

Restoring Soil Calcium Reverses Forest Decline

John J. Battles,^{*,†} Timothy J. Fahey,[‡] Charles T. Driscoll, Jr.,[§] Joel D. Blum,^{||} and Chris E. Johnson[§]

[†]Department of Environmental Science, Policy, and Management, University of California, Berkeley, 130 Mulford Hall, Berkeley, California 94720, United States

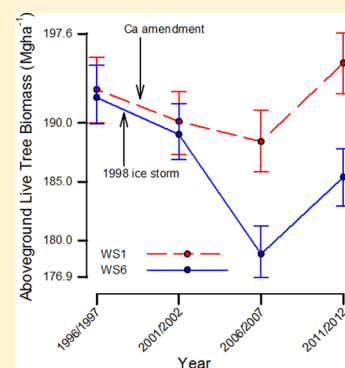
[‡]Department of Natural Resources, Cornell University, G16 Fernow Hall, Ithaca, New York 14853, United States

[§]Department of Civil and Environmental Engineering, Syracuse University, 151 Link Hall, Syracuse, New York 13244, United States

^{||}Department of Earth and Environmental Sciences, University of Michigan, 1100 North University Avenue, Ann Arbor, Michigan 48109, United States

Supporting Information

ABSTRACT: Forest decline in the northeastern United States has been linked to the effects of acid deposition on soil nutrients. To test this link, we added a calcium silicate mineral to a paired watershed at the Hubbard Brook Experimental Forest, New Hampshire, in an amount designed to gradually replace the estimated amount of calcium lost as a result of human activity in the 20th Century (primarily because of acid deposition). The experimental restoration resulted in a recovery of tree biomass increment. The improved calcium nutrition also promoted higher aboveground net primary production and increased the photosynthetic surface area in the treated watershed relative to that in the reference watershed. These results demonstrated that soil acidification accelerated by acid deposition has contributed to the decline of forest growth and health on naturally acidic soil in the northeastern United States and that decline can be reversed by the addition of calcium.



INTRODUCTION

Air pollution can stress forest health and productivity both directly by damaging sensitive tree species and indirectly through the cumulative effects on soil fertility.¹ Air pollutants derived mostly from burning fossil fuels are precursors to acid deposition, which has contributed to the acidification of soils and surface waters in industrialized regions.^{2,3} Since the late 20th Century, environmental regulations in North America and Europe have reduced acid deposition, resulting in the gradual recovery of surface waters;^{4,5} however, severe depletion of soil nutrient cations, for example, calcium (Ca) and magnesium (Mg), may cause persistent acidification effects on sensitive forests.^{6–8} Moreover, acid deposition is projected to increase in regions undergoing rapid economic expansion,^{9,10} and more of the world's forests are likely to experience acidification in the near future. Understanding the consequences of acid deposition and developing strategies to restore the health of acid-stressed ecosystems are therefore research priorities.⁵

Here we report the results of a 15-year field experiment designed to evaluate the role of acid deposition in forest decline, a connection that has proven to be difficult to document because trees are subject to many stresses, abiotic and biotic, natural and human-induced. Forest decline is a complex disorder resulting from various stresses and is manifested in declining growth rates and unexpected or unusual mortality.¹¹ The likely role of acid deposition in some forest declines has been suggested by field surveys and controlled experiments,⁶ with several studies demonstrating the

sensitivity of particular tree species to soil nutrient depletion, especially Ca.^{7,8}

At Hubbard Brook Experimental Forest (HBEF), in the northeastern United States, long-term monitoring of biogeochemical mass balances in small forested watersheds have provided a quantitative basis for evaluating acid deposition effects on soil and streamwater chemistry. In earlier reports, researchers quantified the depletion of soil nutrient cations, particularly Ca, for small watershed ecosystems at HBEF and noted that coincidentally the midaged forest on these watersheds had unexpectedly ceased to accumulate biomass;² however, they could not conclusively link the lack of forest productivity to the observed soil acidification. Ca is an essential plant nutrient, playing key roles in physiological functions and the structural integrity of tree tissue.¹² In addition, two of the important tree species at HBEF, sugar maple (*Acer saccharum* Marsh) and red spruce (*Picea rubens* Sarg.), are known to be sensitive to Ca limitation.^{6,7} We thus hypothesized that soil Ca depletion contributed to forest decline in the experimental watersheds at HBEF. To test this hypothesis, we added calcium silicate to an experimental watershed at HBEF to gradually return soil base saturation to preindustrial levels. We compared biomass accumulation and net primary productivity on this treated watershed with those on a reference (i.e., untreated)

Received: August 13, 2013

Revised: September 6, 2013

Accepted: September 9, 2013

watershed to evaluate effects on forest health. These observations have important implications for understanding the environmental and social costs of continuing emissions of air pollutants.

MATERIALS AND METHODS

Research Sites. HBEF is located in north-central New Hampshire (43°56' N, 71°45' W). The climate is humid continental with short, cool summers and long, cold winters.¹³ Soils are a spatially variable mix of moderately well-drained Spodosols (Haplorthods) and Inceptisols (Dystrochrepts) of sandy-loam texture formed from glacial till.¹⁴ Overstory vegetation is dominated by northern hardwood trees: sugar maple, American beech (*Fagus grandifolia* Ehrh.), and yellow birch (*Betula alleghaniensis* Britt.), which comprise >80% of the forest basal area (Table S1 of the Supporting Information). At the higher elevations, the forest canopy includes significant components of two coniferous tree species, red spruce and balsam fir [*Abies balsamea* (L) Mill.], along with the deciduous tree paper birch (*Betula papyrifera* Marsh.). HBEF is mostly second-growth forest developed following logging in the late 19th and early 20th Centuries. Some areas were also affected by the 1938 hurricane and subsequent salvage logging¹⁵ and by an intense ice storm in 1998.¹⁶ While the ice storm caused considerable structural damage to the forest, most of the damage was restricted to the mid to upper reaches of the watersheds,¹⁶ and by 2001, leaf area had recovered in affected areas¹⁷ (details in the Supporting Information).

This research was conducted in the experimental watersheds on a south-facing slope located in the northeast corner of HBEF (Figure S1 of the Supporting Information). Watershed 6 (WS6, 13.23 ha in size and 549–792 m in elevation) serves as the biogeochemical reference watershed and has been systematically monitored since 1963. The Ca amendment was applied to watershed 1 (WS1, 11.8 ha in size and 488–747 m in elevation). These two watersheds were carefully paired in terms of elevation, topography, soils, and disturbance history (Figure S1 of the Supporting Information). Prior to treatment, there were only minor differences in live tree biomass (Figure 1A) and species composition (Table S1 of the Supporting Information).

