

Nitrate dynamics of forested watersheds: spatial and temporal patterns in North America, Europe and Japan

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Abstract The relationships of nitrogen biogeochemistry are reviewed, focusing on forested watersheds in North America, Europe and Japan. Changes in both local and global nitrogen cycles that affect the structure and function of ecosystems are described. Within northeastern United States and Europe, atmospheric deposition thresholds of ~ 8 and ~ 10 kg N ha⁻¹ year⁻¹, respectively, result in enhanced mobilization of nitrate. High nitrate concentrations and drainage water loss rates up to 22 kg N ha⁻¹ year⁻¹ have also been found near Tokyo. Although atmospheric deposition may explain a substantial portion of the spatial pattern of nitrate in surface waters, other factors also play major roles in affecting the spatial patterns of nitrogen biogeochemistry. Calcium availability influences the composition of the vegetation and the biogeochemistry of nitrogen. The abundance of sugar maple is directly linked to soil organic matter characteristics and high rates of nitrogen mineralization and nitrification. Seasonal patterns of nitrate concentration and drainage water losses are closely coupled with differences in seasonal temperature and hydrological regimes. Snow-dominated forested catchments have highest nitrate losses during snowmelt. Watersheds in the main island of Japan (Honshu) with high summer temperatures and precipitation inputs have greatest losses of nitrate occur during the late summer. Understanding future changes in nitrate concentrations in surface waters will require an integrated approach that will evaluate concomitantly the influence of both biotic and abiotic factors on nitrogen biogeochemistry.

Keywords Biogeochemistry · Europe · Japan · Nitrogen · North America

Introduction

The biogeochemical cycle of nitrogen in forested ecosystems includes a wide range of biotic pools and transformations that affect both inorganic and organic nitrogen forms (Fig. 1). Nitrogen is often a limiting nutrient in both terrestrial and aquatic ecosystems (Vitousek and Howarth 1991; Galloway et al. 2008). Increased inputs of fixed nitrogen from anthropogenic sources result in both beneficial and deleterious impacts to ecosystems. The effect of nitrogen on ecosystems has been greatly altered by substantial anthropogenic inputs over the past century, including contributions from nitrogen fertilizer, human waste products and atmospheric pollutants. These anthropogenic nitrogen inputs have greatly augmented the amount of available nitrogen at local, regional and global scales (Galloway and Cowling 2002). An example of an important beneficial aspect of added nitrogen is the stimulation of primary production in those terrestrial ecosystems where enhanced production is sought such as in agronomic systems where enhanced crop yields are needed to maximize food production (Oberle and Keeney 1990; David et al. 1997). However, there has been increasing awareness (Vitousek et al. 1997; Fenn et al. 1998; Aber et al. 2003; Binkley et al. 2004) about the potential deleterious impacts of this additional nitrogen to various ecosystems, but particularly for forested watersheds that may be particularly sensitive to elevated nitrogen inputs. For some forested watersheds, particularly in relatively pristine areas in the western mountains of the United States, deleterious effects associated with unwanted enhanced production and changes

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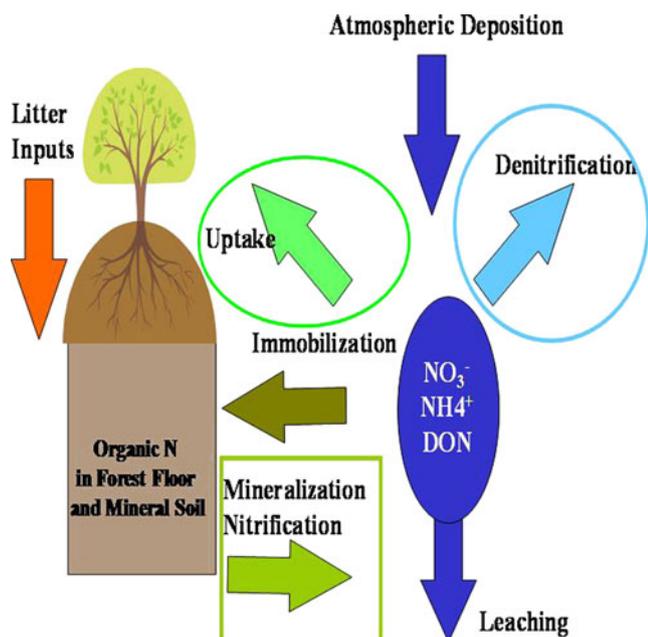


