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Temporal patterns of nitrogen leakage from mid-Appalachian forested watersheds: Role of insect defoliation

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Abstract. Fluxes of dissolved nitrogen (N) as nitrate from forested watersheds in the mid-Appalachian region have important water quality ramifications for small acid-sensitive streams and for downstream receiving waters such as the Chesapeake Bay. Previous studies of N leakage have suggested that annual dissolved N fluxes from small watersheds can vary by several orders of magnitude and may be increasing as second-growth forests gradually become N saturated from the accrual of atmospheric N loadings. In this study, we examined the temporal (intra-annual and interannual) variability in dissolved nitrate fluxes from five small (area < 15 km^2) forested watersheds in the mid-Appalachian region from 1988 to 1995. At all sites, nitrate concentrations were observed to increase dramatically during storm flow events, with nitric acid contributing significantly to depressions in *p*H and acid-neutralizing capacity; annual nitrate fluxes were dominated by high-discharge periods. Interannually, the fluxes at each site varied by 1–2 orders of magnitude, but the patterns of N leakage displayed considerable synchrony with outbreaks of gypsy moth caterpillar defoliation that began in the late 1980s and early 1990s in this region. N leakage from forested watersheds apparently lagged the initial defoliation by several months to perhaps a year or more. Defoliation outbreaks by the gypsy moth caterpillar (or other herbivorous pests) thus provide an alternative explanation of N leakage from forest ecosystems. Poorly documented insect defoliations, rather than premature N saturation of intact forest ecosystems, need to be considered as a possible explanation of N leakage from forested watersheds in the mid-Appalachian region and elsewhere.

1. Introduction

Fluxes of dissolved nitrogen (N), primarily in the form of nitrate, from forested watersheds can contribute both to the acidification of acid-sensitive surface waters [e.g., *Schaefer et al.*, 1990] and to the eutrophication of downstream receiving waters, particularly coastal and estuarine ecosystems [e.g., *Fisher et al.*, 1992]. The scientific literature is replete with studies of dissolved N leakage from forested watersheds in the northeastern United States, Canada, and western Europe [*Hemond and Eshleman*, 1984; *Henriksen and Brakke*, 1988; *Hauhs et al.*, 1989; *Driscoll et al.*, 1989; *Stoddard and Murdoch*, 1991; *Murdoch and Stoddard*, 1992; *Likens and Bormann*, 1995]. Several of these studies have interpreted long-term increases or short-term patterns in dissolved N concentrations in surface waters as evidence of a situation known as "nitrogen saturation" in which the supply of N from the atmosphere to a forest exceeds the demand for N by watershed floral and microbial organisms [*Aber et al.*, 1989; *Driscoll et al.*, 1989; *Stoddard and Murdoch*, 1991; *Stoddard*, 1994]. Since N is often a limiting macronutrient in temperate forests, most of these ecosystems are usually considered to be strongly retentive of atmospheric N inputs, at least during the normal aggradation phase following major disturbance [*Bormann and Likens*, 1979].

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Using long-term data from the Hubbard Brook Experimental Forest (HBEF) in the White Mountains of New Hampshire, *Bormann and Likens* [1979] noted that in normally aggrading northern hardwood forests, N cycling is particularly tight with only ~5% of the total mineralized N being leaked to surface waters on an annual basis. The HBEF studies of N biogeochemistry also suggested that (1) there is a reproducible seasonal pattern of nitrate concentrations in effluent stream water, characterized by lowest concentrations during the growing season and highest concentrations during the dormant season and (2) the rate of accumulation of N in living and dead biomass (excluding the mineral soil) decreases as a function of ecosystem age. Experimental deforestations at the HBEF and elsewhere have clearly demonstrated the effects of vegetative disturbances on the transport and transformation of N from forested watersheds; N export rates and stream water dissolved N concentrations both increased by several orders of magnitude for a period of several years following commercial and experimental clear-cuts but returned to antecedent values within a few years after biotic regulation had been restored [*Bormann and Likens*, 1979]. Finally, *Mitchell et al.* [1996] recently attributed an observed temporal synchrony in nitrate concentrations in four forested watersheds in the northeastern United States to a climatic perturbation, particularly the occurrence of an anomalous cold period during the winter of 1990. *Mitchell et al.* further speculate that climatic variations attributable to global change thus need to be considered in

understanding the transport and transformation of nutrients in forest ecosystems, which strongly supports the need for long-term studies that include comparisons of watersheds within a region.

Studies of N fluxes from forested watersheds in the Appalachian Highlands physiographic region are far less common despite the documented acid sensitivity of surface waters in this region [*Kaufmann et al.*, 1991; *Herlihy et al.*, 1991] and despite the presence of several large estuaries of regional and national importance. *DeWalle and Pionke* [1994] reviewed the literature on N export rates from forested lands in the Chesapeake Bay region and documented more than 2 orders of magnitude variation in mean annual nitrate export (0.04–5.2 kg N ha^{-1} yr^{-1}) among 25 small forested watersheds in the region while showing that maximum annual N export differed from the minimum value by less than a factor of 2–3 at most of the sites; unfortunately, few of the studies cited by *DeWalle and Pionke* [1994] had more than a 5-year record of N export. A watershed manipulation experiment in which granular fertilizer was applied at the Fernow Experimental Forest (FEF) in West Virginia does not unequivocally support the N saturation hypothesis; a significant increase in dissolved N export was observed in only one of two treated watersheds following 3 years of treatment [*Adams et al.*, 1993]. However, more recent studies at FEF [*Gilliam et al.*, 1996; *Peterjohn et al.*, 1996] are more consistent with the N saturation hypothesis.

There is also a practical reason for understanding N transport and transformation within forested lands of the Chesapeake Bay watershed. A plan to reverse a long history of nutrient overenrichment from point and nonpoint source loadings of N and phosphorous (P) is currently being implemented under the Chesapeake Bay Agreement (CBA) of 1987 with the goal of a 40% reduction in the flux of these nutrients to the Chesapeake Bay by the year 2000 [*Chesapeake Executive Council*, 1989]. Because ~60% of the Chesapeake Bay watershed is forested, the regional-scale flux of N from forested lands to the Bay must obviously be taken into account [*Gardner et al.*, 1996]. *Linker et al.* [1996] recently used a watershed simulation model known as Hydrologic Simulation Program-Fortran (HSPF) [*Bicknell et al.*, 1993] to guide nutrient reduction strategies in the Chesapeake Bay watershed; they assumed that the annual N export from forested lands is 3.4 kg ha^{-1}, the lowest export rate of all land uses examined. Despite this relatively low N export rate, the HSPF model predicted that the N flux from forested lands is the largest single nonpoint source of N to the Chesapeake Bay (L. C. Linker, U.S. Environmental Protection Agency, personal communication, 1996), which begs the question of how N fluxes from mid-Appalachian forests might change in the future. Although N fluxes from forests are generally considered uncontrollable sources and thus are not subject to the reductions targeted under the CBA, reductions in atmospheric deposition of N to forests brought about by the Clean Air Act Amendments of 1990, increases in N deposition attributable to population increases in the Chesapeake Bay watershed [*Chesapeake Executive Council*, 1988; *Galloway et al.*, 1994], or the occurrence of regional-scale N saturation could dramatically affect the export of N from forested lands to the Chesapeake Bay. This adds substantial uncertainty to the legitimacy and efficacy of the targeted reductions in controllable N sources.

