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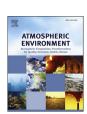
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Declining nitrate-N yields in the Upper Potomac River Basin: What is really driving progress under the Chesapeake Bay restoration?

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HIGHLIGHTS

- U.S. NO_x emission control programs were shown to be the primary driver of improving water quality across most of the UPRB.
- The MKNSM explained large proportions of the variation in annual nitrate-N yield through time and among the watersheds.
- The MKNSM allowed the annual nitrate-N yield to be separated into "responsive" and "non-responsive" components.
- NO_x emission controls have rapidly reversed nitrogen saturation across most of the UPRB.

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ABSTRACT

Reducing nutrient pollution of surface and coastal waters in the U.S. and elsewhere remains a major environmental and engineering challenge for the 21st century. In the case of the Chesapeake Bay restoration, we still lack scientific proof that watershed-based management actions have been effective at reducing nonpoint-source nutrient loads from the land to this estuary in accordance with restoration goals. While the conventional wisdom is that implementation of best management practices (BMP's) and wastewater treatment have turned the tide against nutrient pollution, we examined long-term (1986present) nitrate-N trends in streams and major tributaries of the Upper Potomac River Basin (UPRB) and found that: 1) dramatic reductions in annual discharge-weighted mean nitrate-N concentrations and yields across the UPRB can be almost universally attributed to reductions in atmospheric N deposition as opposed to on-the-ground management actions such as implementation of BMP's; 2) observed water quality changes generally comport with a modified kinetic N saturation model (MKNSM); 3) the MKNSM can separate the nitrate-N yield that is responsive to atmospheric deposition from a "non-responsive" yield; and 4) N saturation from atmospheric N deposition appears to be an inherently reversible process across most of the landscape. These unanticipated region-wide water quality benefits can be attributed to NO_x emission controls brought about by the 1990 Clean Air Act Amendments (and subsequent U.S. NO_X control programs) and reflect a water quality "success story" in the Chesapeake Bay restoration. © 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Reducing nutrient pollution of streams, rivers, lakes, and coastal waters in the U.S. and elsewhere remains a major environmental and engineering challenge for the 21st century (NRC, 2000; Howarth et al., 2000, 2002). In the U.S., the Clean Water Act passed in 1972 and amended in 1977 and 1987 established water pollution control regulations, provided funding for water treatment systems, and created a federal-state administrative program that

has significantly reduced some types of water pollution—especially wastewater from municipal and industrial point source discharges (Dzombak, 2011). Controlling nonpoint source pollution (e.g., nutrient pollution from agricultural and urban runoff), has proven to be a much more vexing problem, however, due at least in part to a lack of regulatory and enforcement actions that can be used under the Clean Water Act (Dzombak, 2011). Perhaps nowhere in the U.S. has solving this nutrient pollution problem been more challenging than in the Chesapeake Bay—the nation's largest estuary—which has been plagued by excessive nutrient pollution and widespread hypoxic conditions that developed over many decades. Now on the third iteration of a state/federal agreement and partnership to restore this valuable ecosystem by dramatically reducing nutrient

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pollution through implementation of a total maximum daily load (TMDL) allocation process, and nearly thirty years of water monitoring and scientific study, progress has been frustratingly slow (NRC, 2011). In the case of the Chesapeake Bay restoration, we still lack scientific proof that watershed-based management actions have been effective at reducing nonpoint-source nutrient loads from the land to this estuary in accordance with the restoration goals. It has been suggested that improvements in nitrogen use efficiency resulting from implementation of agricultural best management practices (BMP's) combined with advanced municipal wastewater treatment are primarily responsible for observed declining nitrogen yields in some Chesapeake rivers (Shenk and Linker, 2013), despite the fact that some of the greatest percentage declines in N yields have been observed in predominantly-forested watersheds (Eshleman et al., 2013).

The availability of long-term water quality datasets, however, allows us to take a different tack that focuses more explicitly on the analysis of water quality trends in streams and major tributaries of Chesapeake Bay to determine whether the water quality improvements might be explained by drivers that have been largely overlooked or not properly accounted for in previous modeling efforts (e.g., Shenk and Linker, 2013). Our particular interest is in understanding watershed responses to atmospheric N deposition which was first implicated as a contributor to riverine nitrogen loads to Chesapeake Bay in the 1990's (Fisher and Oppenheimer, 1991; Jaworski et al., 1992, 1997). In the early 2000's, some researchers concluded that reducing N emissions and associated atmospheric N deposition was not an effective management tool for reducing total N loads to estuaries in the eastern U.S (Castro and Driscoll, 2002; Whitall et al., 2003). Recently, the role of atmospheric N deposition has been more accurately accounted for using the NANI (i.e., net anthropogenic nitrogen input) concept, but this approach has been used exclusively to examine spatial (rather than temporal) variability in N inputs and responses (Howarth et al., 2012; Hong et al., 2013). Chesapeake Bay Program data indicate that steep declines in atmospheric N inputs to the Chesapeake Bay watershed brought about through federal NO_x emission controls dwarf any declines in inputs of agricultural N sources (e.g., manures and fertilizers), but the possibility that declining atmospheric N deposition might by itself provide a universal explanation for recent improvements in water quality in both forested and mixed land use watersheds has not been fully assessed (Shenk and Linker, 2013; Linker et al., 2013; Boyer et al., 2002).

