Evaluating the influence of road salt on water quality of Ohio rivers over time

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ABSTRACT

Anthropogenic inputs have largely contributed to the increasing salinization of surface waters in central Ohio, USA. Major anthropogenic contributions to surface waters are chloride (Cl^-) and sodium (Na^+), derived primarily from inputs such as road salt. In 2012–2013, central Ohio rivers were sampled and waters analyzed for comparison with historical data. Higher Cl^- and Na^+ concentrations and fluxes were observed in late winter as a result of increased road salt application during winter months. Increases in both chloride/bromide (Cl^-/Br^-) ratios and nitrate ($N-NO_3^-$) concentrations and fluxes were observed in March 2013 relative to June 2012, suggesting a mixture of road salt and fertilizer runoff influencing the rivers in late winter. For some rivers, increased Cl^- and Na^+ concentrations and fluxes were observed at downstream sites near more urban areas of influence. Concentrations of Na^+ were slightly lower than respective Cl^- concentrations (in equivalents). High Cl^-/Br^- mass ratios in the Ohio surface waters indicated the source of Cl^- was likely halite, or road salt. In addition, analysis of $^{36}Cl/Cl$ ratios revealed low values suggestive of a substantial dissolved halite component, implying the addition of "old" Cl^- into the water system. Temporal trend analysis via the Mann–Kendall test identified increasing trends in Cl^- and Na^+ concentration beginning in the 1960s at river locations with more complete historical datasets. An increasing trend in Cl^- flux through the 1960s was also identified in the Hocking River at Athens, Ohio. Our results were similar to other studies that examined road salt impacts in the northern US, but a lack of consistent long-term data hindered historical analysis for some rivers.

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1. Introduction

Many natural and anthropogenic factors affect the geochemistry of surface waters in both rural and urban areas. Much of the impact on fresh water quality in the US comes from non-point sources, like road and agricultural runoff, with population and land use playing an important role in determining water quality. The salinization of fresh water occurs by means of surface runoff related to agricultural practices and increasing suburban and urban coverage of roadways. Major components of anthropogenic inputs into rivers and streams are chloride (Cl^-) and sodium (Na^+), derived primarily from modern contributions of halite, or road salt, whereas anthropogenic input of nitrate largely comes from agricultural fertilizers and treated wastewater. The application of salt to highways and roads lowers the freezing point of water, thereby melting snow and ice, but also creates saline water that runs off roadways into the local environment. This saline water is difficult to control and can travel a variety of pathways, flowing into the surrounding rivers and streams and infiltrating into the ground. Consequently, high-quality fresh water resources decrease, and roadside terrestrial habitats and aquatic ecosystems are degraded. Aesthetic effects arising from the increased salinization of water resources, such as salty taste, coupled with chloride's ability to corrode steel and damage pipes can result in increased treatment costs for drinking water. Elevated Cl^- concentrations in natural waters can degrade bridges, road beds, and other infrastructure as a result of its corrosive nature (Transportation Research Board, 1991). The acidification of streams, alteration of the mortality and biodiversity of aquatic biota, and the mobilization of toxic trace metals due to organic matter mobilization and ion exchange can all result from the salinization of fresh waters (Heath et al., 1992; James et al., 2003; Amrhein and Strong, 1990).

After World War II, large-scale implementation of road salt use began in the US by replacing abrasives as the main agent in clearing roadways. Coupled with the expanding US highway system at this time, salt usage increased quickly in the 1950s and 1960s,

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leveling off in the 1970s when the conversion from abrasives to road salt was nearly complete and highway agencies began implementing better salt application practices as salt's adverse effects were recognized (Transportation Research Board, 1991). Although elevated levels of Cl^- and Na^+ have become more common in surface and ground waters in northern regions of the US, road salt is presently not regulated as a primary contaminant to fresh waters (USEPA, 1988). While no limit for Na^+ concentration exists, the USEPA has only a secondary standard of 250 mg/L for Cl^- in drinking water to account for salty taste and a recommended chronic criterion of 230 mg/L Cl^- for at least 4 days in surface waters for aquatic life (USEPA, 2002; USEPA 1988). Surface waters that are exposed to abundant halite dissolution may present a different type of pollution not specifically covered by environmental policies and regulations. As a result, road salt use in the northeastern, midwestern, and Great Lakes region of the US and its impact on the quality of surface water and groundwater has been examined by a number of investigations in recent years (e.g. Chapra et al., 2009; Foos, 2003; Kaushal et al., 2005; Kelly et al., 2012; Koryak et al., 2001; Mason et al., 1999; Panno et al., 2006).

The rate of salt usage for highway deicing in the US has increased yearly by about 2–3% since the late 1970s, while Ohio had an annual road salt application rate of 200 kg/ha in the 1990s (Chapra et al., 2009; Kelly et al., 2012). The Ohio Department of Transportation (ODOT) uses around 700,000 tons of road salt per year statewide, and the city of Columbus has used about 23,000 tons yearly over the past decade (ODOT, 2008; City of Columbus, OH, 2014). During the 2012–2013 winter season, nearly 22,000 tons of salt were used by the city, compared to less than 6000 tons used during the previous, milder winter of 2011–2012 (City of Columbus, OH, 2014).

