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Abstract

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Road salt (NaCl) application around Lake George, New York, resulted in nearly tripling in-lake salt concentration between 1980 and 2009. Salt concentrations measured in 8 streams between 2007 and 2009 ranging in development from pristine to moderate resulted in 4 significantly different groups based on chloride concentrations. Chloride concentrations were significantly correlated to the amount of roadway surface within each sub-watershed, with chloride concentrations in the most impacted stream approaching 200 mg/L. In the more impacted streams, chloride concentrations were significantly and inversely correlated to discharge rate. The high road density around the more developed south end accounted for ∼30% of the estimated road salt application, which created a chloride gradient within the lake that decreased as water flowed north to the single outlet. The continual and disproportionate input of road salt near the southern end of the lake strengthened the gradient over time to create 4 significantly different regions within the lake. While the consistent lake-wide increase seems to be open-ended, a steady-state salt concentration within the lake may occur in the near future. Based on change of lake chloride mass and export from the lake (2007–2009), an average lake-wide steady-state chloride concentration of ∼17 mg/L is expected, with some variation anticipated due to interannual variation in precipitation and the salt gradient within the lake.

Key words: chloride, lake, Lake George, road salt, sodium, streams

Road salt (NaCl) became the standard winter deicing practice in the United States during the 1940s to ensure safe driving conditions during winter storms. Since then, road salt usage has increased more than 120-fold, with 6 states (New York, Ohio, Michigan, Illinois, Pennsylvania, and Wisconsin) accounting for three-quarters of the salt applied nationally (Jackson and Jobbágy 2005). Average annual application throughout the United States approximates 18.6 million tonnes, based on average use between 2006 and 2009 (Kostick 2011). New York State Department of Transportation (NYS DOT) uses ∼900,000 tonnes of road salt annually (Kelting and Laxson 2010), while the entire state (including counties and municipalities) applied more than 3 million tonnes in 2009 (Kostick 2011).

Based on available published data, the long-term application of road salt has resulted in contamination of surface and groundwater near high application and storage areas. In southern Ontario, ponds near 2-lane, ∼4-lane, and ≥6-lane roads had average (range) chloride concentrations of 95 mg/L (0–368), 124 mg/L (0–620), and 952 mg/L (49–3950), respectively (Environment Canada 2001). Similarly, in Baltimore, Maryland, peak stream chloride concentrations approached 5000 mg/L in urban areas (>40% impervious), up to 100 times greater than streams draining forest and agricultural watersheds (Kaushal et al. 2005). In 1970, Hutchinson documented average chloride concentrations of 171 mg/L in wells along major highways in Maine, with 20% exceeding the Environmental Protection Agency potable threshold of 250 mg/L. Typically, groundwater concentration is <10 mg/L (Jones and Jeffery 1992). Average chloride concentrations in springs around Southern Ontario measured 2.1 mg/L in conservation areas, ∼100 mg/L in rural areas, and 1092 mg/L in urban areas (Williams et al. 1997). Even higher concentrations have been documented in groundwater near salt storage facilities. Potable water wells near salt storage facilities in Canada measured average chloride concentrations of 722 mg/L, with a

maximum of 5050 mg/L, and average shallow (0.26–5.25 m) groundwater chloride concentrations measured 2600 mg/L, with a maximum of 24,000 mg/L (Environment Canada 2001). A chloride concentration of 24,000 mg/L has 25% more chloride than salt water, which has an average salinity of 35,000 mg/L, of which 55% is chloride, equaling a concentration of 19,250 mg/L (Evans and Frick 2001).

Road salt is a major concern in the Adirondack Park because the >16,000 km of roads within the park intersect areas of surface water and groundwater. Surface water and groundwater encompass 1300 km^2 (6%) and >4000 km^2 (18%) of the surface area within the Adirondack Park, respectively, making the likelihood of salt contamination a serious threat (Kelting and Laxson 2010). About one-half of the salt applied for deicing is removed from the catchment by surface run-off before the next winter (Church and Friesz 1993, Howard and Haynes 1993); the remainder presumably infiltrates, incorporating into both soil and groundwater. The location and extent of contamination depends on the quantity of road salt applied, soil composition (permeability, ion exchange capacity, organic matter), and precipitation.

Road salt, which is >95% sodium chloride, percolates through soil at different rates (Environment Canada 2001). Sodium can become incorporated into soil by displacing other cations (K^+, Mg^{+2}, Ca^{+2}, Al^{+3}, H^+), while chloride is more conserved because of its small size (MW 35.45) and negative charge (Environment Canada 2001, Ramakrishna and Viraraghavan 2005). While dissolved chloride was believed to have a migration rate identical to water, recent studies have shown that chloride may not be as conservative as once thought. Chloride can be sequestered in the watershed through geochemical sorption, vegetative uptake, vertical transport patterns, and microbial chlorination of soil organic matter (Bastviken et al. 2006). While the extent and relative contribution of these processes remain unclear, watershed sequestration of sodium exceeds that of chloride, resulting in chloride remaining a more effective tracer of road salt.

This study combines data from a 3-year stream assessment project and a 30-year lake-wide monitoring effort to identify streams substantially impacted by road salt application and its effect on lake water. Overall, development around the Lake George Basin has remained relatively limited (~10%), but more intense land use has occurred at the south end of the lake to accommodate a large tourism industry. The disproportional degree of development and road density suggests an equally disproportionate loading of salt to the south end of the lake. The objectives of this research were to identify the impact of deicing practices on chloride concen-

trations in streams and predict a possible steady-state lake concentration.

