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The response of soil and stream chemistry to decreases in acid deposition in the Catskill Mountains, New York, USA[☆]



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ABSTRACT

The Catskill Mountains have been adversely impacted by decades of acid deposition, however, since the early 1990s, levels have decreased sharply as a result of decreases in emissions of sulfur dioxide and nitrogen oxides. This study examines trends in acid deposition, stream-water chemistry, and soil chemistry in the southeastern Catskill Mountains. We measured significant reductions in acid deposition and improvement in stream-water quality in 5 streams included in this study from 1992 to 2014. The largest, most significant trends were for sulfate (SO_4^{2-}) concentrations (mean trend of $-2.5 \mu\text{eq L}^{-1} \text{yr}^{-1}$); hydrogen ion (H^+) and inorganic monomeric aluminum (Al_{im}) also decreased significantly (mean trends of $-0.3 \mu\text{eq L}^{-1} \text{yr}^{-1}$ for H^+ and $-0.1 \mu\text{eq L}^{-1} \text{yr}^{-1}$ for Al_{im} for the 3 most acidic sites). Acid neutralizing capacity (ANC) increased by a mean of $0.65 \mu\text{eq L}^{-1} \text{yr}^{-1}$ for all 5 sites, which was 4 fold less than the decrease in SO_4^{2-} concentrations. These upward trends in ANC were limited by coincident decreases in base cations ($-1.3 \mu\text{eq L}^{-1} \text{yr}^{-1}$ for calcium + magnesium). No significant trends were detected in stream-water nitrate (NO_3^-) concentrations despite significant decreasing trends in NO_3^- wet deposition. We measured no recovery in soil chemistry which we attributed to an initially low soil buffering capacity that has been further depleted by decades of acid deposition. Tightly coupled decreasing trends in stream-water silicon (Si) ($-0.2 \mu\text{eq L}^{-1} \text{yr}^{-1}$) and base cations suggest a decrease in the soil mineral weathering rate. We hypothesize that a decrease in the ionic strength of soil water and shallow groundwater may be the principal driver of this apparent decrease in the weathering rate. A decreasing weathering rate would help to explain the slow recovery of stream pH and ANC as well as that of soil base cations.

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1. Introduction

Acid deposition across the northeastern United States is mainly a result of emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) that originate largely from the combustion of fossil fuels (Driscoll et al., 2001). Acid deposition has caused acidification of soils and surface waters in sensitive ecosystems in the region resulting in deleterious effects to aquatic and terrestrial biota. Across eastern North America acid deposition generally peaked in the late 1970s and the dominant strong acids sulfate (SO_4^{2-}) and nitrate (NO_3^-) have declined sharply by 50 to more than 90% since the 1980s (Burns et al., 2011). Emissions regulations implemented

as part of the US Clean Air Act have largely driven these declines in acid deposition. The EPA's Long-Term Monitoring Program (LTM) was designed to monitor the response of surface water chemistry to implementation of Title IV of the 1990 Clean Air Act Amendments in regions with high sensitivity to acid rain (<https://www.epa.gov/airmarkets/clean-air-markets-monitoring-surface-water-chemistry>).

The Catskill Mountains of New York is a region with demonstrated sensitivity to acid rain, where surface waters have been acidified and harmful effects to aquatic biota have been observed (Baldigo and Lawrence, 2001; Siemion et al., 2014; Stoddard and Murdoch, 1991). The Neversink River and its headwaters are the most sensitive waters in the region with both chronically and episodically acidic streams (Lawrence, 2002; Baldigo and Lawrence, 2001; Wigington et al., 1996a). Past investigations have failed to find strong evidence of recovery in stream acid-base chemistry or in aquatic biota in the Neversink River basin despite sharp declines in acid deposition in the region (Burns et al., 2006, 2008). Stream

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SO_4^{2-} concentrations have declined in parallel with declines in precipitation SO_4^{2-} concentrations (Murdoch and Stoddard, 1993; Stoddard and Murdoch, 1991), but stream NO_3^- concentrations have not shown a parallel decline with those of decreasing NO_3^- concentrations in precipitation (Burns et al., 2006). Base cations (Calcium – Ca^{2+} ; Magnesium – Mg^{2+} ; Potassium – K^+ ; Sodium – Na^+) showed decreasing trends in surface waters in this region while dissolved organic carbon (DOC) increased from 1992 to 2001 (Burns et al., 2006). These patterns of decreasing base cations and increasing organic acidity contributed to the lack of recovery in stream acid-base chemistry in the region through 2001.

High-flow events (storms and snowmelt) have also been shown to cause sharp reductions in acid neutralizing capacity (ANC) in Catskill Mountain streams (Wigington et al., 1996a). These reductions were associated with pulses of NO_3^- and inorganic monomeric aluminum (Al_{im}). Pulses of NO_3^- and Al_{im} were particularly acute during major hydrologic events such as winter and spring melts (Wigington et al., 1996a). Even as streams recover from chronic acidification, episodic acidification may continue to cause sharp declines in ANC and increases in Al_{im} during large hydrologic events such as spring melt (Lawrence, 2002).

