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Forest Ecology and Management

journal homepage: www.elsevier.com/locate/foreco

pH sensitivity of Swedish forest streams related to catchment characteristics and geographical location – Implications for forest bioenergy harvest and ash return

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article info

Article history: Received 1 February 2012 Received in revised form 14 March 2012 Accepted 16 March 2012 Available online 19 April 2012

Keywords: Acidification Catchment characteristics Land cover Forest bioenergy harvest Stream water

ABSTRACT

There is concern that whole-tree harvesting may acidify soils and surface waters. This study is a first attempt to identify the landscape types within Sweden most sensitive to acidification and potentially in need of protection from excessive biomass harvest or management treatments such as ash application. The pH sensitivity was defined from stream water pH and related to catchment characteristics and stream water acid–base chemistry. At the national level, catchments with till soils and a large proportion of forested wetlands form the most pH sensitive areas. Because of regional variability in acidification history, amount and distribution of quaternary deposits, vegetation cover, etc. pH sensitivity is determined by different landscape elements in different regions. At the regional level, lakes and forests on mineral soils were also identified as sensitive landscape types. Southwestern Sweden, historically the most acidified, is the least pH sensitive due to the high buffering capacity at low pH. In order to develop effective management guidelines across Sweden, it is most critical to better understand streams with the highest pH sensitivity, those within the pH range 5–6.2. The patchy spatial distribution of sensitive landscape types, makes it difficult from an administrative point of view to develop simple guidelines for where e.g. forest slash harvest should be restricted or where ash applications should be recommended.

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1. Introduction

For decades, excess sulfate (SO $_4^{2-})$ deposition and low weathering capacity of the Swedish bedrock and soils has depressed pH in many streams, particularly in the southwest of Sweden where the accumulated acid deposition is largest ([Bertills et al., 2007\)](#page--1-0). The severity of the pH depression in a stream depended on local catchment features such as geology, land use and amount of acid deposition. The SO $_4{}^{2-}$ deposition in Sweden peaked in the early 1980's ([Westling and Lövblad, 2000\)](#page--1-0) and covaries with climatic gradients such as precipitation and temperature and decreases towards the northeast ([Bertills et al., 2007](#page--1-0)). As a result of decreased air emissions of acid compounds, many streams are recovering from acidification, both in Sweden [\(Laudon and Hemond, 2002; Löfgren](#page--1-0) [et al., 2009](#page--1-0)) and in the northern hemisphere in general ([Skjelkvåle](#page--1-0) [et al., 2005; Monteith et al., 2007](#page--1-0)). Due to the reduction in SO $_4^{2-}$ deposition, the acidification effects connected to land use changes ([Renberg et al., 2009; Sarkkola et al., 2009](#page--1-0)) become more apparent in the surface waters.

The acid neutralizing capacity (ANC) and pH of streams are strongly impacted by differences in land cover [\(Buffam et al.,](#page--1-0) [2007\)](#page--1-0). Catchments in Sweden having a high proportion of wetlands generally have stream waters with a relatively high ANC, but lower pH values (<5) due to high levels of dissolved organic carbon (DOC) ([Cory et al., 2006\)](#page--1-0). Increased concentrations of DOC tend to decrease the pH of stream waters because DOC is a broadly defined group that includes functional groups of both weak and strong organic acids (e.g. carboxylic acid; [Hruska et al., 2003; Lydersen et al.,](#page--1-0) [2004\)](#page--1-0). In more agricultural areas, the soils tend to be of a more sedimentary origin, i.e. well sorted and fine grained. These soils have a high weathering capacity due to the large specific area ([Jury et al.,](#page--1-0) [1991\)](#page--1-0), which in turn results in a higher ANC and pH (>6) in streams draining arable land. The Swedish forest grows foremost on till soils and streams in those areas generally have fairly low ANC and pH ([Löfgren et al., 2010](#page--1-0)). Tree species also affects pH in soils and stream waters. Compared with deciduous forests, coniferous forests have a lower soil pH due to more acidic litter and also a higher interception of acidifying pollutants ([Augusto et al., 2002\)](#page--1-0). Based on a Swedish survey, the acidity seems to be highest in streams draining coniferous forests dominated by Norway spruce Picea abies (L.) Karst. ([Löfgren et al., 2010\)](#page--1-0).

