WATER RESEARCH 46 (2012) 2749–2758

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Regional analysis of the effect of paved roads on sodium and chloride in lakes

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ARTICLE INFO

Article history:
Received 29 June 2011
Received in revised form
20 January 2012
Accepted 16 February 2012
Available online 25 February 2012

Keywords:
Adirondack Park
Road salt
Watersheds
Management
Shoreline
GIS

ABSTRACT

Salinization of surface water from sodium chloride (road salt) applied to paved roads is a widely recognized environmental concern in the northern hemisphere, yet practical information to improve winter road management to reduce the environmental impacts of this deicer is lacking. The purpose of our study was to provide such information by developing baseline concentrations for sodium and chloride for lakes in watersheds without paved roads, and then determining the relationship between these ions and density, type, and proximity of paved roads to shoreline. We used average summer (June–September) sodium and chloride data for 138 lakes combined in a watershed based analysis of paved road networks in the Adirondack Park of New York, U.S.A. The watersheds used in our study represented a broad range in paved road density and type, 56 of which had no paved roads. Median lake sodium and chloride concentrations in these 56 watersheds averaged 0.55 and 0.24 mg/L, respectively. In contrast, the median sodium and chloride concentrations for the 82 lakes in watersheds with paved roads were 3.60 and 7.22 mg/L, respectively. Paved road density (lane-km/km^2) was positively correlated with sodium and chloride concentrations, but only state roads were significantly correlated with sodium and chloride while local roads were not. State road density alone explained 84 percent of the variation in both ions. We also successfully modeled the relationship between road proximity to shoreline and sodium and chloride concentrations in lakes, which allowed us to identify sections of road that contributed more to explaining the variation in sodium and chloride in lakes. This model and our approach could be used as part of larger efforts to identify environmentally sensitive areas where alternative winter road management treatments should be applied.

1. Introduction

Sodium chloride (NaCl, commonly referred to as "road salt") is widely used in the northern hemisphere to maintain clear roads in the winter months (Albright, 2005; Löfgren, 2001; Rodrigues et al., 2010). Over 19 million metric tons of sodium chloride is applied to provincial and state highways annually in North America alone (NCHRP, 2007). Though alternative deicing treatments are available, due to the comparatively low cost and high availability of sodium chloride, it continues to be the most commonly applied chemical used to maintain clear roads and its use on US highways has increased steadily over the last 50 years (Jackson and Jobbagy, 2005).

A number of studies have reported increased sodium, chloride, and conductivity in surface and groundwater near salted roads, with these increases being attributed to road

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doi:10.1016/j.watres.2012.02.032

density, impervious surfaces, and road salt application rates (Kaushal et al., 2005; Löfgren, 2001; Mullaney et al., 2009; Rosfjord et al., 2007; Siver et al., 1996). Sodium chloride has variable impacts on aquatic environments which makes it difficult to generalize effects and establish water quality criteria for aquatic biota (Corsi et al., 2010). This said, recent authors have stated that the steady increase in sodium and chloride concentrations observed in surface waters should be a cause for concern and efforts should be made now to reduce the amount of salt applied to roads (Jackson and Jobbagy, 2005; Kaushal et al., 2005). This is particularly important when considering recently published evidence for sodium and chloride retention in watersheds (Kelly et al., 2008; Kincaid and Findlay, 2009) which suggests that concentrations of these ions in surface water may remain high even if application rates are reduced.

The New York State Department of Transportation (NYSDOT) is the largest user of sodium chloride for winter road management in North America (NCHRP, 2007). The NYSDOT applies 680,000–860,000 metric tons of sodium chloride to state roads every winter with application rates ranging from 10 to 14 metric tons per lane-km per year (Albright, 2005; Kelting and Laxson, 2010). The northern region of New York State is dominated by the 2.4 million hectare Adirondack Park (AP), a mosaic of public and privately owned lands created in 1892 to protect water and timber resources (NYSAPA, 2011). The AP contains several thousand lakes and ponds and over 48,000 km of rivers and streams. The AP is intersected by 4530 lane-km of NYSDOT roads that are consistently treated with sodium chloride and 12,380 lane-km of roads maintained by local municipalities that receive a variety of winter treatments (snow plow only, sand, sand plus salt, and salt).

The AP has comparatively low paved road densities and sodium and chloride concentrations in Adirondack lakes are on average much lower than in other northeastern lakes (Rosfjord et al., 2007). However, given that the AP was created in part to protect water resources and the fact that salinization will steadily increase if current practices continue, we conducted a study with the overall goal of providing science-based information to aid in improving winter road management practices to reduce the loading of sodium and chloride to lakes in the Adirondack Park. Though our study focused on the AP, we believe that our research approach is applicable to other lake regions in the northern hemisphere. The specific objectives of our study were to (1) provide baseline estimates of sodium and chloride in lakes in watersheds with and without paved roads, (2) determine the effects of paved road type and density on sodium and chloride concentrations in lakes, and (3) determine the effect of paved road proximity to shoreline on sodium and chloride concentrations in lakes.

