Integrated Environmental Assessment and Management — Volume 18, Number 6—pp. 1667–1677
Received: 8 November 2021 | Revised: 18 February 2022 | Accepted: 21 February 2022

Health & Ecological Risk Assessment

Impacts of urbanization on chloride and stream invertebrates: A 10-year citizen science field study of road salt in stormwater runoff

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Abstract
The use of deicing agents during the winter months is one of many stressors that impact stream ecosystems in urban and urbanizing watersheds. In this study, a long-term data set collected by citizen scientists with the Missouri Stream Team was used to evaluate the relationships between watershed urbanization metrics and chloride metrics. Further, these data were used to explore the effects of elevated chloride concentrations on stream invertebrate communities using quantile regression. While the amount of road surface in a watershed was a dominant factor in predicting the maximum chloride measurement, the median chloride concentration was also strongly related to the amount of medium-to-high density development in the watershed, suggesting that nonmunicipal salt use is an important contributor to increases in base flow chloride concentrations. Additionally, chloride concentration appears to be one of the many factors that impact invertebrate density and diversity measurements, with decreases in invertebrate diversity corresponding with the US Environmental Protection Agency water quality criteria. Our findings suggest that the use of chloride-based road salt on municipal roads as well as on nonmunicipal settings is contributing to a loss of diversity and density of aquatic invertebrate communities in urban regions. *Integr Environ Assess Manag* 2022;18:1667–1677. © 2022 The Authors. *Integrated Environmental Assessment and Management* published by Wiley Periodicals LLC on behalf of Society of Environmental Toxicology & Chemistry (SETAC).

KEYWORDS: Benthic macroinvertebrates, Deicing, Quantile regression, Urban development, Water quality

INTRODUCTION
In recent years, there has been growing awareness of the deleterious effects of urbanization on aquatic ecosystems. While many factors like sedimentation, channelization, impervious surfaces, altered flow regimes, and pollutants in stormwater are contributing factors, the salinization of streams due to the use of chemical road deicers has drawn particular scrutiny (Blasius & Merritt, 2002; Findlay & Kelly, 2011; Gardner & Royer, 2010). It has been noted in the northern plains of the United States that chloride (Cl^-) concentrations can be 10 times greater in surface waters in urban areas than in forested areas (Findlay & Kelly, 2011). A comparison of two watersheds in central Illinois demonstrated that streams in an urbanized watershed can have up to a 20-fold increase in Cl^- concentrations in winter relative to spring and summer, while an agricultural watershed has a threefold increase (Lax et al., 2017).

In the United States, the use of chloride-based salts for winter road deicing has increased steadily since its initial usage in the 1940s. Between 1975 and 2003, the use of salt for deicing more than doubled from 8 to 18.5 million metric tons (USGS, 2014), with current use in excess of 22 million metric tons annually (Bolen, 2022). Moreover, from 1987 to 2010 road salt sales increased 3.9% per year on average, while urban land cover increased 2.9% per year in the northern United States, indicating that road salt use has been increasing faster than urban development (Corsi et al., 2015). While improved management practices could decrease road salt sales, continued urban expansion may offset progress in reducing the use of chemical deicers. Thus, since road salt use has been correlated with increasing Cl^- concentrations in freshwater streams (Gardner & Royer, 2010), it appears that freshwater streams in urban and urbanizing areas will continue to be at risk for higher Cl^- concentrations in winter storm seasons for the foreseeable future.

Aquatic biota maintains an osmotic balance with their environment, which, in freshwater organisms, is achieved by

This article includes online-only Supporting Information.

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Published 24 February 2022 on wileyonlinelibrary.com/journal/ieam.

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expending energy to prevent the diffusion of their salts into the environment (Cañedo-Argüelles Iglesias, 2020). However, if an increase in salinity within the freshwater environment surpasses the isotonic point of the organisms, then they are required to instead expend energy to block salt uptake or to excrete excess salt. In this case, sublethal or lethal toxicity may occur due to damage at the cellular level (Cañedo-Argüelles Iglesias, 2020). The US Environmental Protection Agency (USEPA) has set the aquatic life criteria for Cl^- at 230 mg/L for chronic exposure and at 860 mg/L for acute exposure (USEPA, 1988). Many studies have noted that freshwater streams can exceed the acute standard, particularly in the winter months when deicing of roads occurs, and thus have tried to determine the lethal effects of various Cl^- levels on a variety of aquatic invertebrate taxa (Benbow & Merritt, 2004; Blasius & Merritt, 2002; Gardner & Royer, 2010; Mangahas et al., 2019). Most of these studies have examined these effects under short-term (24–96 h) exposures in laboratory conditions and found wide variation in taxa responses.

Several mesocosm experiments have attempted to address these discrepancies and to more accurately replicate stream conditions by increasing the length of Cl^- exposure and incorporating multiple species (e.g., Clements & Kotalik, 2016; Kotalik et al., 2017). Very few studies, however, have examined benthic macroinvertebrate communities in the field and over an extended period of time, that is, years rather than days or months. There is some indication that background levels of Cl^- may be increasing in streams in the northern United States and Canada because of incomplete flushing of road salt after winter storm events due to retention in soil and groundwater, resulting in the gradual release of Cl^- into streams (Findlay & Kelly, 2011; Robinson et al., 2017; Wallace & Biastoch, 2016). Thus, over time it is possible that there may be either a general decline in the richness and/or abundance of the macrobiotic community or a change in the structure of the community (Wallace & Biastoch, 2016).