Study site

Lake George, a heavily forested watershed situated in the southeast portion of the Adirondack Park, New York, has ~10% of its watershed developed. The heaviest development is associated with tourism near Lake George Village at the southern end of the lake. The close proximity of New York State (NYS) Route 9N along the western shore has resulted in localized development in Bolton Landing ~15 km north of the southern end, Hague ~40 km north of the south end, and Ticonderoga near the outlet of the lake. The particularly steep slopes and more difficult access have deterred development on the eastern shore.

The lake consists of 2 discrete basins separated by a shallow sill in the Narrows, ~17 km north of the south end of the lake (Fig. 1). The south and north basins are significantly different chemically, with a phosphorus and chlorophyll gradient that decreases as water flows north to the single outlet, the LaChute River. The gradient is exacerbated by inputs from Northwest Bay and Shelving Rock brooks, which were relatively undeveloped and entered the lake directly south of the Narrows. These 2 sub-watersheds of Lake George accounted for ~20% of the total land catchment, diluting the more salt-laden lake water.

The lake surface area is 110 km^2 with an estimated volume of 2.11 km^3 (~550 billion US gallons), approximately equally distributed between the 2 basins. The total land catchment is 508 km^2 with ~two-thirds draining into the south basin. The lake is 51.5 km long with an average width of 2.15 km and a maximum depth of 60 m at an elevation of 97.5 m a.s.l. (Boylen and Kuliopulos 1981, Shuster 1994). The lake mean residence time is 6.8 years with an annual hydrologic budget of 57% stream discharge, 25% direct precipitation, and 18% groundwater discharged directly to the lake (Shuster et al. 1994). Groundwater is the dominant source of water input to the lake during May and June and the second greatest input from February to April, being surpassed only by stream discharge associated with snowmelt runoff. Groundwater inputs generally increase with precipitation events, except during winter and late summer. During winter, precipitation is stored in snowpack, and the lowest fluxes of the year occur in late summer when upland evapotranspiration depletes groundwater levels (Shuster 1994).

Lake George is a typical Adirondack dimictic lake in terms of temperature. Ice cover normally occurs in January, with water temperatures under the ice near 0 C and slightly warmer near the bottom. Ice-out typically occurs in April,
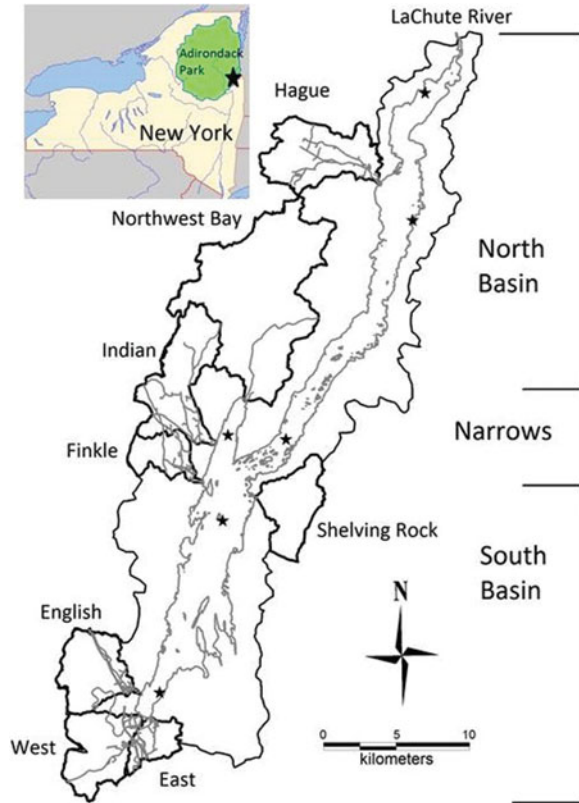
Figure 1. Map of Lake George including sub-watersheds sampled, offshore sampling locations, and the LaChute River. Sub-watersheds are outlined and labeled with roads represented as gray lines; offshore sampling locations are indicated with stars. Moving south to north they include: Tea Island, Dome Island, and Northwest Bay in the south basin, French Point in the Narrows, and Smith Bay and Rogers Rock in the north basin.

and the water column completely mixes soon after. The thermocline establishes in June at 10 to 15 m and, once established, slowly weakens and deepens to 15 to 20 m until turnover in early November.

The current configuration of Lake George resulted from the Wisconsinan Glaciation and the various lakes that formed during the glacial retreat ~13,000 years ago. During the retreat and melting of the Laurentian Ice Sheet, a number of lakes formed in the southern portion of current Lake George with southern flow. Several layers of sand below Lake George Village are believed to be a result of delta formations from these streams. With the glacier's final retreat these southern outflows became dammed by their own deposits, creating the present-day south-to-north flow. This process resulted in markedly different soil composition between the south and north basins, with the north basin comprised of finer sediments and clays associated with a final glacial retreat. Bedrock geology indicates the Lake George basin is a

mix of dominantly granitic gneisses, charnockitic gneisses, garnet-biotite-quartz-plagioclase gneisses, quartzites, meta-anthrosites, metagabbros with smaller quantities of marble, calcsilicates, and amphibolites (Shuster 1994).