Webb et al. [1995] and *Eshleman et al.* [1995] recently documented stream water quality impacts associated with forest defoliation by the gypsy moth caterpillar (*Lymantria dispar* L.) in the early 1990s at White Oak Run in Shenandoah National Park, Virginia. Using 13 years of discharge and stream water chemistry data, *Webb et al.* [1995] showed that stream water changes following defoliation included increasing concentrations of nitrate, strong base cations, and hydrogen ion, with decreasing concentrations of acid-neutralizing capacity (ANC) and sulfate, while *Eshleman et al.* [1995] showed that the intensity of episodic acidification increased at White Oak Run following the initial defoliation, primarily owing to the transient leakage of nitric acid from the watershed during storm flow events. Both studies thus confirmed that nonharvesting forest disturbances can significantly affect rates of loss of solutes to stream water, which is consistent with stream responses following both experimental and natural forest disturbances at the Coweeta Hydrologic Laboratory in North Carolina reported earlier [*Swank*, 1988].

The purposes of this paper are to (1) examine the intra-annual variations in dissolved N concentrations, especially changes in nitrate during storm flow conditions, in streams draining five forested watersheds within the Chesapeake Bay basin of the Appalachian Highlands region, (2) examine the interannual temporal variations in the fluxes of dissolved N from these watersheds, and (3) interpret and discuss the regional-scale synchrony of a recent increase in nitrate concentrations and fluxes in these watersheds in the context of the nitrogen saturation hypothesis and with respect to the issue of the Chesapeake Bay N reduction strategy. In addition, temporal variations in N fluxes from a control watershed at the HBEF [*Likens and Bormann*, 1995] will be shown to be consistent with the interpretation that herbivorous insect defoliations can cause dramatic leakages of N for many years following the initial infestation.

2. Study Sites

The subjects of this paper are five small gaged forested watersheds located in the Chesapeake Bay basin of the Appalachian Highlands physiographic region (Figure 1). This mountain-dominated region of the eastern United States has often been subdivided further into three physiographic subregions or provinces: the Blue Ridge province, the Ridge and Valley province, and the Appalachian Plateaus province [*Fenneman*, 1938]. Four of the five watersheds are located in Shenandoah National Park (SNP), Virginia, wholly within the Blue Ridge province: White Oak Run (WOR), Paine Run (PAIN), Piney River (PINE), and Staunton River (STAN). The geology of SNP is dominated by three general rock types: granites and granodiorites, metamorphosed basalts, and metamorphosed clastic sedimentary rocks [*Gathright*, 1976]. These general groupings have been shown to explain much of the spatial variation in the concentrations of dissolved constituents in SNP streams [*Lynch and Dise*, 1985]. Average annual precipitation in SNP is evenly distributed throughout the year and ranges from ~950 mm yr^{-1} at lower elevations to over 1300 mm yr^{-1} on the highest peaks with annual surface runoff averaging ~525 mm yr^{-1} [*Lynch*, 1987]. The fifth watershed studied is located in Savage River State Forest in western Maryland (Upper Big Run (BIGR)) within the Allegheny Mountain section of the Appalachian Plateaus province. The geology of this section is dominated by gently folded sedimentary rocks of Devonian through Mississippian age including limestones, sandstones, and shales. Average annual precipitation (1150 mm yr^{-1}) and surface runoff (525 mm yr^{-1}) are very similar to

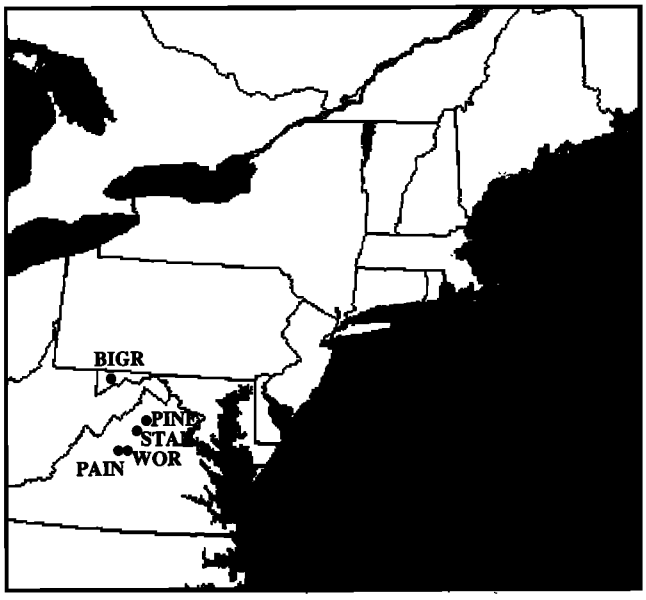

Figure 1. Locations of five small forested watersheds in the mid-Appalachian region of the eastern United States. BIGR, Upper Big Run; PINE, Piney River; STAN, Staunton River; WOR, White Oak Run; PAIN, Paine Run.

those measured in SNP, with snowfall accounting for a somewhat larger percentage (~20%) of mean annual precipitation [*Staubitz and Sobashinski*, 1983]. While similar in size, the five watersheds and their effluent streams encompass a range of geological substrates and acid sensitivities (Table 1); it was for this range of characteristics that the watersheds were initially selected for intensive study. Forest vegetation also varies among the five watersheds, particularly with respect to domination by oak (*Quercus*) species; oak composition is highest in PAIN (~100%), high in WOR (~80%), and moderate in STAN, PINE, and BIGR (~60%).

3. Methods

The chemical composition of stream water at the five sites was monitored using a combination of quarterly grab sampling, weekly grab sampling, and automated sampling (2- to 8-hour frequency) during selected storm flow events, followed by a series of laboratory analyses appropriate for acidification studies [*U.S. Environmental Protection Agency*, 1987]. ANC was measured by an acidimetric Gran titration. Conductivity was measured using a conductivity dip cell and meter; *p*H was measured using an Orion Ross meter and combination *p*H-reference electrode. Major anions (Cl^-, SO_4^{2-}, and NO_3^-) were measured by ion chromatography (IC) using suppressed conductivity detection, while major cations (Ca^{2+}, Mg^{2+}, K^+, and Na^+) were measured by flame atomic absorption/emission spectroscopy or by IC. Exchangeable and nonexchangeable fractions of reactive aluminum were measured using the pyrocatechol violet flow injection analysis. All chemical data were subjected to intensive quality assurance and quality control checks to confirm the internal consistency of the individual measurements [*U.S. Environmental Protection Agency*, 1987].

Presently, all five watersheds are being gaged using standard methods of the U.S. Geological Survey [*Rantz et al.*, 1982], and chemical concentrations in stream water are being measured at least biweekly; analyses of stream water samples collected automatically during selected storm flow events are used to supplement the periodic sampling data. However, the period of record associated with the measurements of stream water chemical composition and stream discharge varied among the five watersheds. The data record is longest for WOR, which has a continuous record of mean daily discharge and weekly measurements of stream water composition dating to 1979. Data for PAIN, PINE, and STAN are shorter, with quarterly chemical composition dating to 1987; however, weekly and episodic composition data and continuous discharge data were collected at these three watersheds from 1992 to present (except at STAN, where a flood in June 1995 washed out the stream gage). Weekly and episodic stream water composition data were collected at BIGR from 1989 to 1992, followed by weekly or monthly sampling from 1992 to 1995; a continuously recording stream gage was installed in 1995.

