9. Nitrogen Saturation in Experimental Forested Watersheds

Ivan J. Fernandez and Mary Beth Adams

Daniel Rutherford is credited with the discovery of nitrogen (N) in 1772. A.L. Lavoisier, the 18th century French chemist, called this element "azote" which translates to "without life" because free N is incapable of supporting life: a fitting name for this element given the paradoxes it presents to society and its role in global ecology. Despite being the most abundant element in the atmosphere, usually quoted as 78% by volume, primarily as N_2, life on the planet does not rely on either the uptake or release of N for the two most important complimentary biological processes, namely respiration and photosynthesis. Nonetheless, N has long been heralded as the most commonly limiting mineral nutrient for plant growth around the globe, and indeed is essential for the formation of amino acids, and thus proteins, for all life forms. No other element has been studied more extensively in the context of agricultural and forest ecosystems than N, yet at no time in the past has there been greater interest in research addressing information needs regarding N.

To add to the irony, N is the most commonly used fertilizer in crop and forest management, with increasing amounts used to maintain production goals. Yet in the past several decades, we have realized that N has its dark side in the environment. Fossil fuel combustion typically releases N gases (nitrogen dioxide [NO_2], nitrogen oxide [NO]) into the atmosphere that

can be transported long distances and, along with sulfur dioxide (SO_2), create strong mineral acids in the atmosphere that return to terrestrial ecosystems in wet and dry forms as acid deposition. Atmospheric N also contributes to the formation of tropospheric ozone (O_3) or "smog." Ironically, N fertilizers have a high energy requirement for their fabrication. This means that high fossil fuel consumption is usually necessary to produce the electricity needed to manufacture a fertilizer form of N to be added to the landscape, while at the same time also contributing to the long-range transport of N in the atmosphere. Vitousek et al. (1997) recently discussed the indisputable human influence on global N cycles, emphasizing the serious and long-term environmental consequences we can expect as a result of our perturbation of this system on a global scale. Galloway (1998) estimated that, in 1997, human activities contributed 150 Tg N to the global terrestrial environment, of which 80% was due to food production (with twice as much attributable to fertilizer production as biological N fixation through cultivation) and 20% due to energy production.

During the past two decades there has been an emerging concern for the consequences of increased atmospheric deposition of N to forested ecosystems, despite the continued interest in certain regions in forest fertilization with N to promote growth. Early concerns were focused on the possibility of N deposition having direct effects on tree foliage and physiology, which later shifted to a focus on soil acidification and related indirect effects on forest health. More recently, it has become evident that chronic N deposition to forests may result in complex responses at both the organismal and ecosystem levels. In the mid-1980s two important experiments were initiated in the eastern U.S. using whole-watershed chemical manipulations to determine the effects of chronic N and sulfur (S) deposition on forested ecosystems. The focus of these studies has been to investigate the biogeochemical response of forested stream catchments to experimental treatments, and more recently research has included other aspects of ecosystem response. These studies have focused on the interaction between forested watersheds and the chemical climate, and the long-term nature of these studies is increasingly providing insight on interactions with physical climatic factors as well. This chapter discusses some of the highlights of these whole-watershed studies during the first six to seven years of experimental manipulation.

Nitrogen Deposition and Nitrogen Saturation

The concept of nitrogen saturation was developed in the early 1980s in Europe by scientists concerned with atmospheric deposition of N. In 1981, Swedish scientist T. Ingestad first presented a model that defined the initial concept (Aber, 1992). Many other European scientists have since worked

on the N saturation concept. One noteworthy example is the "ammonium hypothesis" offered by Nihlgård (1985), based on observations of very high deposition of ammonium (NH_4) in the Netherlands. Ågren and Bosatta (1988) discussed the role of plant vs. soil processes in the evolution of N saturation, concluding that the soil subsystem must saturate first as the condition of N saturation develops.

Europeans have been known for their work in developing the concept of "critical loads" for N and S (Bull, 1991). This concept was defined in a Swedish workshop as "a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" (Nilsson and Grennfelt, 1988). This definition is more conceptual than the earlier definitions of N saturation, but specific policy goals have resulted from the critical loads approach in Europe. Nilsson (1986) summarized the outcome of a Nordic working group on critical loads who concluded approximately 10 to 20 $kg\,N\,ha^{-1}\,yr^{-1}$ was the critical load for forests under conventional management. Ågren and Bosatta (1988) concurred with this estimate based on their own evaluation. Løkke et al. (1996) recently discussed some of the uncertainties that exist with the widespread utilization of this approach in regulating transboundary air pollution in Europe.

In the U.S., the concept of "nitrogen saturation" has received the greatest amount of attention as a condition of "excess" N in the ecosystem. Aber (1992) points out that several definitions of N saturation exist depending on whether this condition is defined as (1) the absence of a growth response in vegetation to N additions, (2) the initiation of NO_3 leaching, or (3) the lack of a net N accumulation in ecosystems as evidenced by an equivalence between inputs and losses. He included in his article a useful table shown here as Table 9.1 that suggests key mechanisms involved with ecosystem response to N, and the nature of those changes.

Several authors have discussed the evolution of N saturation in forested ecosystems and provide a useful framework not only for understanding the processes controlling these phenomena, but also for designing hypothesis-driven research to define mechanisms essential for accurate predictions of future response. Aber et al. (1989) defined N saturation as the availability of NH_4 and nitrate (NO_3) in excess of total combined plant and microbial nutrient demands (and excluding their use as a substrate for denitrification). They described the stages of the development of N saturation we can summarize as Stage 0, N limitations to biological processes ranging from tree growth to litter decomposition; Stage 1, initial increases in foliar N concentrations followed by increased biomass; Stage 2, N saturation where N no longer limits biological function; and finally, Stage 3, forest decline due to a range of potential mechanisms impacting forest health. Aber et al. (1998) recently refined their conceptual

Table 9.1. Characteristics of N-limited and N-saturated Forest Ecosystems (Aber, 1992)

Characteristic	N-limited	N-saturated
Form of N cycled (net, as plant uptake)	100% NH_4	25–50% NO_3, 50–75% NH_4
Soil DOC concentration	High	Low
Ratio of gross NO_3 immobilization to gross nitrification	Near 100%	Near 0%
Ratio of gross NH_4 immobilization to gross mineralization	High (90–95%)	Low (50%?)
Fraction of soil fungi that are mycorrhizal	High	Low
Nitrate loss during snow melt	Low	High
Nitrate loss at base flow	Zero	High
Foliar lignin concentration	High	Low
Foliar N concentration	Low	High
Foliar free amino acid (e.g., arginine) concentration	Zero	High
Soil C/N ratio	High	Low
N_2O production	Zero	High
CH_4 production	High	Low (zero?)

model of N saturation based on recent research. They included in their concept of the evolution of forest ecosystems toward N saturation a decline in both calcium (Ca)/aluminum (Al) and magnesium (Mg)/N ratios in foliage, and a decline in N mineralization in Stage 2, the latter not yet well understood. In this article they point out the paradox that forest studies using N manipulations have shown, and that is the strong retention of added N to forests despite the limited carbon (C) pool in soils to facilitate free-living microbial fixation. They discuss a number of possible mechanisms to explain this phenomenon. Their conclusion is that mycorrhizal conversion of N to organic forms may offer the most plausible mechanism since mycorrhizae have direct access to photosynthate without requiring its conversion to plant biomass and subsequent microbial degradation. This promising hypothesis remains to be tested.