Methods

Offshore chemical monitoring program

The Lake George Offshore Chemical Monitoring Program began in 1980 with the objective to characterize water chemistry throughout the lake, identify areas of concern, and provide data to support remediation recommendations. The program was designed mainly as an open-water testing program with a series of 6 deep-water ($\geq$20 m) sites along the south-to-north axis of the lake. Three sites were located in the south basin and included (south to north) Tea Island, Dome Island, and Northwest Bay. The north basin, which encompassed the Narrows and all sites north, included (south to north) French Point, Smith Bay, and Rogers Rock (Fig. 1). Spring sampling (late Apr to mid-Jun) occurred every 2 weeks until the onset of stratification. Summer sampling (mid-Jun to Sep) was reduced to monthly sampling intervals during stratification; 2-week intervals were reinstated for the fall period (Oct to mid-Nov).

Sampling at each location consisted of collecting epilimnetic and hypolimnetic samples for chemical analyses along with profiles for temperature, dissolved oxygen, and light. The epilimnetic sample was a composite sample (hose-integrated) from 0 to 10 m, while the hypolimnetic sample was a grab sample 1 m off the bottom. Samples were placed in coolers with ice until returned to the lab at the Darrin Fresh Water Institute where they were subsampled and analyzed. Complete chemical analyses included soluble and particulate nutrients, base cations, major anions, and metals; only sodium and chloride are presented here. Sodium was analyzed by atomic absorption using a Perkin Elmer AAnalyst 5000 (Standard Method 3111); chloride was analyzed by ion chromatography using a Lachat 8000 QuikChem (EPA Method 300; Clesceri et al. 1989). Standard quality assurance and quality control protocols included blanks, duplicate samples, spikes, and external check standards every 10 samples.

Stream survey

We monitored 8 major tributaries between 2007 and 2009 for chemistry and discharge. East, West, and English brooks are located around the more developed south end of the lake with the highest road density in the basin (Table 1). Finkle Brook is located in Bolton Landing with localized development, ~15 km north of the south end. Indian Brook, located ~19 km north of the south end, is heavily forested

Table 1. Average (standard deviation) sodium and chloride concentrations measured in each of the major sub-watersheds during baseflow conditions along with drainage area and lane km (percentage of land that is covered by roadways). Salt loading for each sub-watershed was estimated using the NYS DOT annual application rate of 9.36 tonnes salt/lane-km. Letters in the sodium and chloride columns represent significance between sites. Different letters indicate sites were significantly (ANOVA) different for that analyte.

Sub-watershed	Sodium (mM)	Chloride (mM)	Na:Cl	Area (km^2)	Roadway (lane-km)	Salt loading (tonnes)
Finkle	2.15(0.88)[a]	2.38(0.98)[a]	0.90	11.5	40.6(1.2%)	380
East	1.80(0.56)[ab]	2.06(0.64)[ab]	0.87	8.7	85.9(3.3%)	804
English	1.31(0.54)[abc]	1.52(0.65)[b]	0.86	21.8	146.4(2.3%)	1371
West	1.35(0.38)[bc]	1.32(0.42)[b]	1.02	25.5	100.4(1.5%)	939
Hague	0.55(0.18)[d]	0.57(0.25)[c]	0.96	29.1	69.1(0.8%)	647
Indian	0.50(0.14)[d]	0.64(0.16)[c]	0.78	31.3	118.5(1.3%)	1109
Shelving Rock	0.08(0.06)[e]	0.03(0.01)[d]	2.67	18.9	0	0
Northwest Bay	0.06(0.02)[e]	0.04(0.01)[d]	1.50	76.0	0*	0

*73.6 (0.3%) lane km of roads existed within the Northwest Bay watershed, but no paved roads were located upstream of the sampling location.

with sparse housing at higher elevations in the watershed. Northwest Bay is the largest sub-watershed of Lake George and remains relatively undeveloped ∼22 km north of the south end. NYS Route 9N is located within the Northwest Bay watershed, but the sampling location was located upstream of the roadway and therefore was considered to have no roadways influencing water chemistry. Shelving Rock is located on the east side of the lake ∼17 km north of the south end and is undeveloped with no paved roads within its sub-watershed. Hague Brook is located ∼40 km north of the south end and exhibits localized development with roads in close proximity to the stream corridor. The LaChute River, the only outlet from the lake, was monitored for chemistry but not flow due to its large width.

Discharge rate along with baseflow and storm event samples were collected from the 8 major tributaries and the LaChute River. Baseflow grab samples were collected every 2 weeks between April and November, with monthly sampling from December to March. Storm events and snowmelt were sampled when discharge was expected to rise substantially, >3-fold baseflow, using ISCO automatic samplers. Samples were collected and transported to the Darrin Fresh Water Institute where they were subsampled. The instrumentation and protocols were the same as described for the Offshore Chemical Monitoring with the exception that sodium was analyzed at the KECK Water Research Laboratory on the Rensselaer Polytechnic Institute campus in Troy, New York.