Because road salt use predominates in urban and suburban areas, it is difficult to differentiate the effects of Cl^- from the effects of other urban stressors such as temperature, toxic contaminants, sedimentation, and habitat loss (Hassett et al., 2018; Heugens et al., 2001). This challenge results not only from the presence of multiple stressors but also the potential for compounding effects of the stressors, including additive or multiplicative interactions (Heugens et al., 2001). For instance, salt increases the mobilization of some heavy metals, potentially increasing their toxicological effect and leading to consequences throughout the food web (Schuler & Relyea, 2017).

In this study, we examine a long-term set of field data collected across an urban gradient in St. Louis County, Missouri, to evaluate how differences in land use impact Cl^- concentration and how increasing Cl^- concentration affects stream invertebrate communities. This data set has been collected by citizen scientists with the Missouri Stream Team (MST) Program. We first evaluate the land use parameters that show the greatest impact on Cl^- metrics. Further, we utilize a quantile regression analysis of invertebrate responses to Cl^- metrics, increasing our ability to identify the impacts of road salt in a system impacted by multiple stressors.

MATERIALS AND METHODS

Study area

Study sites are located in St. Louis County, Missouri, USA (Figure 1). The St. Louis region has a temperate climate that generally results in several winter storm events per year. In the Climate Normal Period of 1981–2010, the St. Louis area averaged a three-month winter period minimum temperature of −3.4 °C and average daily temperatures of 13.9 °C (NOAA, 2020). Thus, winter precipitation that falls as snow or as a snow/sleet/ice mix is often followed soon after by a melt or rainfall resulting in a runoff.

Roads may receive applications of rock salt or liquid brine more often than winter storm events are recorded because of the difficulty of predicting whether temperatures will be above or below 0 °C when a storm threatens the region. In 2019, the 55 municipalities that share a stormwater permit with the Metropolitan St. Louis Sewer District (MSD) used 66 157 tons of road salt (R. Biehl, MSD, personal communication, December 21, 2020), while the Missouri Department of Transportation (MODOT) used 12 000 tons of salt on the 2800 miles of road they maintain in the area (R. Becker, MODOT, personal communication, December 9, 2020). These figures do not include salt use on 3165 miles of county-maintained roads, on roads in the other 33 municipalities in the county, or on private roads, parking lots, and sidewalks.

Urban growth in the St. Louis region began in the second half of the 1700s along the Mississippi River riverfront of the City of St. Louis (which is not part of St. Louis County) and

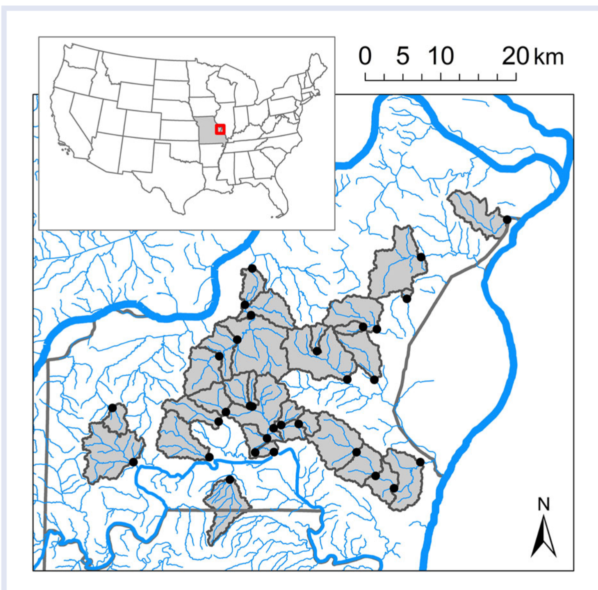

FIGURE 1 Study sites (black points), streams (blue lines), watersheds (gray areas), and county boundaries (black lines) in the St. Louis region, Missouri. The widened blue lines are the Missouri River to the North, the Mississippi River to the East, and the Meramec River to the South

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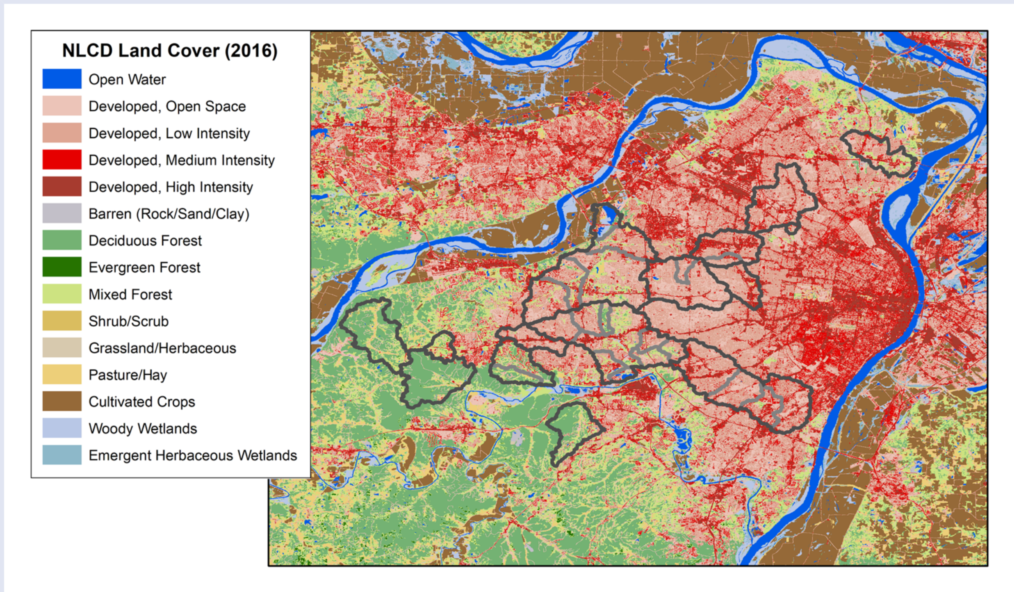

FIGURE 2 Land uses in study watersheds and sub-watersheds (outlined in dark and light gray, respectively) and the surrounding areas of the Saint Louis, Missouri region

has gradually expanded westward. The eastern edge of St. Louis County is dominated by high-density development, while the western edge of the county is predominantly low-density development and semi-intact forest patches (Figure 2). On average, land cover in St. Louis County is 40.5% deciduous woody vegetation, 28.1% urban/impervious, and 25.2% grassland (including lawns) (East-West Gateway, 2017).