Stoddard (1994) has been widely credited with defining the evolution of N saturation based on watershed retention of N and surface-water chemical response. Accordingly, he defined Stage 0, net N retention due to microbial and forest uptake with the exception of spring snowmelt; Stage 1, as with Stage 0 but with amplified export of NO_3 during snowmelt or spring episodes; Stage 2, decreased control of biological uptake of N and an increase in base flow NO_3 concentrations; and Stage 3, no biological N deficiency is evident and the watershed becomes a net source of N. Both approaches have provided conceptual templates for understanding the process of N saturation and the important terrestrial–aquatic linkages involved. Jeffries and Maron (1997) question the adequacy of

either the N saturation or critical loads approach to defining ecosystem response to N enrichment alone, and argue for the addition of a third approach, that of careful monitoring of ground-layer species and mosses sensitive to soil N status to fulfill a critical gap in our understanding of the mechanisms of N transfer within and between ecosystems. Their recommendation seems sound, and would provide another tool to evaluate ecosystem condition that uses factors that integrate many processes, much as we used stream chemistry in watershed studies.

The greatest concern for N saturation in the U.S. has been in the northeastern states, and particularly at high elevations where deposition rates are greatest. Driscoll and Van Dreason (1993) reported increasing NO_3 concentrations in nine lakes of the Adirondack mountains of New York, despite the lack of any significant temporal change in N deposition throughout this region. They also noted 13 of the 17 lakes they studied showed declines in SO_4 concentrations commensurate with the documented declines in SO_4 deposition throughout the northeastern U.S. Sullivan et al. (1997) evaluated several extensive data sets on surface-water chemistry for this region, and concluded that the focus on S in the past may have underestimated the contribution of N to historical acidification, and that ignoring the role of N in surface-water acidification in the region may result in serious overestimations of recovery. McNulty et al. (1990) examined properties of forest soils in high-elevation spruce–fir sites throughout the Northeast from New York to Maine. They reported a spatial coincidence between the gradient of N deposition throughout the Northeast corridor, and N mineralization and nitrification potentials. They concluded that indicators of high rates of N cycling and available N in the systems receiving the higher atmospheric inputs of N might be evidence for the migration of these ecosystems to a condition of N saturation. McNulty et al. (1993, 1996) used N amendments to spruce–fir plots at a high-elevation site in southeastern Vermont to evaluate the mechanisms of response to treatments. They showed evidence that N enrichment resulted in changes in foliar N concentrations, basal area growth, nutrient balances, N cycling, and species composition based on initial differences in regeneration success on the treated plots. These results support and refine the model presented above for the terrestrial response to N saturation as described by Aber et al. (1989). Other N-saturated systems in the U.S. include the pine forests of southern California, unusual because of the very high levels of dry N deposition (Fenn et al., 1996), and high-elevation catchments in the Rocky Mountains, saturated because of very low uptake by biota (Williams et al., 1996). Land use history (e.g., fire, harvesting, blowdown) also appear to have a lasting effect on the susceptibility of forests to N saturation and influences ecosystem C storage in soils (Aber and Driscoll, 1997). It should be noted that the net effects of atmospheric N deposition on C sequestration in northern forests remains uncertain. Nadelhoffer et al. (1999) recently suggested that

N deposition is unlikely to explain the reported C sink in midlatitude forests attributing <20% of current C uptake by forests to N.

In order to study the effects of chronic elevated N deposition on forested ecosystems in situ, we can either (1) watch the evolution of existing ecosystems, over long periods of time, that are subjected to elevated or changing atmospheric deposition of N, (2) study differences among forest sites along carefully delineated spatial gradients of N deposition on the landscape, or (3) experimentally manipulate the deposition of N to the ecosystem. Sullivan (1997) points out the dangers of utilizing numerical simulation models based on an imperfect understanding of processes in ecosystems, and underscores the value of ecosystem manipulation experimentation as a means of testing these models. Plot-scale studies, such as the ones discussed above by McNulty and Aber (1993), are excellent and cost-effective ways to address specific hypothesis regarding mechanisms of response. However, this scale of study is not adequate for defining whole ecosystem responses, particularly with respect to biogeochemical processes that are strongly related to ecosystem hydrological properties. Nor do plot-scale studies allow us to study how different ecosystem components are integrated across landscapes. It is for this reason that watershed studies have been so valuable in understanding the integration of processes at the whole-ecosystem scale. Readers are encouraged to visit special issues of the journal Forest Ecology and Management, Vol. 71, 1/95 and Vol. 101, 2/98 (Wright and van Breeman, 1995; Wright and Rasmussen, 1998) for a collection of papers on the NITREX project, where whole catchments or large forest stands at 12 sites across a modern gradient of N deposition in Europe were used to study N deposition effects on forested ecosystems. Throughout these sites they employed a range of techniques to both add N as well as remove ambient levels of N to study both the risk of N saturation and the characteristics of recovery.

Two whole-watershed experimental N manipulation programs have also been conducted in forested ecosystems in the eastern U.S. since the 1980s, in Maine and West Virginia, specifically designed to study the effects of elevated N and S deposition. We know of no other comparable studies on the continent, and the following sections describe some of the findings to date.

Experimental Watersheds

Watershed Descriptions

The Bear Brook Watershed in Maine (BBWM) is located in eastern Maine (lat 44°52′N, long 68°6′W) approximately 50 km from the Atlantic Ocean. The site lies on the southeastern slope of Lead Mountain, with total relief of 210 m and a maximum elevation of 475 m. Two nearly perennial, low dissolved organic carbon (DOC), low acid neutralizing capacity (ANC)

Table 9.2. General Characteristics of the Study Sites

	BBWM	FEF
Watershed Pair	East Bear (Reference) West Bear (Treated)	WS4 (Reference) WS3 (Treated)
Watershed Area	10.2/10.9 ha	34/39 ha
Relief	265–475 m	735–860 m
Soil Types	Typic Haplorthods	Typic Dystrochrepts
Soil Depths	<1 m	<2 m
Soil Texture	Fine sandy loam	silt loam
Bedrock	Siltstone/granite	sandstone/shale
Stream Yield	65%	45%
Annual Temp	5°C	10°C
Annual Precip	130 cm	145 cm
Forest Types	Northern hardwood/ spruce	Central Appalachian hardwoods

streams (East Bear Brook and West Bear Brook) drain 10.2 and 10.9 ha contiguous watersheds (Table 9.2). Vegetation at the site is dominated by northern hardwoods (*Fagus grandifolia* Ehrb., *Acer rubrum* L., *Acer saccharum* Marsh., *Betula alleghaniensis* Britt., *Betula papyrifera* Marsh., and *Acer pennsylvanicum* Marsh.) with stands of softwoods (*Picea rubens* Sarg., *Abies balsamea* Mill., and *Tsuga canadensis* [L.] Carr.) at higher elevations. Soils are coarse, loamy, mixed, frigid Typic Haplorthods developed on till. Bedrock consists of quartzites and meta-pelites intruded locally by granite.

Chemical additions of N and S, as ammonium sulfate ($[NH_4]_2SO_4$), have been delivered bimonthly via helicopter to the West Bear watershed beginning in November 1989 and continuing through the period of data reported and remain ongoing. The adjacent untreated East Bear watershed serves as a reference. Total experimental loadings were 1800 eq ha^{-1} yr^{-1} (~25.2 kg N ha^{-1} yr^{-1} and ~28.8 kg S ha^{-1} yr^{-1}), which is ~3× and ~2× the ambient-wet-plus-estimated-dry deposition of N and S, respectively (Rustad et al. 1994). Total N deposition was approximately 1.5× that of the highest estimated wet-plus-dry deposition in the U.S. (NADP, 1990), but was less than 70% of the total N deposition in areas of central Europe (Dise and Wright, 1992). Additional details on the BBWM site can be found in Kahl et al. (1993), Norton et al. (1994) and Norton and Fernandez (1999).

