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Spruce-fir forest changes during a 30-year nitrogen saturation experiment

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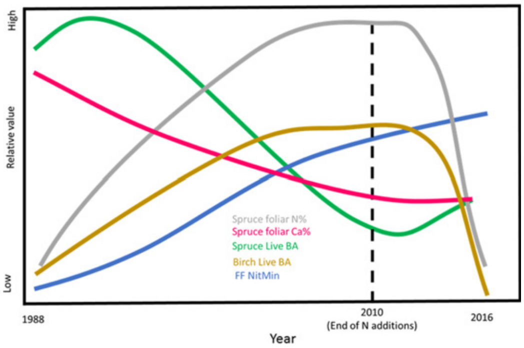
HIGHLIGHTS

- Indicators of nitrogen saturation were observed in plots having a total inorganic nitrogen input of ≥20 kg N ha^{-1} yr^{-1}.
- Control plots aggraded forest floor nitrogen at a rate equal to net N mineralization plus inorganic N deposition.
- Nitrogen treatment plots had a net loss in forest floor nitrogen.
- Drought, heat and thaw-freeze events in conjunction with nitrogen inputs likely contributed to forest decline.
- Despite reduced nitrogen deposition, climate change will make spruce-fir forest regeneration unlikely.

GRAPHICAL ABSTRACT



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ABSTRACT

A field experiment was established in a high elevation red spruce (*Picea rubens* Sarg.) – balsam fir (*Abies balsamea*) forest on Mount Ascutney Vermont, USA in 1988 to test the nitrogen (N) saturation hypothesis, and to better understand the mechanisms causing forest decline at the time. The study established replicate control, low and high dose nitrogen addition plots (i.e., 0, 15.7 and 31.4 kg NH_4Cl-N ha^{-1} yr^{-1}). The treatments began in 1988 and continued annually until 2010, but monitoring has continued to present. During the fertilization period, forest floor C:N, net *in situ* N mineralization, spruce foliar Ca%, and live spruce basal area decreased with increasing N addition, while foliar spruce N% and forest floor net nitrification increased with increasing N addition. The control plots aggraded forest floor N at a rate equal to the sum of the net *in situ* N mineralization plus average ambient deposition. Conversely, N addition plots lost forest floor N. Following the termination of N additions in 2010, the measured tree components returned to pre-treatment levels, but forest floor processes were slower to respond. During the 30 year study, site surface air temperature has increased by 0.5 °C per decade, and total N deposition has decreased 5.5 to 4.0 kg N ha^{-1} yr^{-1}. There have also been three significant drought years and at least one freeze injury year after which much of the forest mortality on the N addition plots occurred. Given that there was no control for the air temperature increase, discussion of the interactive impacts of climate and change and N addition is only subjective. Predicted changes in climate, N deposition and other stressors suggest that even in the absence of N saturation, regeneration of the spruce-fir ecosystem into the next century seems unlikely despite recent region-wide growth increases.

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1. Introduction

Large-scale forest decline was observed across parts of New England beginning in the 1960s, with much of the mortality located within the high elevation red spruce (*Picea rubens* Sarg.) and balsam fir (*Abies balsamea*) forests (Johnson, 1983). Researchers agreed that acidic atmospheric deposition of nitrous and sulfur oxides (i.e., $NO_{x,}$ and SO_x) were the primary cause of this decline, but the mechanisms for the mortality were uncertain (Pitelka and Raynal, 1989).

Agren and Bosatta (1988) and Aber et al. (1989) were the first to discuss the nitrogen (N) saturation hypothesis as a contributing cause of forest mortality. The theory proposes that under conditions of very high N availability (alone), adverse physiological and morphological changes could occur at the tree and forest level. The nitrogen saturation hypothesis proposed that the elevated chronically increased supply of plant available N would initially stimulate net primary productivity and foliar biomass, but both would reverse several years after the termination of N additions. Additionally, root biomass would decrease, while foliar N concentration and total tree N content would increase. At the ecosystem level, N saturation would increase net N mineralization, nitrification, N_2O emissions and NO_3 leaching. If N additions continued, the tree would progress through four N saturation stages that would ultimately lead to tree mortality and overall forest decline (Aber et al., 1989, 1998).

The N saturation hypothesis needed to be tested to prove (or disprove) the theory. The ultimate goal of the research was to minimize future forest decline through changes in forest management or regulatory constraints on pollutant emissions. Therefore, a pair of field studies were conducted to examine the nitrogen saturation. The objective was to first observe the initial stages of N saturation in real time by first locating high elevation red spruce and balsam fir forests across New England with similar soils and climate, but with different rates of N deposition. From 1987 to 1988, over 150 spruce-fir sites were sampled for foliar nitrogen, macro- and micronutrient concentration, forest floor N and carbon (C) concentration, and N mineralization and nitrification potential (McNulty et al., 1990, 1991). Several components of the nitrogen saturation hypothesis were well correlated with observed measurements including foliar and forest floor N concentration. The study also suggested that a forest floor N concentration of 1.4% strongly correlated with the onset of nitrification and N saturation in these highly acidic ($pH < 3.0$) soils (McNulty et al., 1991). Based on the regional survey, a field site located on Mt. Ascutney in southeastern Vermont was selected in 1988 to test the impact of chronic, low-dose, N additions in combination with and ambient atmospheric deposition.

Many studies have been conducted since the Mt. Ascutney study was initiated 30 years ago, particularly as N saturation relates to forest productivity and carbon sequestration. A fertilization study at Harvard Forest found that 20 years of N addition lead to lower fungal abundance and higher rates of lignin accumulation compared to non-fertilized plots in both deciduous and coniferous N additions (Frey et al., 2014). Globally, Tian et al. (2016) found that N additions of 5–6 g m^{-2} yr^{-1} created a threshold for severely reducing net primary productivity (NPP), while De Schrijver et al. (2008) estimated that 25% of European forests are N saturated. Other studies examined the relationship between N saturation and soil N mineralization and nitrification (Lovett and Goodale, 2011), nitrate leaching (Fenn et al., 1998), and relationships between root and foliar N isotopes and N saturation (Pardo et al., 2006, 2007).

However, very few N saturation studies have examined so many structural and functional variables over so long a period as was conducted on Mt. Ascutney. This paper brings together previous and current data as well as other studies to more fully explore the ecological and physiological changes that have occurred since 1988 on the Mt. Ascutney N saturation study plots. The paper also projects how high elevation New England forests may change in the coming years and decades under changing patterns of atmospheric deposition and climate.

2. Materials and methods

2.1. Study plots

After a scouting trip to the site in the fall of 1987, a long-term site in southeastern Vermont adjacent to the Mt. Ascutney State Park (43°26′ N, 72°27′ W) was chosen to test the N saturation hypothesis of forest decline in 1988 for several reasons. First, the initial sampling of red spruce and balsam fir foliar N concentration, and forest floor N concentration suggested that the Mt. Ascutney site had not yet begun to experience symptoms of N saturation. However, the site had elevated N levels compared to other spruce-fir forests further east (McNulty et al., 1991). Nitrogen deposition increases from east to west in New England (NADP, 2017) so placing the site just to the east of forests showing signs of N saturation should predispose the selected plots to develop the initial phase of N saturation with supplemental N additions. Second, Mt. Ascutney had a large area of red spruce (*Picea rubens*) – balsam fir (*Abies balsamea*) forest above 750 m elevation. Finally, the site was located within 200 m of an access road, allowing for convenient movement of equipment, fertilizer, samples and personnel.

Red spruce often grows in pockets surrounded by other deciduous species on Mt. Ascutney. Therefore, plot size needed to be relatively small to accommodate the replicate plots within the same slope and elevation. The establishment ten 15 × 15 m red spruce dominated plots occurred in June 1988 at elevations between 750 and 770 m. Initially, red spruce comprised >80% of the total basal area; with the remainder divided between balsam fir, mountain maple (*Acer spicatum*), red maple (*Acer rubrum*) and birch (*Betula* spp.) (McNulty and Aber, 1993). The location of study plots and treatments within the larger forest stand was randomly selected. Pre-treatment sampling found no statistically significant (paired *t*-test, $p \le 0.05$) differences in stand species composition, stand structure, net nitrification and net N mineralization of the forest floor or foliar chemistry among plots (McNulty et al., 1991; McNulty and Aber, 1993).

The site soil class is a Houghtonville, classified as frigid Typic Haplorthods located on broad areas between rock outcrops. These soils have 3 to 15% slopes and are well-drained (USDA Soil Conservation Service, 1989). Houghtonville soils have a discontinuous and narrow mineral layer. The mineral soil was not often present on flat surfaces but instead accumulated in rock crevasses. Tree root mass was almost exclusively located in the forest floor and not in this nutrient poor mineral horizon. For these reasons, only forest floor material was collected, and not the mineral horizon.

Climatically, the area has warm summers (average July air temperature 21 °C), and cold winters (average January air temperature −12 °C) (https://www.esrl.noaa.gov/gmd/ccgg/trends/index.html). Total annual precipitation was the only meteorological variable directly measured (1989 to 1991) on the site (McNulty and Aber, 1993).

On-site, bulk deposition inorganic N deposition measured on Mt. Ascutney averaged 5.4 kg N ha^{-1} yr^{-1} in open areas adjacent to the research plots from 1988 to 1991 (McNulty and Aber, 1993). Estimates total inorganic N deposition for Mt. Ascutney after 1991, were derived from the nearest National Atmospheric Deposition Program monitoring station (VT99) located approximately 75 km northwest of Mt. Ascutney. From 1988 to 2016, Station VT99 recorded total inorganic N deposition ranged from a high of 6.8 kg N ha^{-1} yr^{-1} (1990) to a low of 3.7 kg N ha^{-1} yr^{-1} (2009), with a mean of 5.0 kg N ha^{-1} yr^{-1} (NADP, 2017). These deposition values are consistent with the 5.4 kg N ha^{-1} yr^{-1} total inorganic N deposition values measured in open areas adjacent to the research plots from 1988 to 1991 (McNulty and Aber, 1993). The total inorganic N deposition values were not significantly different ($p = 0.48$) during the time of Mt. Ascutney and VT99 measurement overlap (i.e., 1988 to 1991). VT99 total inorganic N deposition has been declining at a rate of approximately 0.5 kg N ha-1 yr-1 during the experiment (i.e., 5.5 to 4.0 kg N ha^{-1} yr^{-1} from 1988 to 2016).

2.2. Nitrogen additions

Nitrogen deposition increases from east to west across New England (NADP, 2017). The Mt. Ascutney plots are located slightly east of the area of N saturated forests (McNulty et al., 1990, 1991), and the plots did not exhibit any of the early symptoms of N saturation as outlined by Aber et al. (1989) when initially established. A pair of controls plots received no N-fertilizer. Two other plot pairs were randomly selected and amended with two different levels of ammonium chloride (NH_4Cl) during June, July, and August from 1988 to 2010 (Table 1). The fertilizer was divided into three increments to minimize the potential for N leaching from the forest floor (Johnson, 1992).

