Biogeochemistry (2007) 84:333–348
DOI 10.1007/s10533-007-9125-z

ORIGINAL PAPER

Symptoms of nitrogen saturation in an aggrading forested watershed in western Maryland

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Received: 28 July 2006 / Accepted: 26 February 2007 / Published online: 31 May 2007

Abstract The objective of this study was to evaluate the nitrogen (N) biogeochemistry of an 18–22 year old forested watershed in western Maryland. We hypothesized that this watershed should not exhibit symptoms of N saturation. This watershed was a strong source of nitrate (NO_3^-) to the stream in all years, with a mean annual export of 9.5 kg N ha^{-1} $year^{-1}$ and a range of 4.4–18.4 kg N ha^{-1} $year^{-1}$. During the 2001 and 2002 water years, wet deposition of inorganic N was 9.0 kg N ha^{-1} $year^{-1}$ and 6.3 kg N ha^{-1} $year^{-1}$, respectively. Watershed N export rates in 2001 and 2002 water years were 4.2 kg N ha^{-1} $year^{-1}$ and 5.3 kg N ha^{-1} $year^{-1}$, respectively. During the wetter water years of 2003 and 2004, the watershed exported 15.0 kg N ha^{-1} $year^{-1}$ and 18.4 kg N ha^{-1} $year^{-1}$, rates that exceeded annual wet deposition of N by a factor of two (7.5 kg N ha^{-1} $year^{-1}$ in 2003) and three (5.5 kg N ha^{-1} $year^{-1}$ in 2004). Consistent with the high rates of N export, were high concentrations (2.1–3.3%) of N in foliage, wood (0.3%) and fine roots, low C:N ratios in the forest floor (17–24) and mineral soil (14), high percentages (83–96%) of the amount of mineralized N that was nitrified and elevated N concentrations (up to 3 mg N l^{-1}) in soil solution. Although this watershed contained a young aggrading forest, it exhibited several symptoms of N saturation commonly observed in more mature forests.

Keywords Nitrogen saturation · Forested watersheds · Nitrogen dynamics · Atmospheric nitrogen deposition

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Introduction

Nitrogen saturation is a set of changes that occurs in the biogeochemical cycling of N in terrestrial ecosystems when N availability, usually from chronically elevated atmospheric N deposition, exceeds the capacity of the ecosystem to store or cycle it (Aber et al. 1989, 1998). The capacity of undisturbed forested ecosystems to retain N or to eventually display symptoms of N saturation is believed to be related to several factors, including forest type, atmospheric N deposition rates, forest age, and

disturbance history. In the absence of disturbance, young, early successional forests are expected to actively accumulate and store N as forest biomass increases, with relatively low losses of NO_3^- to stream and ground waters (Aber et al. 1989). A condition of N availability exceeding the demand of vegetation and soils to store and cycle N is expected to occur as growth slows in older forests. Older forests are more likely to export NO_3^- in stream waters, one symptom of N saturation (Aber et al. 1989; Stoddard 1994; Peterjohn et al. 1996, 1999; Goodale et al. 2003). For example, Vitousek (1977) reported that young northern hardwood forests exhibited lower, seasonally variable NO_3^- export, with lows occurring during the growing season. More mature forested watersheds, however, had high and relatively constant stream water NO_3^- export. Goodale et al. (2003) resampled these streams and reported lower NO_3^- concentrations in all streams compared to the earlier sampling (Vitousk 1977), but streams in old-growth forests still had higher NO_3^- concentrations than streams in successional forests. Clearly, forest age may be an important controller of stream water NO_3^- export and may influence the onset of other symptoms of N saturation.

Atmospheric N deposition may also influence regional patterns of NO_3^- export from forested watersheds. For example, Binkley et al. (2004) reported that NO_3^- concentrations tended to be higher in watersheds dominated by hardwood forests in the northeast relative to other regions in the United States (U.S.). They attributed these higher stream water NO_3^- concentrations to higher atmospheric N deposition rates. This suggests that forests in areas with high rates of atmospheric N deposition could have relatively high stream water NO_3^- concentrations and export rates. This is consistent with the high rates of NO_3^- export from the mature forested watersheds at the Fernow Experimental Forest in West Virginia (Peterjohn et al. 1996).

Forested watersheds in western Maryland receive some of the highest wet deposition rates of N in the eastern U.S. (Castro and Morgan 2000). Previous work showed that 60–80 year old forested watersheds were significant sources of NO_3^- to stream waters (Williard et al. 1997; Castro and Morgan 2000). Another study in this region reported elevated stream water NO_3^- export from a 24−year old aggrading forested watershed that nearly equaled NO_3^- exported from an undisturbed mature forested watershed (Peterjohn et al. 1996). Nitrogen export from this young forest is not consistent with studies that report young forests are strong sinks for N with little to no NO_3^- export to surface and ground waters (Emmett et al. 1993; Ohrui and Mitchell 1997).

To increase our understanding of N dynamics in young forests in regions with chronically elevated N deposition rates, we evaluated the N biogeochemistry of a young actively growing forest in western Maryland. More specifically, we developed annual N and water budgets for several years, and measured key variables associated with forest N cycling, including: tissue N concentrations; rates of N resorption from foliage; carbon (C) and N concentrations in the forest floor and mineral soil horizons; net N mineralization and net nitrification; and concentrations of dissolved N in stream water and soil solution. Because of the young age of the aggrading forest in our study watershed, we hypothesized that this watershed should not display symptoms of N saturation.

Site description and sampling locations

This study was conducted in a 3 ha watershed of the East Branch of Neff Run, locally known as TNEF (Fig. 1). The TNEF watershed (39° 35′ 47″ N; 78° 54′ 29″ W) is in the Appalachian Plateau physiographic province of western Maryland. TNEF is on the northwest slope of Dan's Mountain near Frostburg, Maryland. TNEF is relatively high in elevation

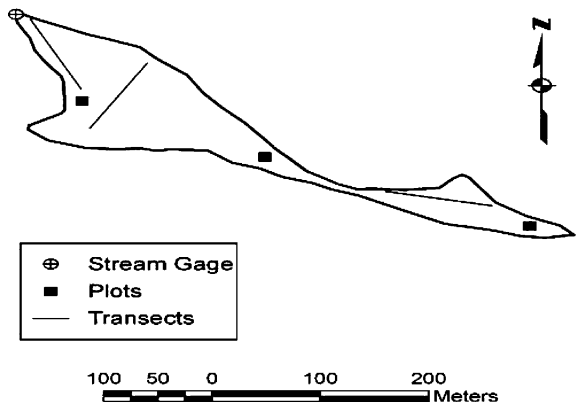

Fig. 1 Map of the TNEF watershed with the permanent sampling plots and permanent transects

(671–780 m above sea level.) with steep (averaging 9.9°) slopes. TNEF is drained by an ephemeral stream that can be dry in late summer and early fall (Negley and Eshleman 2006).

