

A Fresh Look at Road Salt: Aquatic Toxicity and Water-Quality Impacts on Local, Regional, and National Scales

STEVEN R. CORSI,^{*,†}
DAVID J. GRACZYK,[†] STEVEN W. GEIS,[‡]
NATHANIEL L. BOOTH,[†] AND
KEVIN D. RICHARDS[†]

U.S. Geological Survey, Middleton, Wisconsin, and Wisconsin State Laboratory of Hygiene, Madison, Wisconsin

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A new perspective on the severity of aquatic toxicity impact of road salt was gained by a focused research effort directed at winter runoff periods. Dramatic impacts were observed on local, regional, and national scales. Locally, samples from 7 of 13 Milwaukee, Wisconsin area streams exhibited toxicity in *Ceriodaphnia dubia* and *Pimephales promelas* bioassays during road-salt runoff. Another Milwaukee stream was sampled from 1996 to 2008 with 72% of 37 samples exhibiting toxicity in chronic bioassays and 43% in acute bioassays. The maximum chloride concentration was 7730 mg/L. Regionally, in southeast Wisconsin, continuous specific conductance was monitored as a chloride surrogate in 11 watersheds with urban land use from 6.0 to 100%. Elevated specific conductance was observed between November and April at all sites, with continuing effects between May and October at sites with the highest specific conductance. Specific conductance was measured as high as 30 800 $\mu\text{S}/\text{cm}$ ($\text{Cl} = 11\,200\text{ mg/L}$). Chloride concentrations exceeded U.S. Environmental Protection Agency (USEPA) acute (860 mg/L) and chronic (230 mg/L) water-quality criteria at 55 and 100% of monitored sites, respectively. Nationally, U.S. Geological Survey historical data were examined for 13 northern and 4 southern metropolitan areas. Chloride concentrations exceeded USEPA water-quality criteria at 55% (chronic) and 25% (acute) of the 168 monitoring locations in northern metropolitan areas from November to April. Only 16% (chronic) and 1% (acute) of sites exceeded criteria from May to October. At southern sites, very few samples exceeded chronic water-quality criteria, and no samples exceeded acute criteria.

Introduction

Road-salt runoff poses an increasing threat to aquatic ecosystems that are influenced by urban land use and transportation corridors. Four broad issues suggest that road-salt runoff is a serious and increasing threat to the nation's receiving waters. First, there is a multitude of historical evidence documenting detrimental effects of road salt on water chemistry and aquatic life. This issue was recognized at least as early as the 1960s (1). Studies have continued each

decade since then, with more comprehensive evidence of water-quality impacts from road salt. A small sampling of some representative topics studied includes specific water-quality impacts such as increased chloride and sodium concentrations, seasonality, climatic and land-use influence, vertical density gradients, and influence on sediment pore water, mixing and alteration of turnover in lakes (2–5), and aquatic toxicity impacts (2, 6, 7). Second, road salt usage in the United States has increased steadily beginning in the 1940s through the current decade (8, 9). Average annual salt sales in the United States for deicing purposes by decade beginning in 1940 were 0.28 (1940s), 1.1 (1950s), 4.1 (1960s), 8.7 (1970s), 8.8 (1980s), 13.0 (1990s), and 16.0 (2000–2008) million metric tons per year. Third, urban development is increasing each year (10), which increases the amount of impervious area on which winter deicing operations are conducted. This collective information suggests that the increasing road-salt usage trends of the previous seven decades will likely continue under current management conditions. Fourth, chloride, and to a large degree sodium, the two primary ions in road salt, remain in solution, making it difficult with present-day technology to design effective management practices for reduction of road-salt loadings to receiving waters after application. Currently, reduction in usage appears to be the only effective road-salt-runoff management strategy.

The objective of this study was to investigate the influence of road-salt runoff on surface water and aquatic organisms. Water-quality sampling was conducted on a local and regional scale. In the Milwaukee metropolitan area (local scale), streams were sampled for chloride, specific conductance, and aquatic toxicity to assess direct impact on aquatic organisms. In southern and eastern Wisconsin (regional scale), streams were monitored continuously for specific conductance, a surrogate for chloride, to assess potential impact on aquatic organisms. On a national scale, analysis of historical data was conducted for 17 metropolitan areas. Data were retrieved from the U.S. Geological Survey (USGS) National Water Information System (NWIS) for chloride concentrations from streams sampled between 1969 and 2008. Data were compared to U.S. Environmental Protection Agency (USEPA) water-quality criteria and analyzed for seasonal differences.