The Fernow Experimental Forest, located near Parsons, West Virginia (lat 39°3′15″N, long 79°42′15″W) is situated on the unglaciated Allegheny Plateau in the Allegheny Mountain Section, within the Central Appalachian Broadleaf Forest Province (Bailey et al., 1994). The two watersheds (Watershed 3 (WS3) = treatment watershed; Watershed 4 (WS4) = reference watershed) are drained by second-order streams. Precipitation is distributed evenly between dormant and growing seasons, and averages 145 cm per year. Average annual precipitation pH is 4.2 but lower values

are common in summer (Adams et al., 1994). The predominant soils on both watersheds are loamy-skeletal, mixed mesic Typic Dystrochrepts. Elevation on the watershed ranges from 735 to 870 m.

Watershed 3, the treatment watershed (34 ha, see Table 9.2), contains a young stand that originated in 1969 from natural regeneration following clearcutting. Dominant tree species include *Prunus serotina* Ehrh. *Acer rubrum* L., *Betula lenta* L., and *Fagus grandifolia* Ehrh. Watershed 4, the control watershed (39 ha), contains a relatively undisturbed second growth stand of central Appalachian hardwoods. The current stand is approximately 90 years old, but some residual trees may be up to 200 years old (J.N. Kochenderfer, unpublished data). Dominant tree species include *Acer saccharum* Marsh., *Acer rubrum* L., *Fagus grandifolia* Ehrh., and *Quercus rubra* L.

Chemical additions of $(NH_4)_2SO_4$ fertilizer have been applied to WS3 since January 1989. Fertilizer was applied at a rate double the ambient N and S throughfall inputs, which was considered to approximate the combined N and S inputs of wet and dry deposition. Because ambient deposition varied seasonally, three applications were made each year, in March, July, and November. Yearly rates were $\sim 40\,kg\,S\,ha^{-1}$ and $\sim 35\,kg\,N\,ha^{-1}$. Additional details can be found in Adams et al. (1997).

Streams as Integrators of Response

Our discussion focuses on the behavior of these forested watersheds in response to experimentally increased N deposition. By definition, a watershed represents the land area contributing to a specific point of hydrological export, and integrates the inputs and losses of water from that system. Likewise, watersheds integrate biogeochemical processes reflecting the input and transformation of materials within that same hydrologically defined system.

Fig. 9.1 shows the long-term time series of stream water nitrate (NO_3) concentrations at both BBWM and Fernow. Measurements began in 1987 at BBWM, and, although a longer record of measurements is available for Fernow (Adams et al., 1994), our analyses begin with 1984 to facilitate a comparison between the two experimental watershed study programs. Treatments of $(NH_4)_2SO_4$ were initiated in both watersheds in 1989, starting in November for BBWM and in January at Fernow. Thus, data from 1988 and before shows how comparable the two streams were to each other at each site relative to stream water NO_3 concentrations prior to the onset of experimental manipulations. Although variability exists in these data, evidence presented suggests that the reference watersheds chosen behave nearly identical to the treatment watersheds prior to the commencement of treatments. A noteworthy difference between BBWM and Fernow is that a clear seasonal pattern in NO_3 concentrations exists at BBWM even before the treatments, and that the concentrations of NO_3

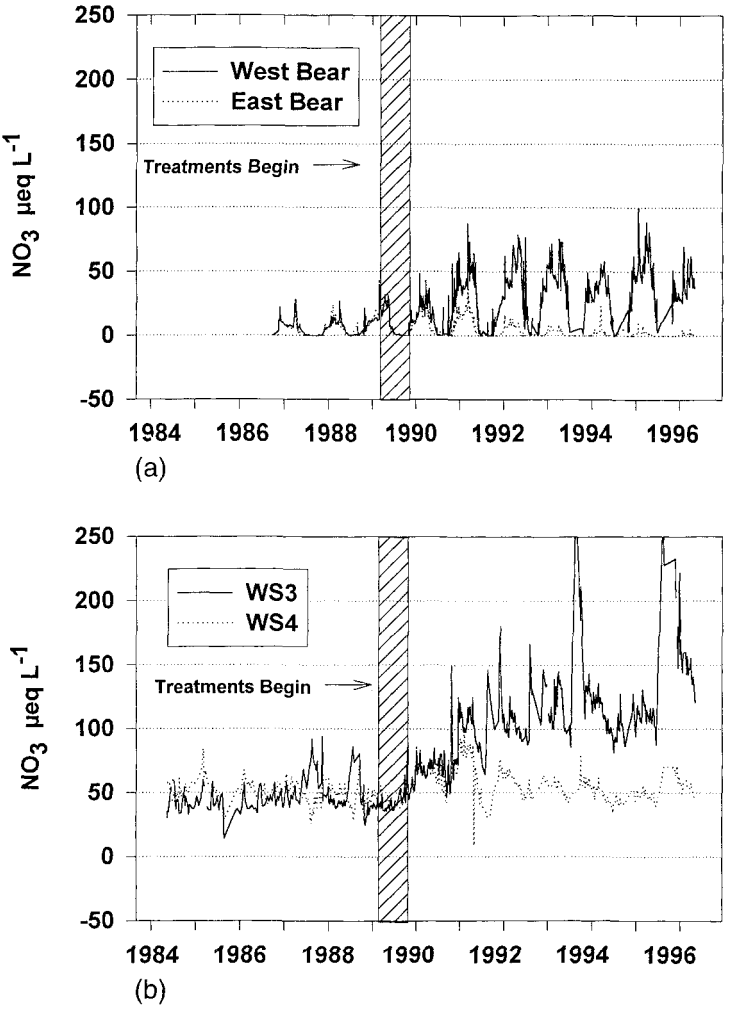


Figure 9.1. Stream NO_3 concentration time series for (a) BBWM and (b) Fernow.

are low and often below detection. This is the behavior we would expect in Stage 0 as described by Stoddard (1994) where N is biologically limiting and only during spring snowmelt, prior to vegetative uptake, are significant losses of inorganic N detected. The situation at Fernow is markedly different. Even before treatment began, there is only a vague representation of a seasonal pattern in stream water NO_3 concentrations, with a relatively constant stream water concentration of $\sim$50 µeq l^{-1} NO_3. This loss of seasonality in NO_3 concentration is indicative of the lack of biological controls on an increasingly N-enriched ecosystem referred to by

both the Stoddard (1994) or Aber et al. (1989) models as Stage 2 in the progression to N saturation. Peterjohn et al. (1996) examined the 23-year record of stream water NO_3 concentrations in WS4 at Fernow. They were able to define three distinct periods of increasing NO_3 export and decreasing seasonality: an initial period of low variability, a period of high peak concentrations and high variability, and, most recently, a period of high concentrations and low variability. While these patterns of response to elevated N deposition seem well demonstrated in forested watersheds, Peterjohn et al. (1996) pointed out the need to better define the mechanisms controlling them. Indeed, as seen later in this chapter, even the treated watersheds exhibiting classic N saturation characteristics are still retaining most of the annual inputs of N. Ecosystem mechanisms controlling the retention of N and the evolution of N saturation characteristics remain poorly understood, with some of the most recent speculation being discussed earlier such as that of Aber et al. (1998).

