



Transport of road salt contamination in karst aquifers and soils over multiple timescales



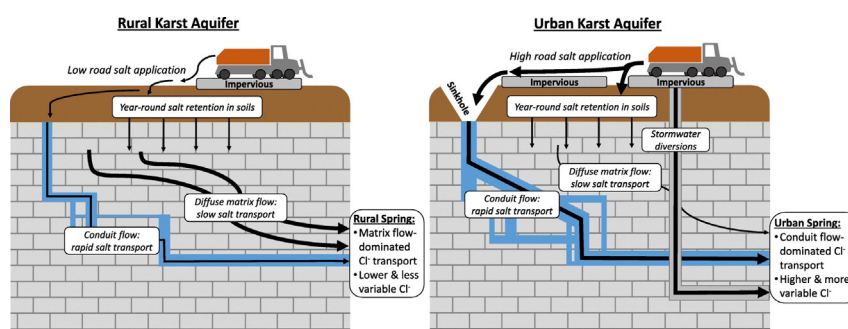
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HIGHLIGHTS

- Continuous karst spring data elucidated salt transport over multiple timescales.
- Conduit flow rapidly transported some salt while the rest moved via diffuse flow.
- An urban spring had more conduit flow while a rural spring had more diffuse flow.
- Salt stored in soil was released year-round by first flush events during flooding.
- Cl^- increased at a rural spring over 20 years due to inter-annual salt retention.

GRAPHICAL ABSTRACT



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ABSTRACT

Road deicing has caused widespread environmental Na^+ and Cl^- release for decades, yet the transport and retention of these contaminants in karst aquifers and soils are poorly understood. We examined the transport dynamics of Na^+ and Cl^- from road salt in shallow groundwater during flooding and over seasonal timescales by intensively monitoring an urban and a rural karst spring over approximately 2 years. Furthermore, we used a 20-year dataset for the rural spring to determine how salt retention affected long-term geochemical trends in the shallow groundwater. Salt transport was governed by hydrologic pathways through karst aquifers: during winter and early spring floods, flow through preferential pathways rapidly transported salty meltwater or stormwater over hours to days, while the remaining salt-contaminated water moved diffusely through the rock matrix on timescales of months to years. Flood hydrograph separations revealed that event water constituted 61.2% of stormflow on average at the urban spring, leading to more extreme variability in salt concentrations during flooding and throughout the year. This variability indicates that baseflow contributions to urban streams overlying karst aquifers with preferential flowpaths are likely less effective at buffering salt concentrations. In contrast, salt concentrations were less variable in the baseflow-dominated rural spring (28.7% event water). Furthermore, salt was episodically released from soils to shallow groundwater throughout the year during first flush events. A Cl^- mass balance indicates that Cl^- applied during previous winters persists within the springs' recharge basins for more than a year, raising baseline concentrations as road salt is introduced faster than it can be flushed from the basin. Inter-annual salt retention by soils or slow groundwater movement likely caused significant Cl^- and specific conductivity (SpC) increases at the rural spring from 1996 to 2016. Accumulation of salt in shallow groundwater can elevate baseflow concentrations in surface waters, where it threatens aquatic organisms.

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1. Introduction

Road salting improves winter travel safety but also releases Na^+ and Cl^- into the environment, altering the chemistry of nearby soil, groundwater, and streams. Following road salt application, Na^+ and Cl^- enter soils and then are delivered to shallow groundwater, which feeds local surface water features via baseflow. Surface water Na^+ and Cl^- concentrations increase with impervious surface area and can remain elevated throughout the year due to temporary salt retention within watersheds (Corsi et al., 2010, 2015; Daley et al., 2009; Kaushal et al., 2005; Ledford et al., 2016; Mason et al., 1999; Novotny et al., 2008a).

Catchment-scale salt retention is important because if Na^+ and Cl^- cannot be flushed completely from watersheds between road salting seasons, these ions build up and increase background concentrations in streams. Long-term Na^+ and Cl^- increases have been observed in several lakes and streams in North America (Bowen and Hinton, 1998; Corsi et al., 2015; Dailey et al., 2014; Daley et al., 2009; Godwin et al., 2003; Kaushal et al., 2005; Kelly et al., 2008; Novotny et al., 2008a; Perera et al., 2013; Sun et al., 2014) and Europe (Müller and Gächter, 2012; Thunqvist, 2004). Groundwater salt concentrations are increasing as well; in the Chicago, Illinois metropolitan area, Kelly (2008) found that groundwater Cl^- concentrations increased from 6 mg/L prior to 1950 to a median value of 20 mg/L from 1990 to 2005. In Connecticut, average groundwater Cl^- increased from 2 mg/L in 1894 to 29 mg/L in 2002–2007 (Cassanelli and Robbins, 2013).

Mass balance studies quantify the annual salt retention that is responsible for these long-term increases. Howard and Haynes (1993) found that 45% of Cl^- applied as road salt left an urban Canadian watershed as stream runoff each year from 1988 to 1991, with the rest being temporarily stored in groundwater. Perera et al. (2013) performed a new mass balance for the same Canadian watershed from 2004 to 2008, this time accounting for dual porosity in the aquifer due to “urban karst” (i.e., a dual porosity aquifer in which the secondary porosity is created by adding permeable fill material around pipes and drains; this secondary porosity is superimposed upon the primary porosity of the rock matrix). They found that about 60% of the applied Cl^- was released from the watershed as runoff or as groundwater flow through preferential flowpaths, with the other 40% entering the aquifer where it flowed diffusely through the rock matrix (Perera et al., 2013). Most mass balance studies show annual Cl^- retention in watersheds ranging from 40 to 78% (Meriano et al., 2009; Novotny et al., 2008b; Ruth, 2003). However, a multi-year mass balance study (1985–2005) of a rural New York stream showed net Cl^- retention within the watershed some years and net Cl^- release other years (Kelly et al., 2008). In these mass balance studies, Cl^- retention is usually attributed to slow groundwater movement, although soils contribute to Cl^- retention as well (Labadia and Buttle, 1996; Lax and Peterson, 2009).

