

ARE NITRATE EXPORTS IN STREAM WATER LINKED TO NITROGEN FLUXES IN DECOMPOSING FOLIAR LITTER?

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Abstract.—The central hardwood forest receives some of the highest rates of atmospheric nitrogen (N) deposition, which results in nitrate leaching to surface waters. Immobilization of N in foliar litter during litter decomposition represents a potential mechanism for temporal retention of atmospherically deposited N in forest ecosystems. When litter N dynamics switch to the N-release phase, litter N may contribute to N exports. We tested the hypothesis that nitrate exports in stream water are positively related to the N dynamics in foliar litter, with generally low exports during N immobilization and generally high exports during N mineralization in litter. We performed a simple regression analysis to test this hypothesis. We used litter N flux and nitrate export data for 2002-2006 from two watersheds at the Fernow Experimental Forest in West Virginia, one receiving ambient and one receiving experimentally increased rates of atmospheric deposition. Linear regression of monthly stream nitrate export values with litter N flux revealed no relationship. We conclude that while foliar litter immobilizes N, and the amount of immobilized N is of similar magnitude as that arriving with deposition, this N-retention mechanism does not translate into lowered nitrate exports in stream. Further, nitrate exports do not increase during the litter N mineralization phase.

INTRODUCTION

Many of the hardwood forests in the eastern United States leach nitrate to surface waters (Stoddard 1994), with potentially far-reaching ecological and economic consequences. During leaching, nitrate combines with base cations such as calcium and magnesium. A prolonged export of both of these elements may lead to soil acidification, a decrease in forest productivity, decline in forest health, and a reduction in the capacity of forest ecosystems to sequester carbon (C) (Adams and others 2000). Additionally, non-point source pollution by nitrate leads to water acidification, and represents a potential health hazard to human and aquatic life.

Nitrate exports from forests have been strongly linked to atmospheric N deposition, land-use history, and the rate and duration of N loading (Aber and others 1989, 1998; Stoddard 1994; Goodale and others 2003; Pregitzer and others 2004; Castro and others 2007). The Central Hardwood Forest receives some of the highest rates of atmospheric N deposition in the eastern United States, reaching 11 kg N ha⁻¹ yr⁻¹ in parts of West Virginia (Adams and others 2000). Mass balance estimates show that most of the atmospheric N is retained in forests (Aber and others 1998, Goodale and others 2009). Studies utilizing isotopic techniques have demonstrated that nitrate in drainage waters is primarily produced by microbial mineralization and nitrification of organic N, which strongly suggests that atmospherically deposited N is cycled (McHale and others 2002, Piatek and others 2005, Campbell and others 2006, Hales and others 2007).

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Mechanisms of atmospheric N retention in impacted forests have been a focus of much research in the last couple of decades (Aber and others 1998). N retention processes remain elusive, however (Magill and others 2000). The hypothesis that abiotic complexation in soils occurred via a “ferrous wheel” mechanism had some merit (Davidson and others 2003) but was unsupported in a large study of different forest types (Coleman and others 2007). Immobilization in microbial biomass, increases in net primary productivity, and mycorrhizal uptake remain among the strongest candidates for likely mechanisms of N retention (Aber and others 1998).

Immobilization of N in the biomass of microbes colonizing fresh foliar litter during litter decomposition represents a potential mechanism for short-term N retention of atmospherically deposited N in forest ecosystems. Immobilization is a microbial process of sequestration of elements limiting microbial reproduction and growth when energy sources in the form of easily decomposable C are abundant (Gallardo and Schlesinger 1994). However, assessments of the capacity of microbial biomass to retain atmospheric N are conflicting, the conflict likely arising from the timing of N-label application and different ways of treating the forest floor (Zogg and others 2000). Namely, N dynamics in fresh hardwood foliar litter exhibit distinct phases. The highest rates of immobilization start shortly after leaf fall in October and last 4-6 months; mineralization begins shortly after peak immobilization (Adams and Angradi 1996, Piatek and others 2009a). Therefore, a summer application of N label when mineralization in fresh foliar litter begins will in effect nullify the microbial N sink activity (Fig. 1). Inclusion of the partly-decomposed forest floor together with fresh litter lowers the effective N sink because older litter exhibits net mineralization rather than immobilization. However, treating fresh litter apart from the old litter layers as well as applying N tracer in the fall is expected to yield a much stronger N immobilization, or N sink activity.

