



Storage and release of road-salt contamination from a calcareous lake-basin fen, western Massachusetts, USA



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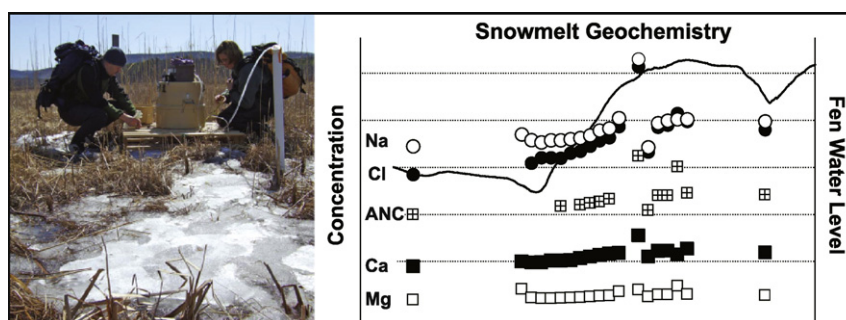
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HIGHLIGHTS

- Field study investigates dissolved salt movement in a fen during hydrologic events.
- We characterize adsorption and desorption reactions for sodium on peat.
- Calculations of selectivity coefficients show that sodium exchanges with magnesium.
- We measured geochemical flux of Na and Cl exiting the fen during snowmelt.
- Non-winter rain events more effectively export salt because snowmelt introduces contamination.

GRAPHICAL ABSTRACT



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ABSTRACT

Road salt (NaCl) applications to highways have increased stream sodium and chloride concentrations due to retention within watersheds. The mechanisms for retention and export of Na^+ and Cl^- from different environments are not fully understood. This field study examines the hydrologic and cation exchange processes that store and release Na^+ and Cl^- from a calcareous fen adjacent to a highway. Despite high concentrations of Ca^{2+} and Mg^{2+} , elevated salt concentrations enable Na^+ to occupy up to 15% of the cation exchange capacity of shallow peat. Calculations of selectivity coefficients show that Na^+ preferentially exchanges with Mg^{2+} , and Na^+ can be desorbed under more dilute conditions caused by precipitation and snowmelt.

Detailed sampling of surface and ground waters during three snowmelt events illustrate early releases of Na^+ and Cl^- at the onset of melting, with maximum fluxes coinciding with peak discharge. From 7 March through 4 April 2005, the flux of dissolved salt exiting the wetland amounts to 13% (Na) and 17% (Cl) of annual rock salt applied to the highway. For all of 2005, the total salt mass leaving the wetland via Kampos Brook is similar to the amount of road salt applied; 50% of the annual salt efflux occurred during the snowmelt season of March through May. In general, exported Na^+ and Cl^- correlate with the number of lane miles of highway crossing the watershed. Large rain events outside of winter months are more effective than snowmelt with reducing dissolved salts because snowmelt also introduces contamination. For this and other wetlands having alkaline geochemistry and high flushing rates, management strategies that reduce rock salt amounts to roadways will assist with reducing salt contamination to levels less toxic to vegetation and aquatic organisms.

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Abbreviations: [X], activity of ion X; CEC_e , cation exchange capacity, equivalents per mass ($\text{cmol}_c \text{kg}^{-1}$); CEC_m , cation exchange capacity, moles per mass (cmol kg^{-1}); $E_{X(\text{CEC}_e)}$, charge fraction of exchangeable ion X of total CEC_e ; $M_{X(\text{CEC}_m)}$, mole fraction of exchangeable ion X of total CEC_m ; K, selectivity coefficient, Vanselow convention; ANC, acid neutralizing capacity ($\mu\text{eq L}^{-1}$), where $\text{ANC} = \sum \text{bases} - [\text{H}^+]$; F_i , flux of ion i (mmol sec^{-1}); Q, stream discharge (L sec^{-1} or $\text{m}^3 \text{sec}^{-1}$); C_i , concentration of ion i .