Movement through soils slows Na^+ and Cl^- transport partly because of the conveyance of these ions depends on the residence time of porewaters and partly because they are retained through interactions with the soil. Na^+ is retained through adsorption to negatively charged sites on clays or organic matter, replacing other major cations or metals and releasing them to the soil porewater (Backstrom et al., 2004; Meriano et al., 2009; Nelson et al., 2009; Robinson et al., 2017; Schweiger et al., 2015). Cl^- has traditionally been considered a conservative ion, but recent evidence has shown that it can be retained in soils (Robinson et al., 2017) through plant or microbial uptake, non-specific adsorption, and chlorination of soil organic matter (Bastviken et al., 2007, 2009; Lovett et al., 2005; McBride, 1994). Soil retention delays the release of Na^+ and Cl^- to shallow groundwater, and then groundwater flow rates control the timing of its delivery to surface waters.

Elevated Na^+ and Cl^- concentrations from road salt contamination have a wide range of ecological impacts for plants and aquatic organisms. Individual species have varying salt tolerances, and road salt contamination causes ecosystems to shift toward dominance by salt-tolerant species, reducing biodiversity among macroinvertebrates (Demers, 1992;

Williams et al., 1997), amphibians (Collins and Russell, 2009), and wetland plants (Panno et al., 1999; Richberg et al., 2001). The USEPA designates a Cl^- chronic exposure limit for aquatic life at 230 mg/L and an acute exposure limit at 860 mg/L (USEPA, 2017). Indeed, acute exposure can cause mortality for amphibians and macroinvertebrates at Cl^- concentrations on the order of 1000 mg/L (Blasius and Merritt, 2002; Collins and Russell, 2009; Sanzo and Hecnar, 2006; Williams et al., 1999).

Springs provide a window into conditions in the shallow subsurface, and they have previously been examined to elucidate spatial patterns in shallow groundwater salt contamination (Foos, 2003; Howard and Beck, 1993; Schweiger et al., 2015). Karst springs are of particular interest because of their dual porosity aquifers, where karst conduits can facilitate rapid and unpredictable transport of some contaminated water, while the rest is retained in the aquifer longer as it travels slowly through the rock matrix. Although carbonate karst lithology is widespread, the transport of road salt contamination in karst aquifers is understudied. Temporal Na^+ and Cl^- variations in karst-hosted shallow groundwater have rarely been investigated over multiple timescales, and the mechanisms driving these variations are not well understood. Toran et al. (2009) credited epikarst storage for year-round elevated Cl^- in three karst springs, as well as for controlling specific conductivity (SpC) patterns during floods. Reisch and Toran (2014) continuously monitored a karst spring during snowmelt and found that the proportion of diffuse versus conduit flow (which have differing transport timescales for road salt contamination) depended on melting rates. However, high temporal resolution sampling of shallow groundwater impacted by road salt has not to our knowledge been combined with long-term data. Consequently, the connection between short-term variability in shallow groundwater salt concentrations and long-term geochemical trends remains unexplored. Additional work is needed to explain how salt transport and retention in shallow karst aquifers affects the timing of Na^+ and Cl^- delivery to surface waters.

In this study, karst springs were monitored to examine the geochemical effects of road salt contamination on shallow groundwater over multiple timescales including flood, seasonal, and inter-annual responses. We used 2 years of intensive water quality monitoring data from an urban and a rural karst spring to evaluate flooding behavior and salt transport pathways, assess seasonal Na^+ and Cl^- variations, and perform a Cl^- mass balance. We also examined geochemical trends for the past 20 years at the rural spring. Our main objectives were to determine: 1) how geology and land use influence road salt transport through partitioning of flow into baseflow and event water, 2) how retention and release of salt in soils affect shallow groundwater Na^+ and Cl^- concentrations, 3) what transport mechanisms govern seasonal Na^+ and Cl^- variations and annual salt retention in karst aquifers, and 4) whether long-term increases in salt concentrations occur in shallow karst aquifers that can feature rapid transport of contaminants through preferential flowpaths. Understanding the mechanisms by which Na^+ and Cl^- are transported and retained in the shallow subsurface helps explain temporal patterns of salt contamination in surface waters, where elevated concentrations are ecologically damaging.

2. Methods

2.1. Study sites

We monitored two karst springs in areas of differing land use in east-central Missouri (Fig. 1). The regional climate is temperate, with an average temperature of 13.5 °C and about 100 cm of precipitation annually (NOAA, 2017a). Snow and ice are common in the winter, with an average of 45.2 cm of snowfall per year (NWS, 2017). The regional lithology is dominated by Paleozoic carbonate rocks (USGS, 2014a) and karst features are common in this area.

We selected Blackburn Spring as our urban land use site in this study. It is located 0.7 km from a major highway in an area of mostly low intensity development (USGS, 2014b) in Webster Groves, Missouri,

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