Both the treated watersheds at BBWM (West Bear) and at Fernow (WS3) responded to the NH_4 amendments with an increase in stream water NO_3 concentrations. This indicates that the added NH_4 was either rapidly nitrified or treatments increased the rate of nitrification of native soil N pools. There is reason to believe that both processes are responsible, but there is evidence to suggest that initial increases in NO_3 export were largely attributable to native soil N (Nadelhoffer et al., 1999). Increased stream-water NO_3 in West Bear retained a strong seasonality, with increasing magnitude of peak concentrations as compared with the reference watershed best representing the characteristics of the response. The time series shows some evidence at BBWM of an increasing length of the period of elevated NO_3, but during the first three years the duration of low NO_3 concentrations during the annual cycle remained constant. In Fig. 9.1 it should be noted that while West Bear appears to show little increase in maximum NO_3 concentrations during the treatment period, the period between 1990 and 1993 actually had an elevated NO_3 export that was partially masked by declining ambient NO_3 export evident in the reference East Bear stream NO_3 concentrations. This pattern of elevated NO_3 export during the early 1990s was evident in forested watersheds throughout the northeastern U.S. Regional NO_3 stream concentrations then appeared to decline and returned to essentially zero in the reference watershed at BBWM. Mitchell et al. (1996) attributed this period of regionwide elevated surface-water NO_3 concentrations to an anomalous cold period in December of 1989, triggering several years of elevated but declining surface-water NO_3 concentrations. At BBWM, overall declining NO_3 concentrations in the reference East Bear watershed between 1990 and 1995 mean that net relative increases continued to occur in the treated watershed during this period when compared with the reference East Bear watershed. This pattern of surface-water NO_3 potentially attributable to relatively short-term climatic trends also points out the value and need for

long-term, whole-ecosystem studies such as these to ascertain the role of climate on these ecosystems.

Fernow watersheds did not exhibit the period of elevated NO_3 export seen further north in the Northeast during the early 1990s, and NO_3 concentrations appear to show a continuous increase over time in response to treatments. Since biological control on stream-water NO_3 patterns are minimal due to excess N deposition at this site, it is logical that increased exports in WS3 beyond the reference WS4 would also not reveal seasonal patterns. There appears to be occasional instances of unusually high NO_3 concentrations but the data and analysis presented here do not attempt to determine their relationship to climatic patterns or antecedent conditions. It is interesting to note that Fig. 9.1 shows a 10-year record of NO_3 concentrations in WS4, with little evidence for increasing or decreasing trends. This may indicate that $\sim 50\,\mu eq\,l^{-1}$ NO_3 represents steady state with current levels of N deposition and stand development in this watershed. The time series suggests that stream NO_3 concentrations in WS3 are becoming more variable, but with the overall trend of steadily increasing concentrations that are twice or more the concentrations in WS4 after six years of treatment.

A great deal of literature has described the importance of atmospheric deposition on soil cation processes, with typical concerns being accelerated base cation leaching and aluminum (Al) mobilization (Likens et al., 1996; Lawrence et al., 1995, 1997; Shortle and Smith, 1988; Shortle et al., 1995). As with ambient deposition, the treatments at both BBWM and Fernow include a source of both N and S, and the effects of each element individually can not be definitively separated from the other in these studies. However, concern for net base cation losses due to chronic atmospheric deposition, and increasing concern for N deposition, has resulted in greater attention to the relationship between N saturation phenomena and base cation depletion. Both BBWM and Fernow are initiating research to specifically address some of these concerns. There is no question that $(NH_4)_2SO_4$ amendments to acid forest soils can result in accelerated depletion of Ca and the mobilization of Al depending on the rate of application. Carnol et al. (1997) recently demonstrated this phenomena in a Norway spruce (*Picea abies*) stand in England where $(NH_4)_2SO_4$ treatments caused soil water concentrations to "switch" from Ca to Al leaching, with relatively low Ca/Al ratios. They concluded that the most sensitive soils to acidification were those with low base saturation, typical of many northeastern forest soils in the U.S.

Fig. 9.2 shows the time series for Ca concentrations in the streams. The graph for BBWM shows the divergence between West Bear and East Bear as streams respond to the treatments. An apparent decline in Ca concentrations in East Bear over the period may reflect the declining contributions of NO_3 in stream water due to the climate-induced regional trends discussed earlier, and thus an overall decrease in the anion export.

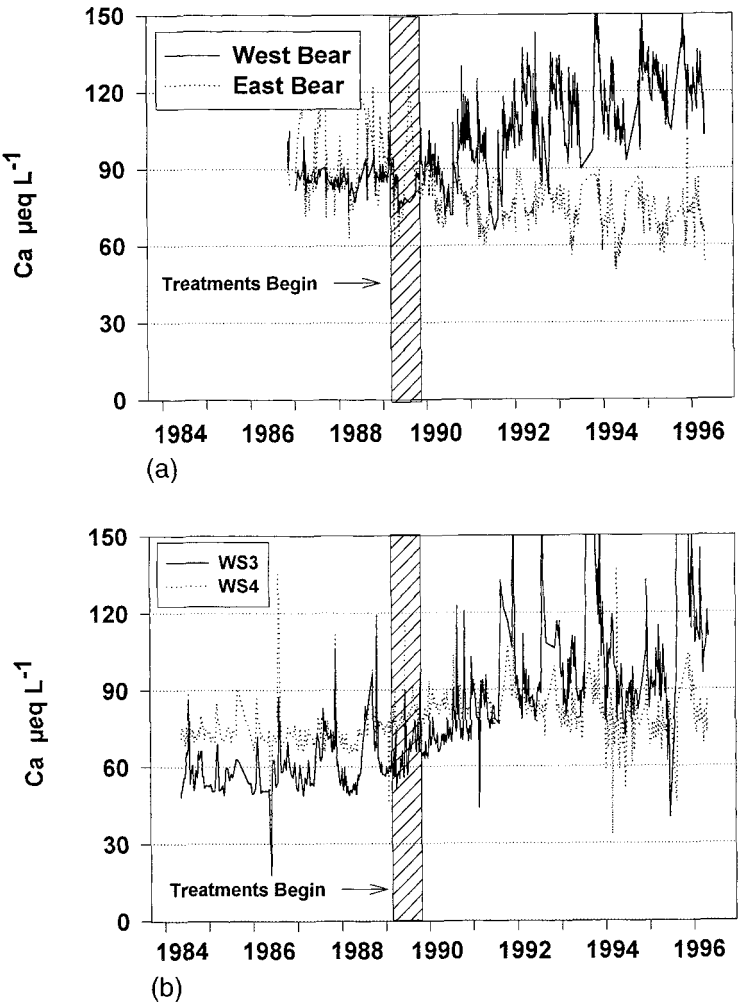


Figure 9.2. Stream Ca concentration time series for (a) BBWM and (b) Fernow.

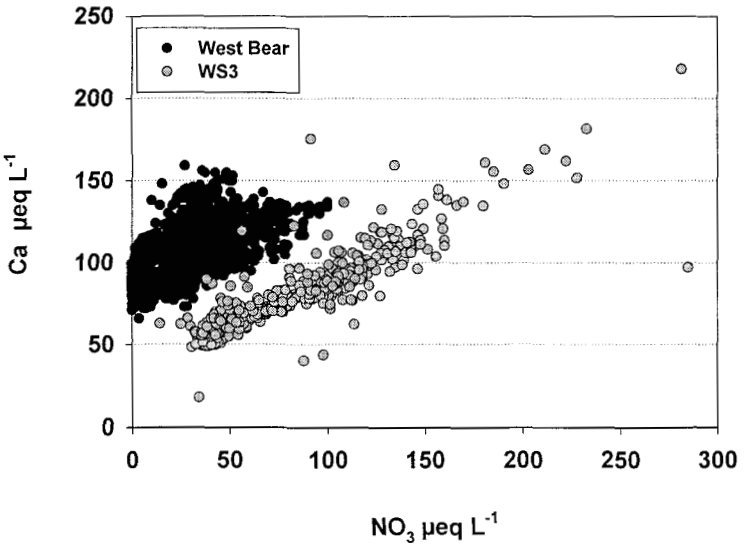
West Bear clearly is showing increased export of Ca with treatment. This accelerated base cation leaching is partly attributable to NO_3 as evidenced by the parallels between Ca and NO_3 concentrations' seasonal patterns, and due to simple ion balance calculations discussed elsewhere (Norton et al., 1994). At Fernow, WS3 appears to have had a lower Ca concentration prior to treatments compared with WS4, but Ca concentrations have increased with the onset of treatment, largely attributable to N export in this highly N-enriched forested ecosystem.