Nitrate fluxes in stream water for the five watersheds were computed as the product of discharge (either mean daily or mean hourly) and instantaneous nitrate concentration. A linear interpolation scheme was used to estimate nitrate concentrations for each sampling interval for which an actual nitrate measurement was not available. Annual nitrate fluxes were computed on a water year basis by aggregation of the daily (or hourly) fluxes. Since discharge data were not available for four of the five sites for the entire period of chemical sampling, data from other gaged watersheds nearby were used to estimate discharge during periods with missing records. Data for WOR were used to estimate flows at PAIN by a simple linear model:

$$Q_{\mathrm{PAIN}} = 0.85 Q_{\mathrm{WOR}} \tag{1}$$

where Q_{PAIN} and Q_{WOR} are both expressed in units of millimeters per time after normalizing discharge values using watershed area. The same method was employed to estimate discharges at PINE and STAN using records reported for a gage on the Rapidan River (RAPR) near Ruckersville, Virginia [*U.S. Geological Survey*, 1997a]; a coefficient of 1.20 was used to adjust the RAPR records. Data from a gage on the

Table 1. Locations and Characteristics of Five Forested Mid-Appalachian Watersheds

Watershed	UTM-E	UTM-N	Area, km^2	Surficial Geology	Baseflow ANC, $\mu eq\ L^{-1}$
Paine Run, Virginia	693,243	4,229,927	12.4	phyllite, quartzite, and metasandstone	0–10
Piney River, Virginia	737,939	4,286,960	12.6	metabasalt and granodiorite	150–350
Staunton River, Virginia	729,381	4,258,242	10.5	granodiorite	75–125
White Oak Run, Virginia	696,960	4,235,772	5.1	phyllite, quartzite, and metasandstone	25–50
Upper Big Run, Maryland	656,509	4,384,218	1.6	sandstone, siltstone, and shale	25–50

UTM-E, universal transverse mercator-easting of stream gage; UTM-N, universal transverse mercator-northing of stream gage; ANC, acid-neutralizing capacity.

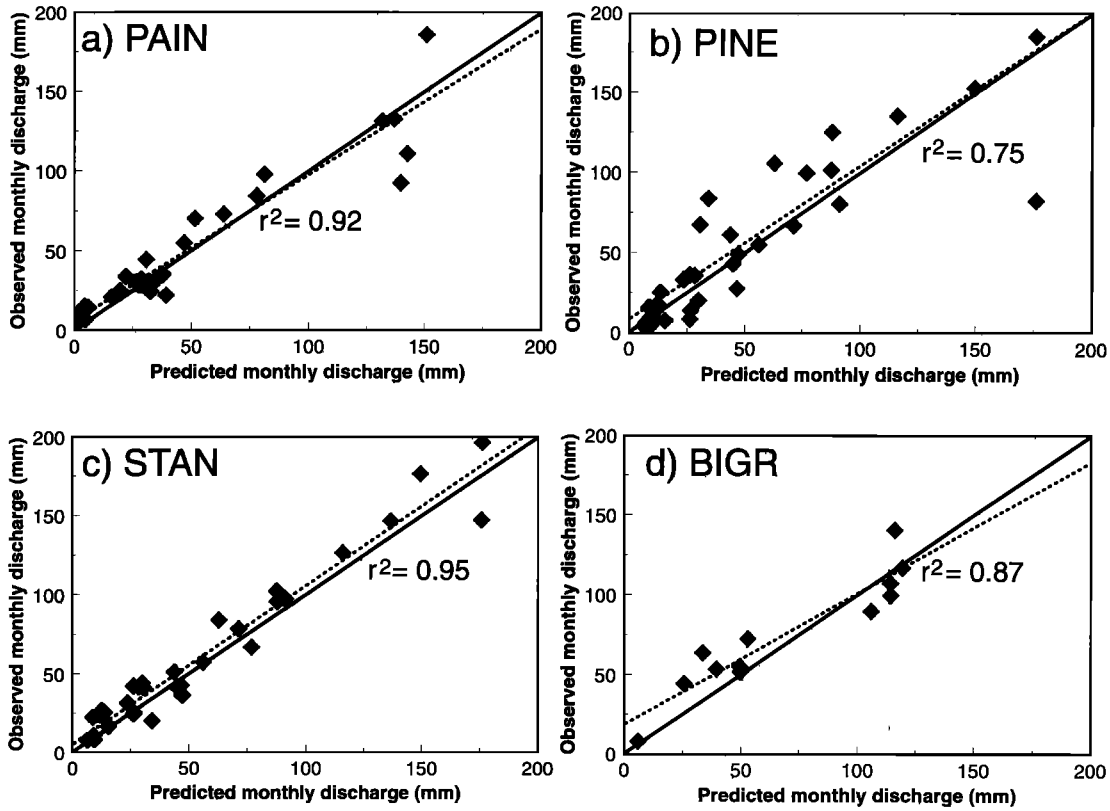

Figure 2. Comparisons of predicted (using a linear flow model) and measured monthly discharge for four streams including (a) PAIN, (b) PINE, (c) STAN, and (d) BIGR. Regression lines (dashed), coefficients of determination (r^2), and 1:1 lines (solid) are also shown.

Savage River (SAVR) near Barton, Maryland [*U.S. Geological Survey*, 1997b] were used to estimate discharges at BIGR using a coefficient of 0.85. Linear regressions of aggregated monthly discharge values (predicted versus observed) indicated that the models provided unbiased estimates of discharge for the four sites (Figure 2), since *t* tests showed that none of the intercepts of the regression lines were significantly different from zero and that none of the slopes of the regression lines were significantly different from unity ($\alpha = 0.05$).

Data from PAIN and PINE (1992–1995) were used to examine the sensitivity of the nitrate flux computations to the frequency of chemical sampling, since these two streams have the most complete records (including episodic sampling) during this period. Annual fluxes were computed on a water year basis after subsetting the chemical databases into (1) quarterly, (2) monthly, and (3) weekly observations; computed fluxes were then compared against fluxes based on the complete stream water composition record. Fluxes for each sampling frequency were recomputed using modeled discharge data to examine the magnitude of errors associated with the absence of on-site discharge monitoring. The results of the sensitivity analysis suggest that computed nitrate fluxes vary as a function of data frequency, with the lowest frequency data (quarterly) generally providing the lowest estimates; annual flux differences based on the use of quarterly, monthly, and weekly data (in combination with measured discharge) fell in the ranges of −40 to −10%, −28 to −2%, and −13 to +2%, respectively (Figures 3a and 3c). Annual flux differences based on the use of quarterly, monthly, and weekly data (in combination with modeled discharge) fell in the ranges of −43 to +9%, −36 to +5%, and −25 to +28%, respectively (Figures 3b and 3d). The results suggest that computational errors in computing annual nitrate flux in this study are likely no greater than ±50% in any 1 year, with a strong probability that the computed values are underestimates. *Swistock et al.* [1997] reported similar errors (−31 to +42%) associated with the computation of annual nitrate flux for other small forested watersheds in the Appalachian region using comparable techniques. In addition, our results indicate that the use of modeled discharge does not add appreciable additional error when quarterly or monthly chemical data are used, but it does add significant errors when weekly (and probably higher frequency) data are employed.

