



Steady-state sulfur critical loads and exceedances for protection of aquatic ecosystems in the U.S. southern Appalachian Mountains



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ABSTRACT

Atmospherically deposited sulfur (S) causes stream water acidification throughout the eastern U.S. Southern Appalachian Mountain (SAM) region. Acidification has been linked with reduced fitness and richness of aquatic species and changes to benthic communities. Maintaining acid-base chemistry that supports native biota depends largely on balancing acidic deposition with the natural resupply of base cations. Stream water acid neutralizing capacity (ANC) is maintained by base cations that mostly originate from weathering of surrounding lithologies. When ambient atmospheric S deposition exceeds the critical load (CL) an ecosystem can tolerate, stream water chemistry may become lethal to biota. This work links statistical predictions of ANC and base cation weathering for streams and watersheds of the SAM region with a steady-state model to estimate CLs and exceedances. Results showed that 20.1% of the total length of study region streams displayed ANC <100 $\mu\text{eq}\cdot\text{L}^{-1}$, a level at which effects to biota may be anticipated; most were 4th or lower order streams. Nearly one-third of the stream length within the study region exhibited CLs of S deposition <50 $\text{meq}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$, which is less than the regional average S deposition of 60 $\text{meq}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$. Owing to their geologic substrates, relatively high elevation, and cool and moist forested conditions, the percentage of stream length in exceedance was highest for mountain wilderness areas and in national parks, and lowest for privately owned valley bottom land. Exceedance results were summarized by 12-digit hydrologic unit code (subwatershed) for use in developing management goals and policy objectives, and for long-term monitoring.

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1. Introduction

Atmospheric sulfur (S) deposition, originating largely from coal-fired electrical power generation and other industrial sources, causes soil, groundwater, and stream water acidification across broad areas of the southeastern United States (U.S. Environmental Protection Agency [USEPA], 2008). Such acidification has been associated with enhanced leaching of sulfate (SO_4^{2-}), depletion of

calcium (Ca^{2+}) and other nutrient base cations from soils, reduced pH and acid neutralizing capacity (ANC) of surface waters, and increased mobilization of potentially toxic inorganic aluminum (Al_i) from soil to streams (Sullivan, 2000). Biological effects include toxicity to fish and aquatic invertebrates (Cosby et al., 2006; USEPA, 2009).

Sulfur is the primary determinant of precipitation acidity and SO_4^{2-} is the dominant anion in streams throughout most of the Southern Appalachian Mountain (SAM) region (Sullivan et al., 2004). Nitrate (NO_3^-) is important at some locations, especially at streams that flow from high-elevation old-growth forests in North Carolina and Tennessee (Cook et al., 1994). Streams are generally dilute and clear-water with limited contributions of naturally occurring organic acidity. Although a substantial proportion of atmospherically deposited S can be retained in watershed soils,

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SO_4^{2-} concentrations in many mountain streams have increased due to atmospheric S deposition and low S retention in soils (Elliott et al., 2008), causing increases in base cation concentrations and decreased stream water ANC.

Acidic soils and streams have developed in this region over a period of many decades in response to high levels of atmospheric S deposition. Many streams in Great Smoky Mountains (GRSM) and Shenandoah (SHEN) national parks and surrounding national forests show signs of acidification, including streams in wilderness areas. Both of these parks and several wildernesses are federally mandated Class I areas, and receive special protection against air pollution impacts under the Clean Air Act. As a result of emissions controls regulation (USEPA, 2009), atmospheric S deposition has decreased throughout the eastern United States since the early 1980s, and further decreases are expected.

Ecosystem sensitivity to acidification is fairly well documented for this region, particularly within the National Acid Precipitation Assessment Program (NAPAP, 1991), the Fish in Sensitive Habitats (FISH) project (Bulger et al., 1999), and the Southern Appalachian Mountains Initiative (SAMI) assessment (Sullivan et al., 2004, 2007).

Stream water ANC is one measure that reflects the ability of a watershed to neutralize acidic inputs. As the rate of acidic deposition increases, ANC often decreases in proportion to the natural re-supply of base cations from the soil. At certain levels of acidification, increases in the hydrogen ion (H^+) and Al_i concentration are directly toxic to fish, including brook trout (*Salvelinus fontinalis*; Baldigo et al., 2007; Bulger et al., 1999), a favored native game fish of cold, high-elevation streams. Various ANC thresholds are associated with different levels of biological effects. In the SAM region and in mountainous areas of the northeastern United States, moderate effects on macroinvertebrate and fish species richness are associated with ANC concentrations between ~ 50 and $100 \mu\text{eq}\cdot\text{L}^{-1}$ (Cosby et al., 2006; Sullivan et al., 2006). More substantial effects have been observed at ANC concentrations $< 50 \mu\text{eq}\cdot\text{L}^{-1}$. Most aquatic species, including the relatively acid-tolerant brook trout, can be extirpated at ANC concentrations $< 0 \mu\text{eq}\cdot\text{L}^{-1}$ (Bulger et al., 1999; Cosby et al., 2006; Sullivan et al., 2006; USEPA, 2009).

