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Experimental Inducement of Nitrogen Saturation at the Watershed Scale

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Introduction

Atmospheric deposition is altering the nitrogen cycle in many northern forested ecosystems (*1–3*). Most low-elevation forested ecosystems in midlatitude temperate regions are N-limited, with efficient internal N cycling leading to minimal NO_3^- losses to surface waters and groundwaters (*4–6*). Increased leaching of N below the rooting zone is indicative of a condition of "nitrogen saturation" (*1, 3*). Nitrogen saturation represents a fundamental shift in forest N dynamics, with adverse implications for forest health and water quality (*4, 7, 8*). In some systems, symptoms of forest decline have closely followed the onset of N loss at the catchment level (*9*), perhaps due to N–Mg imbalances (*6*), nutrient cation loss as counterions with NO_3^-, or mobilization of inorganic Al at toxic concentrations at the lowered pH (*10, 11*).

Nitrate concentrations are increasing in some surface waters draining remote forested watersheds of the northeastern United States and northern Europe (*9, 12–16*). To date, field evidence for nitrogen saturation has been largely from trends in NO_3^- concentrations for a few streams with long-term records (*12, 13, 16*) or from differences in concentrations from populations of lakes sampled years apart (*15*). In some streams and lakes of the mid-Appalachian, New York, and Pennsylvania regions of the United States, NO_3^- concentrations are now elevated for much of the year (*12, 13*). In southern Norway, lake surveys conducted 10 years apart demonstrated widespread increases in NO_3^-, presumably due to continued atmospheric loading of nitrogen (*15*).

To address this issue, we have experimentally increased the N deposition to one of two watersheds of the Bear Brook watershed, Maine (BBWM). The site is in eastern Maine, (44°52′ N, 68°06′ W), 60 km west of New Brunswick, Canada, and 50 km inland from the Gulf of Maine. The paired East and West Bear Brook watersheds are on the upper 210 m of Lead Mountain (475 m). Forest cover at BBWM is 40–60-year-old mixed northern hardwood dominated by beech (*Fagus grandiflora*), with red spruce (*Picea rubens*) and balsam fir (*Abies balsamea*). Spodosols are developed on 0–5 m of till, which overlies intermediate-grade metapelites intruded by granite near the summit.

Experimental Section

Dry $(NH_4)_2SO_4$ has been applied bimonthly via helicopter since November 1989 to the 10.2-ha West Bear Brook catchment. The adjacent 10.9-ha East Bear Brook catchment serves as an untreated reference. Annual loading of N (total of wet plus dry) has been increased from 600 equiv ha^{-1} yr^{-1} ambient wet + dry to a total of 2400 equiv ha^{-1} yr^{-1} (Table I). The N additions have been isotopically enriched with ^{15}N since April 1991, enabling us to track the fate of the N addition over time (*17*).

Total N deposition to the West Bear Brook catchment is now ~25% higher than in some of the high-elevation, high-N-deposition regions of North America, but is less than that in areas of central Europe (Table I). Ammonium deposition in parts of central Europe is nearly as high as the experimental loading at BBWM (Table I). Our experimental treatment is modest relative to total N pools and to vegetative requirements. The loading rate of 25 kg of N ha^{-1} yr^{-1} is less than 25% of estimated vegetative requirements of 80–120 kg ha^{-1} yr^{-1} (*18*) and only ~10% of typical forest fertilization application rates of 200 kg ha^{-1} yr^{-1} (*19*). The annual experimental loading adds only 10% to the estimated total N pool of 2500 kg of N ha^{-1} in upper soil horizons at BBWM.

Precipitation inputs are measured weekly on a National Atmospheric Deposition Program schedule with Aerochem Metrics wet-only collectors located at the East Bear Brook outlet weir, a midelevation station, and at the summit. Stream discharge is recorded at 5-min intervals. Complete stream chemistry is determined for samples collected at least weekly. During episodes and periods of high base flow, samples are collected at least daily and as frequently as hourly in order to accurately characterize watershed mass balances.