As part of our small watershed research program, TNEF is our control watershed for comparison with other small watersheds that were disturbed by coal mining activities. TNEF is a completely forested watershed with plant species, biomass and soils characteristics (described below) typical of other regrowing forests in the region.

TNEF is in the humid-continental climate zone, with mild summers and cold winters. Long-term (1971–2000) average monthly air temperatures range from −8.6°C in January to 26°C in July, with annual average (1971–2000) precipitation of 1132 mm uniformly distributed throughout the year (Maryland State Climatologist's Office: http://www.atmos.umd.edu/~climate/).

The TNEF forest was dominated by sugar maple (*Acer saccharum* Marsh., 28.6%), black cherry (*Prunus serotina* Ehrh., 24.5%), northern red oak (*Quercus rubra* L., 10%) and red maple (*Acer rubrum* L., 9.5%). Sugar maple and black cherry accounted for 60% of the total aboveground living biomass of 108.6 Mg ha^{-1}(K. Kuers, unpublished data). Sugar maples were equally distributed across TNEF with larger individuals in the middle of the watershed. Black cherry was most prevalent in the lower portion while oaks were more common in the upper portion.

Based on historical records and extensive field observations, the stand structure in TNEF indicates that a commercial grade clear-cut took place 18–22 years before our study (K. Kuers, unpublished data). This commercial clear-cut left a small number of older, non-merchantable stems, and a few smaller stems ranging in diameter from 3–5 inches. TNEF is clearly dominated by a young (18–22 year old) cohort, which accounts for 80% of the total basal area of 22 m^2 ha^{-1} and 95% of the 5916 stems ha^{-1}. Inventory of the ground vegetation across TNEF revealed few tree seedlings (only a small number of black cherry seedlings <1 year-old), very little herbaceous vegetation, and obvious mortality of smaller (<6 inches) stems of the more shade intolerant species. The majority of the smaller stems (black birch (*Betula lenta* L., black cherry, sassafras, aspen, and black locust) require moderate to high light conditions. These features could only have developed with open conditions typical of clear-cuts and would not occur in a stand developing from a selective harvest. These observations suggest that TNEF is structurally and developmentally representative of a young, aggrading forest.

TNEF is underlain by soils of the Cockport soil series, which are very stony silt loams formed from weathered acid sandstone (Stone and Matthews 1974). The Oe/Oa soil horizon had an average bulk density of 0.032 ± 0.015 g cm^{-3} (n = 45), an average pH of 4.21 ± 0.02 in 0.01 M $CaCl_2$ (n = 38), an average thickness of 6.4 ± 0.6 cm (n = 33), a C:N ratio of 24 (n = 45), total P concentration of 2.5 ± 0.6 mg g^{-1} (n = 30) and a Ca^{+2} concentration of 2.3 ± 0.8 mg g^{-1} (n = 30) (Simmons and Currie 2005). The upper 10-cm of mineral soil had an average bulk density of 0.98 ± 0.034 g cm^{-3} (n = 45), pH of 3.97 ± 0.03 in 0.01 M $CaCl_2$ (n = 44), a C:N ratio of 14 (n = 45), total P concentration of 1.2 ± 0.8 mg g^{-1} (n = 30) and Ca^{+2} concentration of 2.2 ± 0.8 mg g^{-1} (n = 30) (Simmons and Currie 2005).

Methods

Permanent plots and transects were established in the summer of 1999 (Fig. 1). The 3 permanent plots were positioned along the length of the watershed to allow sampling across an elevation gradient. Each permanent plot was 20 × 20 m and was divided into sixteen 5 × 5 m subplots. These subplots were used to measure net N mineralization and net nitrification rates and to sample litterfall and soil solution. Each of the 3 permanent transects was 100 m in length and oriented 120° from the other two. These transects were located across the length of the watershed. Along each transect, samples of the forest floor, mineral soil and fine roots were collected for chemical and physical analyses. Nineteen 40 m^2 circular plots were established in a fixed 80 × 80 m grid across TNEF in order to systematically sample aboveground vegetation (plots not shown in Fig. 1).

Carbon and nitrogen in vegetation and soils

In mid July 2001, tissue samples (green foliage, stems, branches and boles) were collected from 162 trees; 27 trees from 6 different species: black cherry,

sugar maple, red maple, northern red oak, black birch, and chestnut oak (*Quercus prinus* L.). These 6 tree species represent over 85% of the basal area in TNEF. To examine the effect of tree size and elevation on C and N concentrations, the 27 trees from 6 different species were selected from each of 3 size classes (0–5 cm, 5.1–15 cm and greater than 15 cm) in each of 3 elevation zones: 671–713 m, 714–735 m and 736–774 m.

Sample collection varied based on the type of tissue. For trees less than 5 cm diameter at breast height (dbh), the stem was cut at ground level and a 2 cm thick disk was collected at a height of 1.3 m. Twig samples, approximately 2 cm long, were collected at mid-length of 3–5 branches from each tree. Fifty to 75 leaves were collected from the upper third of the crown of each tree. For trees greater than 5 cm dbh, a 3 cm thick bole sample was removed from the tree with a drill at 1.3 m height. Twig and leaf samples were collected by pruning pole from smaller trees and by shotgun for tall trees. Twig samples consisted of 2–4 cm branch segments from 3 to 5 branches and 50 to 75 leaves from the upper third of the crown of the tree.

Samples were also collected from the dominant shrub in the watershed, *Hamamelis virginia* L. (witch hazel). Five stems less than 2.5 cm (at ground level) and 5 stems greater than 2.5 cm were sampled in each of the 3 elevation zones, for a total of 30 stems. Stem samples consisted of 2 cm stem samples at 0.5-m intervals along the stem. Leaf samples consisted of 50–75 leaves from the entire crown of the shrub.

Foliar tissue samples were pooled for each species by size class in each elevation zone for a total of 9 samples per species. Bole and branch samples were pooled by species and size class for a total of 3 samples per species. Witch hazel samples were pooled into 2 size classes at each elevation zone for a total of 6 foliage and 6 stem samples. Samples were dried at 70°C, ground in a Wiley mill to pass through a 60-mesh sieve, and analyzed for total N and total C using dry combustion on a CHN analyzer (NC-2100; Carlo Erba Instruments, Inc.). Subsamples were dried at 105°C and the total N and total C concentrations were corrected for a drying temperature of 70°C (typically less than 1% difference compared to 105°C). Average N concentrations and the C:N ratios were computed for all composited aboveground tissues (foliage, stems, branches, and boles).