Maps (1:24,000 scale) of forest defoliation by the gypsy moth caterpillar, based on annual (1986–1993) aerial surveys conducted in the areas surrounding the five watersheds, were obtained from Shenandoah National Park and from Maryland Department of Agriculture. These defoliation maps had been produced from sketch maps based on statewide or park-wide aerial infrared photographs from low-level reconnaisance flights during late July of each year when defoliation was at a peak. Digitized areas within each watershed associated with heavy or moderate defoliation (>30% leaf area consumed) were computed using geographic information system (GIS) software.

4. Results

4.1 Intra-annual Variability in Nitrate Concentrations and Fluxes

Concentrations of nitrate in stream water at each of the five sites during the 1993 and 1994 water years displayed significant

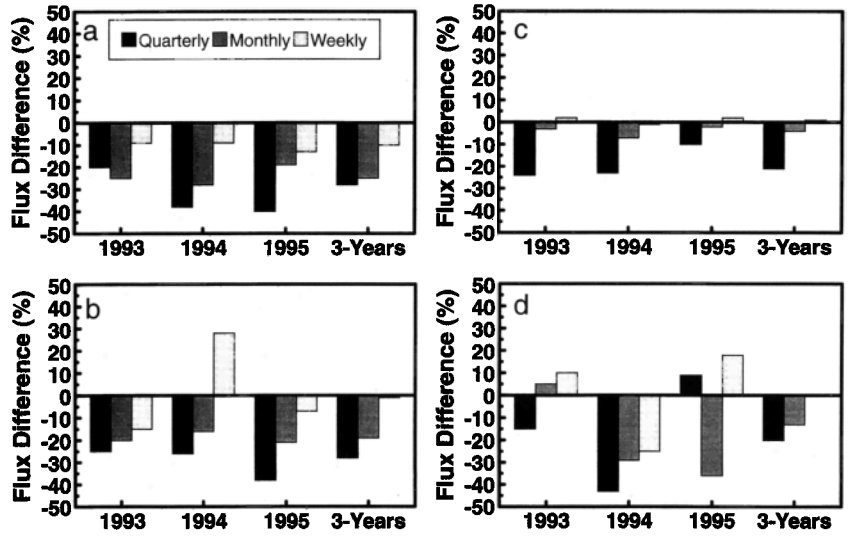

Figure 3. Computed differences in annual nitrate flux (%) based on the use of quarterly, monthly, and weekly nitrate data and either continuous measured discharge or continuous modeled discharge for two streams. Measured and modeled discharges are given for (a and b) PAIN and for (c and d) PINE. Differences are computed with respect to the annual nitrate flux computed using measured discharge and the complete stream water nitrate record.

intra-annual variability. Concentrations at all sites were lowest (near detection limit of 1–2 μeq L^{-1}) at the end of the summer of 1993, and they were highest (40–100 μeq L^{-1}) near the peak of major storm flow events. At those sites with high-intensity data associated with storm flow events during the 1993–1994 water years (PAIN, PINE, and STAN), the graphs of nitrate concentration versus time actually appear to resemble discharge hydrographs with both rising and recession limbs. Nitrate concentrations were particularly high during fall rainstorms in 1993, following the moderately dry summer of that year (Figure 4).

Variability in nitrate concentrations at the sites is associated with a variety of other changes in acid-base status. As an example, data for PAIN suggest that nitric acid is a contributor (along with sulfuric acid) to episodic acidification (defined as the loss of ANC during storm flow events) when not fully balanced by increases in concentrations of strong base cations during these periods (Figure 5). During several rainstorms in the fall of 1992, ANC in PAIN actually dropped to zero or below zero several times, with one rainstorm in late November causing a 0.5 unit depression in *p*H and an increase of nearly 100 μg L^{-1} in exchangeable reactive Al (Figure 6). ANC and *p*H remained depressed and Al concentrations remained elevated for several days following the flood peak (Figure 6).

Dissolved nitrate fluxes from the five watersheds were dominated to varying degrees by the flux during storm flow periods. This situation is illustrated using data for PAIN and PINE for the 1993–1995 water years. At both sites, more than 50% of the nitrate flux occurred in less than 10% of the time, primarily during major storm flow periods when both discharge and nitrate concentration were elevated. The situation was even more pronounced in PAIN than in PINE, presumably owing to the flashier hydrological response of the former watershed (Figure 7).

4.2 Interannual Variability in Nitrate Concentrations and Fluxes

Interannual variability in nitrate fluxes and flow-weighted concentrations in the five watersheds was very large during the 1988–1995 water years and displayed considerable synchrony (Figures 8a and 8b). Annual nitrate fluxes from the four sites for which we have data were extremely low during the 1988 and 1989 water years but increased dramatically (by 1–2 orders of magnitude) at PINE and STAN in 1990 and at PAIN and WOR in 1991; the annual nitrate flux was relatively high at BIGR in 1990 also, but the absence of data during the 1988–1989 water years makes it impossible to determine whether the flux increased. At all sites, nitrate fluxes peaked during the period 1991–1993 and remained dramatically elevated above 1988–1989 baseline levels through the 1995 water year, although they began to steadily decline in 1993 despite highly variable runoff conditions during this period (Figures 8a and 8c). The temporal patterns of discharge-weighted annual nitrate concentrations in the five streams were strikingly similar to the corresponding nitrate flux patterns (Figures 8a and 8b).

Long-term nitrate flux data for WOR (1980–1995) suggest that the dramatic increase and subsequent decrease in nitrate flux in the early 1990s was an isolated occurrence. Annual nitrate fluxes from WOR were all <30 eq ha^{-1} yr^{-1} during the 11-year period from 1980 to 1990, averaging <7 eq ha^{-1} yr^{-1}; from 1991 to 1995, however, the average annual nitrate flux from WOR averaged nearly 220 eq ha^{-1} yr^{-1}, an increase of more than 3000% (Figure 9). While greater runoff contributes to greater nitrate export, all else being equal, it should be emphasized that the increased nitrate flux at WOR during the period 1991–1995 cannot be explained on the basis of increased runoff alone, despite the observations that (1) the extremely low annual nitrate export values measured in 1981, 1985, and 1988 were all associated with below-average runoff and (2) annual runoff during water years 1991–1993 was well above average. Water years 1983 and 1984 were also high runoff years, yet the increases in nitrate export in these years were quite modest compared with values measured during the mid-1990s (Figure 9).