By combining the long-term inputs of anthropogenic N deposition with N-fertilizer at a rate equal to high levels of N deposition found elsewhere (e.g., 30 to 40 kg N ha^{-1} yr^{-1}, Grennfelt and Hultberg, 1986), the study attempted to induce N saturation on these plots. Ammonium chloride was used as the N fertilizer to avoid including sulfur (e.g., $(NH_4)_2SO_4$) that would interfere with testing whether or not N alone could cause the observed decline forest decline observed across the region. Other plots not presented in this paper received either $NaNO_3$ or a combination of $NaNO_3$ and NH_4Cl.

The trend in reduced N deposition (NADP, 2017) and consideration by the Environmental Protection Agency (EPA) to further limit atmospheric N emissions (US EPA, 2010) lead to the decision to discontinue on all treatment plot N additions in 2010 to observe forest recovery response.

2.3. Reoccurring measurements and other studies

The combination of small plot size and the long-term nature of the research could negatively impact plot integrity if sampling were too frequent. Plots were intensively measured at the start of the experiment (i.e., 1987–1991) to establish baseline conditions and suggest trends, but were then converted to a four-year sampling cycle with the limited number of base ecosystem measurements detailed below and (Table 2). Many other studies have periodically assessed changes on Mt. Ascutney (McNulty et al., 2005). Measurements included; trace gases (Castro et al., 1993), tree physiology (Perkins et al., 2000; Schaberg et al., 1997; Schaberg et al., 2002), root morphology (Rustad unpublished), near ground and forest floor temperatures (Boggs and McNulty, 2010), forest floor ion exchange bags (McNulty and Aber, 1993), and soil respiration (Wallenstein et al., 2006). These were self-contained studies and provided additional understanding of tree and ecosystem response to long-term N additions, but were not part of the core project. Therefore, this paper does not provide the complete methodology for each of those studies.

2.4. Total live basal area change

Basal area (BA), tree condition (i.e., living vs. dead) and species type were recorded for all trees >2.54 cm at 1.5 m above the forest floor for each plot before the start on N additions in 1988. Basal area was re-measured eight times between 1990 and 2016 (Table 2).

2.5. Forest floor C and N concentrations and macro-nutrients

Before the first N addition in June 1988, 20 forest floor samples, each with an average area of 150 cm^3 were collected from each plot and randomly composited into five samples per plot. A 5 × 5-mm mesh screen removed all twigs, coarse roots, and stones from the samples. Carbon and N were measured in these samples using a Perkin-Elmer model 240B CHN analyzer. The total elemental analysis that included calcium (Ca), magnesium (Mg), and aluminum (Al) was determined by mixing the forest floor samples with nitric acid and digesting them in a microwave. Additional deionized water brought the digest to volume, and the cations of interest were determined simultaneously on an Inductively Coupled Plasma Spectrograph (Kissel and Sonon, 2008). Seven additional forest floor surveys were completed during the study (Table 2) and analyzed for C and N concentration and macro-nutrients.

2.6. Forest floor mass and total C and N

As frigid Typic Haplorthods located on a granite bedrock, mineral soils of Mt. Ascutney are highly variable. Often non-existent, the A, B, and C horizons can also be either thin (i.e., <0.5 cm in depth) or (rarely) much thicker (>10 cm in depth) between rock pockets. Therefore, sample collection included only the O_e and O_a forest floor layers.

To measure forest floor mass, a 1 m^2 grid was then placed on the 15 × 15 m^2 plot. Researchers recorded forest floor depth at each grid corner with (exposed rock entered as 0). Bulk density and C and N concentrations from collected forest floor N mineralization samples were used to calculate total forest floor C and N content on a per hectare basis. Forest floor mass and C and N content were determined two times during the study (Table 2).

2.7. In situ, annual N mineralization, and nitrification

Forest floor samples were collected monthly from each plot (except over-winter samples that were incubated in situ from October to May). Twenty samples from each plot were randomly combined into five samples per plot and sieved through a No. 18 mesh screen (1 mm) to remove twigs, coarse roots, and stones. Combining samples ensured sufficient sample mass for analysis. Technicians placed approximately 10 g of each of the five composite samples from each plot into 150 ml of 1 M KC1 for 48 h. After 48 h, 20 ml of solution extract was removed and frozen at 0 °C until analyzed for NH_4-N and NO_3-N concentrations. The remaining 20 samples from each plot were placed into 1-ml (2.54 × 10–4 mm) thick polyethylene bags, replaced into the forest floor and incubated *in situ* for 28 days. The 20 incubated samples were then composite, sieved, and extracted as previously described. Initial and incubated extracts were run on a TRAACS 800 Auto-analyzer to determine NH_4-N (Technicon Industrial Systems, 1978) and NO_3-N (Technicon Industrial Systems, 1977) concentrations. Net annual N mineralization equaled the sum of monthly incubated sample (NH_4-N + NO_3-N) minus initial monthly sample (NH_4-N + NO_3-N). Net annual nitrification equaled the accumulation of monthly incubated sample NO_3-N minus initial monthly sample minus NO_3-N. *In situ* N mineralization and nitrification measurements were collected for six separate years during the study (Table 2).

Table 1
Year of plot establishment and treatment additions from 1988 to 2010, and total inorganic N deposition for the Mt. Ascutney, VT spruce-fir experimental plots.

Plot number	Monthly amendment (NH_4Cl-N kg ha^{-1})	Total N addition (kg ha^{-1} yr^{-1})	Average total inorganic N deposition (kg ha^{-1} yr^{-1})	Total N (kg ha^{-1} yr^{-1})
1,6	0	0	5.0	5.0
4, 10	5.2	15.7	5.0	20.7
3,7	10.5	31.4	5.0	36.4

Table 2
Year and measurement from Mt. Ascutney plots. *Pretreatment measurements.

Sampling	Year collected									
	1988*	1990	1994	1998	2000	2002	2006	2010	2012	2016
Total live basal area change	X	X	X	X		X	X	X	X	X
Foliar chemistry	X	X	X	X		X	X	X	X	X
Forest floor C and N concentrations and macro-nutrients	X		X	X	X		X	X	X	X
Forest floor mass and total C and N	X						X			
In situ, annual, N mineralization and nitrification	X	X				X	X	X	X	
Potential N mineralization and nitrification	X		X		X	X	X		X	X
Species regeneration							X	X	X	X

2.8. Potential N mineralization and nitrification

Before treatment additions in May 1988, and in July of six additional years (Table 2), 40 forest floor samples were collected and sieved as previously described for in situ N mineralization and nitrification. Technicians composited the forest floor and placed half of the samples in KC1 for 48 h, extracted, and measured on the auto-analyzer to quantify initial NO_3-N and NH_4-N concentration, while the other half of the sample was incubated at 22 °C for 28 days. After 28 days the samples were composited as previously described, placed in KC1 for 48 h, extracted, and run on an auto-analyzer to measure incubated NO_3-N and NH_4-N concentration.

2.9. Foliar chemistry

Six dominant red spruce trees were randomly tagged on each plot at the beginning of the study. In July 1988, and in July of seven other years (Table 2) during the experiment, three randomly oriented, sunlit branch samples, each containing needles from all age classes were removed from all tagged tree using a pruning pole. The clipped branches from each tree were put into a paper bag and dried at 70 °C for 48 h. All needles had fallen into the bag after drying. The contents of the bag were shaken to homogenize the age classes, and technicians randomly selected sub-samples to sieve through a 2 × 2-mm mesh screen to remove twigs and detritus, and then ground through a No. 10 mesh using a Wiley Mill. The samples were placed in 60-ml acid-washed glass jars, dried for 24 h at 70 °C, and stored in darkness at 22 °C. Foliar cation percentages were measured using the Dry Ash procedure where 2.5 g of leaf tissue was heated to 500 °C overnight. The samples were then dehydrated with 6 N HCl and then brought to volume (50 ml) with distilled water in a volumetric flask. The analysis for Ca, Mg, and Al were conducted using an Ion Coupled Plasma Analyzer. Quality assurance and quality control practices used Standards that spanned the range of foliar cation concentrations. Foliar percent N and C were measured using a Perkin-Elmer Model 240B CHN analyzer. Technicians used National Institute of Standards and Technology (NIST) protocols that included National Bureau of Standards (NBS) 1572 (citrus leaves) and NBS 1567 (wheat flour) standards. If one of the sample trees died during the experiment, another tree would be randomly selected to avoid a loss of sample size over time.

2.10. Statistical analysis

All data analyses were completed using JMP Pro 12 (JMP, 2012) and treatment values were derived from plot level samples (i.e., $n = 6$ for foliage and $n = 5$ for forest floor). JMP Pro 12 analyzed year versus year within treatment, and treatment versus treatment within a year relationships with the total live basal area, forest floor, and foliar chemistry, and nutrient cycling using a paired *t*-test. The *t*-test was selected, and the significance level was set to$\alpha \leq 0.05$ to determine which treatments ($n = 2$) were statistically different from each other. A tight grouping (i.e., small intra-treatment variability) of foliar or forest floor chemistry within treatments could indicate that N fertilization is a major controlling factor for foliar and forest floor chemistry. A broad grouping (i.e., high intra-treatment variability) of foliar or forest floor chemistry within treatments could indicate that other non-N fertilization factors are controlling intra-treatment chemistry. Statistical relationships and standard errors trends are presented in the Results and Discussion section.

3. Results and discussion

3.1. N deposition and climate variability

Average precipitation during that period was averaged 1015 mm yr^{-1} (25 mm yr^{-1}, standard error (se) in parenthesis), with 70% of precipitation falling as snow or ice (McNulty and Aber, 1993). Between 1988 and 2016, PRISM (2015) provided estimates of site level average annual air temperature and total annual precipitation. Average annual precipitation varied between 880 and 1472 mm yr^{-1} and averaged 1122.6 mm yr^{-1} (30.8) mm yr^{-1} between 1988 and 2016. There was not a significant trend in total annual precipitation over time ($p = 0.08$, Fig. 1a). From 1988 to 2016, mean annual air temperature varied from 5.2 to 8.9 °C, with a mean value of 6.8 °C (0.2 °C). During the study, ambient global atmospheric CO_2 concentrations increased from 348 to 406 ppm (16%) (Mauna Loa station, https://www.esrl.noaa.gov/gmd/ccgg/trends/full.html). Although not considered at the beginning of the experiment, the local air temperature has changed significantly from 1987 to 2016. Average annual air temperature increased at a rate 0.5 °C per decade ($p = 0.0005$, Fig. 1b).

The 1990 Clean Air Act significantly reduced atmospheric SO_x and NO_x emissions and deposition across much of New England since the start of the experiment in 1988. Nationally, total inorganic N deposition peaked in 1995 and had since been declining (Du, 2016). Mt. Ascutney was consistent with this trend. Nitrogen deposition fell at a rate of approximately 0.5 kg N ha^{-1} yr^{-1} (i.e., 5.5 to 4.0 kg N ha^{-1} yr^{-1}) from 1988 to 2016.

Given that there was no control for increasing air temperature (i.e., all of the sites warmed), there is no way to determine if (or how much) of the structural and functional changes observed on the sites were caused by climate, changes in N inputs, or a combination thereof. Also, aside from 1988 to 1991, the four-year measurement cycle did not allow for examination of intra- or inter-annual climate variability impacts.

