Decreased atmospheric nitrogen deposition in eastern North America: Predicted responses of forest ecosystems☆

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ABSTRACT

Historical increases in emissions and atmospheric deposition of oxidized and reduced nitrogen (N) provided the impetus for extensive, global-scale research investigating the effects of excess N in terrestrial and aquatic ecosystems, with several regions within the Eastern Deciduous Forest of the United States found to be susceptible to negative effects of excess N. The Clean Air Act and associated rules have led to decreases in emissions and deposition of oxidized N, especially in eastern U.S., representing a research challenge and opportunity for ecosystem ecologists and biogeochemists. The purpose of this paper is to predict changes in the structure and function of North American forest ecosystems in a future of decreased N deposition. Hysteresis is a property of a system wherein output is not a strict function of corresponding input, incorporating lag, delay, or history dependence, particularly when the response to decreasing input is different from the response to increasing input. We suggest a conceptual hysteretic model predicting varying lag times in recovery of soil acidification, plant biodiversity, soil microbial communities, forest carbon (C) and N cycling, and surface water chemistry toward pre-N impact conditions. Nearly all of these can potentially respond strongly to reductions in N deposition. Most responses are expected to show some degree of hysteresis, with the greatest delays in response occurring in processes most tightly linked to "slow pools" of N in wood and soil organic matter. Because experimental studies of declines in N loads in forests of North America are lacking and because of the expected hysteresis, it is difficult to generalize from experimental results to patterns expected from declining N deposition. These will likely be long-term phenomena, difficult to distinguish from other, concurrent environmental changes, including elevated atmospheric CO_2, climate change, reductions in acidity, invasions of new species, and long-term vegetation responses to past disturbance.

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1. Introduction

Gaseous N_2 comprising 78% of the atmosphere (non-reactive N—N_{nr}) neither undergoes photochemical transformations nor enters metabolic pathways in organisms, other than through fixation by a small group of prokaryotes and archaea. By contrast, there are numerous forms of reactive N (N_r) (e.g., NH_3, NH_4^+, NO, NO_2, NO_3^-, N_2O_5, and HNO_3—Horii et al., 2005), which are all capable of undergoing transformations in the atmosphere and entering into biogeochemical cycles of terrestrial and aquatic ecosystems. In addition to emissions of N via internal combustion and power production, ever-increasing amounts of N_{nr} are converted to N-based fertilizer via the Haber-Bosch (H-B) industrial process; currently, fixation via H-B exceeds natural N fixation by >20% (Canfield et al., 2010).

Historical awareness of increasing rates of atmospheric deposition of N_r, along with projections by some models of further increases in the future, provided the initial impetus toward research

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on excess N in forest ecosystems (Lamarque et al., 2013). Based on estimates from Galloway et al. (2004), total global atmospheric deposition of NH_4^+ and NO_3^- in terrestrial ecosystems has increased from $17\,Tg\,N\,yr^{-1}$ in 1860 to $64\,Tg\,N\,yr^{-1}$ in the early 1990s, with projected further increases to $125\,Tg\,N\,yr^{-1}$ by 2050, suggesting that atmospheric deposition of N will have increased by over 7-fold during this 190-year period. Bobbink et al. (2010) predicted similar increases in N deposition by 2030.

Much of the initial work on effects of elevated atmospheric N deposition on terrestrial ecosystems in North America focused on biogeochemical responses, with many studies examining spatial variation in stream/surface water chemistry. Findings generally indicated that with increases in N deposition, leaching rates and stream concentrations of NO_3^- and cations (Ca^{2+}, Mg^{2+}, K^+, Al^{3+}, H^+) coupled to NO_3^- mobility also increased (Murdoch and Stoddard, 1992; Aber et al., 2002; Adams et al., 2006). Large areas of temperate forests in North America, especially those of the eastern U.S., have been shown to be sensitive to N saturation, a phenomenon wherein the rate of atmospheric N deposition exceeds the ability of an ecosystem to retain it (Lovett and Goodale, 2011). More recent work has emphasized the effects of excess N on plant biodiversity, microbial responses, and health of temperate forests (Thomas et al., 2010; Simkin et al., 2016), with broad consensus indicating that excess N has decreased biodiversity and compromised the health of some forests of the eastern U.S. (Clark et al., 2013).

The biogeochemistry of N cycling has received extensive research emphasis (Gorham and Kelly, 2018), especially regarding effects of excess N on ecosystem biogeochemistry and biotic communities. Beginning in the 19th century, with the initiation of the Park Grass Experiment in Rothamsted, England, there is a rich history of experimental studies investigating the effects of N on plant species diversity (Richardson, 1938; Silvertown et al., 2006). As the longest on-going N experiment, Park Grass offered the first evidence that excess N can reduce plant species diversity, results confirmed in the U.S. by ongoing investigations at the Cedar Creek Natural Area in Minnesota (e.g., Clark and Tilman, 2008). Research was initially directed primarily toward herb-dominated communities, but increasingly has since focused on forested ecosystems (Gilliam, 2006; Sutton et al., 2014; Clark et al., 2013; Gilliam et al., 2016).

Empirical data show that emissions of N_r from vehicles and power plants in the U.S. have decreased >50% since the 1980s (Lloret and Valiela, 2016), largely the result of the Clean Air Act (CAA). Because the focus of the CAA has been on oxidized, rather than reduced, forms of N, temporal patterns of atmospheric deposition have differed among N forms. Data collected by the National Atmospheric Deposition Program (http://nadp.slh.wisc.edu/) and by the Clean Air Status and Trends Network (https://www.epa.gov/castnet) indicate that both wet and dry deposition of oxidized N increased from the 1980s, peaked in the mid-to late-1990s, and then decreased notably toward recent years (Du, 2016; Lloret and Valiela, 2016). By contrast, absolute and relative rates of deposition of reduced N have either increased or remained level at most sites (Warner et al., 2017), with increases driven primarily by decreases in SO_2 and NO_x emissions (due to decreases in scavenging of atmospheric NH_4^+ by SO_4^{2-} and NO_3^-) and increases in soil temperature (Fig. 1; Du et al., 2014).

Spatial patterns of total deposition of oxidized versus reduced N over time reveal even sharper contrasts. Whereas total oxidized N deposition reached uniformly high values throughout the eastern U.S. in 2000, by 2014 elevated deposition was confined to major metropolitan areas (Fig. 2a and b). Deposition of reduced N shows a patchy distribution throughout the U.S., driven mainly by agricultural practices, including animal husbandry (Fig. 3a and b)
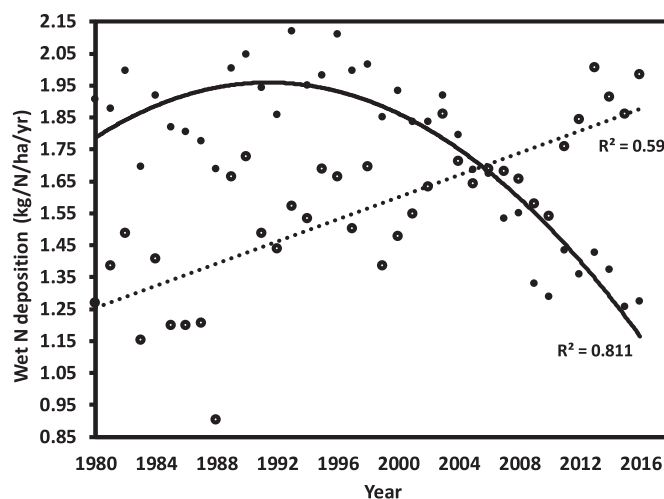

Fig. 1. Annual wet deposition of NH_4^+-N (open symbols/dashed line) and NO_3^--N (closed symbols/solid line) for the U.S. from 1980 to 2015. Data were taken from the National Atmospheric Deposition Program (http://nadp.slh.wisc.edu/) and are averaged across all sample stations in the network.
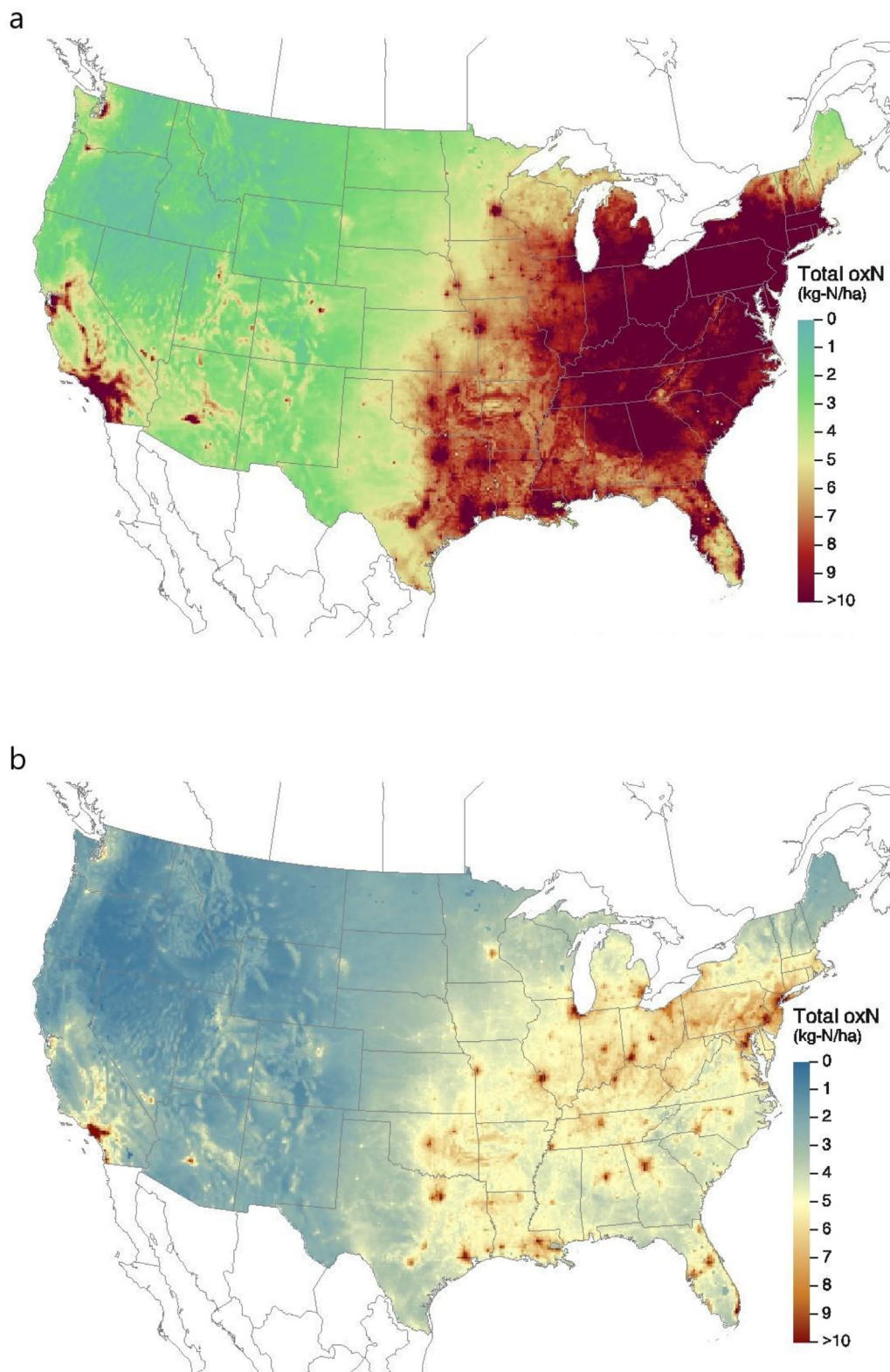
(Costanza et al., 2008). Overall, the eastern U.S. is currently experiencing notable decreases in total N deposition accompanied by a shift towards a greater proportion of reduced N.

The 1980s marked a profound change in how forest ecologists thought about impacts of N on forest ecosystems. Previously, ecologists and forest managers had considered N primarily as the nutrient most limiting forest growth. Later that decade, however, following the publication of several seminal papers about N saturation (e.g., Ågren and Bosatta, 1988; Aber et al., 1989), N deposition was considered potentially detrimental to ecosystem structure and function. Now, however, in contrast to the recent research investigating effects of increasing N on terrestrial ecosystems, research needs to focus on how these impacted ecosystems will change under conditions of decreasing deposition of oxidized N, especially those in eastern North America where deposition has been chronically high.

The purpose of this paper is to describe temporal and spatial patterns of past response and to project likely future scenarios of how forest ecosystems in eastern North America will respond to declines in atmospheric N deposition. We examine these responses in each of five areas: soil acidification, plant biodiversity, soil microbial communities, forest C and N cycling, and surface water chemistry.

We propose a hysteretic model as a conceptual framework for anticipating future change in forests of eastern North America, given current patterns of decreasing atmospheric N deposition. Hysteresis describes any phenomenon in which the state of a property lags behind changes in the effect causing it. It is a property of a system wherein output is not a strict function of corresponding input, but one that incorporates factors, such as response lags and history dependence, resulting in a response to decreasing input that follows a different trajectory than the response to increasing input.