Materials and Methods

Study Sites. Local Scale. Twelve streams in the Milwaukee metropolitan area and one reference stream north of Milwaukee were sampled in February and March 2007 for determination of water chemistry and aquatic toxicity (Table 1, Figure 1). Twelve of the streams had substantial urban land-use contribution and the reference stream had 80% natural areas and no urban land use (Parnell Creek). Drainage areas of these streams ranged from 16.4 km^2 (6.33 mi^2) at Willow Creek to 1833 km^2 (872 mi^2) at the Milwaukee River (Table 1). A 14th stream, Wilson Park Creek, was monitored selectively from 1997 through 2007 during deicing periods. Sample results from these 14 streams include chloride concentrations, specific conductance values, and bioassays using *Pimephales promelas* and *Ceriodaphnia dubia*.

Regional Scale. Eleven streams in southeast Wisconsin were monitored using continuous specific conductance sensors with resulting data used as an indication of road-salt runoff (Table 1, Figure 1). These streams represent a gradient of land use with urban influence ranging from 6.0 to 100%.

National Scale. Individual water-quality samples for chloride in 17 major metropolitan areas around the country

* Corresponding author phone: (608) 821-3835; fax: (608) 821-3817; e-mail: scorsi@usgs.gov.

[†] U.S. Geological Survey.

[‡] Wisconsin State Laboratory of Hygiene.

TABLE 1. Watershed Characteristics for Study Sites in Wisconsin Organized by Geographic Location

monitoring location	USGS site ID	drainage area (km²)	land use percentage			aquatic toxicity sampling dates	continuous specific conductance
			urban	agriculture	natural areas ^a		time period, months of complete record
Milwaukee metropolitan area							
Lincoln Creek at Milwaukee	040869416	24.8	98	0	2	2/26/2007	June 2003–Oct 2005, 29 months
Menomonee River at Menomonee Falls	04087030	89.9	30	44	25	3/7/2007	Jan 2004–Oct 2004, 10 months
Little Menomonee at Milwaukee	04087070	51	44	38	18	2/26/2007	
Underwood Creek at Wauwatosa	04087088	47.1	87	4	9	2/26/2007	
Honey Creek at Wauwatosa	04087119	26.7	99	0	1	2/26/2007	
Menomonee River at Wauwatosa	04087120	318	61	25	15	2/26/2007	
Kinnickinnic River at Milwaukee	04087159	48.7	98	0	2	2/26/2007	
Milwaukee River at Milwaukee	04087000	1800	16	54	30		May 2002–Oct 2004, 27 months
Milwaukee River at Clybourne Ave	04087012	1833	17	53	30	3/7/2007	
Oak Creek at South Milwaukee	04087204	64.7	63	21	16	3/7/2007	Jan 2004–Feb 2005, 14 months
Root River at Greenfield	04087214	38.1	92	3	6	3/7/2007	
Root River near Franklin	04087220	127	67	15	18	3/7/2007	
Willow Creek near Germantown	040870195	16.4	24	47	29	3/7/2007	
Wilson Park Creek at Milwaukee	040871488	29.4	100	0	0	1997–2008	Jan 2001–Oct 2008, 75 months
Green Bay area, Madison area, small communities, and rural							
Duck Creek near Howard	04072150	280	6	74	20		May 2001–Oct 2003, 28 months
Garners Creek at Kaukauna	04084468	53.6	69	25	6		Dec 2003–Dec 2004, 10 months
Parnell Creek near Dundee	04086175	21.8	0	20	80		
Pheasant Branch Creek at Middleton	05427948	47.4	26	67	7		Jan 2007–Apr 2008, 16 months
W. Branch Starkweather Creek at Madison	05428600	31.3	50	42	8		Jan 2004–Jan 2005, 11 months
Delavan Lake Inlet at Lake Lawn	05431017	56.5	6	66	28		Jan 2007–Sep 2008, 23 months
Badger Mill Creek at Verona	05435943	52.6	39	49	12		May 1998–Sep 2007, 102 months

^a Natural areas include forest, grasslands, wetlands, and water.



FIGURE 1. Location of study sites in Wisconsin and metropolitan areas in the United States used for aquatic toxicity evaluation from road salt.

were retrieved from NWIS, the U.S. Geological Survey national water-quality database (Figure 1). Candidate streams were selected based on the latitude and longitude of the monitoring location and its proximity to major urban land-use areas. Streams ultimately chosen for this study included those that were sampled for chloride between 1969 and 2008, had at least 12 samples in the cold-weather months (November to April), 12 samples in the warm-weather months (May to October), and a drainage area of less than 2600 km². A total of 12 005 samples from 162 sites in the northern part of the United States and 2378 samples from 50 sites in the southern part of the United States (south of St. Louis) were used.