Three large rivers form boundaries in the St. Louis region: Mississippi River, Missouri River, and Meramec River (Figure 1). Tributaries to these rivers are influenced by differences in geology, hydrology, and land development (Criss & Wilson, 2003).

Data collection and selection

MST. The MST was formed in 1989 and is a partnership of the Missouri Department of Conservation (MDC), the Missouri Department of Natural Resources (MDNR), and the Conservation Federation of Missouri. The citizen science program of MST is focused on volunteer water quality (WQ) monitoring. Training of volunteers is tiered, with Introductory Level training on stream hydrology and biological monitoring, Level 1 training on water chemistry and physical habitat assessment, Level 2 quality assurance (QA) verification for chemistry methods and invertebrate identification, and Level 3 QA of all field methods. Data from Level 2 and 3 citizen scientists can be used by the MDNR for regulatory and enforcement purposes (MDNR, 2014). Statewide since 1993, 5812 individuals completed Introductory Level training, 3356 completed Level 1 training, 1081 achieved Level 2, and 111 participants reached Level 3 (C. Riggert, MDC, personal communication, April 16, 2021).

Further details about the history of MST and the training of citizen scientists, including QA measures, in the program may be found in Supporting Information Appendix A.

All biological data and most of the Cl^- data in this field study were collected by trained citizen scientists with MST. In the past 15 years, there has been much discussion about the value of citizen science in scientific research (Aceves-Bueno et al., 2017; Albus et al., 2019; Bonney et al., 2009; Cooper et al., 2007; Kosmala et al., 2016; Lewandowski & Specht, 2015; Loperfido et al., 2010; Scott, 2017; Specht & Lewandowski, 2018). While several acknowledge the value of citizen science for community engagement and education (e.g., Bonney et al., 2009; Stepenuck & Genskow, 2017), the accuracy of citizen science data and therefore its credibility has been questioned (Aceves-Bueno et al., 2017). Where it has been compared to the same standard, professional data have not been shown to be more accurate than citizen science data (Lewandowski & Specht, 2015). In fact, research shows that, with adequate training and testing, citizen science data can approach the quality of professional data (Kosmala et al., 2016; Lewandowski & Specht, 2015). Additionally, for projects with long-term citizen science participation and projects where the volunteers have a stake in the research, accuracy can be quite high (Aceves-Bueno et al., 2017).

The present study utilized a long-term data set collected by a cohort of dedicated citizen scientists who participated in at least two years of biological monitoring and completed over 20 h of training with professional scientists (Supporting Information Appendix A). Moreover, since MST allows individuals to select their own sampling site(s), most chose locations that were of interest or concern to them and were geographically near their area of residence or recreation,

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that is, they were motivated to participate. Equipment for both the biological and the Cl^- monitoring was supplied by the MST, and so was standardized. All the above measures were aimed at increasing the value and accuracy of the citizen science data.

Macroinvertebrate sampling. For each sampling event, aquatic macroinvertebrates were collected at three locations within a 91 m stream reach following MST methods, which were adapted from the Izaak Walton League's Save Our Streams program (Firehock & West, 1995). Each of the three subsamples was collected by disturbing approximately 0.9 m^2 of sediment immediately upstream of a 0.84 m^2 kick net with a 500 μm mesh. Large stones were rubbed and removed; then the substrate was disturbed to a depth of 7–15 cm to dislodge invertebrates and wash them into the net. Where possible, all three subsamples were collected from riffles, proceeding in a downstream to upstream direction.

Invertebrates from each subsample were removed from the net, identified to appropriate taxonomic group (generally order or family), counted, and recorded. The time spent removing invertebrates from the net was recorded as (number of minutes) x (number of people picking). A WQ rating was calculated for the entire sample based on taxa richness and sensitivity to pollution (Supporting Information Appendix A). The WQ ratings range from 0 to 49, with ratings above 23 considered excellent, 18–23 considered good, 12–17 considered fair, and ratings below 12 considered poor. To ensure invertebrate collection and identification were conducted by well-trained citizen scientists, only data from volunteers with Level 2 certification or above were included in this study.

Chloride monitoring. The Cl^- measurements by MST citizen scientists were made using Hach Chloride QuanTab® Test Strips. In response to high winter Cl^- measurements by a small group of citizen scientists in 2011, volunteers organized a region-wide, long-term winter sampling effort to characterize the magnitude and extent of Cl^- pollution in area streams. Volunteers were asked to monitor approximately once a week (or as frequently as their schedule permitted) beginning in late November and ending in the spring when Cl^- concentrations returned to prewinter concentrations. Due to the simplicity of the test method, no specialized QA training is provided by MST for Cl^- monitoring. Therefore, data collected by volunteers with Level 1 certification or above were included. In a test comparing Cl^- measurements using QuanTab test strips with measurements by silver nitrate titration in water samples collected within the study area, test strips consistently measured at or below titration values with relative percent differences ranging from 46.8% to −3.2% with a median of 20.4% and a mean concentration difference of 47.5 mg/L; concentrations, as measured by titration, ranged from 75 to 1430 mg/L.