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1. Introduction

Large quantities of road salt (NaCl) are commonly applied to highways and other impervious surfaces to improve driving conditions in snowbelt regions. Yet, pollution from road salt and other deicing agents has lowered surface water and groundwater quality (Scott, 1980; Rhodes et al., 2001; Godwin et al., 2003; Thunqvist, 2004; Kaushal et al., 2005; Ramakrishna and Viraraghavan, 2005; Kelly et al., 2008; Daley et al., 2009; Mullaney et al., 2009; Likens and Buso, 2010; Trowbridge et al., 2010; Novotny and Stefan, 2012; Corsi et al., 2015) and stressed ecosystems (Wilcox, 1986a; Richburg et al., 2001; Green et al., 2008; Corsi et al., 2010; Findlay and Kelly, 2011). Usage of road salt in the United States has increased steadily since the 1940s due to increased urban (Novotny et al., 2009; Corsi et al., 2010) and suburban (Likens and Buso, 2010) development. In Massachusetts, applications of road salt to state and interstate highways average ~500,000 tons (453,590 metric tons) per year with annual amounts varying by ~200,000 tons (181,440 metric tons) based on the severity of the winter season (MassDOT, 2012).

Observed increases in salinity of streams over decadal timeframes have been attributed to long-term retention and accumulation of dissolved salts within watersheds (Godwin et al., 2003; Kaushal et al., 2005; Kelly et al., 2008; Daley et al., 2009; Novotny et al., 2009; Trowbridge et al., 2010). For example in the rural watershed of the Mohawk River, New York, chloride concentrations increased from 7.7 mg/L in the 1950s to 20.4 mg/L in the 1990s, whereas other dissolved constituents decreased or remained the same (Godwin et al., 2003). In New Hampshire (Daley et al., 2009; Trowbridge et al., 2010), high road-salt application rates due to harsh winter climates and increased urbanization have caused chloride concentrations in streams to exceed chronic toxicity levels of 230 mg/L (USEPA, 1988) in both winter and summer months. Retention of dissolved salts in soils, surface water, and groundwater from past road-salt applications result in high concentrations in streams year round, not just when road salt is applied (Rhodes et al., 2001; Kelly et al., 2008; Daley et al., 2009; Novotny et al., 2009; Trowbridge et al., 2010). As an example of high retention, large metropolitan watersheds in Minneapolis/St. Paul, Minnesota have an average chloride retention rate of 72% of road salt applied (Novotny et al., 2009). Shallow groundwater retains significant amounts of dissolved salts, with concentrations as high as 2000 mg L⁻¹, and these urban watersheds have not reached equilibrium conditions where inflow = outflow even after decades of road salt use (Novotny et al., 2009).

Wetlands, which are often crosscut by interstate and state highways (e.g. Forman and Deblinger, 2000), can contribute to retention of road salt within watersheds. The retained salts subsequently can have adverse effects on wetland vegetation, including alteration of plant community attributes and spread by invasive species (Isabelle et al., 1987; Panno et al., 1999; Richburg et al., 2001). As an example, vegetation in peatlands—whose ecology is largely defined by their chemistry—can be particularly sensitive to geochemical alterations (Wilcox, 1986a; Schot and Wassen, 1993; Panno et al., 1999; Richburg et al., 2001). Bogs, acidic peatlands isolated from regional groundwater flow, retain dissolved salts in peat pore spaces (Wilcox, 1986b; Panno et al., 1999), and sodium readily exchanges with H⁺ on peat surfaces in these acidic environments (Pugh et al., 1996). The extent of sodium retention in more alkaline peatlands, such as fens, is less understood, however. The high concentration of divalent cations (Ca²⁺ and Mg²⁺) in fens may inhibit cation exchange with Na⁺, as has been shown for organic material in calcareous soils (Green and Cresser, 2008). Additionally, unlike bogs, groundwater and surface water flow through fens provides a potential mechanism for release of road-salt pollution during hydrologic events, such as snowmelt.