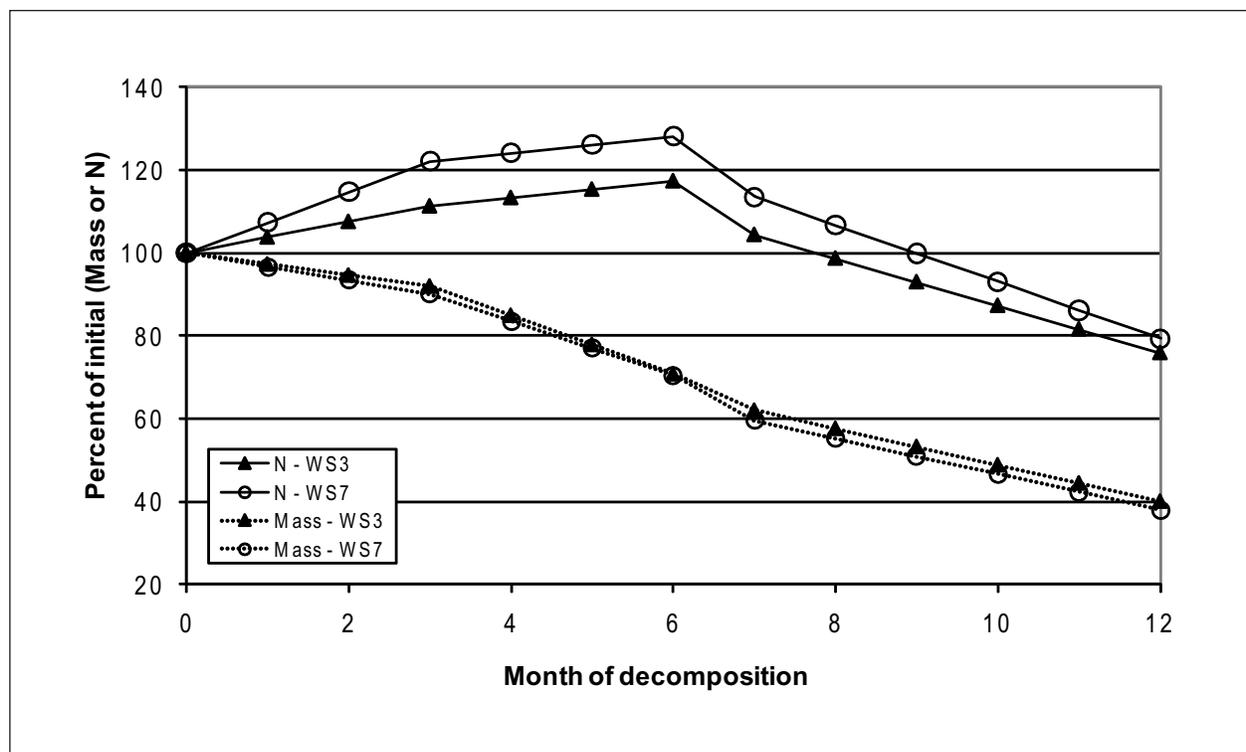


Figure 1.—Foliar litter N and mass loss dynamics (in percent of initial amounts) during 12 months of decomposition starting with litterfall in October (month 0) for two watersheds – reference (WS7) and high deposition (WS3). Data adapted from Piatek and others (2009a). “0” is October and “12” is October of the following year.

Several other lines of reasoning support the role of microbial N immobilization in foliar litter in N retention. First, rates of N immobilization in deciduous foliar litter are often of the same magnitude as rates of N deposition (Magill and Aber 1998, Piatek and others 2009a). Second, fresh foliar litter is the first point of interception of throughfall N with the soil. Third, immobilization in litter, and subsequent litter N mineralization and nitrification, yields a microbial isotopic signature of nitrate, such as is commonly found in drainage waters (McHale and others 2002, Piatek and others 2005, Campbell and others 2006, Hales and others 2007).