In mid-July 2003, tissue sampling was repeated on chestnut oak, red maple, and northern red oak to compare tissue N concentrations in dry (2001) and wet (2003) years. These species were selected because they had the highest (chestnut oak), mid-range (northern red oak) and lowest (red maple) foliar N concentrations in 2001. Three trees from each of the same size classes (0–5 cm; 5.1–15 cm; and greater than 15 cm) were sampled for each species, using the same sampling and analysis protocols. Samples were composited by size class for a total of 3 samples per species for each of the tissue types (foliage, bole, and small branches).

Thirty samples of the forest floor and mineral soil were collected at evenly spaced locations (approximately 3.3 m) along each of the 3 permanent transects for a total of 90 samples. Adjacent samples were composited to generate a total of 45 samples for analysis. We used a 10×10 cm square template to quantitatively sample the Oi (litter) and Oe/Oa (humus) layers. The upper 10 cm of mineral soil below the template was sampled using a 5-cm diameter bulk density core. Subsamples of forest floor and mineral soil were dried in the laboratory at 70°C for 48 h and then ground to pass through a 60-mesh sieve using a Wiley mill and jar mill, respectively. Both forest floor and mineral soils were analyzed for total C and total N with the CHN analyzer. A 70°C to 105°C drying correction was applied to all samples. The C:N ratios were calculated for the Oi, Oe/Oa and upper 10 cm of mineral soil.

Total C and N contents were measured in fine roots in the forest floor and mineral soil from 15 composite samples collected along each transect using the 10×10 cm template. Six adjacent template samples of 90 forest floor and 90 mineral soil samples were composited to ensure adequate material for analysis, keeping the horizons separate. Each dried (at 70°C) composite sample was ground in a Wiley mill to pass through a 60-mesh sieve and analyzed on the CHN analyzer as above.

Nitrogen in litterfall and N resorption

From September through November of 1999 and 2000, autumn litterfall was collected on a monthly basis in each plot using 10 randomly located 2320 cm^2 baskets per plot. Every 4 weeks, litter in each basket was collected and pooled within each plot. It was then sorted by species, dried at 70°C, and

weighed. Subsamples of each species were ground in a Wiley mill to pass a 60-mesh sieve and analyzed for total C and total N concentrations.

Nitrogen resorption efficiency was calculated for each species using the average litter N concentration (1999 and 2000) and the 2001 green tissue concentrations as follows: % N resorption = (green foliar percent N concentration—litter percent N concentration)/green foliar N percentage × 100. While this value is not completely comparable to resorption calculated for leaves that are produced and shed during a single growing season, the method was reasonable because litter N concentrations and litter biomass did not differ over the 2 year period and green foliage N concentrations remained the same or increased slightly in a second year of measurement. Our N resorption estimates are conservative because green foliage N concentrations were the same or higher in a second year of measurement.

Soil solution

In July 2000, 6 tension lysimeters (Soil Moisture Equipment Corporation, Model 1900) were installed in each of the three 20 × 20 m plots. Three tension lysimeters sampled soil solution 15 cm below the forest floor and 3 lysimeters sampled soil solution 60 cm below the rooting zone. Soil solution samples were collected monthly in the fall of 2000 and in the spring, summer and fall of 2002, 2003 and 2004. To collect soil solution, pressure inside the lysimeter was reduced to -50 centibars 24 h before sample collection in clean high-density polyethylene bottles. In the laboratory, samples were filtered through 0.45 μm filters and a 50 ml subsample was frozen for subsequent analysis of NH_4^+, NO_3^- and total dissolved N using a Lachat QuickChem 8000 Flow Injection analysis system. In 2000, NH_4^+ and NO_3^- concentrations were measured. In 2002, 2003 and 2004, NH_4^+, NO_3^- and total dissolved N concentrations were measured. The dissolved organic N concentration was estimated as the difference between total dissolved N and the sum of the NH_4^+–N and NO^-_3–N concentrations.

Net nitrogen mineralization and net nitrification

During the 2000 and 2002 growing seasons, monthly *in situ* rates of net N mineralization and net nitrification were measured in 3 randomly selected subplots in each of the 3 permanent plots using the buried bag technique (Aber et al. 1993), for a total of 9 replicates per 4-week sampling period. During each monthly sampling period, we collected 10 cm of mineral soil in adjacent pairs of 5.4-cm diameter cores. These cores were separated into organic and mineral horizons and placed into separate gas permeable polyethylene bags. One core from each pair was placed into the original location and incubated *in situ* for approximately 4 weeks. The other member of each pair was brought to the laboratory, where it was homogenized through a 2-mm mesh sieve and a 10 g field moist sub-sample was extracted with 100 ml of 2 M KCl for 1 h on an orbital shaker. The extract was filtered (nominal pore size 0. 45 μm) and frozen until it was analyzed for NH_4^+ and NO_3^- using the Lachat system. Another 10 g sub-sample was oven-dried at 105°C for 48 h to determine the moisture content. At the end of each 4-week incubation period, the incubated member of each pair was brought to the laboratory and treated the same as the initial sample. The net N mineralization rate was calculated as the difference between the extractable NO_3^- and NH_4^+ in the *in situ* incubated sample and the extractable NO_3^- and NH_4^+ in the initial sample per unit dry mass of soil. Net nitrification was calculated as the difference between NO_3^- in the incubated and initial samples per unit dry mass of soil. Soil bulk densities were then used to scale these rates to kg N ha^{-1} over each time period, which were then summed to produce annual rates (kg N ha^{-1} $year^{-1}$).

Stream water discharge and N fluxes

Stream water discharge was continuously monitored using a pre-fabricated Parshall flume attached to a stilling well equipped with a Stevens Type A water level recorder (later replaced with a Unidata Model 6541 C digital recorder); analog stage records were field checked and digitized in the laboratory prior to applying the flume rating obtained from the manufacturer (Free Flow, Inc., Omaha Nebraska). A polyethylene liner was placed in the stream channel in front of the flume and between the wing-walls to direct all of the water into the flume.

Grab samples of stream water were collected weekly, biweekly or less frequently (during dry periods) and returned to the laboratory for analysis of NH_4^+ and NO_3^- concentrations. Samples were

filtered through 0.45 μm filters within 24 h and frozen until analyzed. Nitrate was measured by ion chromatography on a Dionex DX 500 instrument and NH_4^+ was measured using the Lachat system. Daily stream water N flux was computed as the product of a linearly-interpolated daily N concentration and the mean daily discharge. Monthly and annual (October 1–September 30 water-year) N fluxes for years 2000–2004 were computed by aggregating the continuous record of daily N fluxes over the appropriate time period.