3.2. Basal area

Chronic N additions altered many ecosystem components associated with stand structure and function compared to the control plots, but the changes in total live BA were the most pronounced (Table 3). Live and dead standing spruce BA increased significantly (i.e., 21% and 2100%), respectively, between 1988 and 2016 in the control plots. The amount of dead standing spruce BA represented 1% of the spruce volume in 1988, and 17% of the volume in 2016 (Table 3). Much of this loss occurred through self-thinning as the average stem density declined from 4778 live spruce ha^{-1} in 1988 to 2711 live spruce ha^{-1} in 2016.

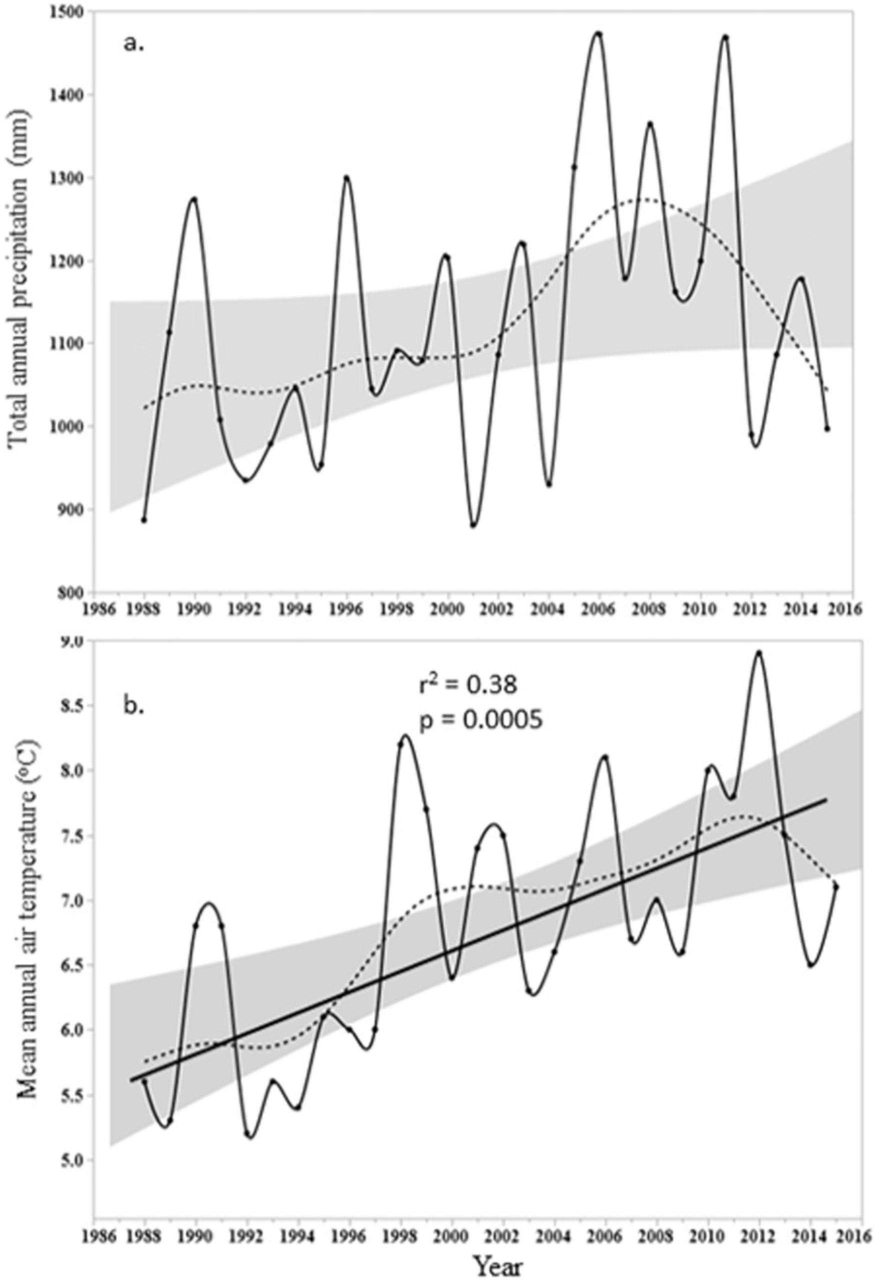

Fig. 1. Estimates of (a) total annual precipitation (mm) and (b) mean annual air temperature (°C) on Mount Ascutney, VT (43°26′ N, 72°27′ W) from 1988 to 2015 (PRISM, 2015). The thick black line represents the linear regression and the dotted line represents the trend when year to year variability is smoothed out. The shaded area depicts 95% confidence interval for the trend.

The changes in spruce BA on the treatment plots were even larger as the low and high N addition plots had a reduction in live spruce BA of 35% and 47%, respectively, between 1988 and 2016. Following the termination of added N in 2010, total live BA increased significantly in the high treatment N plot and was 32% larger in 2016 compared to the BA measured in 2010 (Table 3). Were it not for the cessation of N additions in 2010; there was no indication that the high N addition plots would not have continued to lose living basal area over time. In contrast, low N addition plot total live BA showed no change following the termination of N additions and stabilized at 22.1 m^2 ha^{-1}.

The fraction of dead standing spruce increased in low and high N addition plots, with a 1400% and 1800% increase between 1988 and 2016, respectively. Although both the control and treatments had increases in dead spruce BA, the plots were structurally very different. Control plots maintained canopy closure throughout the experiment, and spruce mortality appeared to be due to self-thinning. Conversely, the high N addition plots had gap opening within the first five years of treatment, and the low N addition plot gaps followed a few years later.

Engel et al. (2016) noted an increase in red spruce basal area growth since the start of the 21st century in a gradient study across New England. While spruce growth changes were not a focus of this study, the sum of live plus dead basal area change between remeasurement periods provides some indication of changes in site productivity. There were no statistically significant changes in spruce BA growth rates from 2002 to 2016 on the control plots.

The three most severe droughts since study site establishment occurred in 2001, 2004 and 2012 (Kunkel et al., 2013a; Fig. 1), along with a severe winter injury event in 2003 (Lazarus et al., 2004). In both instances, the low N addition plots experienced a decrease in total live BA during the following measurement period (i.e., 2006 for

Table 3
Live and dead red spruce, balsam fir, birch spp., maple spp., and total basal area on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 1988 to 2016. Means with the same letter are not significantly different, year versus year within treatment (lowercase) and treatment versus treatment within year (uppercase). *Total includes maple spp. Maples are not listed in the table because they represent a small part of the basal area.

Treatment $kg\ N\ ha^{-1}\ yr^{-1}$	Year	Live Basal Area				Dead Basal Area			
		Red spruce	Balsam fir	Birch spp.	*Total	Red spruce	Balsam fir	Birch spp.	*Total
		$m^2\ ha^{-1}$				$m^2\ ha^{-1}$			
Control	1988	31.2 (1.6)bA	3.7 (2.4)abA	1.9 (0.2)aB	36.7 (0.7)aA	0.4 (0.1)dA	0.8 (0.8)aA	0.6 (0.6)aA	1.7 (0.3)dA
	1990	31.7 (1.0)bB	3.2 (1.3)abA	2.0 (0.2)aA	36.8 (0.3)aA	0.6 (0.2)dB	1.0 (1.0)aA	0.8 (0.2)aA	2.3 (0.6)dAB
	1994	33.0 (0.2)bA	3.9 (1.9)aA	2.2 (0.7)aA	39.0 (1.0)aA	0.9 (0.4)cdA	1.5 (1.1)aA	0.4 (0.1)aA	2.8 (0.8)dA
	1998	34.5 (4.2)abA	2.5 (0.4)abA	1.3 (0.5)abA	38.3 (4.4)aA	1.0 (0.3)cdB	1.0 (0.4)aA	0.7 (0.1)aA	2.6 (0.2)cdB
	2002	37.5 (2.9)abA	1.7 (0.7)abA	1.4 (1.0)abA	40.5 (3.1)aA	2.9 (1.3)bcB	0.3 (0.3)aA	0.5 (0.3)aA	3.6 (1.3)cdB
	2006	43.8 (7.9)aA	0.4 (0.4)abA	0.7 (0.7)abA	44.8 (8.9)aA	2.0 (0.0)cdA	0.0 (0.0)aA	0.4 (0.4)aA	2.4 (0.4)dA
	2010	38 (1.3)abA	0.5 (0.5)abA	0.9 (0.4)abA	39.4 (2.2)aA	4.6 (1.5)bA	0.0 (0.0)aA	1.2 (0.4)aA	5.7 (1.9)bcB
	2012	37.7 (2.7)abA	1.2 (0.2)abA	0.6 (0.0)abA	39.5 (2.6)aA	4.6 (0.1)bA	1.5 (0.9)aA	1.2 (0.4)aA	7.3 (1.2)abA
	2016	37.8 (0.6)abA	0.2 (0.2)bA	0.0 (0.0)bA	37.9 (0.4)aA	8.3 (0.1)aA	0.4 (0.4)aA	1.1 (0.2)aA	9.8 (0.6)aA
15.7	1988	34.1 (3.3)abA	1.1 (1.1)aA	2.8 (0.3)abAB	38.8 (5.0)abA	1.0 (0.6)dA	1.1 (0.6)aA	0.7 (0.3)bA	2.7 (0.8)dA
	1990	35.7 (1.0)aA	1.7 (1.7)aA	3.1 (0.2)aA	40.9 (3.3)aA	2.3 (0.1)dA	0.8 (0.3)aA	0.7 (0.3)bA	3.8 (0.1)dA
	1994	34.6 (3.2)abA	1.8 (1.8)aA	4.4 (1.6)aA	41.0 (3.7)aA	3.2 (2.9)dA	1.1 (0.7)aA	1.0 (0.8)abA	5.8 (2.5)dA
	1998	30.1 (2.2)abAB	0.3 (0.3)aA	3.1 (0.2)aA	33.4 (2.2)abAB	8.9 (1.1)cA	1.9 (1.4)aA	1.1 (0.1)abA	11.9 (2.5)cA
	2002	28.4 (3.0)abAB	0.3 (0.3)aA	2.8 (0.4)abA	31.7 (3.9)abAB	13.4 (2.0)bcA	0.5 (0.5)abA	0.7 (0.3)bA	15 (2.7)bcA
	2006	22.2 (6.0)abAB	0.1 (0.1)aA	2.9 (1.0)abA	25.1 (6.8)abAB	19.5 (1.5)aA	0.0 (0.0)aA	1.5 (1.2)abA	20.9 (2.6)aA
	2010	21.5 (3.8)bB	0.4 (0.4)aA	0.9 (0.3)bcA	23.0 (4.7)bB	16.8 (1.7)abA	0.6 (0.6)aA	2.3 (0.3)abA	19.6 (1.5)abA
	2012	25.2 (4.5)abAB	0.4 (0.4)aA	1.0 (0.4)bcA	26.9 (5.6)abAB	13.7 (0.1)bcA	0.1 (0.1)aA	2.5 (0.4)aA	16.2 (0.4)abcA
	2016	22.1 (7.5)bA	0.5 (0.5)aA	0.0 (0.0)cA	22.9 (8.3)bA	14.2 (1.6)bA	0.0 (0.0)aA	0.8 (0.1)bA	15.0 (1.5)bcA
31.4	1988	32.1 (0.4)aA	0.9 (0.3)aA	3.1 (0.3)aA	36.1 (0.0)aA	0.5 (0.1)bA	0.4 (0.4)abA	0.5 (0.2)aA	1.3 (0.7)cA
	1990	32.9 (0.5)aAB	0.9 (0.2)aA	3.0 (0.4)aA	36.8 (1.0)aA	0.5 (0.4)bB	0.1 (0.1)bA	0.7 (0.3)aA	1.3 (0.6)cB
	1994	30.3 (0.4)aA	0.4 (0.4)aA	3.1 (0.2)aA	33.7 (0.9)aA	5.2 (4.8)abA	0.5 (0.5)abA	1.0 (0.8)aA	6.8 (3.5)bcA
	1998	19.9 (0.9)bB	0.1 (0.1)aB	2.1 (0.5)aA	22.0 (0.5)bB	12.6 (1.1)abA	1.1 (0.3)aA	0.9 (0.4)aA	14.6 (0.4)abA
	2002	18.2 (3.5)bcB	1.5 (1.5)aA	2.2 (1.0)aA	21.8 (4.0)bB	14.4 (1.6)aA	0.3 (0.3)abA	0.9 (0.5)aA	15.6 (1.4)abA
	2006	13.7 (0.0)cB	0.3 (0.3)aA	0.0 (0.0)bA	14.0 (0.3)cB	17.7 (7.5)aA	0.0 (0.0)bA	2.2 (0.5)aA	19.8 (7.9)aA
	2010	13.8 (2.0)cB	0.4 (0.4)aA	0.4 (0.4)bA	14.5 (2.0)cB	13.6 (4.8)abA	0.0 (0.0)bA	1.7 (0.3)aA	15.3 (4.5)abAB
	2012	15.1 (2.3)bcB	0.8 (0.5)aA	0.0 (0.0)bA	15.8 (1.8)cB	8.7 (4.9)abA	0.1 (0.1)bA	1.5 (0.8)aA	10.2 (4.1)abcA
	2016	18.2 (0.9)bcA	0.8 (0.8)aA	0.0 (0.0)bA	19.0 (0.1)bcA	9.1 (4.8)abA	0.0 (0.0)bA	1.7 (0.6)aA	10.9 (5.5)abcA