Soils in this region have developed from the slow weathering of parent rock material, some of which is inherently low in base cations. Adequate amounts of available Ca, magnesium (Mg), and potassium (K) are all essential to maintain an acid-base chemistry that will support persistence of native fish and aquatic invertebrate species. Land managers and regulators have a legally mandated concern for the current and future health of native aquatic species within the SAM. Where the existing stream water acidity is too high to support the native biota, and where ambient stream water ANC is insufficient for buffering, policy-makers may need to call for added air pollution emissions reductions to enable recovery of impacted species, and to prevent further impacts. Thus, to inform public policy regarding air pollutant emissions reductions, it is important to determine 1) the emission and atmospheric deposition levels that are associated with varying degrees of chemical effects and 2) the linkages between water and soil chemistry and subsequent biological impacts.

One approach to addressing these issues is to construct model estimates of regional surface water acid-base chemistry and critical loads (CLs). The CL for S acidification is the level of sustained atmospheric S deposition below/above which harmful effects to sensitive ecosystems are unlikely/likely, based on current understanding (Nilsson and Grennfelt, 1988). The CL is typically calculated as a steady-state value, using models such as the Steady State Water Chemistry (SSWC) model (Henriksen and Posch, 2001). However, data used to inform the steady-state CL calculation may

also be derived dynamically using mass balance equations in a process model such as the Model of Acidification of Groundwater in Catchments (MAGIC; Cosby et al., 1985).

The long-term maintenance of well-buffered aquatic ecosystems depends primarily on maintaining atmospheric S levels that are lower than the natural re-supply rate of base cations through weathering (BC_w). Thus, base cations derived from weathered substrates are generally most influential in determining CLs (McDonnell et al., 2010). However, because BC_w predictions can contribute substantial uncertainty to CL estimation (Li and McNulty, 2007; USEPA, 2009), it is essential to continue to improve the certainty of weathering estimates.

Steady-state CL calculations have been developed and applied across northern Europe (Gregor et al., 2004) and eastern Canada (Oumet et al., 2006; Watmough and Dillon, 2002), providing a basis for political and economic negotiations and national and international air pollution legislation. Some recent efforts in the United States have focused on process-based dynamic model estimates of critical or associated target loads (cf., Sullivan et al., 2005, 2008). Land managers and regulators are also interested in regional predictions at watershed locations where current stream water ANC is affecting the health of aquatic biological communities (USEPA, 2009).

This work incorporates results from recent regional statistical modeling to predict stream water ANC and soil BC_w throughout the SAM region. The BC_w modeling results are used here together with other model input parameters to estimate steady-state S CLs and CL exceedances. Stream ANC estimates are used to assess potential biological effects associated with modeled S deposition.

Previous regional efforts to characterize stream water sensitivity to acidic deposition have been based on stratified random sampling of a subset of streams (cf., Whittier et al., 2002). Results were used to make general statements about ecosystem sensitivity throughout the full population of surface waters; however, methods were insufficient for determining the location of sensitive reaches. This study resolves that problem by generating spatially explicit CL and exceedance estimates for all streams within the SAM region.

As a result of emissions regulation and advances in hydraulic fracturing technology for natural gas production, reduced S emissions at coal-fired power plants and shifts to natural gas-powered electric generation have primarily been responsible for significant reductions in acidic deposition throughout the United States (NAPAP, 2011). Results reported here also consider how changes in exposure to S deposition over time relate to the inherent acid sensitivity of the landscape and expected future stream conditions with respect to acidification.

The SAM region comprises an irregular patchwork of land ownerships, protection status, resource management goals, and sensitivity to degradation from S deposition. National parks and wildernesses are home to terrestrial and aquatic ecosystems that are afforded more legal protections than those that exist on other lands (Organic Act 16 U.S.C. §1 (1997); Organic Act 16 U.S.C. §1601(a) (1997); Wilderness Act 16 U.S.C. §1131 (1997)). It is therefore highly desirable to determine the extent to which acid-sensitive streams occur within these protected areas. Furthermore, present-day land managers need to determine acid-sensitive locations to make informed resource management decisions and recommendations to air quality regulators and policy-makers.

2. Methodology

The study area covers the SAM region and surrounding terrain, from northern Georgia to southern Pennsylvania, and from eastern

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