Recent work at the Fernow Experimental Forest in West Virginia showed that foliar litter composed of about 50 percent oak, 20 percent yellow-poplar, 15 percent maple, and smaller quantities of magnolia, pin and black cherry, and birch immobilizes at least 30 percent more N than initially present in fresh litterfall (Piatek and others 2009a). Depending on the total mass of litter, these litter N dynamics result in immobilization of 6 to 11 kg N ha⁻¹ yr⁻¹. For N deposition levels of 11 kg ha⁻¹ yr⁻¹, the above rates of N immobilization represent a potential retention of 55 to 100 percent of the amount of N in throughfall (Adams and others 2000). Lower rates of immobilization are common under conditions of increased rates of N deposition, while higher rates of immobilization are possible in stands with a higher oak component (Templer and others 2005; Piatek and others 2009a, 2010). When leaf litter begins to mineralize around April, plant N uptake can assume the primary role in N retention. The strength of these retention mechanisms is likely to decrease and then possibly subside when plant uptake diminishes and a new litter layer (Oi) is yet to form at the end of the summer into early fall.

We hypothesized that a positive relationship exists between N flux in decomposing foliar litter and stream nitrate dynamics. According to this hypothesis, lower nitrate exports should generally coincide with the N immobilization phase and higher nitrate exports should generally coincide with the mineralization phase. The objective of this study then was to test this hypothesis and to investigate a potential link between N dynamics in foliar litter and nitrate exports from forests. For this investigation we conducted a simple regression analysis of monthly litter N dynamics on stream nitrate exports for two watersheds at the Fernow between 2002 and 2006.

METHODS

LEAF LITTER NITROGEN DYNAMICS

Nitrogen dynamics (rates of immobilization and mineralization) in fresh litter were adapted from a study by Piatek and others (2009a) in a central hardwood forest at the Fernow Experimental Forest in Parsons, WV. Briefly, known amounts of fresh litter collected in the long-term soil productivity plots at the Fernow were allowed to decompose in nylon mesh bags for 12 months in 2006-2007. Litter was collected from, and later allowed to decompose in, reference (untreated) plots and plots treated with annual additions of 36 kg N and 40 kg sulfur (S) ha⁻¹ as ammonium sulfate (increased rates of deposition). Species composition and specific weight proportions reflecting the actual litter mix were (in percent of dry weight): oak (48), yellow poplar (19), maple (14), magnolia (12), cherry (4), and sweet birch (3). Mass loss and N contents were quantified at 0, 3, 6, 7, and 12 months after the start of the experiment. Monthly data were obtained by linear interpolation between data points (Fig. 1).

Mass loss and litter N dynamics from the above plot-level study (Fig. 1) were applied to litterfall mass for two watersheds at the Fernow for 2002–2006 (Table 1). Watershed WS3 receives annual additions of 36 kg N ha⁻¹ yr⁻¹ and 40 kg S ha⁻¹ yr⁻¹ in an effort to accelerate soil acidification and N leaching (Adams and others 2006). Watershed WS7 is a reference watershed. Both watersheds are approximately 40-year-old central hardwood forests. Nitrogen dynamics in litter differ between the reference and the treated stands in that immobilization levels are significantly higher in litter decomposing in the reference than in the treated stands (Fig. 1; Piatek and others 2009a). Therefore, treatment-specific N dynamics were applied to the correspondingly treated watersheds.

To calculate litter N dynamics, first, annual litterfall mass data for WS3 and WS7 were obtained for the watersheds from annual collections in litter traps. Detailed collection methods are provided in Adams (2008). Mass loss of this litter during a 12-month decomposition period was calculated as the product of initial litterfall mass and percent mass remaining from Figure 1 (Table 1).

Second, monthly N content in these litters was calculated separately for each watershed as the product of initial litter mass (watershed-level data) and initial litter N concentration and percent of initial N content remaining (plot-level data). Nitrogen immobilization was indicated by increases over initial litter N content (concentration times mass). Nitrogen mineralization started when litter N content decreased from maximum immobilization; net N mineralization was the decrease in N content below initial litter N (initial equals 100 percent) (Fig. 1, Table 2).

Third, N fluxes in litter were calculated as the difference in N contents between months (Table 3). Nitrogen fluxes were used in the regression in Figures 2 and 3. For the regression analysis, N flux during immobilization was treated as a loss (negative) because immobilization in effect temporarily removes N from the system. Nitrogen flux during mineralization was treated as an addition (positive) because mineralization contributes N to the system.