Wet deposition of inorganic nitrogen

Precipitation was measured continuously using a Belfort Model 5-780 recording rain gauge fitted with a Belfort windshield. Weekly-integrated wet deposition samples for chemical analysis were collected using an AeroChem-Metrics Model 301 wet/dry collector following the procedures of the National Atmospheric Deposition Program (NADP) (NADP 2000; Castro and Morgan 2000). Ammonium and NO_3^- concentrations were measured in the laboratory using the Lachat system. Annual rates of NH_4^+ and NO_3^- deposition were estimated by multiplying the total annual precipitation from the Belfort by the annual volume-weighted average concentrations of NH_4^+ and NO_3^-. Annual rates are reported on an October 1–September 30 water year to be consistent with the stream water fluxes.

Statistical analyses

The General Linear Model (SPSS, Inc.) was used to test for effects of tree size, trees species, and elevation on foliar N concentrations, to test for effects of tree size on N concentrations in bole and small twig samples, and to test for the effect of year (wet vs. dry years) for those species sampled in more than one growing season. ANOVA and student's t-test were used to test for significant differences ($P < 0.05$) in soil solution N concentrations.

Results and discussion

Water balance

Total annual precipitation from 2000–2004 ranged from 933 mm to 1399 mm with a mean of 1138 mm (Table 1), nearly identical to the long-term mean (1132 mm) from the Frostburg weather station (Maryland State Climatologist's Office: http://www.atmos.umd.edu/~climate/). Water years 2000 and 2002 were relatively dry years, with total annual precipitation nearly 20% less than the long-term (1971–2000) mean. Total annual precipitation in water year 2001 was only 5% below the long-term mean. Total annual precipitation in 2003 and 2004 was approximately 20% greater than the long-term mean. Thus, our five-year study was conducted over a wide range of annual precipitation inputs.

Not surprisingly, annual stream water runoff was highly variable during this period (2000–2004), ranging from 172 mm to 657 mm (Table 1), with a mean of 367 mm. Annual water yields (stream discharge divided by precipitation input) ranged from 18% to 49%. During water years 2000–2002, average annual stream water runoff was 190 mm, average percent water yield was 19%, and evapotranspiration accounted for 81% of the water loss from TNEF. These water yields are well below the 50−year average water yield (49%) for the nearby Savage River (Negley and Eshleman 2006). During the wetter water years of 2003 and 2004, runoff exceeded 600 mm $year^{-1}$ and the annual water yields (44 and 49%) were similar to the long-term average water yields for the Savage River and other temperate forests (Campbell et al. 2004).

Nitrogen input-output budget

Atmospheric N deposition

Over our five-year study, annual wet deposition of inorganic N ranged from 4.8 kg N ha^{-1} $year^{-1}$ to 9.0 kg N ha^{-1} $year^{-1}$, with an average of 6.6 kg N ha^{-1} $year^{-1}$ (Table 2). For this period, annual wet deposition to TNEF usually exceeded wet deposition rates for nearby NADP sites (http://nadp.sws.uiuc.edu/) in West Virginia (WV18, 5 to 5.8 kg N ha^{-1} $year^{-1}$), central Pennsylvania (PA 42, 5.5 to 7.0 kg N ha^{-1} $year^{-1}$) and the eastern shore of Maryland (MD13, 3.8 to 6.3 kg N ha^{-1} $year^{-1}$). Elevated N inputs at TNEF were caused primarily by elevated wet deposition of NH_4^+. Most forest watersheds in rural areas in the eastern U.S. have volume-weighted annual average NH_4^+ concentrations in wet deposition ranging between 0.1 and 0.2 mg NH_4^+ l^{-1}

Table 1 Annual water balance for the TNEF watershed for water years 2000–2004[a]

Year	Total precipitation (mm)	Stream water runoff (mm)	Evapotranspiration (mm)	Water yield %	Evapotranspiration %
2000	933	209	724	22	78
2001	1072	189	883	18	82
2002	944	172	772	18	82
2003	1399	610	789	44	56
2004	1341	657	684	49	51

[a] Based on an October 1–September 30 water year. Data for water years 2000–2002 from Negley and Eshleman (2006)

Table 2 Volume-weighted annual average concentration of inorganic nitrogen in precipitation, annual wet depostion rates of inorganic nitrogen, and stream water export of nitrogen from the study watershed[a]

Year	Ammonium concentration (mg $NH_4^+ l^{-1}$)	Nitrate concentration (mg NO^{-1})	Wet deposition inorganic N (kg N ha^{-1} $year^{-1}$)	Stream water export ammonium (kg N ha^{-1} $year^{-1}$)	Stream water export (kg N ha^{-1} $year^{-1}$)
2000	0.31	1.21	4.78	Not measured	4.44[b]
2001	0.49	2.09	9.00	Not measured	4.16
2002	0.44	1.44	6.29	0.006	5.32
2003	0.35	1.18	7.54	0.011	15.02
2004	0.26	0.92	5.51	0.049	18.35
Average	0.37	1.37	6.62	0.022	9.46

[a] Precipitation N concentrations and deposition rates and stream water N export rates are calculated on an October 1–September 30 water year

[b] Stream water sampling began in December 1999. As a result, annual NO_3^- export for water year 2000 was calculated using data from December 1999 thru September 2000

(Campbell et al. 2004). However, volume-weighted NH_4^+ concentrations in wet deposition at TNEF ranged from 0.26 to 0.49 mg NH_4 l^{-1}, which is consistent with the elevated NH_4^+ concentrations reported for watersheds in western Maryland and West Virginia (Castro and Morgan 2000). These elevated NH_4^+ concentrations may reflect local agricultural operations. We have not measured dry deposition to TNEF, but nearby dry deposition sites in West Virginia, Pennsylvania, and central Maryland estimate dry deposition rates between 1.7 and 3.4 kg N ha^{-1} $year^{-1}$ (http://www.epa.gov/castnet/). If dry deposition at TNEF is similar to dry deposition at these sites, total N deposition to TNEF would range between approximately 8.2 and 9.9 kg N ha^{-1} $year^{-1}$. This does not account for cloud water deposition of N, which has been shown to contribute 5–35 kg N ha^{-1} $year^{-1}$ to high elevation forests in the northeastern U.S. (Lovett and Kinsman 1990). Cloud water deposition has not yet been quantified at TNEF, but TNEF is in the clouds for extended periods of time, particularly in the spring and fall. As a result, average total annual atmospheric deposition (dry, wet and cloud water) of N at TNEF may exceed 13 kg N ha^{-1} $year^{-1}$.