Water-Quality Sampling. For the 13 Milwaukee area streams, sampling periods were targeted at runoff events during road-salt application periods. Real-time specific conductance data from Wilson Park Creek were used as an indicator of road salt in Milwaukee area streams and used to initiate this sample collection. Water-quality samples were collected manually during the February 26 and March 7, 2007 sampling periods. Samples were collected either by submerging sample bottles directly into the center of the channel for wadeable streams or by using a weighted-bottle sampler to collect cross-section integrated samples from a bridge for nonwadeable streams (11). Comparison of the relation between chloride and specific conductance was used to assess potential bias in results. All samples were within 10% of the resulting linear regression except those with chloride concentrations less than 230 mg/L, where chloride and sodium were a less dominant influence on specific conductance. Flow-weighted composite samples were collected at Wilson Park Creek from 1997 through 2007. Specific details of the sampling protocol used from this site have been previously published (12).

Weather data were retrieved from three nearby NOAA weather stations (General Mitchell International Airport, Mount Mary, and Germantown). On February 24, 25, and 26, 2007, average snowfall was 16, 15, and 2 cm (0.9, 1.7, and 0.2 cm water equivalent), and maximum air temperatures were -0.5, 2.8, and 2.8 °C, respectively. This snowfall triggered plowing operations and salt application. On March 7, there was an average of 5.7 cm of snow (0.4 cm water equivalent), and maximum air temperature of 0.5 °C. This was not enough snow to trigger general plowing; however, salt was applied on paved surfaces to melt snow and ice. Salt application and temperatures greater than 0 °C for both of these events resulted in runoff from impervious areas flowing to storm sewers and receiving streams.

Measurements from continuously deployed specific conductance sensors were recorded at least every hour and as frequently as every 5 min depending on the individual site and specific hydrologic conditions. Instantaneous specific conductance was measured in the 13 Milwaukee area streams at the time of the 2007 sampling periods. All specific conductance sensors were maintained in accordance with standard USGS methods (13).

Analytical Methods. Chloride and toxicity tests were conducted at the Wisconsin State Laboratory of Hygiene using well established methods that are presented in the Supporting Information.

Results

Runoff Samples in the Milwaukee Area. Toxicity was exhibited in samples from 7 of the 12 urban-influenced watersheds in the Milwaukee metropolitan area that were collected during road-salt application periods in February and March, 2007 (Figure 2). Adverse response in *C. dubia* tests occurred in samples with chloride concentrations of 1610 mg/L or greater. Adverse response in *P. promelas* tests occurred in samples with chloride concentrations of 2940 mg/L or greater. The IC₂₅ values (the concentration at which

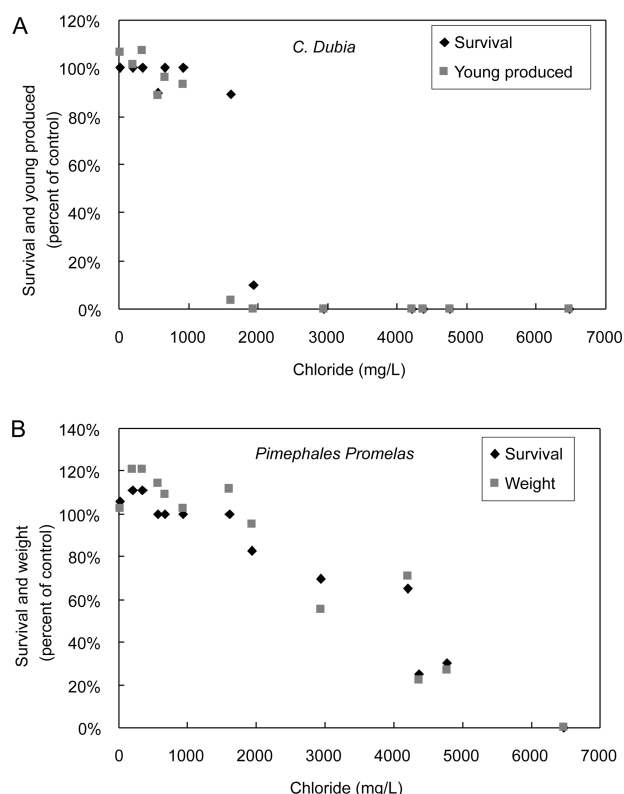


FIGURE 2. Chronic bioassay results in relation to chloride concentration in samples collected from 13 streams in the Milwaukee, WI metropolitan area, February–March, 2007: (A) *C. dubia* survival and mean young produced, and (B) *P. promelas* survival and mean weight.

there is 25% inhibition) computed using measured chloride concentrations in these stream samples were 1050 mg/L for *C. dubia* and 1810 mg/L for *P. promelas*. Chloride concentrations were elevated above the EPA acute water-quality criteria concentration of 860 mg/L in eight of these samples and above the EPA chronic water-quality criteria concentration of 230 mg/L (14) in 11 of these samples, indicating potential for aquatic toxicity effects. A sample collected at the rural reference site during the February sampling period had a chloride concentration of 20.4 mg/L and did not exhibit toxicity.

