Environmental Pollution 192 (2014) 91–103

Contents lists available at ScienceDirect

Environmental Pollution

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

Wetlands as long-term sources of metals to receiving waters in mining-impacted landscapes

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ARTICLE INFO

Article history:
Received 13 February 2014
Received in revised form
18 April 2014
Accepted 5 May 2014
Available online 4 June 2014

Keywords:
Metals
Deposition
Wetlands
Mining
Acidification

ABSTRACT

Wetlands are prevalent in the Sudbury, Ontario region and often operate at the interface between terrestrial and aquatic ecosystems, modifying water chemistry and potentially affecting the recovery of impacted lakes. The deposition of metals and sulphur in Sudbury in 2010–2012 was far below that reported in the 1970's, but still higher than background values. Wetlands in the area have accumulated large quantities of metals, and high concentrations of these metals in streams occurred primarily in response to SO_4-related acidification events or associated with high dissolved organic carbon production in early summer. Concentrations of most metals in streams exceeded provincial guidelines and fluxes of some metals from catchments exceeded deposition inputs to lakes by as much as 12 times. The release of metals long after emissions reductions have been achieved must be considered in ecosystem recovery studies, particularly as dry conditions may become more prevalent in boreal regions affected by mining.
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1. Introduction

Wetlands retain atmospherically deposited metals through ionic exchange with organic matter (Brown et al., 2000; Crist et al., 1996) and are often used as records of past deposition (Cole et al., 1990; Martínez-Cortizas et al., 1999; Shotyk et al., 1998) or are constructed to improve downstream water quality (Sobolewski, 1999). It is estimated that 25% of the world's wetlands are in Canada's boreal region, and a wide range of wetland types exist in this area, although fens and bogs are most common (Wells et al., 2011). Many of these boreal wetlands have been and/or will be subject to elevated metal deposition because of mining and smelting activity (Wells et al., 2011). Elevated metal concentrations in peat have been reported in wetlands in Ontario (Gignac and Beckett, 1986; Taylor and Crowder, 1983), Quebec (Kettles and Bonham-Carter, 2002), and Manitoba in Canada (Outridge et al., 2011), as well as in parts of Finland (Nieminen et al., 2002) and the Czech Republic (Novak et al., 2011). These wetlands are often at the interface between terrestrial and aquatic systems and can affect surface water quality in downstream systems.

Although wetlands retain metals, they can also become sources of metals to surface waters under certain conditions. For example, reductions in pH can result in a release of metals from peat through competition of binding sites between metal cations and protons (Brown et al., 2000; Gambrell et al., 1991). When dry periods result in a lowering of the water table, sulphur (S) can become oxidized to sulphate (SO_4) and subsequent rewetting events then lead to the formation of sulphuric acid (Devito and Hill, 1999; Eimers et al., 2007, 2003). The associated protons compete for binding sites and can result in the release of metals (Juckers and Watmough, 2013; Lucassen et al., 2002; Tipping et al., 2003), which has been documented hundreds of kilometres from the originating smelters (Adkinson et al., 2008; Landre et al., 2009). Increases in ionic strength in response to other sources of ions (e.g.: road salts) can also affect metal binding capacity (Amrhein et al., 1992; Löfgren, 2001). Some metals will also preferentially bind to dissolved organic ligands and form mobile organo-metal complexes, so seasonal increases in dissolved organic carbon (DOC) concentration can result in increased mobility of metals (Antoniadis and Alloway, 2002; Cory et al., 2006; Olivie-Lauquet et al., 2001). Furthermore, wetlands can be significant sources of metals derived from within the catchment (e.g.: Fe, Mn, and Al) during periods of high groundwater infiltration (Shotyk, 1988).

The mining region of Sudbury, Canada is one of the most widely-studied boreal regions in the world in terms of acid- and metal-impacts, emissions reductions, and subsequent wide-scale ecosystem recovery. Nearly a century of smelter emissions

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http://dx.doi.org/10.1016/j.envpol.2014.05.009
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resulted in elevated deposition of S and metals in the region, including Cu, Ni, Co, Zn, Fe, Pb, Cr, and Cd (Adamo et al., 2002; Hazlett et al., 1984; Hutchinson and Whitby, 1977). Major reductions (>95%) in atmospheric emissions of pollutants have occurred since the 1970s, but soils and surface waters remain contaminated in many areas (Keller et al., 2007; Meadows and Watmough, 2012; Nriagu et al., 1998). Improvements in terrestrial and aquatic ecosystems are well documented in Sudbury (Keller et al., 2007; McCall et al., 1995), and research in the area has contributed to our understanding of natural recovery processes and restoration efforts elsewhere (Kozlov and Haukioja, 1999; Schindler, 1997). There are over 33,000 ha of wetlands in the city of Greater Sudbury, many of which are connected to the 47,000 ha of lakes, rivers, and streams in the area (Monet, 2013). Although we know that wetlands close to the smelters were contaminated with metals in the 1980's (Gignac and Beckett, 1986; Taylor and Crowder, 1983) and that they play an important role in modifying surface water quality, the biogeochemistry of these wetlands and their impact on water quality has remained relatively understudied.

The objective of this study was to examine the biogeochemical variability of mining-impacted catchments with a range of characteristics to identify key metal-regulating processes that may be occurring regionally. We focus on catchments with wetland outflows draining into lakes, because of the potential for metals released from these wetlands to affect aquatic communities in receiving waters. Four questions are addressed: (Q1) How does current deposition of smelter-related metals and S compare with historical deposition in Sudbury? (Q2) What are the biogeochemical processes governing the spatial and temporal variability of pore water chemistry? (Q3) Is the stream outflow chemistry from these catchments related to pore water chemistry, and does this result in high concentrations of metals in outflow streams? And finally (Q4), are these wetland-influenced catchments currently losing or retaining metals? The implications of these results are discussed in terms of potential toxicity and metal loads to receiving waters, and the information gained will be applicable to other boreal regions where mining-related impacts are ongoing and expanding and climate is also changing.

