Salt has a long history as the main tool in deicing roads in the U.S., beginning in the 1940’s, with use increasing exponentially since then. Approximately 22 million tons of salt is applied to roads in the US (EPA, 2016), and 91,000 tons were used in Maryland in the 2017-18 winter (MDOT, 2018). The development of roadways and success in increasing winter road safety by salting has driven its wide spread use. However, concerns of elevated chlorides in the 1970’s (Huling and Hollocher, 1972) has prompted numerous investigations into the implications of deicing. Initially centered on elevated chloride concentrations of surface waters, investigations have expanded into impacts on animal and plant species, hydrogeology, interactions with soils, and water quality.

The environmental impacts of increased road salt use are well documented and wide-ranging. Many researchers have reported on elevated chloride and sodium concentrations in surface waters and groundwater (Cassanelli and Robbins 2013, Howard and Beck 1999; Kauschal et al. 2005; Kelly et al. 2008; Ledford, et al. 2016; Moore et al. 2017; Mullaney et al. 2009; Snodgrass et al. 2017; Strank et al. 2013). In EPA’s region 3 states of Delaware, District of Columbia, Maryland, Pennsylvania, Virginia and West Virginia, over 2,747 miles of rivers or streams have been identified as impaired by chlorides. Declining aquatic species populations and diversity has been noted by several studies (Corsi et al. 2010; Fay and Shi 2012; Karraker et al. 2008; Jones et al. 2017; Stranko et al. 2013). A variety of changes in soil properties and chemical reactions have been observed in research studies including: transformation of soil structure and reduced microbial decomposition (Amrhein et al. 1992; and Kaushal et al. 2005); decreased soil permeability (Ramakrishna and Viraraghavan, 2005); mobilization of soil base cations (Price and Syzranski. 2014; Robinson et al. 2017, Kaushal et al. 2017, Kaushal et al. 2018); increased leaching of dissolved organic C (Haq, et al. 2018); increased leaching of nitrate (Green et al. 2008), total dissolved nitrogen (Haq et al. 2018) and total kjeldahl N (Duan and Kaushal 2015); and breakdown of organic matter and release of dissolved organic carbon (Duan and Kaushal 2015; Green et al. 2009). Further, deicing salts can also affect drinking water quality, e.g. increased corrosivity on plumbing (Edwards and Triantafyllidou 2007; Kaushal 2016; St. Clair et al. 2016; Stets al et, 2018); increased mobilization of heavy metals (Amrhein, et al. 1994; Backstrom, et al 2004; Novotny et al. 1998; radionuclides (Alam and Cheng 2014; McNaboe, et al. 2017; Riedel and Kubeck, 2018). These impacts present major ramifications on drinking water quality and protection of public health. The extent of influence of salts are dependent on many factors including degree and longevity of salt application, local geology, aquifer type, soil type and composition, water chemistry, and plumbing infrastructure.

**Elevated Chloride and Sodium**

Chloride and sodium concentrations in surface and ground waters originate from several sources including agricultural chemicals, atmospheric deposition, bedrock and native soils, road salts,
sewage, and water softening treatment. Kelly et al. (2008) concluded that in a large New York watershed (91% forested or open land and 9% residential/commercial), 98% of the chloride and sodium input was anthropogenic. Specifically, 91% of the sodium chloride came from deicing salts, 4% from sewage, 3% from water softeners, and only 2% contributed by atmospheric deposition and weathering of rock/soils. Kauschal (2005) shows the increase in stream chloride concentrations of various land use in a Baltimore watershed (Figure 1).

Chloride is not readily involved in oxidation-reduction reactions, does not form complexes under most groundwater conditions, and is not absorbed onto mineral surfaces. Therefore, once chloride enters groundwater, it is highly mobile. Sodium, however, is less mobile being involved in ion exchanges competing with calcium and magnesium (Acosta et al 2011; Backstrom et al. 2004; Cooper et al. 2014; Lofgren, 2001; Norrstrom and Jacks 2009) resulting in their leaching, which leads to changes in soil chemistry and structure, as well as increases in soil colloid mobilization, including dissolved organic carbon (DOC) (Green et al. 2009). Sodium can also reduce potassium, ammonium, copper, zinc, and manganese ions in the soil, leading to reduced permeability, soil stability, and pH (NRCS 2017).