Specific conductance results from continuous monitoring in Wilson Park Creek in Milwaukee during 2007 indicate that conditions similar to the February and March 2007 sampling periods were common during the cold-weather period of 2007 (Figure 3).

Long-Term Toxicity from Road Salt. Results from 37 samples collected from 1997 to 2007 at Wilson Park Creek in Milwaukee demonstrate long-term toxicity effects in numerous samples and a distinct relation to chloride concentration (Figure 4). Concentrations at which chronic result effects were observed from this long-term sampling program were very similar to corresponding concentrations where chronic effects were observed from the 2007 sampling events in the Milwaukee metropolitan area. In chronic *C. dubia* assays, no young were produced when chloride concentration was 1770 mg/L or greater (43% of samples) and complete mortality was observed at chloride concentrations of 2420 mg/L and greater (38% of samples). Initial toxic effects began between 600 and 1100 mg/L. Mortality was observed in acute *C. dubia* assays for all samples with chloride concentrations greater than 1900 mg/L. In chronic *P. promelas* assays, reduced weight and survival was present when concentrations were 2920 mg/L or greater. Results for both of these organisms in chronic bioassays indicate scatter

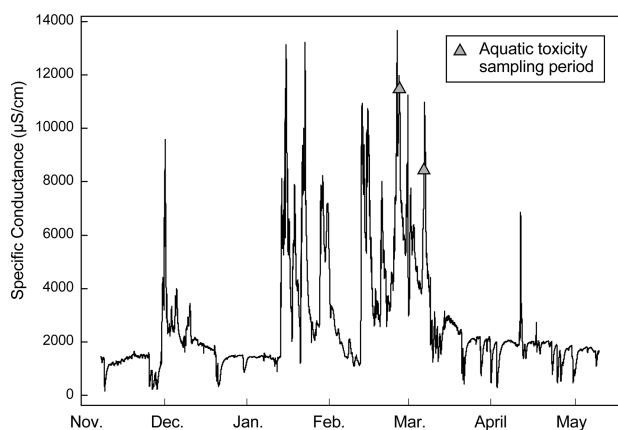


FIGURE 3. Specific conductance in Wilson Park Creek in Milwaukee, WI during 2007 in reference to aquatic toxicity sampling periods (triangles) for 13 Milwaukee area streams.

or uncertainty with lower chloride concentrations. This could be due to variability in the actual test, confounding contaminants in urban runoff, or a combination of these factors. It was difficult to determine the exact concentration at which road-salt effects began for chronic *C. dubia* assays due to this variability. In *P. promelas* acute assays, only two samples were influenced with initial effects occurring between 4660 and 6290 mg/L.

Regional-Scale Influence: Continuous Monitoring of Road-Salt Runoff. Eleven streams in urban regions of Wisconsin were monitored with continuous specific conductance sensors during cold- and warm-weather periods from 1998

to 2008 (Table 1). Between 1 and 10 years of data were available depending on the individual site. Linear regression was conducted from results of concurrent analysis of chloride and specific conductance in samples from these streams. To reduce negative bias in residuals when other ions influence this relation at low concentrations (specific conductance <1400 $\mu\text{S}/\text{cm}$), data in the regression were constrained to those samples with specific conductance greater than 1400 $\mu\text{S}/\text{cm}$. This resulted in a slope of 0.374 and intercept of -328 ($R^2 = 0.997$, Supporting Information Figure S1). This regression is used for the remainder of this paper to provide chloride concentration estimates (referred to as Cl_{est}) from measurement of specific conductance.

The maximum observed specific conductance in these streams increased with increasing urban land use (Figure 5). The maximum Cl_{est} for seven of these sites exceeded the USEPA acute water-quality criteria value of 860 mg/L. The maximum Cl_{est} exceeded the USEPA chronic water-quality criteria value of 230 mg/L at each of the 11 sites with a maximum Cl_{est} of 289 mg/L for the least impacted stream.