Flow monitoring stations were located as close to the lake as possible to ensure a representative sample of tributary discharge, with the exception of Northwest Bay Brook which was sampled upstream of NYS Route 9N. This site was established by the US Geological Survey in 1967 and was utilized as a control site because no development existed upstream. Each site consisted of a staff gauge and Telog

pressure transducer attached to a data recorder with a sampling rate of 15 minutes. Traditional stream gauging was conducted using a Brinkmann electromagnetic flow meter with measurements taken at 0.4 D at 1-foot (0.3 m) increments across the width of the stream (Gordon et al. 1992). Discharge measurements were conducted during ice-free periods throughout the 3-year study to establish reliable rating curves. Discharge volume from the LaChute River was provided by the LaChute Hydro Company, a subsidiary of Enel North America. Discharge was estimated using power generation, gate opening, lake level, and other variables; consequently, a specific formula was not available due to proprietary constraints. The discharge estimates generally agree with previous discharge calculations made by Shuster (1994) during 1987 and 1988. Relationships between annual discharge and precipitation data for 1987–1988 and 2007–2009 resulted in a coefficient of determination (r^2) of 0.85, demonstrating the general agreement of precipitation and resulting discharge volume between the 2 studies.

Statistical analyses

Statistical analysis was conducted using SPSS or SigmaPlot and focused on chloride concentration due to its more conserved nature. All analyses were done on raw data with no data transformations. Pairwise comparisons in the ANOVA analysis were performed using Holm-Sidak when data were normally distributed and Kruskal-Wallis when normality failed. Correlations were conducted using the Spearman Rank Order when data were not normally distributed. The term "significant" in this paper indicates the a priori *P* value ≤0.05 was met, with the statistical test indicated in parenthesis.

Percent impervious area attributed to roadways was calculated by determining the length and type of roadway in each sub-watershed, calculating the area covered, and then di-

viding by total area of the sub-watershed. Shape files for each sub-watershed and road maps were obtained from the Warren County Geographic Information System Program (Table 1). The total watershed area was calculated for each sub-watershed, and the length of each road within the sub-watershed was calculated and adjusted for the number of lanes in MapInfo. Interstate 87 had 6 lanes, NYS Route 9 had 4 lanes near Lake George Village, and all local roads were considered 2 lanes. All lanes were assumed to be 11-feet wide.

To assess the in-lake steady-state chloride concentration, lake export and change in lake chloride mass were determined. The more direct approach of determining loading estimates from surface and groundwater inputs was not favored because of the substantial contribution of groundwater to the lake's hydrologic budget and the lack of information on chloride concentrations in groundwater. Additionally, the streams sampled accounted for less than half of the land catchment draining the Lake George watershed, and the limited number of storm events sampled would result in additional estimation error. For whole-lake estimates, export and change in lake chloride mass provide a more accurate estimate than inputs.

The annual Lake George chloride export from 2007 through 2009 was calculated by multiplying the estimated annual discharge provided by the LaChute Hydro Company by the annual average chloride concentration in the LaChute River. Export was also calculated for each month by multiplying the monthly discharge and the average monthly chloride concentration in the LaChute River:

$$Cl\ export\ = LaChute\ discharge_{year}$$
$$*LaChute\ [Cl]_{year}. \qquad (1)$$

The annual and monthly calculations were within 5% agreement of each other for each year, indicating only small changes in concentration occurred throughout the year.

The annual change in lake chloride mass was calculated by subtracting the mass of chloride in the lake from the previous year. The average epilimnetic chloride concentration in the 6 deep-water sites were averaged with their hypolimnetic concentrations to obtain an average annual lake-wide chloride concentration. To determine the mass of chloride in the lake, the lake-wide chloride concentration was multiplied by the lake volume of $2.11e^{11}$ L (Boylen and Kuliopulos 1981):

$$\Delta Lake\ Cl\ mass\ = ([Cl]_{year} * 2.11e^{11}L)$$
$$-([Cl]_{year-1} * 2.11e^{11}L). \quad (2)$$
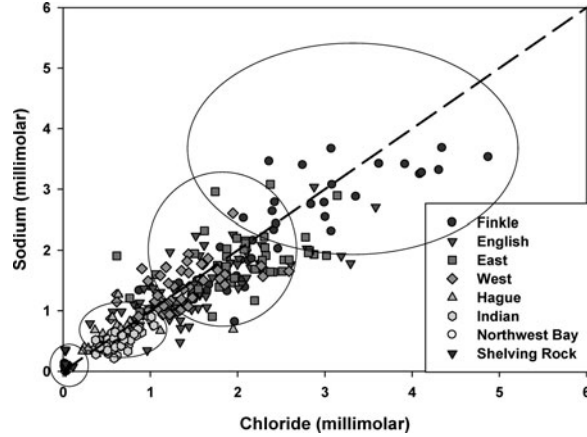
Figure 2. Sodium and chloride concentrations measured in 8 of the largest tributaries to Lake George during baseflow conditions between 2007 and 2009. Four significantly (ANOVA) different groups identified based on chloride concentrations.

Lake volume remained relatively stable because lake level was controlled by the hydroelectric power plant, and law required a stable lake level to be maintained for recreation and navigation.