Field Sampling. Since 1992, the trees on WS6 have been consistently measured every 5 years as part of the HBEF long-term monitoring program. We use a nested design that includes a complete inventory of all standing trees ≥ 10 cm in diameter at breast height (dbh, 1.37 m in height) and an extensive sample of the smaller trees (2.0–9.9 cm dbh). During the 2002 inventory, we tagged all ≥ 10 cm trees and now track the fate of every standing tree through subsequent inventories. The same sampling regime has been used in WS1 since its first measurement in 1996. Trees were first tagged in WS1 in 2007. Prior to 1992, tree measurement regimes in WS6 varied but were sufficiently similar to produce comparable estimates of tree biomass (details in the Supporting Information).

Fine litterfall (leaves, small twigs, buds, seeds, and fruits) was collected with a stratified random network of 84 litter traps (0.1 m² each, raised 1.5 m above ground) in reference areas adjacent to WS6 from 1992 to 2011 and a network of 48 traps in WS1 from 1998 to 2011. Hardwood foliar leaf litter was sorted by species for each litter trap each year, providing a count of foliage number per 0.1 m². The area per leaf was measured on both reference and treated sites in 4 years to convert leaf counts to leaf area.

Experimental Ca Amendment. In October 1999 (soon after leaf fall), 40.8 Mg of powdered and pelletized wollastonite (CaSiO₃) was evenly distributed by helicopter on WS1.¹⁸ The wollastonite application resulted in a Ca addition of approximately 1028 kg of Ca/ha. This delivery rate was chosen to roughly double soil base saturation and consequently to increase the soil pH to estimated preindustrial levels. By summer 2000, the treatment had significantly increased the pH in the Oie horizon (pH 5.45 in WS1 vs pH 4.29 in WS6). The pH differences have persisted to the present. The wollastonite addition also significantly increased the level of exchangeable Ca in organic soils across the elevation gradient in WS1. With time since the addition, the impacts of the treatment (e.g., increasing pH and exchangeable Ca) moved through the soil profile.¹⁴ For example, by 2002, a significant increase in pH was detected in the Oa horizon¹⁹ as well as a significant increase in the level of exchangeable Ca in the mineral soil in the upper reaches of WS1.

Analytical Methods. The biomass of living trees was calculated from site-specific allometric equations in which the parabolic volume of the tree bole is used to estimate biomass.²⁰ To estimate the uncertainty in our biomass estimates, we followed the best practices for Monte Carlo propagation of error as recommended in a recent review.²¹ The leaf area index (LAI) was estimated by counting leaves in traps and using measurements of average area per leaf for each species and site. The aboveground net primary productivity (ANPP) was calculated as the annual increase in the biomass of perennial woody tissue plus production of ephemeral tissues (e.g., foliage and fruits).^{22,23} We used the nonoverlap of 95% confidence intervals as a conservative standard for detecting statistically significant differences in biomass and productivity between sampling intervals and between treated and reference watersheds (details in the Supporting Information). Comparisons of LAI between watersheds were conducted each year (*t* test) and for 5 year intervals (one-way analysis of variance).

RESULTS AND DISCUSSION

The experimental restoration of soil Ca has resulted in a marked recovery of forest biomass accumulation on WS1. On reference WS6, live tree biomass remained constant from 1982 to 1997 as losses from mortality offset gains from slowing rates of tree growth (Figure 1). Between 1997 and 2007, forest biomass decreased, in part because of damage from a severe ice storm in 1998 that affected the midelevations of both WS1 and WS6.¹⁶ After 2001, the declining biomass on WS1 was reversed, with a significant increase occurring from 2006 to 2011. In contrast, live tree biomass on WS6 in 2012 was still lower than in 1997 (Figure 1A). These observations strongly suggest that depletion of available soil Ca in WS6 at HBEF contributed to the pattern of declining live biomass accumulation during the late 20th Century. Measurements of the components of forest productivity provide additional insights into the mechanisms behind forest decline at Hubbard Brook.

The ANPP of the WS1 forest, calculated as the sum of annual woody biomass increment and fine litterfall flux, was significantly higher than on the reference watershed throughout the post-treatment period [2001–2012 (Table 1)]. Similarly, the LAI on the treated watershed was significantly higher than on the reference forest after both sites recovered from ice storm damage in 1998 (Figure 1B). The relative contributions of wood growth and litterfall flux to the ANPP response in WS1 and WS6 were similar in post-treatment years 7–12 (Table 1),

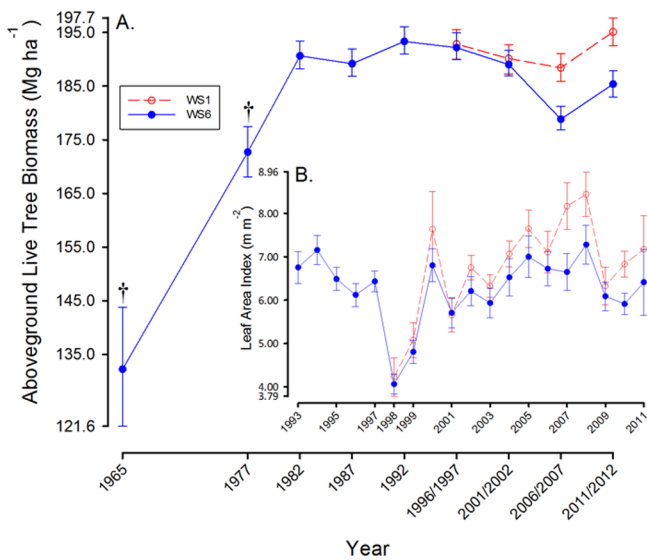


Figure 1. Trends in aboveground live tree biomass and leaf area index in the reference watershed (WS6) and the Ca-treated watershed (WS1) in Hubbard Brook Experimental Forest, New Hampshire. (A) Aboveground live tree biomass (trees ≥ 2 cm in diameter at breast height, 1.37 m) from 1965 to 2012 for WS6 and from 1996 to 2011 for WS1. Means and 95% confidence intervals calculated using Monte Carlo error propagation to account for allometric errors. For 1965 and 1977 (\dagger), different methods were used to estimate uncertainty. See the Supporting Information for details. (B) Leaf area index from 1993 to 2011 for WS6 and from 1998 to 2011 for WS1. Means are plotted with 95% confidence intervals.

