining the mobilization differences among these salts.

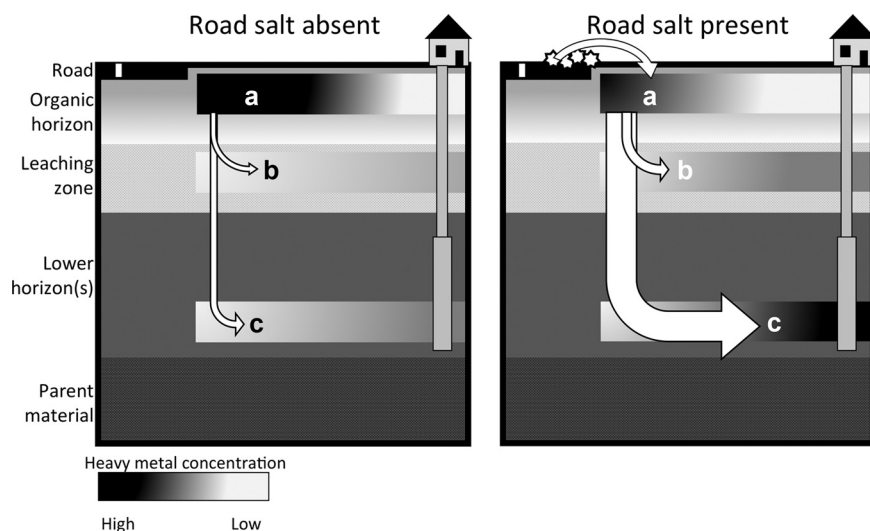
Collectively, these studies suggest that road salts increase the rate of movement of heavy metals (relative to areas without road salt), alter the distribution of heavy metals in soils (figure 1), and affect the fractionation of bioavailable heavy metals in soil and in freshwater ecosystems.

Understanding the relative fractions of various metals in soils is important for calculating the mobility of heavy metals out of soil and into freshwater systems (e.g., streams, lakes, wetlands, and groundwater). Chemical reactions and chemical complexation of heavy metals with organic and inorganic soil components greatly affect the mobility, bioavailability, and therefore toxicity of heavy metals (reviewed in Violante et al. 2010). Quantifying the fractionation of heavy metals in soil can increase our understanding of the risks associated with the bioaccumulation of metals, which could threaten species diversity, ecosystem function, and ecosystem services in freshwater systems. Some researchers have investigated the fractionation of heavy metals in roadside soils and shown that road salts can increase the exchangeable fraction of metals (Howard and Sova 1993, Norrström and Jacks 1998). Cadmium typically has the highest exchangeable fraction, because it is commonly found in aqueous solutions and not adsorbed to soils (Norrström and Jacks 1998). Lead typically has a low exchangeable fraction because of strong associations with iron, magnesium oxides, and organic matter in soil. However, the application of road salts can alter soil chemistry (e.g., pH, ligands, and dissolved organic carbon), which could increase the exchangeable fraction of heavy metals such as lead (Clozel et al. 2006).

More information is needed to understand whether the accumulation of metals in food webs might be affected by the application of salts. Currently, there has not been a rigorous investigation of the precise mechanisms by which road salts affect soil chemistry and heavy-metal fractionation in soils (see Green et al. 2008). If salts increase the bioavailability of heavy metals and salts more easily mobilize the metals, then this could increase the toxic effects of heavy metals as they enter freshwater systems. For example, in soils of freshwater systems, cadmium and lead often have high percentages of easily transformed and bioavailable species compared with those of copper and chromium (Clozel et al. 2006). This is important, because lead is often shown to have a high organic-matter-bound fraction in soils, which means that the application of road salts could be mobilizing lead and increasing its bioavailability, thereby increasing the risk of bioaccumulation in freshwater systems.

### **The potential effects of heavy metals and salts on aquatic communities**

Freshwater ecosystems provide numerous ecosystem services, including clean drinking water, food resources, recreational benefits, and economic benefits (Dudgeon et al. 2006). Most research efforts have focused on the effects of single contaminants on individual species or a small group of species. However, studying the toxicological and ecological consequences of multiple contaminants at the community and ecosystem levels will improve our understanding of the threats that freshwater systems face, especially because many of those threats could have consequences for human health.



**Figure 1.** A depiction of the effects of road salt on heavy metal transport in soil along roads near homes or agriculture. (a) Heavy metals accumulate along a gradient (black and gray gradient bar), decreasing with distance from roads. The addition of road salt leads to fewer heavy metals being present in the organic layer at the surface because of rapid downward movement through the soil, indicated by the heavier arrows. (b) Heavy metals become disassociated from organic materials in the upper horizons, moving into leachate zones where they might enter freshwater systems (e.g., streams, lakes, and wetlands). (c) The presence of road salt might also increase the rate of movement across the leachate zones into groundwater. Collectively, these processes move heavy metals down into groundwater and slowly move horizontally, where they can enter wells and aquifers used for human consumption and agriculture.

Road salts and heavy metals are toxic to freshwater organisms, but there is very little information on their combined toxicological effects. One study on salmon (*Salmo salar*) egg development showed that the toxic effects of heavy metals and road salts together were more severe than the effects of each alone (Mahrosh et al. 2014). However, a related study on the alevin stage of *S. salar* showed that the toxic effects of salt were strongest, regardless of heavy-metal concentrations (Mahrosh et al. 2017). These contrasting results highlight the need for continued rigorous investigations of how contaminants interact to affect organisms, especially invertebrates that might be more susceptible to multiple contaminants or stressors compared with vertebrates.