Whole tree harvesting is increasing and there is concern that the large biomass removal may acidify soils and streams, or delay the recovery, in the long-term perspective due to the removal of base cations (BC) from the catchments, which potentially reduces ANC in the streams. Both steady state models such as mass

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balances [\(Akselsson et al., 2007](#page--1-0)) and dynamic models such as MA-GIC ([Aherne et al., 2008, 2012](#page--1-0)) indicate that increased removal of base cations (BC) increases acidification. This is based on the assumption that weathering cannot compensate for the loss of BC due to the biomass removal within one rotation period (approximately 60 years in S Sweden, and 120 years in N Sweden). Data on long-term (i.e. decades) changes in soil and stream water chemistry related to forest biomass removal are rare. The field studies available indicate, however, that the degree of soil acidification and base cation depletion tends to be much less than expected from theoretical model assessments ([Thiffault et al., 2011](#page--1-0) and references therein) and that excess stream water acidity often is related to nitrification some years following harvest ([Rosén et al.,](#page--1-0) [1996; Feller, 2005](#page--1-0)).

Additionally, recent research shows that the methods for estimating weathering give conflicting results and hence there is a large uncertainty in the weathering estimates ([Klaminder et al.,](#page--1-0) [2011\)](#page--1-0). This uncertainty is higher than the amount of weathered BC needed for compensation of the removal of BC in whole-tree utilization. Therefore, model simulations based on weathering estimates are very uncertain [\(Köhler et al., 2011\)](#page--1-0) for assessing whether whole-tree harvest poses a real threat for acidifying streams in the long-term.

Another approach is to relate the pH sensitivity (see below) in streams to catchment characteristics. This approach has been used successfully in the 68 km² Krycklan catchment in northern Sweden (64°23′ N, 19°23′ E) ([Ågren et al., 2010](#page--1-0)). [Löfgren et al. \(2010\)](#page--1-0) used a similar approach to detect areas where forest streams are sensitive to elevated concentrations of inorganic aluminum. By finding out what kind of streams that are most sensitive, and what type of landscape they are draining, those systems can be monitored in order to detect a potential acidifying effect of whole-tree harvest and remedies such as ash treatment can be targeted in areas where it is most needed.

It is important to differentiate between acidity and pH sensitivity. Acidity is defined by proton $(H⁺)$ activity or pH, while the pH sensitivity is defined by the ANC. In clear water with the partial pressure of carbon dioxide $(CO₂)$ in equilibrium with air (\approx 380 ppm CO₂), pH is most sensitive to additions of an acid or base at pH around 5.6 ([Stumm and Morgan, 1981\)](#page--1-0). As the difference from pH 5.6 increases, the pH sensitivity decreases. That is to say, altering the ANC would have a relatively greater effect on the final pH of stream water when the initial water pH is nearer to 5.6 than when the initial pH is further away from 5.6. On the high end of the pH spectra, bicarbonate buffers against substantial pH drops. On the low end, organic acids and aluminum buffer against large pH drops. Hence, in colored waters the picture is more complicated and depending on pH, the DOC acts either as an acid or as a base. In high pH water the organic matter dissociates and releases H^+ (acid) and in low pH water, there is an uptake of $H⁺$ by the DOC (base). Thus, there is a sigmoid relation between pH and ANC in natural waters and the shape (pH sensitivity) of this relationship is to a large extent determined by the DOC concentration ([Laudon](#page--1-0) [et al., 2001,](#page--1-0) see below).