The trend of elevated chloride and sodium concentrations in groundwater has been observed in several studies, and in some cases, exceeding EPA’s guidelines. A national study of well water by DeSimone (2009) showed chloride from most wells tested ranging from 1.3 to 62.8 mg/L, and 2.1 percent of wells exceeded the EPA Secondary Maximum Concentration Limit (SMCL) of 250 mg/L. Mullaney et al (2009) analyzed wells in the Northeast and noted that 1.7 percent exceeded the SMCL. Further, Howard and Beck (1993) observed chlorides ranging up to 700 mg/L for domestic wells and 2,840 for urban springs in shallow groundwater in the Toronto area. In studies by the Maryland Geological Survey, sodium in groundwater ranged from 1.4 to 69 mg/L (Bolton, 1998) from 97 wells in the Piedmont region of MD, and 1 to 1,300 mg/L.
averaging

Figure 1. Average annual stream chloride concentration and percent impervious surface in Baltimore area watershed (adapted from Kaushal et al. 2005).

92.6 mg/L in the Aquia and Piney point aquifers, major Maryland coastal plain aquifers used for drinking water (Drummond and Bolton, 2010).

Water with elevated chloride and sodium would likely be noticeable by taste, and the saltiness may cause people to reduce their water consumption. The taste threshold for sodium varies, ranging from 30 mg/L to 460 mg/L (EPA, 2003). The EPA recommends sodium concentrations not exceed 30 to 60 mg/L to reduce taste issues, and 20 mg/L for those on a sodium-restricted diet. The EPA has an SMCL of 250 mg/l for chlorides due to taste.

**Corrosiveness on Plumbing**

The effect of salts on corrosivity of plumbing and relationship to incidences of elevated harmful contaminants such as metals is well studied. St Clair et al. (2016) points to increased salts displacing metals from aging water infrastructure and associated sediments, and older distributions systems being at greater risk of metal leaching. Old lead pipes, lead solder on copper pipes, brass faucets, fittings, and valves, including newer "lead-free," fixtures have been well documented as the key sources of metals in drinking water (Deberry et al. 1982; Nguyen et al. 2010; Shock and Neff 1998; Viraraghavan et al. 1999; Walker and Oliphant 1982). A key chemical mechanism associated with the degree of corrosivity of plumbing is related to the chloride sulfate mass ratio (CSMR), and described by numerous recent research studies (Edwards and Triantafyllidou 2007; Ng and Lin 2016; Nguyen et al. 2010; Stets et al 2018; Willison and Boyer 2012). These studies suggest that a CSMR ratio of >0.5 would enhance metal leaching. Willison and Boyer (2012) explained that increased chloride forms soluble lead
complexes thereby increasing lead concentrations in water, whereas sulfate forms insoluble lead sulfate complexes, which precipitates onto the pipe reducing further corrosion. These precipitates or scaling can build up overtime and help hold metals within the plumbing.

In examination of several significant metal toxicity incidents in the US, Edwards and Triantafyllidou (2007) showed that in each case the CSMR ratio was greater than 0.5, often reaching as high as 5.0. They further showed that in all of these cases changes in the type treatment coagulant used, e.g. alum to ferric chloride, resulted in the increase of CSMR and subsequent lead leaching. Interestingly, the authors also describe an incident of lead toxicity unrelated to change in coagulant use, rather the introduction of ion exchange treatment for arsenic. In this case, the ion exchanger, replaced sulfate with chloride, in effect lowering the ratio and forming soluble lead chloride. Pieper et al. (2017) further describes the relationship of CSMR and lead toxicity in the infamous Flint Michigan lead toxicity case. Here the authors show that the water CMSR was 0.45 before the utility switched water sources and certain water treatment protocols including using ferric chloride and discontinuation of the corrosion inhibitor, orthophosphate. Once lead was detected in the drinking water, CSMR had reached 2.04 due to increased chloride and lack of the orthophosphate and subsequent destabilization of the protective scales in plumbing and release of lead and iron into the water. As the authors state, even slight changes in water chemistry can cause disruptions with lead and iron scales, thereby releasing lead and iron particulates in water.