2. Materials and methods

The bedrock geology in the central part of the AP is dominated by metaplutonic rocks such as granite gneiss, metanorthosite, and metagabbro with metasedimentary rocks forming the bedrock on the periphery of the AP (Isachsen et al., 2000). This bedrock is overlain by glacial deposits of till and outwash that form the parent materials for the coarse textured soils that dominate the region (Isachsen et al., 2000). These soils support a largely forested region that receives about 100 cm of precipitation per year (Sullivan et al., 2006). The forests cover over 95% of the land area and consist of hardwood, conifer, and mixed forests. The land area of the AP is 43% public lands that are protected and cannot be developed or harvested and 57% private lands that are managed largely for timber (NYSAPA, 2011). The AP has a very low population density with only about 130,000 year-round residents living on its 2.4 million hectares of land.

We utilized Geographic Information Systems (GIS) and water quality data to investigate the effects of paved roads on sodium and chloride concentrations in 138 lakes throughout the Adirondack Park (Fig. 1). Lake area ranged from 0.01 to 83.94 km^2 with a median area of 0.66 km^2. Watershed characteristics for our study region are summarized in Table 1. The watershed areas for each individual lake were digitized on top of a USGS 1:100 k topographical map using ArcGIS software (ESRI, Redlands, CA). Land cover, road network, and geologic features were clipped to each of the watershed boundaries. The percentage of each land cover class and surface geology type was calculated by dividing the total area of the cover class by the total watershed area. Road densities (lane-km/km^2) were calculated on a lane-km basis by multiplying the total length of each road type by the number of lanes (state = 2, county = 1.5, town = 1.5, local = 1.5, interstate = 4, US highway = 2) and dividing this value by the watershed areas. All GIS layers were obtained from the NYS Adirondack Park Agency GIS database (NYSAPA, 2011).

2.1. *Lake chemistry data sources*

The sodium and chloride dataset for the 138 lakes was compiled from two sources. Data for 84 of the lakes were from the Adirondack Watershed Institute (AWI) lake monitoring program that represents a large range in paved road density (some also without roads) and type. One surface water sample was collected from each of these lakes once a month by boat (May–October, 2010). The samples were taken at the deepest part of each lake by deploying a Kemmerer water bottle to 1.5 m depth. Samples were analyzed for sodium by inductively coupled plasma optical emission spectroscopy (Varian Instruments, 720-ES, Walnut Creek, CA) and chloride by suppressed ion chromatography (Lachat Instruments, QC8500, Loveland, CO). Data for the 54 remaining lakes were obtained from the Adirondack Lake Survey Corporation (ALSC), whose long term dataset includes analysis of sodium and chloride. ALSC data were collected from the surface water of lakes in least impacted watersheds (though some with roads) on a monthly basis and analyzed for sodium by atomic absorption spectroscopy and chloride by ion chromatography. For this study, the dataset was compiled and analyzed as average summer concentration of sodium and chloride (June–September) for each lake using 2010 data from the AWI and 2009 data from the ALSC (the 2010 ALSC dataset was not yet available at the time of this analysis). In addition, the complete time series of ALSC average summer concentration of sodium and chloride from 1992 through 2009 was used to examine the variation in these ions through time: this analysis was done in part to check the validity of comparing

