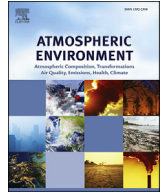




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A modified approach for estimating the aquatic critical load of acid deposition in northern Saskatchewan, Canada

Colin J. Whitfield ^{a,*}, Aidan C. Mowat ^a, Kenneth A. Scott ^b, Shaun A. Watmough ^c^a Global Institute for Water Security & Centre for Hydrology, University of Saskatchewan, 117 Science Place, Saskatoon, SK, S7N 5C8 Canada^b Saskatchewan Ministry of Environment, Regina SK, S4S 5W6 Canada^c Environmental and Resource Studies, Trent University, 1600 West Bank Drive, Peterborough, Ontario, K9J 7B8 Canada

HIGHLIGHTS

- Empirical SSWC relationships did not hold for lakes in northern Saskatchewan.
- An alternate method for SSWC critical load determination was employed.
- Accurate atmospheric deposition is needed to reduce uncertainty in CL and runoff.
- Sulphur retention appears to be an important feature in many catchments.

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ABSTRACT

Acid-sensitive ecosystems are found in northern Saskatchewan, which lies downwind of major sulphur (S) and nitrogen (N) emissions sources associated with the oil sands extraction industry. In order to protect these ecosystems against acidification, tolerance to acid deposition must be quantified. The suitability of the central empirical relationship used in the Steady-State Water Chemistry (SSWC) model to predict historical sulphate (SO₄) concentrations was investigated, and an alternate approach for determining aquatic critical loads of acidity (CL(A)) was employed for the study lakes (n = 260). Critical loads of acidity were often low, with median values of 12–16 mmol_c m⁻² yr⁻¹, with the lower value reflecting a region-specific limit for acid-neutralizing capacity identified in this study. Uncertain levels of atmospheric deposition in the region, however, are problematic for characterizing acidification risk. Accurate S and chloride (Cl) deposition are needed to identify catchment sources (and sinks) of these elements in the new approach for CL(A) calculation. Likewise, accurate depiction of atmospheric deposition levels can prove useful for evaluation of lake runoff estimates on which estimates of CL(A) are contingent. While CL(A) are low and exceedance may occur according to projected increases in S deposition in the near-term, S retention appears to be an important feature in many catchments and risk of acidification may be overstated should long-term S retention be occurring in peatlands.

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1. Introduction

The issue of acid rain is a long-standing global concern. In Canada, elevated deposition of sulphur (S) and nitrogen (N) to natural ecosystems across large areas has been well documented (Environment Canada, 2004). In southeastern Canada, widespread damage to acid-sensitive systems was observed (Beamish and Harvey, 1972; Duchesne et al., 2002; Watt, 1987). International

efforts to reduce emissions of acid precursors during the 1980s and 1990s, including the Acid Rain Control Program (1985) and Canada-U.S. Air Quality Agreement (1991), were successful and subsequent improvements in water chemistry of affected lakes have been observed, although increases in alkalinity and pH have lagged behind decreases in lake sulphate (SO₄) concentration (Jeffries et al., 2003; Whitfield et al., 2006).

Over the past twenty years, the steady-state critical load (CL) concept has been applied to assess acid-sensitivity of aquatic and terrestrial ecosystems of Canada. The critical load of acidity (CL(A)) is the maximum load of acid deposition below which damage to sensitive biota will not occur over the long-term (Nilsson and

* Corresponding author.

E-mail address: colin.whitfield@usask.ca (C.J. Whitfield).

Grennfelt, 1988). During the 21st century, much of the focus on acid rain in Canada has shifted to western provinces (e.g. Aherne and Shaw, 2010). While the western part of the country does not have the same historical legacy of pollution from acid deposition as the east, except in local areas around smelters, significant sources of S and N to the atmosphere associated with transportation and natural resource extraction are emerging in some areas. Critical load studies in the western provinces (e.g. British Columbia: Mongeon et al., 2010; Alberta: Whitfield et al., 2010b, Saskatchewan (SK): Scott et al., 2010; Manitoba: Jeffries et al., 2010) commonly identified acid-sensitive terrain (lakes or upland forest soils).

In Canada and around the world, aquatic CL(A) are most often determined using the Steady-State Water Chemistry model (SSWC: Henriksen and Posch, 2001). For lakes in northern SK, which are downwind of major emission sources of S and N associated with the oil sands industry, previous investigations of CL(A) indicated widespread acid sensitivity. The fifth percentile aquatic CL(A) was estimated to be $<6 \text{ mmol}_c \text{ m}^{-2} \text{ yr}^{-1}$ (Scott et al., 2010; Jeffries et al., 2010). Improved understanding of CL(A) in northern SK is critical, as even small uncertainties could have important consequences. Likewise, poorly quantified atmospheric acid deposition could lead to large inaccuracies when determining exceedance of the CL(A). Given the acid-sensitive nature of large numbers of lakes in the region, small changes in deposition could have a significant impact.