the 2004 drought and 2003 freeze-thaw event, and 2016 for the 2012 drought, Table 3). Fenn et al. (1998) postulated that decreased fine root mass would be a characteristic of N saturation. Preliminary root ingrowth studies on the Mt. Ascutney plots support the hypothesis of reduced root mass on the high N treatments compared to the controls (Rustad, unpublished data). Wallenstein et al. (2006) observed a slight decreased fungal:bacterial activity ratio in the high N addition treatment plots. Reductions in mycorrhizal biomass could negatively impact both tree water and nutrient uptake. However, in the high N addition plots remeasurements after the 2012 drought, there was a net increase in total live BA. Reductions in stand density reduce water stress and mortality risk during drought periods (Zhang et al., 2017). Previous reductions in total live BA within the high N treatment plots may have been enough that competition for the reduced soil water was not sufficient to cause further tree mortality during the drought period. Additionally, the high N addition plots could be responding favorably to the cessation of N fertilization in 2010.

Aside from red spruce, balsam fir was the next most abundant tree species in the control plots at the beginning of the study (comprising 11% of the total BA). However, by 2016, fir was virtually gone from the control plots but persisted on both the low and high N addition plots.

Birch species (*Betula alleghaniensis*, and *Betula papyrifera*) comprised between 6 and 9% of the total BA across all plots at the beginning of the experiment. There was a gradual decline of birch on the control plots with a shift toward increased mortality following the 2006 sampling. By 2016, birch species with a DBH >2.5 cm were all dead in the control plots (Table 3). On the low N addition plots, total live birch BA peaked in 1994 (i.e., 4.4 $m^2\ ha^{-1}$), and then began to decline, a pattern similar to the control plots. On the low N addition plots the amount of dead birch BA almost doubled between the 2006 and 2010 sampling (Table 3), and by 2016, only dead birch was present. On the high N addition plots, live birch declined quickly, and there was no live birch during the 2006 sampling period (Table 3). On the high N addition plots during the 2010 sampling, a few of live birch were observed, and BA did not change significantly in 2012 or 2016. In this study, there were no physiological measurements on the birch from these plots, but other investigators have examined the role of the rapid frost freeze cycle on cellular damage (including increased water loss) and birch mortality (Cox and Zhu, 2003; Bourque et al., 2005). Schaberg et al. (1997, 2002) well characterized the physiological response of red spruce to freezing and thawing on Ascutney plots. These same destructive physical changes to cell structure (i.e., cellular damage) and function (i.e., increased water loss) could also be impacting the birch. However, given that the decline also occurred on the control plots (that themselves are slowly, naturally aggrading in total N), the birch mortality may have as much or more to do with climate than with N deposition.

3.3. Forest floor mass, C, N and selected macro-nutrients

From 1987 until 2006, average forest floor mass changed from 68.0 to 81.2 Mt ha^{-1} on the control plots and from 89.7 to 76.8 Mt ha^{-1} and 96.0 to 77.5 Mt ha^{-1} on the low and high N additions respectively. However, the differences were not statistically significant due to the low number of treatment replicates (i.e., $n = 2$), and large site variability.

Differences in initial forest floor mass across treatments also made statistically significant changes in total N and C associated with N treatments difficult to detect. However, when normalized using either a percentage or ratio term, N treatment impacts become significant. Forest floor N% increased on both treatments and in the control plots over time. On the control plots, forest floor N concentration changed from 1.22% (0.22) in 1988 at the beginning of the experiment to a high of 1.64% (0.04) in 2010 but then began to decrease to 1.36% (0.04) by 2016 after N additions were terminated. Both the low and high N treatments exhibited a similar pattern for forest floor N% with both peaking in 2010 at approximately 1.8% and then declining back to 1.5 N% by 2016 (Table 4).

Annual changes in the forest floor N budget were estimated by multiplying total forest floor mass per ha by average N%, then

Table 4
Forest floor chemistry (%) on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 1988 to 2016. Standard error is in parenthesis. Means with the same letter are not significantly different, year versus year within treatment (lowercase) and treatment versus treatment within year (uppercase).

Treatment (kg N ha^{-1} yr^{-1})	1988	1994	1998	2000	2006	2010	2012	2016
Nitrogen								
0.0	1.22 (0.22)bA	1.36 (0.11)abA	1.5 (0.03)abA	1.56 (0.07)aA	1.50 (0.05)abA	1.64 (0.04)aA	1.48 (0.03)abA	1.36 (0.04)abA
15.7	1.47 (0.01)cdA	1.47 (0.06)cdA	1.5 (0.04)cdA	1.65 (0.02)abcA	1.43 (0.06)dA	1.81 (0.13)aA	1.71 (0.07)abA	1.55 (0.05)bcdA
31.4	1.30 (0.06)cA	1.40 (0.08)bcA	1.49 (0.05)abcA	1.65 (0.06)abcA	1.46 (0.08)abcA	1.79 (0.03)aA	1.66 (0.25)abA	1.54 (0.10)abcA
Carbon								
0.0	35.7 (6.1)bA	43.5 (2.7)abA	43.8 (1.7)abA	45.2 (1.4)aA	43.7 (3.2)abA	51.6 (0.5)aA	46.4 (2.3)aA	46.8 (0.9)aA
15.7	42.9 (0.4)bA	44 (1.7)abA	42.9 (0.4)bA	46.4 (0.6)abA	36.9 (0.3)cA	47.7 (2.4)aA	46 (1.7)abA	46 (0.1)abA
31.4	38.9 (0.2)aA	41.6 (2.7)aA	42.0 (1.5)aA	44.2 (0.3)aA	38.2 (2.6)aA	44.6 (1.3)aA	44.8 (4.1)aA	44.5 (2.0)aA
Calcium								
0.0	0.07 (0.02)bA	0.07 (0.02)bB	0.07 (0.02)bA	0.09 (0.00)abA	–	0.13 (0.02)aA	0.10 (0.01)abA	0.06 (0.01)bA
15.7	0.10 (0.06)aA	0.08 (0.02)aAB	0.12 (0.02)aA	0.16 (0.04)aA	–	0.13 (0.03)aA	0.13 (0.04)aA	0.07 (0.01)aA
31.4	0.10 (0.02)bcA	0.19 (0.04)aA	0.14 (0.03)abcA	0.16 (0.01)abA	–	0.09 (0.01)bcA	0.10 (0.02)bcA	0.08 (0.01)cA
Magnesium								
0.0	0.030 (0.005)aA	0.014 (0.002)bA	0.029 (0.001)abA	0.036 (0.001)aA	–	0.030 (0.002)abA	0.037 (0.011)aA	0.025 (0.000)abA
15.7	0.039 (0.004)aA	0.024 (0.008)bcA	0.029 (0.002)abcA	0.035 (0.003)abA	–	0.023 (0.003)cA	0.021 (0.002)cA	0.019 (0.003)cA
31.4	0.040 (0.007)aA	0.017 (0.002)dA	0.032 (0.002)abcA	0.036 (0.002)abA	–	0.022 (0.001)bcdA	0.019 (0.002)dA	0.023 (0.001)cdA
Aluminum								
0.0	0.276 (0.03)aA	0.146 (0.01)bA	0.258 (0.01)aA	0.272 (0.04)aA	–	0.157 (0.01)bA	0.155 (0.03)bA	0.173 (0.02)bA
15.7	0.324 (0.03)aA	0.126 (0.02)dA	0.248 (0.03)bA	0.217 (0.03)bcA	–	0.149 (0.00)cdA	0.146 (0.01)cdA	0.181 (0.02)bcdA
31.4	0.376 (0.02)aA	0.143 (0.02)dA	0.298 (0.06)abA	0.284 (0.01)abcA	–	0.202 (0.03)cdA	0.148 (0.01)dA	0.208 (0.00)bcdA
Carbon/nitrogen								
0.0	29.4 (0.1)bA	32.1 (0.8)abA	29.3 (0.7)bA	29.1 (0.5)bA	29.3 (1.3)aA	31.6 (1.0)abA	31.5 (1.1)bA	34.6 (1.5)aA
15.7	29.2 (0.4)aA	29.9 (0.1)aB	28.7 (1.3)abA	28.3 (0.9)abcA	26.0 (0.8)dA	26.4 (0.6)cdB	27.0 (0.1)bcdA	29.9 (0.7)aB
31.4	30.0 (0.9)aA	29.7 (0.4)abB	28.3 (0.2)abA	26.9 (1.5)abcA	26.8 (0.9)bcA	24.9 (0.3)cB	27.7 (2.0)abcA	29.0 (0.6)abB
Nitrogen/magnesium								
0.0	40.6 (0.4)cA	99.4 (8.7)aA	52.3 (2.3)bcA	43.6 (2.7)bcA	–	60.5 (0.7)bA	43.5 (11.8)bcB	53.8 (1.3)bcA
15.7	38.7 (3.8)cA	69.1 (25.3)abcA	51.7 (4.2)abcA	47.1 (4.3)bcA	–	88.6 (4.7)aA	81.4 (5.6)abA	83.3 (10.5)abA
31.4	34.3 (7.2)dA	84.5 (4.7)abA	46.4 (2.0)bcA	46.4 (3.8)cdA	–	67.0 (11.7)abcA	87.7 (2.8)aA	65.8 (5.8)bcA
Calcium/aluminum								
0.0	0.26 (0.04)cA	0.48 (0.13)bcB	0.29 (0.09)cA	0.36 (0.07)cA	–	0.87 (0.12)aA	0.76 (0.05)abA	0.40 (0.12)cA
15.7	0.35 (0.20)aA	0.74 (0.19)aB	0.5 (0.12)aA	0.79 (0.32)aA	–	0.91 (0.25)aA	0.92 (0.33)aA	0.39 (0.03)aA
31.4	0.28 (0.04)cA	1.41 (0.08)aA	0.58 (0.21)bcA	0.57 (0.06)bcA	–	0.54 (0.07)bcA	0.69 (0.15)bA	0.37 (0.03)bcA

