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Modeling salinization and recovery of road salt-impacted lakes in temperate regions based on long-term monitoring of Lake George, New York (USA) and its drainage basin

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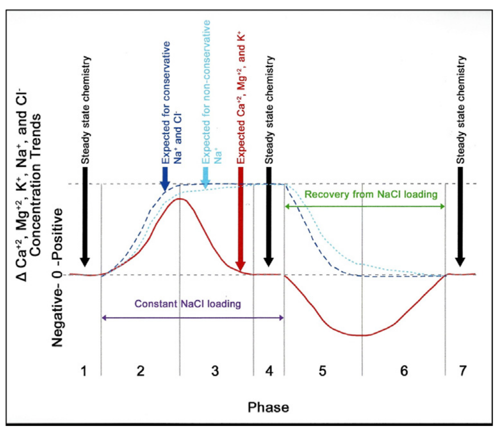
HIGHLIGHTS

- Long-term road salt application accumulates salt in soils and receiving waters.
- Salt (NaCl) concentration in Lake George, NY increased 30-fold from 1940 to 2009.
- Ca^{+2}, Mg^{+2}, and Na^{+} increased in runoff and the lake from soil ion exchange of Na^{+}.
- Models are presented for trends of tributary cations and Cl^{-} in temperate lakes.
- Na^{+} from salt and H^{+} from acid deposition displace soil cations in a similar manner.

GRAPHICAL ABSTRACT



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ABSTRACT

Road salt mitigates winter highway icing but accumulates in watershed soils and receiving waters, affecting soil chemistry and physical, biological, and ecological processes. Despite efforts to reduce salt loading in watersheds, accumulated cations and Cl^{-} continue to impact tributaries and lakes, and the recovery process is not well understood. Lake George, New York (USA) is typical of many temperate lakes at risk for elevated Cl^{-} concentrations from winter deicing; the lake salt concentration increased by ~3.4% $year^{-1}$ since 1980. Here, we evaluated the ionic composition in Finkle Brook, a major watershed draining to Lake George, studied intermittently since 1970 and typical of other salt-impacted Lake George tributaries. Salt loading in the Lake George basin since the 1940s displaced cations from exchange sites in basin soils; these desorbed cations follow a simple ion-exchange model, with lower sodium and higher calcium, magnesium and potassium fluxes in runoff. Reduced salt application in the Finkle Brook watershed during the low-snow winter of 2015–2016 led to a 30–40% decline of Cl^{-} and base cations in the tributary, implying a Cl^{-} soil half-life of 1–2 years. We developed a conceptual model that describes cation behavior in runoff from a watershed that received road salt loading over a long period of time, and then recovery following reduced salt loading. Next, we developed a dynamic model estimating time to steady-state for Cl^{-} in Lake George with road salt loading starting in 1940, calibrating the model with tributary runoff and lake chemistry data from 1970 and 1980, respectively, and forecasting Cl^{-} concentrations in Lake

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George based on various scenarios of salt loading and soil retention of Cl^-. Our Lake George models are readily adaptable to other temperate lakes with drainage basins where road salt is applied during freezing conditions and paved roads cover a portion of the watershed.

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1. Introduction

New Hampshire was the first US state to adopt sodium chloride (NaCl, road salt) as a deicing compound during the winter of 1941–1942 (Kelly et al., 2010). Seven decades later, about 15 million t were applied to US roads during the winter of 2013–2014 (Riley, 2015). The increasing use of road salt to mitigate winter icing in temperate regions has caused widespread salinization of lakes throughout North America and Europe (Novotny et al., 2008; Müller and Gächter, 2011). At least 7770 US lakes may be at risk for elevated Cl^- concentrations from winter highway deicing (Dugan et al., 2017). Sustained applications of road salt cause accumulation of Na^+ and Cl^- in watershed soil and soil water where ion-exchange processes alter the cation composition of runoff into receiving waters and ecosystems (Norrstrom and Bergstedt, 2001; Rosfjord et al., 2007; Rhodes and Guswa, 2016). Effects can cause mortality of forest vegetation from direct impact of aerosols and changes of chemical and physical soil properties in close proximity to treated highways (Fleck et al., 1988; Langen et al., 2006; Kelting and Laxson, 2010). Salinity increases in lakes can affect phytoplankton community composition toward cyanobacteria (Tonk et al., 2007; Pade and Hagemann, 2015), and alter circulation and water column overturn (Bubeck and Burton, 1989; Ramakrishna and Viraraghavan, 2005; Novotny et al., 2008; Sibert et al., 2015). Excess Na^+ in drinking water is a health concern for individuals on low sodium diets due to hypertension. The US EPA requires monitoring of drinking water for Na^+ and public water suppliers to report concentrations above 20 mg L^{-1} to local health authorities (US EPA, 2003).