The increase in nitrate fluxes from the five watersheds can also not be attributed solely to computational errors associated with different sampling frequencies or runoff estimation meth-

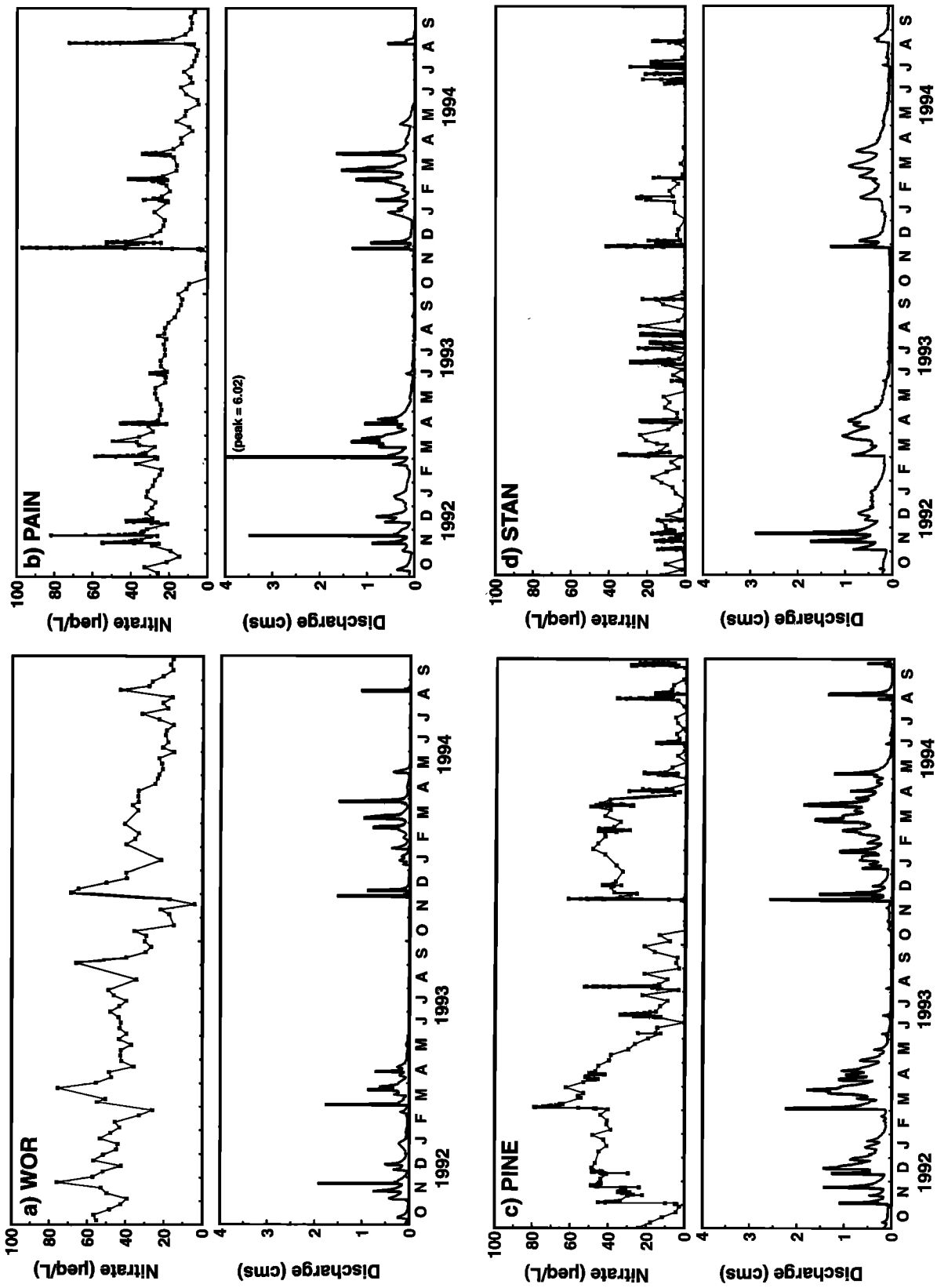

Figure 4. Nitrate concentrations and mean daily discharge versus time for five forested watersheds including (a) WOR, (b) PAIN, (c) PINE, (d) STAN, and (e) BIGR, for the 1993 and 1994 water years.

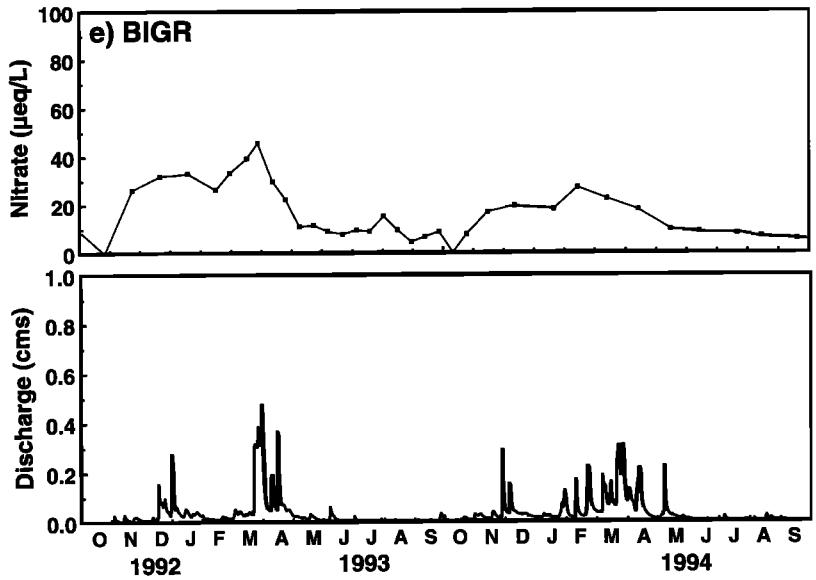

Figure 4. (continued)

ods. Allowing for computational errors of the magnitude ±50% based on the sensitivity analysis for PAIN and PINE, these errors could only account for interannual flux differences of about a factor of 2, not 1–2 orders of magnitude. Clearly, the observed nitrate pulse cannot be explained by computational errors alone, although some of the interannual variations in nitrate flux, particularly for watersheds such as BIGR, which required extensive estimation of discharge, can certainly be attributed to this factor.

The dramatic increase in annual nitrate export from the five

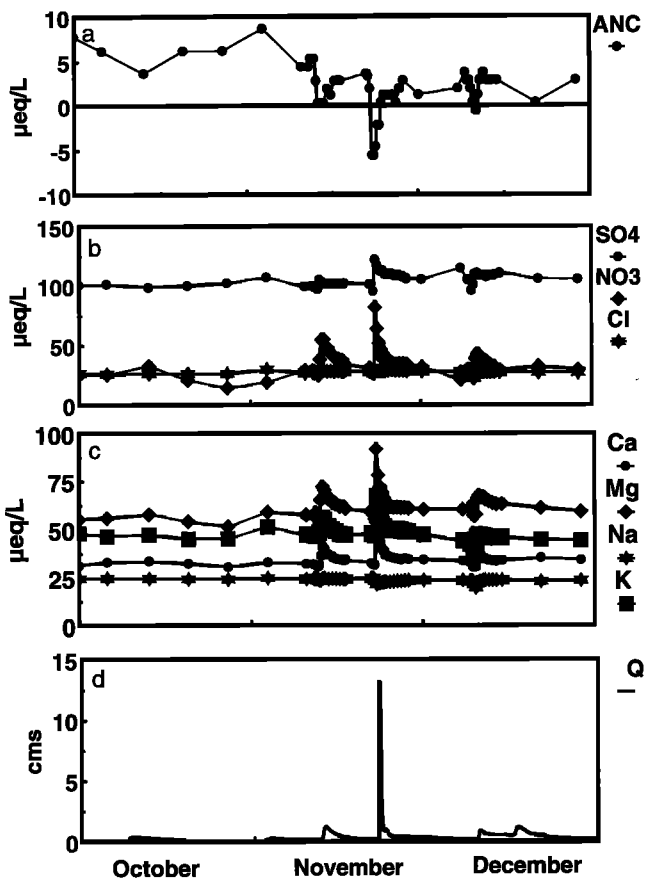

Figure 5. Temporal changes in (a) acid-neutralizing capacity (ANC), (b) major anion concentrations, (c) major cation concentrations, and (d) discharge in PAIN during autumn 1992.

