Atmospheric Environment xxx (2016) 1–13

Contents lists available at ScienceDirect

Atmospheric Environment

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

Impediments to recovery from acid deposition

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HIGHLIGHTS

- Precipitation and stream chemistry measured at Plastic Lake for 30 years.
- Reduction in SO_2 and NO_2 emissions reflected in lower SO_4^{2-} and NO_3^- in precipitation.
- Number of drought days correlated with peak fall SO_4^{2-} in the wetland-draining stream.
- Fall peaks in stream Al and depressions in pH in the 2000s are as high as the 1980s.
- Soil acidification and excess S export delay recovery from acid deposition.

ARTICLE INFO

Article history:
Received 24 November 2015
Received in revised form
24 February 2016
Accepted 8 March 2016
Available online xxx

Keywords:
Acid deposition
Forested catchments
Streams
Recovery
Base cations
Drought
Aluminum

ABSTRACT

In response to large reductions in sulphur (S) emissions over the past 30 years, sulphate (SO_4^{2-}) concentrations in precipitation at Plastic Lake in south-central Ontario, Canada, have declined by more than 70%. More recent decreases in NO_x emissions have similarly led to a reduction in nitrate deposition (NO_3^-) and consequently the pH of bulk precipitation has increased by approximately 0.8 pH units since 1980. Despite the large decrease in acidic deposition, chemical recovery of the streams, as measured by an increase in pH and decrease in aluminum (Al), has been much less than expected, primarily due to losses of base cations from the shallow, base-poor soils. While nitrogen (N) is almost totally retained within the terrestrial catchment, S export continues to exceed inputs measured in bulk deposition and during the early part of the record approximately 70% of the anions in streams were buffered by calcium (Ca^{2+}) and magnesium (Mg^{2+}) compared with only 60% in 2011/12. In the wetland-draining stream (PC1), peak depressions in stream pH and peaks in SO_4^{2-} and Al concentration in the fall are significantly positively correlated with annual drought days defined as the number of days when stream flow ceases. Even though reductions in SO_2 and NO_x emissions in Canada have resulted in large improvements in precipitation chemistry, the combined influence of soil acidification and climate-mediated biogeochemical processes occurring in wetlands cause acidification-related issues to persist. Forecasting the longer-term response of soils and surface waters in light of these observations is required to fully assess the need for further emission reductions.

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1. Introduction

Acid deposition has been a major environmental concern in Europe and eastern North America since the 1960s–1980s (Likens et al., 1996). In both regions, large numbers of lakes were found to be acidic and there were widespread concerns over adverse impacts on forested and aquatic ecosystems (Likens et al., 1979). Beginning in the late 1960s various policy actions were implemented to greatly reduce SO_2 emissions and more recently NO_x emissions (Environment Canada, 2004). Large reductions in SO_4^{2-} deposition were expected to promote chemical and biological recovery in aquatic ecosystems, but initial responses to reduced emissions were mixed (Clair et al., 2011; Jeffries et al., 2003; Stoddard et al., 1999). Both Stoddard et al. (1999) and Skjelkvale et al. (2005) for example, synthesized trends in aquatic chemistry in Europe and eastern North America and found that in many regions the decline in S deposition was greater than the corresponding decline in surface water SO_4^{2-}. Furthermore, the increase in pH and ANC (acid neutralizing capacity) was often less than expected and was attributed to corresponding declines in base cation export and increasing dissolved organic carbon (DOC) concentrations (Evans et al., 2006; Lawrence et al., 2011; Monteith

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http://dx.doi.org/10.1016/j.atmosenv.2016.03.021
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Please cite this article in press as: Watmough, S.A., et al., Impediments to recovery from acid deposition, Atmospheric Environment (2016), http://dx.doi.org/10.1016/j.atmosenv.2016.03.021

et al., 2007).

More recently, Strock et al. (2015) reported that there has been an acceleration in the rate of recovery from acidification in the northeastern United States. This assessment was primarily based on changes in surface water SO_4^{2-} concentrations as Strock et al. (2015) reported that both ANC and pH continue to have variable trends and the authors concluded that continued monitoring is needed to ascertain the potentially antagonistic or additive interactions between reduced S emissions and changes in the frequency and intensity of extreme wet and dry years. Indeed, Mitchell and Likens (2011) suggested that S biogeochemistry was shifting from atmospheric dominance to climatic regulation and as atmospheric S deposition declines, climate will play an increasingly important role in regulating S budgets and the amount of SO_4^{2-} mobilized from internal watershed sources (Mitchell et al., 2013). In Quebec, Houle et al. (2010) reported that improvements in acid base status at 47 lakes could not be attributed solely to a decrease in SO_4^{2-} deposition, but that climate variations were more important. In particular, Houle et al. (2010) noted that higher annual air temperatures were more often correlated with the alkalinity and pH of lakes than were rates of decreasing SO_4^{2-} deposition. Similar influences of temperature on surface water chemistry have been reported for alpine lakes in Europe (Sommaruga-Wögrath et al., 1997).