– no data.

subtracting the sampling year total N from the 1988 (pretreatment) N, and dividing by the number of years between sampling. The control plots were aggrading forest floor N at a rate of 20.4 kg N ha yr^{-1}. This rate is nearly equal to the total average inorganic N deposition (i.e., 5.0 kg N ha^{-1} yr^{-1}) plus average annual net N mineralization (i.e., 17.2 kg N ha^{-1} yr^{-1}) on the control sites. Conversely, the high N addition plots had a net loss of N at a rate of 3.0 kg N ha yr^{-1} despite the addition of N to those plots.

The control plot forest floor C% changed from a low of 35.7% (6.1%) in 1988, to 51.6% (0.5%) in 2010, before declining to 46.8% (0.9%) in 2016 (Table 4). Forest floor carbon % changed less in the two treatments, with the low N plots ranging from 36.9% (0.3%) in 2006 to 47.7% (2.4%) in 2010. The forest floor C% of the high N plots ranged from a low of 38.2% (2.6%) in 2006 to a high of 44.8% (4.1%) C in 2012.

Formation of canopy gaps leading to patches of increased soil temperature (Ritter et al., 2005) and associated rates of forest floor decomposition (Kirschbaum, 1995), along with changes in above- and belowground detrital inputs, may have contributed to increased forest floor N% and C:N variability. Additionally, if dead trees blow over as opposed to breaking off along the stem tip-up mounds can be formed. This study did not characterize either gap dynamics or tip-up mounds so it is not possible to determine if increases in forest floor percent C and N variability may also be a function of increased soil disturbance.

Although individually, the forest floor C and N percentages were often not statically significantly different between treatments and across years, there were small, opposite directional changes such that the forest floor C:N that were significantly different across treatments (Table 4). The control plot forest floor C:N was relatively constant between 1988 and 2006, before increasing from 2010 to 2016 (Table 4). Over the course of the study, the forest floor C:N was significantly higher in the controls compared to the treatments in three out of eight measurement periods (i.e., 1994, 2010, and 2016). Conversely, the forest floor C:N of both treatment plots decreased until N additions ended in 2010, after which time, forest floor C:N began to rise (Table 4).

3.4. Forest floor N mineralization and nitrification

Within the control plots, rates of *in situ* net N mineralization did not change significantly during the period of study, ranging from 13.7 to 26.3 kg N ha^{-1} yr^{-1} (Fig. 2). *In situ* net N mineralization was more variable in both the high and low N addition plots compared to the control plots. Therefore, while the average annual net N mineralization values

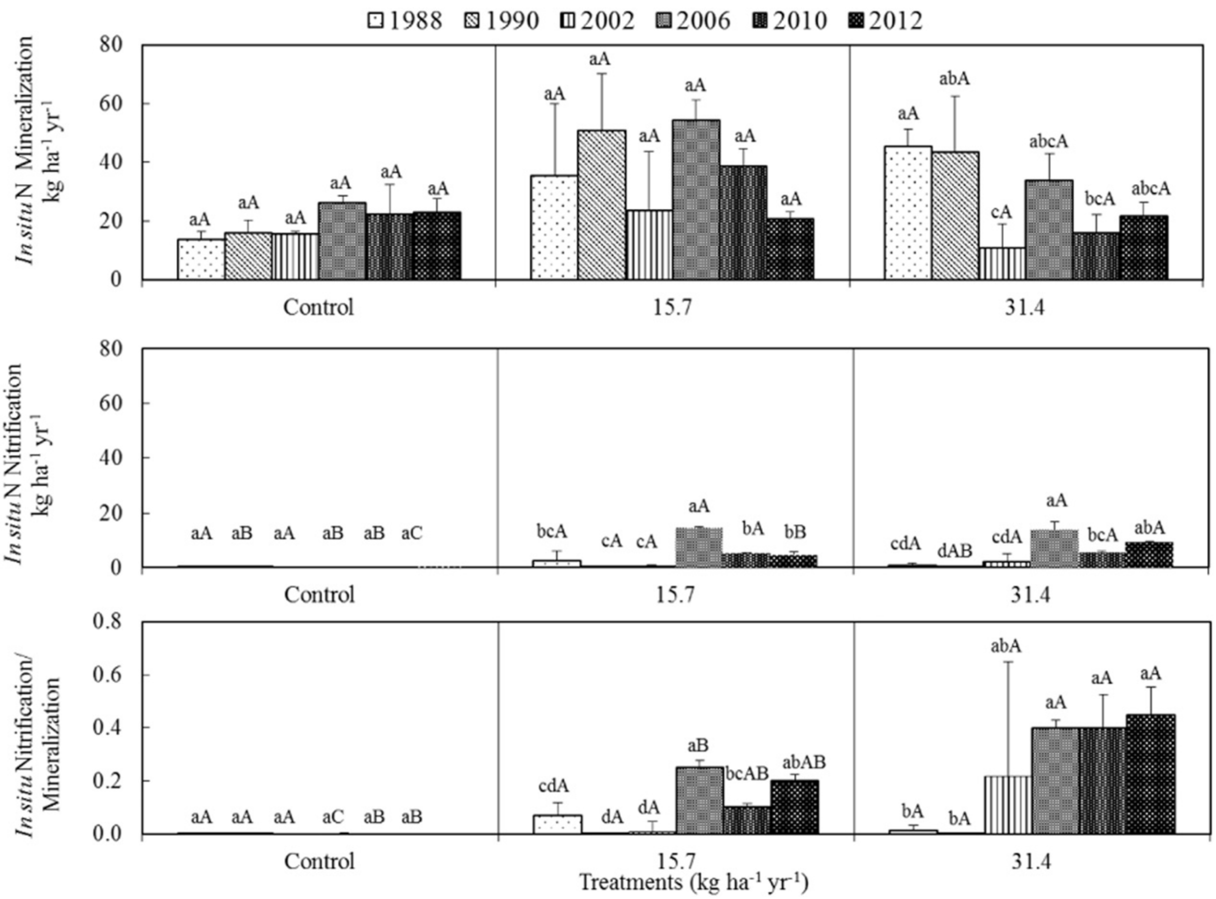

Fig. 2. *In situ* net N mineralization, net nitrification and net nitrification/mineralization on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 1988 to 2012. Standard error bars are shown. Means with the same letter are not significantly different, year versus year within treatment (lowercase) and treatment versus treatment within year (uppercase).

varied, they were not significantly different from the controls. No *in situ* nitrification was ever recorded on the control plots, but nitrification has occurred on both low and high N treatments. For both N treatments, rates of *in situ* nitrification were largest in 2006, at 14.6 kg N ha^{-1} yr^{-1}, and then fluctuated over time. The standard error was very low (i.e., generally $<$1.0 kg N ha^{-1} yr^{-1}) for all years and treatments. Notably, 2002 was a year of low net N mineralization (and nitrification) across all plots. The decline in net *in situ* N mineralization for both low and high N addition plots since N additions ended in 2006 suggests that the ecosystem can quickly respond to changes in N input. Pardo et al. (2006, 2007) found a similar reduction in N saturation using foliar isotopic analysis samples collected before and after the 1990 Clean Air Act reduced N deposition across the region. Increased net N mineralization was one of the original hypotheses of N saturation (Aber et al., 1989). However, given the large volume of woody material inputs associated with tree decline on the treatment plots, much of the added and deposited N may be immobilized in the slowly decaying wood (Kaňa et al., 2015). Eventually, the decomposition will proceed to a point where mineralization exceeds immobilization that could lead to a nitrate pulse from the ecosystem.

A nitrification ratio is calculated by dividing the *in situ* net nitrification by *in situ* net N mineralization rate. The *in situ* nitrification ratio of the control plots was 0.0 over all time periods because no *in situ* nitrification was measured on the control plots. However, nitrification was measured on the treatments, and the ratio has increased over time for both the low and high N addition plots with 2012 values of 0.2 and 0.4 respectively. The rates of nitrification potential for the low and high N addition plots were similar, but the net N mineralization rates on the high N addition plots were approximately half those of the low N addition plots. Therefore, the nitrification ratio of the high N addition plots was almost twice that of the low N addition plots and near 1.0 at the peak in 2006. Nitrification converted nearly all the mineralized N on the high N addition plots into nitrate. The lack of increasing forest floor N% may be partially due to the high relative rate of nitrification, and probable nitrate leaching from the ecosystem.

Net N mineralization and nitrification potential were measured seven times during the study. Control plots rates of potential net N mineralization increased from 6.7 to 21.0 kg ha^{-1} 28 days^{-1} from 1988 to 2006, and decreased back to 5.0 kg ha^{-1} 28 days^{-1} after 2006, but were not significantly different in each treatment over time (Fig. 3).