Hysteresis is particularly applicable to ecosystem response to N deposition. The ascending response (Fig. 4) resembles earlier predictions of N saturation (Aber, 1992; Stoddard, 1994), many of which have been supported by empirical data for both biogeochemical and plant community responses (Peterjohn et al., 1996; Gilliam et al., 2016). Because N deposition has varied as a function of time (i.e., increasing in North America up to the mid-1990s—Fig. 1), the x-axis of Fig. 4 reflects the increase in excess N as

Fig. 2. Total deposition of NO$_x$ for 2000 (a) and 2014 (b). Maps were taken from the National Atmospheric Deposition Program (http://nadp.slh.wisc.edu/).

N deposition has increased over time, then reflects the decrease in excess N as N deposition decreases (i.e., decreasing in North America after the mid-1990s—Fig. 1). Our conceptual hysteretic model predicts varying lag times in recovery of soil acidification, plant biodiversity, soil microbial communities, forest C and N cycling, and surface water chemistry toward pre-N impact conditions as deposition of N continues to decline.
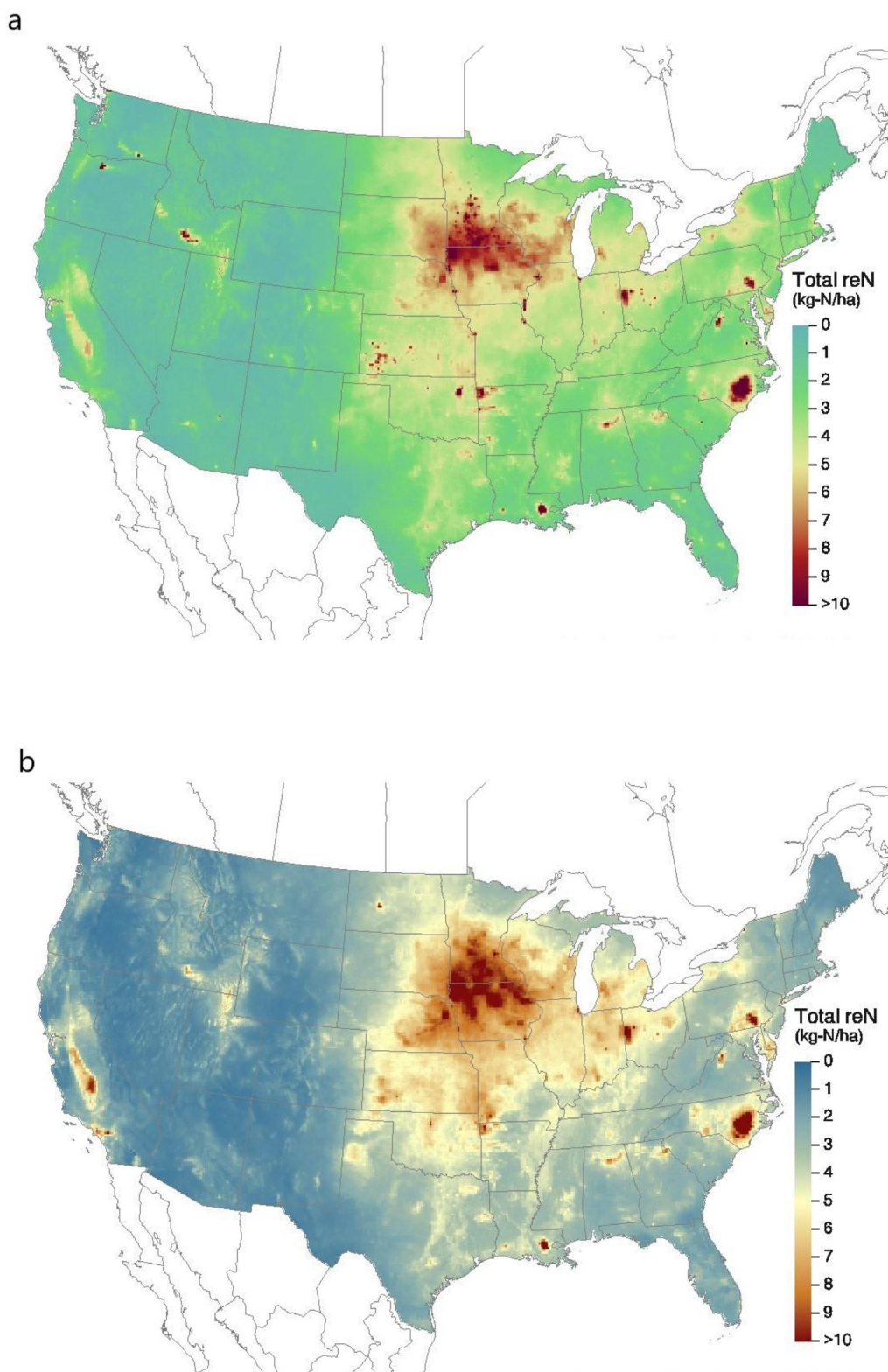
Fig. 3. Total deposition of NH$_y$ for 2000 (a) and 2014 (b). Maps were taken from the National Atmospheric Deposition Program (http://nadp.slh.wisc.edu/).

2. Soil acidification

The CAA targets several criteria pollutants, among them the oxides of S and N that contribute to acid deposition, a phenomenon that has been shown to accelerate soil and water acidification.

Although soil acidification is a natural phenomenon, occurring in regions where precipitation quantity exceeds evapotranspiration (van Breeman et al., 1983; Jones et al., 2012), acid deposition can accelerate natural soil acidification with adverse consequences for acid-sensitive ecosystems. Acid sensitive regions typically are
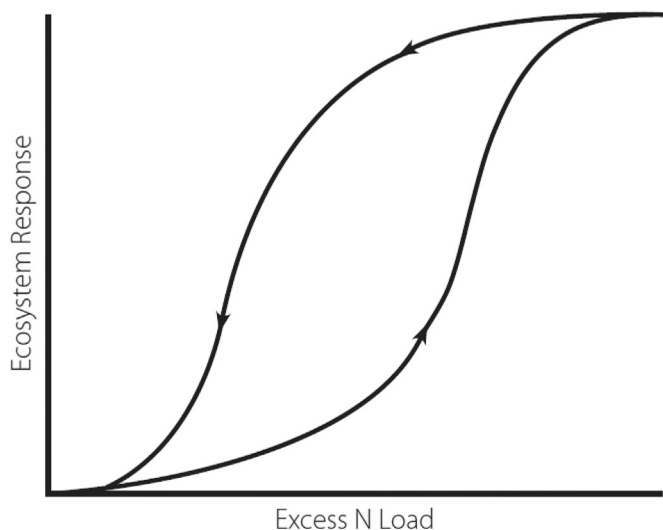
Fig. 4. A hysteretic model proposed for the response of forest ecosystems of eastern North America to decreases in atmospheric deposition of N.

forested, usually occurring in upland regions with soils that are characterized by a relatively low supply of calcium (Ca) and magnesium (Mg) from mineral weathering (Greaver et al., 2012). Soils that have been altered by acid deposition are less able to neutralize inputs of strong acids, provide poorer growing conditions for plants, alter the quality of drainage waters, and extend the time needed for ecosystems to recover from acid deposition. Acid deposition has altered and continues to impact soils in sensitive regions by depleting available Ca and Mg from soil exchange sites and mobilizing dissolved inorganic aluminum (Al_i) into soil and surface waters.

Understanding the cycling of Ca, Mg, and potassium in forest ecosystems involves consideration of the inputs and losses of these important nutrient cations. For many forest ecosystems, the primary supply of Ca and other nutrient cations largely occurs by weathering (i.e., the breakdown of rocks and minerals in soil). Calcium and other nutrient cations may also enter forests through atmospheric deposition. Although this pathway is generally small compared with chemical weathering, exceptions to this pattern occur in extremely Ca-poor ecosystems (Bailey et al., 1996). For example, natural sources such as airborne soil particles originating from arid regions contribute to high atmospheric Ca deposition in Northeast Asia, and human emissions of particulate matter from cement production and other industrial activities result in high atmospheric Ca deposition in South Asia (Larssen and Carmichael, 2000; Zhao et al., 2011).
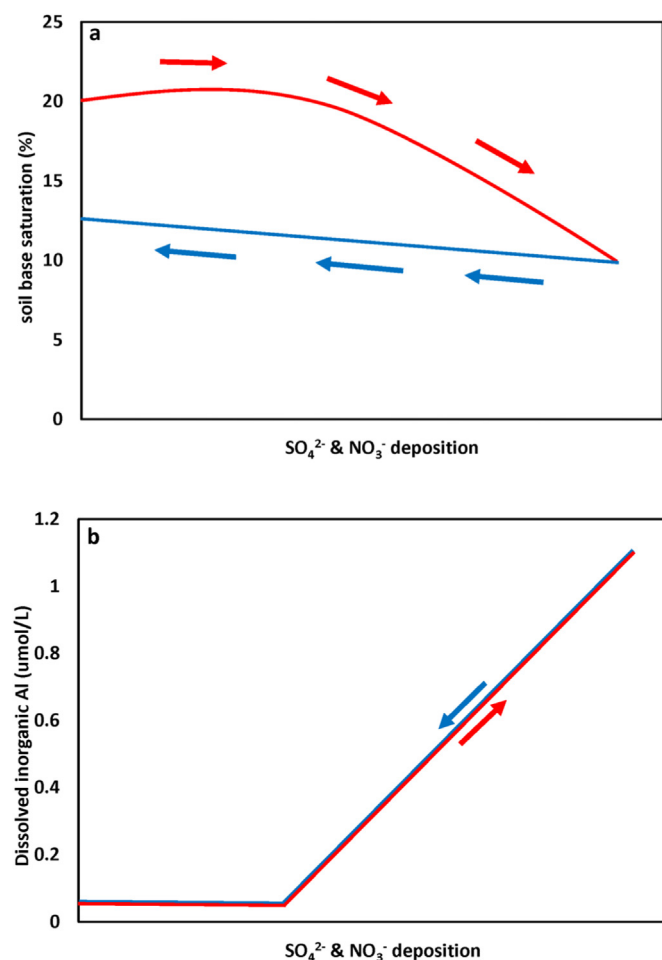
Ecosystem losses of Ca occur largely by vegetation uptake and subsequent harvesting, as well as by leaching in drainage waters. The soil available pool or soil cation exchange complex provides an important and readily available source of C and other nutrient cations. Plants are generally able to utilize these nutrients from solution in equilibrium with soil exchange surfaces. Forest ecosystems that are naturally sensitive to acid deposition are often characterized by low rates of weathering and low quantities of available nutrient cations. Under conditions of elevated acid deposition and subsequent transport of SO_4 and NO_3 in drainage waters, nutrient cations are displaced from available pools and leached from soil (Ruess and Johnson, 1986). This condition is not problematic in areas with high weathering rates and abundant pools of available nutrient cations. However, in acid-sensitive areas with shallow soil characterized by minerals with high resistance to weathering, enhanced loss of Ca and other nutrient cations can result in

depletion of soil available pools. Both NO3 and SO4 contribute to this process but with different seasonal patterns (Ruess and Johnson, 1986). Sulfate leaching generally occurs chronically throughout the annual cycle, due to the fact that it is largely controlled by abiotic processes (i.e., atmospheric deposition and soil adsorption). In contrast, NO_3 leaching often shows strong seasonal patterns driven by biotic processes and is strongly dependent on hydrologic conditions (Stoddard, 1994).

Over the last century, acid deposition and associated leaching by strong acid anions has accelerated the loss of relatively large amounts of available Ca and Mg from soil in acid-sensitive areas (Likens et al., 1996; Warby et al., 2009). This depletion occurs when nutrient cations are displaced from the soil by acid deposition at a rate faster than they can be replenished by the slow breakdown of rocks and minerals or deposition from the atmosphere. Depletion of nutrient cations can fundamentally alter soil development, compromise the nutrition and health of sensitive trees species, influence adjacent surface waters, and limit the capacity of sensitive soils to recover. For example, based on a hindcast analysis, more than half of the available Ca has been lost from soil at the Hubbard Brook Experimental Forest, New Hampshire over the past 70 years (Likens et al., 1996). Note that while acid deposition to acid-sensitive areas is decreasing and there is some associated recovery of the acid neutralizing capacity (ANC) of surface waters, it appears that forest soils continue to exhibit depletion of exchangeable nutrient cations or at best are recovering soil pools of available nutrient cations at a very slow rate (Warby et al., 2009; Fakhraei et al., 2014; Lawrence et al., 2011).

Aluminum (Al) can be released from soil to soil water, lakes, and streams in forested regions with high acid deposition, low stores of available Ca, and acidic soil (Driscoll and Postek, 1995; Driscoll et al., 2001). One of the most significant ecological effects of acid deposition is the mobilization of Al from soil and a shift in the form of Al in water from non-toxic organic forms to toxic Al_i. Concentrations of Al increase markedly with decreases in pH below 6, particularly Al_i. Note that because of short-term temporal variations in NO_3 concentrations, seasonal and episodic variations in Ali generally closely track concentrations of NO_3^- in chronically and episodically acidic waters (Driscoll and Van Dreason, 1993; Driscoll and Postek, 1995). High concentrations of Al_i can be toxic to plants, fish, and other organisms (Gensemer and Playle, 1999). Concentrations of Al_i in acid-impacted surface waters in eastern North America and Europe are above levels considered toxic to fish (~2 µmol/L) and much greater than concentrations observed in forest watersheds that receive low inputs of acidic deposition (Driscoll and Postek, 1995; Driscoll et al., 2001).