2. Methods

2.1. Site selection and characterization

A preliminary survey of vegetation and annual water table fluctuation in 29 wetlands was conducted in the region in 2010. Peat samples were collected from a subset of 18 of the wetlands (primarily poor or transitional fens) revealing a range in metal and nutrient content. We then selected six wetlands that varied naturally in wetland and catchment characteristics, but that were considered representative of the fens in the region. Each selected wetland was in a watershed sub-catchment with a stream draining directly into a lake (henceforth termed "wetland-influenced catchments") (Table 1). Two of the fens (D4 and D5) had shallow organic soil layers (<30 cm) overlaying mineral soils and had lower organic matter content. These more minerotrophic wetlands were dominated by a variety of grasses (primarily *Muhlenbergia uniflora* at D4 and *Calamagrostis canadensis* at D5) with some terrestrial grasses at D5 as well (mostly *Schizachne purpurascens*). The other four fens had deep organic layers (>50 cm) and were dominated by shrubs (primarily *Chamaedaphne calyculata*), sedges, and rushes.

The catchments were delineated with Whitebox Geospatial Analysis Tools (Lindsay, 2012) using digital elevation models compiled in ArcMap (ESRI ArcGIS version 9.3, Redlands, CA) from Ontario Basic Mapping and Ontario Ministry of the Environment Forest Inventory data. Wetland areas and stream lengths were interpreted from 1:40,000 aerial photos (taken by the City of Greater Sudbury) and corrected with in-field data. The catchments vary in distance from the active Copper Cliff smelter (7.3–13.6 km), total area (11.0–100 ha), proportional wetland area (1.39–20.8%), and length of the outflow to the lake (45.0–156 m) (Table 1, Fig. 1). They are all in an area of heavy historical smelter-related deposition but the extent of damage to upland vegetation varies among sites. Catchments LU, D4, and D5 are in an area that was classified as barren to semi-barren, impacted primarily by fumigation from the active Copper Cliff smelter and the now-closed Coniston smelter (McCall et al., 1995). The Daisy Lake catchments (D4 and D5) suffered a loss of pine-dominated forest in the 1940's followed by

Table 1
Catchment characteristics and mean (SD) concentrations of metals, cations, and nutrients in the wetlands of six catchments. Five peat samples were taken at 0–15 cm depth and are compared to the medians and ranges from a 1981 survey of 25 wetlands in the Sudbury area (Taylor and Crowder, 1983). The pH is given as a mean from pore water samples collected across the two study years.

	C1	C2	BR	LU	D4	D5	1981 Median (Range)
Metals and other cations (mg/kg)							
Al	5345 (1738)	1716 (50.8)	10456 (2589)	3356 (777)	20950 (4503)	18618 (2683)	
Co	9.35 (3.32)	8.36 (0.200)	10.4 (0.888)	17.3 (4.35)	11.8 (1.52)	12.0 (1.57)	
Cu	844 (179)	635 (83.3)	1039 (143)	1238 (428)	711 (183)	512 (116)	371 (21–6912)
Fe	7656 (2660)	8179 (2177)	3091 (108)	7866 (733)	15290 (812)	17272 (4137)	12452 (2090–45448)
Mn	39.2 (5.88)	29.1 (0.590)	42.5 (7.75)	47.1 (13.8)	62.5 (22.6)	92.4 (12.8)	188 (25–573)
Ni	472 (193)	348 (11.2)	540 (47.2)	920 (231)	275 (47.2)	429 (186)	481 (38–9372)
Zn	42.9 (15.7)	38.7 (8.08)	43.0 (12.6)	64.7 (20.2)	41.3 (14.0)	45.2 (2.14)	92 (23–481)
Na	507 (106)	705 (84.0)	572 (57.2)	1245 (336)	491 (194)	468 (28.2)	
K	674 (96.1)	834 (240)	591 (61.2)	1019 (171)	1044 (399)	1117 (197)	
Ca	2524 (603)	5432 (1043)	1741 (294)	5837 (2326)	723 (159)	1727 (403)	15575 (2650–38068)
Mg	586 (127)	385 (13.5)	384 (42.9)	403 (68.6)	1721 (978)	2315 (533)	2775 (683–7220)
Nutrients and pH							
% C	31.8 (4.34)	48.4 (1.24)	39.6 (2.28)	49.2 (0.490)	17.3 (3.40)	12.1 (5.83)	
% N	1.31 (1.04)	1.73 (0.234)	2.11 (0.204)	1.86 (0.230)	1.34 (0.212)	0.713 (0.260)	
% S	0.390 (0.339)	0.695 (0.159)	0.756 (0.093)	0.675 (0.094)	0.238 (0.065)	0.224 (0.103)	
pH	4.85 (0.436)	4.08 (0.566)	4.83 (0.459)	4.20 (0.623)	4.22 (0.460)	4.86 (0.480)	NA (3.6–5.6)
Catchment Characteristics							
Area (km^2)	1000	275	268	110	324	292	
% Wetland	13.5	10	5.15	20.8	1.39	1.64	
Stream length (m)	145	92.0	45.0	156	48.0	148	
Dist. to smelter (km)	12.8	12.2	11.6	7.30	13.6	13.2	
Receiving Lake	Clearwater	Clearwater	Broder 23	Laurentian	Daisy	Daisy	
Lake Area (ha)	76.0	76.0	36.9	157	36.6	36.6	

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