whereas the difference in wood growth dominated the large ANPP difference observed in the earlier interval (2–7 years post-treatment). Forest production efficiency and woody biomass production efficiency were comparable between watersheds in the later interval, but wood production efficiency was much higher on the treated watershed in the earlier interval (Table 1). These results suggest that improved forest Ca nutrition promoted higher ANPP both by stimulating increased photosynthetic surface area and by facilitating recovery from canopy damage by the ice storm. Watershed-scale measurements of actual evapotranspiration indicated a transient increase in forest water use on the treated catchment in 2001–2002, suggesting Ca-induced changes in tree physiological processes.²⁴

Understanding the specific role of soil Ca depletion on tree vigor is complicated by the fact that other tree species at HBEF have been afflicted by biotic or other stresses, including American beech,²⁵ red spruce,⁶ paper birch,²⁶ yellow birch,²⁷ and sugar maple.⁷ The decline of beech, because of an exotic disease complex, is not much affected by soil Ca availability as beech growth and survivorship were not enhanced by the Ca treatment. The declining growth and high mortality of both species of birch have contributed to a reduced level of accumulation of biomass on WS6, but increased Ca availability has not significantly ameliorated these trends on WS1. Other regional studies have suggested that high soil Ca reduced ice storm-related mortality of paper birch.²⁶ Although the susceptibility of red spruce to winter injury was reduced by our Ca treatment,²⁸ this response has not yet translated to a significant increase in growth or survivorship.

The most responsive tree species in the Hubbard Brook forest to soil Ca restoration was sugar maple. The overall trend in biomass increment (Figure 1A) was driven largely by the increase in sugar maple biomass in WS1 relative to that in WS6 (Figure S2 of the Supporting Information). This forestwide result supports initial observations that the Ca treatment improved the crown condition of adult sugar maple trees and the performance of sugar maple seedlings.²⁹ Evaluations of canopy trees on WS1 in 2005 indicated significant decreases in canopy dieback and crown transparency, typical symptoms of maple decline.²⁹ These qualitative assessments of improving vigor were consistent with the measured increase in sugar maple LAI (Figure S3 of the Supporting Information). However, the mechanisms whereby Ca deficiency is manifested in canopy dieback, reduced growth, and increased mortality in sugar maple are not fully understood. Colonization of roots of both seedlings and mature sugar maple trees by beneficial arbuscular mycorrhizae was greatly enhanced by the Ca treatment.²⁹ The LAI response suggests improved physiological performance, but photosynthetic rates of sugar maple seedlings did not respond significantly to the Ca treatment.²⁹ Previous studies indicated soil edaphic factors can predispose sugar maple trees to decline by canopy disturbances (e.g., insect defoliation³⁰). The strongest responses of sugar maple to Ca addition on WS1 were observed in the upper elevations of the watershed (Figure S4 of the Supporting Information). The soils are thinner and more base-poor in the upper reaches of WS1,³¹ and the largest increase in exchangeable soil Ca was measured at the higher elevations.³² In the middle elevations, ice storm damage in 1998 contributed to increased mortality, and Ca

Table 1. Comparison of Productivity Components between the Watersheds at HBEF^a

	2001–2002 to 2006–2007, 5 year mean (95% confidence interval)		2006–2007 to 2011–2012, 5 year mean (95% confidence interval)	
	reference	treatment	reference	treatment
wood production (Mg ha ⁻¹ year ⁻¹)	2.27 (2.14–2.39)	* 3.05 ^b (2.90–3.20)	3.48 (3.34–3.62)	* 3.77 (3.63–3.93)
fine litter flux (Mg ha ⁻¹ year ⁻¹)	2.86 (2.76–2.96)	2.88 (2.74–3.02)	2.73 (2.63–2.83)	* 3.12 (2.98–3.26)
aboveground NPP (Mg ha ⁻¹ year ⁻¹)	5.13 (4.90–5.35)	* 5.93 ^b (5.64–6.22)	6.21 (5.97–6.45)	* 6.89 (6.61–7.19)
leaf area index (m ² /m ²)	6.22 (5.96–6.48)	6.68 (6.28–7.08)	6.44 (6.18–6.70)	* 7.34 (6.94–7.74)
ANPP efficiency (ANPP/LAI)	0.82 (0.76–0.90)	0.89 ^b (0.80–0.99)	0.96 (0.89–1.04)	0.94 (0.86–1.04)
wood production efficiency (wood production/LAI)	0.36 (0.33–0.40)	* 0.46 ^b (0.41–0.51)	0.54 (0.50–0.59)	0.51 (0.47–0.56)

^aNet primary productivity components, forest leaf area index and production efficiency for an untreated reference forest (WS6) and for a forested watershed in which soil Ca was experimentally restored (WS1) at the Hubbard Brook Experimental Forest, NH, USA. Comparisons with a * indicate non-overlap of 95% confidence intervals between WS1 and WS6 for each of the metrics. ^bResults relied on indirect means to estimate mortality and ingrowth corrections to production calculations. See the Supporting Information for details.

addition facilitated wound closure and injury recovery in nearby plot-scale studies.⁸ Recent observations of increased evapotranspiration and higher sapflow suggest the possibility of improved physiological performance on the Ca-treated watershed, possibly associated with observed root and mycorrhizal responses, but additional research is needed.²⁴

Sugar maple is among the most important tree species in the northern forest.³³ The species has exhibited decline symptoms in many locations over the past few decades, and previous research has demonstrated that Ca fertilization in the form of relatively high-dose liming can correct its decline on acid forest soils.³⁴ Our study is the first to document that replacement of Ca lost from acid soils as a result of anthropogenic activity in the 20th Century is capable of restoring forest health. Thus, we conclude that soil acidification caused by acid deposition has contributed to the decline of forest growth and health on naturally acidic soils in the northeastern United States. It is also likely that species other than sugar maple have been affected (e.g., red spruce and paper birch). Foliar Ca, soil base saturation, and the ratio of Ca in soil solution to dissolved inorganic aluminum (Al) in soil solution have been recommended as indicators of acidification stress in forest ecosystems.³⁴ These indicators increased from values symptomatic of acidification stress prior to the WS1 Ca treatment to values suggesting that acidification stress is alleviated following treatment (details in the Supporting Information).

Despite emission controls and decreased acid deposition, the recovery of soil base status on these sites has been slow because of the severe depletion of soil available Ca and the slow process of its natural restoration by mineral weathering.⁴ Recent efforts in the United States have focused on critical loads of acidity for aquatic ecosystems.³⁵ A quantitative understanding of air pollution effects on forests is fundamental to extend these efforts and ultimately establish a Secondary National Ambient Air Quality Standard for oxides of nitrogen and sulfur to protect aquatic, forest, and other vulnerable ecosystems (details in the Supporting Information). Rapid recovery of forest health on these acidified soils seems unlikely, and the economic costs of reduced productivity are undoubtedly very high. Continued vigilance in the control of acid rain precursors will be needed to correct these problems, and aggressive options to reduce pollution in rapidly industrializing regions of Latin America, Asia, and Africa⁹ are warranted.