In this article, we evaluate the pH sensitivity of forest streams in relation to the catchment characteristics of four regions of Sweden. Between regions, there are large differences in landscape features such as climate, marine influence, atmospheric SO_4^2 deposition and forest production. The objective is to test whether the pH sensitivity is related to land cover, forest status or geographical location of the streams. Additionally, possible implications for the localization of forest bioenergy harvest and ash return have been assessed.

2. Material and methods

2.1. Map variables

The database consists of stream water chemistry data from a total of 325 sampling sites located in eight different catchments and four different regions of Sweden [\(Fig. 1\)](#page--1-0). For each site, catchment characteristics and regional climatic data have been collected. The map variables were already collected in the project by [Temnerud et al. \(2007, 2009\)](#page--1-0) and the methodology is thoroughly described there. In short, the catchments delineation was determined using Arc/Info version 9.1 by using VSQ [\(Nisell et al.,](#page--1-0) [2007](#page--1-0)) and a digital elevation model (DEM). Four different sources of map variables were used to characterize the catchments; the road map (1:100,000), quaternary deposits map (1:50,000), land cover (CORINE, satellite data 50 m grid) and forest data (kNN, satellite data 50 m grid). Based on the position of the highest marine coastline (HC) according to the National Atlas of Sweden, the proportion of the catchment area above HC was calculated. In total, 114 map variables were collected.

The numbers of variables used in this study are greatly reduced ([Table 1\)](#page--1-0). First because many variables covary, e.g. the dataset produced four sets of numbers for the lake surface area (one for each map). Where several numbers/sources existed for one variable, we chose the one that was based on the best geographical resolution and therefore was the least generalized. Due to statistical constraints (few observations and not normally distributed), some variables were excluded and some were aggregated to increase the number of observations (for example; sand, silt, alluvial sediments were aggregated to the term coarse sediments). After aggregation, 24 catchment characteristics ([Table 1\)](#page--1-0) and regional climatic data from the catchments ([Table 2\)](#page--1-0) were used for the statistical analysis.

For each region, we also collected regional climatic data and deposition data on SO_4^2 and chloride (Cl⁻) from the Swedish Metrological and Hydrological Institute (SMHI) ([Table 2](#page--1-0)). In the streamwater 60%, 25%, 20% and 20% of $SO_4{}^{2-}$ is of marine origin in the southwest, south, central and north Sweden, respectively. Chloride was included in order to examine if the deposition of neutral NaCl solution could induce ion exchange in soils and affect the pH sensitivity (cf. [Löfgren et al., 2011](#page--1-0)).

Nitrogen (nitrate-N and ammonium-N roughly 50% each) and sulfur depositions are highly co-varying across Sweden [\(Löfgren](#page--1-0) [et al., 2010](#page--1-0)), which in the OPLS analysis erroneously would indicate acidification effects by the nitrogen deposition. Studies at the four Swedish IM sites located within the survey areas indicate that there are no acidification effects by inorganic nitrogen in soil water, groundwater and stream water ([Löfgren et al., 2011\)](#page--1-0). Therefore, we do not include nitrogen deposition in the analysis.

Due to methodological problems with BC deposition measurements, there are no official BC deposition figures available for Sweden. However, attempts have been made to estimate the total deposition of BC at the four Swedish IM sites [\(Löfgren et al., 2011;](#page--1-0) [Köhler et al., 2011\)](#page--1-0). Based on these estimates, ANC in deposition was approximately zero or positive in 2007 [\(Löfgren et al., 2011\)](#page--1-0). Hence, the acidification effects on stream water coupled to the deposition were primarily due to leakage of the mobile anions SO_4^2 (primarily anthropogenic) and Cl⁻ (primarily of marine origin) according to the mobile anion theory ([Reuss and Johnson,](#page--1-0) [1986\)](#page--1-0). Due to the assimilation of nitrate and ammonium by the forest ecosystem, acidification effects coupled to nitrate leakage were negligible. Additionally, there was a large leakage of organic acids affecting pH [\(Löfgren et al., 2011](#page--1-0)). Hence, including BC deposition (positive side of ANC) in the statistical analyses would not

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