In Ontario, Reid and Watmough (2016) found that declines in calcium (Ca^{2+}) and magnesium (Mg^{2+}) concentrations were similar to decreases in SO_4^{2-} concentration at 104 lakes between the 1980s and 2000s resulting in minimal improvement in surface water chemistry. Watershed studies examining ion mass balance in the early 1980s and late 1990s suggest that export of base cations from acid-sensitive catchments in Ontario and elsewhere exceeds the combined inputs from atmospheric deposition and mineral weathering (Watmough et al., 2005). In addition, summer droughts result in elevated SO_4^{2-} export primarily due to oxidation of S in organic soils, further contributing to the delay in recovery from acidic deposition (Kerr et al., 2012). The impact of summer drought on stream SO_4^{2-} has been shown to persist for several months (Eimers et al., 2008a), although peaks in Al and depressions in pH, which are critical to biota, may occur over relatively short periods (days) (Neff et al., 2009). These short-lived acidic episodes could be missed in routine monitoring programs that have weekly, fortnightly or monthly sampling regimes.

In light of these observations, the objective of the present study was to evaluate long-term (30+year) patterns in bulk deposition and stream chemistry at a regionally representative forested watershed in central Ontario to determine whether the large reductions in SO_2 and more recently, NO_x emissions have prompted chemical recovery. A major focus of this work was to provide insight into the biogeochemical processes that may be delaying chemical recovery of aquatic ecosystems.

2. Methods

2.1. Study area

The two study sites are located in the Plastic Lake watershed in Haliburton County on a southern extension of the Precambrian Shield in Ontario, Canada (45°11′ N, 78°50′ W), and is typical of acid-sensitive watersheds in this region (Fig. 1). The 32 ha headwater-lake is fed by one major stream (PC1) and 6 ephemeral streams, which together drain an area of approximately 90 ha. Plastic Lake-1 (PC1) is the largest sub-catchment (23.3 ha) in the watershed, and is characterized by thin (average depth 0.37 m to bedrock) orthic humo-ferric and ferro-humic podzols (Watmough and Dillon, 2004), formed from thin, sandy basal tills. The forest at Plastic Lake is dominated by white pine (*Pinus strobus*), eastern hemlock (*Tsuga canadensis*), red oak (*Quercus rubra*), and red maple (*Acer rubrum*) in the upland areas, and by white cedar (*Thuja occidentalis*) and black spruce (*Picea mariana*) in the swamp. A 2.2 ha conifer-*Sphagnum* swamp is located approximately 50 m upstream of the catchment outflow and more than 85% of the runoff from the Plastic Lake catchment drains through the swamp before discharging to the lake. A short (<250 m) ephemeral stream (PC1-08) drains the northeastern upland part of PC1 before discharging into the swamp.

2.2. Bulk precipitation

Deposition data were obtained from collections of bulk deposition, defined as that caught in a continuously open, 0.25 m^2 collector. The collectors were fitted with Teflon-coated funnels that are screened (80 μm Nitex mesh) to prevent contamination by insects and debris. Precipitation samples were removed from collectors when there was sufficient volume for chemical analyses, typically weekly. Analytical methods for Ca^{2+} (CA8), Mg^{2+} (MA4) K^+ (PH1), Na^+ (SC1), SO_4^{2-} (SH13), NO_3^- (NB1) and NH_4^+ (NA1) are outlined in detail in Ontario Ministry of Environment (1983) and were unchanged during the study period. Briefly, base cations were measured by atomic absorption spectrophotometry, SO_4^{2-} by ion chromatography and NH_4^+ and NO_3^- were determined through colorimetry. Precipitation volume and volume-weighted concentrations were calculated for each water year (June 1—May 31). On the rare occasions when data were missing the average precipitation values from three nearby (within 50 km) bulk collectors (or stream flow from nearby weirs with highly correlated discharge relationships) were used.

2.3. Stream export

Water level or stage was recorded continuously at a V-notched weir installed at each catchment outflow (PC1 and PC1-08), and daily stream discharge ($m^3\ d^{-1}$) was computed using established stage—discharge relationships. Stream data were unavailable for water years 2003/04 and 2004/05 at PC1 and PC1-08 due to weir replacement and upgrades, and also between 1995/96 and 1999/00, inclusive at PC1-08 due to monitoring cutbacks. Monitoring of PC1-08 stream chemistry began in 1983. Water samples for chemical analyses were collected at the weir at regular intervals (at least biweekly) when there was flow, although sampling was more frequent during the spring melt period. Water samples were filtered through 80 μm Nitex mesh in the field, and transported to the laboratory for chemical analyses in insulated containers. Filters (Nitex mesh) were rinsed in deionized water in the laboratory and rinsed in stream water immediately prior to sampling. Stream samples were analyzed for Ca^{2+}, Mg^{2+}, K^+, Na^+, SO_4^{2-}, NO_3^- and NH_4^+ as defined above. In addition, samples were analyzed for Kjeldahl N (TKN) (NE1) and dissolved organic carbon (DOC) (CB13) and Al (MD21) using standard methods (Ontario Ministry of Environment (1983)). The contribution of organic acids to the ion balance was estimated from measured DOC concentrations and pH (Oliver et al., 1983) and ANC (acid neutralizing capacity) was estimated as the difference between base cations (Ca^{2+}, Mg^{2+}, Na^+, K^+) and acid anions (SO_4^{2-}, NO_3^-, Cl^-).

2.4. Intensive stream monitoring: climate effects analysis

Stream samples at PC1 were collected daily (once every 24 h) between May 1st and October 31st, 2012 using an automated ISCO™ sampler housed in a heated hut above the v-notch weir, where streamflow is continuously gauged. The ISCO™ sampling

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