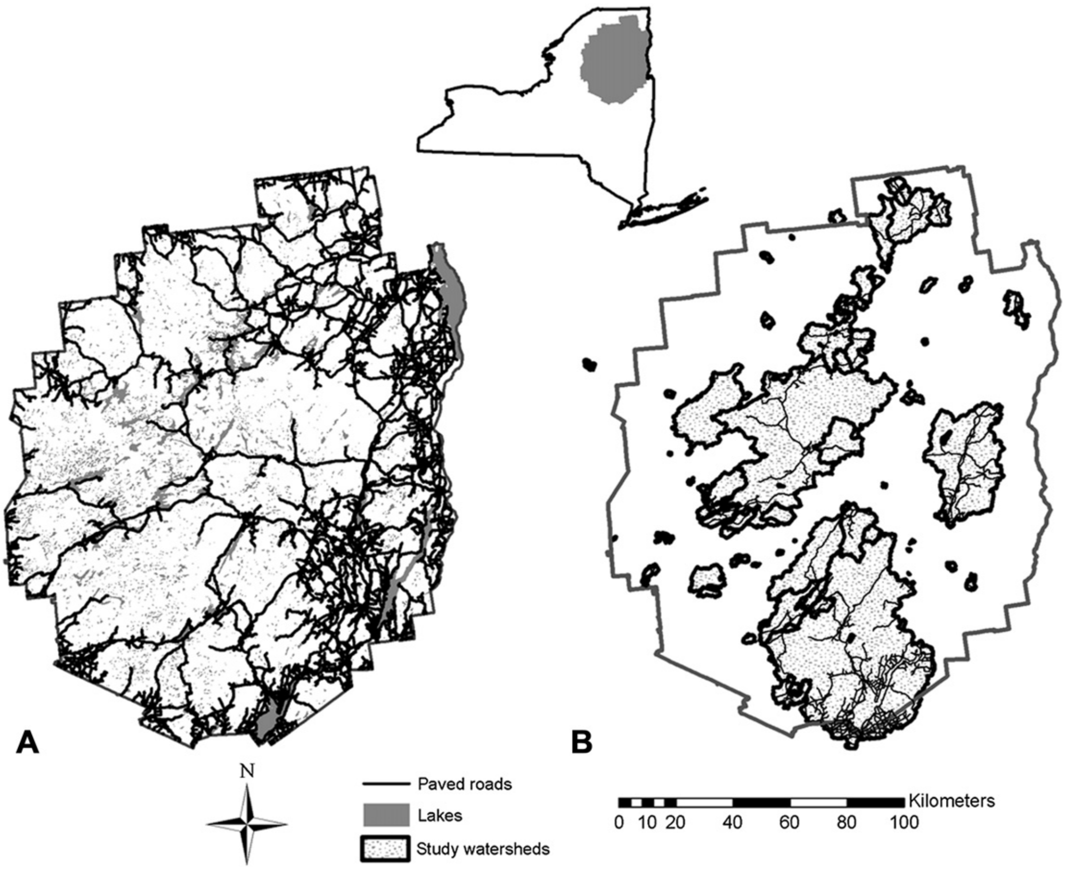

Fig. 1 – Paved road network and lakes (A) and locations of 138 study watersheds with associated paved road networks (B) in the Adirondack Park of New York, U.S.A.

datasets collected from different years but also to understand the historical trends in sodium and chloride concentrations under least impacted watershed conditions.

2.2. *Effect of road presence*

To examine the effect of presence of paved roads on sodium and chloride concentrations in our study lakes the data were split into two groups, watersheds with paved roads ($n = 82$) and those without paved roads ($n = 56$). General watershed characteristic in these two groups were similar, only differing in terms of presence of paved roads, watershed size, and area of surface water, though the ratios of watershed area to surface water area were very similar (Table 1). The sodium and chloride datasets did not follow the normal distribution, so differences in sodium and chloride concentrations between these two groups were tested for using the two-sample Wilcoxon rank sum test. In addition, cumulative frequency distributions were constructed to examine the distribution patterns between the two groups.

2.3. *Effect of road density and type*

The effects of road density and type on sodium and chloride concentrations were examined using only the 82 lakes in watersheds with paved roads. Simple linear regression was used to examine the effect of total road density on sodium and chloride concentrations and multilinear regression was used to examine the influence of road type for the six types of paved roads that exist within the Adirondack Park: county roads, interstate highways, local roads, state roads, town roads, and US routes. As multicollinearity is possible in multiple regression, co-correlations among road types were first evaluated using simple linear correlations, for which no statistically significant correlations among the road types were found (maximum $r = 0.3$ and all p-values were greater than 0.2).

2.4. *Influence of road proximity to shoreline*

We used GIS to capture the length of paved roads that existed in a range of proximities to our 82 study lakes. Our approach was to add a series of buffers of increasing width to each of the shorelines in the watersheds. Buffer widths of 10, 20, 40, 80, 160, 320, 640, and 1280 m were added to each water body, and then clipped to the watersheds to prevent inter-watershed overlap. The road network shapefile was clipped to each of the eight buffer widths, and then road density was calculated for each buffer width. The relationship between road density in each of the buffer widths and sodium and chloride concentration in the lakes was analyzed using simple linear regression. A nonlinear equation was fit to the regression results to model the relationship between the percent

Table 1 – General characteristics for watersheds without paved roads ($n = 56$) and with paved roads ($n = 82$) used in an analysis of the effects of paved road type and density on sodium and chloride concentrations in lakes located in the Adirondack Park of New York, U.S.A.

Characteristic	1st quartile		Median		3rd quartile	
	No roads	Roads	No roads	Roads	No roads	Roads
Watershed area (km^2)	1.2	5.3	2.6	17.1	6.7	96.1
Surface water (km^2)	0.1	0.9	0.2	2.0	0.6	8.9
WA/SW ratio[a]	6.0	5.6	10.5	8.8	16.1	14.1
Land cover						
Developed (%)	0	0	0	0	0	1
Deciduous (%)	51	63	69	73	81	79
Conifer (%)	4	6	8	11	19	18
Mixed (%)	5	7	10	9	16	13
Forested (%)	89	91	97	94	99	98
Agriculture (%)	0	0	0	0	0	1
Wetlands (%)	0	0	1	4	8	7
Surface geology						
Sand and gravel (%)	0	0	0	13	11	31
Glacial till (%)	57	48	80	68	94	78
Bedrock (%)	0	1	6	8	19	13

a WA/SA ratio = watershed area divided by surface water area.

variation in sodium and chloride explained by road density and the corresponding buffer width from the lake shoreline.

3. Results

3.1. *Historical sodium and chloride concentrations*

Analysis of eighteen years of lake monitoring data from the ALSC program dataset showed that median summer sodium and chloride concentrations have remained relatively stable over time although a slight downward trend was observed for chloride (Fig. 2). The maximum year to year difference in median sodium and chloride concentrations were 0.1 and 0.05 mg/L, respectively. Over the entire sampling period median sodium was within a range of 0.35–0.85 mg/L and median chloride was within a range of 0.20–0.35 mg/L. The median sodium concentration across all years was 0.54 mg/L and the median chloride concentration across all years was 0.27 mg/L.