Results

Stream salt concentration around Lake George ranged from near background levels (0–2 mg/L) to ~175 mg/L (i.e. moderately impacted) during baseflow. Chloride concentrations in 8 major streams around Lake George indicate 4 signif-
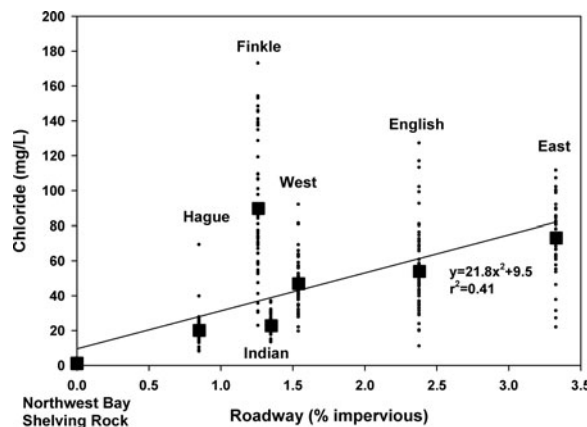
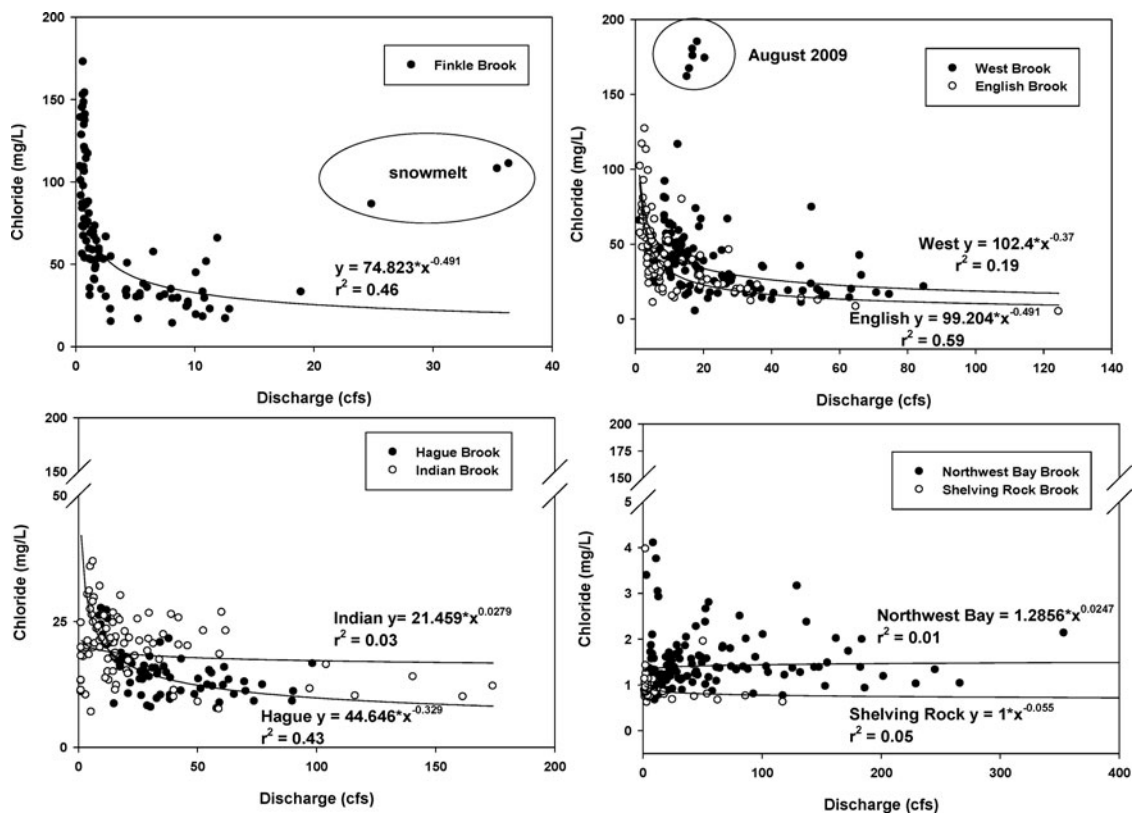**Figure 3.** Regression of stream chloride concentrations based on the percentage of impervious area created by roadways within each sub-watershed. Black circles represent individual baseflow measurements ($n = 432$) while the large squares represent the average baseflow chloride concentration. The percentage of roadway within each sub-watershed was significantly correlated (Spearman) to chloride concentration.

icantly (ANOVA) different groups based on baseflow concentrations (Fig. 2; Table 1). The chloride concentrations in the undeveloped watersheds of Northwest Bay and Shelving Rock brooks were near background concentrations and significantly different from all other streams. Indian and Hague brooks were slightly impacted and significantly different from all other stream concentrations. The more impacted streams, which included Finkle, East, English, and West brooks, separated into 2 significantly different groups. Finkle concentrations were the highest and significantly different from English and West brooks, while East Brook was not significantly different from either group (Table 1). As salt concentrations increased by stream, the average molar Na:Cl ratio decreased. In the least-developed watersheds, sodium concentrations often exceeded chloride concentrations, with a gradual transition to excess chloride in the more impacted streams (Fig. 2; Table 1). In the more impacted streams, the frequency of sodium exceeding chloride increased during fall, winter, and spring compared to summer when baseflow concentrations favored excess chloride.

The percentage of impervious land in each sub-watershed due to roadways was significantly correlated (Spearman) to stream chloride concentrations (Fig. 3). Plotting the significant correlation provided a linear regression to predict chloride concentration based on roadways within a watershed. The average measured chloride concentrations in East, English, and West brooks were 89.0, 87.8, and 109.0% of those calculated based on the amount of roadway surface in each sub-watershed, respectively. The largest underestimate of chloride concentration occurred in Finkle Brook, where the average measured concentration exceeded the calculated concentration by 242%.