This investigation characterizes the conditions under which sodium and chloride are mobilized and retained in a large (70 ha) calcareous, graminoid, lake-basin fen that receives road-salt runoff from an

interstate highway. In particular, we measure the geochemical response of the fen to significant snowmelt and rain, and we determine the flux of sodium and chloride exported from the fen during snowmelt. Measurement of exchangeable cations on peat is used to interpret the chemical reactions with groundwater that affect sodium retention on peat surfaces. Ultimately, understanding the storage and release of road-salt pollution by fens will help inform conservation and management decisions for other salt-sensitive wetland environments.

2. Background

The study site is Kamposoa Bog (42.293N, 73.305W, elev. = 300 m, 70 ha), located in the Paleozoic marble region of the Berkshire Mountains of western Massachusetts and in the towns of Stockbridge and Lee. Misnamed a bog, Kamposoa Bog is the largest and most biologically diverse calcareous lake-basin fen remaining in Massachusetts. It is a member of the Massachusetts Areas of Critical Environmental Concern (ACEC) based on the ecological significance of its habitat and stewardship by local landowners, regional planning commissions, and state conservation agencies (Mass EEA, 2015). Approximately 19 state-listed rare plant and animal species occur in Kamposoa Bog; descriptions of plant assemblages are found in Richburg (1999). The diversity of wetland plants has been diminished by road-salt contamination from the Massachusetts Turnpike, an adjacent interstate highway (Richburg et al., 2001). In their study of plant diversity and shallow groundwater chemistry, Richburg et al. (2001) showed an association between lower plant species richness, cover, and abundance in areas of the fen having high concentrations of sodium (>112 mg/L) and chloride (>54 mg/L). In some localities within the fen, the road-salt contamination may have facilitated spread of the invasive plant *Phragmites australis australis* (Richburg et al., 2001), a salt-tolerant, giant reed that can colonize rapidly in wetlands located alongside transportation corridors (Brisson et al., 2010).

Fens are peatlands that have a strong connection to alkaline, minerotrophic groundwater rich in calcium, magnesium and bicarbonate. Some water movement—albeit slow—is required in fens to provide nutrients to plant root zones and to remove organic acids (Schot and Wassen, 1993; Almendinger and Leete, 1998; Bedford and Godwin, 2003; Duval and Waddington, 2012). Otherwise, the dissolved minerals are removed by vegetation and not replaced, resulting in acidic, nutrient-poor environments more characteristic of bogs. However the slow movement of water in fens lowers the rate of plant decomposition, which enables the buildup of peat (Bedford and Godwin, 2003). Even though water chemistry and hydrology strongly control the ecological structure within Kamposoa Bog, little is known about the major flow paths of water that supply nutrients and dissolved minerals to vegetation, in addition to those that mobilize salt contamination.

2.1. Site description

Kamposoa Bog lies within a 4.7 km² watershed with 160 m of relief (Fig. 1). The Bog is a circumneutral wetland complex that consists of a central open pond (1.3 ha) surrounded by an open, graminoid fen (9.2 ha), a shrub fen (4.4 ha), and a red maple, calcareous seepage swamp (55.8 ha, Richburg et al., 2001). The four-lane Massachusetts Turnpike crosses the northern edge of the wetland, and a two-lane state highway (Rt. 7) crosses the southern margin. The primary inlet stream to the wetland is Marsh Brook, which is renamed Kamposoa Brook after it flows beneath the Massachusetts Turnpike and enters the wetland complex. Kamposoa Brook (KB) flows south through the fen, the open pond, and eventually beneath Rt. 7. A second, unnamed inlet stream enters the wetland through the red maple swamp in the southeastern region of the complex and flows southwest, parallel to Rt. 7, before joining Kamposoa Brook. In addition to receiving runoff

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