Empirical evidence suggests that the temporal pattern of soil available nutrient cations in acid sensitive soils likely follows a hysteretic pattern as described herein for N (Fig. 5a). During the acidification phase, increases in deposition and leaching of strong acid anions facilitate the depletion of exchangeable nutrient cations. Due to low weathering rates, decreases in acid deposition and NO_3^- and SO_4^{2-} leaching will not concurrently result in parallel increases in soil base saturation resulting in slowly recovering soil fertility. In contrast, soil exchangeable Al and concentrations of Al_i in surface waters are highly reversible in response to changes in acid deposition and leaching of strong acid anions in soils, even those with low base saturation (Cronan and Schofield, 1990; Fig. 5b; Lawrence et al., 2011). When soil base saturation is below a critical level (~12–15%), increases in concentrations of strong acid anions associated with increases in acid deposition will result in increases in concentrations of Al_i, because the mobilization/immobilization of Al_i are controlled by fast processes (dissolution/precipitation of secondary Al minerals, adsorption/desorption of Al on soil surfaces).

Fig. 5. Conceptual model of (a) soil % base saturation and (b) soil solution concentrations of dissolved inorganic aluminum (Ali) response to changes in acid deposition in acid sensitive regions. The red line shows the hypothetical trajectory under increases in acid deposition. The blue line shows the trajectory in response to decreases in acid deposition.

The emerging issue of 'browning of surface waters' is associated with the recovery of forested watersheds in North America and Europe from the effects of acid deposition as manifested by increases in the mobilization of dissolved organic matter (Monteith et al. 2007). This process has implications for the recovery of ecosystems from acid and N deposition. First, a component of naturally occurring organic matter functions as organic acids (Fakhraei and Driscoll, 2015). As a result, elevated leaching of dissolved organic matter can also contribute to cation leaching and the acidification of soil and water, albeit as a natural acidification process. These organic acids can also facilitate the mobilization of Al (Driscoll and Postek, 1995; Fakhraei and Driscoll, 2015). However, in this case the form is dissolved organic Al, which is less toxic due to organic complexation. Finally, spatial stream surveys have shown an inverse relationship between concentrations of dissolved organic carbon and NO_3^- (Goodale et al., 2005; Taylor and Townsend, 2010). The mechanism for this empirical relationship is unclear, but may be due to the role of dissolved organic matter as an electron donor during denitrification, or to microbial immobilization of NO_3^- associated with the processing of dissolved organic matter as an energy source. Regardless, if this empirical relationship reflects a biogeochemical mechanism, then the enhanced mobilization of dissolved organic matter following decreases in acid deposition could accelerate the retention or denitrification of NO_3^-, an

additional process that may affect ecosystem recovery.

An unresolved question remains—to what extent will the temporal shift in chemical speciation of atmospheric N deposition observed in the eastern U.S. affect N cycling processes and ecosystem recovery? Ammonium increased from <40% of wet-deposited N in the 1980s to >60% currently (Fig. 1). Does such a change have implications for forest ecosystem response? Certainly, these shifts could impact the recovery of the acid-base chemistry of soil and surface waters. In theory, NH_4^+ deposition followed by nitrification and leaching of NO_3^- results in greater soil acidification than atmospheric NO_3^- deposition and resulting leaching of NO_3^-. Both oxidation and biotic uptake of NH_4^+ are acidifying processes (van Breeman et al. 1983). In contrast, ecosystem uptake of NO_3^- is an alkalization process. We know of no studies that have quantified shifts in soil acidification associated with decreasing atmospheric N deposition and a shift it its speciation. Nevertheless, the overall decreases in atmospheric N deposition and associated decreases in NO_3^- leaching should diminish soil and surface water acidification. However, because this decrease has largely occurred due to decreases in oxidized N deposition, residual N deposition is relatively enriched in NH_4^+ and will have a relatively greater acidification potential (per mol of N deposition) than historical N deposition.

3. Plant biodiversity

In addition to its biogeochemical implications, soil acidification substantially alters availability of essential nutrients for uptake by plants. Thus, there is a strong potential for related changes in forest community structure and diversity. Excess N can have a direct fertilizer effect on plant communities, especially those of the most sensitive stratum of forest vegetation—the herbaceous layer (Gilliam, 2007).

Evidence is ubiquitous that chronically increased N deposition has decreased biodiversity of herb strata in essentially all forest types (Sutton et al., 2014). Clark et al. (2013) estimated, via hindcast modeling, N-mediated species loss for this region of up to 30% from 1985 to 2010. Using data from >15,000 sites, Simkin et al. (2016) demonstrated that negative relationships between atmospheric N deposition and plant species richness are quite common. Gilliam et al. (2016) found a pronounced shift in herb layer composition in response to 25 years of experimental N additions to a central Appalachian hardwood forest. This change arose from increases in a nitrophilic species (*Rubus allegheniensis*) that competitively eliminated numerous N-efficient herbaceous species, resulting in a loss of plant diversity.

Less work has focused on effects of excess N on forest overstory communities. Thomas et al. (2010) modeled the potential effects of N on the sequestration of tree biomass C in temperate forests, along with survivorship of tree seedlings. They found that chronically-elevated N deposition enhanced C storage, but decreased survivorship in eight of 11 common temperate tree species. Not considered in Thomas et al. (2010) is the degree to which herbaceous species can determine the success of tree seedling emergence out of the herb layer, often referred to as an 'ecological filter' (George and Bazzaz, 2014). Thus, N-mediated changes in forest herb communities can represent an indirect effect of N on tree species.

Although few published data directly assess forest diversity response to decreased N deposition, there are studies—virtually all on the forest herb communities—that allow us to address this question to hypothesize on how N-impacted forests may respond to decreased N. Boxman et al. (1998) reported on results of transparent roof studies—the NITREX project in Europe. This networked study integrated results from seven sites throughout Europe wherein high ambient N deposition was excluded experimentally

by roofs to simulate decreased pollutant N inputs. Results from the Netherlands showed that above-ground biomass of nitrophilous species decreased rapidly under the roof that excluded N to the forest floor over a 5 year period (Boxman et al., 1998), suggesting a short-term recovery of the forest herb community to decreased N inputs.

Strengbom et al. (2001) employed two separate forest fertilization experiments in northern Sweden assessing legacy effects of increased N loading following cessation of added N. The first experiment added NH_4NO_3 at rates up to 108 kg N $ha^{-1} y^{-1}$ from 1971 to 1990 (nine years prior to the Strengbom et al. study). Herb layer composition varied significantly between control and treatment areas, with herb diversity significantly lower on treatment areas, arising primarily from sharp declines in ericaceous species and increases in the nitrophilous grass, *Deschampsia flexuosa*. Strengbom et al. (2001) concluded that effects of increased N deposition on the herb layer can be long lived.

In the grass-dominated communities of the Park Grass Experiment, Storkey et al. (2015) demonstrated a positive response of biodiversity to decreases in N from either atmospheric deposition or fertilizers. However, Stevens (2016) reviewed studies from throughout Europe and concluded that plant species composition is likely to be slow to recover during this current period of decreasing atmospheric N deposition. In contrast, she concluded that soil N dynamics are potentially more responsive to such decreases. She further identified possible barriers to recovery, including continued critical load exceedance and lack of either a seed bank or local seed source. Thus, the potential exists for plant communities to reach an alternative stable state wherein species loss from excess N may not allow recovery to pre-impact plant community composition.

That all these studies are from Europe represents a challenge, and a research opportunity, for forests of the eastern U.S. Among these challenges is the wide range of micro-environmental conditions that precludes broad generalizations. The response time of forest herb communities to increases in N load is typically inversely related to ambient N deposition and the antecedent (pre-N enhanced) N status of the forest. Sites with low N availability respond more quickly to increasing N than high N sites (Gilliam et al., 2016). Working in upstate New York and Massachusetts, which receive low levels of N deposition (relative to other regions of the eastern US; Fig. 2), Hurd et al. (1998) and Rainey et al. (1999), respectively, found loss of herb diversity to added N 1—2 years following initiation of treatment. By contrast, Gilliam et al. (2006) found no similar response following 5 year of N additions in a central hardwood forest of West Virginia, a region experiencing chronically high levels of N deposition.

Similar to the effect on soil acidity, the increase in ratio of NH_4^+ to NO_3^- in atmospheric deposition (Fig. 1) previously discussed (see *Soil acidification*) has potential implications for future responses of plant species. Most plant species exhibit little discrimination in form of N for uptake, with the ability to take up both forms of mineral N in soil. In fact, most plant species perform best with a relative balance between the two forms of mineral N (Marschner, 1995). On the other hand, several species adapted to acidic soils (often called *calcifuges*) show a notable preference for NH_4^+, whereas those adapted to neutral to alkaline soils (*calcicoles*) prefer NO_3^- (Marschner, 1995; Nordin et al., 2001). Thus, potential exists for future plant communities to change toward calcifuge dominance, especially those of the herbaceous stratum.

We hypothesize that the degree of hysteresis (i.e., the distance between lines, Fig. 4) will vary directly with the N status of the impacted forest ecosystem. The results of Boxman et al. (1998) for the NITREX study and Storkey et al. (2015) for the Park Grass Experiment suggest that herb diversity can return to pre-impact levels in a decadal time frame. Although the greater

physiognomic complexity of forests (compared to grasslands) should increase recovery time for the forest herb communities (up to 40 yr—Strengbom et al., 2001), we anticipate recovery of biodiversity of such communities under future decreases in N deposition. On the other hand, conclusions of Stevens (2016) are convincing that the composition may be quite different from the pre-impact condition, given alteration of seed pools and sources of dispersal. That is, the number of species may recover, but species composition may remain quite different, relative to pre-impact levels.

4. Soil microbial communities

Excess N deposition that alters both tree and herb communities of impacted forests also affects soil microbiota, both directly in forest floor and mineral soil, and indirectly through linkages between tree and herb strata and soil microbial communities (Gilliam et al., 2014). To predict how these communities in temperate forests may respond to a regional decrease in atmospheric N deposition, it is first important to understand microbial responses to decades-long soil N enrichment and the concomitant changes in soil properties (e.g., pH, nutrient concentrations) and plant communities. Considerable research has documented microbial responses to ambient and simulated N deposition, with general consensus that chronic soil N enrichment reduces total soil microbial biomass, active biomass, and the fungal:bacterial biomass ratio (Frey et al., 2004; Wallenstein et al., 2006; Frey et al., 2014); reduces microbial growth (Kamble et al., 2013) and the activities of extracellular enzymes, especially those associated with lignin breakdown (Carreiro et al., 2000; Sinsabaugh et al., 2002; Deforest et al., 2004; Frey et al., 2004); and reduces the capacity of the microbial community to utilize a suite of C substrates typically found in soil (Frey et al., 2004; Kamble et al., 2013; van Diepen et al., 2017). The mycorrhizal component of the microbial community is particularly sensitive to excess soil N, showing reductions in sporocarp production, root colonization, and mycorrhizal species richness (Egerton-Warburton and Allen, 2000; Lilleskov et al., 2002; Treseder, 2004; Morrison et al., 2016; Carrara et al., 2018).

Recent molecular analyses also indicate significant N-induced shifts in the diversity and composition of both the bacterial (Fierer et al., 2012; Turlapati et al., 2012) and total fungal communities (Entwistle et al., 2013; Mueller et al., 2014; Freedman et al., 2015; Hesse et al., 2015; Morrison et al., 2016), with increased relative abundances of copiotrophic bacteria (Fierer et al., 2012), weak fungal decomposers (e.g., yeasts, cellulolytic ascomycetes; Morrison et al., 2016, 2018), and nitrophilic taxa (Morrison et al., 2016). Thus, chronic soil N enrichment appears to favor the growth and competitive ability of fast-growing, N-tolerant copiotrophs, taxa that may have a lower capacity for efficient organic matter degradation. These shifts in temperate forest microbial communities in response to chronic soil N enrichment are consistently associated with reduced rates of leaf litter (Magill and Aber, 1998; Knorr et al., 2005) and organic matter decomposition (Pregitzer et al., 2008; Liu and Greaver, 2010; Zak et al., 2011; Lovett et al., 2013; Frey et al., 2014), leading to an accumulation of soil organic matter with altered chemistry (Pregitzer et al., 2008; Zak et al., 2008; Lovett et al., 2013; Frey et al., 2014).

It is a reasonable assumption that changes in microbial communities resulting from long-term N additions are reversible if decreases in N deposition are coupled with recovery in soil N cycling rates, pH, base cation and trace nutrient concentrations, and plant community structure. However, a limited number of studies have examined the impact of decreases in N deposition on soil microorganisms. Available data are equivocal, with some evidence for rapid recovery and other results suggesting that microbial

community composition and at least some microbially-mediated processes may remain impacted for many years. Mycorrhizal diversity and fruiting body abundance has been shown to significantly increase ~5–15 years of N after fertilization ceased (Högberg et al., 2014; Stevens, 2016 and references therein), though this result is inconsistent across studies. For example, in a long-term N addition and recovery experiment, mycorrhizal sporocarp abundance remained significantly different on formerly N-enriched plots compared to untreated controls following more than four decades of recovery (Strengbom et al., 2001). Additionally, differences in mycorrhizal species composition remained, with sporocarp production of N-sensitive mycorrhizal fungi lower in formerly N-treated plots. Bacterial community composition is also slow to recover (Högberg et al., 2014). Microbial community reassembly and soil processes (e.g., decomposition) may be slower to recover due to legacy effects resulting from the hysteretic recovery of soil pH and macro- and micronutrient concentrations (e.g., Ca, Mg, P, Mn). Note that, if some aspects of the microbial community recover relatively quickly (e.g., species richness, mycorrhizal sporocarp abundance), the microbial community, as a whole, may not return to the same state (i.e., composition) as prior to the onset of high N deposition rates.

