



Boreal forest riparian zones regulate stream sulfate and dissolved organic carbon



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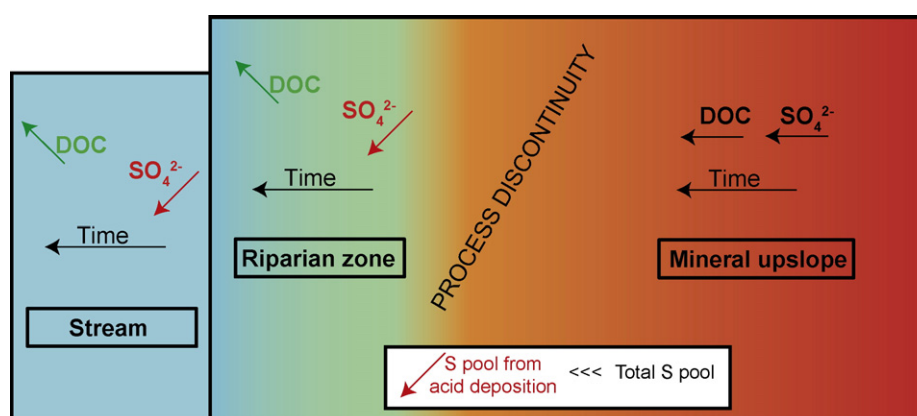
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HIGHLIGHTS

- Sulfate is still an important driver of DOC in boreal catchments.
- Riparian zone processes control stream sulfate and DOC independently of up-slope.
- Usefulness of riparian data as a means to predict stream chemistry is time-scale dependent.

GRAPHICAL ABSTRACT



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ABSTRACT

In boreal forest catchments, solute transfer to streams is controlled by hydrological and biogeochemical processes occurring in the riparian zone (RZ). However, RZs are spatially heterogeneous and information about solute chemistry is typically limited. This is problematic when making inferences about stream chemistry. Hypothetically, the strength of links between riparian and stream chemistry is time-scale dependent. Using a ten-year (2003–2012) dataset from a northern Swedish catchment, we evaluated the suitability of RZ data to infer stream dynamics at different time scales. We focus on the role of the RZ versus upslope soils in controlling sulfate (SO_4^{2-}) and dissolved organic carbon (DOC). A priori, declines in acid deposition and redox-mediated SO_4^{2-} pulses control sulfur (S) fluxes and pool dynamics, which in turn affect dissolved organic carbon (DOC). We found that the catchment is currently a net source of S, presumably due to release of the S pool accumulated during the acidification period. In both, RZ and stream, SO_4^{2-} concentrations are declining over time, whereas DOC is increasing. No temporal trends in SO_4^{2-} and DOC were observed in upslope mineral soils. SO_4^{2-} explained the variation of DOC in stream and RZ, but not in upslope mineral soil. Moreover, as SO_4^{2-} decreased with time, temporal variability of DOC increased. These observations indicate that: (1) SO_4^{2-} is still an important driver of DOC trends in boreal catchments and (2) RZ processes control stream SO_4^{2-} and subsequently DOC independently of upslope soils. These phenomena are likely

Abbreviations: RZ, riparian zone.

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occurring in many regions recovering from acidification. Because water flows through a heterogeneous mosaic of RZs before entering the stream, upscaling information from limited RZ data to the catchment level is problematic at short-time scales. However, for long-term trends and annual dynamics, the same data can provide reasonable representations of riparian processes and support meaningful inferences about stream chemistry.

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1. Introduction

In boreal forest catchments, the transfer of solutes to streams is predominantly controlled by hydrological and biogeochemical processes occurring in the near-stream riparian zone (RZ) (Gregory et al., 1991; McClain et al., 2003; Bishop et al., 2004), linking terrestrial and aquatic systems. RZs influence catchment dynamics of multiple compounds including organic carbon (Fiebig et al., 1990; Dosskey and Bertsch, 1994; Köhler et al., 2009), nitrogen (Fölster, 2000; Petrone et al., 2007), metals (Bishop et al., 1995; Pellerin et al., 2002), and persistent organic pollutants (Bergknut et al., 2011). However, information on RZ chemistry is generally limited and inferences about stream water chemistry are potentially ambiguous at different time scales because RZs are spatially heterogeneous (Ledesma et al., 2015). Hypothetically, any change in hydrological conditions, notably drought-rwetted cycles, will alter riparian groundwater table levels and transient redox conditions, influencing riparian and, subsequently, stream concentration and fluxes of redox-sensitive compounds (Lidman et al., 2011), including sulfate (SO_4^{2-}) (Devito and Hill, 1997).