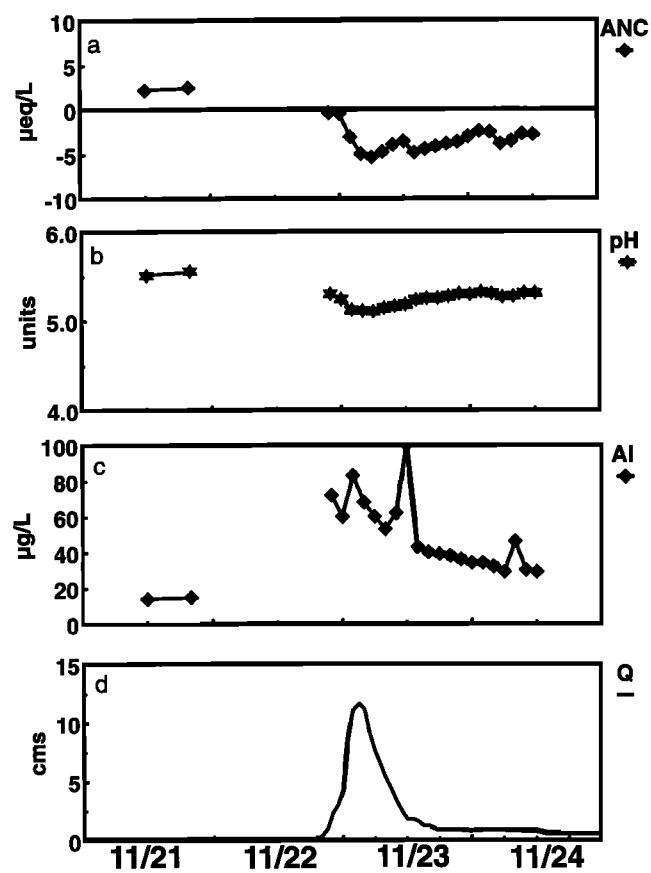

Figure 6. Temporal changes in (a) ANC, (b) *p*H, (c) exchangeable reactive Al concentrations, and (d) discharge in PAIN during the period November 21–24, 1992.

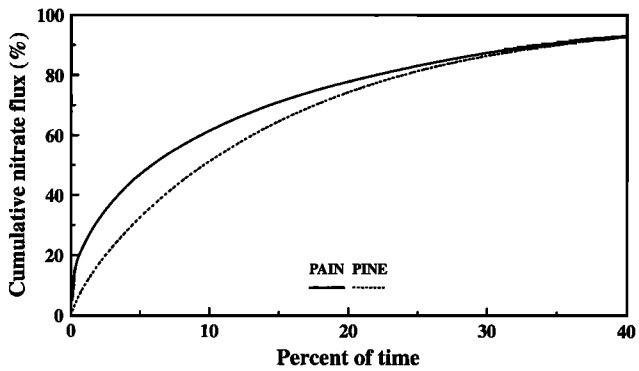

Figure 7. Cumulative frequency distributions of stream water nitrate flux for PAIN and PINE for water years 1993–1995.

watersheds beginning in the early 1990s can be interpreted as a biogeochemical response to gypsy moth caterpillar defoliation, which slightly preceded the stream water nitrate responses in time. There is no evidence of any gypsy moth "outbreaks" (large-scale defoliations) occurring either in western Maryland or in SNP prior to 1986 (D. Cohen, Maryland Department of Agriculture, personal communication, 1997). Beginning in 1986 or 1987, however, each of these areas was defoliated several times by a southward moving gypsy moth wave; computed percentages of the areas of mapped defoliation within each watershed indicate that the two northernmost watersheds (BIGR and PINE) experienced their first defoliation in 1987, that STAN was first defoliated in 1989, and that the two southernmost watersheds (PAIN and WOR) were not first defoliated until 1990 (Table 2). The timing of nitrate export response is generally consistent with this defoliation pattern, with significant export evident for BIGR, PINE, and STAN in 1990 and for WOR and PAIN beginning in 1991 (Figure 8a).

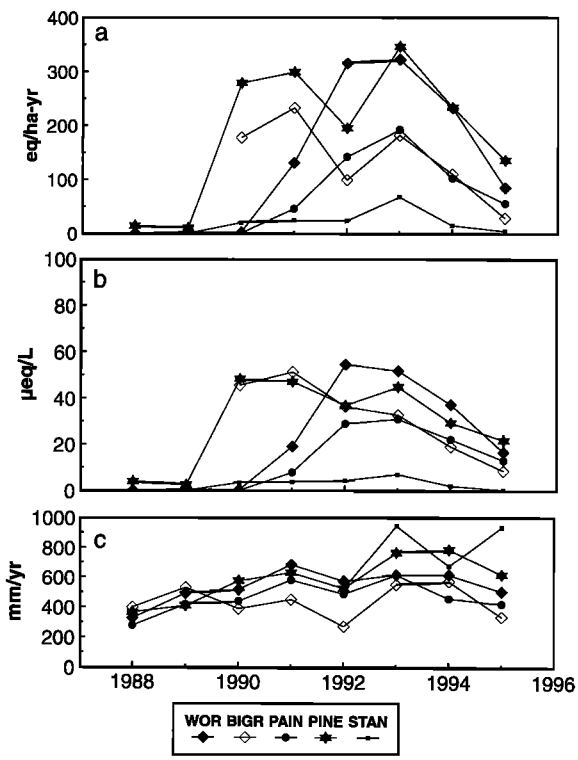

Figure 8. Computed (a) annual nitrate fluxes, (b) annual discharge-weighted nitrate concentrations, and (c) annual runoff values for the five study watersheds for water years 1988–1995 (note that no nitrate flux or concentration data for BIGR were available for years 1988–1989).