STREAM NITRATE EXPORTS

Volume-weighted monthly nitrate-N exports (kg N ha⁻¹ mo⁻¹) and nitrate-N concentrations (mg L⁻¹ mo⁻¹) in stream water were obtained for 2002 through 2006 for both watersheds. Stream chemistry has been monitored at the Fernow in gauged watersheds, and long-term data are available.

Table 1.—Litterfall mass (kg ha⁻¹) and examples of mass decrease during 12 months of decomposition in two watersheds, one treated with increased rates of N deposition (WS3) and a reference (WS7). Initial litter mass from Adams (2008); decomposition mass from Piatek and others (2009a) as shown in Figure 1. Litter mass at 3, 6, and 12 months is calculated from percent of initial mass remaining (Fig. 1). “0” denotes October and “12” denotes October of the following year.

Year	WS3				WS7			
	0	3	6	12	0	3	6	12
2002	2,920	2,686.4	2,067.4	1,168.0	3,100	2,790.0	2,185.5	1,844.5
2003	3,350	3,082.0	2,371.8	1,340.0	3,440	3,096.0	2,425.2	2,046.8
2004	2,910	2,677.2	2,060.3	1,164.0	3,250	2,925.0	2,291.3	1,933.8
2005	3,630	3,339.6	2,570.0	1,452.0	3,490	3,141.0	2,460.5	2,076.6
2006	2,890	2,658.8	2,046.1	1,156.0	3,360	3,024.0	2,368.8	1,999.2

Table 2.—Example litter N contents^a (kg ha⁻¹) during 12 months of litter decomposition^b in two watersheds, one treated with increased rates of N deposition (WS3) and a reference (WS7). Nitrogen immobilization is indicated by increases over initial litter N, gross mineralization is a decrease in litter N since peak immobilization, and net mineralization is a decrease in litter N below initial content. Nitrogen dynamics adapted from Piatek and others (2009a). “0” denotes October and “12” denotes October of the following year.

Year	WS3				WS7			
	0	3	6	12	0	3	6	12
2002	35.3	39.32	41.4	26.8	33.5	40.9	42.9	26.6
2003	40.5	45.1	47.5	30.7	37.2	45.4	47.7	29.5
2004	35.2	39.2	41.3	26.7	35.1	42.9	45.0	27.8
2005	43.9	48.8	51.5	33.3	37.7	46.0	48.3	29.9
2006	35.0	38.9	41.1	26.6	36.3	44.3	46.5	28.8

^aLitter N content is calculated as litter N concentration * litter mass; Initial N concentration for WS3 was 1.21 percent and for WS, 1.1 percent (Piatek and others, in press).

^bLitter N content during decomposition is calculated as initial litter N concentration * percent of initial N remaining (from Piatek and others, in press; Fig. 1).

Table 3.—Monthly litter N fluxes (differences between months in kg ha⁻¹ mo⁻¹) during 12 months of decomposition in two watersheds, one treated with increased rates of N deposition (WS3) and a reference (WS7). Negative fluxes denote immobilization and reflect N removal from the system. Positive fluxes denote mineralization from litter, or N input.

Month	WS3					WS7				
	2002	2003	2004	2005	2006	2002	2003	2004	2005	2006
Oct	0	0	0	0	0	0	0	0	0	0
Nov	-1.3	-1.5	-1.3	-1.6	-1.3	-1.3	-1.4	-1.3	-1.4	-1.4
Dec	-1.3	-1.5	-1.3	-1.6	-1.3	-1.3	-1.4	-1.3	-1.4	-1.4
Jan	-1.5	-1.3	-1.5	-1.3	-1.7	-0.7	-4.9	-5.4	-5.1	-5.5
Feb	-0.8	-0.8	-0.8	-0.7	-0.9	-0.7	-0.7	-0.8	-0.7	-0.8
Mar	-0.8	-0.8	-0.8	-0.7	-0.9	-0.7	-0.7	-0.8	-0.7	-0.8
Apr	-0.8	-0.8	-0.8	-0.7	-0.9	4.9	-0.7	-0.8	-0.7	-0.8
May	5.3	4.6	5.3	4.6	5.7	2.3	5.0	5.5	5.2	5.6
Jun	2.0	2.0	2.3	2.0	2.5	2.3	1.9	2.1	2.0	2.2
Jul	2.0	2.0	2.3	2.0	2.5	2.3	1.9	2.1	2.0	2.2
Aug	2.0	2.0	2.3	2.0	2.5	2.3	1.9	2.1	2.0	2.2
Sep	2.0	2.0	2.3	2.0	2.5	2.3	1.9	2.1	2.0	2.2