Site and data selection. The study period was December 2009 to April 2020. The Cl^- data were limited to the winter and early spring (December–April) to compensate for unbalanced data collection at sites where volunteers tended to collect data only seasonally (either winter or summer). To be included in the analysis, a site needed a minimum of 10 Cl^- measurements with at least seven taken in January, February, and March to ensure that data include sampling in months likely to witness winter storm events; regardless of the total number of Cl^- measurements, at least 50% of the measurements had to come from these months. If more than two samples were taken in a day, the highest value and lowest value of the day were included.

In addition to the Cl^- data requirement, at least one complete macroinvertebrate sample must have been collected within 400 m of the Cl^- monitoring location during the study period. Invertebrate samples were excluded if a tributary entered the stream between the Cl^- and invertebrate collection locations. Complete macroinvertebrate samples include three full net-sets and recorded the amount of time spent collecting invertebrates from the nets, with the exception of Fishpot Creek data, which did not include time. Based on the selection criteria, 31 sites were included in this study.

Watershed urbanization data

Values for a set of seven variables were determined for each of the study watersheds: watershed area; percent of the watershed as road; percent impervious area; percent developed area; percent low, medium, and high (LMH) intensity developed area; percent medium and high (MH) intensity developed area; and percent undeveloped green space. Watershed characteristics were identified using ArcGIS 10.8 (ESRI). Roadway area as observed in 2013 in each watershed was determined using a GIS coverage developed and provided by MSD (2017). Percent imperviousness was determined using the National Land Cover Database (NLCD) imperviousness raster for 2016 (NLCD, 2019a). The NLCD raster for 2016 land cover from the US Geological Survey (NLCD, 2019b) was used to evaluate the amounts of developed and undeveloped land in each watershed. The NLCD land cover includes four classes of developed area: open space (<20% impervious), low intensity (20%–49% impervious), medium intensity (50%–79% impervious), and high intensity (80%–100% impervious). The developed area in each watershed was summarized as the total percent of developed land (interpreted as a measure of all urban and suburban development), the percent of developed land under LMH intensity (interpreted as a measure of residential, commercial, and industrial development, not including parks, large lawns, cemeteries, and other green spaces), and the percent of developed land under MH intensity (interpreted based on visual inspection areas defined as MH intensity in Figure 2 as a measure of commercial and industrial development). Percent undeveloped green space was determined as the combined portion of each watershed that was identified by the NLCD land cover as forest, grassland, or pasture.

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Analysis

Aquatic invertebrate data for each three-net sampling event were quantified using the MST WQ rating, the total number of invertebrates, and the catch per unit effort (CPUE) in invertebrates per minute. The Cl^- data for each site were summarized into five metrics: maximum, 75th percentile, median, mean, and interquartile range. The relatedness among these Cl^- metrics was identified using Pearson correlation, as was the relatedness among watershed variables. To prevent multicollinearity in regression analysis, the number of Cl^- metrics and land-use variables were reduced based on the correlations within each group of variables. Only variables with a correlation coefficient of less than 0.75 were included.

Multiple linear regression was used to evaluate the response of Cl^- metrics to watershed urbanization metrics. The Cl^- metrics were log-transformed to improve the normality of the residuals.

Relationships between aquatic invertebrate measurements and Cl^- variables were evaluated using quantile regression. Quantile regression is a method that allows estimation of the rate of change of the relationship between paired variables in a different portion of data set than is estimated by ordinary least-squares regression. This method is particularly useful in ecological systems for exploring questions that relate to the maximum, minimum, or other quantiles of a data set rather than the measures of central tendency. Because ecological communities generally, and aquatic invertebrates specifically, may be impacted by multiple simultaneous stressors, it is possible for stressors other than the measured variable(s) to impact biological conditions, thereby increasing the heteroscedasticity of the results. Therefore, quantile regression at the 90th and 95th percentiles was used (following Cade & Noon, 2003) to estimate the upper limits of invertebrate metrics as a function of Cl^- concentrations. These upper limits provide an estimate of the biological condition that could be achieved if other limiting factors were reduced or eliminated (Cade & Noon, 2003).

RESULTS AND DISCUSSION

Watershed characteristics and in-stream chloride

In all, 31 sites met the requirements for inclusion in this study. Watersheds ranged from 0.9 to 76.5 km^2 in size and had 2.0–46.7% impervious surface cover. Percent roadway ranged from lows of 1.5%–2.8% in the four least developed watersheds to highs of 9.2%–10.1% in the four watersheds with the highest roadway areas. Additional watershed characteristics can be seen in Table S1.

No correlation between watershed size and land-use variables was detected using Pearson correlation coefficients ($p > 0.05$). Among the six land-use variables, all pairs had correlation coefficients >0.7 or <−0.8 (Figure S1). To decrease the likelihood of Type 1 errors, watershed area and the two least-correlated land-use variables were retained for further analysis. The percent roadway (% Road) was retained as a surrogate for road-based pollution, including road salt, and the percent of MH intensity development (% MH) was retained as a surrogate for pollutants and habitat alteration associated with urban development, including nonroad-based salt application.