Anthropogenic sulfur dioxide (SO_2) emissions and consequent SO_4^{2-} deposition triggered the acidification of terrestrial and aquatic ecosystems in much of Europe and North America (Mylona, 1996; Driscoll et al., 2001; Schöpp et al., 2003). Measures undertaken to reduce emissions in the 1980s effectively decreased SO_4^{2-} deposition and led to acidification recovery in many forest catchments (Fölster and Wilander, 2002; Watmough et al., 2005). Recovery has been slow in some areas (Driscoll et al., 2003; Evans et al., 2014; Futter et al., 2014) in part because the sulfur (S) legacy in the soil, predominantly accumulated in organic S pools (Wieder and Lang, 1988; Houle and Carignan, 1992; Giesler et al., 2005), can be oxidized to SO_4^{2-} and mobilized into surface waters after drought events (Aherne et al., 2008). Redox-mediated SO_4^{2-} pulses have been observed in many forest catchments (Laudon et al., 2004a; Eimers et al., 2008; Laudon, 2008; Vestin et al., 2008; Landre et al., 2009; Kerr et al., 2012) as well as in other ecosystems such as blanket bogs (Clark et al., 2005). However, it is not clear how long such pulses will continue as S pools are depleted in those areas and climate changes. Increased drought frequency is projected for the boreal region (Schlyter et al., 2006; Walker and Johnstone, 2014), potentially leading to an increased severity of redox-driven S acid pulses in the short-term, but also to a more rapid depletion of S pools and thus faster recovery in the medium-term.

In addition to its dominant role as a control on stream acidity in areas affected by anthropogenic acid deposition, S cycling is coupled to dissolved organic carbon (DOC) solubility (Clark et al., 2006; Evans et al., 2012). The decline in SO_4^{2-} deposition is one of the most important drivers of recent increasing DOC trends in European and North American surface waters (Evans et al., 2006; De Wit et al., 2007; Monteith et al., 2007; Erlandsson et al., 2008). Thus, SO_4^{2-} has a fundamental influence on a range of key surface water quality variables, especially in regions historically affected by anthropogenic acidification.

Here, we used ten-year climate, stream flow and chemistry, soil solution chemistry including riparian and upslope profiles in a hillslope transect, groundwater levels, and deposition data from a northern Swedish forest catchment. We aimed to test the suitability of hydrochemical data from a single riparian soil profile to infer stream dynamics at different time scales. We hypothesized that the strength of links between riparian data and stream chemistry is time-scale dependent. For this, we focus on SO_4^{2-} and its connection to DOC. The

specific objectives of the study were: (1) to quantify a long-term boreal forest catchment S mass balance, (2) to investigate the role of the RZ vs. upslope soils in controlling catchment S biogeochemistry, (3) to examine the role of riparian groundwater fluctuations in the mobilization of SO_4^{2-} , and (4) to explore the connection between SO_4^{2-} and DOC over both short-term (episodic, seasonal) and long-term (ten-year) time scales.

2. Material and methods

2.1. Catchment characterization

Västrabäcken, known as C2 (64° 15' N 19° 46' E), is a 12 ha, 100% old forest catchment located in Northern Sweden, within the Krycklan Catchment Study (Fig. 1) (Laudon et al., 2013). Mean annual air temperature is 1.9 °C and mean annual precipitation 632 mm year⁻¹ (1981–2013). Annual water outputs are divided approximately evenly between runoff and evapotranspiration (Köhler et al., 2008; Oni et al., 2013). The 4–6 week period of spring flood contributes between 40% and 60% of the total annual runoff. Nearly 100% of the tree volume is comprised of two species: Scots pine (*Pinus sylvestris*), found primarily on dry upslope podzols, and Norway spruce (*Picea abies*), common in wet low-lying areas near the stream channel. RZs are largely Histosols covered by *Sphagnum* spp. mosses, though *Vaccinium* spp. are present throughout the catchment. The gneiss bedrock is overlain by several meters of locally-derived Quaternary deposits of glacial till. The stream was straightened and deepened in the 1930s following common practices in Fennoscandia to improve drainage for forest production.

During autumn 1995, a hillslope transect monitoring site was installed near the catchment outlet to provide data across an upslope-riparian continuum (Köhler et al., 2009). This is the so-called S-transect (note that S in this case does not stand for sulfur). It consists of three soil profiles located at 4 (S4), 12 (S12), and 22 (S22) meters from the stream following the local topographic slope and thereby the likely dominant hydrological flowpaths (Fig. 1). The profiles were selected to represent, respectively, riparian peats, transitional soils, and upslope podzols within the catchment. Each profile includes six to seven ceramic suction lysimeters placed at soil depths varying between 5–10 cm and 65–90 cm (Fig. 1). Approximately 15% of the catchment, partially including the S-transect, is located below the highest postglacial coastline, which is around 250–260 m above sea level. Hence, pyrite (FeS_2) is likely present in the mineralogy of the soil C-horizon. However, inorganic S colloids have never been detected in the catchment (Skjyllberg et al., 2003) possibly because the influence of deep groundwater at the study catchment is low (Peralta-Tapia et al., 2015). Most of the S stored in the RZ is found in the form of reduced organic S, with lower proportions of sulfonate and SO_4^{2-} (Skjyllberg et al., 2003). The isotopic signature of S is denoted by deviations from the standard ratio of ^{32}S to ^{34}S measured in a meteorite collected in California and expressed as $\delta^{34}\text{S}$ (in ‰ units). In stream samples in the catchment (including high and low flow conditions), $\delta^{34}\text{S}$ is +6‰ to +8‰ (Björkvald et al., 2009), indicating negligible S contributions from weathering as the typical $\delta^{34}\text{S}$ values of S-rich bedrock in the region range from +0.3 to +0.6‰ (Klaminder et al., 2011). Since 2005, the catchment has likely been a net source of S (Björkvald et al., 2009).

Here, we studied the ten-year period 2003–2012, that is from the time when regular monitoring started in the study area until the latest

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