REGRESSION ANALYSIS

Simple regression analyses of monthly litter N contents with monthly stream nitrate-N exports and concentrations were performed using SAS[®] statistical analysis package (SAS Institute, Inc., Cary, NC). Litter N contents during the immobilization phase are considered as “-”, or removal phase, and as “+”, or addition phase, during mineralization. P-values for the regression models were considered significant at $\alpha \leq 0.05$.

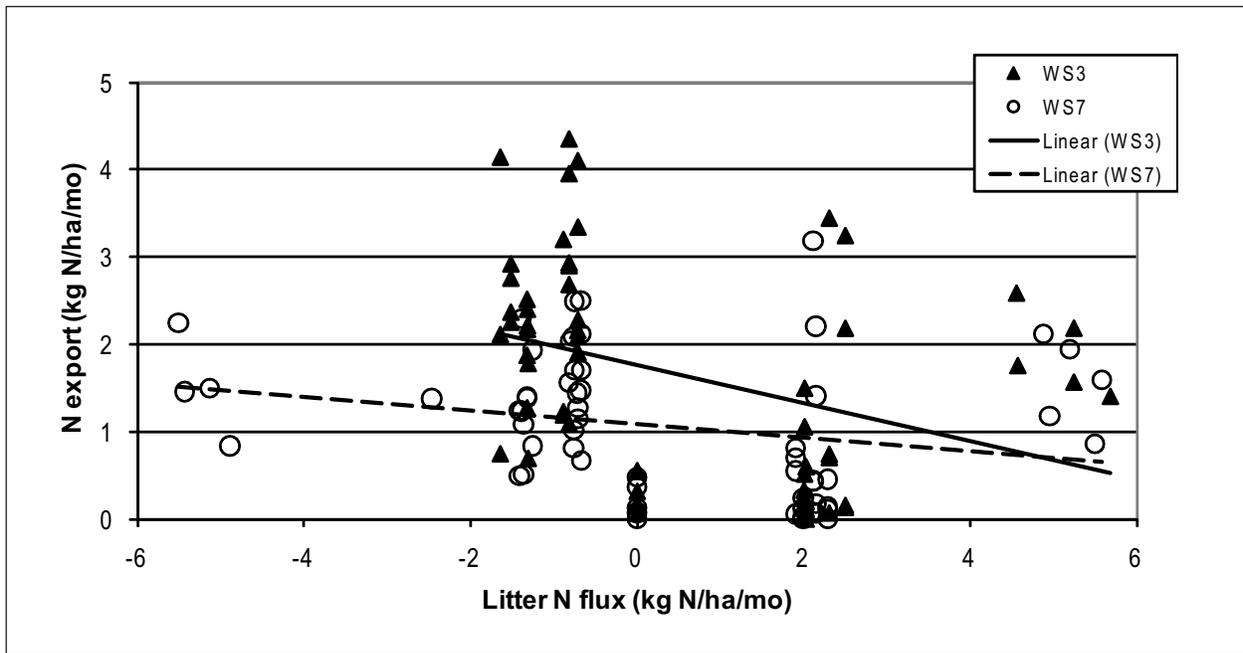


Figure 2.—The relationship of stream N exports to litter N flux. Negative litter N flux denotes microbial N immobilization as a form of N removal, while positive flux denotes N mineralization as a form of N addition. Litter N flux is the difference in N content between that in a corresponding month of decomposition and that in initial litter (from Table 1). Included are 2002-2006 monthly data for a reference (WS7) and a high-deposition watershed (WS3).