Annual stream water N export

Nitrate was the dominant form of inorganic N exported from TNEF, accounting for approximately 99% of the total inorganic N in stream water. Annual stream water export of NO_3^- ranged from 4.2 kg N ha^{-1} $year^{-1}$ in 2001 to 18.4 kg N ha^{-1} $year^{-1}$ in 2004 (Table 2). During water years 2001 and 2002, TNEF was a net sink for N, retaining 53% and 15% of the annual wet deposition of inorganic N, and exporting 4.2 kg N ha^{-1} $year^{-1}$ and 5.3 kg N ha^{-1} $year^{-1}$, respectively. During these years, N export from TNEF was within the range (0.1–5.7 kg N ha^{-1} $year^{-1}$) of annual N export rates for forested watersheds that had

not been disturbed for more than 50 years (Campbell et al. 2004). Amongst these relatively undisturbed watersheds, the highest NO_3^- export was from Watershed 4 at the Fernow Experimental Forest in West Virginia (Campbell et al. 2004). This watershed supports a mature deciduous forest that was last cut in 1905. Annual NO_3^- export rates from Watershed 4 ranged from 0.5 to 9 kg N ha^{-1} $year^{-1}$ from 1971–2004 (Christ et al. 2002; Wood, personnel communication) and may represent the best example of a N saturated forest in the U.S. (Peterjohn et al. 1999). During both normal and dry water years, stream water NO_3^- export from TNEF was similar in magnitude to NO_3^- export from Watershed 4 at the Fernow Experimental Forest and more mature and undisturbed forested watersheds in the northeastern U.S. (Peterjohn et al. 1999; Campbell et al. 2004; Binkley et al. 2004).

During the wet years of 2003 and 2004, TNEF was a net source of N, exporting 15 kg N ha^{-1} $year^{-1}$ and 18.4 kg N ha^{-1} $year^{-1}$, respectively. These annual N export rates were 3 to 4 times greater than the annual export rates in 2001 and 2002 and also 2 to 3 times greater than the annual wet deposition of inorganic N. These higher NO_3^- export rates were evidently due to the wetter conditions and increased hydrologic flow, which moved NO_3^- from storage in the watershed into the stream. For comparison, N export from Watershed 4 at the Fernow Experimental Forest was 5.5 kg N ha^{-1} $year^{-1}$ and 6.8 kg N ha^{-1} $year^{-1}$ for the 2003 and 2004 water years, respectively (Wood, personnel communication). Thus, TNEF exported 2 to 3 times more N than the more mature undisturbed forested watershed at the Fernow Experimental Forest. These high NO_3^- export rates are clearly unusual and strongly suggest that TNEF, with its young aggrading forest, is N saturated.

Seasonal patterns in runoff and N export

There were strong seasonal patterns in both water runoff and NO_3^- export (Fig. 2a, b). Highest runoff and NO_3^- export generally occurred between February and June with little runoff and NO_3^- export in the late summer, particularly in 2000, 2001 and 2002. During these water years, runoff and NO_3^- export reached a maximum of 70 mm $month^{-1}$ and 2 kg N ha^{-1} $month^{-1}$. During the wet years of 2003 and 2004, the same seasonal patterns were observed, but the runoff and export rates were much higher. Runoff rates peaked at 144 mm $month^{-1}$ with several monthly rates greater than 70 mm $month^{-1}$ and the monthly export rates approached 4 kg N ha^{-1}. Most of the monthly rates were close to 2 kg N ha^{-1}, which was the maximum in the dry years.

Seasonal patterns in the volume-weighted stream water NO_3^- concentrations were consistent with a forest that is N saturated (Fig. 2c). As a forest becomes N saturated, the reduced biotic demand for N is reflected in seasonal patterns of stream water NO_3^- concentrations. When a forest is N limited, NO_3^- concentrations in stream waters are very low or below detection limits throughout the entire year

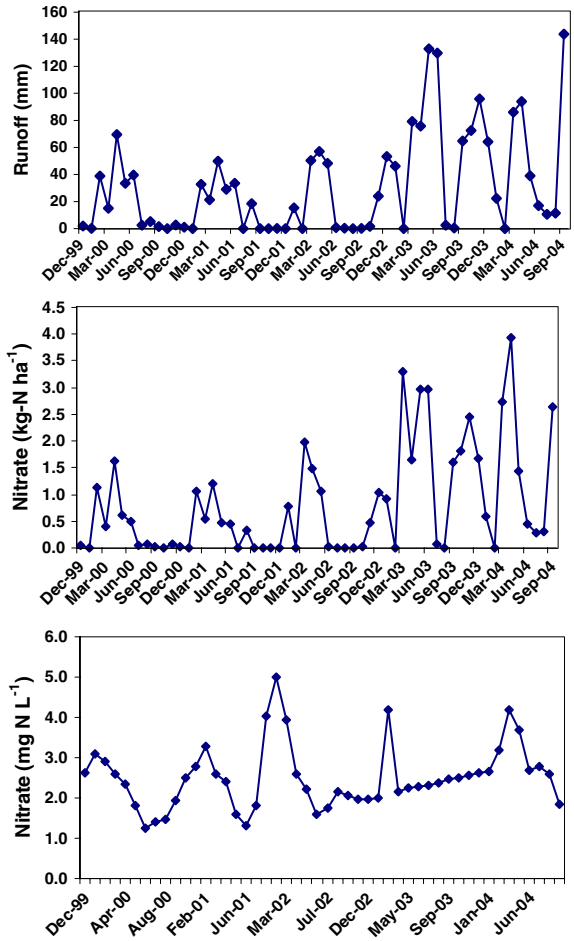

Fig. 2 Monthly water runoff (**A**), NO_3^- export (**B**) and volume-weighted monthly average stream water NO_3^- concentrations (**C**) for TNEF for the October to September 2000–2004 water years

(Stoddard 1994; Peterjohn et al. 1996). At the other extreme, a forest that is severely N saturated will have elevated stream water NO_3^- concentrations year round with little to no seasonal changes because the biotic demand is always satisfied by the elevated N pools. A forest in transition between these two extremes is likely to have elevated NO_3^- concentrations during much of the non-growing season, when the biotic sinks for N are minimal. During the growing season, transitional forests are likely to have low NO_3^- concentrations in stream waters because of the high biotic N demand in the watershed. TNEF had elevated stream water NO_3^- concentrations throughout dry, normal and wet water years. During the growing seasons, the volume-weighted stream water NO_3^- concentrations were always above 1.0 mg N l^{-1}. During the non-growing season, stream water NO_3^- concentrations increased, peaking at concentrations between 3 and 5 mg N l^{-1}. During the wet water years of 2003 and 2004, stream water NO_3^- concentrations in the summer growing season increased from 1.5 to 2.5 mg N l^{-1}. Stream water NO_3^- concentrations in TNEF were higher than those reported for the N saturated Watershed 4 at Fernow Experimental Forest that ranged from 0.4 to 1.6 mg N l^{-1} (Peterjohn et al. 1996) and other forested watersheds in the eastern U.S. (Campbell et al. 2004; Binkley et al. 2004). Based on the seasonal stream water NO_3^- concentrations, the forest in TNEF appears to be severely N saturated.