The research we report on here focuses on an issue of great importance to scientists and watershed managers alike by addressing two questions: 1) have controls on atmospheric N deposition reduced N yields from the land to surface waters (and, if so, how and by how much); and 2) will future reductions in atmospheric N deposition result in additional water quality improvements? Our previous work on these questions focused exclusively on nine predominantly-forested (i.e., >75% forest cover) watersheds located in the mountainous headwaters of the Chesapeake Bay basin (Eshleman et al., 2013). The study provided evidence that reductions in atmospheric N deposition—brought about through controls on NO_x emissions from stationary sources under the Acid Rain Program (ARP) of the Clean Air Act Amendments of 1990 (and subsequent federal air quality regulatory actions that reduced both stationary and mobile NO_x sources)—had produced dramatic (~40%) reductions in nonpoint-source nitrate-N yields during the period from the mid-1990's to the present. Our results also provided support for the application of a kinetic N saturation model—based on the simple concept of a watershed N mass balance—that attributed long-term changes in nitrate-N yields from forests to changes in atmospheric N deposition (Eshleman et al., 2013). The specific focus of this follow-up study is on the Upper Potomac River Basin (UPRB)—after the Susquehanna River, the 2nd largest source of freshwater to Chesapeake Bay—although we believe that the methods are relevant to understanding N dynamics throughout the larger Chesapeake Bay watershed.

We evaluate long-term changes in nitrate-N yields across the UPRB using monitoring data from 12 subwatersheds and the mainstem station at Washington, DC (POTW); data from five other Chesapeake Bay watersheds (not located in the UPRB, but analyzed previously by Eshleman et al., 2013) are also included in the present analysis. The data are used to test the following hypotheses: 1) reductions in annual nitrate-N concentrations and yields across the entire UPRB, including watersheds dominated by non-forested land, can be attributed to reductions in atmospheric N deposition; and 2) the observed water quality changes comport with a conceptual model of kinetic N saturation.

In our previous analysis (Eshleman et al., 2013), we interpreted empirical relationships between annual nitrate-N yield (Y) and annual wet N deposition (D) as evidence of a process of kinetic forest N saturation first suggested by Lovett and Goodale (2011):

$$Y = D - A - G \tag{1}$$

where A is the net annual incorporation of N into forest vegetation and soil organic matter and G are gaseous N losses. A simple solution to Eq. (1) was obtained by: 1) neglecting G; and 2) making A a linear function of D: A = aD where a ($0 \le a \le 1$) represents the proportion of D that is taken up and stored in forest vegetation and soil (i.e., a is a forest N retention factor) and the y-intercept ($Y_0 \ge 0$) provides a measure of the (assumed constant) annual nitrate-N yield from non-forested land considered to be non-responsive to changes in atmospheric N deposition (Eshleman et al., 2013):

$$Y = Y_0 + (1 - a)D (2)$$

For cases where $Y_0=0$, it is easily shown that a=A/D=1-Y/D where a is a forest N retention factor (i.e., the average proportion of atmospheric N retained by a forest system in a year) which can be readily measured in watershed input-output studies (e.g., Grigal, 2012). In the present analysis of data from both predominantly forested and mixed land use watersheds, we test whether there is statistical support for a more general, *modified* kinetic N saturation model (MKNSM) in which Y increases exponentially with increasing atmospheric N deposition such that:

$$Y = Y_0 exp[kD] (3)$$

where Y = annual watershed nitrate-N yield (kg N ha⁻¹); Y_0 is a baseline annual watershed nitrate-N yield (kg N ha⁻¹) that is considered non-responsive to changes in atmospheric N deposition; and k is a constant (ha kg⁻¹). Exponential relationships between N outputs and inputs have been observed in several other studies (Howarth et al., 2006, 2012; Gao et al., 2014), but our analysis is the first to examine such relationships using long-term, temporal datasets for individual watersheds. In testing this relationship for mixed land use watersheds in particular, we are effectively assuming that other non-atmospheric N inputs to these systems (i.e., N from point sources and nonpoint sources) are static and independent of D.

2. Materials and methods

We supplemented our own long-term data from two UPRB watersheds by obtaining nitrate-N concentration records from state water quality databases or USEPA STORET (http://www.epa.gov/storet) and stream/river discharge data from U.S. Geological Survey (http://waterdata.usgs.gov/nwis/sw) for 16 additional stations;

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