Fig. 1 Simplified diagram of nitrogen biogeochemical pools, fluxes and transformations in a forested ecosystem

in species composition have also been noted (Fenn et al. 2003). In addition, for those areas which are sensitive to acidification due to their geochemical attributes, a major concern with excess nitrogen is the contribution of nitrification in generating excess acidity and the resultant deleterious impacts of this additional acidity on terrestrial and aquatic ecosystem structure and function (Van Miegroet and Cole 1984; Rabalais 2002). The relative importance of atmospheric nitrogen inputs has increased as the atmospheric sulfur inputs have declined in the United States (Fig. 2) and Europe. This enhancement of nitrogen has multifaceted impacts on a wide range of ecosystem processes including acidic deposition, global energy balance and changes in biotic production and community diversity. The integration of the various influences of this added nitrogen has been described as the “nitrogen cascade” (Galloway et al. 2003), which details how an individual nitrogen atom can be transformed and transferred within and among ecosystems resulting in a broad range of biogeochemical and environmental effects.

Linkages of nitrate in surface waters and atmospheric deposition of nitrogen

The linkage between atmospheric nitrogen deposition and responses to surface waters has been shown in North America, Europe and Asia (Japan). In a synthesis of results from the northeastern United States [including New York (Adirondack and Catskill Mountains), Vermont, New

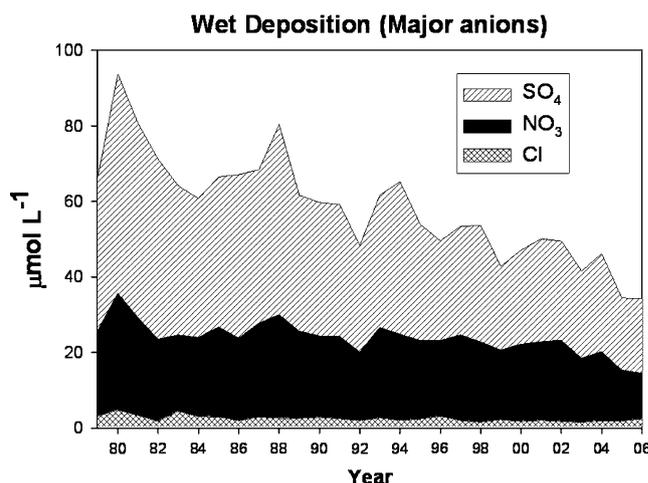


Fig. 2 Changes in solute concentration of major anions (sulfate, nitrate and chloride) at the Huntington Forest in the Adirondack Mountains of New York State (USA). Over the period of analyses, the relative proportion of nitrate is increasing. Adapted from Mitchell et al. (2009)

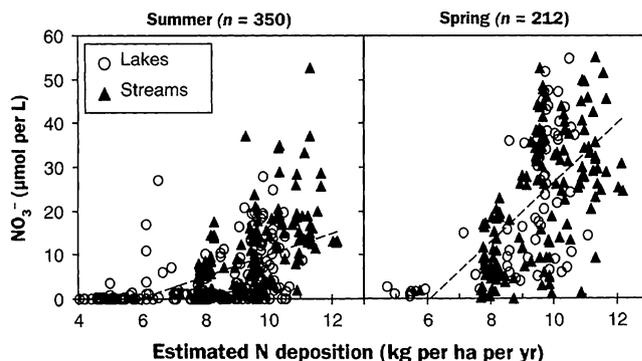


Fig. 3 Influence of atmospheric deposition on surface water nitrate concentrations in the northeastern United States. From Aber et al. (2003)

Hampshire and Maine], Aber et al. (2003) showed that there was a deposition threshold of $\sim 8 \text{ kg N ha}^{-1} \text{ year}^{-1}$ that resulted in significant increases in nitrate concentrations of lakes and streams of these regions (Fig. 3). Using summer or spring surface water concentrations, atmospheric N deposition explained 30 and 38%, respectively, of the spatial variation in surface water nitrate concentrations in this region. Studies in Europe of forested ecosystems have suggested somewhat different thresholds. Below a deposition threshold of $\sim 10 \text{ kg N ha}^{-1} \text{ year}^{-1}$, there was no substantial nitrogen leaching in forests, while at deposition levels of $10\text{--}25 \text{ kg N ha}^{-1} \text{ year}^{-1}$, leaching occurred at some sites. Above $25 \text{ kg N ha}^{-1} \text{ year}^{-1}$, significant leaching occurred at all European sites (Dise and Wright 1995). The influence of atmospheric N deposition on nitrate concentrations has also been shown for Japan with highest atmospheric N inputs and high concentrations of nitrate in surface waters found within the Tokyo region