Previous work in Ohio has also demonstrated the detrimental environmental impact of point source pollution related to road salt. The Ohio EPA has found evidence in five Ohio communities of runoff from road salt storage piles contaminating public and private wells since 2009. Wells had to be abandoned in Preble County in southwest Ohio when the salty taste in drinking water could no longer be tolerated (Ohio EPA, 2011). In central Ohio, drinking water concerns pertinent to storm water runoff from salt piles has recently lead state organizations to create road salt storage guidelines for businesses and cities. Although these are not laws and have not been officially adopted, the guidelines request that road salt be stored outside of flood prone areas, a minimum of 90 m from streams and wells, and at least 30 m from ditches and storm drains (Ohio Water Resources Council, 2013).

The metropolitan Columbus, Ohio, area has seen population growth in the past two decades, resulting in urbanization and suburban sprawl. Little is known about the correlation between urban and suburban development across the US and its impact on long-term changes in baseline salinity of fresh surface water. Historical data on fresh water quality exist for many rivers and streams throughout Ohio, however much of it has never been utilized to characterize trends through time. Due to chloride's conservative nature, low natural background levels, and ability to be measured with reasonable precision, it serves as a reliable indicator of anthropogenic influence on water quality (Chapra et al., 2009). The overall goal of this research was to investigate the application of road salt, or halite, as the main source of Cl^- input to the Ohio surface waters. Additionally, possible anthropogenic induced trends in Cl^- and Na^+ concentration through time were evaluated in a number of Ohio rivers that are located in areas of urban and suburban influence. Rivers studied include the Hocking River, Olentangy River, Little and Big Darby Creeks (Darby Creek), Alum Creek, and Little Miami River (Fig. 1).

2. Methods

Water quality data from two main sources were tabulated to identify long-term changes in ion concentrations in rivers at multiple locations throughout central Ohio. Historical water chemistry and flow data were obtained from the US Geological Survey (USGS) National Water Information System. Previously unpublished water chemistry data from the Lyons research group for the 2000s at The Ohio State University (OSU) School of Earth Sciences were also utilized in this study. A total of 14 sites including 12 sites examined in the past corresponding to USGS gauge stations and 2 new sites (OA, HA) were sampled in June 2012 and March 2013 (Fig. 1; Appendix A). Not every site sampled in 2012 was sampled again in 2013 either due to difficulties in reaching the water because of ice and snow or due to a lack of long-term consistent historical data for the sample site. For this study, sample locations have identifiers beginning with O, A, L, D, and H for the Olentangy River, Alum Creek, Little Miami River, Darby Creek, and Hocking River, respectively.

Water samples were collected in two bottles at each sample location, one with a precleaned 125 mL wide mouth low-density polyethylene bottle and one with a 20 mL plastic scintillation vial with a poly cone cap. Before filling each bottle, water from the stream was used to rinse the bottles three times. The 125 mL sample at each location was for anion and cation analysis via ion chromatography using a Dionex Ion Chromatograph (DX-120). The 20 mL sample was analyzed for H_2O isotopes using a Picarro L1102-i water isotope analyzer. The samples for ion analysis were filtered either on-site or shortly after collection in the Environmental Geochemistry lab on the OSU campus. Samples were filtered through Whatman 0.45 μm polypropylene filters using a plastic disposable filtering device. All samples were stored at 4 °C and in the dark until analyses were completed. Historical data utilized from OSU are from samples that were filtered similarly to the samples in this study. USGS data are from samples filtered according to USGS standard procedure using a 0.45 μm disposable capsule filter (Wilde et al., 2004).

In order to resolve water contamination issues, it is essential to identify the source of contamination. Due to chemical similarities between the elements of chlorine and bromine, the concentration of Br^- relative to Cl^- in the Ohio rivers was utilized to aid in determining the origin of Cl^-. Bromide, like Cl^-, behaves conservatively in aquatic systems; however there are geochemical processes, such as precipitation of halite, that fractionate Br^- from Cl^-. As halite is precipitated, Br^- is preferentially excluded from the NaCl crystal lattice due to its much larger ion size relative to Cl^- (Oosting and Von Damm, 1996). Thus, waters affected by the dissolution of halite will have higher Cl^-/Br^- mass ratios due to the addition of Cl^- with little accompanying Br^- input. Previous studies have examined Cl^-/Br^- mass ratios of waters affected by the dissolution of halite (Davis et al. 1998, 2001; Panno et al. 2006).

In addition, the analysis of the cosmogenic isotope ^{36}Cl was utilized to interpret the origin of Cl^- in the Ohio rivers. Chlorine-36 is produced in the upper atmosphere by cosmic-ray spallation and has a long half-life of over 301,000 years. More recently, ^{36}Cl was released into the atmosphere in large amounts from 1950s nuclear weapons testing, establishing it as a useful hydrologic tracer worldwide (Davis et al., 2001). It behaves the same as stable chlorine and mixes with marine derived stable chlorine in the atmosphere, which results in relatively low but measurable ratios of $^{36}Cl/Cl_{total}$ in precipitation. The $^{36}Cl/Cl_{total}$ ratio in the ocean has been estimated at $0.5 \pm 0.3 \times 10^{-15}$ (Argento et al., 2010). Consequently, halite taken from deposits formed by the evaporation of saline or seawater long ago has little ^{36}Cl. As distance from the ocean increases, $^{36}Cl/Cl_{total}$ ratios in groundwater generally increase near