The highest continuous specific conductance results at these eleven sites occurred during cold-weather months (Figure 6). The most dramatic impacts from road-salt runoff were observed in the two highly urban watersheds Lincoln and Wilson Park Creeks in Milwaukee with specific conductance often exceeding 10 000 $\mu\text{S}/\text{cm}$ ($\text{Cl}_{\text{est}} = 3410$ mg/L) and at times exceeding 20 000 $\mu\text{S}/\text{cm}$ ($\text{Cl}_{\text{est}} = 7150$ mg/L, Figure 6A). Maximum monthly specific conductance at four sites with intermediate influence ranged between 3000 and 8000 $\mu\text{S}/\text{cm}$ (Figure 6B). These sites had 26–69% urban land use. Maximum monthly values at four sites with low influence were still substantially impacted by chloride in cold-weather

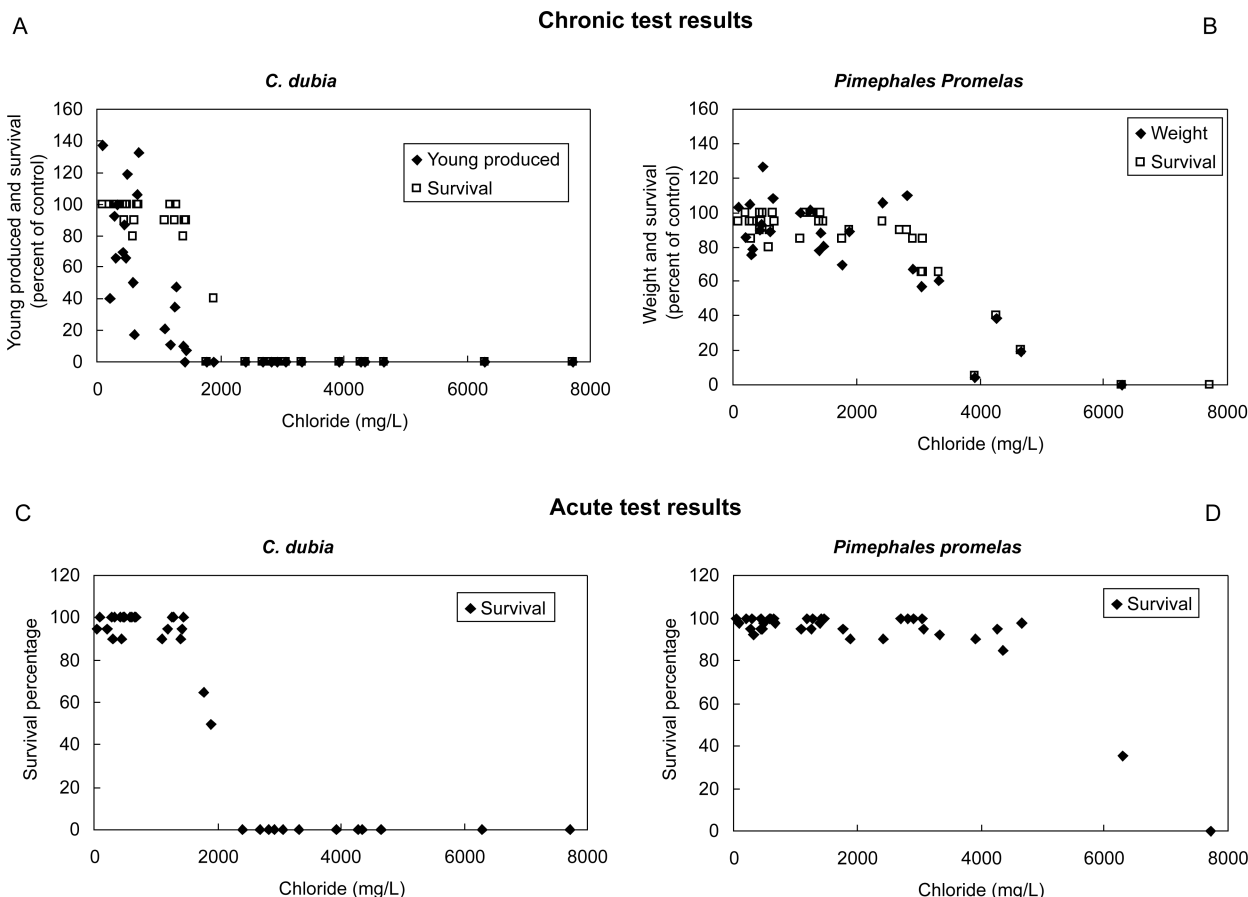


FIGURE 4. Bioassay results in relation to chloride concentration in samples collected from Wilson Park Creek in Milwaukee, Wisconsin, 1997–2007: (A) *C. dubia* survival and mean young produced in chronic bioassays, (B) *P. promelas* survival and mean weight in chronic bioassays, (C) *C. dubia* survival in acute bioassays, and (D) *P. promelas* survival in acute bioassays.