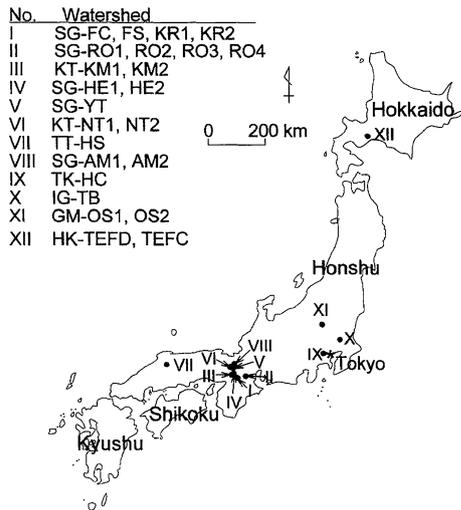


Fig. 4 Location of study watersheds in Japan. From Mitchell et al. (1997)

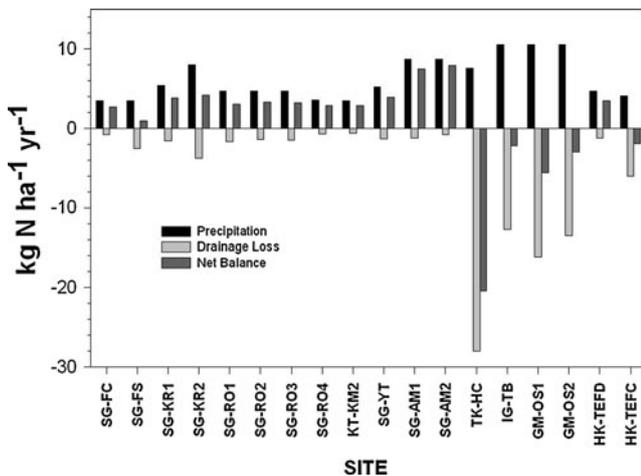


Fig. 5 Nitrogen mass balances in watersheds in Japan. From Mitchell et al. (1997)

with its high levels of anthropogenic sources of nitrogen (Ohruai and Mitchell 1997). These elevated nitrate concentrations result in relatively large (up to 22 kg N ha⁻¹ year⁻¹) drainage water losses of inorganic nitrogen (Figs. 4 and 5) (Mitchell et al. 1997).

Although clear linkages have been made between atmospheric nitrogen deposition and nitrate concentrations and export in surface water, atmospheric deposition explains only about 30–38% of the spatial variation in surface water nitrate concentrations (Aber et al. 2003). For watersheds in the northeastern United States with similar atmospheric nitrogen inputs, there is substantial variation in nitrate drainage losses (Fig. 6). In an analysis of factors affecting nitrogen leaching in soils in Europe, Dise et al. (2009) found that inorganic nitrogen in deposition, mean annual temperature, mean altitude, site drainage (plot vs.

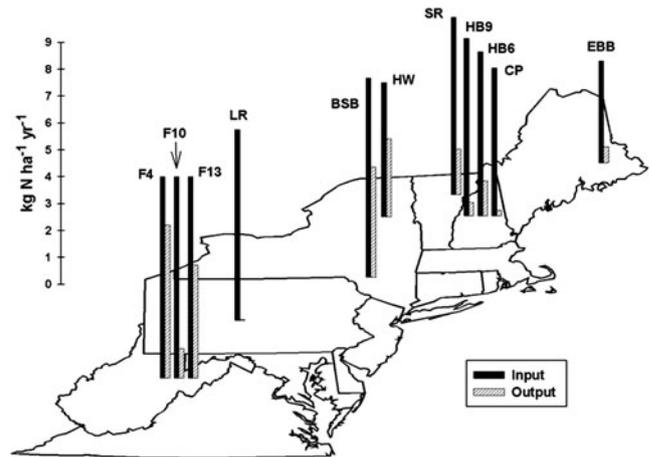


Fig. 6 Nitrogen mass balances in forested watersheds in the northeast United States. From Campbell et al. (2004) with kind permission from Springer Science+Business Media B.V.

catchment), needle- and litter-N concentration, organic horizon C:N ratio, and subsoil pH were all statistically significant. These results suggest the complexities of the biogeochemical cycling of nitrogen in forest ecosystems where atmospheric influences, biotic (both vegetation and soil microbes) and a variety of watershed attributes all influence the dynamics of nitrogen.