■ ASSOCIATED CONTENT

● Supporting Information

Detailed methods, species-specific responses to Ca amendment, indicators of acidification stress, one table, and four figures. This material is available free of charge via the Internet at <http://pubs.acs.org>.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: jbattles@berkeley.edu. Phone: (510) 643-0684.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This research has been supported by the National Science Foundation (NSF) through Grant DEB-042359 and the Long-Term Ecological Research (LTER) program (NSF Grant 0423259). We appreciate the contributions of the many

researchers involved in making a study of this spatial and temporal scale possible. In particular, we acknowledge Thomas Siccama (forest inventory, tree biomass), Cynthia Wood (litterfall, leaf area measurements), Natalie van Doorn (forest inventory, database management), Alexis Heinz (litterfall data analysis), Natalie Cleavitt (forest inventory, data auditing), and Debra Larson (forest inventory, manuscript proofing) for their efforts. This work is a contribution of the Hubbard Brook Ecosystem Study. Hubbard Brook is part of the LTER network, which is supported by the NSF. HBEF is operated and maintained by the U.S. Department of Agriculture Forest Service, Northern Research Station, Newtown Square, PA.

■ REFERENCES

- (1) Schulze, E.-D. Air pollution and forest decline in a spruce (*Picea abies*) forest. *Science* **1989**, *244*, 766–783.
- (2) Likens, G. E.; Driscoll, C. T.; Buso, D. C. Long-term effects of acid rain: Response and recovery of a forest ecosystem. *Science* **1996**, *272*, 244–246.
- (3) Schindler, D. Effects of Acid-Rain on Fresh-Water Ecosystems. *Science* **1988**, *239*, 149–157.
- (4) Warby, R.; Johnson, C.; Driscoll, C. Chemical recovery of surface waters across the northeastern United States from reduced inputs of acidic deposition: 1984–2001. *Environ. Sci. Technol.* **2005**, *39*, 6548–6554.
- (5) 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. B.; Goodale, C. L.; Herlihy, A. T.; Lawrence, G. B.; Liu, L.; Lynch, J. A.; Novak, K. J. Ecological effects of nitrogen and sulfur air pollution in the US: What do we know? *Frontiers in Ecology and the Environment* **2012**, *10*, 365–372.
- (6) Eagar, C.; Adams, M. B., Eds. *Ecology and Decline of Red Spruce in the Eastern United States*; Springer-Verlag: New York, 1992; pp 429.
- (7) Long, R. P.; Horsley, S. B.; Hallett, R. A.; Bailey, S. W. Sugar maple growth in relation to nutrition and stress in the northeastern United States. *Ecol. Appl.* **2009**, *19*, 1454–1466.
- (8) Huggett, B. A.; Schaberg, P. G.; Hawley, G. J.; Eagar, C. Long-term calcium addition increases growth release, wound closure, and health of sugar maple (*Acer saccharum*) trees at the Hubbard Brook Experimental Forest. *Can. J. For. Res.* **2007**, *37*, 1692–1700.
- (9) Dentener, F.; Drevet, J.; Lamarque, J. F.; Bey, I.; Eickhout, B.; Fiore, A. M.; Hauglustaine, D.; Horowitz, L. W.; Krol, M.; Kulshrestha, U. C.; Lawrence, M.; Galy-Lacaux, C.; Rast, S.; Shindell, D.; Stevenson, D.; Van Noije, T.; Atherton, C.; Bell, N.; Bergman, D.; Butler, T.; Cofala, J.; Collins, B.; Doherty, R.; Ellingsen, K.; Galloway, J.; Gauss, M.; Montanaro, V.; Mueller, J. F.; Pitari, G.; Rodriguez, J.; Sanderson, M.; Solomon, F.; Strahan, S.; Schultz, M.; Sudo, K.; Szopa, S.; Wild, O. Nitrogen and sulfur deposition on regional and global scales: A multimodel evaluation. *Global Biogeochem. Cycles* **2006**, *20*, GB4003.
- (10) Smith, S. J.; van Aardenne, J.; Klimont, Z.; Andres, R. J.; Volke, A.; Arias, S. D. Anthropogenic sulfur dioxide emissions: 1850–2005. *Atmos. Chem. Phys.* **2011**, *11*, 1101–1116.
- (11) Manion, P. D. *Tree Disease Concepts*; Prentice Hall: Englewood Cliffs, NJ, 1991; pp 402.
- (12) McLaughlin, S.; Percy, K. Forest health in North America: Some perspectives on actual and potential roles of climate and air pollution. *Water, Air, Soil Pollut.* **1999**, *116*, 151–197.
- (13) Bailey, A. S.; Hornbeck, J. W.; Campbell, J. J.; Eagar, C. Hydrometeorological database for Hubbard Brook Experimental Forest: 1955–2000. General Technical Report GTR-NE-305, 2003.
- (14) Cho, Y.; Driscoll, C. T.; Johnson, C. E.; Blum, J. D.; Fahey, T. J. Watershed-level responses to calcium silicate treatment in a northern hardwood forest. *Ecosystems* **2012**, *15*, 416–434.
- (15) Schwarz, P. A.; Fahey, T. J.; McCulloch, C. E. Factors controlling spatial variation of tree species abundance in a forested landscape. *Ecology* **2003**, *84*, 1862–1878.

