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# High elevation watersheds in the southern Appalachians: Indicators of sensitivity to acidic deposition and the potential for restoration through liming

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ABSTRACT

Southern Appalachian high elevation watersheds have deep rocky soils with high organic matter content, different vegetation communities, and receive greater inputs of acidic deposition compared to low elevation sites within the region. Since the implementation of the Clean Air Act Amendment in the 1990s, concentrations of acidic anions in rainfall have declined. However, some high elevation streams continue to show signs of chronic to episodic acidity, where acid neutralizing capacity (ANC) ranges from 0 to 20 $\mu eq\,L^{-1}$. We studied three 3rd order watersheds (North River in Cherokee National Forest, Santeetlah Creek in Nantahala National Forest, and North Fork of the French Broad in Pisgah National Forest) and selected four to six 1st order catchments within each watershed to represent a gradient in elevation (849–1526 m) and a range in acidic stream ANC values (11–50 $\mu eq\,L^{-1}$). Our objectives were to (1) identify biotic, physical and chemical catchment parameters that could be used as indices of stream ANC, pH and Ca:Al molar ratios and (2) estimate the lime required to restore catchments from the effects of excess acidity and increase base cation availability. We quantified each catchment's biotic, physical, and chemical characteristics and collected stream, O-horizon, and mineral soil samples for chemical analysis seasonally for one year. Using repeated measures analysis, we examined variability in stream chemistry and catchment characteristics; we used a nested split-plot design to identify catchment characteristics that were correlated with stream chemistry. Watersheds differed significantly and the catchments sampled provided a wide range of stream chemical, biotic, physical and chemical characteristics. Variability in stream ANC, pH, and Ca:Al molar ratio were significantly correlated with catchment vegetation characteristics (basal area, tree height, and tree diameter) as well as O-horizon nitrogen and aluminum concentrations. Total soil carbon and calcium (an indicator of parent material), were significant covariates for stream ANC, pH and Ca:Al molar ratios. Lime requirement estimates did not differ among watersheds but this data will help select catchments for future restoration and lime application studies. Not surprisingly, this work found many vegetation and chemical characteristics that were useful indicators of stream acidity. However, some expected relationships such as concentrations of mineral soil extractable Ca and $SO_4$ were not significant. This suggests that an extensive test of these indicators across the southern Appalachians will be required to identify high elevation forested catchments that would benefit from restoration activities.



## 1. Introduction

Ecosystem responses to acidic deposition were a significant concern and a focus of research in the later part of the 20th century in the eastern United States (Johnson et al., 1982, 1992). The Clean Air Act of 1970 and the Clean Air Act Amendments of 1990 (CAAA), along with other emission reduction regulatory programs, have resulted in declining concentrations of sulfate ($SO_4$) and hydrogen ($H^+$) in wet deposition, consistent with the declines in sulfur dioxide ($SO_2$) emissions across the eastern US (Driscoll et al., 2003). For example, the U.S. Environmental Protection Agency (2015) reported that the three-year average for 1989–1991 and

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2009–2011 sulfur (S) and total nitrogen (N) deposition (dry plus wet) decreased by 55% in the eastern US. Similarly, NADP reported a decline in $SO_4$ deposition in most southern Appalachian monitoring sites beginning in 1990 (NADP, 2007). This decline was also evident in data from Great Smoky Mountains National Park (Pardo and Duarte, 2007) and the Coweeta Hydrologic Laboratory in southwestern North Carolina, US (Knoepp, unpublished data). Despite reductions in emissions, many areas of the US still exhibit evidence of the negative impacts of acidic atmospheric deposition (Greaver et al., 2012). For example, some high elevation streams in the eastern U.S. continue to show signs of chronic to episodic acidity (Sullivan et al., 2007). Modeled patterns of $SO_4$ + nitrate ($NO_3$) deposition and ecosystem critical loads, exceeded the capacity of forest soils in approximately 17% of forested sites across the conterminous United States (McNulty et al., 2007) and numerous aquatic ecosystems in the southern Appalachians (McDonnell et al., 2014).

Patterns of atmospheric S and N deposition in mountainous terrain varies across landscapes and is related to rainfall amount, the presence of clouds and fog, elevation, forest edges, aspect, and vegetation composition (Weathers et al., 2000; Sullivan et al., 2007) with an estimated a 4–6-fold range in spatial variability (Weathers et al., 2006). Within the southern Appalachian Mountains high elevation sites receive higher rainfall (Swift et al., 1988) and greater inputs of nutrient and pollutant deposition (Swank, 1988; Swank and Vose, 1997; Sullivan et al., 2007) than low elevation sites. High elevation watersheds also have deep rocky soils with high organic matter content (Knoepp and Swank, 1998; Knoepp et al., 2000) and vegetation communities similar to forests in the northeastern U.S. (Elliott et al., 1999; Elliott and Swank, 2008). The deposition of $SO_4$ and $NO_3$ anions and their movement through the forest floor (soil O-horizon) and mineral soil profile results in the removal of base cations (calcium (Ca), magnesium (Mg), and potassium (K)) from soils. When base cations are removed from soils without adequate buffering capacity, soil pH decreases and aluminum (Al) is solubilized resulting in increased Al concentrations in soil solution and streams. As a result, stream acid neutralizing capacity (ANC) and stream pH decline. Examination of the effects of regional changes in acidic deposition, using biogeochemical models such as NuCM (Elliott et al., 2008) and MAGIC (Sullivan et al., 2007, 2011) found sensitivity to $SO_4$ deposition was related to soils and parent material; soils and parent material with low base cations concentrations were particularly sensitive to $SO_4$ deposition.