In a national study of surface and river waters, Stets et al. (2018) found that chloride, CSMR and Larson ratio (LR), a measure of the sum of chloride and sulfate to bicarbonate, increased in urban areas between 1992 and 2012. They further concluded that elevated chloride, CSMR, and LR, were correlated with snow affected urban areas. In a study in Virginia, Pieper et al., (2018) reported that 89% of wells tested had CSMR values above 0.5. They further noted median CSMR values of 7.7, 4.5, and 1.9 in wells near salt barns, near major roadways, and wells not in proximity to either source respectively.

The cost of corrosion to public water supply infrastructure has been estimated at $22 billion annually in 2002 (Edwards 2004). Corrosion related damages in private homes is estimated to cost 2-20 times higher than that of public water supplies (Levin et. al 1989; and Ryder, 1980). Edwards (2004) describes varied types of costs to homeowners including repair of water damages due to leaks, repairing/replacing failed pipes, increases in insurance premiums, and hot water heater repair and replacement. Another significant cost to homeowners is water filtration systems and their maintenance.

**Heavy Metal Mobility**

In addition to the effect of salt on corrosivity of plumbing and subsequent metal discharge, road salt affect the movement of metals within the soils, influencing surface and ground water quality. Novotny et al (1998) states that the partition coefficient (the concentration ratio of a chemical between two immiscible liquids at equilibrium) of metals is inversely proportional to the salt content of snow and ice, suggesting that salted ice may have a greater concentration of dissolved metals, observed as divalent ions. They also state that salted snowmelt may have a lower pH,
which can affect partitioning and increase metal solubility. In a study at two sites in Sweden, Backstrom et al. (2004) noted a 1-2 unit decrease in soil pH at two sites with salt deicing applications and noted a decrease in total organic carbon as associated with increasing electrical conductivity. The authors concluded that soil solution concentrations of cadmium (Cd), copper (Cu), lead (Pb) and zinc (Zn) are directly related to the use of deicing compounds due the strong correlation with electrical conductivity and chloride. They further suggest the major mechanisms of mobilization are ion exchange, pH decrease, formation of chloride complexes and possibly colloid dispersion, while noting that organic matter can counteract mobilization of metals such as copper and lead. Lumsdon et al. (1995) supports mechanism of chloride complexation with certain metals and noted affinities being greatest with Zn followed by Pb, then Cd and lastly Hg.

Several other studies support some of these mechanisms of metal motility and offer additional ones. Results of field studies by Nelson et al. (2009) described several mechanisms of metal motility exist including cation exchange, chloride complex formation, and colloid dispersion release of organic matter and/or clays that can complex metals. They further noted that sodium chloride showed the greatest release of copper and lead by means of indirect mobilization of organic matter. Their metal mobility rates varied between two test sites. The authors noted that soil characteristics greatly influence metal mobility, observing greater cadmium mobility in the site with higher clay content. The natural association of metals with organic matter and salts disruption/dispersion of soil colloids and release of organic matter resulting metal mobility is supported Linde et al. (2007) and Weng et al. (2002). They noted that in two urban soils, chromium, lead and mercury mobility was tied with the concentration of dissolved organic matter, whereas cadmium, nickel and zinc dissolution was more readily effected by salting. In their research of coastal aquifers, two watersheds receiving deicing salts and laboratory studies, Sun et al. (2015) noted similar correlation of lead and mercury mobility and increased sodium due to soil dispersion effect. They suggested that continued sea level rise and subsequent saltwater intrusion will likely increase mobilization of lead and mercury.

In a study of soils in the urbanized Yangtze river estuary, Zhao et al. (2013) noted that increasing salinity promoted the mobility of metals in the following order: Cd > Mn > Cu > Pb. The authors stated that properties of sediment, specifically organic carbon; and metal speciation, controlled the release of Cd and Mn respectively, while Cu and Pb were regulated by both factors. They suggested the varied processes of metal release influenced by increasing salinity might be due to chloride complexation for Cd, Cu and Pb affinity to FE-Mn oxides and organic matter, and acidity changes for Mn. They further noted that lower clay and TOC content of soils increased leaching of Cd, Cu and Pb. Acosta et al. (2011) noted that other road salts such as magnesium chloride and calcium chloride have even a greater effect on metal mobilization via ion displacement than the more commonly used NaCl.