- (16) Rhoads, A. G.; Hamburg, S. P.; Fahey, T. J.; Siccama, T. G.; Hane, E. N.; Battles, J.; Cogbill, C.; Randall, J.; Wilson, G. Effects of an intense ice storm on the structure of a northern hardwood forest. *Can. J. For. Res.* **2002**, *32*, 1763–1775.
- (17) Weeks, B. C.; Hamburg, S. P.; Vadeboncoeur, M. A. Ice storm effects on the canopy structure of a northern hardwood forest after 8 years. *Can. J. For. Res.* **2009**, *39*, 1475–1483.
- (18) Nezat, C. A.; Blum, J. D.; Yanai, R. D.; Park, B. B. Mineral sources of calcium and phosphorus in soils of the northeastern United States. *Soil Sci. Soc. Am. J.* **2008**, *72*, 1786–1794.
- (19) Groffman, P. M.; Fisk, M. C.; Driscoll, C. T.; Likens, G. E.; Fahey, T. J.; Eagar, C.; Pardo, L. H. Calcium additions and microbial nitrogen cycle processes in a northern hardwood forest. *Ecosystems* **2006**, *9*, 1289–1305.
- (20) Whittaker, R. H.; Bormann, F. H.; Likens, G. E.; Siccama, T. G. The Hubbard Brook Ecosystem Study: Forest biomass and production. *Ecol. Monogr.* **1974**, *44*, 233–254.
- (21) Yanai, R. D.; Levine, C. R.; Green, M. B.; Campbell, J. L. Quantifying uncertainty in forest nutrient budgets. *J. For.* **2012**, *110*, 448–456.
- (22) Fassnacht, K. S.; Gower, S. T. Interrelationships among the edaphic and stand characteristics, leaf area index, and aboveground net primary productivity of upland forest ecosystems in north central Wisconsin. *Can. J. For. Res.* **1997**, *27*, 1057–1067.
- (23) Pregitzer, K. S.; Burton, A. J.; Zak, D. R.; Talhelm, A. F. Simulated chronic nitrogen deposition increases carbon storage in northern temperate forests. *Global Change Biology* **2008**, *14*, 142–153.
- (24) Green, M. B.; Bailey, A. S.; Bailey, S. W.; Battles, J. J.; Campbell, J. L.; Driscoll, C. T.; Fahey, T. J.; Lepine, L. C.; Likens, G. E.; Ollinger, S. V.; Schaberg, P. G. Decreased water flowing from a forest amended with calcium silicate. *Proc. Natl. Acad. Sci. U.S.A.* **2013**, *110*, 5999–6003.
- (25) Houston, D. R. Major New Tree Disease Epidemics: Beech Bark Disease. *Annu. Rev. Phytopathol.* **1994**, *32*, 75–87.
- (26) Halman, J. M.; Schaberg, P. G.; Hawley, G. J.; Hansen, C. F. Potential role of soil calcium in recovery of paper birch following ice storm injury in Vermont, USA. *For. Ecol. Manage.* **2011**, *261*, 1539–1545.
- (27) Bourque, C.; Cox, R.; Allen, D.; Arp, P.; Meng, F. Spatial extent of winter thaw events in eastern North America: Historical weather records in relation to yellow birch decline. *Global Change Biology* **2005**, *11*, 1477–1492.
- (28) Hawley, G. J.; Schaberg, P. G.; Eagar, C.; Borer, C. H. Calcium addition at the Hubbard Brook Experimental Forest reduced winter injury to red spruce in a high-injury year. *Can. J. For. Res.* **2006**, *36*, 2544–2549.
- (29) Juice, S. M.; Fahey, T. J.; Siccama, T. G.; Driscoll, C. T.; Denny, E. G.; Eagar, C.; Cleavitt, N. L.; Minocha, R.; Richardson, A. D. Response of sugar maple to calcium addition to Northern Hardwood Forest. *Ecology* **2006**, *87*, 1267–1280.
- (30) Bailey, S.; Horsley, S.; Long, R.; Hallett, R. Influence of edaphic factors on sugar maple nutrition and health on the Allegheny Plateau. *Soil Sci. Soc. Am. J.* **2004**, *68*, 243–252.
- (31) Johnson, C. E.; Driscoll, C. T.; Siccama, T. G.; Likens, G. E. Element fluxes and landscape position in a northern hardwood forest watershed ecosystem. *Ecosystems* **2000**, *3*, 159–184.
- (32) Cho, Y.; Driscoll, C. T.; Johnson, C. E.; Siccama, T. G. Chemical changes in soil and soil solution after calcium silicate addition to a northern hardwood forest. *Biogeochemistry* **2010**, *100*, 3–20.
- (33) Lovett, G. M.; Mitchell, M. J. Sugar maple and nitrogen cycling in the forests of eastern North America. *Front. Ecol. Environ.* **2004**, *2*, 81–88.
- (34) Fenn, M. E.; Lambert, K. F.; Blett, T. F.; Burns, D. A.; Pardo, L. H.; Lovett, G. M.; Haeuber, R. A.; Evers, D. C.; Driscoll, C. T.; Jeffries, D. S. Setting limits: Using air pollution thresholds to protect and restore U.S. Ecosystems. *Issues in Ecology*; Ecological Society of America. Washington, DC, 2011; Vol. 14.
- (35) Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur. U.S. Environmental Protection Agency: Washington, DC, 2009; EPA-452/P-09-004a (http://www.epa.gov/ttnnaqs/standards/no2so2sec/cr_rea.html).

SUPPORTING INFORMATION

Title: Restoring Soil Calcium Reverses Forest Decline

Authors: John J. Battles, Timothy J. Fahey, Charles T. Driscoll Jr., Joel. D. Blum, and Chris E. Johnson

METHODS

Research site. On average, Hubbard Brook Experimental Forest (HBEF) receives 1,395 mm (std. dev. = 189 mm) of precipitation per year, part of which is captured in snow pack persisting from December until April. The mean annual temperature is 5.5 °C (std. dev = 0.61 °C); daily temperatures average from -8.5°C in January to 18.8°C in July¹. The forest of HBEF is considered representative in vegetation and climate of the northern hardwood forest complex, which extends from the north-central United States into southeastern Canada². The soils of HBEF are moderately well-drained Spodosols (Haplorthods) and Inceptisols (Dystrochrepts) of sandy-loam texture formed from glacial till. There is a high degree of spatial variability in the experimental watersheds with shallower, more acidic soils at higher elevations³. These soils exhibit a pattern common in mountainous forest landscapes throughout the Northeast – there are low rates of calcium (Ca) supplied by weathering and the effects of acidic deposition intensify with increasing elevation³.

Forest inventory. The entire reference watershed (WS6) is divided into 208, 25 by 25 m square plots (0.0625 ha, slope corrected). At every five-year interval since 1992, we measure all standing trees ≥ 10 cm dbh in the plot, identify the species, and determine a vigor class based on the health of live trees and the decay class of standing dead trees. Small trees (2.0 to 9.9 cm dbh) are sampled in a 3-m strip along the southern edge of each plot (approximately 75 m²).

Prior to 1992, tree sampling regimes in WS6 varied. In 1965, all trees ≥ 2 cm dbh were measured in a 100 m² square subplot in each of the 208 plots. In 1977, the tree inventory was conducted (i.e., all trees ≥ 10 cm in the 208 plots were measured) but no small trees were measured. In 1982 and 1987, the tree inventory was completed but small trees were measured on random subsets of the 0.0625 ha plots (32 plots in 1982, 35 plots in 1987). These data provide the core information needed to document biomass increment trends and quantify forest productivity responses to the Ca amendment.