Environmental and public health concerns about adverse effects from road salt may stimulate efforts to reduce salt loading in watersheds. However, salt accumulated in roadside soils, and altered concentrations of exported cations will continue to impact streams and lakes following reductions of salt input. The recovery time in down-gradient receiving waters in response to reductions has received little investigation and is poorly understood. Meanwhile, salt loading from winter deicing has been increasing for up to six decades in many watersheds (Jackson and Jobbágy, 2005) and drainage basins, including Lake George, New York (USA), where the lake salt concentration has increased by about 3.4% $year^{-1}$ since 1980 (Boylen et al., 2014), and has been raised by a factor of ~30 since the early 1940s (inferred from Lipka and Aulenbach, 1976; NADP, 2017).

Here, we evaluated the evolution of ionic concentrations in Finkle Brook, a major Lake George watershed, which has been monitored intermittently since 1970 and is typical of the salt-impacted drainage from other Lake George tributaries. The warm, low-snow winter of 2015–2016 at Lake George (SM Figs. S1 and S2 + text) provided an opportunity to evaluate the response of a typical road-salt impacted watershed and tributary following an abrupt 30–40% reduction of road salt application. Finkle Brook discharge and chemical data from 1970 to 2016 allowed the development of a qualitative predictive model for changing soil chemistry and cation concentrations in runoff from salt-impacted watersheds, and predictions of future trends of cation export to the lake during recovery. Similarly, the long term data record from Lake George permitted us to develop a quantitative model of Cl^- concentration in the lake from 1940 to 2009. Then, we predicted Cl^- concentrations in Lake George based on various scenarios of salt loading and retention of salt in watershed soils. Our primary objective in developing this predictive model was to provide guidance regarding how rapidly salt concentrations in the lake would respond to policy-directed efforts to reduce un-necessary excessive salt loading in the watersheds, recognizing the likely adverse environmental consequences of continued unrestrained applications of roadway de-icing salt.

2. Materials and methods

2.1. Study area

Lake George, NY USA has a surface area of 114 km^2 with a 209-km irregular shoreline containing numerous bays (Sutherland et al., 1983) (Fig. 1). There are 1094 km of roads within the Lake George drainage basin that drain to ground water, tributaries and eventually to Lake George (Swinton et al., 2015). The terrestrial basin surface area is 492 km^2, resulting in a drainage basin:lake surface ratio of 4.3. The topography of the drainage basin generally is steep, with elevation differences in excess of 700 m between the lake surface and the drainage divide. >95% of the Lake George drainage basin and 100% of the three watersheds described herein are underlain by Grenville-aged (ca. 1 billion years old; Precambrian) syenite and granite, and lesser amounts of granitic and metasedimentary gneiss (Newland and Vaughn, 1942). The latter includes minor quartzite (mostly SiO_2) and marble ($CaCO_3$). Neither marble nor evaporites are known in the three watersheds and there are no known evaporite sediments in the Lake George basin. Only trace amounts of Cl^- occur in relatively minor slow-weathering silicate minerals. Post-glacial surficial deposits (till, kame terraces, and possibly small perched deltas) are composed largely of locally derived material but may have detritus from bedrock outside the basin (see SM text on geology).

The Finkle Brook watershed, located on the west side of Lake George (Fig. 1) within the Town of Bolton, is moderately developed (8.9% developed; 0.45% impervious), and has a volume-weighted mean annual discharge of 0.16 $m^3 s^{-1}$ (United States Geological Survey StreamStats Beta 4 v. 4; URL: http://streamstatsags.cr.usgs.gov/streamstats/). Detailed daily discharge, cumulative discharge, precipitation, cumulative precipitation, and snowpack for Finkle Brook in 2014, 2015, and 2016 are in SM Figs. S1 and S2. The watershed has 16.1 km of roads that receive winter road salt application, 12% of the total road mileage salted within the Town of Bolton. From 2008 through 2015, the Town purchased 1447 ± 440 t $year^{-1}$ of road salt, or 174 t $year^{-1}$ for the Finkle Brook watershed. Earlier Town records were not available. The winter 2015–2016 salt reduction in the Finkle Brook watershed was ~61 t (pers. comm., Bolton, NY, Highway Superintendent). Minor human sources of Na^+, Ca^{+2}, and Cl^- within the watershed include residences (ca. 100, population estimated at 240 people; Town of Bolton records) not on the public sewage system (source of Cl^-), and several concrete culverts and a spillway (source of Ca^{+2}). Sodium chloride, Ca^{+2}, and Mg^{+2} in applied granular road salt, supplemented by sand, ranged from 98 (prior to mid-winter of 2014–2015) to 96% (mid-winter 2014–2015 onward), 0 to <1%, and 0 to 0.5%, respectively (SM Table S1 + text), for all years for which data were available. Most concrete structures (spillway, bridge, culverts) have been in place for the duration of our time series. Thus, increases in Ca^{+2}, Mg^{+2}, and K^+ concentrations that occurred during increased loading of NaCl from 1970 to 2016 cannot be attributed to the leaching of concrete structures, nor to impurities in the salt.

2.2. Data sources and protocol

2.2.1. Lake George Cl^- and cation monitoring

Lake George is dimictic, with complete overturn of the water column in spring and fall. Most roadway de-icing salt within the Lake George

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