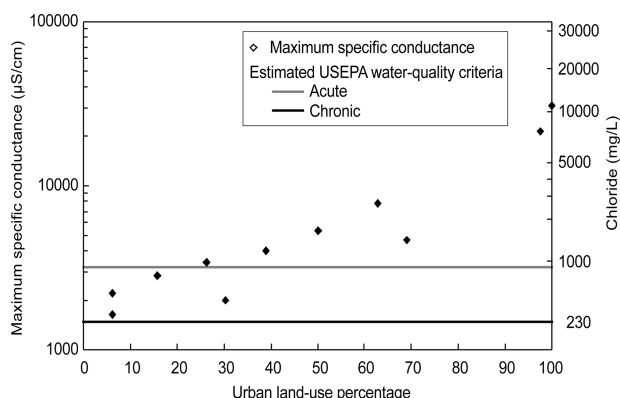


FIGURE 5. Maximum specific conductance compared to urban land-use percentage in 11 Wisconsin streams with reference to U.S. Environmental Protection Agency water quality criteria for chloride (19).

months, but maximum monthly specific conductance was less than 3000 $\mu\text{S}/\text{cm}$ (Figure 6B). These sites had 6.0–30% urban land use. Although most of these watersheds were small to medium in size with a drainage area of 25–280 km^2 , the Milwaukee River at Milwaukee has a drainage area of 1800 km^2 and still was impacted by road-salt runoff with a maximum specific conductance of 2850 $\mu\text{S}/\text{cm}$. In all months, the average monthly maximum specific conductance was greatest in the sites with urban land use of 98% or greater, followed by those with 26–69% urban land use, and least in sites with less than 26% urban land use (Table 1, Figure 6).

In some cases, specific conductance decreased through the warm-weather months, reaching a minimum in October (Figure 6C). Specific conductance in the two highly urban watersheds decreased from May through October by 34% and 39%. The average monthly maximum in these two streams was greater than 1200 $\mu\text{S}/\text{cm}$ throughout the entire year. Specific conductance data from Oak Creek (63% urban land use) also decreased by 26% from May through October. Other sites either did not have sufficient data to evaluate warm-weather conditions or did not exhibit this effect.

National Scale. USGS chloride sample results from streams near metropolitan areas were retrieved from 1969 to 2008 for assessment of potential road-salt influence throughout the country and to provide context for the more intensive Wisconsin study results (Figure 7). The maximum number of sites per metropolitan area was 29 (Denver) and the maximum number of samples per metropolitan area was 1690 (Cleveland).

A total of 898 samples were collected and analyzed for chloride at 21 monitoring locations within the Milwaukee area. Results exceeded 230 mg/L chloride in at least one sample at 90% of monitoring sites during cold-weather months and 33% of monitoring sites during warm-weather months (Figure 7A). Similarly, 57% of these monitoring sites exceeded 860 mg/L chloride in at least one sample during cold-weather months, and none during warm-weather months (Figure 7B).

Most northern metropolitan areas included in the analysis demonstrated the same pattern as the Milwaukee area sites. A total of 51% of all 168 northern monitoring locations had at least one sample with concentrations exceeding 230 mg/L during cold-weather months and 15% exceeded that concentration during warm-weather months. A total of 23% of northern monitoring locations had at least one sample with concentrations exceeding 860 mg/L during cold-weather months and 1% exceeded that concentration during warm-weather months. Ten of 13 metropolitan areas had more monitoring sites that had a chloride sample result exceeding 230 mg/L during cold-weather months than during warm-weather months. Nine metropolitan areas had more moni-