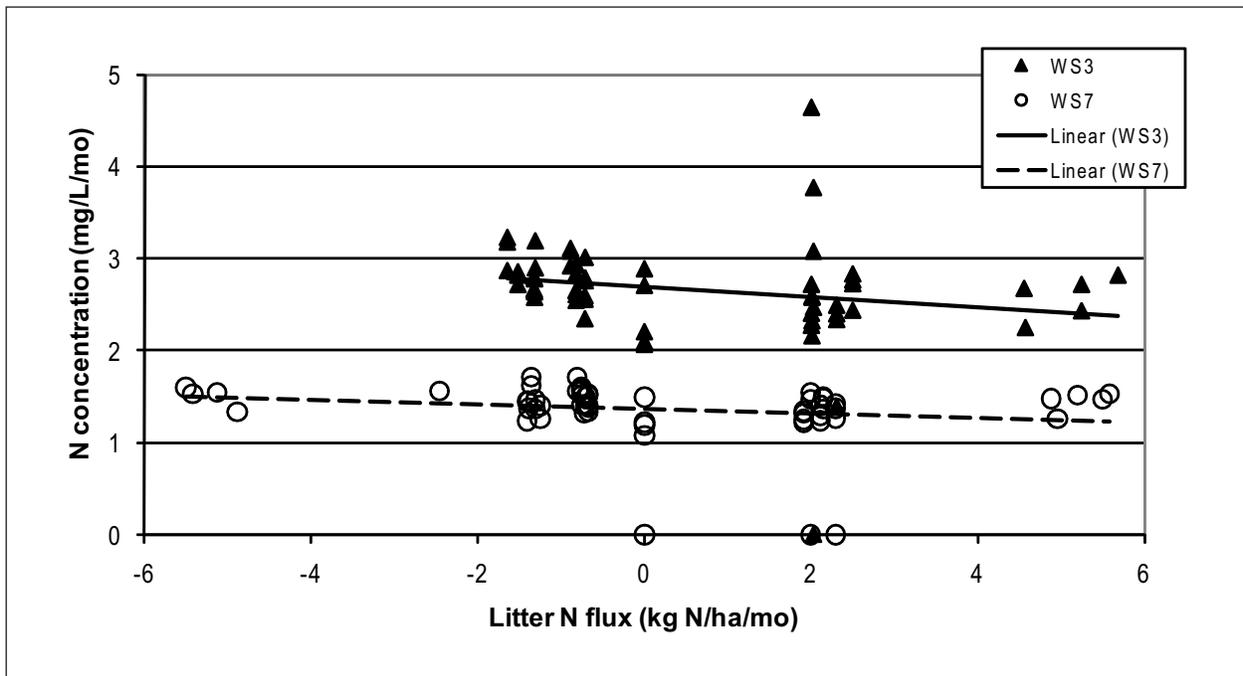


Figure 3.—The relationship of stream N concentrations to litter N flux. Negative litter N flux denotes microbial N immobilization as a form of N removal, while positive flux denotes N mineralization as a form of N addition. Litter N flux is the difference in N content between that in a corresponding month of decomposition and that in initial litter (from Table 1). Included are 2002-2006 monthly data for a reference (WS7) and a high-deposition watershed (WS3).

RESULTS

The reference watershed WS7 exhibited somewhat higher masses of litterfall than did the treated watershed WS3 for all years included except 2005. Decomposition decreased the mass of litter in both watersheds by about 40 percent in 12 months (Table 1). Nitrogen immobilization peaked in April, 6 months after decomposition started. Net mineralization started 2 months later in June (Fig. 1, Table 2). The highest monthly immobilization rates occurred during the first 3 months of decomposition. The monthly mineralization fluxes were almost two times greater than immobilization fluxes (Table 3).

We found no relationship between nitrate-N exports in stream water and litter N flux in the reference watershed (Fig. 2). Regression coefficients for the model evaluating the effects of litter N flux on stream nitrate-N exports were 0.12 ($p = 0.007$) for WS3, and 0.05 ($p = 0.079$) for WS7. There was also no relationship between stream nitrate-N concentrations and litter N flux (Fig. 3). Regression coefficients for the model evaluating the effects of litter N flux on stream nitrate-N concentrations were 0.03 ($p = 0.162$) for WS3, and 0.05 ($p = 0.106$) for WS7.

DISCUSSION

Under ambient N deposition in the reference watershed WS7, N immobilization in foliar litter did not coincide with low stream nitrate-N, and N mineralization did not coincide with high stream nitrate-N. In WS3, however, that relationship was significant, and litter N dynamics appeared to explain a small amount of variation in stream nitrate-N levels (r^2 for the regression model = 0.12). However, the relationship in the treated watershed was inverse, with higher nitrate exports during periods of N immobilization into litter, opposite of what we hypothesized (Fig. 2). We conclude that our hypothesis about the effects of decomposing foliar litter on stream nitrate levels was not supported in either watershed.