5. Discussion

The dramatic leakage of dissolved N as nitrate from small forested watersheds in the mid-Appalachian region during the early 1990s to mid-1990s occurred contemporaneously with a period during which defoliation of these watersheds by the gypsy moth larva was taking place. While our evidence that an outbreak of gypsy moth caterpillar defoliation was the cause of this biogeochemical response is largely circumstantial, we believe that this conclusion is reasonable for several reasons. First, long-term data from WOR suggest no appreciable N leakage during the 11-year period prior to 1991 (Figure 9); while isolated infestations of the gypsy moth probably existed in SNP and in western Maryland before 1986, no known large-scale defoliations can be attributed to the pest prior to this time [*Doane and McManus*, 1981]. Second, all five watersheds are dominated to varying degrees by oak species (gypsy moth caterpillars have a strong preference for oak foliage) and were all partially defoliated by the southward moving gypsy moth wave during the period when N leakages from the watersheds occurred (Table 2); the N leakages occurred sequentially along a north-south gradient, which is consistent with the general direction of movement of the gypsy moth defoliation wave. Third, the synchrony of the observed biogeochemical responses among the five different watersheds suggests a regional-scale perturbation, rather than a local disturbance; once an infestation is present, gypsy moth outbreaks usually occur over very large areas, rather than as localized events, but are notoriously difficult to predict [*Liebhold and Elkinton*, 1989]. Despite the difficulty of predicting the timing of outbreaks, the interannual patterns in outbreaks have also been shown to be quite synchronic [*Liebhold and McManus*, 1991]. Fourth, the estimated time lags between the water year of initial defoliation and the water year in which N leakage was observed were consistent for three of the four watersheds for which this could

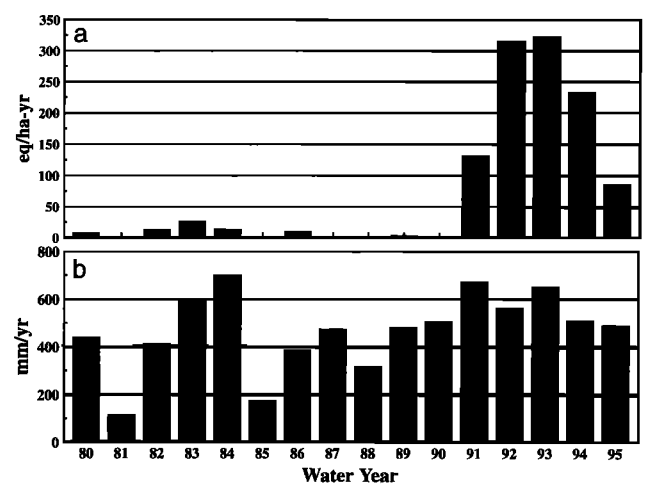

Figure 9. Computed (a) annual nitrate fluxes and (b) annual runoff values for WOR for the period 1980–1995.

Table 2. Timing and Magnitude of Gypsy Moth Defoliation in Five Forested Mid-Appalachian Watersheds During the Period 1987–1993

	Area of Mapped Defoliation, %						
Watershed	1987	1988	1989	1990	1991	1992	1993
Paine Run	0.0[a]	0.0[a]	0.0[a]	8.8	63.4	85.2	0.0
Piney River	3.8	32.3	34.0	15.2	0.0	14.2	0.0
Staunton River	0.0	0.0	31.4	59.3	0.0	0.0	0.0
White Oak Run	0.0[a]	0.0[a]	0.0[a]	46.5	92.9	90.4	0.0
Upper Big Run	61.6[b]	0.0	0.0	34.8	7.3	0.0	0.0

Areas mapped as "heavy defoliation" in the Virginia watersheds and as "heavy defoliation" or "moderate defoliation" in the Maryland watershed.

[a]Defoliation maps for the portion of Shenandoah National Park including these watersheds are not available for these years; only minor defoliation may have occurred in these watersheds in 1989.

[b]Mapped defoliation in 1987 was partly attributed to a cankerworm.

be estimated; for STAN, WOR, and PAIN the lag was ~1 year, while the lag for PINE was estimated as 3 years. The 1-year lag is somewhat deceptive, since peak defoliation usually occurs by midsummer (near the end of the water year), while N leakage was, in several cases, observed to begin during the first autumn following defoliation (near the beginning of the next water year); for example, *Webb et al.* [1995] presented data from WOR that indicated that nitrate losses can begin as early as September of the year of initial defoliation. In the case of PINE, the estimated 3-year lag may reflect a less severe defoliation in 1987 (Table 2) combined with the effect of subnormal water yields (60–70% of normal) in 1988 and 1989 on N transport from the watershed (Figure 8c). Since each of the watersheds was defoliated more than once during the study period, it is not possible to determine the duration of nitrate losses associated with a single defoliation event without making additional assumptions.

Many previous studies have documented the effects of watershed-scale disturbances on dissolved N loss, but few studies have shown synchronic timing of insect defoliations and biogeochemical responses. At the Coweeta Hydrological Laboratory in North Carolina, however, nitrate losses from two watersheds could be attributed to defoliation outbreaks of the fall cankerworm during the 1970s. The highest fluxes of nitrate apparently occurred in years with the greatest amounts of defoliation, and nitrate losses gradually declined once the cankerworm population declined [*Swank et al.*, 1981]. *Swank* [1988] reported a maximum annual loss of nitrate during defoliation at Coweeta of 450 g N ha^{-1} yr^{-1} (32 eq ha^{-1} yr^{-1}), compared with a range of 70–350 eq ha^{-1} yr^{-1} for the five watersheds in this study (Figure 8). Leakages of N due to deforestation are more commonly reported, however. For example, *Likens et al.* [1978] showed that deforestation of northern hardwood forests in New Hampshire by commercial and experimental clear-cutting resulted in a peak in nitrate losses in the second year following disturbance with annual fluxes in the range of 57–140 kg N ha^{-1} yr^{-1} (4100–10,000 eq ha^{-1} yr^{-1}). Therefore it appears that peak dissolved N losses due to deforestation are at least 1–2 orders of magnitude larger than comparable losses due to defoliation (when expressed on a watershed area basis) with a similar lag period.

While the timing of the biogeochemical response to defoliation appears quite similar to the response to deforestation, a mechanism for increased N losses following defoliation has not been experimentally confirmed. *Bormann and Likens* [1979] noted that increased N losses from defoliation could result from a variety of effects on the forest floor, including (1) increased discharge of water due to reductions in transpiration rates, (2) decreased uptake of nutrients by green plants, (3) higher soil temperatures due to increased radiant energy flux to the forest floor, and (4) alterations in microbiological activity in the soil leading to increased rates of nitrification. *Swank et al.* [1981] proposed that N losses will increase following forest disturbance if plant and microbial uptake are not sufficient to retain the mobilized N, while *Swank* [1988] proposed that decomposition, net primary production, and plant uptake of N are the most important factors controlling the magnitude and timing of N export from disturbed forest ecosystems. Recognizing the potential importance of the N content in insect biomass, frass (pellets of insect excrement), and green leaf debris, which is added to the forest floor during defoliation, several workers have proposed an increased N availability from mineralization of N from these sources [*Seastedt and Crossley*, 1984; *Schowalter et al.*, 1986; *Hollinger*, 1986]. However, a recent study by *Lovett and Ruesink* [1995] demonstrated rapid net immobilization, rather than the expected net mineralization, of N by gypsy moth frass in laboratory incubations. *Lovett and Tobiessen* [1993] showed that N deficient oak trees have a strong capacity for uptake of available ammonium even when they are defoliated. These two latter studies seem to provide a mechanism by which N losses from defoliated forests might be avoided altogether.