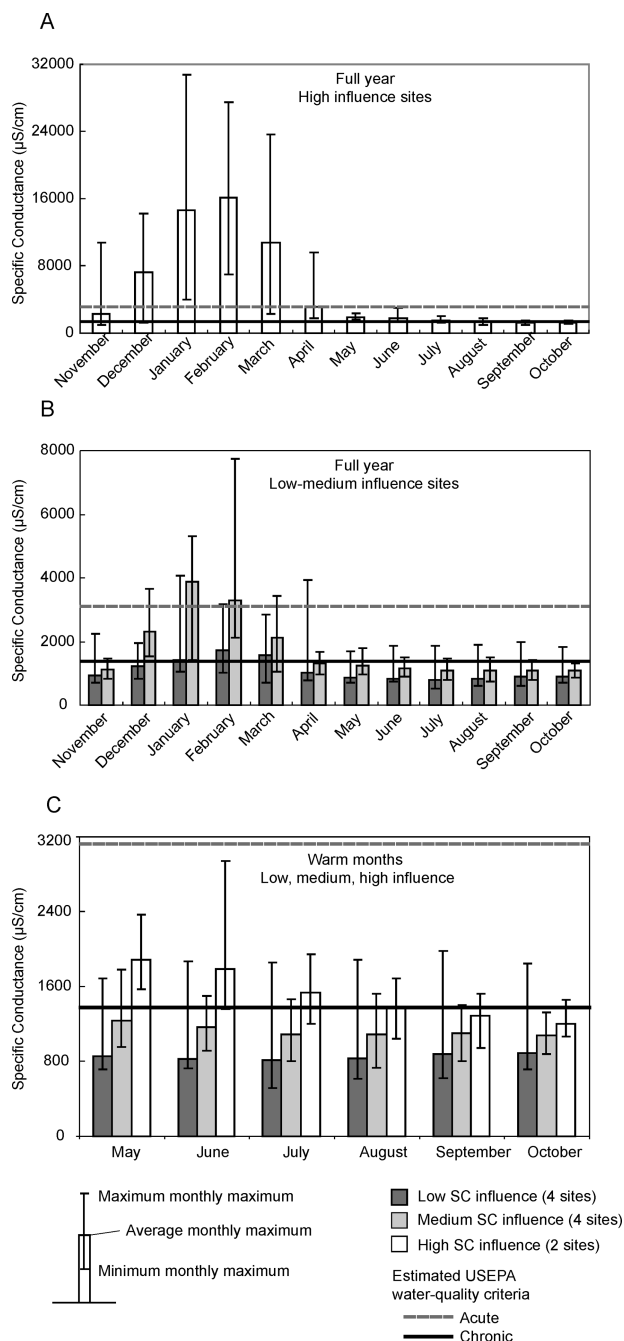


FIGURE 6. Monthly maximum specific conductance from continuous monitoring at 11 sites in Wisconsin over a gradient of urban influence.

toring sites with sample results that exceeded 860 mg/L during cold-weather months than during warm-weather months. Only two northern metropolitan areas had monitoring sites with concentrations greater than 860 mg/L during warm-weather months.

At monitoring locations in the four southern metropolitan areas, few samples exceeded the water-quality criteria concentrations and no common seasonal pattern was detected. Only 2 and 4% of monitoring locations had samples exceeding 230 mg/L during warm- and cold-weather months, respectively; samples from the southern sites did not exceed 860 mg/L. Several other southern metropolitan areas were analyzed but not included in this study because monitoring locations either had insufficient data or marine influence.

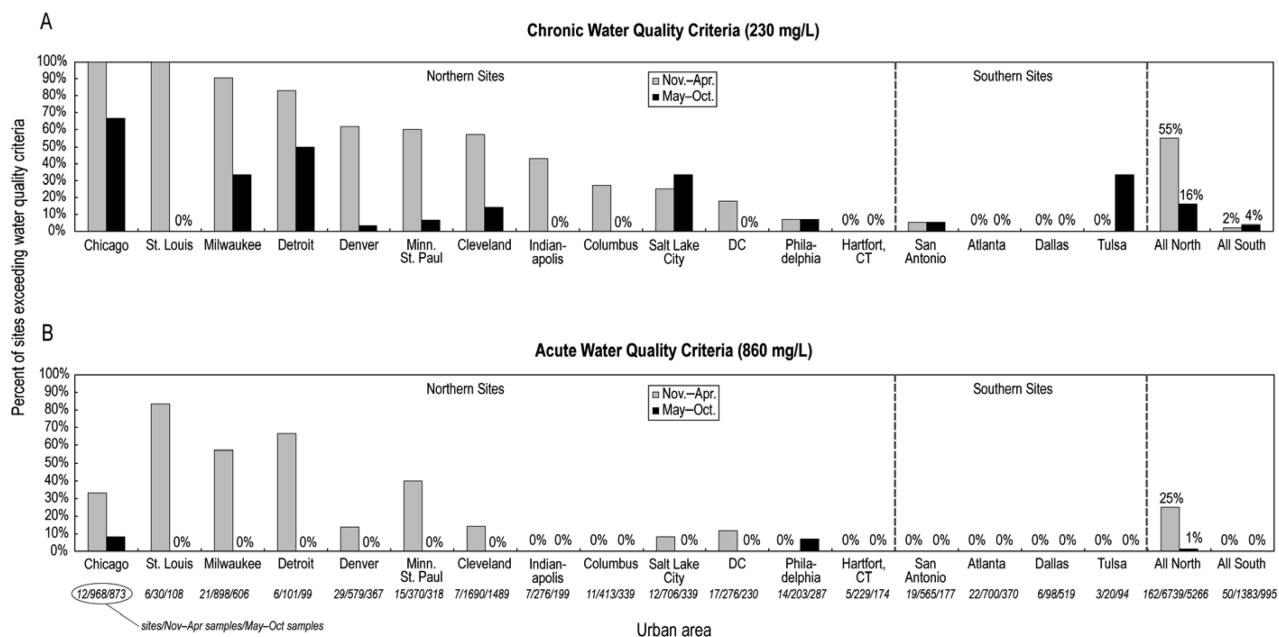


FIGURE 7. Comparison of chloride concentrations to chronic (A) and acute (B) USEPA water-quality criteria for warm-weather months and cold-weather months in streams from northern and southern urban areas. Bars indicate the percent of sites for each metropolitan area that had at least one sample result greater than the water-quality criteria.