Data analysis. As noted, the biomass of living trees was calculated from site-specific allometric equations in which parabolic volume of the tree bole is used to estimate biomass⁴. Given the dependence of biomass on accurate estimates of tree diameter and tree height⁵, we revised our inventory protocols to reduce measurement errors in dbh. We also refit our dbh-to-height equations based on more than 1,300 samples (stratified by species and elevation) collected across WS6 and WS1 in 1996 and 1997 (i.e., prior to the application of Ca). Our approach to estimating biomass at the HBEF has been repeatedly refined and validated against actual measured biomass⁵⁻⁷. On average, there was less than 5% difference between harvest-based and allometric-based estimates of aboveground tree biomass^{6,7}.

We used Monte Carlo randomizations to propagate and combine the errors that contribute to the uncertainty associated with our measurements of tree biomass. The primary sources of error include: 1) measurement error of dbh; 2) variation in the saturating equations predicting tree height from tree diameter; 3) variation in the power equations predicting tree biomass from parabolic volume; and 4) spatial variation among sample plots. It is important to note that at HBEF some populations have been completely inventoried at the watershed scale (e.g., trees ≥ 10 cm dbh). In these instances, there is no spatial variation even though we still use plots as a

convenient accounting tool. In our forest biomass estimates, we followed the best practices for Monte Carlo propagation of error as recommended in a recent review⁸. We report means and 95% confidence intervals calculated from 1,000 Monte Carlo simulations.

Leaf area index (LAI) was estimated by counting leaves in traps and using measurements of average area per leaf for each species and site. Area per leaf was measured on both reference and treated sites in four years; no significant differences (one-way ANOVA) were observed across years within sites. A correction to hardwood LAI was needed to account for conifer LAI (not sorted from litter traps). This correction was based on allometric estimates of conifer (red spruce, balsam fir) leaf area calculated from the complete survey of trees on the two watersheds described earlier. We also compared watershed-wide estimates of total LAI between the litterfall and allometric approaches, prior to treatment (1997 surveys). These comparisons indicated close agreement between methods (<4% difference). Allometric comparisons also indicated pre-treatment LAI was very similar between watersheds prior to treatment (1% difference). The LAI values in these sites are similar to those reported for other sugar maple dominated forests^{9,10}.

Aboveground net primary productivity (ANPP) was calculated as the annual increase in the biomass of perennial woody tissue plus production of ephemeral tissues (e.g., foliage, fruits)^{4,7}. We used the demographic data to correct for wood production of dying trees¹¹ and ingrowth. Note that for WS1 from 2001 to 2006, we lacked comprehensive demographic data. Thus we used the observed mortality and ingrowth rates from 2006 to 2011 to correct the contribution of wood production to ANPP. This approach assumes demographic rates between 2001 and 2006 were the same as the observed rates for 2006-2001. Three lines of evidence support this assumption. A cohort of trees in WS1 (> 800 individuals) tracked since 1998 showed no difference in annual mortality rate for the two intervals. Furthermore there was no

change in the relative density of standing dead trees in WS1 from 2006 to 2011. In both instances standing dead trees accounted for 12% of the population. Finally, there were only minor fluctuations (<3% change) in live tree density between 2006 and 2011. The annual production of ephemeral tissues (leaf, twig, bud, and fruit) was estimated from the litterfall collections, described above.

Sugar maple response. The dynamics of the vegetation during the course of this study (1996-2012) has been particularly complex as evidenced by the fluctuations in biomass increment (Figure 1A) and productivity (Table 1) of the reference watershed (i.e., WS6). The 1998 ice storm that occurred a year before the study contributed to the volatility. Here we provide details on the species-specific responses to the Ca amendment and further support for our contention that sugar maple was the most responsive species to the treatment.

For the major tree species (S), we calculated the net change in live tree biomass in the watersheds for each of the three five-year measurement intervals (i). We then normalized these changes to the trends observed in WS6. Specifically:

$$\Delta Biomass_{S_{\Delta i}} = WS1_{S_{\Delta i}} - WS6_{S_{\Delta i}} \quad \text{[Equation S1]}$$

where S = species and Δi represents the net change in live tree biomass ($Mg\ ha^{-1}$) for species S over the measurement interval i. If there are no difference in species-specific changes between WS1 and WS6, $\Delta Biomass = 0$. Values > 0 indicate a greater increment in biomass during the interval in WS1 relative to WS6; Values < 0 indicate a smaller increment in biomass in WS1. We expressed changes in $\Delta Biomass$ through time as cumulative function with the initial inventory (1996/1997) set to 0.

The overall increase in live tree biomass observed in WS1 (Fig. 1A) was largely driven by a corresponding increase in sugar maple biomass (Figure S2). Differences between the watersheds for most of the other major species were minor. The exception was beech. Its biomass declined on WS1 relative to WS6 (Figure S2). Beech is not considered to be as sensitive to soil Ca availability¹² and thus the absence of a positive response to the Ca amendment on WS1 is not surprising. While the reason for the relative decline of beech in WS1 is unknown, we suspect that competition with a vigorous sugar maple population on WS1 limited its growth and recruitment relative to WS6.

The observed increase in leaf area in WS1 (Figure 1B) can also be attributed to an increase in sugar maple leaf area (Figure S3). For each year we calculated the relative difference in sugar maple leaf area as:

$$\Delta LAI_i = WS1_i - WS6_i \quad \text{[Equation S2]}$$

where i represents the year, $WS1_i$ is the sugar maple leaf area index for WS1 in year i , and $WS6_i$ is the sugar maple leaf area index for WS6 in year i . Sugar maple leaf area was greater in WS1 for every year measured (Figure S3). The differences peaked in 2008 and remained more than $0.9 \text{ m}^2\text{m}^{-2}$ higher through 2010.

We also compared the relative growth rate of sugar maple trees ($\text{dbh} \geq 10 \text{ cm}$) for the most recent five-year interval (2006/2007 to 2011/2012). Relative growth rate was calculated as:

$$RGR_k = \frac{\ln \text{dbh}_{k,j+1} - \ln \text{dbh}_{k,j}}{t} \quad \text{[Equation S3]}$$

where k refers to the individual tree, j refers to the census, and t refers to the time interval between j and $j+1$ ¹³. We used hierarchical Bayesian analysis to account for error propagation in tree demographic parameters¹⁴. We report the median values and the 95% credibility intervals. For the entire watershed, sugar maple trees in WS1 grew more than 40% faster than trees in WS6: 1.57% yr⁻¹ (95%CI: 1.52 - 1.62 % yr⁻¹) versus 1.10% yr⁻¹ (95%CI: 1.03 -1.16 % yr⁻¹). There was also a strong elevation gradient with differences in relative growth rate greater at the higher elevations (Figure S4).