Relationships between geology, soils, vegetation and nitrogen biogeochemistry

A key factor with respect to nitrogen cycling is linked with the role of vegetation. For example, Mitchell et al. (1992) found that, in a comparison of a site in southeastern Canada (Turkey Lakes) with a site (Huntington Forest) in the Adirondack Mountains of the northeast United States, the former site had much higher soil nitrate concentrations. This difference was attributed to the predominance of sugar maple and an absence of American beech at Turkey Lakes. The importance of sugar maple in forest stands as a generator of nitrate was also further confirmed by a comparison of sites across the northeast United States that showed that the proportion of sugar maple in a forest stand was an excellent predictor of nitrate concentration in soils (Fig. 7) (Lovett and Mitchell 2004). The role of vegetation in affecting nitrate concentrations was further explored by Lovett et al. (2002) in an examination of the relationships between vegetation, soil characteristics and surface water nitrate concentrations in the Catskill Mountains of New York State (Fig. 8). There was a strong positive relationship between nitrate concentration in those watersheds with a high proportion of sugar maple and a strong negative relationship between surface water nitrate concentration

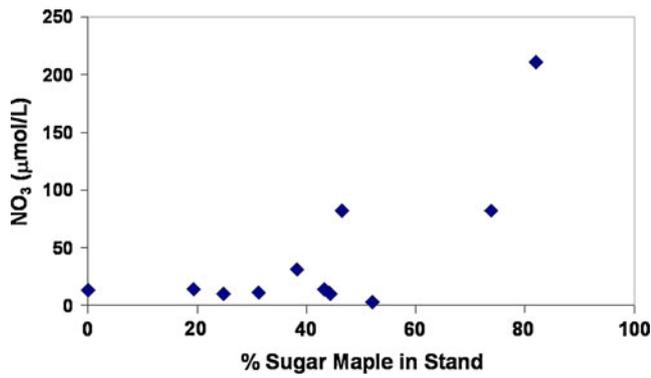


Fig. 7 Effect of sugar maple abundance on soil nitrate concentrations in forest stands in the northeastern United States. From Lovett and Mitchell (2004)

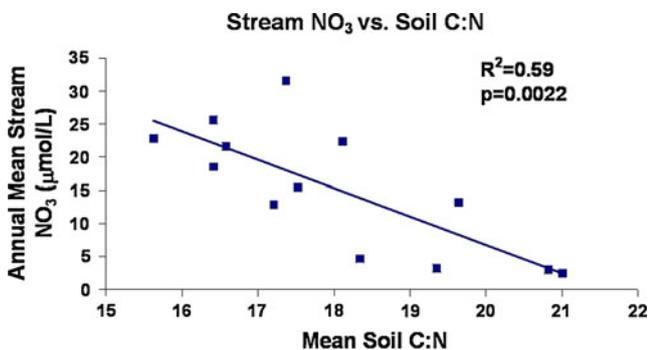


Fig. 8 Effect of C:N soil ratios on nitrate concentrations in streams of the Catskill Mountains of New York State. The variance in C:N ratios is primarily explained by the relative abundance of sugar maple (low C:N) and red oak (high C:N) within these watersheds. From Lovett et al. (2002)

and the proportion of red oak. These relationships confirm previous studies that have shown how litter quality affects soil organic matter quality with respect to nitrogen mineralization and nitrate generation (Melillo et al. 1982). Leaf litter generated from sugar maple has a relatively low C:N ratio and low lignin concentration compared to leaf litter from American beech. These differences in litter quality affect soil organic matter with sugar maple stands having lower soil C:N ratios than soils associated with American beech (Lovett et al. 2002).