3.2. *Effect of road presence*

With median concentrations of 0.55 mg/L for sodium and 0.24 mg/L for chloride in lakes in watersheds without paved roads, these values were nearly identical to their respective historical median concentrations in the ALSC dataset. The median sodium concentration in lakes in watersheds without paved roads was 5.5 times lower than the median sodium concentration of 3.60 mg/L measured in lakes in watersheds with paved roads ($p < 0.001$). The median chloride concentration in lakes in watersheds without paved roads was 29

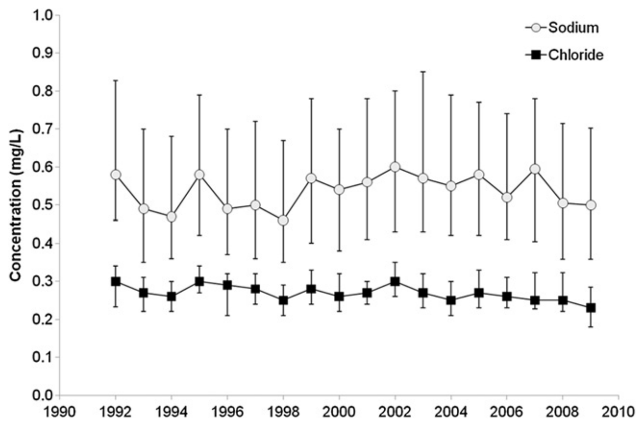

Fig. 2 – Yearly median summer (June–September) sodium (open circles) and chloride (solid squares) concentrations (mg/L) for lakes in the ALSC program from 1992 through 2009. Vertical bars represent first and third quartiles ($n = 54$ each year).

times lower than the median chloride concentration of 7.22 mg/L measured in lakes in watersheds with paved roads ($p < 0.001$).

Sodium concentrations in lakes in watersheds without paved roads ranged from 0.1 to 3.7 mg/L with 98% of lakes having concentrations less than 1.5 mg/L (Fig. 3A). Sodium concentrations in lakes in watersheds with paved roads ranged from 0.1 to 32.8 mg/L with 70% of lakes having concentrations greater than 1.5 mg/L. Chloride concentrations in lakes in watersheds without paved roads ranged from 0.1 to 5.3 mg/L with 90% of lakes having concentrations less than 2.5 mg/L (Fig. 3B). Chloride concentrations in lakes in watersheds with paved roads ranged from 0.1 to 58.4 mg/L with 80% of lakes having concentrations greater than 2.5 mg/L.

The molar equivalent concentration of sodium to chloride in lakes in watersheds without paved roads showed a weak positive correlation (slope = 2.5, slope $p = 0.001$, $r^2 = 0.26$), while the molar equivalent concentration of sodium to chloride in lakes in watersheds with paved roads showed a strong positive correlation and a significantly lower slope coefficient compared to without paved roads (slope = 0.85, slope $p < 0.001$, $r^2 = 0.96$). The 95% confidence interval on the slope coefficient for paved roads was 0.82–0.89.

3.3. *Effect of road density and type*

Sodium and chloride concentrations were both positively correlated with total paved road density in the watersheds, with road density explaining 22% of the variation in sodium and 26% of the variation in chloride (Fig. 4). Though both linear regressions were statistically significant ($p < 0.001$ for all model coefficients), the relationships were weak and there was a lot of residual error around the fitted lines, particularly at higher road density.

When paved road density was analyzed by road type in a multilinear regression model, the variation in both sodium and chloride explained by paved road density increased substantially, with road density by type explaining 85% of the

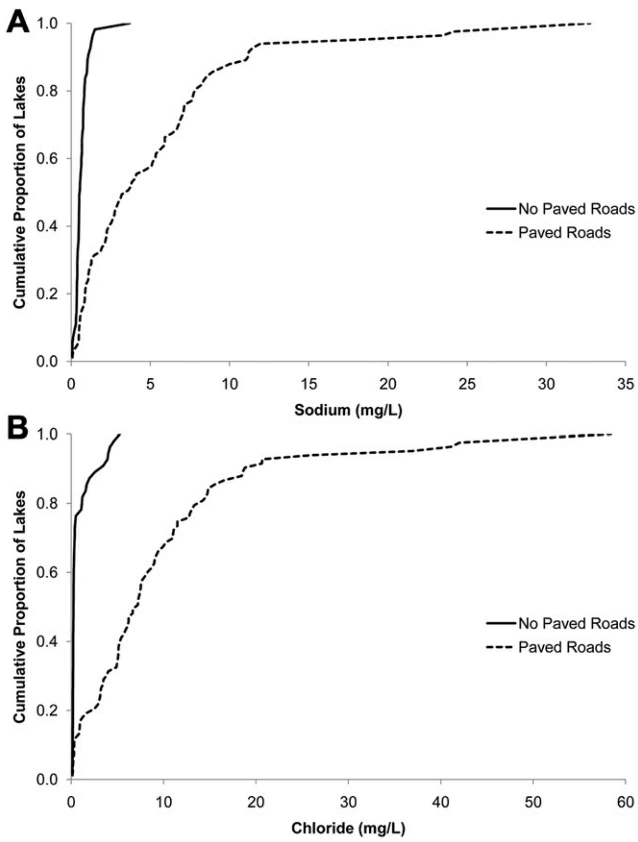

Fig. 3 – Cumulative proportions of sodium (A) and chloride (B) concentrations in lakes in watersheds with no paved roads (solid lines) and lakes in watersheds with paved roads (dashed lines) for 138 watersheds located in the Adirondack Park of New York, U.S.A.