Indicators of acidification stress. Several metrics and limits have been suggested for the protection of forest ecosystems from stress associated with elevated acidic deposition¹⁵, including foliar Ca (> 0.55%)¹⁶, soil % base saturation (> 10-15%)¹⁷ and the molar ratio of calcium to aluminum in the soil solution (Ca/Al > 1)¹⁷. While these metrics and associated thresholds have been widely used to determine critical loads of acidity, they have rarely been evaluated in the context of field observations demonstrating recovery of impacted ecosystems. To help evaluate the applicability of metrics for forest acidification stress, we calculated their values prior to and following the WS1 Ca addition experiment. Foliar Ca concentrations in sugar maple and yellow birch increased significantly following the treatment¹⁸. Prior to treatment, foliar Ca of sugar maple was below the limit (0.55%), indicative of canopy decline in this species¹⁶ over much of the watershed. After treatment, values exceeded this threshold throughout the watershed. Application of soil % base saturation and soil solution Ca/Al as metrics is complicated by spatial heterogeneity in soil chemistry. Both the soil acidity (%BS) and soil solution (Ca/Al) vary with depth as does the root density. Nevertheless our measurements support the use of these indicators as a measure of sugar maple health in WS1. Prior to treatment, the overall base saturation of the mineral soil was approximately 9.6%. Not

surprisingly much higher values were observed in the surface organic horizons of the forest floor (Oie 48.7%, Oa 32.9%) and the upper mineral soil (0-10 cm, 12.1%). After Ca addition, forest floor % base saturation increased markedly (Oie 78.6%, Oa 56.0%), while upper mineral soil did not increase significantly (14.3%)²⁰. Molar ratios of Ca to inorganic monomeric Al (Ali) in soil solution may be a better indicator of acidification stress than Ca to total Al ratio, as Ali is thought to be the toxic fraction^{18,21}. Our observations of Ca/Ali show that values in the Oa horizons increased from 1.6 pre-treatment to 6.4 post-treatment, while mineral soil solutions increased from 0.97 to 3.8²⁰. In high elevation (~730 m) deciduous stands where sugar maple response was most apparent, the mean Ca:Ali molar ratio was 1.2 in Oa horizon solutions, decreasing to 0.59 in Bh horizon solutions and 0.39 in Bs horizon solutions²² prior to treatment, and increased to 25, 3.5 and 1.5, respectively after Ca addition.

References

1. Bailey, A. S.; Hornbeck, J. W.; Campbell, J. J.; Eagar, C. Hydrometeorological database for Hubbard Brook Experimental Forest: 1955-2000. **2003**, *Gen. Tech. Rep. GTR-NE-305*.
2. Hubbard Brook Ecosystem Study: Site Description and Research Activities. USDA Forest Service, Northeastern Forest Experiment Station; **1996**; NE-INF-96-96R.
3. Cho, Y.; Driscoll, C. T.; Johnson, C. E.; Blum, J. D.; Fahey, T. J. Watershed-level responses to calcium silicate treatment in a northern hardwood forest. *Ecosystems* **2012**, *15*, 416-434.
4. Whittaker, R. H.; Bormann, F. H.; Likens, G. E.; Siccama, T. G. The Hubbard Brook Ecosystem Study: Forest biomass and production. *Ecol. Monogr.* **1974**, *44*, 233-254.
5. Arthur, M. A.; Hamburg, S. P.; Siccama, T. G. Validating allometric estimates of aboveground living biomass and nutrient contents of a northern hardwood forest. *Can. J. For. Res.* **2001**, *31*, 11-17.
6. Siccama, T. G.; Hamburg, S. P.; Arthur, M. A.; Yanai, R. D.; Bormann, F. H.; Likens, G. E. Corrections to allometric equations and plant tissue chemistry for Hubbard Brook Experimental Forest. *Ecology* **1994**, *75*, 246-248.

7. Fahey, T. J.; Siccama, T. G.; Driscoll, C. T.; Likens, G. E.; Campbell, J.; Johnson, C. E.; Battles, J. J.; Aber, J. D.; Cole, J. J.; Fisk, M. C.; Groffman, P. M.; Hamburg, S. P.; Holmes, R. T.; Schwarz, P. A.; Yanai, R. D. The biogeochemistry of carbon at Hubbard Brook. *Biogeochemistry* **2005**, *75*, 109-176.
8. Yanai, R. D.; Levine, C. R.; Green, M. B.; Campbell, J. L. Quantifying uncertainty in forest nutrient budgets. *J. For.* **2012**, *110*, 448-456.
9. Fassnacht, K. S.; Gower, S. T. Interrelationships among the edaphic and stand characteristics, leaf area index, and aboveground net primary productivity of upland forest ecosystems in north central Wisconsin. *Can. J. For. Res.* **1997**, *27*, 1057-1067.
10. Pregitzer, K. S.; Burton, A. J.; Zak, D. R.; Talhelm, A. F. Simulated chronic nitrogen deposition increases carbon storage in northern temperate forests. *Global Change Biol.* **2008**, *14*, 142-153.
11. Binkley, D.; Arthur, M. A. How to count dead trees. *Bull. Ecol. Soc. Amer.* **1993**, *74*, 15-16.
12. Bailey, S.; Horsley, S.; Long, R.; Hallett, R. Influence of edaphic factors on sugar maple nutrition and health on the Allegheny Plateau. *Soil Sci. Soc. Am. J.* **2004**, *68*, 243-252.
13. Condit, R.; Ashton, P.; Bunyavejchewin, S.; Dattaraja, H. S.; Davies, S.; Esufali, S.; Ewango, C.; Foster, R.; Gunatilleke, I. A. U. N.; Gunatilleke, C. V. S.; Hall, P.; Harms, K. E.; Hart, T.; Hernandez, C.; Hubbell, S.; Itoh, A.; Kiratiprayoon, S.; LaFrankie, J.; de Lao, S. L.; Makana, J.; Noor, M. N. S.; Kassim, A. R.; Russo, S.; Sukumar, R.; Samper, C.; Suresh, H. S.; Tan, S.; Thomas, S.; Valencia, R.; Vallejo, M.; Villa, G.; Zillio, T. The importance of demographic niches to tree diversity. *Science* **2006**, *313*, 98-101
14. van Doorn, N. S.; Battles, J. J.; Fahey, T. J.; Siccama, T. G.; Schwarz, P. A. Links between biomass and tree demography in a northern hardwood forest: a decade of stability and change in Hubbard Brook Valley, New Hampshire. *Canadian Journal of Forest Research-Revue Canadienne De Recherche Forestiere* **2011**, *41*, 1369-1379.
15. Fenn, M. E.; Lambert, K. F.; Blett, T. F.; Burns, D. A.; Pardo, L. H.; Lovett, G. M.; Haeuber, R. A.; Evers, D. C.; Driscoll, C. T.; Jeffries, D. S. Setting limits: Using air pollution thresholds to protect and restore of U.S. Ecosystems. *Issues in Ecology* **2011**, *14* (www.esa.org/issues).
16. Hallett, R. A.; Bailey, S. W.; Horsley, S. B.; Long, R. P. Influence of nutrition and stress on sugar maple at a regional scale. *Canadian Journal of Forest Research-Revue Canadienne De Recherche Forestiere* **2006**, *36*, 2235-2246.
17. Cronan, C.; Schofield, C. Relationships between Aqueous Aluminum and Acidic Deposition in Forested Watersheds of North-America and Northern Europe. *Environ. Sci. Technol.* **1990**, *24*, 1100-1105.