There is also a linkage between geology and soils and the vegetation composition of forested watersheds. Christopher et al. (2006) found dramatic differences in nitrate concentration for two nearly adjacent watersheds in the Adirondack Mountains of New York State that were similar in all attributes except calcium availability and vegetation composition (Fig. 9). The watershed with higher nitrate concentration had a higher proportion of sugar maple and American basswood and a lower proportion of American beech. Further analyses by Page and Mitchell (2008a, b) found that there was a strong relationship between soil

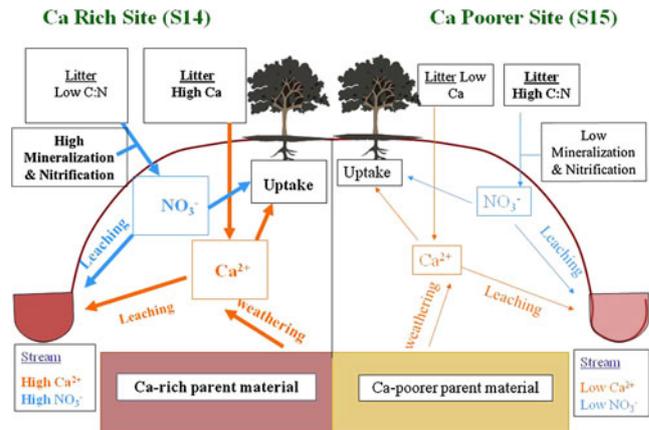


Fig. 9 Relationships among calcium availability, vegetation response and microbial processes in generating different levels of soil and surface water nitrate in the Adirondack Mountains of New York State. From Christopher et al. (2006)

nitrate levels and the presence of calciphilic trees (e.g., sugar maple and American basswood) throughout the Adirondack Mountains. The importance of calcium in affecting forest ecosystems, including the decline of sugar maple, has been clearly shown in a variety of studies (e.g., Schaberg et al. 2006; Long et al. 2009). It has been suggested that acidic deposition, especially in the northeast United States, has resulted in a depletion of soil calcium in some sites (Shortle and Bondietti 1992; Bailey et al. 2005). To evaluate the response of a forest watershed to calcium availability, an experimental calcium silicate (wollastonite, CaSiO₃) addition was done at Watershed 1 at the Hubbard Brook Experimental Forest in the White Mountains of the State of New Hampshire (Juice et al. 2006). Foliar calcium concentration of canopy sugar maples increased markedly after the experimental addition. Crown condition of sugar maple was much healthier in the treated watershed compared with the reference watershed. Following calcium addition, there was also a marked increase in sugar maple seed production and a concomitant increase in sugar maple density and survivorship. Further analyses are needed in evaluating how soil calcium depletion is affecting nitrogen biogeochemistry both with respect to temporal and spatial patterns.

Seasonal patterns in nitrate concentrations in surface waters

The seasonal patterns of nitrate concentration in surface water have been evaluated for sites in North America, Europe and Japan. For those sites with substantial amounts of snow pack (e.g., southeast Canada, northeast United States, northern Europe and Hokkaido Japan), most nitrate lost through drainage waters is associated with snowmelt, with lowest concentrations of nitrate generally associated

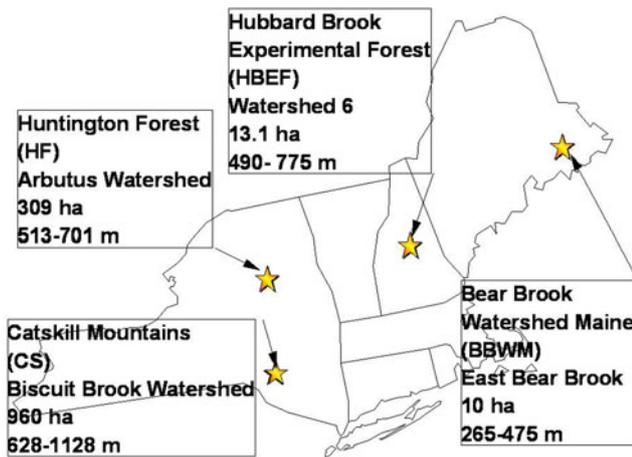


Fig. 10 Location and short site description of four watersheds in the northeastern United States used for evaluating seasonal and annual patterns of surface water nitrate concentrations. From Mitchell et al. (1996)

with the summer period when biotic demand for nitrogen is highest (Figs. 10, 11 and 12) (Stoddard 1994; Mitchell et al. 1996; Christopher et al. 2008). Stable isotopic analysis of nitrate has established that the vast majority of the nitrate is derived from microbially-mediated nitrogen mineralization and nitrification in the soil (Kendall 1998; Piatek et al. 2005) (Fig. 13). During some events, however, there may be some limited direct transfer of atmospherically deposited nitrate to surface waters (Sebestyen et al. 2008). There are important regional differences in the timing of highest nitrate concentration. For warmer regions in the southeast United States (e.g., Coweta, North Carolina) and the main island (Honshu) of Japan, the peak in nitrate concentrations occurs in the midsummer which coincides with the period of highest temperature and maximum biotic activity (Ohruai and Mitchell 1997; Ohte et al. 2001). In Japan, these high summer nitrate concentrations also coincide with the highest discharge rates that are a direct reflection of the high precipitation inputs of the summer. These high discharge rates are in marked contrast to most sites in the United States and Europe where the summer period is associated with low discharge due to precipitation patterns and high evapotranspiration (Stoddard 1994; Mitchell et al. 1996; Mitchell 2001). Future studies that evaluate how these geographical differences in nitrogen biogeochemistry should provide insight into how various factors including climate change may be important.