Nitrogen concentration in vegetation

Aber et al. (1989) suggest that one of the first signs of N saturation of temperate forests would be increased N concentrations in living (green) foliage. Magill et al. (1997) reported results consistent with this expectation from a N fertilization experiment at the Harvard Forest. They reported an increase in foliar N concentrations over the first 6 years of their fertilizer additions. In our study at TNEF, there are no long-term foliar N data. However, we measured foliar N concentrations from several tree species for comparison with results from comparable forests and forests used in fertilization studies. Tree species in TNEF would be expected to have elevated foliar N concentrations relative to the same tree species in regions with lower atmospheric N deposition.

Based on samples collected in mid-July 2001, tree species differed significantly in their foliage N concentrations (Table 3). Chestnut oak, black birch and black cherry had average foliar N concentrations over 3%. Northern red oak and sugar maple had foliar N concentrations that averaged 2.7% and 2.4%, respectively. Red maple had the lowest average foliar N concentration at 2.1%. The overall species-weighted average was 2.7% N. Neither tree size nor elevation had a significant effect on foliar, stem, or small twig N concentrations. This may be due to the lack of a dramatic elevation gradient (100 m at TNEF). In contrast, other studies have reported increased foliar N concentrations with increased elevation (Yin 1994).

Of the 3 tree species re-sampled in mid-July 2003, there was a statistically significant increase (compared to 2001) in the foliage N concentrations for red maple (2.1% vs. 2.5%) and northern red oak (2.7% vs. 3.1%) but not for chestnut oak (3.3% vs. 3.2%). Small branch N concentrations were also significantly higher in 2003 than 2001 for all three species (data not shown). This increase may reflect better growing conditions in 2003 because of the relatively high total annual precipitation or it may be due to natural variations that can range up to 25% between years (Magill et al. 1997; Magill et al. 2000).

Foliar N concentrations for tree species in TNEF, particularly black birch, black cherry, northern red oak and red maple, are among the highest reported in the literature (Table 3). These high foliar N concentrations are consistent with the relatively high foliar N concentrations measured in other areas of western Maryland and West Virginia (Townsend et al. 2003; May et al. 2005). Maples and oaks in TNEF had N concentrations 10–30% higher than concentrations in the same species in control plots at the Harvard Forest and were similar to or slightly greater than those reported for maples and oaks in fertilized plots (Magill et al. 2000). Red maple, black cherry and black birch in TNEF had N concentrations similar to or greater than those of a 24 year-old forest at the Fernow Experimental Forest in West Virginia that had been fertilized with $(NH_4)_2SO_4$ (Gilliam et al. 1996). Increased foliar N concentration is one of the key forest responses to chronic N additions (Aber et al. 1995) and provides evidence that TNEF is expressing another symptom of N saturation.

Table 3 Tissue nitrogen concentrations (std.dev.) for foliage, small twigs (<1 cm), and boles of the dominant watershed overstory and shrub species

Species	Nitrogen concentration (std. dev.)				Foliage ilterature range	Reference[a]
	Foliage		Small twig	Bole		
	2001	2003				
Acer rubrum	2.14 (0.22)	2.47 (0.025)[b]	0.32 (0.03)	0.32 (0.05)	1.3–2.2	1–4, 7–9, 11–15, 17, 19
Acer saccharum	2.36 (0.23)		0.35 (0.08)	0.28 (0.01)	1.5–2.2	3,6,10,14,16–18
Betula lenta	3.09 (0.40)		0.41 (0.09)	0.34 (0.13)	2.1–2.8	2–3, 11–13, 15
Hamamelis virginiana	2.46 (0.20)			0.39 (0.30)	1.7–1.8	5, 7
Prunus serotina	3.05 (0.42)		0.42 (0.11)	0.19 (0.08)	1.7–3.2	1–3, 8, 17, 19
Quercus prinus	3.25 (0.40)	3.2 (0.054)	0.45 (0.10)	0.25 (0.01)	1.3–2.8	4, 7–10, 13–15
Quercus rubra	2.74 (0.21)	3.1 (0.041)[b]	0.39 (0.07)	0.21 (0.07)	1.4–2.8	4, 7–8, 10–11, 13–15
Weighted average	2.68 (0.51)		0.39 (0.08)	0.27 (0.19)		

[a] Numbers refer to the following references: 1 (Abrams and Mostoller 1995), 2 (Adams et al. 1995), 3 (Bard 1945), 4 (Boerner 1984a), 5 (Boerner 1984b), 6 (Burton et al. 1993), 7 (Day and Monk 1977a), 8 (Johnson and Todd 1998), 9 (Johnson and Henderson 1989), 10 (Jose and Gillespie 1996), 11 (Lovett et al. 2004), 12 (Magill et al. 1997), 13 (Martin et al. 1998), 14 (Mitchell 1936), 15 (Mitchell et al. 1999), 16 (Morrison 1985), 17 (Ricklefs and Matthews 1982), 17 (Whittaker et al. 1979) and 18 (May et al. 2005)

[b] Statistically significant

High N concentrations were not limited to the foliage in TNEF. Average N concentrations in wood (0.3% N) was higher than N concentrations (0.03%–0.2% N) in trees in many eastern hardwood forests (Adams et al. 1995; DeWalle et al. 1995; Johnson and Todd 1998; Martin et al. 1998), although lower than the N concentration (0.5%–0.6%N) in southern Appalachian forests (Day and Monk 1977b). Elevated N concentration in wood is somewhat surprising because several N fertilization studies have not shown increased N concentrations in wood (DeWalle et al. 1995; Kashuba 1992). Unlike the N concentration in foliage, there were no statistically significant differences in the N concentration in small twigs and boles among different tree species (Table 3). Nitrogen in fine roots was quite high compared to roots in most northern hardwood forests (Hendricks et al. 2000) because of the large fine root biomass (7632 kg ha^{-1}) and low C:N ratio (21).