Across all study sites, 2004 Cl^- measurements were recorded during the winter and early spring from December

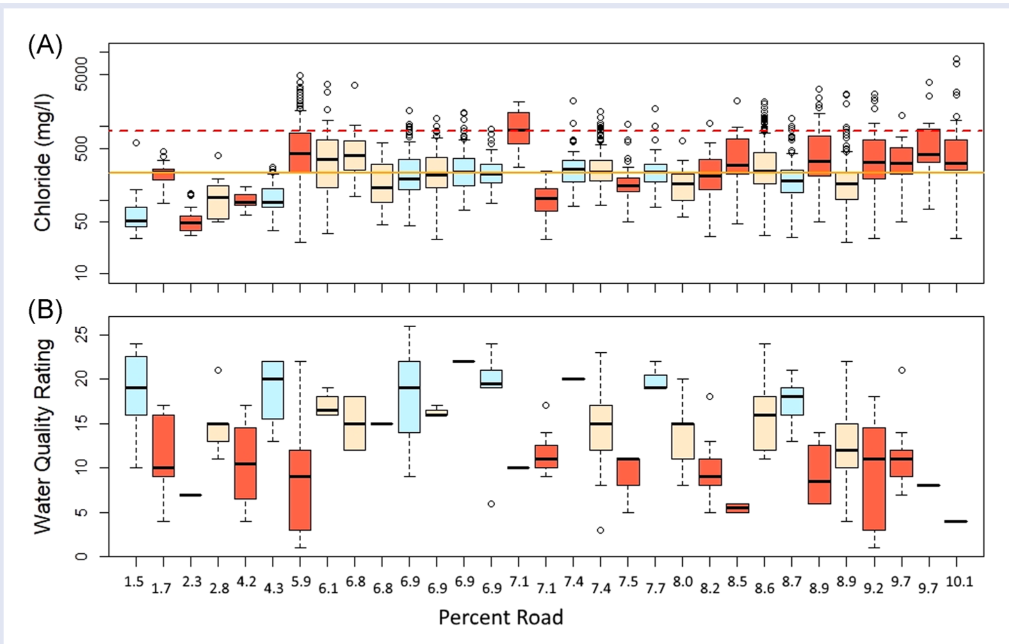

FIGURE 3 Regional results for (A) chloride measurements and (B) aquatic macroinvertebrate water quality ratings for each site in St. Louis County, Missouri. Bar color represents the categorization of the median water quality rating with light blue denoting good (18–23, $n = 8$), tan for fair (12–17, $n = 9$), and red for poor (<12, $n = 14$). The dashed red line and solid orange line represent the US Environmental Protection Agency acute and chronic toxicity thresholds of 860 and 230 mg/L, respectively

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TABLE 1 Results of multiple regression for chloride metrics (maximum chloride concentration [Cl Max] and median chloride concentration [Cl Med]) against watershed metrics (watershed area, percent of watershed as road [% Road], percent of watershed as medium and high-intensity development [% MH], and the interaction)

	Log (Cl Max)			Log (Cl Med)		
	Estimate	SE	*p* Value	Estimate	SE	*p* Value
Watershed area	−0.004	0.003	0.167	−0.003	0.002	0.077
% Road	0.113	0.055	**0.0498**	0.105	0.030	**0.002**
% MH	0.026	0.021	0.767	0.038	0.011	**0.003**
% Road × % MH	0.0002	0.003	0.922	−0.004	0.002	**0.019**

Note: *p* values <0.05 are shown in bold.

2009 to April 2020 (Figure 3A). The overall maximum recorded concentration was 8000 mg/L.

Several metrics were calculated to represent the Cl^- conditions at each site. To prevent overparameterization, these metrics were reduced to two based on the lowest value of the Pearson correlation (Figure S2). This resulted in the selection of maximum Cl^- (Cl Max) and median Cl^- (Cl Med) as representative metrics of the amount of Cl^- at each location. These metrics are considered surrogates for the acute and chronic conditions, respectively, at each site. The relationship of each Cl^- metric (log-transformed) was evaluated against the watershed area, % Road, % MH, and the % Road × % MH interaction using multiple regression (Table 1).

For Cl Max, a significant multiple linear regression model was found, $F(4, 26) = 6.689$, $p = 0.0008$, with an adjusted R^2 value of 0.431. Within this model, only % Road was a significant predictor of Cl Max ($p = 0.0498$; Table 1). Based on this model, the likelihood of a maximum Cl^- concentration that exceeds the USEPA acute toxicity threshold of 860 mg/L increases when the proportion of land in the watershed that is paved road exceeds 5% (Figure 4).

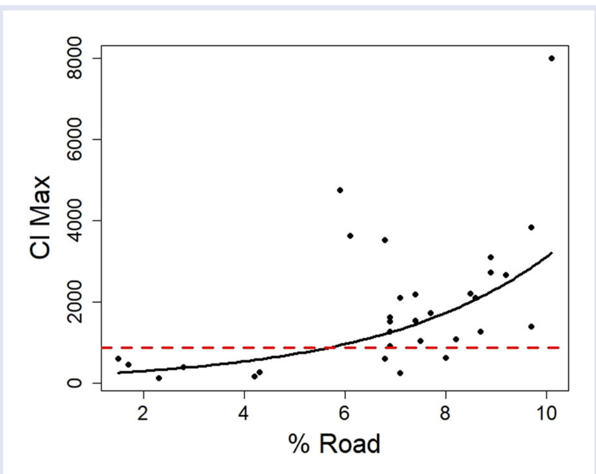

FIGURE 4 Relationship between maximum chloride concentration (Cl Max) and percent of the watershed covered by roads (% Road). The dashed line represents the US Environmental Protection Agency acute toxicity threshold of 860 mg/L

Similarly, a significant model was found for Cl Med, $F(4, 26) = 9.50$, $p < 0.0001$, with an adjusted R^2 value of 0.531. In this case, % Road, % MH, and their interaction were all significant predictors of Cl Med ($p = 0.002$, 0.003, 0.019, respectively; Table 1) with increasing Cl^- concentrations at sites with increased roadway or development. As with Cl Max, the increase in Cl Med within our study area is notable when % Road is greater than 4.5% (Figure 3A).