One of the surprises of N deposition research has been the high retention for added N in northeastern U.S. forests (Aber and Magill, 2004). Even systems exposed for decades to high N fertilization rates show retention of greater than 70% of total N inputs (Aber et al., 1989; Aber et al., 1998), with most of the added N being retained in the forest floor and to a lesser extent in the mineral soil (Nadelhoffer et al., 1999, 2004; Templer et al., 2012). So, although soil inorganic N pools may recover to background levels within a few years (Stevens, 2016), total soil N and its overall availability may remain elevated for many years, resulting in cascading effects on other soil properties (e.g., pH, macro- and micronutrient concentrations). As already discussed (see *Plant biodiversity*), plant communities may also show slow recovery, with nitrophilic species self-perpetuating in the understory through mechanisms such as allelopathy, shading of neighbors, and via litter accumulation. Microbial community recovery to its pre-N enriched state may be also be impeded or delayed if the composition of the microbial spore bank was significantly altered (Glassman et al., 2016) or if microbes lost their capacity, over decades of soil N enrichment, to perform particular functions (van Diepen et al., 2017).

5. Forest carbon and nitrogen cycling

Clearly, both increases and decreases in N supply to terrestrial ecosystems can have a profound influence on soil microbial dynamics, which in turn influence the cycling of C and N in impacted forest ecosystems. In addition, two complexities make predicting responses of forest C and N cycling to declining N deposition especially challenging. First, declines in N deposition can cause multiple, sometimes offsetting, responses. For instance, if N is retained by vegetation, it can have a fertilization effect, increasing productivity, whereas if NO_3^- leaches from the ecosystem it can acidify soils and reduce productivity (Lovett and Goodale, 2011) (Fig. 6). Similarly, low levels of N addition can increase plant tissue N concentrations, stimulating N mineralization, but high levels of N addition can decrease decomposition, inhibiting N mineralization rates (Fig. 6). Thus, we may expect different responses at different positions along the trajectory of response to decreases in N deposition.

Second, forest ecosystem pools and fluxes related to C and N cycling are likely to vary widely in the rate at which they respond to decreases in N deposition. Those processes that respond slowly are associated wit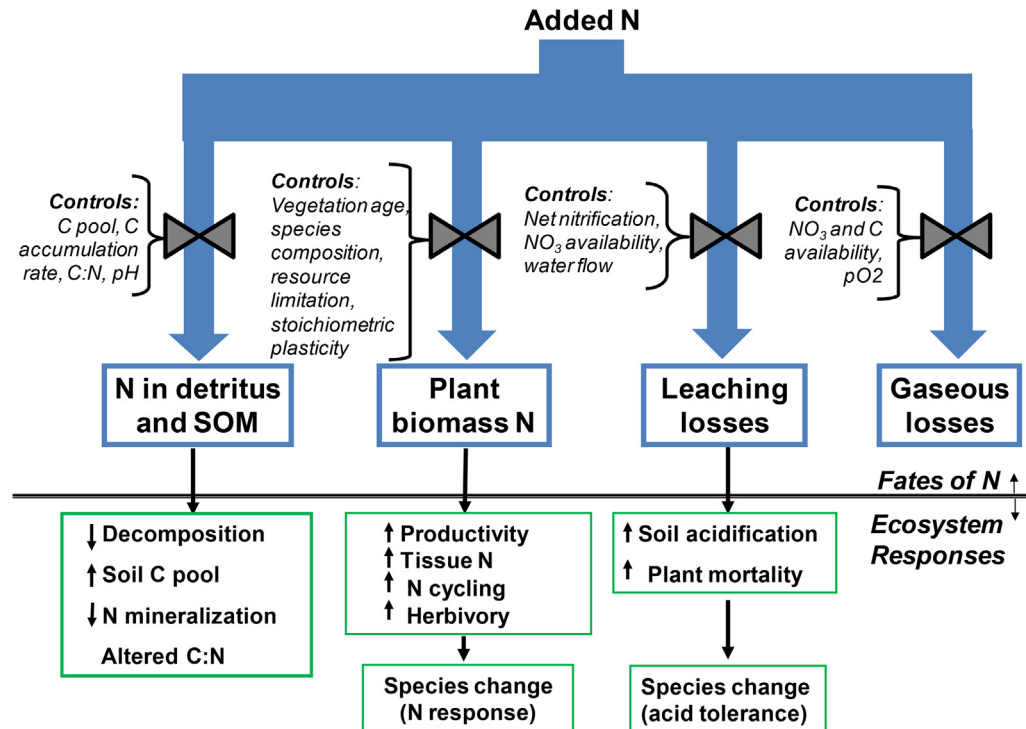h the slow-turnover pools of C and N in the ecosystem, principally soil organic matter (SOM) and wood. Faster-responding processes are associated with pools of N that often have a shorter turnover time such as soil mineral (inorganic) N, microbial biomass, and sometimes foliage and fine roots. Lags in response of the slow pools can produce a hysteresis by slowly feeding N to the fast-response components of the system. In addition, the slow pools can sometimes increase in N content more quickly than they decrease; for example, N in microbial exudates can quickly be incorporated into stable SOM which may take decades or more to turn over. We hypothesize that the SOM and wood pools are likely to respond slowly to declines in N deposition and exhibit pronounced hysteresis, whereas fast pools such as soil mineral N will respond rapidly and exhibit less hysteresis. Further, we predict that the response of ecosystem N cycling processes will vary depending on the extent to which they depend on these slow or fast pools. For example, when the forest C and N cycling model Spe-CN (Crowley et al., 2016; Crowley and Lovett, 2017) is used to simulate the response of a second-growth forest to an increase followed by a decrease in N deposition, net primary production (NPP) responds more slowly and shows more hysteresis than N leaching (Fig. 7a and b). This difference occurs because NPP depends strongly on N availability which is primarily controlled by N mineralization from the slow SOM pool, while leaching (primarily occurring in the dormant season) is controlled much more directly by deposition levels acting through the fast-turnover soil mineral N pools.

These hypotheses are largely consistent with results of field studies of decreases in N deposition in European forests (Stevens, 2016). In the Netherlands and Germany, soil inorganic N fluxes declined by over 50% in coniferous forest plots within 2 years after roofs were placed over the soil to exclude N deposition (Boxman et al., 1995, Bredemeier et al. 1998). Tree-associated variables have changed more slowly in these studies. Tree growth increased significantly in the Dutch N-exclusion plots after 6 years of treatment (Boxman et al., 1998), indicating that the trees had been experiencing negative effects of soil acidification that outweighed the benefits of N fertilization. In Scots pine stands in Sweden, foliar %N and ectomycorrhizal function recovered to control plot levels within 6–15 years after the termination of a 20-year N addition experiment (Hogberg et al., 2011).

Although similar N exclusion experiments have not been reported for forests of eastern North America, N addition experiments have shown that soil NO_3^- and NH_4^+ concentrations, soil solution NO_3^-, and stream NO_3^- generally show relatively fast (i.e., 1–2 years) responses to N addition (e.g., Gilliam et al., 2001; Edwards et al., 2006; Lovett and Goodale, 2011; Fatemi et al., 2012), indicating that those variables are governed by fast-turnover pools of N and are likely to decrease rapidly in response to decreases in N deposition.

In contrast, net N mineralization rates often exhibit no observable change in response to experimental N additions (Wang and Fernandez, 1999; Lovett and Goodale, 2011; Lovett et al., 2013) or changes may develop slowly (Jefts et al., 2004). Gilliam et al. (2018) found that the temporal pattern of net N mineralization and nitrification at the Fernow Experimental Forest in West Virginia was determined more by year-to-year changes in temperature than by experimental N additions through 25 years treatment. Forest growth showed mixed responses to experimental increases in N inputs, with some sites exhibiting increases in vegetation growth (e.g., Fowler et al., 2015; Magill et al., 2004), some showing no change (e.g. Lovett et al., 2013), some showing declines in growth and increased tree mortality (e.g. Magill et al., 2004; Wallace et al., 2007; McNulty et al., 2005). Some sites show increases in tree growth after N fertilization, followed by later decreases, presumably because acidification effects eventually overwhelms fertilization effects (e.g., DeWalle et al., 2006). Differences in the direction

Fig. 6. Conceptual model of the fates of N added to an ecosystem. N can simultaneously move to two internal sinks (plant biomass and detritus/SOM) and two loss mechanisms (leaching and gaseous loss). The fate of the added N determines the response of the ecosystems (boxes below the horizontal line). Reduction in N deposition should cause reduced losses and reduced rate of accumulation in the sinks. Because there are multiple fates of N, there can be multiple effects, potentially offsetting one another, such as increased productivity from N fertilization and decreased productivity from soil acidification. Diagram from Lovett and Goodale (2011).

and timing of tree growth response can probably be attributed to differences in forest age, species composition, and the sensitivity of the site to soil acidification.

Another approach to evaluating patterns of response is to track C and N cycling properties of a forest as ambient N deposition declines. For example, in the Netherlands, decreases in ambient N deposition by about 25% (from ~60 to ~45 kg N ha^{-1} y^{-1}) led to rapid declines in inorganic N concentrations in soil solution and soil leachate (Boxman et al., 2008).
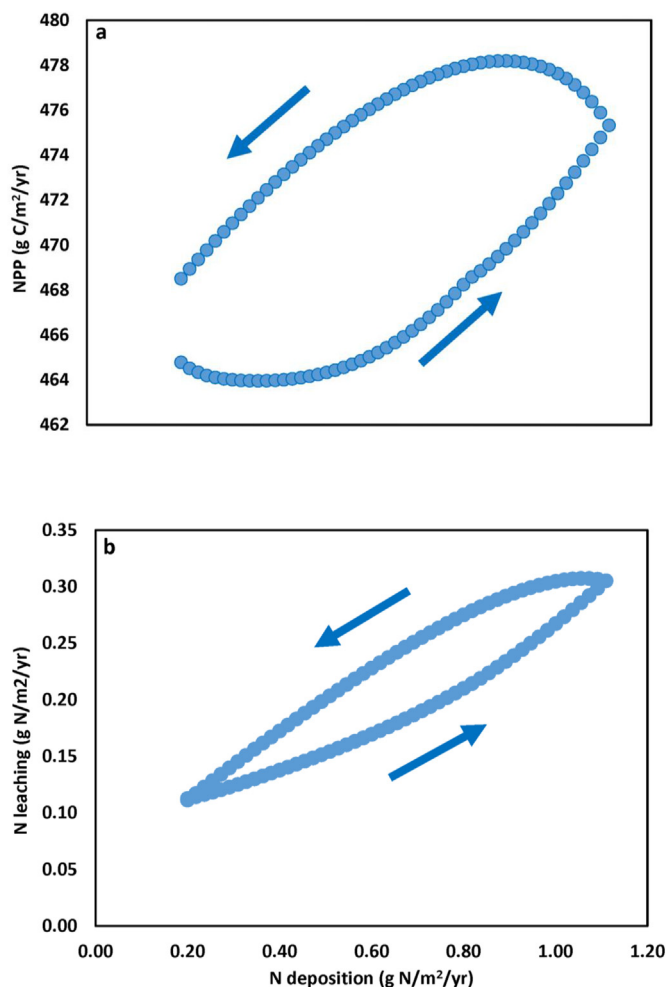
Mathias and Thomas (2018) studied red spruce forests of the Central Appalachian region using a multiproxy approach with 75-year tree ring chronologies of basal area growth, C isotope discrimination ($\Delta^{13}C$, a proxy for leaf gas exchange), and $\Delta^{15}N$ (a proxy for ecosystem N status) to examine individual tree and ecosystem-level responses to global change, including atmospheric CO_2 concentrations, and acid and N deposition. They documented a notable recovery of red spruce ecosystems—including increases in basal area increment (BAI), photosynthesis, stomatal conductance, and water use efficiency—beginning in 1989, which they ascribed to the efficacy of the Clean Air Act and its reduction in several pollutants, including N, but especially acid deposition. Although improvements in soil acidity exhibited the best correlation with recent increases in tree growth, decreases in NO_x emissions from 1989 to the present were significantly correlated with increased BAI in red spruce during this period. Similarly, Wason et al. (2017) found that red spruce in the Adirondack region of New York are showing recent increases in growth rates attributed largely to declines in atmospheric deposition of SO_4 and NO_3 as well as a minor contribution from a warming climate.

At the Hubbard Brook Experimental Forest in NH, USA, one of the best-studied forest research sites in North America, bulk N deposition has declined by 50% from about 8 kg N ha^{-1} y^{-1} in 1990

to about 4 kg N ha^{-1} y^{-1} currently (Rosi-Marshall et al., 2016). During this period there has also been a decline in potential net N mineralization, nitrification and soil inorganic N pools, which Duran et al. (2016) attributed primarily to climate change. There has been a slight decline in aboveground biomass in the reference watershed, attributed primarily to soil acidification (Battles et al., 2014). Soil solution and stream water N export has remained at the lowest levels in the 50-year record at this site (notwithstanding several spikes likely due to disturbance events); the mechanism for these low levels of export is still unknown (Yanai et al., 2013; Fuss et al., 2015), but has been attributed to successional processes (Bernal et al., 2012). All of these trends are consistent with what is expected from declining N deposition, yet in each case the authors attribute them primarily to other causes, illustrating the complexity of distinguishing forest responses to N deposition from responses to other concurrent environmental drivers.