The significance of the relationship between nitrate exports and litter N in the treated watershed appears to be statistical rather than biological. The treated watershed exported more N, on average $0.5 \text{ kg N ha}^{-1} \text{ mo}^{-1}$, reaching up to $2 \text{ kg N ha}^{-1} \text{ mo}^{-1}$ more in any one month and year than the reference. At the same time, rates of litter N immobilization were lower in the litter in the treated watershed than in the reference (Fig. 2). Increased nitrate exports and decreased levels of N immobilization are typical forest-ecosystem responses to elevated levels of atmospheric N deposition (Aber and others 1989, 1998). Further, in this study, nitrate exports in summer were similar for both watersheds, probably due to comparable levels of N uptake by the vegetation; N mineralization levels from litter at that time were also similar. Therefore, the differences between the watersheds appear to consist of a combination of higher nitrate exports and lower immobilization in the treated watershed than in the reference.

Litter in the reference watershed has a greater capacity for immobilization than litter in the treated watershed (Fig. 1; Piatek and others 2009a). Even with the higher capacity for potential N retention, however, litter in the reference apparently neither removed sufficient N from the system to affect stream nitrate levels, nor did it contribute enough N to result in a significant increase in stream nitrate. In view of some new evidence that N mineralization from litter may not yield soluble forms of N as is typically assumed (Piatek 2011), it is possible that decomposing litter does not contribute soluble N. Such a functional detachment of N processes in fresh litter from N processes in partly decomposed forest floor (Oa+Oe) has been observed in hardwood forests (Currie and Nadelhoffer 1999, Fisk and Fahey 2001), and may apply to stream N exports.

Nitrate expression in stream is a function of both nitrate production and hydrologic control. Even if litter N entered soil horizons, it would become part of the soil-available N pool. Soil N mineralization rates under hardwood forests are high at 10.5 kg N ha⁻¹ mo⁻¹ for WS7 and 11.2 kg N ha⁻¹ mo⁻¹ for WS3, with very high rates of nitrification, at 91 and 105 percent of mineralization (Gilliam and others 2001). By comparison, monthly litter N fluxes averaged 2 kg ha⁻¹. Soil N cycling rates are sufficiently high to dilute the flux of N from litter, as well as from atmospheric sources. According to the flushing-draining hypothesis of nitrate movement, once nitrate is produced in soils, it may be flushed to the stream if the nitrate-source area is hydrologically connected to the stream during high levels of soil moisture. Alternatively, nitrate may drain to groundwater, when hydrologic connectivity with stream is lost at low soil-moisture levels (Creed and others 1996). This mechanism has been confirmed for some forested watersheds in the northeastern United States, and may operate at the Fernow (Piatek and others 2009b). The flushing-draining mechanism can disconnect soil N dynamics spatially and temporally from stream nitrate exports. Therefore, the next step in an effort to explain the dynamics of monthly nitrate exports in these watersheds will be to address the effects of hydrologic fluxes.

We now discuss the validity of our approach of applying litter N dynamics from one area, albeit nearby, to watersheds which may have a different species assemblage. Foliar litter invariably immobilizes N (e.g., Adams and Angradi 1996, Piatek and Allen 2001) as a result of N-limitation in forested ecosystems, including soil microbial biomass. Therefore, starting after litter fall in October, N contents in litter steadily increase over the initial level, until N mineralization begins, at which point N contents in litter decrease. In mixed-species litters, the magnitude of the increase varies depending on the relative amounts of easily decomposable and recalcitrant litters (Piatek and others, in press), but the overall immobilization/mineralization dynamics are preserved. Therefore, while total N numbers in litter may be affected under different species assemblages, immobilization will temporarily retain N and mineralization will contribute N to the system, preserving the relationship as analyzed here. As such, the validity of using litter N dynamics across similar but not the same watersheds is fully supported.

CONCLUSIONS

Nitrogen dynamics in foliar litter were not related to monthly nitrate levels in stream water under ambient or experimentally elevated rates of N deposition for the 5 years evaluated. It appears that litter N dynamics may be disconnected from stream nitrate dynamics.

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