The direction of the interaction of % Road and % MH suggests that, in watersheds with little medium-to-high density urban development (dashed line in Figure 5), the amount of roadway has a strong influence on the median amount of Cl^- in the stream. Meanwhile, watersheds with a large amount of medium-to-high density urban development (solid line in Figure 5) have a high median Cl^- concentration that increases only slightly as road surface area increases.

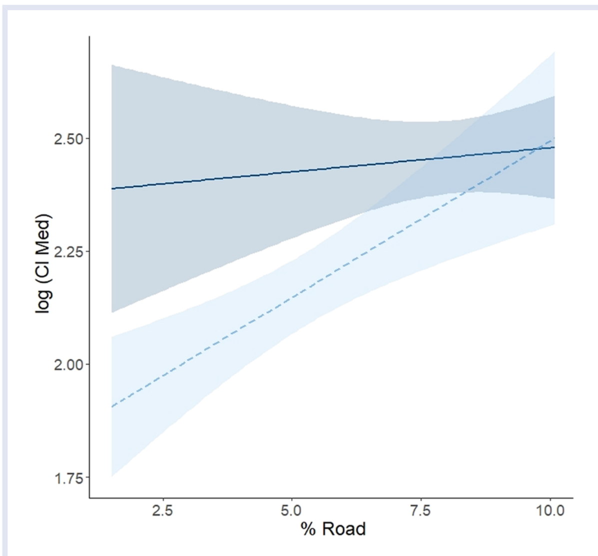

FIGURE 5 Graphical representation of how median chloride concentration (Cl Med) is affected by the interaction between percent of the watershed covered by roads (% Road) and percent of the watershed as medium and high intensity (% MH) development. The solid line represents the relationship between Cl Med and % Road when % MH development is 1 SD above the mean (when % MH development is relatively high); the dashed line represents this relationship when MH development is 1 SD below the mean (when % MH development is relatively low). Shaded areas are the 90% confidence interval

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It is unsurprising that the amount of road surface would be a dominant factor in the concentrations of Cl^- in streams in a region where road salt is the primary deicing agent. Public works departments use salt on roadways, most of which convey runoff quickly to streams (Oswald et al., 2019). Some of this salt is retained in soils and groundwater and released gradually over the ensuing months (Oswald et al., 2019; Robinson et al., 2017), which is reflected in the contribution to median concentrations.

The importance of medium-to-high density development on median Cl^- concentrations may be indicative of a greater contribution of nonroad sources to residual salt. This contribution may take several forms: overapplication that results in salt remaining on the pavement after winter storms have passed, uncovered or improperly managed salt storage piles (Ohio Water Resources Council, 2013), and increased infiltration of salt-laden runoff into stormwater retention basins (Snodgrass et al., 2017). The authors have observed evidence of all three of these mechanisms within the study watersheds (Supporting Information Appendix B). While overapplication and improperly managed salt storage may also occur on roadways, municipal users have been compelled by a combination of stormwater regulations under the National Pollution Discharge Elimination System and budgetary considerations to moderate their use of road salt and adopt salt management practices.

One site receiving runoff from a highly developed watershed had a surprisingly low maximum Cl^- concentration. The furthest downstream site on Creve Coeur Creek (site 3) had the third lowest maximum concentration (246 mg/L), despite having a watershed that was over 7% covered by road and being relatively near an upstream site with comparable % Road and a maximum Cl^- concentration of 1540 mg/L. This site with an unexpectedly low Cl^- concentration is situated downstream of a natural 320-acre oxbow lake that is a remnant of an earlier course of the Missouri River. The residence time and dilution of Cl^- into the large volume of the lake could account for these lower readings.

Invertebrate response

A total of 215 benthic macroinvertebrate sample sets were collected between 2010 and 2019. The WQ ratings for these samples ranged from 1 to 26 with a median rating of 14. Of the samples, 80 were categorized as poor, 76 as fair, 55 as good, and only 4 as excellent (Figure 3B). During the study period, considerable temporal variability in WQ ratings was observed at several sites, though no trends were apparent regionally or within watersheds (Figure S3)

Across all sites and samples, a total of 28 259 aquatic macroinvertebrates were collected and identified. Taxa encountered in the greatest number of samples were sowbugs (Isopoda), caddisflies (Trichoptera), aquatic worms (Oligochaeta), mayflies (Ephemeroptera), leeches (Hirudinea), and midges (Nematocera). The most numerous taxa were sowbugs, scuds (Amphipoda), and caddisflies, with over 5000 individuals of each.

The WQ ratings were highly variable relative to % Road (Figure 3B), with poor ratings at sites with less than 2.5% Road and with good ratings at sites with as much as 8.7% Road. In addition, the relationships between measures of the invertebrate density and metrics for both Cl^- and watershed urbanization were heteroscedastic, with greater variability in the data at low Cl^- concentrations (Figures 6 and S4). The low quality of invertebrate communities and the heteroscedasticity of the data may be attributed to the influence of non-Cl^- stressors (Hassett et al., 2018; Heugens et al., 2001). For example, at Williams Creek (site 55; Table S1), the southernmost watershed with only 2.3% road, the WQ rating was poor even though Cl^- concentrations and development were low. In this location, the depauperate invertebrate community may be a result of its proximity to several large horse farms, which may contribute sediment and nutrient loads to the stream as well as reduce riparian vegetation (Agouridis et al., 2005). In addition, within-site variability may be high due to invertebrate sample collections occurring throughout the year.