Long-term changes in nitrate concentrations in surface waters

There has been considerable conjecture on the cause for long-term changes in nitrate concentrations in forested

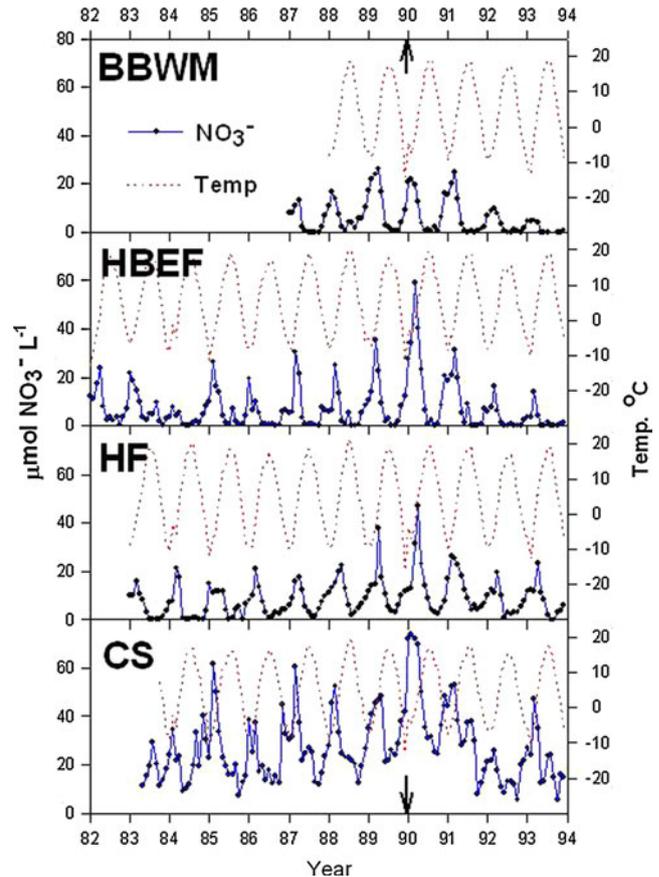


Fig. 11 Temporal changes in nitrate concentrations and above ground temperature for four watersheds in the northeastern United States. Arrows indicate a month with extremely cold temperatures and low snow cover that is followed by high nitrate concentrations during snowmelt. Dotted line temperature and solid line nitrate concentrations. From Mitchell et al. (1996)

watersheds. The “nitrogen saturation” hypothesis would suggest that, over time, the concentration of nitrate in surface waters should increase especially as nitrogen biotic demand is reached (Aber et al. 1989; Stoddard 1994), but long-term monitoring studies of surface waters in the northeast United States have not always found such an increase and in some cases there have been decreases in concentration (e.g., Goodale et al. 2003). The causes for these long-term temporal patterns have not been fully ascertained (Aber et al. 1991; Mitchell et al. 1996; Groffman et al. 2009). However, the coincidence of annual patterns among surface waters within a region suggest some type of temporal synchronization such as that associated with climate (Goodale et al. 2003). A climatic factor that has been shown to have a marked impact on nitrate loss from forests is the effect of soil frost (Groffman et al. 2001; Fitzhugh et al. 2003). Mitchell et al. (1996) suggested that a major increase in nitrate concentration in four watersheds in the northeast United States was preceded by