A previous high elevation spruce-fir gradient study (McNulty et al., 1990, 1991; Boggs et al., 2007) found that the onset of forest floor potential nitrification correlated with a spruce-fir forest floor C:N of 30 (Fig. 4), and a forest floor N concentration of approximately 1.5% (Fig. 5). The Mt. Ascutney treatment plots were well outside the predicted range of potential nitrification given their forest floor C:N and N% values (Fig. 5). However, the relationship between both forest floor C:N and N% and nitrification potential, the 2012–2016 sampling period are more in line with the region-wide relationship compared to the 1988–2006 sampling. This shift may suggest that the treatment plots are tending toward but have not reached N cycling equilibrium. With the termination of N additions, forest floor C:N an N% could continue to decrease for years or decades to come as immobilization declines with reduced debris inputs and decomposition of existing material progresses. However, there is much uncertainty. A study by de Vries et al. (2014) suggests that improvements in the ability to predict forest C cycling are hindered by a

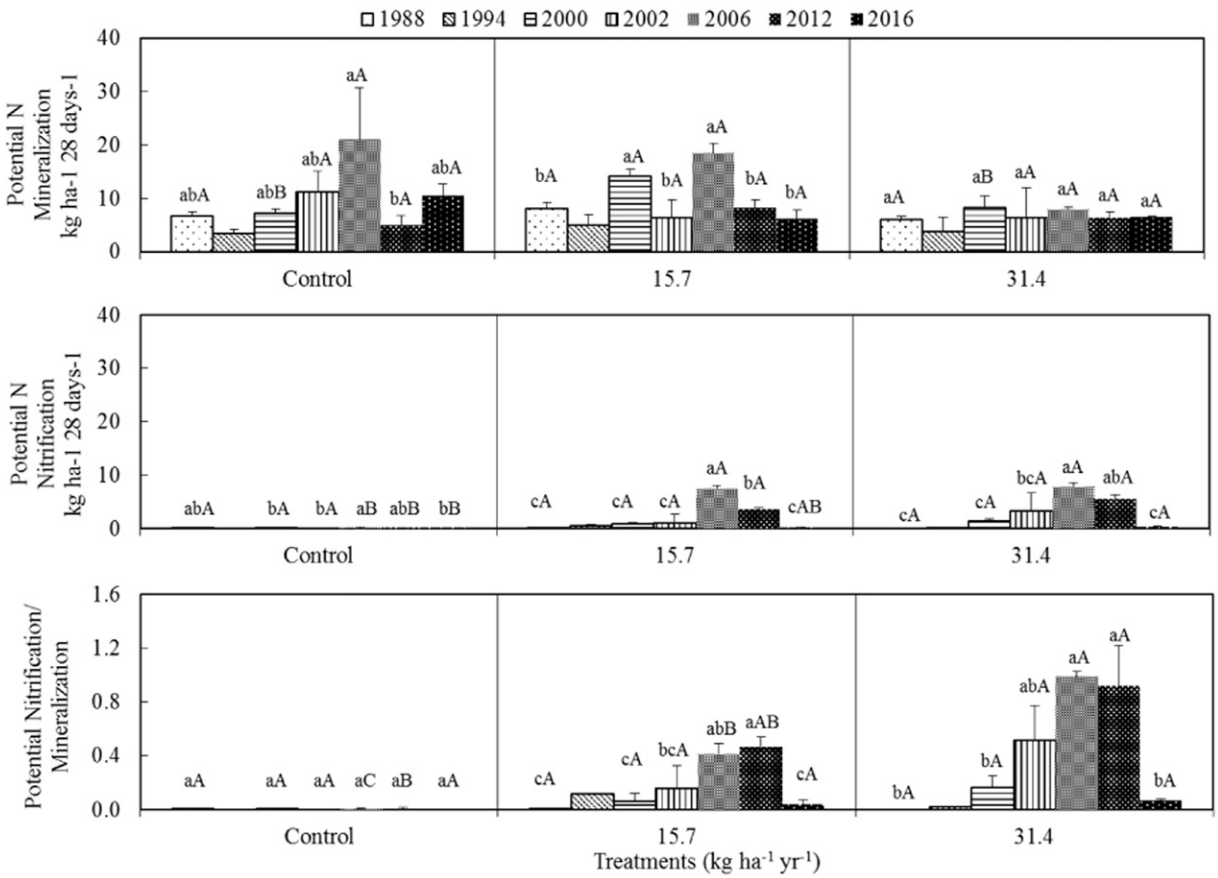

Fig. 3. Potential net N mineralization, net nitrification and net nitrification/mineralization on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 1988 to 2016. Standard error bars are shown. Means with the same letter are not significantly different, year versus year within treatment (lowercase) and treatment versus treatment within year (uppercase). Letters are not shown for 1994 because only the treatment level data could be found; thus 1994 was not included in the statistics.

lack of understanding of plant-microbe interaction. This study would support that conclusion.

3.5. Foliar chemistry

All age class foliage was collected and sampled from the red spruce samples across all treatments, with the first samples collected in June 1988, immediately before any treatments. At that time, there were no statistical differences between any of the plots, and the needles had a foliar N concentration of about 0.90% (Table 5). Over time, there was an upward shift in the control plot foliar N, and it reached a peak of 1.04% (0.07) in 2006 but drifted back down during subsequent years (Table 5). Foliar N concentration on the low N treatment plots peaked in 1998 at 1.57% (0.03) and had since declined back to 1988 pretreatment

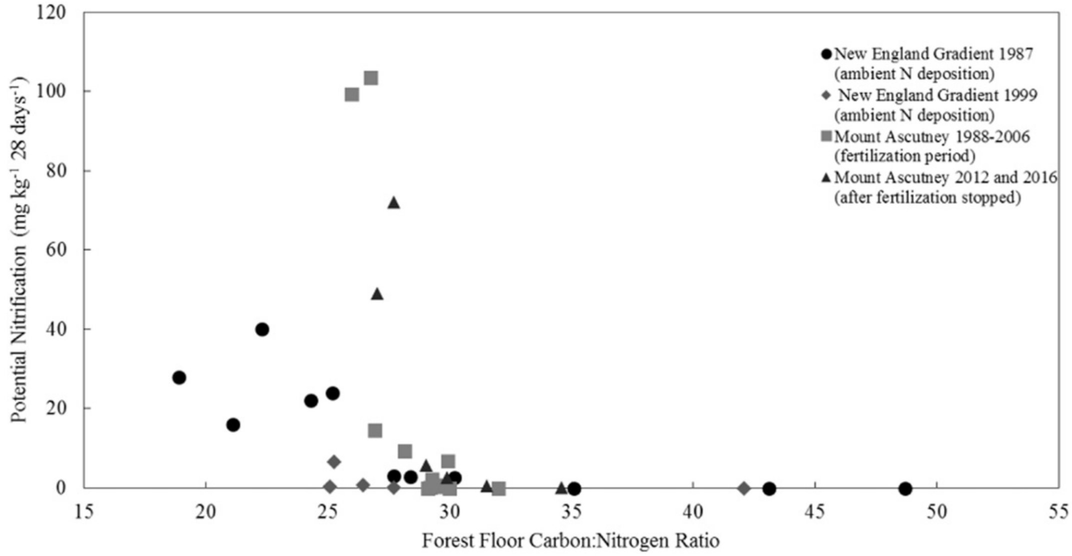

Fig. 4. Comparison of the relationship between potential nitrification and forest floor carbon/nitrogen concentration from McNulty et al. (1990, 1991) (New England Gradient study 1987) and Boggs et al. (2007) (New England Gradient Study 1999) and Mount Ascutney 1988–2016.

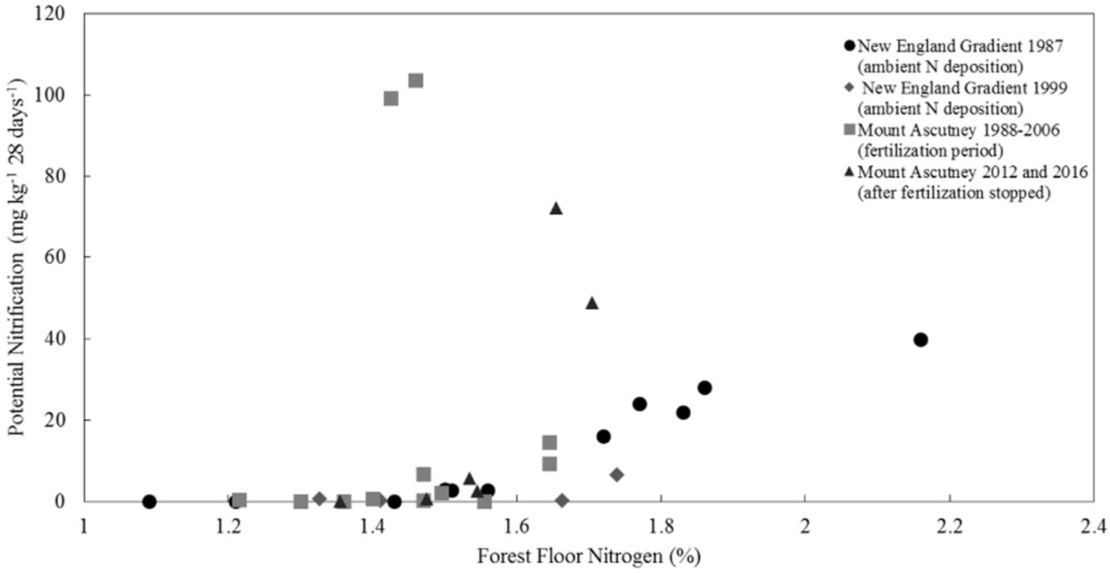

Fig. 5. Comparison of the relationship between potential net nitrification and forest floor nitrogen concentration from McNulty et al. (1990, 1991) (New England Gradient study 1987) and Boggs et al. (2007) (New England Gradient Study 1999) and Mount Ascutney 1988–2016.

Table 5
Foliar chemistry (%) on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 1988 to 2016. Standard error is in parenthesis. Means with the same letter are not significantly different, year versus year within treatment (lowercase) and treatment versus treatment within year (uppercase).