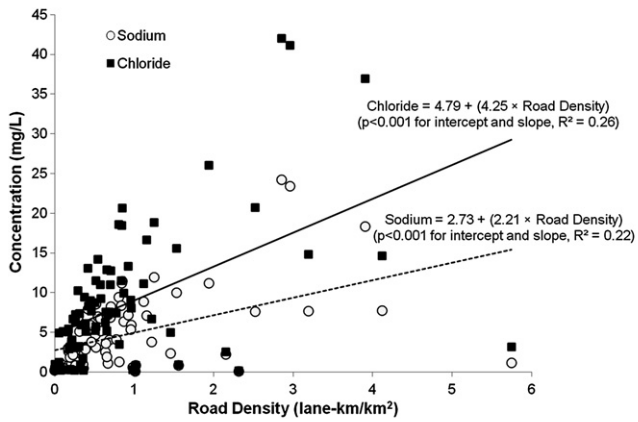

Fig. 4 – Relationship between sodium (open circles) and chloride (solid squares) concentrations (mg/L) in lakes and total paved road density (lane-km/km^2) for 82 watersheds located in the Adirondack Park of New York, U.S.A. The dashed and solid lines represent simple linear regression fits through the sodium and chloride data, respectively.

variation in sodium and 87% of the variation in chloride (Table 2). In the sodium model interstate, state, and US routes were significant variables ($p < 0.05$) in the regression model, with these three road types explaining a combined total of 82% of the variation in sodium. In the chloride model local roads and interstate, state, and US routes were significant variables in the regression model, though the standard error for the slope coefficient of local roads was very high and local roads explained less than 1% of the variation in chloride. Interstate, state, and US routes explained a combined total of 82% of the variation in chloride. Thus, for both sodium and chloride local roads (county, local, and town) were not significant contributors to explaining the concentration of these ions in lakes, while state roads (interstate state, and US routes) explained almost all of the variation for both ions.

When the relationship between sodium and chloride concentrations in lakes and road density was reanalyzed using only paved state road density (=interstate + state + US routes), state road density explained 84% of the variation in both sodium and chloride (Fig. 5). The strength of this relationship was improved over that shown for total road density, with clear positive correlations shown between concentration and state road density for both ions. The slope coefficients show that the chloride concentration increases at a higher rate than sodium for each lane-km per km^2 of state roads in the watershed. The ratio of the sodium to chloride slopes is 0.56, which is equivalent to a molar ratio of 0.86, the same molar ratio determined for the correlation between sodium and chloride for lakes in watersheds with paved roads.

3.4. *Influence of road proximity to shoreline*

Road density in all lakeshore buffer widths contributed to explaining some portion of the variation in sodium and chloride, with the percent variation explained by road density increasing with buffer width in a nonlinear manner (Table 3). Road density in the 10 m buffer explained 36 and 30% of the variation in sodium and chloride, respectively. While at 320 m, 78 and 76% of the variation in sodium and chloride was explained by road density in this buffer. Roads closer to the shoreline contribute disproportionally more to explaining the variation in sodium and chloride than roads further away. For example, 22% of the total road length lies within the 160 m buffer, yet these roads explain 64% of the variation in both ions, while doubling the amount of road length in the 320 m buffer only explains an additional 14 and 12% of the variation in sodium and chloride, respectively. All of the variation in sodium explainable by road density (84%) is captured within the 640 m buffer, which constitutes 66% of the total road length in the study watersheds. We were able to model the variation in sodium and chloride as a function of road proximity to shoreline using a nonlinear asymptotic function, with buffer width explaining 87% of the variation (Fig. 6).

4. Discussion

The lake sodium and chloride concentrations in the undeveloped watersheds in our study represent reference conditions for the Adirondack Park and these values agree closely with data reported for lakes in other undeveloped watersheds

Table 2 – Summary of regression and analysis of variance results of a multilinear regression analysis of sodium and chloride concentrations (mg/L) in lakes versus the density (lane-km/km^2) of county, local, town, interstate, state, and US roads in 82 watersheds with paved roads located in the Adirondack Park of New York, U.S.A.

Variable	Regression			Analysis of variance		
	Coefficient	Standard error	*p*-Value	d.f.	Sums of squares	% contribution
Sodium						
Constant	1.805	0.288	<0.001			
County Route	0.084	0.540	0.877	1	19.16	1.1
Local Road	28.950	24.100	0.234	1	0.82	0.0
Town Road	−0.110	0.203	0.588	1	37.02	2.1
Interstate	5.073	0.735	<0.001	1	399.65	22.3
State Route	5.536	0.325	<0.001	1	985.71	54.9
US Route	8.764	1.766	<0.001	1	88.01	4.9
Regression				6	1530.37	85.3
Residual Error				75	264.57	
Total				81	1794.94	
Chloride						
Constant	3.108	0.482	<0.001			
County Route	0.145	0.902	0.873	1	46.41	0.8
Local Road	86.340	40.280	0.035	1	16.26	0.3
Town Road	0.097	0.339	0.777	1	209.42	3.6
Interstate	7.584	1.229	<0.001	1	1201.07	20.9
State Route	9.827	0.542	<0.001	1	3037.28	52.9
US Route	20.721	2.952	<0.001	1	491.99	8.6
Regression				6	5002.43	87.1
Residual Error				75	738.82	
Total				81	5741.25	

