



Original Articles

Relationships between indicators of acid-base chemistry and fish assemblages in streams of the Great Smoky Mountains National Park

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ABSTRACT

The acidity of many streams in the Great Smoky Mountains National Park (GRSM) has increased significantly since pre-industrial (~1850) times due to the effects of highly acidic atmospheric deposition in poorly buffered watersheds. Extensive stream-monitoring programs since 1993 have shown that fish and macroinvertebrate assemblages have been adversely affected in many streams across the GRSM. Matching chemistry and fishery information collected from 389 surveys performed at 52 stream sites over a 22-year period were assessed using logistic regression analysis to help inform the U.S. Environmental Protection Agency's assessment of the environmental impacts of emissions of oxides of nitrogen (NO_x) and sulfur (SO_x). Numerous logistic equations and associated curves were derived that defined the relations between acid neutralizing capacity (ANC) or pH and different levels of community richness, density, and biomass; and density and biomass of brook trout, rainbow trout, and small prey (minnow) populations in streams of the GRSM. The equations and curves describe the status of fish assemblages in the GRSM under contemporary emission levels and deposition loads of nitrogen (N) and sulfur (S) and provide a means to estimate how newly proposed (and various alternative) target deposition loads, which strongly influence stream ANC, might affect key ecological indicators. Several examples using ANC, community richness, and brook trout density are presented to illustrate the steps needed to predict how future changes in stream chemistry (resulting from different target deposition loads of N and S) will affect the probabilities of observing specific levels of selected biological indicators in GRSM streams. The implications of this study to the regulation of NO_x and SO_x emissions, water quality, and fisheries management in streams of the GRSM are discussed, but also qualified by the fact that specific examples provided need to be further explored before recommendations concerning their use as ecological indicators could be proposed.

1. Introduction

Watersheds of the Great Smoky Mountains National Park (GRSM) receive high levels of acid deposition resulting from atmospheric emissions of nitrogen (NO_x) and sulfur (SO_x) oxides (Cook et al., 1994; Zhou et al., 2015). Acidic deposition has reduced acid neutralizing capacity (ANC) and calcium (Ca) concentrations and increased acidity and aluminum (Al) concentrations in soils and surface waters and affected forest health as well as fish and macroinvertebrate assemblages across the southern Appalachian Mountain region (Pardo and Duarte, 2007; Schwartz et al., 2014; Weathers et al., 2006). Despite recent declines in acidic deposition nationwide, acid-base chemistry for a group of study streams in the Shenandoah National Park, Virginia has generally not changed appreciably between the early 1990s and the 2000s (USEPA, 2009). Hindcast simulations of pre-industrial chemistry in two groups of streams in the southern Appalachian Mountains (Blue

Ridge and GRSM) estimate that all had ANC greater than 20 μeq L⁻¹ and median ANC was generally near 50 μeq L⁻¹ around 1850 to 1860 (Fakhraei et al. 2016; Sullivan et al. 2011). Sullivan et al. (2011) estimated that the median ANC of 66 streams in the region had declined 25 μeq L⁻¹ by 2005, whereas, Zhou et al. (2015) estimated the mean ANC of 12 streams had decreased 38 μeq L⁻¹ by 2007. In the GRSM, the median ANC of waters in sensitive streams (with ANC less than 100 μeq L⁻¹) declined from about 45 μeq L⁻¹ in 1850 to about 6 μeq L⁻¹ in 2014 (Fakhraei et al., 2016), yet the ANC in 77% of GRSM streams had either not changed (67 of 92 sites) or decreased significantly (4 of 92 sites) between 1993 and 2009 (Schwartz et al. 2014). Twelve streams on the Tennessee-side of the GRSM National Park are listed on the Clean Water Act's 303d list of impaired surface waters for falling below the pH standard (6.00) as a result of atmospheric acidic deposition – mainly nitrate and sulfate (Schwartz et al., 2014). These changes in acid-base chemistry have adversely affected fish and

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macroinvertebrate assemblages in numerous streams across the GRSM (Schwartz et al., 2014). As one example, whole-body losses of sodium (10–20%) have approached lethal levels for caged brook trout during storm events and seven brook trout populations have been lost in 13 miles of headwater streams in the GRSM over the last 40 years due to acid deposition (M.A. Kulp, May 2016, Personal communication; Neff et al., 2009).

The U.S. Environmental Protection Agency (EPA) is developing secondary national ambient air quality emission standards for NO_x and SO_x that should indirectly protect terrestrial and aquatic species and their communities from further adverse impacts and help promote the recovery of acidified ecosystems to an unimpaired or, at least, an acceptable condition/level (USEPA, 2009). These standards will rely heavily on critical loads research which estimate threshold or target deposition loads of nitrogen (N) and sulfur (S) to watersheds, below which significant harmful effects to sensitive elements of terrestrial and (or) aquatic ecosystems should not occur (Greaver et al., 2012; Porter et al., 2005). A number of acid-base chemistry parameters such as pH, ANC, and inorganic monomeric aluminum have relatively fixed effect thresholds for selected aquatic and terrestrial species, which when exceeded can impair their health, cause mortality, reduce population density, shift species distributions, and decrease overall community diversity (Sullivan et al., 2012, 2008). A clearer understanding of the relations between acid-base chemistry and biological responses, however, is needed to evaluate the specific target loads (for N and S deposition) that will best protect species populations, communities, and entire ecosystems (USEPA, 2009).