Due to the heteroscedasticity of the data sets and to the potential for negative impacts on invertebrates from unknown stressors, we did not evaluate the central tendencies of the data. Instead, quantile regressions of Cl Max and Cl Med were plotted against three invertebrate metrics: CPUE and Total Invertebrates as measures of invertebrate density along with WQ Rating as a measure of invertebrate diversity (Figure 6). In all cases, Cl^- concentrations were negatively related to aquatic macroinvertebrates, both in terms of their density and diversity. This negative relationship is consistent with prior studies (Cañedo-Argüelles Iglesias, 2020; Hintz & Relyea, 2019; Wallace & Biastoch, 2016).

Assuming that the use of quantile regression was successful in decreasing the impacts of confounding factors, it seems that the total number of invertebrates collected at a site may drop by roughly half when maximum Cl^- measurements reach the 3000–5000 mg/L range and when median Cl^- concentrations reach 700–900 mg/L (Figure 6). Additionally, fewer invertebrates were collected per minute (as measured by the CPUE) in streams with high Cl^- concentrations (Figure 6), suggesting a decreased abundance of invertebrates in these streams.

Invertebrate diversity, as measured by the MST WQ Rating, was also negatively related with both Cl^- metrics (Figure 6). Based on the 95^{th} quantile regression for the WQ rating measurement, it appears that WQ ratings in streams in the St. Louis region can be expected to drop from the excellent category (>23) to the good category (18–23) once sites exceed a Cl Max of 900 mg/L and a Cl Med of 100 mg/L. In addition, sites appear unlikely to attain a good WQ rating when Cl Med is above 250 mg/L (Figure 3A).

Implications for salt management

Variation in stream Cl^- levels may be influenced by differing road deicing methods. Snow and ice removal on roads in this study area is provided by multiple entities: the

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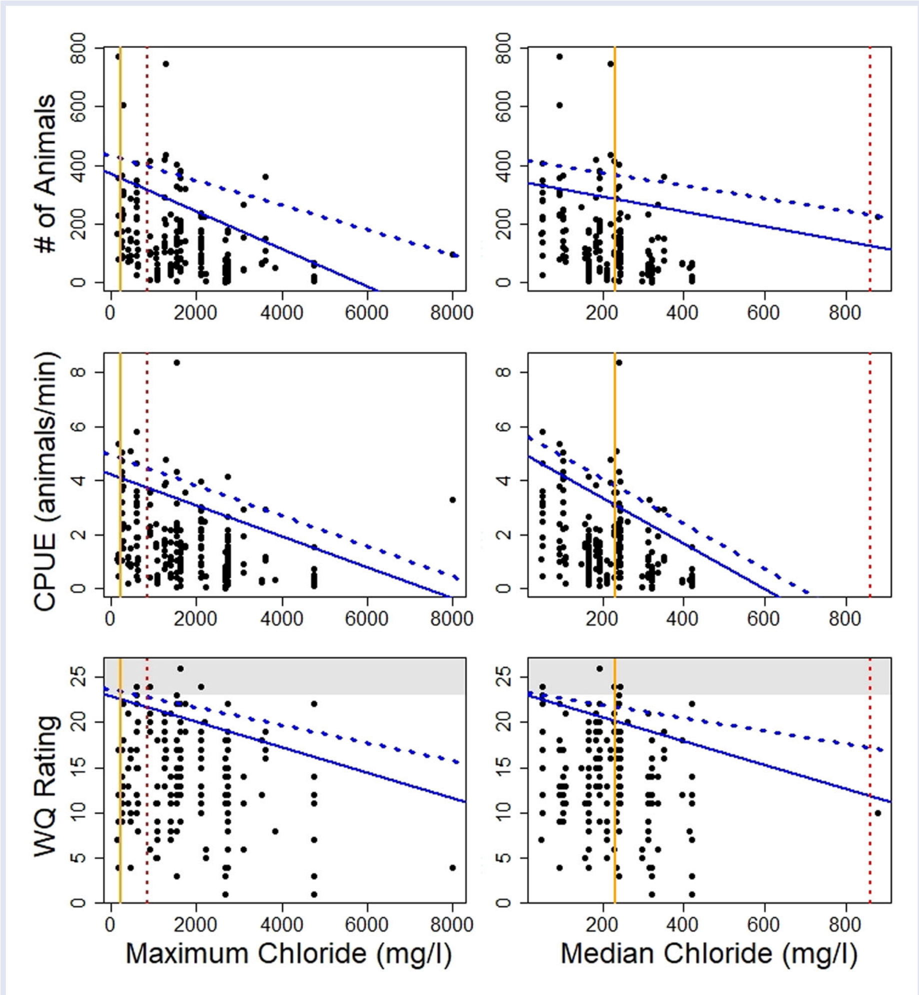

FIGURE 6 Quantile regressions of invertebrate (animal) metrics on chloride metrics. Vertical solid orange line is the chronic toxicity threshold (230 mg/L), vertical dashed red line is the acute toxicity threshold (860 mg/L), solid blue line is the 90th quantile regression, dashed blue line is the 95th quantile regression, and gray area represents "excellent" water quality ratings (>23)

MODOT (for state roads), the St. Louis County Department of Transportation (for county roads); and various municipal Public Works Departments (for secondary roads within the 88 municipalities in St. Louis County). In addition, a large number of nonmunicipal, independent entities are responsible for snow and ice removal in the many commercial and industrial parking lots within the study area that are primarily managed by private services. This pattern of multilevel responsibility for the deicing of pavement in urban and suburban areas is not unique to the study region.