Discussion

Detrimental impacts from road-salt runoff to surface water presented in this study were evident on local, regional, and national scales. The presented long- and short-term runoff sampling programs in Wisconsin demonstrate a substantial effect from road salt on streamwater quality and aquatic life. Bioassay results from runoff events confirm that the observed high concentrations of road salt caused acute and chronic toxicity to aquatic organisms. In addition, continuous specific conductance results indicate that elevated levels of road salt were present multiple times per year each year of monitoring. Populations of aquatic organisms in these streams and others with such road-salt influence are likely limited to salt-tolerant species. Effects on aquatic organisms have previously been demonstrated using a salt tolerance biotic index (chloride contamination index, CCI) in Toronto area streams (15). When compared to a review published by Environment Canada including laboratory and field assessments of road salt impact on aquatic species, concentrations observed in the present research indicate that detrimental effects would be present over numerous taxa in addition to the organisms used in this study (7). This review of road salt effects concluded that high concentrations may have immediate or long-term ecosystem population effects, and that lower levels of increased chloride concentrations may affect community structure, diversity, and productivity (7).

Results from continuous specific conductance monitoring in Lincoln and Wilson Park Creeks in Milwaukee indicate that elevated levels of chloride in these streams were common for extended periods of time, even through the summer months. These results have broad implications considering that traditional "chronic" toxicity assessments consider relatively short time periods of 7–14 days. Exposures over multiple months add a level of complexity to traditional toxicity assessments. Similar to results from Lincoln and Wilson Park Creeks in Milwaukee, study of groundwater influence on stream chemistry in Massachusetts confirmed that chloride from highway-deicing applications persisted throughout the year as a source of contamination in groundwater, interflow, and streamwater even during warm-weather months (16).

The analysis of historical chloride data from urban areas around the country indicated potential for considerable and widespread impact from road salt on surface water quality and aquatic life. Despite the limitation that sample results from these selected areas were from numerous studies not necessarily designed to capture periods of road-salt runoff, the influence of road salt was clear. Streams with urban influence throughout the country in areas where road salt is applied are at risk for substantial contamination and detrimental effect on aquatic life. Impacts of road salt are present in other geographic regions that commonly apply these deicers as well. For instance, in a study of urban streams in Finland, chloride concentrations during road-salt runoff periods varied over 9-fold within one day with NaCl concentrations from weekly composite samples observed as high as 567 mg/L (17). Another study of five catchments in Sweden, reported that road salt had a "profound effect on the soil- and stream-water chemistry". Salinity in these streams increased in direct proportion to accumulated application of road salt (18).

Research on the influence of urban land use on aquatic life in streams has identified a level of 7–12% impervious surface where decreases in biological integrity were observed (19–21). Much work investigating aquatic life degradation has focused on ambient water chemistry, habitat, and other physical, hydrologic, and hydraulic factors (22). The relation of chloride concentrations and specific conductance with urban land use shown in this study and a recent study of the northern United States (23) indicates that road-salt runoff and other anthropogenic uses of chloride are important factors in the biological integrity of urban streams in the northern United States. Although chloride sampling has been included in previous evaluations of urban streamwater quality (21), water-quality sampling did not specifically focus on periods of winter runoff and may not fully represent the severity of road-salt influence.

To better understand the relation between urban land use and stream biology, focused monitoring would be needed to characterize the range of chloride concentrations and duration of road-salt influence in streams during deicing periods. However, because of the episodic nature of road-salt runoff, the full range of in-stream road-salt influence is

difficult to characterize without use of continuous- and event-based monitoring focusing on deicing periods. A periodic or fixed-interval sampling plan will not fully characterize road-salt influence except by happenstance.

Environmental management or mitigation of this issue is complex. Solutions would require consideration of environmental, political, economic, and safety issues. Added to this complexity is the diversity of applicators in urban areas. City maintenance crews de-ice roadways, public parking lots, and sidewalks while a host of private applicators de-ice commercial, institutional, and industrial areas, and homeowners apply de-icers to residential driveways and sidewalks. Alternative chemicals each have unique environmental and/or economic impacts as well. For example, use of organic salts such as calcium magnesium acetate would reduce chloride loading, but may increase biochemical oxygen demand thereby increasing potential for oxygen depletion in receiving waters. Greater aquatic toxicity and water-quality impacts seem likely if increasing trends in road-salt usage and expanding urban development continue. Regardless of methods chosen, reducing the impact of road salt on the environment would take a substantial and sustained effort coupled with consideration of numerous interconnected factors.

Acknowledgments

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Supporting Information Available

Description of analytical methods and a graph of the relation between chloride and specific conductance. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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