Nitrogen resorption from foliage

Currently, it is not clear how chronic N deposition affects N resorption by different tree species. Aber et al. (1989) suggested that chronic N additions may reduce N resorption, which would in turn lead to higher N concentrations in litterfall. However, Staaf and Stjernquist (1986) demonstrated that tree species appear to vary in their efficiency of N resorption and their response to variations in nutrient availability. Some species decrease resorption rates with increased N availability while others do not. In contrast, Aerts (1996) reported that N resorption by deciduous tree species did not change in response to increased nutrient supply.

Nitrogen resorption per unit leaf mass for vegetation in TNEF ranged from a low of 48% for the shrub witch hazel to a high of 78% for chestnut oak (Table 4). With the exception of witch hazel, all of the species studied resorbed over 50% of their leaf N. Chestnut oak and red maple are at the upper end of their respective ranges (Table 4). Sugar maple, northern red oak, black cherry and witch hazel are below the ranges reported in the literature (Table 4).

An intriguing result of our study is the high resorption efficiency of chestnut oak (78%), the species with the highest foliar N concentration. This result is consistent with the findings of Ostman and Weaver (1982) and Boerner (1984a) indicating no decrease in resorption for chestnut oak as site fertility

Table 4 Mass based % N resportion from senescing foliage of 5 tree species, 1 shrub species, and the average for the whole watershed

Species	Leaf litter N %8[a]	% N resorption	Literature range (Reference) [d]	
Acer rubrum	0.87	59[b]	26–76	(1,4,7)
Acer saccharum	1.14	52[b]	56–64	(3)
Hamamelis virginiana	1.32	48[b]	65–72	(2)
Prunus serotina	1.34	56[b]	60–79	(7)
Quercus prinus	0.71	78[b]	55–80	(1,5)
Quercus rubra	1.23	55[b]	60–83	(6)
Species average	1.16	58[b]		
Site Average		64[c]		

[a] Average of 2 years, 1999–2000

[b] %Resorption = (green N conc.—litter N conc.)/green N conc.× 10

[c] Site avg. % resorption = 1—(total litter N content/green foliage N content)

[d] References: 1 (Boerner 1984a); 2 (Boerner 1984b); 3 (Burton et al. 1993); 4 (Magill et al. 1997); 5 (Ostman and Weaver 1982) 6 (Son and Gower 1991) and 7 (May et al. 2005)

increased and shows further that this is true even when chestnut oak has extremely high foliar N. Boerner (1984a) also found that chestnut oak had a higher N resorption than red maple on a fertile, northeast facing slope. Interestingly, the opposite was true on a drier, southwest facing slope. The high resorption efficiency of chestnut oak in our study is one more example of a species that does not respond to high N inputs with reduced N resorption. Magill et al. (1997) also found that five years of N fertilization did not change foliar N resorption in black oak, red maple, black birch, or red pine at the Harvard Forest N Experiment. It was also interesting that sugar maple had a low rate of N resorption and a higher concentration of N in litter (1.14%) than is generally reported in the literature (0.43–1.0%; Burton et al. 1993). If sugar maple responds to increased fertility with decreased resorption efficiency and increased litter N concentrations, it could explain why studies have shown higher stream NO_3^- export from watersheds dominated by sugar maple (Mitchell et al. 2003).

Northern red oak and black cherry had the lowest N resorption and highest litter N concentrations. Low resorption and high litter N of northern red oak was surprising given the high resorption efficiency of chestnut oak and the findings that streams draining watersheds dominated by oak forests generally have lower NO_3^- concentrations than streams in watersheds without oaks (Lovett et al. 2000). For example, NO_3^- concentrations in watersheds dominated or co-dominated by northern red oak in Pennsylvania were 3–7 times less than those dominated by non-oak species (Lewis and Likens 2000). A similar pattern was observed in the Catskill Mountains of New York where streams that drained oak-dominated watersheds had the lowest NO_3^- concentrations, compared to streams that drained watersheds without oaks (Lovett et al. 2000). Since northern red oak, a species with a lower resorption efficiency, was dominant in these oak-dominated watersheds, low N resorption by northern red oak does not explain the low NO_3^- concentrations in stream waters.

Net nitrogen mineralization and net nitrification

In situ growing season annual average net N mineralization rates were 61 kg N ha^{-1} $year^{-1}$ in 2000 and 101 kg N ha^{-1} $year^{-1}$ in 2002. These rates are within the range (30–200 kg N ha^{-1} $year^{-1}$) of net annual N mineralization rates reported for temperate forests (Nadelhoffer et al. 1992; Pastor et al. 1984). In addition, our rates were within the range of annual rates (60–100 kg N ha^{-1} $year^{-1}$) reported for the control hardwood forest at the Harvard Forest N Experiment, but lower than those reported for the fertilized (15 g m^{-2} $year^{-1}$) hardwood stand (100–200 kg N ha^{-1} $year^{-1}$, Magill et al. 2000). Our rates were similar to rates for the deciduous forests in Watersheds 3 and 4 at the Fernow Experimental

Forest (67–78 kg N ha^{-1} $year^{-1}$; Peterjohn et al. 1999), but lower ($\sim$ 125 kg N ha^{-1} $year^{-1}$) than those reported by Gilliam et al. (2001). These higher rates may have been due to temporal variations and/or differences in analytical methods.

Another important change that occurs as a forest moves toward N saturation is increased rates of nitrification, which can increase the soil NO_3^- pool and NO_3^- leaching losses. Net nitrification is often very low in acid forest soils but it can be stimulated by increased N inputs. TNEF has acid soils (pH = 3.97) and therefore would be expected to have relatively low net nitrification rates. In contrast, TNEF has relatively high rates of net nitrification compared to the net mineralization rates. *In situ* net nitrification rates in TNEF in 2000 and 2002 were 50 kg N ha^{-1} $year^{-1}$ and 97 kg N ha^{-1} $year^{-1}$, respectively. Another key indicator of N saturation is the ratio of net nitrification to net N mineralization. In TNEF mineral soils, this ratio was 83% and 96% in 2000 and 2002, respectively. These are much greater ratios than typically reported for undisturbed temperate forests. For example, Aber et al. (2003) reported that only two out of approximately 100 temperate forests had percent net nitrification rates greater than 80%. Thus, there appears to be significant net nitrification in TNEF soils, producing NO_3^-, which is lost to soil solution and stream water.