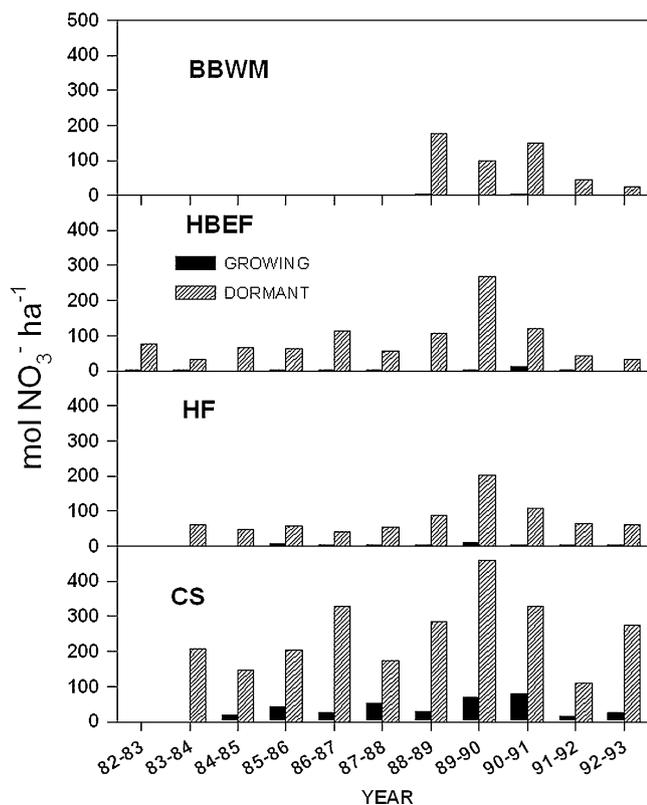


Fig. 12 Nitrate losses in four watersheds in the northeastern United States showing the importance of the winter/snow melt period as the time of maximum nitrate drainage water losses. Most (>87%) of the nitrate in these watersheds is lost during the dormant season (1 October–30 May). From Mitchell et al. (1996)

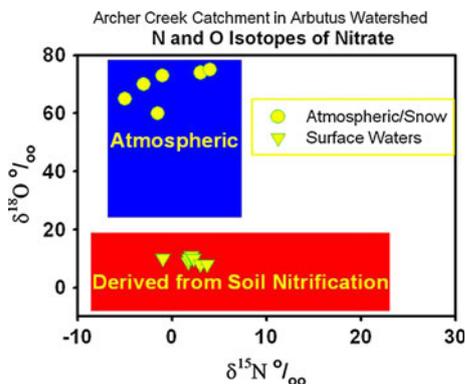


Fig. 13 Use of the stable isotopes of oxygen and nitrogen to evaluate nitrate sources in surface waters of the Archer Creek Catchment of the Arbutus Watershed in the Adirondack Mountains of New York State. From Piatek et al. (2005)

an unusually cold period with little snow cover that would have likely resulted in substantial soil freezing (Fig. 11). Park et al. (2003), working at the Archer Creek watershed in the Adirondacks, showed the close coupling nitrate drainage losses and above ground temperature affecting the snowmelt (Fig. 14). A close correspondence was found

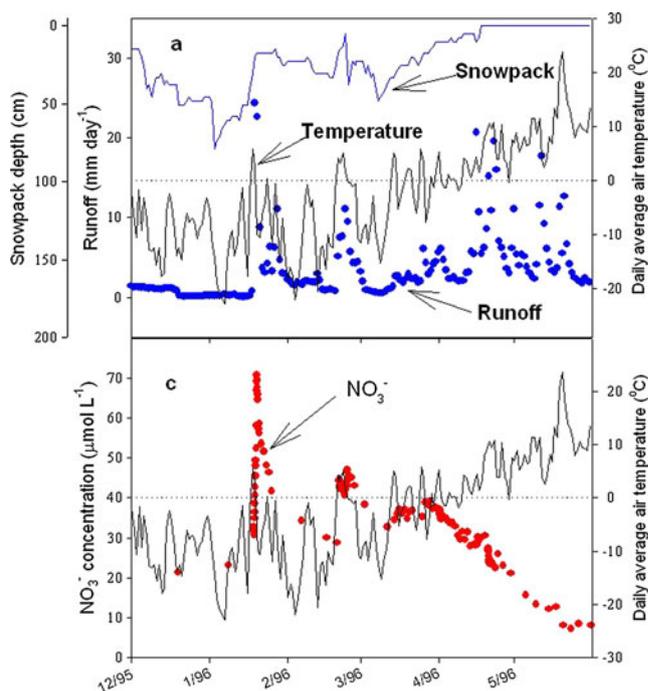


Fig. 14 Responses of snowpack depths, runoff amounts, and nitrate concentrations in drainage water from the Archer Creek catchment to temperature changes from December 1995 to May 1996. From Park et al. (2003)

between above ground temperature (especially during the winter) and the variation in nitrate losses among years (Fig. 15). However, further studies are needed to more fully ascertain what are the actual climatic drivers that are affecting the long-term temporal patterns in surface water nitrate concentrations (Fitzhugh et al. 2003; Goodale et al. 2003).