The relations among emission rates and deposition loads of N and S, acid-base chemistry, and the health of terrestrial and aquatic species and their assemblages (population and/or community metrics) are complicated, regionally variable, and difficult to characterize (Greaver et al., 2012; Pardo et al., 2011). In an extensive literature review and assessment of GRSM water quality, fish (population metrics for brook trout *Salvelinus fontinalis* and rainbow trout *Oncorhynchus mykiss*), and aquatic macroinvertebrate data, Schwartz et al. (2014) identified several acid-base chemistry thresholds, and streams exceeding these limits, yet indicated a more comprehensive analysis was needed to characterize a broader range of biological effects caused by acidification. Fortunately, water chemistry has been routinely monitored at 43 to 357 stream sites in the GRSM since 1993, fish assemblages were quantified at nearly 300 sites from 1990 to 2014, and macroinvertebrate communities were assessed at more than 118 sites from 1990 to 2014 (Schwartz et al., 2014). Thus, acid-base chemistry and biological data from thousands of chemistry samples and hundreds of fish surveys done across the GRSM over the past 30 years are available to derive a wide assortment of biological-response equations and associated curves. The equations and curves can be used to postulate and evaluate how changes in various ecological (chemical) indicators such as ANC or pH (expected to result from different target loads of N and S), will likely affect biological indicators that reflect the health of stream ecosystems (e.g., community richness and diversity, abundance of species populations) in the GRSM.

The primary objective of the present study is to increase our understanding of the relations between acid-base chemistry and the condition of fish populations and communities in streams sampled throughout the GRSM between 1993 and 2014. Resulting response equations that quantify the relations between chemical indicators and fishery metrics (biological indicators) will help inform the EPA's Integrated Review Plan for Secondary National Ambient Air Quality Standards for NO_x and SO_x (USEPA, 2009). Specific goals of this paper are to: (1) characterize the significant relations between chemical indicators of acidification stress (e.g., pH, ANC, total Al, SO₄²⁻, NO₃⁻ and Ca²⁺), and selected population and community metrics or indicators (e.g., species absence-or-presence, abundance, richness, and diversity), (2) examine the relations for biological-effect thresholds, and (3) identify the range of responses that selected biological indicators

exhibit in streams of the GRSM that are affected by acidification or which are expected to recover from declines in acid deposition. These equations and associated curves can be used not only to characterize how acidification currently affects fish assemblages in local streams, but to also forecast how future changes in stream chemistry (resulting from various target loads of N and S) will affect the recovery of fish communities in streams of the GRSM across variable time steps.

2. Methods

In 1993, a long-term Inventory and Monitoring (I&M) Program was initiated to investigate the impacts of acid deposition on water quality, aquatic macroinvertebrate communities, and fish populations in streams across the GRSM (Schwartz et al., 2014). Although the specific study sites, sampling frequency at individual sites, and number of sites inventoried each year varied over time with changes in program objectives and funding, more than 2700 water-chemistry samples were collected from at least 387 sites and about 590 quantitative fish inventories were completed from nearly 300 sites between 1993 and 2014. This dataset was appended to include chemistry and fish information from an additional 20 sites, where fish assemblages were surveyed (during 2015) but no individuals were observed, to ensure that the chemistry gradient was complete. Data from 26 inventories done at several sites with ANC greater than 150 µeq L⁻¹ were excluded from all analyses because the biological responses to acid-base chemistry above that ANC level were nominal and because the extended range in acid-base chemistry tended to obscure the response gradients. From the remaining 564 inventories, corresponding water chemistry data (1 to 13 samples collected during the same year that fish were inventoried) were available from 52 sites. Because fish assemblages at many sites were inventoried multiple times over the 22-year period (some were surveyed annually for the entire period), a total of 389 observations of paired fishery and chemistry data were available for analysis. The number of sites and generalized design for monitoring water chemistry and fish assemblages between 1986 and 2009, and which continue through 2014 (and today), are described in detail by Schwartz et al. (2014). The locations and characteristics of the 52 study sites, water-chemistry sampling and laboratory methods, fish-survey procedures, data sources, and the data compilation and analysis steps are summarized in the following subsections.

2.1. Study area

Fish and chemistry data from 52 study sites located in the GRSM were evaluated for the present investigation; see Figure S1 and interactive map in Baldigo et al. (2017). The stream order ranged from 1 to 5, however, most sites were second or third order reaches with drainage areas that ranged from 3.1 to 2756 ha and elevations that ranged from 335 to 1469 m as noted in Table S1 of Baldigo et al. (2017). The site identification (ID), GRSM station name, years of surveys, coordinates, stream order, elevation, and drainage area for each site are listed in Table S1 of Baldigo et al. (2017).

2.2. Water chemistry

Samples for analysis of water chemistry were collected manually at least once and as many as 13 times at each of the 52 stream sites during the same years that fish were inventoried between 1993 and 2014. All samples were collected in 250-ml plastic bottles, stored on ice or refrigerated, then transported generally within 24 h of collection to the University of Tennessee (UT) Department of Civil and Environmental Engineering laboratory for analysis. All water-chemistry samples were analyzed by UT staff for determination of pH, ANC, conductivity, acid anions (Cl⁻, SO₄²⁻, NO₃⁻), ammonia (NH₄⁺), and base cations (Ca²⁺, Mg²⁺, Na⁺, K⁺) with the addition of dissolved metals (Al, Cu, Fe, Mn, Si, and Zn) in 2003 following standard analytical methods and quality

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