In summary, the evidence presented here leads us to hypothesize that as N deposition declines in eastern North America the processes of N mineralization, soil C storage and forest NPP will require decades to respond because they rely on the soil N pool, much of which turns over quite slowly. However, we hypothesize that the soil inorganic N pools and NO_3^- leaching will track that decline with a lag of a year or two, although these pools will remain somewhat elevated for decades as N is mineralized from the soil N pool.

Observing this sort of long-term response in forests will be difficult because other factors that influence forest ecosystem processes are changing simultaneously. The C and N cycling of forests in the Northeast is affected by ongoing climate change and increases in atmospheric CO_2 as well as many types of disturbance, including wind storms, ice storms, and attacks by insects and diseases, all of which can recur on time scales shorter than the

Fig. 7. Simulation of hysteresis in responses of net primary productivity (a) and N leaching (b) to changing N deposition. This simulation uses the Spe-CN model (Crowley and Lovett, 2017). Simulation is for a mixed forest of *Acer saccharum*, *Betula alleghaniensis* and *Fagus grandifolia*, harvested in 1915 and allowed to regrow as a second-growth forest. N deposition increases from 0.2 g N m^{-2} y^{-1} in 1940 to 1.1 in 1990 and then decreases to 0.2 in 2040 (50 years of increase followed by 50 years of decrease). Response variables are plotted from 1940 to 2040, and the arrows indicate the direction of the temporal sequence. The forest ages from 25 to 125 years old during this simulation, and climate is assumed not to change.

recovery from N deposition. Thus, the signal of a long-term N deposition response will need to be observed through the noise resulting from multiple disturbances. Further, acid deposition is declining concurrently with N deposition, and the de-acidification of the forest can have profound effects on the N cycle (Rosi-Marshall et al., 2016).

6. Surface water chemistry

Also related to effects of de-acidification and decreased atmospheric N loading on N cycling are the direct effects of these declines on the chemistry of surface waters. Four forms of N are generally present in surface waters: NO_3^-, dissolved organic N (DON), NH_4^+, and particulate N (PN). Particulate N, which is generally associated with organic colloids, is usually a minor component of surface water N in minimally-disturbed catchments, except during high flow, when PN can be dominant (Inamdar et al., 2015). Dissolved organic N, an operational term applied to a complex mix of organic N-bearing constituents, is often the dominant form of
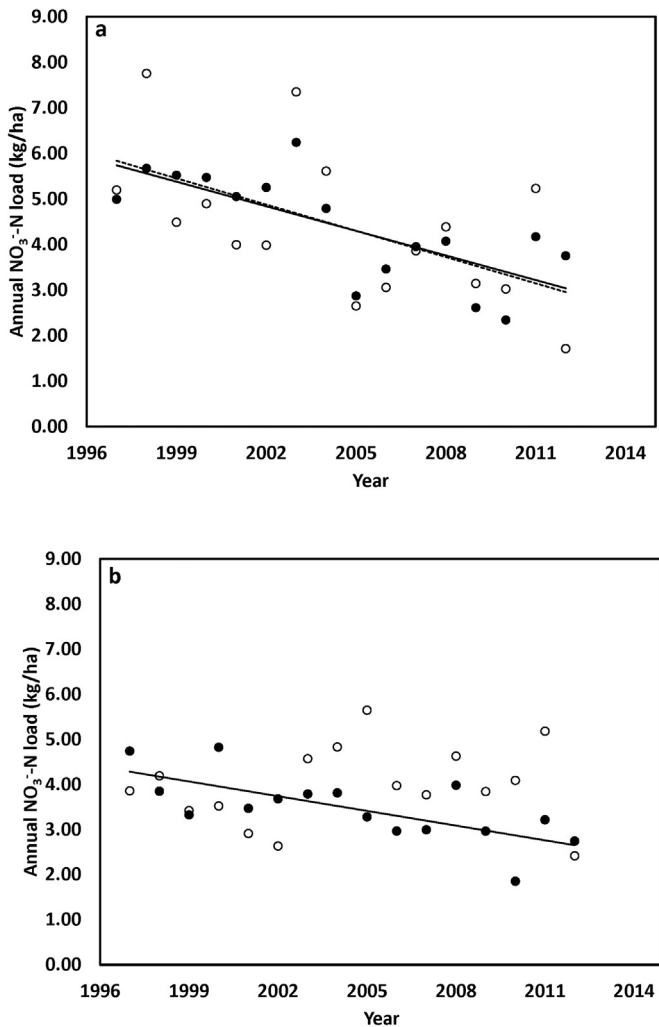
dissolved N in many surface waters (Berman and Bronk, 2003). Although DON is infrequently measured in surface water monitoring programs, DON loads increase with increasing atmospheric N deposition (Brookshire et al., 2007), indicating potential importance as a response indicator. Transformations among DON and dissolved inorganic N (DIN) are observed, with DON serving as both nutrient and energy source in aquatic ecosystems (Wymore et al., 2015). In this section we will largely focus on DIN, of which NO_3^- is typically the dominant form and the most frequently studied and modeled.

In catchments of the northeastern U.S. and southeastern Canada minimally affected by direct human land use, atmospheric N deposition is generally the dominant source of N to watersheds. Two other potential sources—bedrock and N-fixation—may be important in some settings, such as catchments underlain by metasedimentary bedrock and those with significant N-fixing species often associated with wetlands (Williard et al., 2005; Morford et al., 2011).

Elevated NO_3^- leaching from forested catchments in the eastern United States and elsewhere is usually only apparent above an atmospheric N deposition threshold of approximately 5 kg N ha^{-1} yr^{-1} (Aber et al., 2003; Watmough et al., 2005). Above this threshold, which encompasses much of the eastern U.S., there is considerable variability in NO_3^- export such that adjacent catchments receiving the same N deposition can exhibit up to a ten-fold difference in stream NO_3^- export (Aber et al., 2003). There are numerous factors that contribute to this variation in NO_3^- leaching such as those that affect NO_3^- production and consumption in forest soils and NO_3^- transport from soils to surface waters. The balance between NO_3^- production and consumption is primarily influenced by stand age, tree species composition, land use history, and soil physical and chemical characteristics (Lovett and Mitchell, 2004; Lovett et al., 2010; Aber et al., 2003; Lovett et al., 2002). Hence, factors that may lead to changes in tree species composition, e.g., pest or pathogen outbreaks, forest management (silviculture), or climate change, can potentially lead to changes in stream NO_3^- export irrespective of changes in atmospheric N deposition (Crowley and Lovett, 2017).

Factors that alter soil nitrification and plant uptake can greatly alter NO_3^- export, leading to considerable spatial variability. Forest disturbance, including timber harvesting, ice storm damage, soil freezing, and pest or pathogen outbreaks, can lead to differences in NO_3^- export among forested catchments (Aber et al., 2002; Burns and Murdoch, 2005; Eshleman et al., 1998; Nielsen et al., 2001). Increased NO_3^- export following disturbance events is attributable to a combination of reduced plant uptake as well as changes in soil properties (moisture, temperature, fine root death) and these impacts can persist for many years (Compton and Boone, 2000; Goodale et al., 2000).

Spatial variation in NO_3^- export within and among forested catchments may also originate from a variety of other factors including the mix of tree species, differences in flow paths, and the presence of a riparian zone (Burns et al., 1998; Creed and Band, 1998; Lovett et al., 2002). Given the extremely large inherent spatial variability in stream NO_3^- export among forested catchments it is not surprising that the response of surface waters to reductions in atmospheric NO_3^- deposition has been mixed (Argerich et al., 2013). In some cases, a clear response to reductions in atmospheric NO_3^- deposition has been observed (Kothawala et al., 2011; Eshleman et al., 2013; Driscoll et al., 2016), whereas in some cases, no response is evident (Lawrence et al., 2011; McHale et al., 2017) (Fig. 8a and b). Kothawala et al. (2011) reported significant decreases in stream NO_3^- export in response to decreases in N deposition, but only in streams draining catchments with a low wetland fraction. The explanation is that NO_3^- accumulated in the winter

Fig. 8. Annual NO_3^--N loads in wet deposition and stream water during 1997–2012 for (a) Black Lick Run, Maryland, a stream that is responding to decreases in atmospheric deposition, and (b) Biscuit Brook, New York, a stream that is not responding to decreases in atmospheric deposition. For both, closed symbols/solid lines is wet deposition; open symbols/dashed line is stream flow. Linear regression relations at Black Lick Run, wet deposition: slope = −0.18, r^2 = 0.50, p < 0.001, stream: slope = −0.19, r^2 = 0.27, p = 0.022; Biscuit Brook, wet deposition: slope = −0.11, r^2 = 0.46, p = 0.002, stream: no relation, p = 0.60. Data source for Black Run—Eshleman and Sabo (2016), and for Biscuit Brook—McHale et al. (2017).

snowpack was more readily flushed in the spring in catchments with small riparian zones and hence there was a stronger connection to the decline in atmospheric NO_3^- deposition. This strong connection however can be masked by climate change that will likely alter snowpack dynamics and rain-on-snow events that have been shown to increase NO_3^- export (Casson et al., 2014; Crossman et al., 2016). However, as the climate warms further and snowpack depth and duration decrease there will likely be less opportunity for rain-on-snow events and the spring NO_3^- flush may diminish leading to a decline in NO_3^- export (Crossman et al., 2016). Other factors such as temporal changes in air temperature and hydrology, and the response to anthropogenic (e.g., forest harvesting) and natural disturbances (e.g., invasive insects) are believed to affect the long-term trajectory of surface water NO_3^- export and to cause an apparent decoupling of the relationship with declining atmospheric N deposition (Argerich et al., 2013).

Soil freezing is another contributor to the variability in NO_3^- export from forested catchments (Mitchell et al., 1996; Campbell

et al., 2014), which has serious implications in light of ongoing climate change. Snow manipulation studies have suggested that removal of snow can lead to greater soil freezing and more root damage, which can lead to enhanced NO_3^- export in the spring (Campbell et al., 2014). Beyond the influence of changing snow cover and winter dynamics, the response of stream chemistry to decreases in N deposition is complex.

Most studies suggest that N will continue to accumulate in forests in eastern North America even with reductions in atmospheric N deposition (Crossman et al., 2016) and ultimately factors that alter the relative rates of soil NO_3^- production and uptake (e.g., tree species change, climate, soil OM) and transport (altered riparian zone, hydrology) will have considerable influence on future stream NO_3^- export. Hence, as much as stream NO_3^- concentrations have varied in response to increasing N deposition, we expect the response of stream chemistry to decreasing N to be just as variable, with only some sites showing clear relationships with atmospheric N deposition.

7. Synthesis and conclusions: forest response to decreased N deposition in a time of rapid environmental change

The components of forest ecosystem structure and function discussed herein recall the classic quote from Frank Egler (Egler,1977):

"Ecosystems are not only more complex than we think, but more complex than we can think."

Furthermore, such discussion exemplifies the words of Frank Golley that an ecosystem is "… more than the sum of its parts" (Golley, 1996). Although we have addressed several aspects of N-impacted forests of the eastern U.S. separately, it is clear that their responses to decreases in atmospheric deposition of N are highly intermeshed, highlighting the complexity inherent in such forest ecosystems.

As N deposition has declined, ecologists are challenged to understand how N limitation and N excess can exist simultaneously, and how forest ecosystems respond to decreases in available N while still experiencing legacy effects of excess N deposition. A case in point is the Hubbard Brook Experimental Forest, which is undergoing what Groffman et al. (2018) describe as "N oligotrophication." They suggest that C-based reductions in soil N availability could limit forest productivity, a response they see as further enhanced by climate change, including enhanced assimilation caused by increasing atmospheric CO_2 and changes in phenology and length of growing season. Ultimately, they conclude, as we do here, with an urgent call for further research on N dynamics, emphasizing the need to re-evaluate the nature and extent of N cycling in temperate forests. This will lead to a better understanding of how oligotrophication will influence forest ecosystem response in the context of global change (Groffman et al., 2018).

We reach several overarching conclusions from our examination of processes and review of the literature on responses to declining N deposition:

1) Nearly all the processes and conditions that we examined, including soil acidification, microbial populations and processes, C and N cycling, NPP, plant species composition, and stream chemistry, can potentially respond strongly to reductions in N deposition. Similar to the period of increasing atmospheric N deposition, the responses will likely vary considerably from site to site due to variation in forest age, soil type, N status, canopy and understory species composition, and hydrology.

2) Most responses are expected to show some degree of hysteresis, with the greatest delays in response occurring in the processes most tightly linked to the "slowly changing pools" of N in wood and soil organic matter.

3) Experimental studies of declines in N loads in forests of North America are lacking, so it is necessary to infer responses from studies in Europe (where deposition levels have been higher) or from studies in which N loads were increased experimentally (e.g., Oulehle et al., 2011). Because of the expected hysteresis, it is difficult to generalize from experimental increases to patterns expected from declining N deposition. Good data are available from North America tracking stream chemistry as N deposition has declined, and recent studies have used retrospective analysis of tree rings to better understand tree physiological responses.