Schuler and Relyea (2019) summarized salt impact on heavy metals in roadside soils showing heavy metals accumulation in soils along a gradient that decreases with distance from roads. The addition of salts results in lower concentration of metals in the upper organic soil horizon due to downward movement. Heavy metals then are disassociated with organic matter as they move downward through the horizons and moving into leachate zones and subsequently surface
waters. They noted that road salts may increase the rate of metal movement into groundwater and affect well water quality.

In a study of roadside infiltration trench soils in Sweden, Norrström (2005) observed similar Cl complex mechanism for Cd and Zinc (Zn) mobilization. Confirmation of the relative immobility of Pb was noted however, the author also stressed the point that since the accumulation of Pb in roadside soils is large; a threat to groundwater still exists due to colloidal transport. Hernadez-Soriano and Lopez (2012) studied bioavailability of metals formed in soils amended with various types of organic matter. They also noted that bioavailability is related to speciation of the metals in solution and metal organo complex formation. Amrhein et al (1992, 1994) showed that Cd, Cr, Fe, Ni and Pb concentrations tend to increase as salt concentrations increase and that Cr, Cu, Fe, Ni, and Pb, mobilization from near road soils was mainly controlled by organic matter mobilization. Further, organic matter mobilization was noted in soils with high exchangeable Na and low electrolyte concentrations.

Several studies have explored methods to remediate soils with heavy metal concentrations. Plants, as in the case of phytoremediation, plant byproducts and soil oxides have shown promise in remediation of metals. Rahman et al. (2016) provide a comprehensive review of numerous studies on this subject. Komarak et al. (2012) studied a chemical based stabilization of metals and showed that iron oxide application resulted in reducing water-soluble As, Cd, Pb and Zn by 39-95%. Zhang et al. (2013) studied the use of biochar to stabilize metals in soil thereby reducing mobilization and plant uptake. The high phosphorus and carbonate content of biochar acts to stabilize heavy metals and therefore reduce their bioavailability. In addition, biochar tends to be alkaline and therefore can increase pH and alkalinity, which contributes to a decrease in metal mobility. This increase in pH and cation exchange capacity by use of biochar is supported by Uchimiya et al. (2010) showing increased immobilization of metals. Park et al. (2011) showed that poultry manure biochar significantly increased Cd and Pb soil immobilization.

An important implication of increasing metal mobilization by road salts is the cost for treatment in both public and private water supplies. Reverse osmosis is the most commonly used water treatment approach to remediating heavy metal contamination for drinking water

**Mobilization of Radionuclides**

Uranium (U), radium (Ra), and radon (Rn) are naturally occurring radionuclides found in rock and soils and, therefore can be in surface and groundwater. Uranium is highly persistent and decays to radium which has a half-life of 1600 years and which then decays to radon with a half-life of less than four days. These elements release various types of high-energy radiation including alpha, beta, or gamma particles. The health risks associated with excessive exposure to radionuclides include nasal anemia, kidney damage, lung disease and cancer (EPA, 2002). The Center for Disease Control (CDC) states that elevated levels of Ra in groundwater can also produce lead to cataracts and osteosarcoma (CDC 2015). Exposure is through inhalation or ingestion of drinking water. EPA has established MCL’s for radionuclides as follows: gross alpha particle - 15 picocuries per liter (pCi/L); radium-226 and radium-228 - 5 pCi/L; and uranium - 30 micrograms per liter (μg/L).
Uranium in groundwater results from natural geologic processes including dissolution from rock and sediments (Alam and Cheng 2014) and is found in most natural waters at low concentrations. The authors noted that uranium was most associated with Fe-Mn hydroxides and silicate minerals, and further, release of U is controlled by several processes including dissolution of minerals, desorption from mineral surface, formation of aqueous U complexes, and reductive precipitation of U. They also noted U desorption increased with increasing pH, however in their study, U release by mineral dissolution was high at both high and low pH, and near neutral pH showed the lowest U concentration.

In a national study of 94 wells from public water supplies, Focazio et al. (1998) observed that 21 samples exceeded the current combined radium standard. Bolton (2000) tested 203 wells in MD and observed that radium-226, radium-228, gross alpha particle and gross beta particle activity increased with increasing total dissolved solids and sodium and chloride concentrations. He noted further that all the samples having greater than 10 and 15 mg/Liter sodium and chloride respectively had radium-226 plus radium-228 concentrations greater than the MCL of 5 pCi/L picocuries per liter.