in the region. D'Arcy and Carignan (1997) reported a median sodium concentration of 0.51 mg/L and a range of 0.27–0.92 mg/L for 30 lakes in undeveloped watersheds with similar granitic geology and glacial till and outwash soils located on the Canadian Shield in Quebec, Canada. Our sodium and chloride concentrations also overlap the range of 0.63–1.49 mg/L for sodium and 0.22–0.54 mg/L for chloride reported in the USEPA Eastern Lakes Survey for 10 lakes in

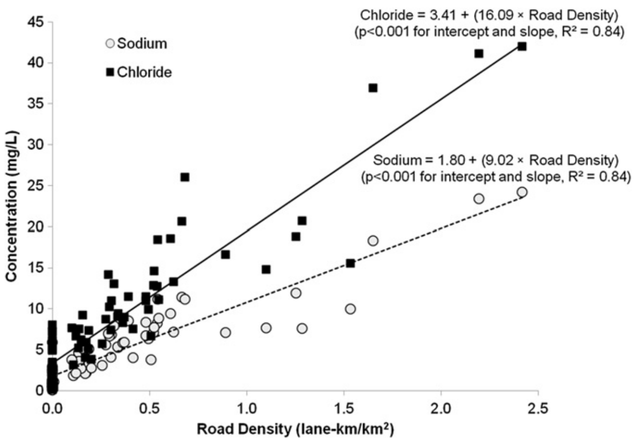

Fig. 5 – Relationship between sodium (open circles) and chloride (solid squares) concentrations (mg/L) in lakes and paved state road density (lane-km/km^2) for 82 watersheds located in the Adirondack Park of New York, U.S.A. State roads are the sum of road lane kilometers of state, interstate, and US highways in each watershed. The dashed and solid lines represent simple linear regression fits through the sodium and chloride data, respectively.

undeveloped watersheds located in the interior of Maine with similar geology and soils as in our study (Landers et al., 1988). The low concentrations of sodium and chloride reflect the low annual inputs of these ions from the combination of atmospheric deposition and rock weathering. Atmospheric deposition contributes about 0.8 kg/ha/yr of sodium and 1.6 kg/ha/yr of chloride to watersheds in the Adirondacks (Johnson and Lindberg, 1992), which are comparable to atmospheric deposition rates on the Canadian Shield (Ouimet and Duchesne, 2005) and the interior of Maine (NADP, 2009). With respect to rock weathering sources, studies on similar granitic parent materials on the Canadian Shield and in the northeastern US report rock weathering releases from 2.6 to 6.0 kg/ha/yr of sodium (Lovett et al., 2005; Ouimet and Duchesne, 2005), while the amount of chloride released via rock weathering from these same materials has been estimated at less than 0.12 kg/ha/yr (Lovett et al., 2005) with this input generally considered insignificant to the overall chloride budget in watersheds with similar granitic parent materials (Lovett et al., 2005; Nimiroski and Waldron, 2002; Rosfjord et al., 2007). The higher concentration of sodium relative to chloride in our lakes in watersheds without paved roads most likely reflects the higher input of sodium from atmospheric deposition plus rock weathering.

The high correlation between road density and sodium and chloride concentrations points clearly to road salt as the primary source of salt loadings to the lakes in our study, but there are other potential anthropogenic sources of sodium and chloride that may have contributed to the higher concentrations in the lakes in watersheds with paved roads, as homes are also present in these watersheds. Two other potentially significant sources of sodium and chloride are

Table 3 – Percent of total variation in sodium and chloride in lakes explained by paved state road density within buffers created from the shoreline of increasing width in 82 watersheds with paved roads located in the Adirondack Park of New York, U.S.A. State road length and percent of total state road length associated with each buffer width are also shown. Variation explained was obtained via simple linear regression, and all regression *p*-values were less than 0.001 (*n* = 82 per regression).

Buffer width (m)	Sodium (%)	Chloride (%)	Road length (lane-km)	% of total road length
10	36	30	13	1
20	34	29	25	2
40	39	36	69	4
80	48	42	170	10
160	64	64	361	22
320	78	76	673	41
640	84	82	1065	66
1280	84	82	1489	92
All roads	84	84	1624	100

sewage disposal systems and water softeners (Rosfjord et al., 2007). Kelly et al. (2008) estimated that sewage disposal and water softeners accounted for 4 and 3%, respectively, of the total sodium chloride load while road salt accounted for 91% of the total sodium chloride load in a rural watershed in southeastern NY. Another study in a more developed watershed in Connecticut but also on similar geology and soils as in our study estimated the load from individual sewage disposal at 15% for sodium and about 1% for chloride, with road salting estimated to contribute 66 and 90% of the sodium and chloride load, respectively (Nimiroski and Waldron, 2002). Given that our watersheds are greater than 90% forested and that our population density is much lower than in these other studies (the AP has a population density of 0.05 persons/ha, while the population density in the southeastern NY study was 0.4 persons/ha), we believe that the contribution of sewage disposal and water softeners to the salt load in our study lakes to be negligible relative to road salt.