Ratios of carbon to nitrogen in soils

The C:N ratios of the forest floor (17–24) and mineral soil horizons (14) were at the low end of the range of C:N ratios reported for mineral soils (10–39) and forest floors (15–48) in other temperate forests in the northeastern U.S. (Table 5, Aber et al. 2003; Lovett et al. 2002). They were, however, similar to C:N ratios of mineral soils (13–19) reported for temperate forests in the mid-Atlantic region of the U.S. (Williard et al. 1997). The low C:N ratios in TNEF and other mid-Atlantic forests suggest that these forests are enriched in N. This N enrichment could be related to the chronically elevated rates of atmospheric N deposition. Upper elevation forests in the mid-Atlantic region receive some of the highest wet deposition of N in the U.S. (Castro and Morgan 2000).

Soil solution

We observed consistent patterns in the volume-weighted annual average NH_4^+ and NO_3^- concentrations in soil solution in all years (Fig. 3). First, NH_4^+ concentrations were always significantly lower than the NO_3^- concentrations. This pattern is consistent with our stream water measurements. Annual averaged NH_4^+ concentrations in soil solution ranged from 0.1 to 2.1 mg NH_4^+ l^{-1} for both depths (15 cm and 60 cm) in all years. There were no statistically significant differences in NH_4^+ concentrations between depths and years. Over all depths and all 4 years, average annual NO_3^- concentrations ranged from 3.9 to 13.4 mg NO_3^- l^{-1}. There were no statistically significant differences with depth or years except in 2004. In 2004, the annual average NO_3^- concentrations at both depths were about 2 to 3 times

Table 5 Carbon to nitrogen mass ratios in aboveground tissues, forest floor, mineral soil and fine roots

Tissue	Year sampled	Average C:N ratio	Number of samples
Green foliage	2001	15	54 composite samples
	2003	18	54 composite samples
Branches	2001	121	18 composite samples
	2003	99	18 composite samples
Boles	2001	198	18 composite samples
	2003	162	18 composite samples
Forest floor litter/Oi	1999	24	45 composite samples
Oe/humus	1999	17	45 composite samples
Mineral soil upper 10 cm	1999	14	45 composite samples
Fine roots	1999	21	15 Oe composite samples
			15 mineral soil composite

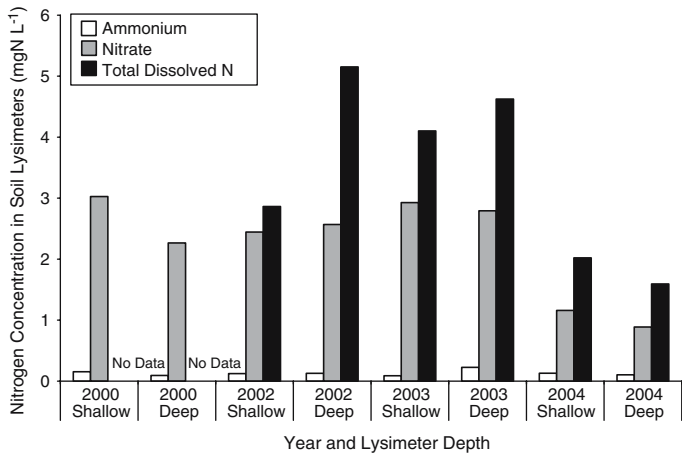

Fig. 3 Annual volume-weighted concentrations of dissolved inorganic N and total dissolved N (TDN) in soil solution collected at 15 and 60 cm depths

lower than in previous years. This may be due to elevated NO_3^- export from the soil solution to stream waters. Average dissolved organic N concentrations ranged from 1.6 to 5.2 mg N l^{-1}. The higher dissolved organic N concentrations, around 4 to 5 mg N l^{-1}, were in soil solutions collected at 60 cm. Nitrate concentrations in soil solution from TNEF were elevated compared to unpolluted temperate forests, but similar to those reported for temperate forests subjected to elevated rates of atmospheric N deposition. For example, soil solution N concentrations in an unpolluted old growth temperate forests were less than 0.02 mg NO_3^- l^{-1} (Hedin et al. 1995). High NO_3^- concentrations were found in soil solutions from Watershed 4 in Fernow Experimental Forest, where NO_3^- concentrations ranged from 2.5 to 12.9 mg NO_3^- l^{-1} in the A horizon and up to 19 mg NO_3^- l^{-1} in the B horizon (Peterjohn et al. 1996; Gilliam et al. 2001; Edwards et al. 2002). Watershed 4 has wet deposition rates of N similar to or less than TNEF (Castro and Morgan 2000).

Summary and conclusions

The overall objective of this study was to evaluate the N biogeochemistry of a structurally and developmentally young (18–22 years old) aggrading forest in western Maryland. We hypothesized that TNEF should N not display symptoms of N saturation, but should instead be N limited. Results from our extensive measurement program, however, are not consistent with our original hypothesis. TNEF clearly exhibited several symptoms of N saturation. First, the vegetation and soils in TNEF were highly enriched in N. Foliar and wood N concentrations were among the highest reported in the literature. The forest floor and mineral soils had some of the lowest C:N ratios reported for temperate deciduous forests (Williard et al. 1997; Aber et al. 2003). These elevated N concentrations are consistent with results from other forest studies in the mid-Atlantic region (Willaird et al. 1997; Peterjohn et al. 1999; Townsend et al. 2003). Collectively, these studies suggest that young aggrading and more mature forests in the mid-Atlantic region are enriched in N and are experiencing several symptoms of N saturation. We speculate that the N enrichment is related to the chronically elevated atmospheric N deposition in the mid-Atlantic region.

Our study also demonstrated the impact of annual precipitation variations on N runoff from a young aggrading N saturated forest. For example, TNEF was a net source of N, exporting 15 kg N ha^{-1} $year^{-1}$ and 18.4 kg N ha^{-1} $year^{-1}$ in the relatively wet water years of 2003 and 2004. These annual export rates were 2 to 3 times greater than the annual wet deposition rates of N. These wet year N export rates were higher than those reported for relatively undisturbed mature temperate forests in the U.S. (Campbell et al. 2004; Binkley et al. 2004). The high export rates from TNEF are likely due to a combination of chronically elevated N deposition coupled with highly variable annual precipitation amounts, ranging from dry to very wet years. We speculate that the dry years created elevated NO_3^- pools in the soils, which

were flushed into stream waters during the wetter conditions. Our results imply that we need to better understand how future climatic variations will affect N export from N saturated terrestrial ecosystems.

Acknowledgement We gratefully acknowledge support from the Andrew W. Mellon Foundation. We acknowledge the help of several students, particularly, Tim Negley, Philip Davis, Matthew Carter and Jack Wallace. We also greatly appreciate the help of the local land owners and the use of their private land. This paper is contribution 4050 of the Appalachian Laboratory of the University of Maryland Center for Environmental Science.

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