As expected, both NO_3 and SO_4 are highly and significantly correlated with Ca leaching in these watersheds based on correlation analyses of the

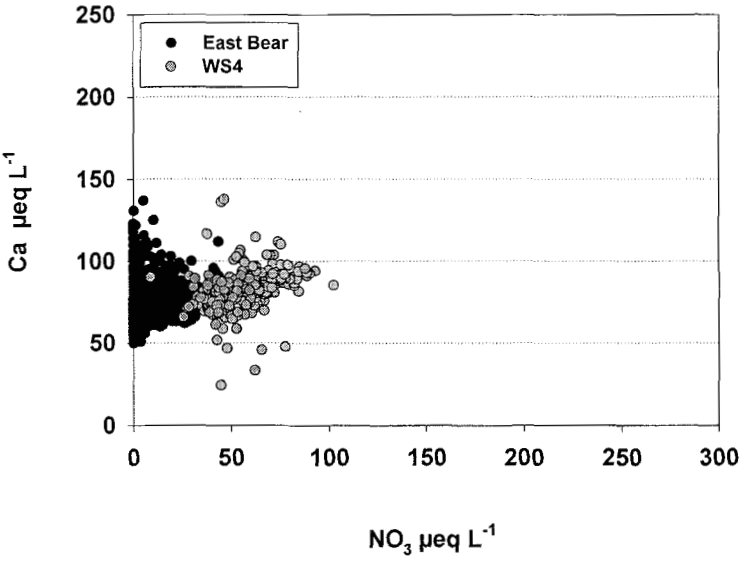
stream data presented here. At BBWM, simple correlation coefficients suggest that both of these strong mineral acid anions are governing Ca export. The correlation with Ca is stronger for SO_4 ($r = 0.83$) than for NO_3 ($r = 0.65$) in West Bear, but NO_3 is dramatically less important to Ca export in East Bear ($r = 0.59$ and $r = 0.17$ for Ca with SO_4 and NO_3, respectively). At Fernow, strong soil adsorption of SO_4 minimizes the effects of SO_4 on cation export but at this site even the reference watershed has high background NO_3 concentrations. Therefore, stream-water concentration data show that the correlation with Ca at Fernow in the treated WS3 is weaker for SO_4 ($r = 0.67$) than for NO_3 ($r = 0.91$). Both anions are much more weakly correlated with Ca in the reference WS4 watershed, which were $r = 0.20$ and $r = 0.38$ for SO_4 and NO_3, respectively.

Fig. 9.3 is a scatter plot of the relationships between stream concentrations of Ca and NO_3. Fig. 9.3a shows that a relationship appears to exist between stream concentrations of this cation–anion pair, although only in WS3 at Fernow is NO_3 apparently a dominant factor governing Ca concentrations in streams as inferred by correlations. In both reference watersheds, East Bear and WS4, there is little evidence of a strong relationship between Ca and NO_3, largely because of the lack of NO_3 in East Bear and the limited range of NO_3 concentrations in WS3 despite the elevated baseline NO_3 concentrations. It should also be noted that soil fertility, as indicated by soil exchangeable Ca concentrations, can influence nitrification and subsequent leaching. Willard et al. (1997) studied N mineralization and nitrification in nine mid-Appalachian forested watersheds including WS4 at Fernow. They concluded high soil fertility promoted nitrification due to significant positive correlations between soil exchangeable Ca and NO_3 production.

Table 9.3 and Fig. 9.4 present N mass balances for these watersheds. Table 9.3 shows that untreated East Bear has nearly total retention of ambient and treatment N in the final years of data presented here (exports of $<0.5\,kg\,N\,ha^{-1}\,yr^{-1}$), while treatments have driven West Bear toward a condition similar to the reference watershed WS4 at Fernow with respect to N mass balance. Fernow clearly receives much greater inputs of atmospheric N compared with BBWM (approximately 2.3× using annual precipitation N data in this chapter), and has a greater export of N as a result (Fernow is approximately 4.8× BBWM using annual stream-water export N data in this chapter). Fig. 9.4 shows net N retention (%) for the study period at the treated watersheds. The most striking feature of these data is the relatively high retention of N in all watersheds despite the increasingly N-saturated character of WS3 and, perhaps most recently, West Bear. The mechanisms responsible for this continued net retention of N and the consequences for both forest management and recovery from N saturation are critically important areas for research. Even at WS3, where long-term N saturation processes are well advanced compared with



(a)



(b)

Figure 9.3. Scatter diagram for the relationship between stream NO_3 and Ca concentrations in the (a) treated and (b) reference watersheds at BBWM and Fernow.

BBWM, most N is retained by the ecosystem. As aboveground biota attain and surpass conditions of biological N limitation, it is logical that soil sequestration is largely responsible for the persistent N retention. Yet

Table 9.3. Inorganic Nitrogen Import–Export Budget for BBWM and Fernow from 1988 to 1995 ($kg\,ha^{-1}\,yr^{-1}$)

	Wet Deposition (+Treatment)[a]		Stream Export	
	East Bear	West Bear	East Bear	West Bear
1988	2.8	2.8	1.5	1.7
1989	4.5	8.7	2.7	3.1
1990	4.6	29.8	1.5	3.1
1991	3.7	28.9	2.2	7.0
1992	3.6	28.8	0.7	5.5
1993	3.1	28.3	0.4	6.0
1994	3.9	29.1	0.2	5.7
1995	3.8	29.0	0.2	5.8
	WS4	WS3	WS4	WS3
1988	8.0	8.2	4.2	3.5
1989	10.0	45.1	6.3	6.1
1990	9.3	44.4	8.6	10.3
1991	8.3	43.4	3.8	6.6
1992	8.1	43.2	4.3	8.8
1993	8.9	44.0	6.3	13.7
1994	10.3	45.4	3.7	7.8
1995	11.2	46.5	5.4	14.6

[a] Wet deposition is the sum of precipitation inputs of NH_4-N plus NO_3-N for the year in the reference watersheds (East Bear, WS4), and the sum of precipitation inputs plus N amendments in the treated watersheds (West Bear, WS3).

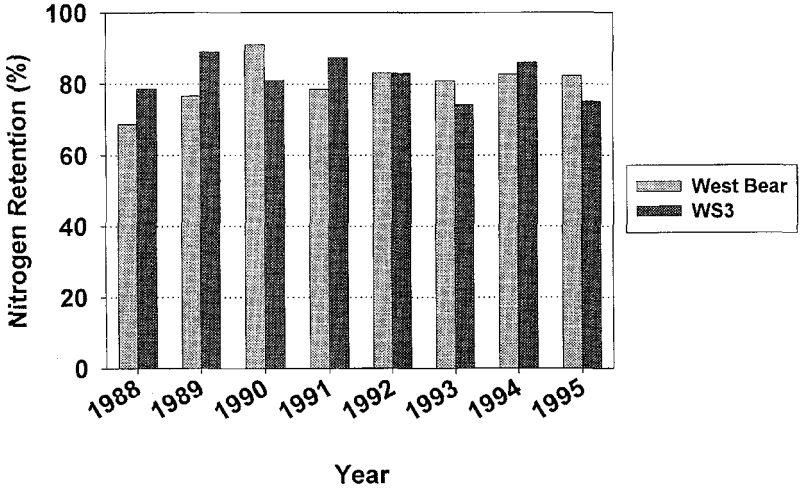


Figure 9.4. Nitrogen retention between 1988 and 1995 for BBWM and Fernow shown as a percentage of total calculated N input retained in the treated watersheds at BBWM and Fernow.

the mechanisms of this phenomena and our ability to define and predict both response and recovery remain incomplete at best.