4) The response to declining N deposition will most certainly be a long-term phenomenon, and will be difficult to distinguish from other, concurrent environmental changes, including elevated atmospheric CO_2, climate change, reductions in acidity, invasions of new species, and long-term vegetation responses to past disturbance. These other environmental factors will govern in part whether measures of ecosystem state (e.g. — soil exchangeable Ca, stream NO_3 concentrations, etc.) will eventually return to an initial condition that existed prior to the advent of anthropogenically-enhanced atmospheric N deposition (Fig. 4), or will reach an alternate stable state, and thus never close the hysteresis loop.

Despite numerous and increasing multi-factorial global change experiments, most understanding of N deposition effects and recovery comes from single-factor fertilization experiments and observational gradient studies. Well-documented single factor responses need to be supplemented with multi-factorial observations on how chronic N additions and their cessation interact with other global change stressors. Indeed, the hypothetical nature of much of our discussion illustrates the need for field data from impacted forests to address the question of recovery from N deposition. These field studies should include both long-term monitoring of ecosystems under declining ambient N deposition, and experimental studies in which current N fertilizations are gradually decreased and ecosystem responses are tracked. Such studies will augment understanding and improve modeling of impacted ecosystems, enhancing evaluation of air pollution policy and prediction of forest responses to future environmental change.

Sir Arthur Tansley famously described ecological succession as "a variable approaching a variable, not a constant" to emphasize the dynamic nature of ecosystem response to an ever-changing environment (Gilliam, 2016). Anthropogenic climate change has created an unprecedented degree of uncertainty in how these forests will respond to future environmental change, including the current trends of decreasing inputs of N from atmospheric deposition. What is certain is that the ambient CO_2 and temperature conditions following any return to pre-impact N status will not be the same as they were prior to historical increases in N deposition.

Acknowledgements

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.09.135.

References

Aber, J.D., 1992. Nitrogen cycling and nitrogen saturation in temperate forest ecosystems. Trends Ecol. Evol. 7, 220—223.

Aber, J.D., Goodale, C.L., Ollinger, S.V., Smith, M.L., Magill, A.H., Martin, M.E., Hallett, R.A., Stoddard, J.L., 2003. Is nitrogen deposition altering the nitrogen status of northeastern forests? Bioscience 53, 375—389.

Aber, J.D., Magill, A.H., 2004. Chronic nitrogen additions at the Harvard Forest (USA): the first 15 years of a nitrogen saturation experiment. For. Ecol. Manag. 196, 1—5.

Aber, J.D., McDowell, W.H., Nadelhoffer, K.J., Magill, A., Berntson, G., Kamakea, M., McNulty, S.G., Currie, W., Rustad, L., Fernandez, I., 1998. Nitrogen saturation in temperate forest ecosystems: hypotheses revisited. Bioscience 48, 921—934.

Aber, J.D., Nadelhoffer, K.J., Steudler, P., Melillo, J.M., 1989. Nitrogen saturation in northern forest ecosystems—hypotheses and implications. Bioscience 39, 378—386.

Aber, J.D., Ollinger, S.V., Driscoll, C.T., Likens, G.E., Holmes, R.T., Freuder, R.J., Goodale, C.L., 2002. Inorganic nitrogen losses from a forested ecosystem in response to physical, chemical, biotic, and climatic perturbations. Ecosystems 5, 648—658.

Adams, M.B., DeWalle, D.R., Hom, J., 2006. The Fernow Watershed Acidification Study. Environmental Pollution Series, vol. 11. Springer, New York.

Ågren, G.I., Bosatta, E., 1988. Nitrogen saturation of terrestrial ecosystems. Environ. Pollut. 54, 185—197.

Argerich, A., Johnson, S.L., Sebestyen, S.D., Rhoades, C.C., Greathouse, E., Knoepp, J.D., Adams, M.B., Likens, G.E., Campbell, J.L., McDowell, W.H., Scatena, F.N., 2013. Trends in stream nitrogen concentrations for forested reference catchments across the USA. Environ. Res. Lett. 8, 014039.

Bailey, S.W., Hornbeck, J.W., Driscoll, C.T., Gaudette, H.E., 1996. Calcium inputs and transport in a base- poor forest ecosystem as interpreted by Sr isotopes. Water Resour. Res. 32, 707—719.

Battles, J.J., Fahey, T.J., Driscoll, C.T., Blum, J.D., Johnson, C.E., 2014. Restoring soil calcium reverses forest decline. Environ. Sci. Technol. Lett. 1, 15—19.

Berman, T., Bronk, D.A., 2003. Dissolved organic nitrogen: a dynamic participant in aquatic ecosystems. Aquat. Microb. Ecol. 31, 279—305.

Bernal, S., Hedin, L.O., Likens, G.E., Gerber, S., Buso, D.C., 2012. Complex response of the forest nitrogen cycle to climate change. Proc. Natl. Acad. Sci. Unit. States Am. 109, 3406—3411.

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.-W., Fenn, M., Gilliam, F., Nordin, A., Pardo, L., de Vries, W., 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity effects of terrestrial ecosystems: a synthesis. Ecol. Appl. 20, 30—59.

Boxman, A.W., Peters, R., Roelofs, J.G.M., 2008. Long term changes in atmospheric N and S throughfall deposition and effects on soil solution chemistry in a Scots pine forest in The Netherlands. Environ. Pollut. 156, 1252—1259.

Boxman, A.W., van der Ven, P.J.M., Roelofs, J.G.M., 1998. Ecosystem recovery after a decrease in nitrogen input to a Scots pine stand at Ysselsteyn, The Netherlands. For. Ecol. Manag. 101, 155—163.

Boxman, A.W., Vandam, D., Vandijk, H.F.G., Hogervorst, R.F., Koopmans, C.J., 1995. Ecosystem responses to reduced nitrogen and sulfur inputs into 2 coniferous forest stands in The Netherlands. For. Ecol. Manag. 71, 7—29.

Bredemeier, M., Blanck, K., Xu, Y.J., Tietema, A., Boxman, A.W., Emmett, B., Moldan, F., Gundersen, P., Schleppi, P., Wright, R.F., 1998. Input-output budgets at the NITREX sites. For. Ecol. Manag. 101, 57—64.

Brookshire, E.N.J., Valett, H.M., Thomas, S.A., Webster, J.R., 2007. Atmospheric N deposition increases organic N loss from temperate forests. Ecosystems 10, 252—262.

Burns, D.A., Murdoch, P.S., 2005. Effects of a clearcut on the net rates of nitrification and N mineralization in a northern hardwood forest, Catskill Mountains, New York, USA. Biogeochemistry 72, 123—146.

Burns, D.A., Murdoch, P.S., Lawrence, G.B., Michel, R.L., 1998. Effect of groundwater springs on NO_3- concentrations during summer in Catskill Mountain streams. Water Resour. Res. 34, 1987—1996.

Campbell, J.L., Reinmann, A.B., Templer, P.H., 2014. Soil freezing effects on sources of nitrogen and carbon leached during snowmelt. Soil Sci. Soc. Am. J. 78, 297—308.

Canfield, D.E., Glazer, A.N., Falkowski, P.G., 2010. The evolution and future of Earth's nitrogen cycle. Science 330, 192—196.

Carrara, J.E., Walter, C.A., Hawkins, J.S., Peterjohn, W.T., Averill, C., Brzostek, E.R., 2018. Interactions among plants, bacteria, and fungi reduce extracellular enzyme activities under long-term N fertilization. Global Change Biol. 24, 2721—2734. https://doi.org/10.1111/gcb.14081.

Carreiro, M.M., Sinsabaugh, R.L., Repert, D.A., Parkhurst, D.F., 2000. Microbial enzyme shifts explain litter decay responses to simulated nitrogen deposition.

Ecology 81, 2359–2365.

Casson, N.J., Eimers, M.C., Watmough, S.A., 2014. Sources of nitrate export during rain-on-snow events at forested catchments. Biogeochemistry 120, 23–36.

Clark, C.M., Morefield, P., Gilliam, F.S., Pardo, L.H., 2013. Estimated losses of plant biodiversity across the U.S. from historical N deposition from 1985–2010. Ecology 94, 1441–1448.

Clark, C.M., Tilman, D., 2008. Loss of plant species after chronic low-level nitrogen deposition to prairie grasslands. Nature 451, 712–715.

Compton, J.E., Boone, R.D., 2000. Long-term impacts of agriculture on soil carbon and nitrogen in New England forests. Ecology 81, 2314–2330.

Costanza, J.K., Marcinko, S.E., Goewert, A.E., Mitchell, C.E., 2008. Potential geographic distribution of atmospheric nitrogen deposition from intensive livestock production in North Carolina, USA. Sci. Total Environ. 398, 76–86.

Creed, I.F., Band, L.E., 1998. Exploring functional similarity in the export of Nitrate-N from forested catchments: a mechanistic modeling approach. Water Resour. Res. 34, 3079–3093.

Cronan, C.S., Schofield, C.L., 1990. Relationships between aqueous aluminum and acidic deposition in forested watersheds of North America and northern Europe. Environ. Sci. Technol. 24, 1100–1105.

Crossman, J., Eimers, M.C., Casson, N.J., Burns, D.A., Campbell, J.L., Likens, G.E., Mitchell, M.J., Nelson, S.J., Shanley, J.B., Watmough, S.A., Webster, K.L., 2016. Regional meteorological drivers and long term trends of winter-spring nitrate dynamics across watersheds in northeastern North America. Biogeochemistry 130, 247–265.

Crowley, K.F., Lovett, G.M., 2017. Effects of nitrogen deposition on nitrate leaching from forests of the northeastern United States will change with tree species composition. Can. J. For. Res. 47, 997–1009.

Crowley, K.F., Lovett, G.M., Arthur, M.A., Weathers, K.C., 2016. Long-term effects of pest-induced tree species change on carbon and nitrogen cycling in northeastern US forests: a modeling analysis. For. Ecol. Manag. 372, 269–290.

Deforest, J.L., Zak, D.R., Pregitzer, K.S., Burton, A.J., 2004. Atmospheric nitrate deposition, microbial community composition, and enzyme activity in northern hardwood forests. Soil Sci. Soc. Am. J. 68, 132–138.

DeWalle, D.R., Kochenderfer, J.N., Adams, M.B., Miller, G.W., Gilliam, F.S., Wood, F., Odenwald-Clemens, S.S., Sharpe, W.E., 2006. Vegetation and acidification. In: Adams, M.B., DeWalle, D.R., Hom, J. (Eds.), The Fernow Watershed Acidification Study. Springer, Dordrecht, Netherlands, pp. 137–188.

Driscoll, C.T., Lawrence, G.B., Bulger, A.J., Butler, T.J., Cronan, C.S., Eagar, C., Lambert, K.F., Likens, G.E., Stoddard, J.L., Weathers, K.C., 2001. Acidic deposition in the Northeastern United States: sources and inputs, ecosystem effects, and management strategies. Bioscience 51, 180–198.

Driscoll, C.T., Postek, K.M., 1995. The chemistry of aluminum in surface waters. In: The Environmental Chemistry of Aluminum, pp. 363–418. Lewis, Chelsea, MI.

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

Driscoll, C.T., Driscoll, K.M., Fakhraei, H., Civerolo, K., 2016. Long-term temporal trends and spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in response to decreases in acidic deposition. Atmos. Environ. 146, 5–14.

Du, E., 2016. Rise and fall of nitrogen deposition in the United States. Proc. Natl. Acad. Sci. Unit. States Am. 113, E3594–E3595.

Du, E., de Vries, W., Galloway, J.N., Hu, X., Fang, J., 2014. Changes in wet nitrogen deposition in the United States between 1985 and 2012. Environ. Res. Lett. 9, 095004.

Duran, J., Morse, J.L., Groffman, P.M., Campbell, J.L., Christenson, L.M., Driscoll, C.T., Fahey, T.J., Fisk, M.C., Likens, G.E., Melillo, J.M., Mitchell, M.J., Templer, P.H., Vadeboncoeur, M., 2016. Climate change decreases nitrogen pools and mineralization rates in northern hardwood forests. Ecosphere 7, e1251. https://doi.org/10.1002/ecs2.1251.

Edwards, P.J., Williard, K.W.J., Wood, F., Sharpe, W.E., 2006. Soil water and stream chemical responses. In: Adams, M.B., DeWalle, D.R., Hom, J. (Eds.), The Fernow Watershed Acidification Study. Springer, Dordrecht, Netherlands, pp. 71–136.

Egerton-Warburton, L.M., Allen, E.B., 2000. Shifts in arbuscular mycorrhizal communities along an anthropogenic nitrogen deposition gradient. Ecol. Appl. 10, 484–496.

Egler, F.E., 1977. The Nature of Vegetation: its Management and Mismanagement. Aton Forest, Norfolk, CT.

Entwistle, E.M., Zak, D.R., Edwards, I.P., 2013. Long-term experimental nitrogen deposition alters the composition of the active fungal community in the forest floor. Soil Sci. Soc. Am. J. 77, 1648–1658.