Based on the NYS DOT annual application rate of 9.36 tonnes NaCl/lane-km (Godwin et al. 2003), an estimated 10,241 tonnes of salt were applied to the 1094 km of roads within the basin annually. The 8 sub-watersheds monitored accounted for 5939 tonnes (58%) of salt applied, with East, West, and English (south end) accounting for 3114 tonnes, 30% of the estimated total annual application for roads within the Lake George basin (Table 1). The area included in those 3 sub-watersheds accounts for ~10% of the total land catchment in the Lake George basin. Chloride concen-


Figure 4. Regression of stream chloride concentrations and instantaneous discharge rates in cubic feet per second (cfs). Each graph represents a significantly (ANOVA) different group based on baseflow chloride concentrations. East Brook was not included due to uncertainty in discharge rates due to beaver activity.
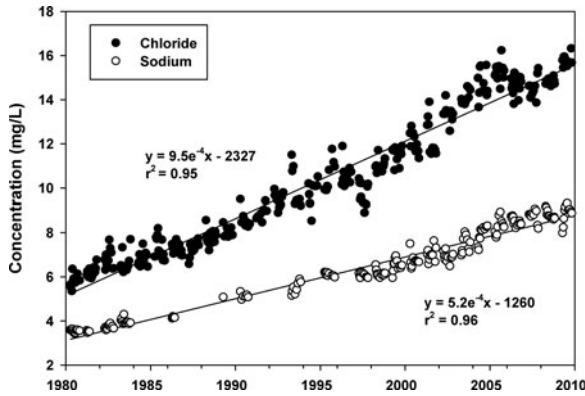
Figure 5. Average lake-wide epilimnetic sodium and chloride concentrations between 1980 and 2009 have nearly tripled. The rate of change is based on days (x = days).

trations in all streams substantially impacted by salt (Finkle, English, West, and Hague brooks) were significantly correlated (Spearman) to discharge, with concentrations decreasing exponentially as discharge increased. The coefficients of determination (r^2) were >0.40 for Finkle, English, and Hague brooks. Chloride concentrations in Northwest Bay, Shelving Rock, and Indian brooks were not significantly correlated to discharge (Fig. 4). East Brook was not included in the analysis due to the presence of beaver activity confounding discharge calculations.

The loading of salt to Lake George resulted in a consistent lake-wide increase in concentrations of chloride and sodium, nearly tripling since 1980. The average lake-wide concentrations in 1980 were 5.8 mg/L (0.163 mM) and 3.5 mg/L (0.154 mM) for chloride and sodium, respectively, and increased to 15.8 mg/L (0.444 mM) and 9.4 mg/L (0.407 mM)
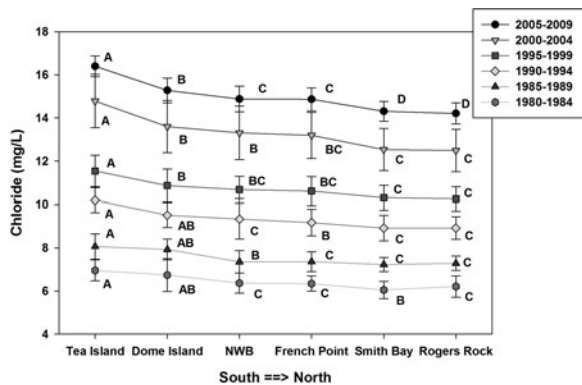
Figure 6. Average chloride concentration (Apr-Nov) at each location averaged over 5 years; error bars represent the standard deviation. Different letters within a 5-year bin indicate the sites were significantly (ANOVA) different from one another.

Table 2. Precipitation, LaChute discharge volume, discharge chloride concentration, and lake chloride export are reported for each year while the annual change in lake chloride represents the difference in Cl^- concentration and mass between the current and previous year. Chloride loading to Lake George was estimated as the sum of lake Cl^- export and the annual change in lake Cl^- mass. Precipitation data were obtained from the NOAA station at Glens Falls Airport located ~11 km from the south end of Lake George.

	2007	**2008**	**2009**	**Average**
Precipitation (cm)	95.0	123.8	101.5	109.1
Discharge volume (km^3/yr)	0.284	0.461	0.294	0.346
LaChute discharge Cl^- (mg/L)	13.8 (2.5)	14.5 (2.0)	14.6 (1.9)	
Lake Cl^- export mass (tonnes)	3918	6682	4284	4961
Δ Lake Cl^- (mg/L)	−0.26	0.43	0.88	
Δ Lake Cl^- mass (tonnes)	−538	898	1854	738
Lake Cl^- loading mass (tonnes)	3380	7580	6138	5699

in 2009 (Fig. 5). While annual concentrations steadily increased lake-wide, a strong in-lake gradient existed with greater salt concentrations in the south end of the lake (Fig. 6). Initially a weak gradient was present that resulted in similar chloride concentrations in the south and north basins based on ANOVA analysis. During the 1990s, a slightly stronger and more consistent gradient became established in the lake, and in the most recent interval (2005–2009) 4 significantly (ANOVA) different regions of the lake were identified. The 2 most southern sites, Tea and Dome islands, were significantly different from each other and all other sites. Northwest Bay and French Point, located near the Narrows, were significantly different from all other sites, and the 2 most northern sites, Smith Bay and Rogers Rock, were significantly different from the rest of the sites. Over the 30 years of record, chloride concentrations at Tea Island, the most southern site, exceeded those for Rogers Rock, the most northern site, by 1.5 mg/L (±1.1 SD). The concentration difference between these sites was 0.9 mg/L (±1.4 SD) during 1980–1984 and increased to 2.2 mg/L (±1.0 S.D.) during 2005–2009.