Results and Discussion

Prior to experimental treatment, NH_4^+ concentrations were generally below detection in the two first-order streams draining each watershed. Nitrate concentrations varied in parallel in the two streams, ranging from virtually zero during the growing season, to a maximum of 43 μequiv L^{-1} in the spring (Figure 1). Approximately 50% of the N in wet deposition (75% of wet and estimated dry deposition of N) was retained within the catchments before treatment. This relatively low retention (*18*) suggests that the site was close to nitrogen saturation prior to treatment. The BBWM site has both a lower C:N ratio in soils and higher forest floor N concentration, compared to other low-elevation sites in Maine (*20*). Both factors may contribute to the potential for N saturation (*21, 22*).

In response to the experimental N applications to the West Bear Brook watershed, NO_3^- concentrations have more than tripled relative to the East Bear Brook reference (Figure 1). Mean values of monthly NO_3^- concentrations for the two streams were significantly different ($p > 0.05$)

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Table I. Comparison of Total Loading of Wet + Dry N to the West Catchment, Bear Brook Watershed, with Other High-Deposition Locations in the Northern Hemisphere[a]

	ambient wet NO_3^- (*a*)	ambient wet NH_4^+ (*b*)	est N dry dep (*c*)	exptl treat. (*d*)	est total (*e*)
this study (BBWM)	210	100	310	1800	2420
US NADP max[b] (*28*)	485	275	750	0	1510
GSMNP,[c] NC (*6*)	230	220	1490	0	1940
Gardsjon, Sweden (*26*)	400	330	730	2900[d]	4360
Solling, Germany (*29*)	700	860	1120	0	2680
Ysselsteyn, Neth. (*29*)	430	1570	1780	0	3780

[a] Ambient precipitation loading data are averages for the period 1988–1990, as available. Dry deposition is assumed to be equivalent to wet inputs when dry deposition estimates are not available at the site. Values are in equiv ha^{-1} yr^{-1}. [b] Bennett Bridge, NY. [c] Great Smokey Mountains National Park. [d] Experimental annual loading at Gardsjon is between 2100 and 3600 equiv ha^{-1} yr^{-1} depending on ambient precipitation. [e] Sum of columns *a*–*d*.

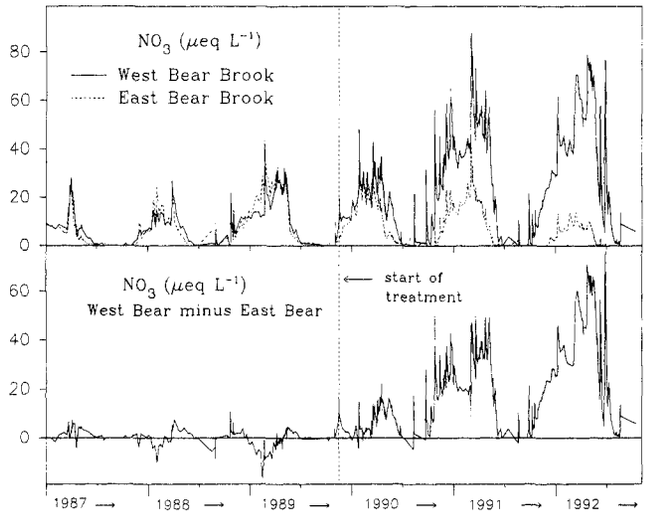

Figure 1. (Top panel) 1987–1992 record of NO_3^- concentrations in West and East Bear Brook. (Lower panel) Difference between NO_3^- concentrations in West Bear Brook draining the experimental catchment and the reference East Bear Brook.

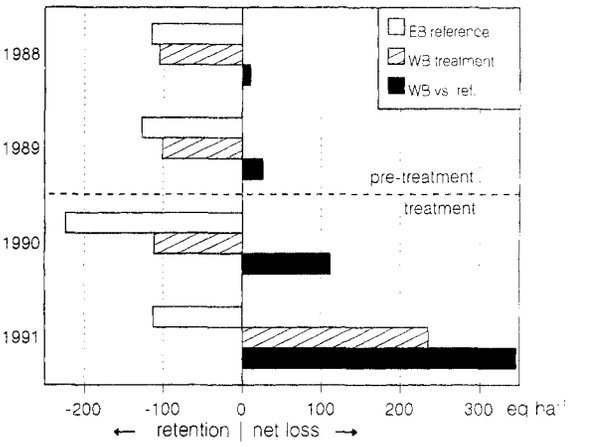

Figure 2. Mass balance estimates for the experimental and reference watersheds, 1988–1991. Data are watershed flux minus total wet inputs of N in precipitation. The black bars represent the response of West Bear Brook relative to the reference, calculated as the difference between East and West Bear. The response (black bar) would have the same value with or without the inclusion of dry deposition in the estimate. Water years are defined as Nov 1 through Oct 31 to accommodate the start of experimental treatment in Nov 1989.