Eshleman, K.N., Morgan, R.P., Webb, J.R., Deviney, F.A., Galloway, J.N., 1998. Temporal patterns of nitrogen leakage from mid-Appalachian forested watersheds: role of insect defoliation. Water Resour. Res. 34, 2005–2116.

Eshleman, K.N., Sabo, R.D., 2016. Declining nitrate-N yields in the Upper Potomac River Basin: what is really driving progress under the Chesapeake Bay restoration? Atmos. Environ. 146, 280–289.

Eshleman, K.N., Sabo, R.D., Kline, K.M., 2013. Surface water quality is improving due to declining atmospheric N deposition. Environ. Sci. Technol. 47, 12193–12200.

Fakhraei, H., Driscoll, C.T., 2015. Proton and aluminum binding properties of organic acids in surface waters of the northeastern U.S. Environ. Sci. Technol. 49, 2939–2947.

Fakhraei, H., Driscoll, C.T., Selvendiran, P., DePinto, J.V., Bloomfield, J., Quinn, S., Rowell, H.C., 2014. Development of a total maximum daily load (TMDL) for acid-impaired lakes in the Adirondack region of New York. Atmos. Environ. 95, 277–287.

Fatemi, F.R., Fernandez, I.J., Szillery, J., Norton, S.A., Rustad, L.E., 2012. Soil solution chemical response to two decades of experimental acidification at the Bear Brook Watershed in Maine. Water Air Soil Pollut. 223, 6171–6186.

Fierer, N., Leff, J.W., Adams, B.J., Nielsen, U.N., Bates, S.T., Lauber, C.L., Owens, S., Gilbert, J.A., Wall, D.H., Caporaso, J.G., 2012. Cross-biome metagenomic analyses of soil microbial communities and their functional attributes. Proc. Nat. Acad. Sci. 109, 21390–21395.

Fowler, Z.K., Adams, M.B., Peterjohn, W.T., 2015. Will more nitrogen enhance carbon storage in young forest stands in central Appalachia? For. Ecol. Manag. 337, 144–152.

Freedman, Z.B., Romanowicz, K.J., Upchurch, R.A., Zak, D.R., 2015. Differential responses of total and active soil microbial communities to long-term experimental N deposition. Soil Biol. Biochem. 90, 275–282.

Frey, S.D., Ollinger, S., Nadelhoffer, K., Bowden, R., Brzostek, E., Burton, A., Caldwell, B., Crow, S., Goodale, C., Grandy, A.S., Finzi, A., Kramer, M., Lajtha, K., Martin, M., McDowell, W., Minocha, R., Templer, P., Wicking, K., 2014. Chronic nitrogen additions suppress decomposition and sequester soil carbon in a temperate forest. Biogeochemistry 121, 305–316.

Frey, S.D., Knorr, M., Parrent, J., Simpson, R.T., 2004. Chronic nitrogen enrichment affects the structure and function of the soil microbial community in a forest ecosystem. For. Ecol. Manag. 196, 159–171.

Fuss, C.B., Driscoll, C.T., Campbell, J.L., 2015. Recovery from chronic and snowmelt acidification: long-term trends in stream and soil water chemistry at the Hubbard Brook Experimental Forest, New Hampshire, USA. J. Geophys. Res.: Biogeosciences 120, 2360–2374.

Galloway, J.N., Dentener, F.J., Capone, D.G., Boyer, E.W., Howarth, R.W., Seitzinger, S.P., Asner, G.P., Cleveland, C.C., Green, P.A., Holland, E.A., Karl, D.M., Michaels, A.F., Porter, J.H., Townsend, A.R., Vörösmarty, C.J., 2004. Nitrogen cycles: past, present, and future. Biogeochemistry 70, 153–226.

Gensemer, R.W., Playle, R.C., 1999. The bioavailability and toxicity of aluminum in aquatic environments. Crit. Rev. Environ. Sci. Technol. 29, 315–450.

George, L.O., Bazzaz, F.A., 2014. The herbaceous layer as a filter determining spatial pattern in forest tree regeneration. In: Gilliam, F.S. (Ed.), The Herbaceous Layer in Forests of Eastern North America, second ed. Oxford University Press, Inc., New York, NY, USA, pp. 340–355.

Gilliam, F.S., 2006. Response of the herbaceous layer of forest ecosystems to excess nitrogen deposition. J. Ecol. 94, 1176–1191.

Gilliam, F.S., 2007. The ecological significance of the herbaceous layer in temperate forest ecosystems. Bioscience 57, 845–858.

Gilliam, F.S., 2016. Forest ecosystems of temperate climatic regions: from ancient use to climate change. New Phytol. 212, 871–887.

Gilliam, F.S., Hockenberry, A.W., Adams, M.B., 2006. Effects of atmospheric nitrogen deposition on the herbaceous layer of a central Appalachian hardwood forest. J. Torrey Bot. Soc. 133, 240–254.

Gilliam, F.S., Yurish, B.M., Adams, M.B., 2001. Temporal and spatial variation of nitrogen transformations in nitrogen-saturated soils of a central Appalachian hardwood forest. Can. J. For. Res. 31, 1768–1785.

Gilliam, F., Walter, C., Adams, M.B., Peterjohn, W.T., 2018. Nitrogen (N) dynamics in the mineral soil of a Central Appalachian hardwood forest during a quarter century of whole-watershed N additions. Ecosystems (in press).

Gilliam, F.S., Hédl, R., Chudomelová, M., McCulley, R.L., Nelson, J.A., 2014. Variation in vegetation and microbial linkages with slope aspect in a montane temperate hardwood forest. Ecosphere 5, 66. https://doi.org/10.1890/ES13-00379.1.

Gilliam, F.S., Welch, N.T., Phillips, A.H., Billmyer, J.H., Peterjohn, W.T., Fowler, Z.K., Walter, C.A., Burnham, M.B., May, J.D., Adams, M.B., 2016. Twenty-five year response of the herbaceous layer of a temperate hardwood forest to elevated nitrogen deposition. Ecosphere 7, e01250. https://doi.org/10.1002/ecs2.1250.

Glassman, S.I., Levine, C.R., DiRocco, A., Battles, J.J., Bruns, T.D., 2016. Ectomycorrhizal fungal spore bank recovery after a severe forest fire: some like it hot. ISME J. 10, 1228–1239.

Golley, F.B., 1996. A History of the Ecosystem Concept in Ecology: More than the Sum of the Parts. Yale University Press, New Haven, CT.

Goodale, C.L., Aber, J.D., McDowell, W.H., 2000. The long-term effects of disturbance on organic and inorganic nitrogen export in the White Mountains, New Hampshire. Ecosystems 3, 433–450.

Goodale, C.L., Aber, J.D., Vitousek, P.M., McDowell, W.H., 2005. Long-term decreases in stream nitrate: successional causes unlikely; possible links to DOC? Ecosystems 8, 334–337.

Gorham, E., Kelly, J., 2018. A history of ecological research derived from titles of articles in the journal "ecology," 1925–2015. Bull. Ecol. Soc. Am. 99, 61–72.

Greaver, T.L., Sullivan, T.J., Herrick, J.D., Barber, M.C., Baron, J.S., Cosby, B.J., Deerhake, M.E., Dennis, R.L., Dubois, J.-J.B., Goodale, C.L., Herlihy, A.T., Lawrence, G.B., Liu, L., Lynch, J.A., Novak, K.J., 2012. Ecological effects of nitrogen and sulfur air pollution in the US: what do we know? Front. Ecol. Environ. 10, 365–372.

Groffman, P.M., Driscoll, C.T., Durán, J., Campbell, J.L., Christenson, L.M., Fahey, T.J., Fisk, M.C., Fuss, C., Likens, G.E., Lovett, G., Rustad, L., Templer, P.H., 2018. Nitrogen Oligotrophication in Northern Hardwood Forests. Biogeochemistry in press.

Hesse, C.N., Mueller, R.C., Vuyisich, M., 2015. Forest floor community metatranscriptomes identify fungal and bacterial responses to N deposition in two maple forests. Front. Microbiol. 6, 337. https://doi.org/10.3389/fmicb.2015.00337.

Högberg, M.N., Yarwood, S., Myrold, D.D., 2014. Fungal but not bacterial soil communities recover after termination of decadal nitrogen additions to boreal forest. Soil Biol. Biochem. 72, 35–43.

Hogberg, P., Johannisson, C., Yarwood, S., Callesen, I., Nasholm, T., Myrold, D.D., Hogberg, M.N., 2011. Recovery of ectomycorrhiza after 'nitrogen saturation' of a conifer forest. New Phytol. 189, 515—525.

Horii, C.V., Munger, J.W., Wofsy, S.C., Zahniser, M., Nelson, D., McManus, J.B., 2005. Atmospheric reactive nitrogen concentration and flux budgets at a North-eastern U.S. forest site. Agric. For. Meteorol. 133, 210—225.

Hurd, T.M., Brach, A.R., Raynal, D.J., 1998. Response of understory vegetation of Adirondack forests to nitrogen additions. Can. J. For. Res. 28, 799—807.

Inamdar, S., Dhillon, G., Singh, S., Parr, T., Qin, Z., 2015. Particulate nitrogen exports in stream runoff exceed dissolved nitrogen forms during large tropical storms in a temperate, headwater, forested watershed. J. Geophys. Res.: Biogeosciences 120, 1548—1566.

Jefts, S., Fernandez, I.J., Rustad, L.E., Dail, D.B., 2004. Decadal responses in soil N dynamics at the bear Brook watershed in Maine, USA. For. Ecol. Manag. 189, 189—205.

Jones, J.A., Creed, I.F., Hatcher, K.L., Warren, R.J., Adams, M.B., Benson, M.H., Boose, E., Brown, W.A., Campbell, J.L., Covich, A., Clow, D.W., Dahm, C.N., Elder, K., Ford, C.R., Grimm, N.B., Henshaw, D.L., Larson, K.L., Miles, E.S., Miles, K.M., Sebestyen, S.D., Spargo, A.T., Stone, A.B., Vose, J.M., Williams, M.W., 2012. Ecosystem processes and human influences regulate streamflow response to climate change at long-term ecological research sites. Bioscience 62, 390—404.

Kamble, P.N., Rousk, J., Frey, S.D., Bååth, E., 2013. Bacterial growth and growth limiting nutrients following chronic nitrogen additions to a hardwood forest soil. Soil Biol. Biochem. 59, 32—37. https://doi.org/10.1016/j.soilbio.2012.12.017.

Knorr, M., Frey, S.D., Curtis, P.S., 2005. Nitrogen additions and litter decomposition: a meta-analysis. Ecol 86, 3252—3257.

Kothawala, D.N., Watmough, S.A., Futter, M.N., Zhang, L., Dillon, P.J., 2011. Stream nitrate responds rapidly to decreasing nitrate deposition. Ecosystems 14, 274—286.

Lamarque, J.F., Dentener, F., McConnell, J., Ro, C.-U., Shaw, M., Vet, R., Bergmann, D., Cameron-Smith, P., Dalsoren, S., Doherty, R., Faluvegi, G., Ghan, S.J., Josse, B., Lee, Y.H., MacKenzie, I.A., Plummer, D., Shindell, D.T., Skeie, R.B., Stevenson, D.S., Strode, S., Zeng, G., Curran, M., Dahl-Jensen, D., Das, S., Fritzsche, D., Nolan, M., 2013. Atmos. Chem. Phys. 13, 7997—8018. https://doi.org/10.5194/acp-13-7997-2013.

Larssen, T., Carmichael, G.R., 2000. Acid rain and acidification in China: the importance of base cation deposition. Environ. Pollut. 110, 89—102.

Lawrence, G.B., Simonin, H.A., Baldigo, B.P., Roy, K.M., Capone, S.B., 2011. Changes in the chemistry of acidified Adirondack streams from the early 1980s to 2008. Environ. Pollut. 159, 2750—2758.

Likens, G.E., Driscoll, C.T., Buso, D.C., 1996. Long-term effects of acid rain: response and recovery of a forest ecosystem. Science 272, 244—246.

Lilleskov, E.A., Fahey, T.J., Horton, T.R., Lovett, G.M., 2002. Belowground ectomy-corrhizal fungal community change over a nitrogen deposition gradient in Alaska. Ecol. 83, 104—115.

Liu, L., Greaver, T.L., 2010. A global perspective on belowground carbon dynamics under nitrogen enrichment. Ecol. Lett. 13, 819—828.

Lloret, J., Valiela, I., 2016. Unprecedented decrease in deposition of nitrogen oxides over North America: the relative effects of emission controls and prevailing air-mass trajectories. Biogeochemistry 129, 165—180.

Lovett, G.M., Arthur, M.A., Weathers, K.C., Fitzhugh, R.D., Templer, P.H., 2013. Nitrogen addition increases carbon storage in soils, but not in trees, in an eastern US deciduous forest. Ecosystems 16, 980—1001.

Lovett, G.M., Arthur, M.A., Weathers, K.C., Griffin, J.M., 2010. Long-term changes in forest carbon and nitrogen cycling caused by an introduced pest/pathogen complex. Ecosystems 13, 1188—1200.

Lovett, G.M., Goodale, C.L., 2011. A new conceptual model of nitrogen saturation based on experimental nitrogen addition to an oak forest. Ecosystems 14, 615—631.

Lovett, G.M., Mitchell, M.J., 2004. Sugar maple and nitrogen cycling in the forests of eastern North America. Front. Ecol. Environ. 2, 81—88.