Treatment (kg N ha^{-1} yr^{-1})	1988	1990	1994	1998	2002	2006	2010	2012	2016
Nitrogen									
0.0	0.88 (0.04)cA	1.00 (0.01)abA	0.85 (0.04)cA	1.00 (0.00)abA	0.88 (0.02)cA	1.04 (0.07)aA	0.84 (0.03)cA	0.91 (0.04)bcA	0.87 (0.03)cA
15.7	0.91 (0.02)dA	1.35 (0.06)bcB	1.30 (0.03)cB	1.57 (0.03)aB	1.46 (0.01)abB	1.53 (0.05)aB	1.47 (0.11)abB	1.21 (0.05)cAB	0.92 (0.01)dA
31.4	0.91 (0.05)eA	1.35 (0.04)dB	1.47 (0.09)bcdB	1.84 (0.07)aC	1.56 (0.03)bcdC	1.64 (0.10)abB	1.58 (0.06)bcB	1.38 (0.12)cdB	0.92 (0.04)eA
Calcium									
0.0	0.33 (0.01)abA	0.35 (0.06)aA	0.35 (0.02)aA	0.24 (0.09)abcA	0.20 (0.01)cA	0.21 (0.02)bcA	0.18 (0.03)cA	0.20 (0.02)cA	0.24 (0.00)abcA
15.7	0.24 (0.03)bcB	0.26 (0.04)bA	0.37 (0.05)aA	0.19 (0.03)bcA	0.18 (0.03)bcA	0.18 (0.03)bcA	0.14 (0.00)cA	0.15 (0.03)cA	0.22 (0.04)bcA
31.4	0.27 (0.01)abAB	0.26 (0.05)bA	0.34 (0.02)aA	0.25 (0.04)bcA	0.16 (0.01)dA	0.15 (0.01)dA	0.18 (0.01)cdA	0.15 (0.01)dA	0.18 (0.03)cdA
Magnesium									
0.0	0.07 (0.01)bcA	0.12 (0.00)aA	0.06 (0.01)cA	0.08 (0.01)bA	0.06 (0.01)cA	0.06 (0.00)bcA	0.07 (0.00)bcA	0.08 (0.01)bA	0.07 (0.00)bcA
15.7	0.07 (0.01)bcA	0.12 (0.01)aA	0.08 (0.00)bA	0.05 (0.01)cdA	0.05 (0.00)cdA	0.04 (0.00)dB	0.05 (0.01)dA	0.05 (0.01)dB	0.07 (0.01)bcA
31.4	0.07 (0.01)bA	0.13 (0.01)aA	0.07 (0.01)bA	0.07 (0.02)bA	0.05 (0.01)bcA	0.04 (0.01)cB	0.05 (0.02)bcA	0.04 (0.01)cB	0.05 (0.01)bcB
Aluminum									
0.0	0.002 (0.000)eA	0.007 (0.001)aA	0.004 (0.000)cdA	0.006 (0.001)abA	0.003 (0.000)deA	0.005 (0.001)abA	0.004 (0.000)cdA	0.005 (0.000)bA	0.004 (0.000)bcA
15.7	0.003 (0.001)cA	0.007 (0.001)aA	0.003 (0.000)bcA	0.005 (0.001)bA	0.003 (0.001)cA	0.004 (0.001)bA	0.004 (0.000)bcA	0.004 (0.001)bcA	0.004 (0.001)bcA
31.4	0.003 (0.000)cA	0.007 (0.000)aA	0.003 (0.000)bcA	0.005 (0.001)bA	0.003 (0.000)cA	0.005 (0.000)bA	0.003 (0.000)cA	0.004 (0.001)bcA	0.003 (0.000)cA
Calcium/aluminum									
0.0	140.0 (5.0)aA	55.5 (7.5)cA	95.0 (4.0)bA	57.0 (6.0)bcA	61.5 (1.5)cA	40.9 (0.3)dA	51.9 (3.9)cdA	41.1 (4.8)dA	59.4 (2.4)cA
15.7	97.5 (7.5)aB	41.5 (7.5)bA	85.0 (42.0)abA	57.5 (17.5)abA	71.0 (10.0)abA	43.9 (3.1)bA	41.1 (1.6)bA	43.5 (2.0)bA	60.3 (3.9)abA
31.4	83.5 (2.5)aA	38.5 (6.5)deA	96.5 (9.5)aA	48.0 (0.0)cdA	48.0 (3.0)cdA	32.2 (1.2)eB	55.7 (3.3)bcA	38.9 (0.6)deA	65.3 (5.1)bA
Magnesium/nitrogen									
0.0	0.08 (0.005)bcA	0.12 (0.000)aA	0.07 (0.002)bcA	0.07 (0.005)bcA	0.06 (0.006)cA	0.06 (0.003)cA	0.08 (0.002)bA	0.09 (0.005)bA	0.08 (0.010)bA
15.7	0.07 (0.000)abA	0.08 (0.000)aB	0.06 (0.000)bA	0.03 (0.006)cB	0.03 (0.000)cAB	0.03 (0.002)cB	0.03 (0.000)cB	0.04 (0.005)cB	0.07 (0.005)bAB
31.4	0.07 (0.000)bA	0.1 (0.011)aAB	0.05 (0.002)cdB	0.03 (0.006)cdeB	0.03 (0.003)eB	0.02 (0.003)eB	0.03 (0.010)deB	0.03 (0.005)eB	0.05 (0.000)cB

levels following the cessation of N additions in 2010. The high N addition plot red spruce foliar N concentrations peaked in 1998 at 1.84% (0.07) and like the low N addition plot foliage, rapidly declined following the end of N addition treatments back to the pretreatment concentration. These findings suggest that caution is needed when assuming that the return of tree condition (e.g., foliar N%) equates to a recovery in ecosystem level state (e.g., forest floor N%).

Foliar Ca and Mg% were highly variability throughout the experiment (Table 5). Across all plots, including the control plots, foliar % were highest in 1990 for Mg and 1994 for Ca (Table 5). After this time, control plot red spruce, foliar Ca and Mg% were lower but with no consistent trend. Red spruce foliage Ca% was just above the concentration range of deficiency as reported by Swan (1971). However, this study used whole branch nutrient analysis. Foliar Ca% generally increases with needle age while N, P, and K decreases with age (Turner et al., 1977; DeHayes et al., 1997), so certain foliar age classes could well have been within the deficiency range. Schaberg et al. (1997, 2002) and Perkins et al. (2000) noted reduced membrane Ca% in the N addition plot red spruce foliage that could have caused both water loss and reduced cold tolerance. At the tree level, the problem of reduced cold tolerance may be amplified because foliage with higher N% tends to have shorter leaf retention times (Reich et al., 1995), and therefore lower overall foliar Ca%. Other elements (e.g., Al) and ratios (e.g., Ca:Al, Ca:N, Mg:N) were highly variable, with no discernible pattern.

Schaberg et al. (1997) found that compared to the control plots, spruce on the N addition plots had higher rates of shoot water loss and foliar respiration. A subsequent study by Schaberg et al. (2002) found that N addition plots had lower membrane Ca% (which may have reduced membrane stability and increased respiration), higher levels of electrolyte leakage, and reduced cold tolerance. All of these changes could have predisposed the trees to growing season stress during periods of drought or high air temperatures that would increase evapotranspiration and soil water loss (Sun et al., 2011).

3.6. Seedling regeneration

Under the heavily shaded, intact spruce-fir canopy, there was no regeneration for the first several years of the experiment. However, as the canopy began to open, first on the high N addition plots, and then on the low N addition plots, seedling regeneration began to occur. Control plots had spruce regeneration <1 cm in height in 2006, but the low and high N addition plots did not have similar spruce regeneration (Fig. 6) even though they had significant canopy loss. The low and high N addition plots also had no other species regeneration in 2006. By 2012, the <1 cm spruce regeneration increased on the control, low and high N addition plots, with the most regeneration occurring in the high N addition (Fig. 6). Both low and high N treatments had substantial birch regeneration by 2012, but the control plots did not.

In the mid-sized seedling class (1–3 cm in height), spruce is most abundant on the controls, fir most abundant on the low N additions, and birch most abundant on the high N additions (Fig. 7). Finally, in the advanced regeneration seedling class (>3 cm in height), fir was found on all treatments plots, spruce on none, and birch only on the high N addition plots (Fig. 8). Birch appears to have opportunistically grown as gaps and N became available early on in the high N addition plots. By 2016 birch was the dominant advanced (>3 cm in height) regenerating species on the high N addition plots, while fir was dominant in the low N addition and control plots. Advanced spruce regeneration was not present on any of the sites by 2016.

During the initial opening of the stand gaps, the seedlings <1 cm in height were often present but did not survive into the following summer. Boggs and McNulty (2010) installed thermosensors at the height of 2 cm above the forest floor surface and recorded temperatures above 32.5 °C, a temperature that can permanently damage spruce seedlings (Frey, 1983), especially during periods of drought such as occurred in 2001 and 2004 (Fig. 1) when the forest floor would likely have been abnormally dry.

Additionally, Renard et al. (2016) found that a lack of snow cover can cause winter desiccation in seedlings in ambient conditions. Winter

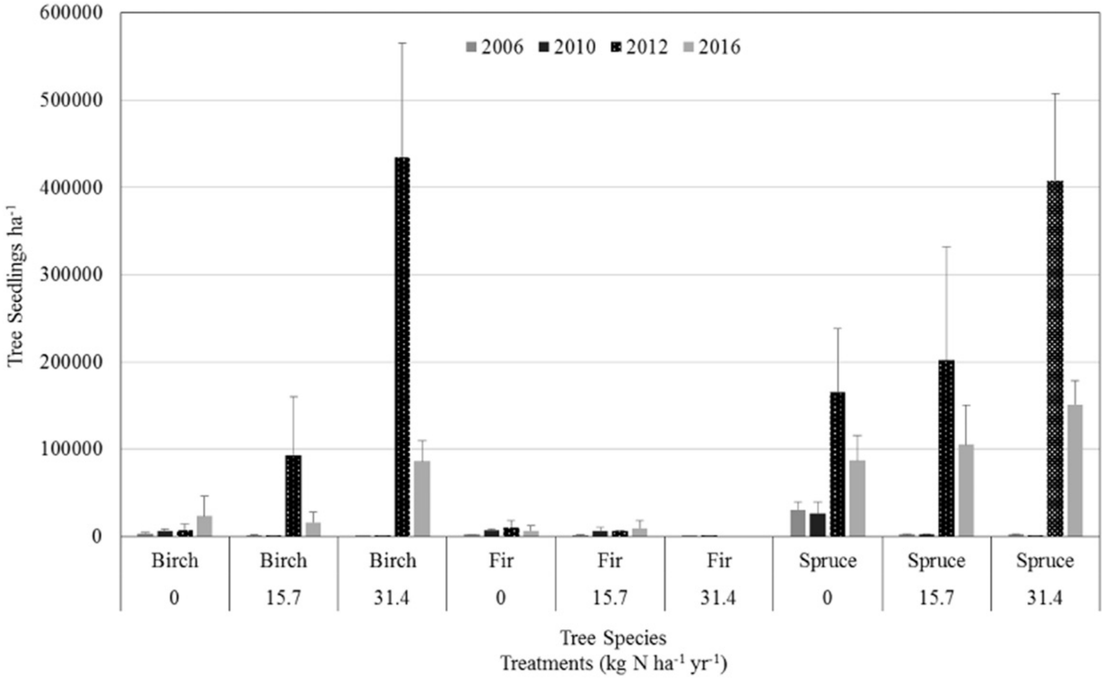

Fig. 6. Birch spp., balsam fir, and red spruce regeneration (<1 cm in height) across treatments on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 2006 to 2016. Standard error bars are shown.

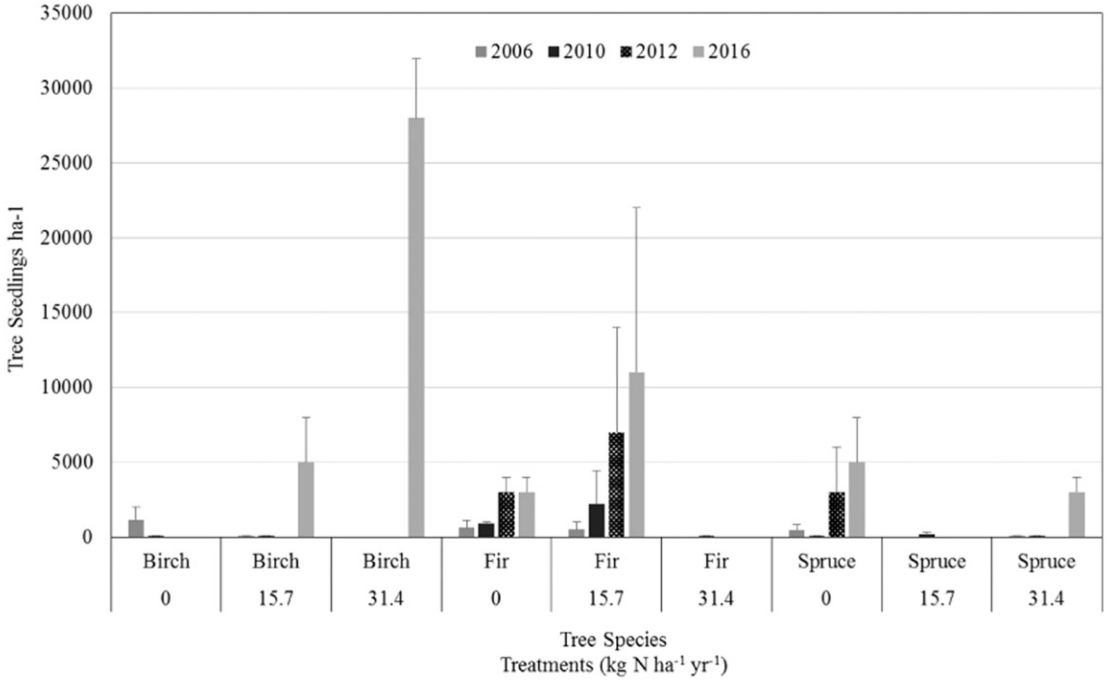

Fig. 7. Birch spp., balsam fir, and red spruce regeneration (1–3 cm in height) across treatments on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 2006 to 2016. Standard error bars are shown.

desiccation was also observed on mature spruce in N addition plots (Perkins et al., 2000; Schaberg et al., 2002). Lazarus et al. (2004) note a severe freeze-thaw event and subsequent spruce mortality across New England in 2003. Both, warmer winters and early thaws followed by freezing conditions, are predicted for the future (IPCC, 2013). These changes to both winter and summer climate could produce an environment that is much less suited for seedling survival compared to the past.