only twice (both at summer low flow) prior to the beginning of experimental treatment. After treatment began, the difference was immediately significant ($p > 0.01$) in November and December 1989, and every month since March, 1990 ($p > 0.0001$), except during summer drought in 1990. Through October 1992, NO_3^- concentrations in West Bear Brook have ranged up to 88 μequiv L^{-1}. Chronic increases in NH_4^+ concentrations in streamwater have not occurred.

The experimental loading has induced concentrations of NO_3^- that often exceed the values presently observed in surface waters of the New York or mid-Atlantic regions. Differences between the treated and reference watersheds were especially pronounced during spring 1992. Nitrate concentrations in 1992 were unusually low in the reference stream due to low spring discharge, but concentrations in the experimental watershed stream remained high (Figure 1). The weighted mean concentration and annual flux of both NO_3^- and NH_4^+ from the East Bear Brook reference watershed have declined since a peak in mid-1989. This change in NO_3^- export from the East Bear catchment is in large part due to a decrease in the frequency of high-discharge events since late 1989. The large increases in NO_3^- concentrations in West Bear Brook have occurred concurrently with the decreases in East Bear Brook.

The flux of NO_3^- from the West Bear Brook catchment has more than doubled, compared either to the pretreatment period of record for West Bear Brook or to the reference East Bear catchment. The increased flux of NO_3^- has doubled total N output, but output of non-NO_3^- forms of N apparently has not changed. On a mass balance basis (watershed flux minus wet input of NO_3^- plus NH_4^+), the mass balance for total inorganic N in the West Bear catchment was relatively constant during 1988–1990 (Figure 2). In 1991, the output flux of inorganic N from the West Bear treatment watershed exceeded the ambient wet input. Compared to the East Bear reference, the West Bear catchment exhibited increasing losses of N even in the first year of treatment (Figure 2).

Retention of the experimental N additions within the watershed has decreased each year. During the first year of treatment (November 1989–October 1990), 94% of the additional N was retained by the system, based on mass balance calculations. In year 2, 81% was retained, and thus far for year 3, 74% has been retained. Nevertheless, the system continues to accumulate N, but at an increasingly slower rate over the last 3 years.

The treatment has resulted in a fundamental shift in the seasonal discharge–NO_3^- concentration relationship in West Bear Brook. We examined the relationship between NO_3^- concentrations and discharge during the spring season transition to summer base flow (generally mid-April through June). Prior to treatment, the regression of NO_3^- concentration as a function of discharge had a zero discharge intercept of approximately zero NO_3^- (Figure 3a). After 6 months of treatment (spring 1990), concentrations of NO_3^- in West Bear Brook had increased relative to the reference stream, increasing the slope of the discharge–NO_3^- regression relative to the reference stream (Figure 3b). However, NO_3^- concentrations in West Bear Brook

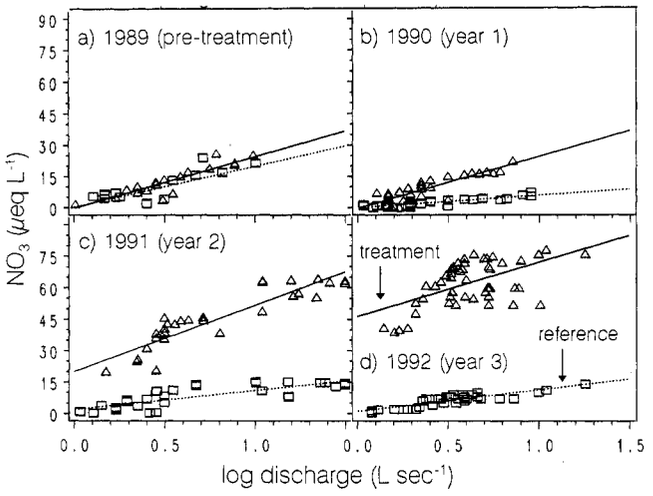

Figure 3. Changes in the relationship between NO_3^- concentrations in streamwater and stream discharge, spring seasons 1989–1992. The experimental catchment (West Bear Brook) is indicated in triangles. The reference stream shows a zero NO_3^- intercept at zero discharge in each year. The treated catchment also shows a zero NO_3^- intercept after 6 months of manipulation, but the slope of the relationship has increased compared to the pretreatment period. The zero discharge intercept increased in each of the next 2 years due to the experiment.