Conclusions

Nitrogen is a critical nutrient and has a major effect on the structure and functioning of terrestrial and aquatic ecosystems. Increased inputs of anthropogenic fixed nitrogen have had major impacts on nitrogen biogeochemistry at local, regional and global scales. Particularly attention has focused on forested watersheds some of which are particularly sensitive to elevated nitrogen inputs. Linkage between atmospheric nitrogen deposition and responses to surface waters has been shown in North America, Europe and Asia (Japan). Above a threshold of atmospheric nitrogen inputs, both increased concentrations of nitrate in surface waters and enhanced inorganic nitrogen inputs are observed. However, other factors affect the temporal and spatial distribution of nitrate concentrations and losses among forested ecosystems. Important factors besides atmospheric deposition include vegetation and soil characteristics. Studies in the

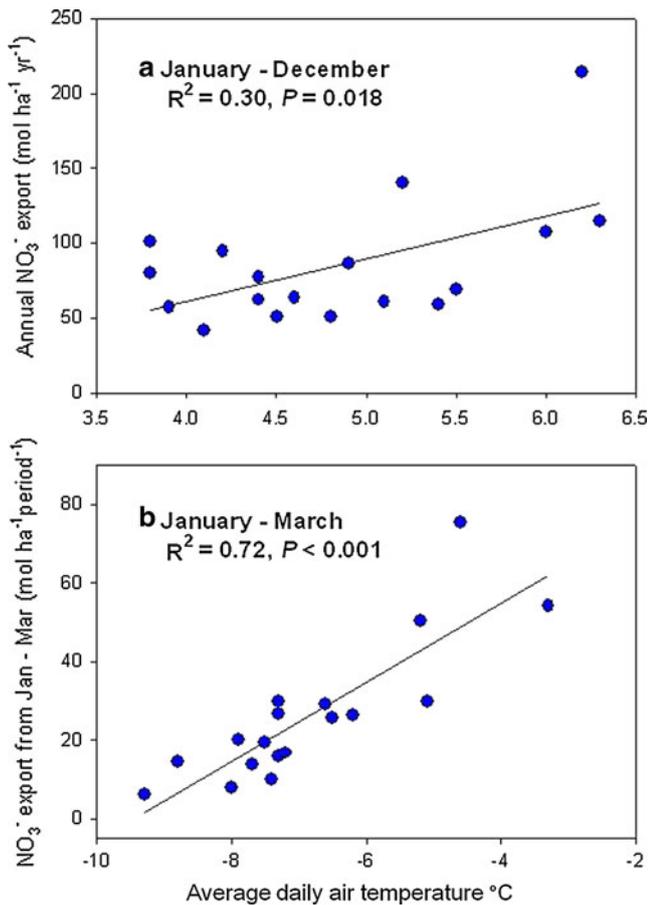


Fig. 15 Relationships between average air temperatures and nitrate export in Arbutus Lake outlet (Adirondack Mountains of New York State) for either the entire year or the months preceding spring snowmelt from 1984 to 2001. Winter air temperature is a major driver of nitrate export. From Park et al. (2003)

northeast United States have shown important relationships between geological characteristics (especially calcium availability), vegetation, and nitrate in soils and surface waters. There are strong seasonal patterns in surface water nitrate concentrations and drainage losses. For snow-dominated forested catchments, highest levels of nitrate are associated with the snowmelt period during which nitrate generated by soil microbial processes is transferred in drainage waters to streams and lakes. For some sites in the southeast United States and in Japan (especially near the Tokyo metropolitan region), highest concentrations and drainage water losses occur during the latter part of summer, a period of warm temperatures and elevated precipitation. There has been considerable conjecture on factors that influence the long-term annual changes in nitrate concentrations. The relative importance of “nitrogen saturation” versus other factors including climatic factors (e.g., soil freezing, changes in precipitation and temperature regimes) have been evaluated, but the actual regulating factors

affecting these long-term changes have not been fully identified or quantified. Further research is needed on evaluating the interactions between geology, vegetation, soils and climate change in affecting both seasonal and long-term changes in the nitrogen biogeochemistry of forested watersheds.

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