Alternately, increased nitrate export due to gypsy moth defoliation might also be expected to occur over a much longer term as a result of oak mortality, since repetitive defoliations increase the probability of tree mortality [*Liebhold et al.*, 1995]. Interestingly, oak mortality is typically caused by secondary agents such as the two-lined chestnut borer (*Agrilus bilineatus*) or the root rotting fungus (*Armillaria mellea*), not by the gypsy moth [*Doane and McManus*, 1981]. Defoliation represents the primary causal agent associated with oak mortality and subsequent shifts in species composition; major forest successional changes, perhaps in response to gypsy moth defoliation, have been reported in the literature [*Allen and Bowersox*, 1989; *Abrams and Downs*, 1990; *Widmann*, 1995], although it is not yet clear whether the regional impacts of gypsy moth defoliation are as dramatic as other perturbations such as the chestnut blight and Dutch elm disease [*Liebhold et al.*, 1995]. Monitoring of nitrate leakage for many years following defoliation is needed to accurately estimate the total duration of response that might result from oak mortality and subsequent forest succession.

The leakages of nitrate reported in this study largely occurred during storm flow conditions when both discharge of water and nitrate concentrations increased (Figures 4, 5, and 7). The increasing concentration of nitrate in stream water during storm flow periods has important implications for quantifying episodic acidification of acid-sensitive surface waters, for identifying the source of nitrate, and for determining the mechanism of N loss. Since nitrate is an extremely mobile anion, nitrate leakage has the potential to acidify surface waters (if it is associated at least in part with protons) or toxify surface waters (if it is associated with Al^{3+} or $Al_x(OH)_y^{+z}$ species). *Molot et al.* [1989] proposed a straightforward method of partitioning the episodic acidification of a surface water (measured as ΔANC) by the corresponding major ion changes measured concurrently. For the example presented in Figure 6 the measured ΔANC of -8 μeq L^{-1} in PAIN during the November 22–23, 1992, event can be partitioned into relative ion changes (Δ[X]/ΔANC) for nitrate, sulfate, and base cations. The results indicate that more than two thirds of the ΔANC can be attributed to the increase in nitric acid concentration, while one third can be attributed to an increase in sulfuric acid; since base cation concentrations increased during the event, none of the ΔANC can be attributed to base cation dilution. The importance of nitric acid in contributing to episodic acidification of PAIN, PINE, and STAN was effectively demonstrated by *Hyer et al.* [1995] through analysis of data from more than 25 distinct storm flow events in SNP during 1992–1994. Overall, an increase in nitric acid concentration was generally the largest relative contributor to ΔANC at PAIN, a secondary contributor at STAN, and a tertiary contributor at PINE [*Hyer et al.*, 1995]. Previous studies of episodic acidification in the Appalachian Highlands area have also demonstrated a variable contribution of nitric acid to ANC depression [*Wigington et al.*, 1996], with the magnitude of the hydrochemical response apparently varying as a complex function of antecedent ANC, storm type, rainfall intensity, and peak discharge [*Morgan et al.*, 1994; *Hyer et al.*, 1995; *Hyer*, 1997].

There is also evidence that the intensity of episodic acidification has increased since the onset of gypsy moth defoliation in SNP. Using 13 years of weekly stream chemistry data and continuous daily flow records, *Eshleman et al.* [1995] showed that the magnitude of ANC depressions at WOR has increased considerably since 1990, the first year of defoliation in this watershed; their analysis showed that the mean ΔANC associated with storm flow events has approximately doubled since 1990. In addition, virtually all of the ΔANC shift was explained by episodic increases in nitric acid in stream water [*Eshleman et al.*, 1995].

The fact that N leakage was shown in this study to occur primarily during storm flow conditions confounds a determination of the N source and provides an alternative to premature N saturation from atmospheric N inputs as a mechanism of N leakage from forested watersheds. Interestingly, this pattern of N loss, dominated by storm flow fluxes, is identical to the pattern of N leakage that has been proposed to describe premature N saturation from excessive inputs of atmospheric N [*Aber et al.*, 1989; *Stoddard*, 1994]. Therefore increased losses of dissolved N during storm flow periods cannot be used alone to unequivocally identify the occurrence of premature N saturation in watersheds receiving atmospheric N inputs. In fact, the interpretation that natural disturbances of forested watersheds by nonendemic insect pests can result in appreciable leakage of dissolved N to stream waters on a regional scale also has relevance in understanding the effects of N deposition or climatic changes on forest ecosystems and in managing watersheds to reduce N losses. Since gypsy moth outbreaks in the northeastern United States have been shown to recur at ~8- to 10-year intervals with outbreaks typically lasting 2–3 years [*Doane and McManus*, 1981], it is possible that N leakages due to undocumented insect defoliations could cause one to wrongly conclude that intact forest ecosystems are leaking N because of a long-term N saturation or because of climatic shifts. At HBEF, for example, high rates of defoliation occurred during 1969–1971 due to an outbreak of a defoliating caterpillar, the saddled prominent (*Heterocampa guttivita*). It was further reported that during peak defoliation, ~44% of the total leaf tissue was consumed [*Bormann and Likens*, 1979]. Somewhat surprisingly, HBEF scientists have never attributed an 8-year period (1969–1976) of above-normal nitrate losses to this defoliation, despite the fact that the defoliation is well documented and that the second highest annual flow-weighted nitrate concentration in watershed 6 was measured during 1969, the year of peak defoliation [*Likens and Bormann*, 1995].

Documentation of N responses to insect defoliation also has relevance to managing N export from large complex watersheds to downstream ecosystems such as the Chesapeake Bay. *DeWalle and Pionke* [1994] recently examined spatial and temporal variations in the magnitude of N export from small forested watersheds in the Chesapeake Bay watershed, and they proposed that there are two geographic "zones" of N export within the watershed: a high N export zone and a low N export zone. Estimates of annual N loss from small forested watersheds form the basis for the computed average nitrogen loads from forest lands used by *Linker et al.* [1996] in their model of nutrient contributions to the Chesapeake Bay. Unfortunately, since few of the studies cited by *DeWalle and Pionke* [1994] had more than a 5-year record of N export, it is difficult to determine the potential uncertainties in N export associated with periodic defoliations. However, using the WOR long-term data set as an example, basing N export on the 5-year period from 1991 to 1995 would have resulted in an estimated annual N load (3.1 kg N ha^{-1} yr^{-1}) that is 30 times higher than the load computed for the 11-year period prior to 1991 (0.1 kg N ha^{-1} yr^{-1}). Since roughly 60% of its watershed is forested, there is a huge uncertainty associated with current estimates of the contribution of N from forested lands to the Chesapeake Bay. The results of this study, though, suggest that the average N load used by *Linker et al.* [1996] of 3.4 kg ha^{-1} yr^{-1} may be more characteristic of disturbance periods rather than periods without significant disturbance. Consistent with the hypothesis of *Bormann and Likens* [1979] for normally aggrading forests of intermediate age, it appears that few, if any, small forested watersheds in the mid-Appalachian region are leaking dissolved N in the absence of natural or human-induced disturbances. This conclusion appears to be generally consistent with recent analyses indicating little or no trend in nitrate concentrations in precipitation in this region during the period 1980–1992 [*Lynch et al.*, 1995]. However, additional studies of the controls on N losses from both disturbed and undisturbed forests in the Appalachian Mountain region are needed to understand the mechanisms by which N can be exported from these systems, either in the short term as the result of disturbance or in the long term as the result of N saturation.

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