Lovett, G.M., Weathers, K.C., Arthur, M.A., 2002. Control of nitrogen loss from forested watersheds by soil carbon: nitrogen ratio and tree species composition. Ecosystems 5, 712—718.

Magill, A.H., Aber, J., 1998. Long-term effects of experimental nitrogen additions on foliar litter decay and humus formation in forest ecosystems. Plant Soil 203, 301—311.

Magill, A.H., Aber, J.D., Currie, W.S., Nadelhoffer, K.J., Martin, M.E., McDowell, W.H., Melillo, J.M., Steudler, P., 2004. Ecosystem response to 15 years of chronic nitrogen additions at the Harvard Forest LTER, Massachusetts, USA. For. Ecol. Manag. 196, 7—28.

Marschner, H., 1995. Mineral Nutrition of Higher Plants, second ed. Academic Press, London, England.

Mathias, J.M., Thomas, R.B., 2018. Disentangling the effects of acidic air pollution, atmospheric CO2, and climate change on recent growth of red spruce trees in the Central Appalachian Mountains. Global Change Biol. 24, 3938—3953.

McHale, M.R., Burns, D.A., Siemion, J., Antidormi, M.R., 2017. The response of soil and stream chemistry to decreases in acid deposition in the Catskill Mountains, New York, USA. Environ. Pollut. 299, 607—620.

McNulty, S.G., Boggs, J., Aber, J.D., Rustad, L., Magill, A., 2005. Red spruce ecosystem level changes following 14 years of chronic N fertilization. For. Ecol. Manag. 219, 279—291.

Mitchell, M.J., Driscoll, C.T., Kahl, J.S., Likens, G.E., Murdoch, P.S., Pardo, L.H., 1996. Climatic control of nitrate loss from forested watersheds in the northeast

United States. Environ. Sci. Technol. 30, 2609—2612.

Monteith, D.T., Stoddard, J.L., Evans, C.D., De Wit, H.A., Forsius, M., Høgåsen, T., Wilander, A., Skjelkvåle, B.L., Jeffries, D.S., Vuorenmaa, J., Keller, B., Kopécek, J., Vesely, J., 2007. Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. Nature 450, 537—541.

Morford, S.L., Houlton, B.Z., Dahlgren, R.A., 2011. Increased forest ecosystem carbon and nitrogen storage from nitrogen rich bedrock. Nature 477, 78—81.

Morrison, E.W., Frey, S.D., Sadowsky, J.J., van Diepen, L., Thomas, W.K., Pringle, A., 2016. Chronic nitrogen additions fundamentally restructure the soil fungal community in a temperate forest. Fung. Ecol. 23, 48—57.

Morrison, E.W., Pringle, A., van Diepen, L.T.A., Frey, S.D., 2018. Simulated nitrogen deposition favors stress-tolerant fungi with low potential for decomposition. Soil Biol. Biochem. 125, 75—85.

Mueller, R.C., Balasch, M.M., Kuske, C.R., 2014. Contrasting soil fungal community responses to experimental nitrogen addition using the large subunit rRNA taxonomic marker and cellobiohydrolase I functional marker. Mol. Ecol. 23, 4406—4417.

Murdoch, P.S., Stoddard, J.L., 1992. The role of nitrate in the acidification of streams in the Catskill Mountains of New York. Water Resour. Res. 28, 2707—2720.

Nadelhoffer, K.J., Colman, B.P., Currie, W.S., Magill, A., Aber, J.D., 2004. Decadal-scale fates of ^{15}N tracers added to oak and pine stands under ambient and elevated N inputs at the Harvard Forest (USA). For. Ecol. Manag. 196, 89—107.

Nadelhoffer, K.J., Emmett, B.A., Gundersen, P., Kjønaas, J., Koopmans, C.J., Schleppi, P., Tietema, A., Wright, R.F., 1999. Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. Nature 398, 145—148.

Nielsen, C.B., Groffman, P.M., Hamburg, S.P., Driscoll, C.T., Fahey, T.J., Hardy, J.P., 2001. Freezing effects on carbon and nitrogen cycling in northern hardwood forest soils. Soil Sci. Soc. Am. J. 65, 1723—1730.

Nordin, A., Högberg, P., Näsholm, T., 2001. Soil nitrogen form and plant nitrogen uptake along a boreal forest productivity gradient. Oecologia 129, 125—132.

Oulehle, F., Evans, C.D., Hofmeister, J., Krejci, R., Tahovska, K., Persson, T., Cudlin, P., Hruska, J., 2011. Major changes in forest carbon and nitrogen cycling caused by declining sulphur deposition. Global Change Biol. 17, 3115—3129.

Peterjohn, W.T., Adams, M.B., Gilliam, F.S., 1996. Symptoms of nitrogen saturation in two central Appalachian hardwood forests. Biogeochemistry 35, 507—522.

Pregitzer, K.S., Burton, A.J., Zak, D.R., Talhelm, A.F., 2008. Simulated chronic N deposition increases carbon storage in northern temperate forests. Global Change Biol. 14, 142—153.

Rainey, S.M., Nadelhoffer, K.J., Silver, W.L., Downs, M.R., 1999. Effects of chronic nitrogen additions on understory species in a red pine plantation. Ecol. Appl. 9, 949—957.

Richardson, H.L., 1938. The nitrogen cycle in grassland soil with particular reference to the Rothamsted Park Grass experiment. J. Agric. Sci. 28, 73—121.

Rosi-Marshall, E.J., Bernhardt, E.S., Buso, D.C., Driscoll, C.T., Likens, G.E., 2016. Acid rain mitigation experiment shifts a forested watershed from a net sink to a net source of nitrogen. Proc. Natl. Acad. Sci. U.S.A. 113, 7580—7583.

Ruess, J.O., Johnson, D.W., 1986. Soil acidification: Fundamental concepts. In: Acid Deposition and the Acidification of Soils and Waters. Springer, New York, pp. 7—13.

Silvertown, J., Poulton, P., Johnston, A.E., Edwards, G., Heard, M., Biss, P.M., 2006. The Park grass experiment 1856—2006: its contribution to ecology. J. Ecol. 94, 801—814.

Simkin, S.M., Allen, E.B., Bowman, W.D., Clark, C.M., Belnap, J., Brooks, M.L., Cade, B.S., Collins, S.L., Geiser, L.H., Gilliam, F.S., Jovan, S.E., Pardo, L.H., Schulz, B.K., Stevens, C.J., Suding, K.N., Throop, H.L., Waller, D.M., 2016. A continental analysis of ecosystem vulnerability to atmospheric nitrogen deposition. Proc. Natl. Acad. Sci. Unit. States Am. 113, 4086—4091.

Sinsabaugh, R.L., Carreiro, M.M., Repert, D.A., 2002. Allocation of extracellular enzymatic activity in relation to litter composition, N deposition, and mass loss. Biogeochemistry 60, 1—24.

Stevens, C.J., 2016. How long do ecosystems take to recover from atmospheric nitrogen deposition? Biol. Conserv. 200, 160—167.

Stoddard, J.L., 1994. Long-term changes in watershed retention of nitrogen: its causes and aquatic consequences. In: Baker, L.A. (Ed.), Environmental Chemistry of Lakes and Reservoirs. American Chemical Society, Washington, DC, pp. 223—284.

Storkey, J., Macdonald, A.J., Poulton, P.R., Scott, T., Köhler, I.H., Schnyder, H., Goulding, K.W.T., Crawley, M.J., 2015. Grassland biodiversity bounces back from long-term nitrogen addition. Nature 528, 401—416.

Strengbom, J., Nordin, A., Nasholm, T., Ericson, L., 2001. Slow recovery of boreal forest ecosystem following decreased nitrogen input. Funct. Ecol. 15, 451—457.

Sutton, M.A., Mason, K.E., Sheppard, L.J., Sverdrup, H., Haeuber, R., Hicks, W.K., 2014. Nitrogen deposition. In: Critical Loads and Biodiversity: Proceedings of the International Nitrogen Initiatives Workshop, Linking Experts of the Convention on Long-range Transboundary Air Pollution and the Convention on Biological Diversity. Springer, New York.

Taylor, P.G., Townsend, A.R., 2010. Stoichiometric control of organic carbon-nitrate relationships from soils to the sea. Nature 464, 1178—1181.

Templer, P.H., Mack, M.C., Chapin, F.S., Christenson, L.M., Compton, J.E., Crook, H.D., Currie, W.S., Curtis, C.J., Dail, D.B., D'Antonio, C.M., Emmett, B.A., Epstein, H.E., Goodale, C.L., Gundersen, P., Hobbie, S.E., Holland, K., Hooper, D.U., Hungate, B.A., Lamontagne, S., Nadelhoffer, K.J., Osenberg, C.W., Perakis, S.S., Schleppi, P., Schimel, J., Schmidt, I.K., Sommerkorn, M., Spoelstra, J., Tietema, A., Wessel, W.W., Zak, D.R., 2012. Sinks for nitrogen inputs in terrestrial ecosystems: a meta-analysis of enriched ^{15}N field tracer studies. Ecol. 93, 1816—1829.

Thomas, R.Q., Canham, C.D., Weathers, K.C., Goodale, C.L., 2010. Increased tree carbon storage in response to nitrogen deposition in the US. Nat. Geosci. 3, 13—17.

Treseder, K.K., 2004. A meta-analysis of mycorrhizal responses to nitrogen, phosphorus, and atmospheric CO2 in field studies. New Phytol. 164, 347—355.

Turlapati, S.A., Minocha, R., Bhiravarasa, P.S., Tisa, L.S., Thomas, W.K., Minocha, S.C., 2012. Chronic N-amended soils exhibit an altered bacterial community structure in Harvard Forest, MA, USA. FEMS Microbiol. Ecol. 83, 478—493.

van Breeman, N., Mulder, J., Driscoll, C.T., 1983. Acidification and alkalinization of soils. Plant Soil 75, 283—308.

van Diepen, L.T.A., Frey, S.D., Landis, E.A., Morrison, E.W., Pringle, A., 2017. Fungi exposed to chronic nitrogen enrichment are less able to decay leaf litter. Ecol. 98, 5—11.

Wallace, Z.P., Lovett, G.M., Hart, J.E., Machona, B., 2007. Effects of nitrogen saturation on tree growth and death in a mixed-oak forest. For. Ecol. Manage. 243, 210—218.

Wallenstein, M.D., McNulty, S., Fernandez, I.J., Boggs, J., Schlesinger, W.H., 2006. Nitrogen fertilization decreases forest soil fungal and bacterial biomass in three long-term experiments. For. Ecol. Manag. 222, 459—468.

Wang, Z., Fernandez, I., 1999. Soil type and forest vegetation influences on forest floor nitrogen dynamics at the Bear Brook Watershed in Maine (BBWM). Environ. Monit. Assess. 55, 221—234.

Warby, R.A.F., Johnson, C.E., Driscoll, C.T., 2009. Continuing acidification of organic soils across the northeastern USA: 1984-2001. Soil Sci. Soc. Am. J. 73, 274—284.

Warner, J.X., Dickerson, R.R., Wei, Z., Strow, L.L., Wang, Y., Liang, Q., 2017. Increased atmospheric ammonia over the world's major agricultural areas detected from space. Geophys. Res. Lett. 44, 2875—2884.

Wason, J.W., Dovciak, M., Beier, C.M., Battles, J.J., 2017. Tree growth is more sensitive than species distributions to recent changes in climate and acidic deposition in the northeastern United States. J. Appl. Ecol. 54, 1648—1657.

Watmough, S.A., Aherne, J., Alewell, C., Arp, P., Bailey, S., Clair, T., Dillon, P., Duchesne, L., Eimers, C., Fernandez, I., Foster, N., 2005. Sulphate, nitrogen and base cation budgets at 21 forested catchments in Canada, the United States and Europe. Environ. Monit. Assess. 109, 1—36.

Williard, K.W., DeWalle, D.R., Edwards, P.J., 2005. Influence of bedrock geology and tree species composition on stream nitrate concentrations in mid-Appalachian forested watersheds. Water Air Soil Pollut. 160, 55—76.

Wymore, A.S., Rodríguez-Cardona, B., McDowell, W.H., 2015. Direct response of dissolved organic nitrogen to nitrate availability in headwater streams. Biogeochemistry 126, 1—10.

Yanai, R.D., Vadeboncoeur, M.A., Hamburg, S.P., Arthur, M.A., Fuss, C.B., Groffinan, P.M., Siccama, T.G., Driscoll, C.T., 2013. From missing source to missing sink: long-term changes in the nitrogen budget of a Northern Hardwood forest. Environ. Sci. Technol. 47, 11440—11448.

Zak, D.R., Holmes, W.E., Burton, A.J., Pregitzer, K.S., Talhelm, A.F., 2008. Simulated atmospheric NO_3 deposition increases soil organic matter by slowing decomposition. Ecol. Appl. 18, 2016—2027.

Zak, D.R., Pregitzer, K.S., Burton, A.J., Edwards, I.P., Kellner, H., 2011. Microbial responses to a changing environment: implications for the future functioning of terrestrial ecosystems. Fung. Ecol. 4, 386—395.

Zhao, Y., Duan, L., Lei, Y., Xing, J., Nielsen, C.P., Hao, J., 2011. Will PM control undermine China's efforts to reduce soil acidification? Environ. Pollut. 159, 2726—2732.