Based on chloride export and the annual change in the lake chloride mass, a steady-state salt concentration in the lake may occur in the near future. Between 2007 and 2009, an annual average of 4961 tonnes of chloride was exported from the lake with a range of 3918 to 6682 tonnes (Table 2). Annual chloride export significantly (linear regression) increased with annual precipitation. Chloride retained in the lake over the same period averaged 2373 tonnes Cl for an

estimated annual average loading of 5699 tonnes Cl (9343 tonnes NaCl). The estimated loading of 9343 tonnes of NaCl based on export and change in the mass of chloride within the lake closely agreed with the estimated 10,241 tonnes applied based on NYS DOT application rates. The average loading estimate and average discharge rate over the 3-year period indicated a steady-state lake chloride concentration of 16.6 mg/L (0.47 mM). Using the same loading and an average residence time of 6.8 years (Shuster 1994), the steady-state chloride concentration would be 18.3 mg/L (0.52 mM).

Discussion

Moderately impacted streams

Streams moderately impacted by road salt around Lake George had average chloride concentrations >1.0 mM Cl (35.5 mg/L), which included Finkle, East, English, and West brooks. These streams exhibited an average Na:Cl molar ratio between 0.86 and 1.02 over the 3-year sampling period (Fig. 2; Table 1). The variation observed in the ratio was dominantly associated with season. The summer salt concentrations almost exclusively favored excess chloride, with the frequency of sodium exceeding chloride greater during the remainder of the year; however, average chloride concentrations exceeded sodium in all months. The variation was most likely a result of the origin of the stream discharge. Baseflow concentrations primarily originated from groundwater, but the depth of groundwater changed with season. During the summer, baseflow was low and most likely originated from deep groundwater, while during the rest of the year the groundwater would have originated as shallow groundwater or a mix of deep and shallow groundwater. Groundwater closer to the soil surface would have a greater opportunity to transport subsurface sodium buildup to the stream, thus the opportunity for sodium concentrations to exceed chloride would be more likely to occur in fall, winter, and spring.

Rhodes et al. (2001) showed that chloride concentrations exceeded sodium in intermediate and highly impacted streams, with chloride being as much as 15% greater when concentrations exceeded 3.0 mM (106 mg/L). Finkle, East, and English brooks demonstrated average stream chloride concentrations exceeding sodium by 11–16%. When chloride concentrations exceeded 3.0 mM, chloride exceedance rose to 28%. West Brook was not included in the assessment due to the indirect input of water from a waste water treatment facility (Aulenbach et al. 1974). The facility operation fluctuates dramatically throughout the year based on the volume of tourism and confounded discharge rate, salt concentrations, and loading estimates.

The significant correlation between the amount of road surface in each sub-watershed and stream chloride concentrations was best represented by East, English, and West brooks. These brooks were located around the south end of the lake, an area that exhibited higher road densities than other sub-watersheds. The close agreement between the measured and estimated concentrations suggested that road surfaces were a driving force in the regression, while the large discrepancy in Finkle Brook indicated factor(s) other than roadways were influencing chloride concentration. The higher than expected chloride concentration in Finkle Brook is believed to be a result of the close proximity of the stream to an uncovered salt storage facility. It is unclear exactly how long the salt storage facility remained uncovered in that location, but it was in close proximity to Finkle Brook for at least several decades, finally being covered in the late 2000s. Snodgrass and Morin (2000) determined that 20% of salt stored uncovered can be lost before application, with spillage during stockpiling and loading identified as a major source of loss. To determine if the stream chloride concentrations were a result of the uncovered salt storage facility, soil samples along with shallow and deep groundwater will need to be collected and analyzed for sodium and chloride. Additionally, continuous monitoring of chloride or conductivity in the stream along with continuous discharge records will allow a comparison between the estimated road salt application and salt export from the stream. If export exceeds annual application substantially, that finding would strengthen the hypothesis that subsurface salt buildup was the cause of the elevated stream chloride concentrations.

Stream chloride concentrations have been shown to be directly correlated to stream discharge when substantially impacted by road salt. Periods of low flow exhibit higher concentrations because they are more representative of groundwater (Trowbridge et al. 2010). Long-term application results in greater groundwater salt concentrations, while periods of high flow are event driven (precipitation and snowmelt) and tend to dilute the stream chloride concentrations. Exceptions can occur during snowmelt when large pulses of salt are washed into streams by runoff, a finding observed in Finkle Brook (Fig. 4). Finkle and English brooks were strongly correlated with discharge rate, while West Brook agreement was only marginal due to the additional input from the waste water treatment facility. East Brook could not be included because the flow records were compromised by the presence of beavers in the watershed.