3.7. Implications for the future New England spruce-fir forests

New England spruce-fir forest condition has improved since the 1980s. Kosiba et al. (2013) found that spruce growth rates in the 21st century were higher than they had been in a hundred years. Reductions in atmospheric N deposition and a lack of winter freeze events after 2003 were cited at the reasons for the increased spruce growth

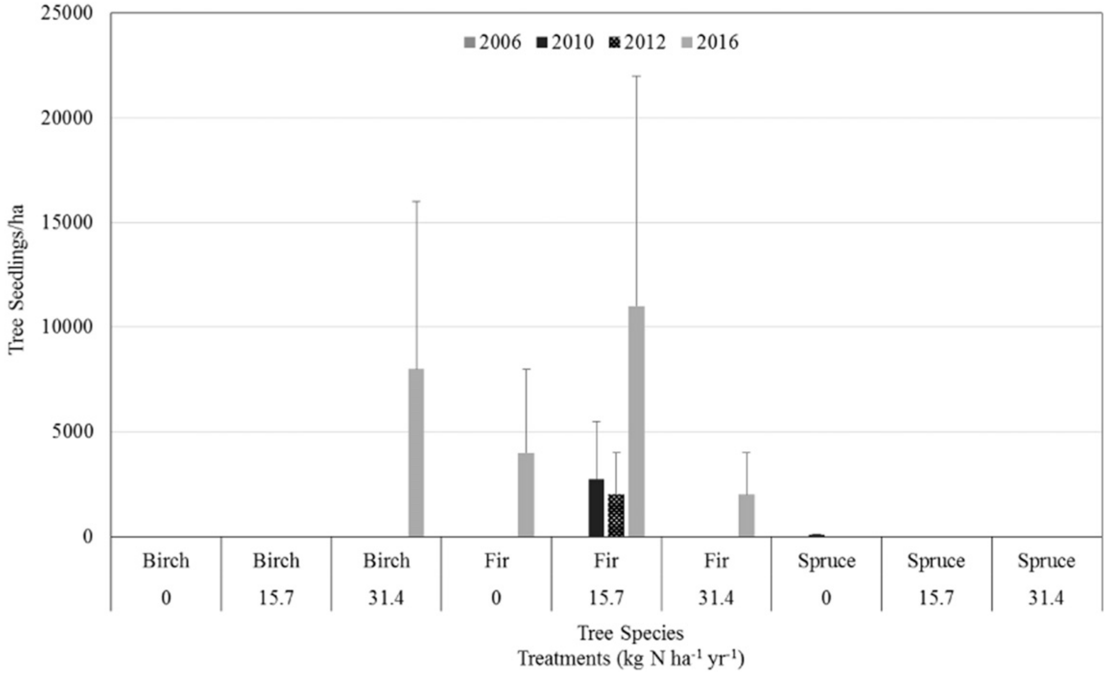

Fig. 8. Birch spp., balsam fir, and red spruce regeneration (>3 cm in height) across treatments on six spruce-fir plots (two paired N addition treatments and one paired control) on Mount Ascutney, Vermont from 2006 to 2016. Standard error bars are shown.

(Kosiba et al., 2013). However, by 2100, a combination of regional warming and increased climate variability (Hayhoe et al., 2007) are predicted to significantly reduce or eliminate, balsam fir, red spruce, birch and maple (*Acer* sp.) from most of New England (Prasad et al., 2007-ongoing). The changes in live and dead BA on Mt. Ascutney support these findings as does a long-term red spruce monitoring study (Ribbons, 2014). Regional ecosystem resiliency could be negatively impacted given that these species comprise the majority of high elevation forested area in New England, (Thompson et al., 2009). However, in areas that are not N saturated, N could increase forest growth (de Vries et al., 2014) of other species (e.g., *Quercus* spp. and *Carya* spp.) that are predicted to become more dominant in warmer, longer growing season environment (Rustad et al., 2012). Additionally, increased base cation weathering and increased N uptake could reduce the area of forest soil critical acid load exceedance (McNulty et al., 2014). Ollinger et al. (2002) predicted that forest growth may remain relatively constant if negative impacts of increased ozone (O_3) counteract positive impacts of N deposition and increased atmospheric CO_2, assuming the forest is not N saturated.

The Mt. Ascutney low N addition plots received annual total N additions input that were below those considered to cause N saturation (Tian et al., 2016), yet significant forest mortality occurred. Wilkins et al. (2016) also observed changes in ecosystem biodiversity at levels below the calculated critical Nitrogen load. Assessment of potential N saturation risk should include N interactions with O_3, drought, winter thaw and freeze events. Reductions in cell wall thickness may predispose a forest to winter freeze-thaw mortality (Schaberg et al., 2002), but if thaw-freeze events to not occur, neither may the mortality. Likewise, reductions in mycorrhiza and fine roots may predispose a tree to drought and or insect related mortality, but if there is no drought, the deaths may be averted. These interactions are complex and can only be seen over years or decades of variable environmental conditions such as observed on Mt. Ascutney.

Interactive stresses likely also impacts forest regeneration. Researchers (e.g., McNulty et al., 1996; Fenn et al., 1998) have suggested that regrowth of more N demanding species may occur after spruce-fir forest decline has ended. On Mt. Ascutney, some intermediate (i.e., 1–3 cm tall) and advanced (i.e., >3 cm tall) birch occurred on the high N addition plots, and low N addition plots had fir regeneration (Figs. 6–8). However, these changes did not occur until N additions ended in 2010. Snow may be protecting these seedlings from the freeze-thaw cycles that appear to be contributing to the death of both spruce and birch on the plots. As these seedling outgrow the protective snow layer, or warmer winters reduce the snow depth, abrupt seedling mortality is possible. If this were to occur, it is not clear what local species could fill the void.

Changes in foliar and forest floor chemistry, and basal area were also observed soon after the N additions stopped, indicating that trees can respond quickly to changes in N inputs. However, regionally elevated N deposition has been occurring for much longer (i.e., 60+ years) than the 20 years associated with this study, but the reduction in N deposition in the region has been ongoing for almost two decades (NADP, 2017). Therefore, New England forests may be moving back from a condition of N saturation and acidification (Strock et al., 2014). Even if N saturation is becoming less of an issue, this does not necessarily mean that these forests are at a reduced risk of decline as climate variability is increasing, and ecosystems are very slow to respond. Therefore, the question may not be whether spruce-fir forests will continue to thrive through the end of the 21st century, but rather what will be the species composition of the next generation of New England forests?

Despite the lack of added N, the control plots of 2016 have changed significantly since the study began in 1988, and now have the characteristics of early stage N saturation (Aber et al., 1989, 1996). Decreases in the root:shoot are one of the main changes that occur early on that leads not only to increased growth but also increased water demand. There has not been a New England drought as severe as the 1961–1967 event in over 50 years (Kunkel et al., 2013b) so it is challenging to assess the impact of a large-scale, severe drought on an ecosystem subjected to such a long record of N deposition, but outcomes could be surprising.

Between 1999 and 2000, the southern Appalachian Mountains experienced a rare and severe drought (US Drought Monitor, 2017). High elevation spruce began to die even though the forest soils were not predicted to be in exceedance of their critical acid load (McNulty et al., 2007). Additionally, the larger, dominant spruce was disproportionately killed compared to the suppressed and subdominant spruce (McNulty and Boggs, 2010). An analysis of the sites suggested that the dominant trees, with their larger leaf area, had much higher water demand and were more chronically stressed based on isotopic wood tissue analysis. Additionally, during the drought, the area experienced a southern pine beetle (*Dendroctonus frontalis*) outbreak. The drought created additional (water) stress on the dominant spruce, and they were unable to produce resins needed to exclude the beetles. The insects subsequently colonized the dominant trees but were unable to colonize the suppressed spruce due to their lower soil water demand and ability to continue to produce resin (McNulty et al., 2014). If New England were to again experience a drought comparable to that in the 1960s, and if a secondary stress such as the eastern spruce beetle, (*Dendroctonus rufipennis*) were to have an outbreak year in conjunction with the drought, the dominant forest trees might be even more severely impacted. None of these type of interactions were part of the original N saturation hypothesis, but could be equally or more important for determining the fate of high elevation spruce-fir forests within the region.

4. Conclusions

The long-term field experiment on Mt. Ascutney has shown that relatively small but chronic inputs of N can cause N saturation as initially described by Aber et al. (1989, 1996). When the annual rate of inorganic N was about triple (approx. 40 kg N ha^{-1} yr^{-1}) the ambient annual net N mineralization rate (12–15 kg N ha^{-1} yr^{-1}), N saturation occurred within a few years. A doubling of N inputs above ambient levels still induced N saturation, but over a longer period. Control plot forest floor N levels were aggrading at a rate equal to the rate of N deposition plus net N mineralization, with no measured nitrate and therefore small a chance of leaching loss (McNulty et al., 1993), and little loss through gaseous emissions recorded (Castro et al., 1993).

The low and high N addition plots failed to reach equilibrium after 18 years of N addition. However, since N additions ended in 2010, the vegetative recovery appears to be progressing quickly. Lovett and Goodale (2011) observed a similar improvement following three years of N additions in a New York deciduous forest. Foliar N concentrations began to decrease within two years of treatment termination, and regeneration has also increased. However, other factors such as nitrate generation and therefore leaching potential continued.

These plots are only at the beginning stages of recovery and continue to be subject to climate variability. Increased mineral weathering, warmer air temperatures and a longer growing season could reduce the amount of forest area negatively impacted by chronic N deposition in the future. Conversely, more winter thaw-freeze cycles, a smaller snow pack, and more intense and frequent drought could once again cause forest decline equal or possibly even worse than that observed in the 1960s. Therefore, long-term N deposition across New England coupled with a changing and more variable climate could ultimately prove to be the longest, largest, and most destructive disturbance yet observed within the region.

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