An important question that has been raised is whether we really understand N mass balances given the difficulties in measuring all phases of N deposition, and the paucity of data on organic N export in streams. Certainly there are weaknesses in the current data from the literature. During the field seasons of 1996 and 1997, a pilot study was conducted that sampled streams from both BBWM and Fernow six times to gather preliminary data on the importance of other forms of N export besides inorganic NH_4 and NO_3. Fig. 9.5 shows the mean concentrations for those six collections, that included dissolved NH_4 and NO_3, NH_4 adsorbed on particulates, and total organic nitrogen on the unfiltered stream samples (includes dissolved and particulate forms of organic N). While these are only a few samples from a pilot study, they suggest that the majority of the stream export response in the treated watersheds at both BBWM and Fernow is in the form of NO_3. Even though the treatment is in the form of NH_4, no evidence of increased NH_4 export was found in the data. This reflects the ability of soil colloids to adsorb the NH_4 cation, and the subsequent nitrification potential of these soils. At Fernow, in both the treated and reference streams, NO_3 accounted for >96% of the N in streams. At BBWM the data were more variable, with organic N accounting for an average of 64% of the total stream N export in East Bear. It is important to note that this high percentage is of a very low concentration since in East Bear almost no N is leaving the ecosystem. In West Bear, these data averaged only 27% organic N with the majority of the treatment response expressed as increased NO_3 concentrations. These preliminary data suggest that organic forms of N

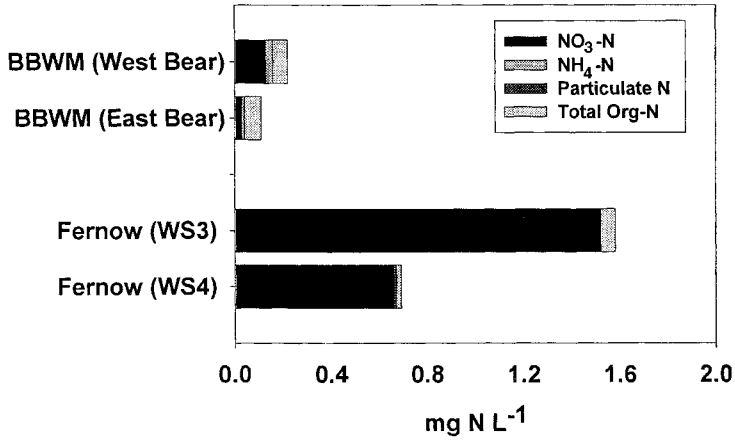


Figure 9.5. Means for the concentrations of NO_3, NH_4, particulate NH_4, and organic N from 1996 and 1997 at BBWM and Fernow ($n = 6$).

can be relatively important, particularly where N export is low. However, most of the response to N amendments in these experimentally manipulated watersheds during the initial years of treatment was clearly expressed as NO_3 increases in streams.

The Role of Soils and Vegetation

The focus of research at both BBWM and Fernow has been on maintaining a schedule of treatments to the watersheds, and tracking the integrated response of these treatments through stream-water monitoring. A number of studies are now in progress designed to evaluate the response of these watersheds to treatments that will provide insight on ecosystem response mechanisms. Typically, the first step in research at the ecosystem-level dealing with biogeochemical information needs is to treat the ecosystem as a "black box" and define inputs and outputs. Once the basic input–output budgets are defined, we then investigate what is taking place "inside the box."

One question about the fate of N in these ecosystems focuses on the role of forest vegetation and whether increased growth or N concentrations can explain any of the net N retention we see in these watersheds. We have initial insights from several studies that suggest that vegetative uptake likely plays a role in the N retention mechanism at both sites. At BBWM, White et al. (1999) showed that the treated West Bear watershed had significantly greater foliar concentrations of N in four of the major tree species, but there was no evidence of significant radial growth increases after four years of treatment in these species. They also reported declines in foliar Ca and increases in foliar Al concentrations on the treated watersheds. Similarly, Gillam et al. (1995) and Adams et al. (1993) reported increased concentrations of foliar N and declines in foliar Ca and Mg for major tree species and understory foliage concentrations at Fernow, but no change in diameter growth. Gilliam et al. (1995) also reported no significant effects on foliar phosphorus (P) concentrations that could be attributed to treatments. Dewalle et al. (1999) studied tree ring cation concentrations at both the West Virginia and Maine experimental watershed study sites. They included in their study a second watershed also treated with $(NH_4)_2SO_4$ in West Virginia (Clover Run), along with the Maine site. They reported significant decreases in Ca and/or Mg for all species in both treated West Virginia watersheds when compared with the reference watersheds, along with significant increases in manganese (Mn). A similar trend was reported for the Maine site, but the base cation declines were not statistically significant at the confidence level they chose. They suggested that sapwood Ca/Mn or Mg/Mn molar ratios were better indices of soil acidification than Ca/Al.

Weber and Wiersma (1997) studied the chemical composition of mosses at BBWM and reported significantly higher concentrations of N in the

treated watershed in *Bazzania trilobata* and *Dicranum fulvum*, with significantly lower concentrations of Ca, Mg, potassium (K) and some trace elements, and found no treatment effects on tissue P concentrations. Magill et al. (1996) reported increases in foliar N in hardwood stands in a plot experiment adjacent to the BBWM watershed that was subjected to wet N treatments. They also reported no net increase in increment growth, but continuously increasing foliar N concentrations with time due to treatments. The limited data to date on these patterns suggest that some sequestration of increased N inputs is possibly attributable to aboveground vegetative uptake, but the studies have not been carried out long enough to see how these differences may result in growth changes by stand or species. There has been no quantification to date at either site of total aboveground biomass differences between treated and reference watersheds that would define whether changes were taking place in minor stand components or understory species.

The other ecosystem compartment that plays a critical role in N accumulation and transformation is the soil. Limited intensive soils research has been conducted to date in these watersheds, although this is the focus of current studies. As yet, there appears to be no evidence for major shifts in forest floor total C and N pools or C/N ratios attributable to the treatments. Both the Maine and West Virginia sites show a significant increase in N mineralization and nitrification in the forest floor in response to elevated N (Wang and Fernandez, 1998; Gilliam et al., 1995). High rates of N mineralization were reported at Fernow in both WS3 and WS4 (6.7 and 7.7 $g\,m^{-2}\,yr^{-1}$, respectively) with between 90 to over 100% of this N release accounted for in net nitrification measurements. No comparable studies have been completed at BBWM, but data reported by Wang and Fernandez (1998) suggests much less nitrification in the BBWM ecosystems. Where increases in N mineralization or nitrification potentials in the forest floor due to treatments occurred at BBWM, they were largely confined to hardwood stands in the watershed, with little response in the softwoods. The influence of vegetation type, particularly the contrast between hardwood and softwood stands due to the influence on litter quality and, therefore, soil properties, should be further investigated and is likely critical in considering the relevance of these studies to landscapes.

Soil solutions at both BBWM and Fernow show differences due to treatments consistent with the changes in stream-water chemistry over time. Adams et al. (1997) described the treatment response in soil solutions at Fernow, with elevated NO_3 concentrations in WS3 soil solutions in the A horizon, diminishing with depth in the soil. They reported that both SO_4 and NO_3 contributed to increased Ca leaching losses, with little influence on Mg. Fernandez et al. (1999) also reported similar patterns in soil solutions of West Bear, but these changes were largely confined to the hardwood stands within the West Bear watershed.