Slightly impacted streams

Hague and Indian brooks were both slightly impacted by road salt with similar stream chloride concentrations around 0.6 mM (21 mg/L) but different Na:Cl ratios. The close

proximity of Hague Brook to roads may explain the near 1:1 Na:Cl ratio and the significant correlation between discharge and stream chloride concentration. NYS Routes 8 and 21 were in close proximity to Hague Brook throughout a substantial portion of the watershed while the majority of roads in Indian Brook were at higher elevations in the watershed, substantially separated from the main stream reach. In Indian Brook, sodium rarely exceeded chloride while Hague Brook concentrations were almost 1:1, on a molar basis. The close proximity of Hague Brook to the roads would allow applied road salt to be quickly flushed into the stream and surrounding area. The longer transport distance in the Indian Brook watershed would provide greater opportunity for sodium to become incorporated into soil via cation exchange reactions thus creating a greater deviation between sodium and chloride concentrations (Norrström and Jacks 1998, Kelly et al. 2008). Additionally the longer transport distance for applied road salt to reach streams in the Indian Brook watershed could explain the lack of a strong correlation between stream chloride concentration and discharge rate. The lower than expected chloride concentrations in Indian Brook, based on the amount of road surface within the watershed, may be explained by the greater proportion of residential roads higher in the watershed. Residential roads do not receive the same attention as more heavily utilized roads and therefore normally have lower salt application rates.

Minimally impacted streams

The minimally impacted sites, Northwest Bay and Shelving Rock brooks, represented background or near background chloride concentrations at 1.3 and 1.0 mg/L, respectively. Streams unimpacted by road salt around Lake George are consistent with streams near Rich Lake in the central Adirondacks that exhibited an average chloride concentration of 1.1 mg/L in a larger stream and ~0.5 mg/L in smaller tributaries (Demers and Sage 1990). Additionally, the greater Na:Cl ratio may reflect the natural weathering of sodium-bearing rocks in these areas. Lake George does possess pockets of orthosite and granitic gneiss that contain sodium-bearing minerals, but no known chloride-bearing minerals occur in the basin (Shuster 1994). Chloride concentrations in precipitation were normally <0.5 mg/L, and therefore loading from precipitation is viewed as minimal.

Salt in lake george

The loading of salt to Lake George has resulted in a steady lake-wide increase since at least 1980 with an in-lake salt gradient that has increased over time. Two-thirds of the Lake George watershed drained into the south basin, which had a higher density of roads that included the only interstate highway in the watershed. The higher density of roads reflected the degree of development within the south basin, accounting for 73% of the total 9009 buildings as of 2009 based on available data from property tax records, building permits, and Adirondack Park Agency permits. The most intensely developed area was centralized around the south end of Lake George. Elevated stream chloride concentrations in East, English, and West brooks reflected this development. Roads within these 3 watersheds accounted for ~30% of the total estimated application or 3114 tonnes NaCl annually. These 3 streams entered Lake George in the area of Tea Island, which consistently and often significantly (ANOVA) exhibited chloride concentrations greater than other in-lake sampling locations. The disproportionate salt loading in the south end has intensified the absolute gradient while the relative difference has remained fairly consistent between the south and north ends. Between 1980 and 1984, the chloride concentration difference between Tea Island in the south and Rogers Rock in the north was 0.88 mg/L ($\pm$1.39 SD) which represented 13% of the Rogers Rock concentration at 6.2 mg/L. Between 2005 and 2009, the chloride difference between the 2 sites was 2.2 mg/L ($\pm$1.0 SD), representing a 15% difference based on the Rogers Rock concentration of 14.2 mg/L. These results implied that relative loading was similar between the time periods but consistently disproportionate. The input of less saline water from Northwest Bay and Shelving Rock brooks has exacerbated the salt gradient between the south and north basins. These 2 sub-watersheds accounted for ~20% of the land catchment in the basin and enter the lake near the Narrows. The addition of less saline water has additionally diluted chloride concentrations in the north end of the lake. This pattern is not likely to change because these areas are dominantly protected by New York State's Forest Preserve statute.

Salt plateau

The steady increase of salt within Lake George does not seem to be changing, but the lake may reach a steady-state in the near future when the quantity of salt entering the lake is equal to the salt exported from the lake. Based on lake chloride export, the annual change in lake chloride mass (Table 2) and the assumption that future road salt application does not increase, the lake-wide average is estimated to reach a steady-state chloride concentration of ~17 mg/L (0.48 mM). The close agreement between the estimated 10,241 tonnes NaCl applied annually and the average loading of 5699 tonnes Cl (9343 tonnes NaCl) strengthens this assumption. If the steady-state chloride concentration within the lake is reached, variation is expected from year to year based on precipitation. Between 2007 and 2009,

precipitation ranged from 95.0 to 123.8 cm, with annual chloride loadings directly related to precipitation received within the same year. The relationship between precipitation and annual changes in lake chloride mass are less clear. For instance, the annual change in lake chloride mass was greatest in 2009 while annual precipitation was highest in 2008 (Table 2), suggesting a lagged concentration response time.

A better understanding of the groundwater loading may help explain the variation in the change in lake chloride mass. With an estimated lake volume of 2.11 km^3, the increased mass of 1854 tonnes Cl in 2009 equated to an increase of almost 1 mg/L; therefore, lake-wide concentration could vary by more than 1 mg/L due to excessive precipitation or drought. Additionally, the $\sim$15% gradient between Tea Island and Rogers Rock will most likely remain to some degree. Assuming the south end of the lake will continue to supply $\sim$30% of the salt to the Tea Island sub-basin and the continued dilution from Northwest Bay and Shelving Rock brooks, the gradient will likely remain.

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