In a Connecticut study, McNaboe et al (2017) showed that groundwater contaminated by deicing salts could mobilize radium and radon in subsurface layers. The authors suggest a strong correlation between sodium and Ra exists due to competition of soil adsorption sites. On the other hand, they observed a weak inverse correlation between salinity and Rn. They also suggest that wells with high levels of salt can result in up to four times the EPA radium MCL of 5 picocuries per liter (pCi/L). In addition, they showed an increased Rn gas flux from the water table. These studies show the extent of natural occurrence of radionuclides, and the potential for salt to increase radionuclide activity and risk to public health.

Conclusions and Implications

A preponderance of research results on the impacts of road salts points to multiple and significant environmental and public health risks. Clear trends of increasing sodium and chloride concentrations in ground and surface waters are widespread noted by the detriment of many ecosystems and aquatic species, and drinking water quality. In the case of groundwater and drinking water quality, several direct and indirect factors come into play to multiply the risk to health. The elevated chloride and sodium concentration in drinking water wells noted in numerous locations near highways have resulted in need for reverse osmosis filtration systems or well replacement in some cases. Elevated chloride increases corrosivity of plumbing, which increases leaching of several heavy metals, notably lead. Further as described, elevated chloride increases mobilization of radionuclides. Associated with these risk factors is an important public health dilemma. Though public water supplies are regularly, tested and treated, private wells are generally not monitored and the responsibility rests on the well owner. Many well owners are unaware of the water quality, risks associated with chlorides, or recommend testing practices. Increased education efforts are needed to reach private well owners and residents who rely on this source for drinking water. Special implementation challenges exist for low-income population, as the cost for well repairs, water testing and treatment may be prohibitive.
Efforts to mitigate deicing salt impacts are challenging and, have to date largely focused on implementing salt application/management strategies. Employing equipment calibration, enhanced weather prediction and decision support tools, pre-wetting of roads, and use of alternative deicers are practices that many state highway and county road management agencies utilize in effort to reduce salt usage. Reductions are promising and increased use of these application practices are needed by more jurisdictions and as well as the private sector responsible for private parking lots, private roads, etc.

Another approach to salt mitigation has been implementation of stormwater catchment, vegetated buffers and similar strategies. The high mobility of chloride in soils however presents difficulty in capturing chloride, though some benefit to heavy metal remediation has been noted in studies. Droste and Johnston (1993) described settleability of snowmelt solids, and noted settling times between 1 and 6 hours provided acceptable removals of TSS (up to 95%) and most metals associated with sediments including Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, and Zn. Guesdon, G. et al. (2016) tested a series of practices including a detention basin, a filtering bed, and a constructed wetland in a Quebec City watershed. They noted a removal efficiency that varied with the season of 16%–20% Cl, 3%–25% Na, 7%–10% Cd and 7%–36% Pb. In contrast, Snodgrass et al. (2017) observed watersheds with stormwater ponds had consistently higher conductivities and chloride concentrations during base flow and occasionally peaks in chloride and conductivity associated with winter storms. They noted that stormwater management practices are not protecting surface waters from road salt contamination and can result in groundwater plumes that transport chloride and sodium to streams during the year. These and other studies show that remediating chloride is difficult, and therefore the emphasis is on implementation of improved salt application strategies.

Shuler et al. (2019) offer an additional emphasis for addressing increasing freshwater salinization. They suggest that policy makers should develop regional and national regulations targeted at adopting sustainable salinization practices, especially in environmentally sensitive locations. The authors note challenges associated with this approach including potential safety risks and economic constraints. In addition to expanded monitoring to better understand threats, they do suggest a suite of management and stewardship practices including: a) assess strategies for various ion inputs; b) develop models that policy makers can use to estimate loading; c) assess costs and benefits of various means of reduction of salt use; and d) develop ion recovery systems and desalinization technologies.

A call to act to reduce both environmental and public health risks is evident. Increasing build up of salts in both ground and surface waters, with associated risks of metal and radionuclide exposures warrant expanded action. Options include enhanced voluntary adoption of proven salt reduction practices, or regulations to require said practices and engage the entire community involved in deicing activities. Further, enhanced education for private well owners on potential health risks, water testing and treatment practices are critical.
References:


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