Several previous studies in the northeastern U.S. have identified depletion of soil exchangeable base cations as a limiting factor on surface water recovery from acid deposition (Lawrence et al., 2015; Likens et al., 1996; Warby et al., 2009). Mineral soils in the Catskills are generally highly acidic with base saturation values as low as 2% in the southern part of the Catskills that includes the Neversink basin (Siemion et al., 2014). The long-term depletion of soil base cations coupled with little or no change in stream water NO_3^- concentrations may delay the recovery of stream water from decreases in acid deposition. Despite these potential negative feedbacks there has been some promising evidence of soil improvement across the northeast that may signal the beginning of a more robust recovery from stream acidification (Lawrence et al., 2012, 2015).

The purpose of this study was to examine the response of stream water and soil chemistry to reductions in acid deposition in 5 Catskill Mountains watersheds that have been the focus of previous acid deposition research (Burns et al., 2006; Murdoch and Stoddard, 1992; Stoddard and Murdoch, 1991). Although previous work has documented reductions in stream water SO_4^{2-} concentrations in the region, decreases in base cations, increases in organic acidity, and little change in NO_3^- concentrations have resulted in a muted increase in stream water pH and ANC (Burns et al., 2006). We also examined trends in stream chemistry during spring melt and changes in chemistry throughout the range in flow conditions in each watershed. Soil resampling was used to determine whether there is evidence of recovery in watershed soils that may indicate a more robust, ecosystem-scale recovery. The broad aim of this work was to evaluate whether declines in S and N emissions have resulted in recovery of stream water and soil chemistry that may promote the recovery of acid-sensitive aquatic biota and the terrestrial ecosystem.

2. Methods

This study examines 23 years of atmospheric and stream water chemical data from water years 1992 through 2014 (a water year is defined as October 1 to September 30 of the year in which it ends). Field data collection methods and laboratory analyses are detailed below.

2.1. Site description

The watersheds included in this study were the Neversink and Rondout reservoir watersheds (Fig. 1) located in the southeastern

portion of the Catskill Mountains in southeastern New York State. This area of the Catskill Mountains is particularly vulnerable to the effects of acid deposition because it is characterized by thin soils overlying sedimentary bedrock composed of shale, siltstone, sandstone and conglomerate of the upper Walton and Slide Mountain Formations (Arscott et al., 2006; Rich, 1934; Ver Straeten, 2013). The sandstones, conglomerates, and siltstones are composed primarily of slow weathering quartz with some sedimentary and metamorphic rock fragments (Ver Straeten, 2013). The shale and related mudstones are composed primarily of illite and chlorite clay minerals that are potassium and magnesium/iron aluminosilicates respectively (Ver Straeten, 2013). Soils in these watersheds are primarily moderately to highly acidic Inceptisols (Arscott et al., 2006). The soils have a low cation exchange capacity because of the base-poor parent material that has been further diminished by several decades of acid deposition (Lawrence et al., 1999; Siemion et al., 2014).

Data from 5 watersheds were included in this study (Fig. 1; Table 1). The watersheds ranged in size from 199 ha to 17,249 ha. Stream water discharge and water quality have been monitored from 1992 to 2014. These same watersheds have been included in a series of studies of stream acidification and recovery during the last 20 years (Baldigo and Murdoch, 1997; Burns et al., 1998, 2006; Lawrence, 2002; Lawrence et al., 2000; Murdoch and Stoddard, 1992; Stoddard and Murdoch, 1991; Wigington et al., 1996b).

2.2. Field methods

Wet-only precipitation chemistry samples were collected weekly at the Biscuit Brook National Atmospheric Deposition Program (NADP) sampling station (NY68; Fig. 1). Precipitation chemical data and wet deposition loads were obtained from the NADP website (<http://nadp.isws.illinois.edu/data/sites/siteDetails.aspx?net=NTN&id=NY68>; accessed on September 30, 2015). Precipitation amount was recorded at the Biscuit Brook NADP station. Stream water chemistry grab samples were collected bi-weekly or monthly throughout the study period and discrete storm samples were collected using automated samplers. Samples from the automated samplers were retrieved approximately every other week. Approximately 6 storms were sampled each year at each station with a total of 5–6 samples per storm. Stream water discharge was calculated at 15 min intervals using a stage-discharge rating curve developed for each station using standard U.S. Geological Survey (USGS) methods (Rantz, 1982).

Soil chemistry samples were collected at 3 locations in the Fall Brook watershed in 2001 and again in 2011 and 6 locations in the Winnisook watershed in 1993 and again in 2012 (Fig. 1) to determine whether there was a change in soil chemistry during the 2 periods. The data from those two soil resampling efforts are included in this study to investigate what effect the large decrease in acid deposition has had on soil chemistry in the Catskill Mountains. At each location in the Fall Brook watershed a central pit was excavated and a full description was made of each soil horizon. A sample of each soil horizon was collected for soil chemical analyses. In addition, 4 “satellite” pits were excavated into the B-horizon surrounding and approximately 10 m from the central pit to provide a measurement of the chemical variability at each sampling location. Samples from the Oa/A and upper B-horizon were collected in each satellite pit. In the Winnisook watershed central pits were excavated, but no satellite pits were excavated. The original samples were collected in 1993 (Johnson et al., 2000; Ruiz, 1995), a representative sample of Oa horizon soil was collected by excavating the entire depth to mineral soil. A single mineral soil sample was collected at each site by compositing 125-mL subsamples taken at 10-cm intervals to a maximum depth of 50 cm. We

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