were still zero when extrapolated to zero discharge. The zero discharge intercept of the regression increased to 20 μequiv L^{-1} by the spring of 1991 (Figure 3c), and to 43 μequiv L^{-1} in 1992 (Figure 3d), indicating increased NO_3^- in soil solution in excess of uptake capacity early in the growing season. In 1992, NO_3^- concentrations in West Bear Brook were chronically elevated for most of the summer (Figure 1), with concentrations reaching unprecedented summer values as high as 67 μequiv L^{-1}. We interpret this nearly chronic NO_3^- loss to indicate leaching of N to the deeper hydrologic flow paths that sustain summer base flow.

Nitrogen saturation in forested watersheds apparently proceeds in a continuum of two stages. The first stage is indicated by chronic or near-chronic NO_3^- export during the nongrowing season. BBWM was transitionally into this stage when treatment began. Recent data from a NH_4NO_3 treatment to a 0.5-ha subcatchment at Gardsjon, Sweden, indicate that N loading at ~2900 equiv ha^{-1} yr^{-1} can induce winter export of N within 1 year (*23*). The second stage of N saturation is manifested by chronic loss of N to surface waters even during summer base flow, such as evident in regions of the eastern United States (*12–14, 16*) and Scandinavia (*15*). We interpret our data to reflect incipient N saturation at BBWM.

Both the rapidity and magnitude of the increase in NO_3^- flux from the treated catchment were unexpected given the historically low rates of N deposition (*24*), the modest increase in loading (Table I), and the generally N-limited status of forests in the northeastern U.S. region. However, these loss rates are consistent with several measures at BBWM indicating high N availability, such as high foliar N, low soil C:N, and high rates of net N mineralization (*20*). Most net retention of chronic N amendments to forests is incorporated into soils (*20, 25*). This retention capacity can be saturated by chronic N additions (*25, 26*). An important result of the research at BBWM is the demonstration that forest ecosystems in areas of low N deposition may be nearly N saturated, even when export of N is very low. Net N retention may rapidly decline when N inputs increase (*23*). These findings have important implications for policy relating to N emissions. Continuing research at BBWM, including analysis of the distribution of ^{15}N added with the treatment (*17*), will further our understanding of the processes controlling N retention in forest ecosystems.

One of the key issues to be addressed at BBWM is whether, or at what rate, N saturation is reversible after experimental treatments cease. The 1990 U.S. Clean Air Act mandates a smaller reduction in N than for S through the year 2000. During the past decade, SO_4 concentrations decreased in both precipitation and surface waters in the northeastern United States (*12, 16, 27*), but acid neutralizing capacity (ANC) in surface waters generally did not increase in response. This lack of response in ANC may be in part the result of the increases in NO_3^- loss from many watersheds. If only modest atmospheric loadings of N are sufficient to induce N leaching to surface waters, the efficacy of the Clean Air Act in aiding recovery of surface waters from acidification will be in question.

Conclusions

Atmospheric nitrogen deposition to forested ecosystems in excess of vegetative demand may lead to a condition of terrestrial "nitrogen saturation". The subsequent increases in N export to surface waters are symptomatic of fundamental changes in terrestrial nutrient processing. We have experimentally induced incipient N saturation in a paired-catchment experiment at the Bear Brook Watershed in Maine (BBWM). Using modest loading rates of N compared to typical forest fertilization rates or the N pool in forest soils, the experiment has resulted in large increases in NO_3^- concentrations in streams. Nitrate flux from the treated catchment has increased from 200 to more than 500 equiv ha^{-1} yr^{-1}. Seasonal patterns in stream NO_3^- concentrations have been fundamentally altered by the treatment, with NO_3^- loss now occurring nearly all year from the experimental watershed. These results suggest that N saturation, soil acidification, and altered N cycling in forested watersheds may be induced at lower rates of N deposition than previously believed.

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