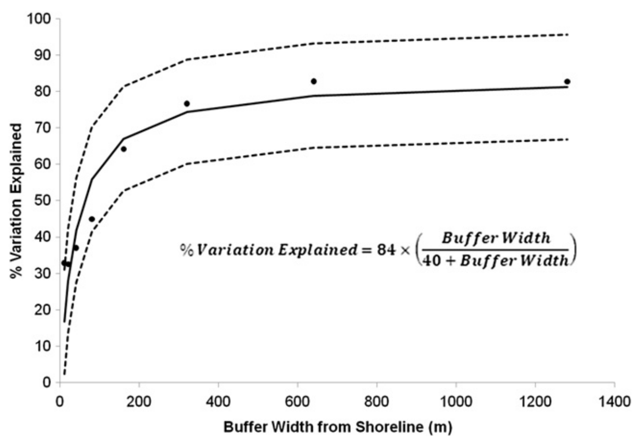

Fig. 6 – Percent of total variation in sodium and chloride explained by the density of paved state roads within increasing buffer widths from the shoreline for 82 watersheds located in the Adirondack Park of New York, U.S.A. The points represent the mean variation of sodium plus chloride explained by paved state road density within each buffer width, the solid line represents a fit of a nonlinear asymptotic function through the data, and the dashed lines represent 95% confidence intervals on the nonlinear fit ($n = 8$, $r^2 = 0.87$).

Though sodium and chloride concentrations in our lakes in watersheds with paved roads were significantly higher than the least impacted condition, interpreting this difference from a lake ecosystem health perspective is challenging. For example, acute toxic response from select invertebrates and fish typically occurs when chloride concentrations are well above 1000 mg/L (Corsi et al., 2010; reviewed by Environment Canada, 2001), while chronic effects can be detected at an order of magnitude lower (Corsi et al., 2010; Elphick et al., 2011; USEPA, 1988). However, road salt may impact lake ecosystems in other ways at concentrations below those documented to be toxic to aquatic organisms. Increased salt concentrations have been linked to colonization of invasive species (Richburg et al., 2001), mobilization of heavy metals from roadside soils (Amrhein and Strong, 1990), and prolonged stratification of the water column (Bubeck et al., 1971). Thus, as suggested by Kelly et al. (2008), there may be significant ecological effects at concentrations lower than those published for aquatic biota.

Although the concentrations detected in our study are still low, it is important to recognize that these increases have occurred in a short period of time in watersheds that are primarily rural. The use of large amounts of road salt in the Adirondack region was initiated in the late 1970s as part of a clear roads policy enacted prior to the 1980 Olympic Winter Games in Lake Placid, New York, thus the bulk of the changes to sodium and chloride concentrations have probably occurred within the last three decades. Other researchers in the U.S and Europe have observed increased salinization of lakes in similarly short time frames (Müller and Gächter, 2012; Rosfjord et al., 2007). For example, Siver et al. (1996) demonstrated a significant increase in both sodium and chloride in a 26 year time period in a study of 40 lakes in Connecticut. Some of the lakes influenced by an increase in residential and urban development saw increases as great as 2.5 times for sodium and 2.9 times for chloride. These increases are substantially lower than the 5.5 fold increase for sodium and 29 fold increase for chloride shown in our study, which would have occurred in about the same timeframe.

We believe there are two major explanations for the more rapid increase in sodium and chloride in lakes in watersheds with paved roads in the AP compared to other studies. First, compared to other states in the northeastern US, the NYSDOT applies the largest amounts of sodium chloride to state roads annually (Kelting and Laxson, 2010), thus the total load of sodium chloride applied to state roads in the AP may be higher than in other regions. Second, the AP is dominated by coarse textured glacial till and outwash soils of granitic origin (Sullivan et al., 2006; and Table 1), these soils have high infiltration and percolation rates, thus sodium and chloride dissolved in the soil pore water can leach rapidly with little opportunity for retention in the soil matrix. Though a portion of infiltrated sodium would be retained via cation exchange, evidence for which is provided by the 0.85 M ratio of sodium to chloride that indicates preferential sodium retention (Jackson and Jobbagy, 2005), much of the sodium would leach to surface or groundwater because of the coarse textured soils. Chloride is retained in soils via chlorination of organic matter, anion exchange, plant uptake, and entrainment in soil micropores (Bastviken et al., 2006; Kelly et al., 2008; Lovett et al., 2005; Svensson et al., 2007). The capacity of these mechanisms to retain chloride in AP soils is most likely lower than in other regions that have finer textured soils, which would generally have more organic matter and micropores (Plante et al., 2004; Brady and Weil, 2010) and thus greater soil retention capacity for chloride. Thus, the combination of higher loads and lower capacity for soil retention probably explains the greater salinization rate in AP lakes.