They, too, reported increased leaching of Ca along with increased Al in soil solutions as a result of the treatments.

The Reversibility Question

The phenomenon referred to as N saturation, or the chronic effects of elevated atmospheric N deposition on forested ecosystems, has the benefit of drawing on a vast scientific literature of N in forested ecosystems either from the forest fertilization or nutrient cycling literature, or more broadly from studies in the ecological and agricultural sciences. What we do not have is a robust literature on the consequences of long-term, chronic N amendments to humid, temperate climate forested ecosystems. More importantly, we have little information on how these ecosystems respond to decreased N inputs following long-term elevated N exposures. Indeed, there is much we do not know, nor can adequately predict, about how forest ecosystems "recover" from N saturation. There is ample evidence in the literature to suggest that soil export of NO_3 in soil solutions rapidly declines following short-term experimental applications of N, such as reported by Rustad et al. (1993, 1996). However, data reported here on BBWM and Fernow show that the majority of N deposition even in N "saturated" ecosystems is retained. Although a significant amount of the retained N could be sequestered in an aggrading forest, a major proportion must be retained in the soil. Therefore, while the symptoms of N saturation may be quickly ameliorated by reduced inputs as judged by the geochemical signal in surface waters, we expect that the forest ecosystem will take much longer to recover, on the order of decades to centuries. Indeed, even the concept of "recovery" from N accumulations could be debated as to meaning and demonstration. Stand growth and composition, litter quality, and subsequent rates of decomposition and nutrient cycling, and short- and long-term soil pools of N are likely to respond slowly to major shifts in N inputs, with many of the controlling mechanisms in this context poorly understood.

Conclusions

The BBWM and Fernow whole-watershed experimental N enrichment studies provide a unique opportunity to study, in situ, the mechanisms governing forest ecosystem response to elevated N deposition and N saturation phenomena. Both sites are paired watershed studies offering a powerful tool to examine long-term changes in ecosystem structure and function, particularly with respect to the biogeochemistry of N. Our understanding of forest ecosystem response to N allows us to predict some changes in these watersheds, but our knowledge is lacking on mechanisms

governing these responses and on the temporal development of the changes expected. Both sites together offer an interesting spectrum of forested stream watersheds ranging from little or no inorganic N export and modest deposition, to chronically elevated N export and rates of N inputs that rival some of the high N deposition European landscapes. Yet in all cases a majority of the added N is retained by the watersheds.

On the other hand, even in the reference watershed in Maine where no N saturation exists and tree growth remains N-limited, small increases in N inputs had almost immediate consequences expressed as increased stream export of NO_3. We suggest that these sites become more valuable each year because of their record of treatment and response, but neither of the treated watersheds has yet attained a new equilibrium with the elevated N inputs from treatments. Some have suggested they never will, others believe a new equilibrium can be attained in a practical time frame. Research on the biological and geochemical mechanisms controlling these response characteristics, and how they will respond in the future to potential changes in N deposition, should be a high priority for the national research agenda and contribute to our basic understanding of ecosystem function regardless of current policy issues. This becomes even more important in a changing climate that couples warming-induced changes in the rates of biogeochemical cycling with chronic exposure to elevated N deposition.

References

Aber JD, Driscoll CT (1997) Effects of land use, climate variation, and N deposition on N cycling and C storage in northern hardwood forests. Global Biogeochem Cycl 1:639–648.

Aber J, McDowell W, Nadelhoffer K, Magill A, Berntson G, Kamakea M, McNulty S, Currie W, Rustad L, Fernandez I (1998) Nitrogen saturation in temperate forest ecosystems—hypothesis revisited. BioScience 48:921–943.

Aber J (1992) Nitrogen cycling and nitrogen saturation in temperate forest ecosystems. Tree 7:220–223.

Aber JD, Nadelhoffer KJ, Steudler P, Melillo JM (1989) Nitrogen saturation in northern forest ecosystems. BioScience 39:378–386.

Adams MB, Angradi TR (1996) Decomposition and nutrient dynamics of hardwood leaf litter in the Fernow Whole-Watershed Acidification Study. For Ecol Manage 83:61–69.

Adams MB, Angradi TR, Kochenderfer JN (1997) Stream water and soil solution responses to 5 years of nitrogen and sulfur additions at the Fernow Experimental Forest, West Virginia. For Ecol Manage 95:79–91.

Adams MB, Edwards PJ, Wood F, Kochenderfer JN (1993) Artificial watershed acidification on the Fernow Experimental Forest, USA. J Hydrol 150:505–519.

Adams MB, Kochenderfer JN, Wood F, Angradi TR, Edwards PJ (1994) *Forty Years of Hydrometeorological Data from the Fernow Experimental Forest, West Virginia.* Gen Tech Rep NE-184. United States Department of Agriculture (USDA) Forest Service, Northeastern Forest Experiment Station, Radnor, PA.

Ågren GI, Bosatta E (1988) Nitrogen saturation of terrestrial ecosystems. Environ Pollut 54:185–197.

Bailey RG, Avers PE, King T, McNabb WH (1994) *Ecoregions and Subregions of the United States*. United States Department of Agriculture (USDA) Forest Service, Washington, DC.

Bull KR (1991) The critical loads/levels approach to gaseous pollutant emission control. Environ Pollut 69:105–123.

Carnol M, Ineson P, Dickinson AL (1997) Soil solution nitrogen and cations influenced by $(NH_4)_2SO_4$ deposition in a coniferous forest. Environ Pollut 97:1–10.

DeWalle DR, Tepp JS, Swistock BR, Sharpe WE, Edwards PJ (1999) Tree-ring cation response to experimental watershed acidification in West Virginia and Maine. J Environ Qual 28:299–309.

Dise NB, Wright RF (1992) *The NITREX Project (Nitrogen Saturation Experiments). Ecosystem Research Report 2*. Commission of European Communities, Brussels, Belgium.

Dise NB, Wright RF (1995) Nitrogen leaching from European forests in relation to nitrogen deposition. For Ecol Manage 71:153–161.

Driscoll CT, Van Dreason R (1993) Seasonal and long-term temporal patterns in the chemistry of Adirondack lakes. Water Air Soil Pollut 67:319–344.

Fenn ME, Poth MA, Johnson DW (1996) Evidence for nitrogen saturation in the San Bernardino Mountains in southern California. For Ecol Manage 82: 211–230.

Fernandez IJ, Rustad LE, David MB, Nadelhoffer KJ, Mitchell MJ (1999) Mineral soil and solution responses to experimental N and S enrichment at the Bear Brook Watershed in Maine (BBWM). Environ Monit Assess 55: 165–185.

Galloway JN (1998) The global nitrogen cycle: changes and consequences. Environ Pollut 102:15–24.

Gilliam FS, Adams MB, Yurish BM (1995) Ecosystem nutrient responses to chronic nitrogen inputs at Fernow Experimental Forest, West Virginia. Can J For Res 26:196–205.

Jeffries RL, Maron JL (1997) The embarrassment of riches: atmospheric deposition of nitrogen and community and ecosystem processes. Tree 12:74–77.

Johnson D (1992) Nitrogen retention in forest soils. J Environ Qual 21:1–12.

Kahl SJ, Norton SA, Fernandez IJ, Nadelhoffer KJ, Driscoll CT, Aber JD (1993) Experimental inducement of nitrogen saturation at the watershed scale. Environ Sci Tech 27:565–568.

