



# Development of a total maximum daily load (TMDL) for acid-impaired lakes in the Adirondack region of New York



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## HIGHLIGHTS

- A total maximum daily load was developed for 128 acid-impaired Adirondack lakes.
- Hindcast simulations were used to estimate pre-anthropogenic conditions.
- Controlling S deposition is much more effective in recovering acidic lakes than N.
- Recovery classes: no additional control; additional control needed; no recovery.
- Adirondack lake-watersheds will not recover to pre-anthropogenic conditions.

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## ABSTRACT

Acidic deposition has impaired acid-sensitive surface waters in the Adirondack region of New York by decreasing pH and acid neutralizing capacity (ANC). In spite of air quality programs over past decades, 128 lakes in the Adirondacks were classified as “impaired” under Section 303(d) of the Clean Water Act in 2010 due to elevated acidity. The biogeochemical model, PnET-BGC, was used to relate decreases in atmospheric sulfur (S) and nitrogen (N) deposition to changes in lake water chemistry. The model was calibrated and confirmed using observed soil and lake water chemistry data and then was applied to calculate the maximum atmospheric deposition that the impaired lakes can receive while still achieving ANC targets. Two targets of ANC were used to characterize the recovery of acid-impaired lakes: 11 and 20  $\mu\text{eq L}^{-1}$ . Of the 128 acid-impaired lakes, 97 currently have ANC values below the target value of 20  $\mu\text{eq L}^{-1}$  and 83 are below 11  $\mu\text{eq L}^{-1}$ . This study indicates that a moderate control scenario (i.e., 60% decrease from the current atmospheric S load) is projected to recover the ANC of lakes at a mean rate of 0.18 and 0.05  $\mu\text{eq L}^{-1} \text{ yr}^{-1}$  during the periods 2022–2050 and 2050–2200, respectively. The total maximum daily load (TMDL) of acidity corresponding to this moderate control scenario was estimated to be 7.9 meq S  $\text{m}^{-2} \text{ yr}^{-1}$  which includes a 10% margin of safety.

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## 1. Introduction

Controls on anthropogenic S and  $\text{NO}_x$  emissions are recognized as an important environmental success; in Europe anthropogenic  $\text{SO}_2$  emissions decreased 73% between 1980 and 2004 (Vestreng

et al., 2007), and in the US emissions of  $\text{SO}_2$  decreased 79% and emissions of electric utility and transportation based  $\text{NO}_x$  decreased 68% and 57%, respectively, between 1980 and 2012 (<http://www.epa.gov/ttn/chief/trends/index.html>). As a result, studies are shifting from assessment of impacts of acid deposition toward evaluation of ecosystem recovery. Consequently, there has been interest in the application of critical loads (CLs) and total maximum daily loads (TMDLs) to guide decisions regarding emission controls and atmospheric deposition that are necessary to recover acid impaired ecosystems (Posch, 2002; Sullivan et al., 2012a, 2012b). Maximum allowable atmospheric S and N (sum of reactive oxidized and

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reduced N species) deposition is typically derived by linking atmospheric inputs to surface water chemistry and setting a target for critical chemical indicator such as acid neutralizing capacity (ANC).

Models of varying complexity have been utilized to provide this linkage (e.g., [Henriksen and Posch, 2001](#); [Sullivan et al., 2012b](#)). The objective of this study is to apply a process-based biogeochemical model, PnET-BGC, as a tool to link changes in atmospheric S and N inputs to lake chemistry to: (1) investigate long-term acidification of soil and surface waters of impaired Adirondack ecosystems in response to historical changes in atmospheric deposition and potential future deposition scenarios; and (2) determine the TMDLs of acidity that would allow for their chemical recovery.

## 2. Sites selection

Under Section 303(d) of the Clean Water Act, in 2010, the New York State Department of Environmental Conservation (NYSDEC) identified 128 lakes within the Adirondack Forest Preserve that are impaired due to elevated acidity. These lakes were subject of this study. In addition 13 lakes were utilized from the Adirondack Long Term Monitoring (ALTM) program ([Driscoll et al., 2007, 2003](#)) to facilitate model calibration/confirmation.

Monthly water chemistry from the ALTM program is available since 1982 or 1992. Five of the impaired lakes are part of the Temporally Integrated Monitoring Environmental (TIME) program ([Waller et al., 2012](#)), for which annual water chemistry has been collected since 1991. Water chemistry data were also collected for all TMDL lakes by the NYSDEC during late summer or early fall for a few years, over the period 2007–2012.

The study sites generally have low ANC, and are sensitive to acid deposition. Among the study lakes 85%, 9% and 6% of 141 have ANC  $\leq 50$   $\mu\text{eq L}^{-1}$ , between 50 and 100  $\mu\text{eq L}^{-1}$ , and  $\geq 100$   $\mu\text{eq L}^{-1}$ , respectively, based on annual average values for ALTM lakes and summer/fall measurements for other study lakes. The mean atmospheric S and N deposition (wet and dry deposition) during 2009–2011 for the study lakes ranged from 20 to 34 and from 24 to 33  $\text{meq m}^{-2} \text{yr}^{-1}$ , respectively.