Heavy metals are more toxic to macroinvertebrates than to fish; they reduce macroinvertebrate diversity, alter ecosystem services, and affect food-web structure of macroinvertebrate communities (Maltby et al. 1995, Clements et al. 2000, Hirst et al. 2002, Carlisle and Clements 2005). In addition, road salts are far more toxic to invertebrates and microbial communities than to fishes (Blasius and Merritt 2002, Linton et al. 2008, Gillis 2011, Tyree et al. 2016, Hintz and Relyea 2017). Comprehensive field and lab studies investigating the potential interactive effects of heavy metals and salts on freshwater communities will provide insights into the potential effects that multiple toxins might have on organismal diversity, ecosystem function, and ecosystem services. For

example, a greater focus on the potential effects of heavy metals and salts on mollusks, especially bivalves, will potentially show that the two contaminants are contributing to the rapid decline in bivalve diversity around the world, because they are very sensitive to salts and are known to accumulate heavy metals.

To our knowledge, no studies have investigated whether road salts increase the rate of accumulation of heavy metals in freshwater food webs. Road salts can increase the bioavailability and ionic presence of heavy metals in soils, which would then likely lead to the metals accumulating faster in plants and consumers, especially filter-feeding organisms such as bivalves. Even if the organisms can uptake high concentrations of heavy metals into tissue storage without dying (e.g., *Betula spp.* and *Salix spp.*; Gallagher et al. 2008), the presence of heavy metals might make them more susceptible to the toxic effects of road salt. As an example, multiple stressors (e.g., predation, pesticides, and fertilizers) negatively affect amphibian populations (Boone et al. 2007). Although organisms might be able to tolerate a single stressor, changes in predation pressure, competition, or climate warming

might alter the way that these stressors affect populations and communities (e.g., O’Gorman et al. 2012). Furthermore, multiple contaminants might only affect a few organisms, but the absence of these organisms in the food web can have a cascading effect, leading to ecosystem consequences (Relyea 2005). It is also paramount that researchers maintain a focus on field investigations, because the toxicity and effects of these contaminants found from lab studies do not directly transfer to natural systems (Giller et al. 1998).

We also suggest that future research efforts focus on ecosystem functions and ecosystem services. Toxicological and ecological studies at the organismal level might underestimate the long-term consequences of multiple contaminants, such as heavy metals and road salts accumulating in aquatic systems. As an example, many shallow wetlands and ponds might be metal limited, which is important because the abundance of metals such as nickel regulate processes affecting methane and other greenhouse-gas production (e.g., Basiliko and Yavitt 2001). The limited quantity of ionic nickel in wetlands is important, because the methane cycle requires ionic nickel (Krüger et al. 2006). Increased mobilization and increased bioavailability of heavy metals such as nickel that are important for biogeochemical cycles in wetlands and shallow aquatic communities could increase microbial activity, magnifying the production of greenhouse gases and exacerbating the effects of climate change.

Finally, we suggest that future studies investigate the mobilization trends in heavy metals (e.g., lead) that could eventually become concentrated in groundwater used for human consumption and agriculture. Norrström and Jacks (1998) suggested that the increased mobilization of lead due to road-salt pollution increased the concentrations of lead in wells near roads. Walraven and colleagues (2014) demonstrated that lead quickly moves into groundwater after being released from soil by road salt. Therefore, further research on the transport of metals such as lead could help in the development of predictive models for understanding areas of high risk, such as areas that have shallow groundwater and large roadways in states that apply large quantities of road salts per lane kilometer. These models would be especially useful in rural areas that do not rely on city water systems and farmlands, which could be more easily monitored. Modeling the potential mobilization of lead into drinking water, given the amount of traffic (i.e., concentrations of metals), the amount of road-salt use, and soil structure, could inform landowners and farmers about the potential risks associated with using groundwater. It is also important for urban communities to monitor increasing concentrations of salts in water used for human consumption because of the potential for salts to mobilize heavy metals into drinking water and to release toxic levels of lead from pipes (Kaushal 2016).

### Conclusions

Vehicular use along roadways has increased the concentrations of heavy metals in soils. In addition, salts used for deicing roads mobilize heavy metals, transporting them away from roadways, deeper into soil, and eventually into freshwater ecosystems. The rapid increase in road-salt use over the last two decades could mean that more heavy metals are being transported away from roads and into freshwater systems. Heavy metals and road salts accumulate in freshwater ecosystems, and both have been shown to be individually toxic to freshwater organisms and to affect ecosystem function and ecosystem services. However, there has been a great lack of research on the potential interactive effects of road salts and heavy metals. With this article, we hope to inspire more research on the interactive effects of road salts and heavy metals. These interactive effects include the direct effects that salts have on the bioavailability, mobility, and transport rate of heavy metals; the toxicological effects that metals and salts have on organisms and communities; and the ecosystem effects that these contaminants might have. We also urge increased focus on the potential contamination of groundwater sources used for human consumption and farming. Increased field research and modeling efforts will be vital for estimating the potential effects that these contaminants will have with increased vehicular use and road-salt application.

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