The impact of winter road maintenance on the chemistry of receiving waters has been well covered in the literature, yet in order to mitigate these impacts it is essential to have an understanding of the relationship between road density and surface water salinization. Although numerous studies have demonstrated the relationship between road salt use and increase salinization of surface waters, few studies have carefully examined the impact of varying road densities, and no studies have looked specifically at their impact on lake chemistry, only stream chemistry. Most studies only relate salinization to roads in a general sense by examining percent impervious surfaces (Kaushal et al., 2005), road density in one watershed (Kelly et al., 2008), or a small number of watersheds (Gardner and Royer, 2010). One exception is Daley et al. (2009) who determined that total road pavement density in 44 watersheds explained 78% and 75% of the variability in sodium and chloride concentrations in stream water in New Hampshire. In contrast, our analysis of lake chemistry of 82 watersheds revealed that a weak relationship exists with total road density but a much stronger relationship exists when road type is considered. The combination of Interstate highways, US routes, and State routes explain 84% of the variation in both sodium and chloride concentrations in lakes. The weak relationship we found between total road density and sodium and chloride concentration is likely due to highly variable winter road maintenance procedures across state, local, and county roads. For example, Interstate highways, US routes, and State routes are all managed by the New York State Department of Transportation. The NYSDOT relies heavily on road salt to prevent snow and ice from bonding to the pavement (anti-icing) and to loosen snow and ice that has already bonded (deicing) (NYSDOT, 2006). Because local, town and county roads in the AP receive less road salt and are typically managed with a combination of plowing, sanding, and salting, their contribution to explaining the total variation in sodium and chloride in lakes is significantly less than state roads. Specific data on how local municipalities treat non-state roads would improve our understanding of the effect of paved roads on sodium and chloride concentrations in lakes.

As recommended by Jackson and Jobbagy (2005) managers need tools to help identify sensitive areas to receive alternative treatments in order to reduce salt loads to surface waters. In New York State, the NYSDOT has begun an environmental initiative that includes protection and restoration of water quality as a principal goal (NYSDOT, 2011). Attaining this goal first requires identifying sensitive areas along NYSDOT roads within the Adirondack Park. Our model could be used as part of the NYSDOT planning process in a GIS framework to help identify sections of road that contribute more to sodium and chloride concentrations in lakes based on road proximity to shoreline.

For illustration purposes only, we applied our model to the entire 4530 lane-km state road network in the AP (see Fig. 1A), identifying all road segments that were within 80 m of lakeshore. There are 320 lane-km of state roads within 80 m of lakeshore within the AP, which represents only 7% of total state roads. Assuming that our model represents the AP as a whole, and we believe that it does given the spatial extent and range of watershed characteristics captured in our dataset, this 7% or 320 lane-km of state roads located within 80 m of shoreline would account for 45% of the variation in sodium and chloride (see Fig. 6).

An assumption we make is that targeting road segments within buffers for alternative treatment will reduce sodium and chloride loads and ultimately lake concentrations. We don't know the rate at which lakes will respond in terms of observing reduced concentrations of sodium and chloride; this would depend on the treatments and how long it takes for accumulated salt to be released from the watersheds. No reductions in stream sodium concentrations were observed in a study on similar geology and soils that monitored concentrations for 10 years after reducing the road salt application rate by 60% (Nimiroski and Waldron, 2002). These authors suggested that the stream sodium concentration was being maintained by mobilization of accumulated exchangeable soil sodium by the calcium in their alternative deicing treatment of calcium chloride. A significant portion of the chloride in road salt accumulates in groundwater and this contaminated groundwater contributes a significant load of chloride to surface waters (Kelly et al., 2008), which may maintain a high chloride concentration even after the road salt application rate is reduced. Thus, though the concentrations increased rapidly with the onset of road salting in our study, given that retention and accumulation have occurred, the reverse trend would likely be slower. But, stabilization at current concentrations may be an acceptable and reasonably achievable goal, and we believe that our approach of linking regional monitoring data to road networks would be applicable to other regions and countries interested in identifying more sensitive areas to achieve similar goals.

5. Conclusions

- Lakes in least impacted watersheds without paved roads in the AP have very low sodium and chloride concentrations, with the majority having concentrations less than 1 mg/L for both ions.
- Compared to concentrations in least impacted watersheds, road salting has increased sodium and chloride concentrations dramatically in watersheds with paved roads.
- Roads maintained in winter following NYSDOT deicing protocols are the greatest contributors to salinization of lakes in the Adirondack Park.
- The contribution of state roads to lake sodium and chloride concentrations can be modeled as a function of road

proximity to shoreline, which provides a useful tool for identifying areas to treat differently to reduce environmental impacts.

Acknowledgments

The impetus for this work came from AdkAction.org who first asked us the question about road salt and lakes. We are particularly grateful to the Adirondack Lakes Survey Corporation for the use of their valuable long term dataset. We would also like to thank our volunteer water quality monitors and our partner organization in the Adirondack Lakes Assessment Program, Protect the Adirondacks. We thank Mike DeAngelo for coordinating our water monitoring program. Data were also used from AWI monitoring projects sponsored by the Hamilton County Soil and Water Conservation District, Blue Mountain Lake WaterWatch, Great Sacandaga Lake Advisory Council, Litchfield Park, Rainbow Lake Association, Simon Pond, and the Upper Saranac Lake Foundation. A special thanks to the anonymous reviewers, whose comments and suggestions greatly improved the quality of this manuscript. We also acknowledge the National Science Foundation for awarding us a grant to purchase the laboratory instruments used in this study (award number 0722927).

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