Lawrence GB, David MB, Shortle WC (1995) A new mechanism for calcium loss in forest-floor soils. Nature 378:162–165.

Lawrence GB, David MB, Vailey SW, Shortle WC (1997) Assessment of calcium status in soils of red spruce forests in the northeastern United States. Biogeochemistry 38:19–39.

Likens GE, Driscoll CT, Busco DC (1996) Long-term effects of acid rain: response and recovery of a forest ecosystem. Science 272:244–246.

Løkke H, Bak J, Falkengren-Grerup U, Finlay RD, Ilvesniemi H, Nygaard PH, Starr M (1996) Critical loads of acidic deposition for forest soils: Is the current approach adequate? Ambio 25:510–516.

Magill AH, Downs MR, Nadelhoffer KJ, Hallet RA, Aber JD (1996) Forest ecosystem reponse to four years of chronic nitrate and sulfate additions at Bear Brooks Watershed, Maine, USA. For Ecol Manage 84:29–37.

McNulty S, Aber JD (1993) Effects of chronic nitrogen additions on nitrogen cycling in a high-elevation spruce–fir stand. Can J For Res 23:1252–1263.

McNulty S, Aber JD, McLellan TM, Katt SM (1990) Nitrogen cycling in high elevation forests of the northeastern US in relation to nitrogen deposition. Ambio 19:30–40.

McNulty S, Aber JD, Newman SD (1996) Nitrogen saturation in a high elevation New England spruce–fir stand. For Ecol Manage 84:109–121.

Mitchell MJ, Driscoll CT, Kahl JS, Likens GE, Murdoch PS, Pardo H (1996) Climatic control on nitrate loss from forested watersheds in the northeast United States. Environ Sci Technol 30:2609–2612.

Nadelhoffer K, Downs M, Fry B, Aber J (1999) Controls on N retention and exports in a forested watershed. Environ Monit Assess 55:187–210.

Nadelhoffer KJ et al. (1999) Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. Nature 398:145–148.

NADP (1990) National Atmospheric Deposition Program (IR-7)/National Trends Network: 1990. Annual Report: NADP/NTN Coordination Office. Natural Resource Ecology Laboratory, Colorado State University, Fort Collins, CO.

Nihlgård B (1985) The ammonium hypothesis—an additional explanation to the forest dieback in Europe. Ambio 14:2–8.

Nilsson J (1986) *Critical Loads for Nitrogen and Sulphur*. Nordsk ministerrånd. Miljørapport 11. Copenhagen, Denmark.

Nilsson J, Grennfelt P (1988) *Critical Loads for Sulphur and Nitrogen. Report from a Workshop Held at Skokloster, Sweden, 19–24 March 1988*. Miljørapport 1988:15, Nordic Council of Ministers, Copenhagen, Denmark.

Norton SA, Fernandez IJ (eds) (1999) *The Bear Brook Watershed in Maine: A Paired Watershed Experiment—The First Decade (1987–1997)*. Kluwer Academic, Boston, MA.

Norton SA, Kahl JS, Fernandez IJ, Rustad LE, Scofield JP, Haines TA (1994) Response of the West Bear Brook Watershed, Maine, USA, to the addition of $(NH_4)_2SO_4$: 3-year results. For Ecol Manage 68:61–73.

Peterjohn WT, Adams MB, Gilliam FS (1996) Symptoms of nitrogen saturation in two central Appalachian hardwood forest ecosystems. Biogeochemistry 35: 507–522.

Rustad LE, Fernandez IJ, David MB, Mitchell MJ, Nadelhoffer KJ, Fuller RB (1996) Experimental soil acidification and recovery at the Bear Brook Watershed in Maine. Soil Sci Soc Am J 60:1933–1943.

Rustad LE, Fernandez IJ, Fuller RB, David MB, Nodvin SC, Halteman WA (1993) Soil solution response to acidic deposition in a northern hardwood forest. Agric Ecosyst Environ 47:117–134.

Rustad LE, Kahl JS, Norton SA, Fernandez IJ (1994) Underestimation of dry deposition by throughfall in mixed northern hardwood forests. J Hydrol 162:319–336.

Shortle WC, Smith WC (1988) Aluminum-induced calcium deficiency syndrome in declining red spruce. Science 240:1017–1018.

Shortle WC, Smith WC, Minocha R, Alexeyev VA (1995) Similar patterns of change in stemwood calcium concentrations in red spruce and Siberian fir. J Biogeogr 22:467–473.

Stoddard JL (1994) Long-term changes in watershed retention of nitrogen: its causes and aquatic consequences. In: Baker LA (ed) *Environmental Chemistry of Lakes and Reservoirs*. American Chemical Society, Washington, DC, pp 223–284.

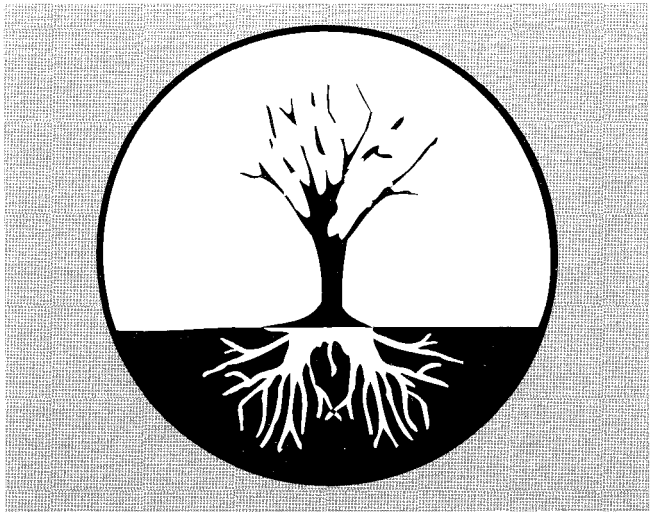
Sullivan TJ, Eilers JM, Cosby BJ, Vaché KB (1997) Increasing role of nitrogen in the acidification of surface waters in the Adirondack Mountains, New York. Water Air Soil Pollut 95:313–336.

Sullivan TJ (1997) Ecosystem manipulation experimentation as a means of testing a biogeochemical model. Environ Manage 21:15–21.
Vitousek PM (1997) Human alteration of the global nitrogen cycle: sources and consequences. Ecol Applic 7:737–750.
Wang Z, Fernandez IJ (1998) Soil type and forest vegetation influences on forest floor nitrogen dynamics in the Bear Brook Watershed in Maine (BBWM). Environ Monit Assess 55:221–234.
Weber KL, Wiersma GB (1997) Trace element concentrations in mosses collected from a treated experimental forest watershed. Toxic Environ Chem 65:17–29.
White GJ, Fernandez IJ, Wiersma GB (1999) Impacts of ammonium sulfate treatment on foliar chemistry of forest trees at the Bear Brooks Watershed in Maine. Environ Monit Assess 55:235–250.
Willard KW, DeWalle DR, Edwards PJ, Schnabel RR (1997) Indicators of nitrate export from forested watersheds of the mid-Appalachians, United States of America. Global Biogeochem Cycl 11:649–656.
Williams MW, Baron JS, Caine N, Sommerfeld R, Sanfrod R Jr. (1996) Nitrogen saturation in the Rocky Mountains. Environ Sci Technol 30(20):640–646.
Wright RF, van Breeman R (1995) The NITREX Project: an introduction. For Ecol Manage 71:1–5.
Wright RF, Rasmussen L (eds) (1998) The whole ecosystem experiments of the NITREX and EXMAN projects. For Ecol Manage Vol. 101.

Ecological Studies 139

Robert A. Mickler
Richard A. Birdsey
John Hom Editors

Responses of Northern U.S. Forests to Environmental Change



Springer