Base cation depletion in southern Appalachian high elevation watersheds is indicated by stream ANC values below 50 $\mu eq\ L^{-1}$, a value that has been defined as acidic (Bulger et al., 2000; Sullivan et al., 2007). Sullivan et al. (2008) examined the possibility of ANC recovery in Shenandoah National Park, Virginia US and found that watersheds located on siliciclastic bedrock would require a 77% decrease in atmospheric $SO_4$ deposition, compared to 1990 levels, to reach an ANC level of 50 $\mu eq\ L^{-1}$ due to low concentrations of base cations in the soil. Sullivan et al. (2011) used the biogeochemical cycling model MAGIC and data from 65 acid sensitive watersheds in eastern Tennessee and western North Carolina to back cast historical ANC values; they estimated that in 1860, ANC was as low as 30 $\mu eq\ L^{-1}$ with a median of 65 $\mu eq\ L^{-1}$. Likens et al. (1996) estimated that pre-industrial revolution stream ANC in the Northeastern US averaged 20 $\mu eq\ L^{-1}$. While stream chemistry at Hubbard Brook Experimental Forest has shown consistent improvement (declining $SO_4$, increasing pH and ANC) since the implementation of the Clean Air Act in 1970, stream ANC values remain low. Likens and Buso (2012) concluded that soil weathering processes have not been rapid enough to replenish stream Ca concentrations, leaving diluted streams with altered cation ratios. Soils in the southern Appalachians have high $SO_4$ retention capacity, which may delay the recovery of stream base cations. Rice et al. (2014) predicted that soils at three locations in western North Carolina will crossover from retaining to releasing $SO_4$ between 2023 and 2025.

Liming is a potential management option to restore streams and forest soils by decreasing acidity and increasing base cation availability. Lime is routinely used in agricultural systems, increasing soil pH, as well as Ca and Mg availability while also reducing Al solubility. Lime contains Ca and Mg, the two major divalent base cations; the ratio of these cations is dependent on the lime sources. Huettl (1993) and more recently Reid and Watmough (2014) review benefits and problems of lime applications in forest liming studies. The meta-analysis conducted by Reid and Watmough (2014) found that 67% of lime treatment trials showed increased soil pH and foliar Ca concentration. Soil pH response was greater in organic compared to mineral soils and Ca foliar response was positively correlated with treatment dose. Huettl (1993) focused on historic liming studies in Germany with reported benefits including increased soil Ca and Mg in O-horizons and surface mineral soils, which was accompanied by increased soil cation exchange capacity (CEC) and percent base saturation (%BS). Conversely, there was also evidence of increased soil $NO_3$ production following liming that resulted in cation leaching from subsoils. Increased rates of nitrification following liming were also found in forested sites in Finland (Priha and Smolander, 1995) and was stated as a concern because of the potential for accelerated cation leaching losses. Elliott et al. (2013) measured the response of soil and soil solution to liming following a wildfire in the Linville Gorge Wilderness Area in western North Carolina, a site previously shown to have low soil cation availability and acidic streams (Elliott et al., 2008). They found significant increases in surface mineral soil ECEC, pH, and Ca and a decline in Al as well as increases in soil solution $NO_3$. However, the lime response was short-lived, which they attributed to the small amount of lime added to the site (1.1 $Mg\ ha^{-1}$). In other forest liming studies, the effects of liming in the O-horizon and surface mineral soils were long-term, up to 21 years (Long et al., 2015), and in some cases resulted in the accumulation of organic material in the O-horizon (Johnson et al., 1995).

The continued sensitivity of southern Appalachian streams to atmospheric deposition emphasizes the need to identify watershed characteristics that influence stream chemistry responses to acidic deposition. For example, McDonnell et al. (2014) developed a screening tool that used a mass balance model to estimate critical loads for watersheds at risk of acidification based on S deposition. Although coarsely modeled at the regional scale, their work suggested that catchment characteristics could be used to identify catchments at risk and help managers prioritize stream monitoring and restoration efforts through liming. Hence, our first objective was to identify catchment biotic, physical and chemical characteristics, that are potential indices of stream acidity measured as ANC, pH, or Ca:Al molar ratio. A better understanding of these catchment scale characteristics and their relationship with stream chemistry could be used to evaluate restoration options such as liming. Our second objective was to estimate catchment lime requirements and consider how liming may improve stream chemistry.

To address these objectives we studied three large watersheds in the southern Appalachian Mountains along a gradient of acidic deposition with differing geology. Within each watershed we selected first order catchments that represented a range of elevations (849–1526 m) and stream ANC values based on previous studies (W. A. Jackson, unpublished data). We characterized all catchments in terms of overstory tree species composition and stand characteristics, site and soil morphology and chemistry, and soil lime requirement. Stream, O-horizon, and mineral soils, were intensively sampled four times over one year to capture sea-