The principles underlying the SSWC are robust and widely applicable; however, the way in which the model is typically applied is premised on empirical relationships for lakes in northern Scandinavia (Henriksen and Posch, 2001). These relationships are known to vary among regions of northern Europe, but nonetheless the SSWC has been used widely in Canada assuming empirical relationships (for Norway) are relevant across this large and geographically diverse country. It remains unknown whether these relationships are appropriate for boreal (or other) lakes in Canada. Likewise, owing to dependence of the SSWC on observations of surface water chemistry and the assumption that SO_4 is mobile, one complicating factor when calculating CL(A) can be catchment sources (or sinks) of SO_4 . Sulphur biogeochemistry is complex, for example as conversion of the mobile SO_4 ion to organic S or sulphides can result in retention. Sources and sinks of SO_4 are often not considered, however appropriate modifications are required in instances where they occur. In the case of chloride (Cl), failure to identify local sources can also be problematic, as correction of base cation concentrations to a non-marine component can produce irreconcilable results (negative concentrations).

The central objective of this study was to improve the capacity for estimating CL(A) by considering alternate approaches for SSWC application, and to describe the uncertainty in CL(A) exceedance using available atmospheric deposition estimates. Modelled atmospheric deposition (total S, N, Cl and base cation (BC)) and lake chemistry data for 260 headwater lakes in northwestern SK were used to characterize their acid sensitivity and determine the risk of acidification. As N is a limiting nutrient in terrestrial systems of the region and lake N is very low, the focus of the study was S. As part of the study, the empirical regression approach used in SSWC for estimating historical SO_4 concentrations was tested for the first time outside Europe. Refinements for SSWC application were considered, and a new approach for SSWC application is presented.

2. Methods

2.1. Study area and data sources

Northwestern SK features a subarctic climate with a combination of snow and rain precipitation, and strongly varying seasonal temperatures. Long-term annual precipitation is between 450 mm

and 530 mm, with mean temperatures of approximately 16°C in July and -24°C in January (Environment Canada, 2015). The study area traverses three boreal ecoregions: Athabasca Plain (AP), Churchill River Upland (CRU), and Mid-Boreal Upland (MBU); all of which vary in soil type, vegetation, regional geology, and geographic landforms. Candidate headwater lakes (10–400 ha) within 300 km of the centre of the oil sands industry near Fort McMurray, Alberta (Fig. 1) were identified in alternating polygons delineated using 10° azimuth angle increments intersected at 25 km radial intervals. The study lakes ($n = 260$) were chosen randomly from the candidate pool and sampled for water chemistry during fall turnover in one or more years (2007, 2008, 2009, 2011). Further details on sample collection, analytical methods and the study area can be found in Scott et al. (2010). Lake runoff estimates according to the isotope mass balance method (Q_{imb}), and a grid-based method (Q_{grid}) were used. The isotope mass balance method takes into account site-specific hydrology, while the grid-based method represents average precipitation surplus within a 0.5° (latitude by longitude) grid (Gibson et al., 2010). Mineral soils in the region are generally well-drained (in particular for boreal shield ecoregions (AP and CRU)); deposits are largely glaciolacustrine or glaciofluvial till overlaying crystalline Precambrian bedrock. Upland forest soil plots ($n = 73$) were sampled across the study area (in the same ecoregions (ecoregion subdivision) as study lakes) and analysed according to Whitfield and Watmough (2012).

2.1.1. Atmospheric deposition

Estimates of atmospheric deposition were used in the alternate approaches for estimating CL(A) for lakes, and for identifying acidification risk. In boreal and taiga regions of Saskatchewan deposition data from monitoring programs are very limited, leading to poorly documented regional deposition patterns. Sulphur deposition was estimated according to the on-line chemical transport model GEM-MACH run for meteorological year 2013 (Makar et al., 2015). Estimates of mapped wet (Ro and Vet, 2003) and dry (Vet and Shaw, 2004) BC and Cl deposition for the period 1994–1998 interpolated to a common grid resolution for total deposition were available for the study catchments; these data were required in the alternate approach for SSWC application described below.

2.2. Critical loads of acidity

2.2.1. Steady-state water chemistry model

The Steady State Water Chemistry (SSWC) model (Henriksen and Posch, 2001) can be used to calculate CL(A) in an empirical fashion using water chemistry and runoff data. The CL(A) was calculated for each study lake with sea-salt corrections for SO_4 and BC (calcium (Ca), magnesium (Mg), potassium (K), and sodium (Na)) concentrations in lakes. Observed lake Cl is used for sea-salt corrections, with correction factors of 0.104 for SO_4 , 0.04 Ca, 0.21 for Mg, 0.86 for Na and 0.109 for K (Evans et al., 2001). Where multiple observations were available, the average lake chemistry condition was used to calculate the CL(A):

$$\text{CL(A)} = ([\text{BC}^*]_0 - \text{ANC}_{\text{limit}}) \times Q_{\text{imb}} \quad [1]$$

where Q_{imb} is the site-specific watershed runoff (m y^{-1}), and $[\text{BC}^*]_0$ is the sea-salt corrected (denoted by $*$) pre-acidification BC concentration. Lake-specific $\text{ANC}_{\text{limit}}$, the lowest (charge balance) acid neutralizing capacity (ANC) above which biota do not respond adversely, was set to $10 \mu\text{mol}_c \text{ L}^{-1}$ (Scott et al., 2010), then scaled to account for organic acidity according to Lydersen et al. (2004):

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