Agencies and municipalities selectively use best management practices (BMPs) as guidelines or codes of practice for winter road deicing and maintenance operations. The selection of BMPs is generally guided by the need to provide the safest winter driving road conditions in the most cost-effective manner while secondarily limiting environmental risks and damage (Shi et al., 2009). Guidelines typically describe operational efficiencies related to the procurement, storage, distribution, and application of road salt with an aim to minimize waste and related labor and material costs (Nixon & Williams, 2001). The BMPs with the greatest potential to prevent Cl^- stream pollution are those that minimize the application of road salt through practices like prewetting (Hossain et al., 2016), brining (Haake & Knouft, 2019), and prevention of salt loss through general housekeeping practices associated with salt transport and storage (Fay et al., 2015; Meegoda et al., 2004; Ostendorf et al., 2006). In addition, the control and optimization of road salt application are affected by the interaction of many variables; the most important of which is the effect of weather (Blackburn et al., 2004). Complex choices of material and application rates are greatly affected by winter severity, stability, and instability indexes of ambient and road temperatures, wind, and precipitation (Blackburn et al., 2004). For example, in Missouri, application rates of road salt are suggested to be varied and optimized between 75 and 300 pounds per road mile, depending on road, snow, and weather conditions (Jang et al., 2011).

The adaptive use of multiple BMPs, deicing chemicals, and application rates by municipal entities is driven by a combination of the cost-effectiveness of these strategies and the requirements for minimum control measures set

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forth in state-issued stormwater permits. This has resulted in improved efficiency of salt application nationwide (Boselly, 2001; Fay et al., 2013). Meanwhile, nonmunicipal (residential and commercial) applications are likely contributing heavily to Cl^- concentrations due to application rates on parking lots and walkways that may be several times those used on roadways (Perera et al., 2010). The overapplication of salt in these developments may be related to a lack of financial or legal incentives for the implementation of such BMPs and the potential liabilities associated with incomplete deicing of pavement. Unfortunately, studies of residential and commercial salt use and BMP implementation are lacking, likely due to the many commercial and individual applicators involved (Sander et al., 2007) and the complexity of the socioeconomic factors that may impact salt use within a relatively small area (Haake & Knouft, 2019).

Studies have shown that many aquatic species can tolerate brief periods of high Cl^- concentrations (Benbow & Merritt, 2004; Jackson & Funk, 2019; Mangahas et al., 2019). On the other hand, studies show that long-term exposures to even moderately increased Cl^- concentrations can lead to impacts on the growth and reproduction of invertebrates (Elphick et al., 2011; Lawson & Jackson, 2021; Soucek & Dickinson, 2015). In this study, peak Cl^- concentration in urbanized streams appear to be driven by salt spread on roadways, but additional nonroadway salt inputs in watersheds with greater medium- and high-density urban land cover are an important driver of median Cl^- concentrations associated with nonstorm periods during the winter. Based on these findings, we suggest that future research should be directed toward studying the current use of salt in nonroadway deicing and the potential impacts of implementing BMPs for the application and storage of road salt for commercial and residential use.

ACKNOWLEDGMENT

We are grateful to the current and former staff of the Missouri Stream Team program for their ongoing support of citizen scientists, especially Chris Riggert, Karen Westin, and Darlene Haun, who provided encouragement and expertise in the development of the chloride monitoring project. This project could not have been successful without the water monitoring contributions of Stream Team volunteers, including Rick Archeski, Earl Barge, Michael J. Engle, Eric Karch, Dave Kuechenmeister, Mark Kuechenmeister, J. Michael Lahm, Steve McCarthy, San Mueller, Claire L. Schosser, Besa Schweitzer, Ed Shafer, Beth Skelton, Coralynne Westcott, Sarah Wilson, and several additional anonymous volunteers. The authors also thank Chris Riggert and two anonymous reviewers for their comments and suggestions, which substantially improved this manuscript. The Volunteer Water Quality Monitoring portion of the Missouri Stream Team Program is jointly run and funded by the Missouri Department of Natural Resources and the Missouri Department of Conservation. This Program provided all training and equipment for the citizen science efforts.

CONFLICT OF INTEREST

The authors declare no conflicts of interest.

AUTHOR CONTRIBUTION

Danelle M. Haake conceived and initiated the study, coordinated volunteer efforts, and performed statistical analyses. Danelle M. Haake, Stephen Krchma, Claire W. Meyners, and Robert Virag collected data, explored the literature, discussed results, and drafted the manuscript.

OPEN DATA BADGE/OPEN MATERIAL BADGE

This article has earned an Open Data Badge and Open Material Badge for making publicly available the digitally shareable data necessary to reproduce the reported results. The data and material are available at https://doi.org/10.5281/zenodo.6206870. Learn more about the Open Practices badges from the Center for Open Science: https://osf.io/tvyxz/wiki.

DATA AVAILABILITY STATEMENT

The following supplements and project data are available at DOI: 10.5281/zenodo.6206870:

- PDF Supplement with additional figures and appendices.
- Excel file with data used in this manuscript.
- Text file with R code used in data analysis.

SUPPORTING INFORMATION

The supporting information file contains a table of site and watershed characteristics, four additional figures, and two appendices. The first appendix describes the training and methods used by Missouri Stream Team volunteers. The second appendix is a compilation of 24 photos or photo sets showing examples of salt management.

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 DOI: 10.1002/ieam.4594 © 2022 The Authors