## 3. Model application

### 3.1. Model description

PnET-BGC is a biogeochemical model that requires meteorological, atmospheric deposition and historical land disturbance data to simulate hydrology and major ion chemistry in vegetation, soil and water. The model was developed by combining two sub-models, PnET-CN and BGC ([Gbondo-Tugbawa et al., 2001](#)). PnET-CN ([Aber et al., 1997](#)) is a lumped-parameter model which simulates the cycling and interactions of carbon, N, and water in forest ecosystems and estimates net primary productivity. The BGC algorithm depicts the dynamics of major elements in vegetation, soil and water and provides a comprehensive model which simulates major element balances including both biotic and abiotic process in forest watershed ecosystems ([Gbondo-Tugbawa et al., 2001](#)). In this paper, we focus our discussion on major chemical parameters in lake water (i.e., sulfate ( $\text{SO}_4^{2-}$ ), nitrate ( $\text{NO}_3^-$ ), calcium ( $\text{Ca}^{2+}$ ), pH and ANC) and soil (i.e., soil base saturation (BS)).

Recently, PnET-BGC has been modified by the addition of two subroutines; a  $\text{CO}_2$  uptake algorithm and a lake solute retention algorithm. The  $\text{CO}_2$  uptake algorithm considers the effects of increases in atmospheric  $\text{CO}_2$  concentration on forest ecosystem processes ([Pourmokhtarian et al., 2012](#)). The lake solute retention algorithm simulates the mass transfer of major elements from the water column of lakes to sediments. The lake solute retention algorithm requires knowledge of lake mean depth, water residence

time and mass transfer coefficients to predict in-lake removal or generation of major ions ([Kelly et al., 1987](#)).

### 3.2. Model data development

Simulations are initiated in the year 1000 allowing for a spin-up period to achieve steady state (e.g., net ecosystem production (NEP) of the simulated forest watershed remains close to zero) before anthropogenic disturbances are applied to the model after 1850. The model was run on a monthly time step. Monthly values of atmospheric deposition of all major elements and meteorological data (minimum and maximum temperature, precipitation, solar radiation) are input for the entire simulation period.

Wet deposition has been monitored at Huntington Forest in the central Adirondacks (43° 58' N, 74° 13' W) since 1978 through the National Atmospheric Deposition Program (NADP NY20). Daily meteorological data (e.g., maximum, minimum air temperature and precipitation) are also available at this site since 1940 provided by State University of New York College of Environmental Science and Forestry (SUNY-ESF). Huntington Forest was chosen as a benchmark to estimate wet deposition for the Adirondack lake-watersheds to which we applied PnET-BGC. Atmospheric wet deposition and climatic drivers for the entire simulation period were reconstructed as follows:

Pre-anthropogenic conditions (i.e., before 1850) were estimated from precipitation chemistry in remote areas, with volume-weighted mean concentration of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  varying from 3 to 10 and from 2 to 5  $\mu\text{eq L}^{-1}$ , respectively ([Galloway et al., 1984](#)). Precipitation concentrations in the middle of the range observed for remote sites were used as pre-anthropogenic conditions and are about 35% of current values observed in the Adirondacks. Note a sensitivity analysis indicated that model simulations of long-term projections of the acid–base status of soil and surface waters are relatively insensitive to the estimated pre-anthropogenic deposition ([Appendix E, Table E1](#)). The reconstruction of atmospheric wet deposition assumed a linear ramp from pre-anthropogenic values in 1850 to estimated values in 1900.

Estimates of wet deposition of major solutes for the historical period were based on historical emission estimates. Linear regression models were developed between national emissions and measured concentrations of wet deposition at an NADP site (NY20) for the years 1979–2010 ([Appendix B, Table B6](#)). These regression models were utilized to reconstruct historical (1900–1978) wet deposition based on historical U.S. emissions ([Nizich et al., 1996](#); <http://www.epa.gov/ttn/chief/trends/index.html>).

PnET-BGC estimates dry deposition of chemical constituents based on user inputs of dry to wet deposition ratios. Uniform spatial dry to wet deposition ratios ([Appendix B, Table B7](#)) for base cations, ammonium ( $\text{NH}_4^+$ ) and chloride ( $\text{Cl}^-$ ) were estimated from throughfall studies at the Huntington Forest ([Shepard et al., 1989](#)). Since a consistent temporal trend was not observed in dry to wet S and N deposition ratios among CASTNET and nearby NADP sites in the northeastern US (<http://epa.gov/castnet/javaweb/index.html>), a constant dry to wet deposition ratio over time was assumed. This assumption induced some uncertainties in the simulations which require further investigation. Spatial patterns in dry to wet deposition for the  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  were calculated based on spatial models developed by [Ollinger et al. \(1993\)](#) and then modified by [Chen and Driscoll \(2004\)](#) to incorporate effects of forest composition ([Cronan, 1985](#)). The forest composition for each study watershed was determined through a GIS data layer obtained from the National Land Cover Database ([http://www.mrlc.gov/nlcd06\\_data.php](http://www.mrlc.gov/nlcd06_data.php)).

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