18. Cronan, C.; Grigal, D. Use of Calcium Aluminum Ratios as Indicators of Stress in Forest Ecosystems. *J. Environ. Qual.* **1995**, *24*, 209-226.
19. Green, M. B.; Bailey, A. S.; Bailey, S. W.; Battles, J. J.; Campbell, J. L.; Driscoll, C. T.; Fahey, T. J.; Lepine, L. C.; Likens, G. E.; Ollinger, S. V.; Schaberg, P. G. Decreased water flowing from a forest amended with calcium silicate. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110*, 5999-6003.
20. Cho, Y.; Driscoll, C. T.; Johnson, C. E.; Siccama, T. G. Chemical changes in soil and soil solution after calcium silicate addition to a northern hardwood forest. *Biogeochem.* **2010**, *100*, 3-20.
21. Sucoff, E.; Thornton, F.; Joslin, J. Sensitivity of Tree Seedlings to Aluminum .1. Honeylocust. *J. Environ. Qual.* **1990**, *19*, 163-171.
22. Likens, G. E.; Driscoll, C. T.; Buso, D. C. Long-term effects of acid rain: Response and recovery of a forest ecosystem. *Science* **1996**, *272*, 244-246.

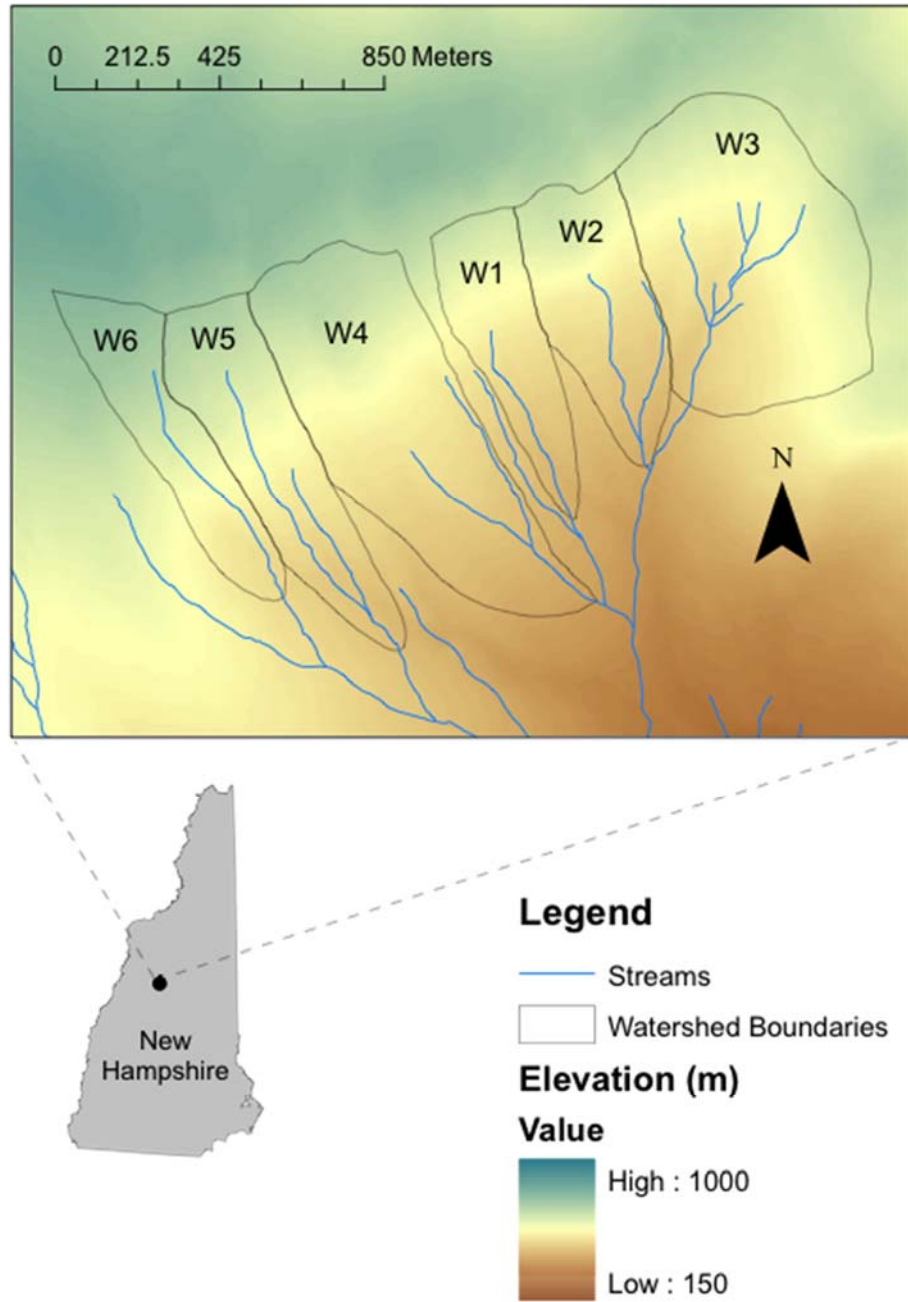


Figure S1. Reference map of watersheds 1 through 6 at the Hubbard Brook Experimental Forest.

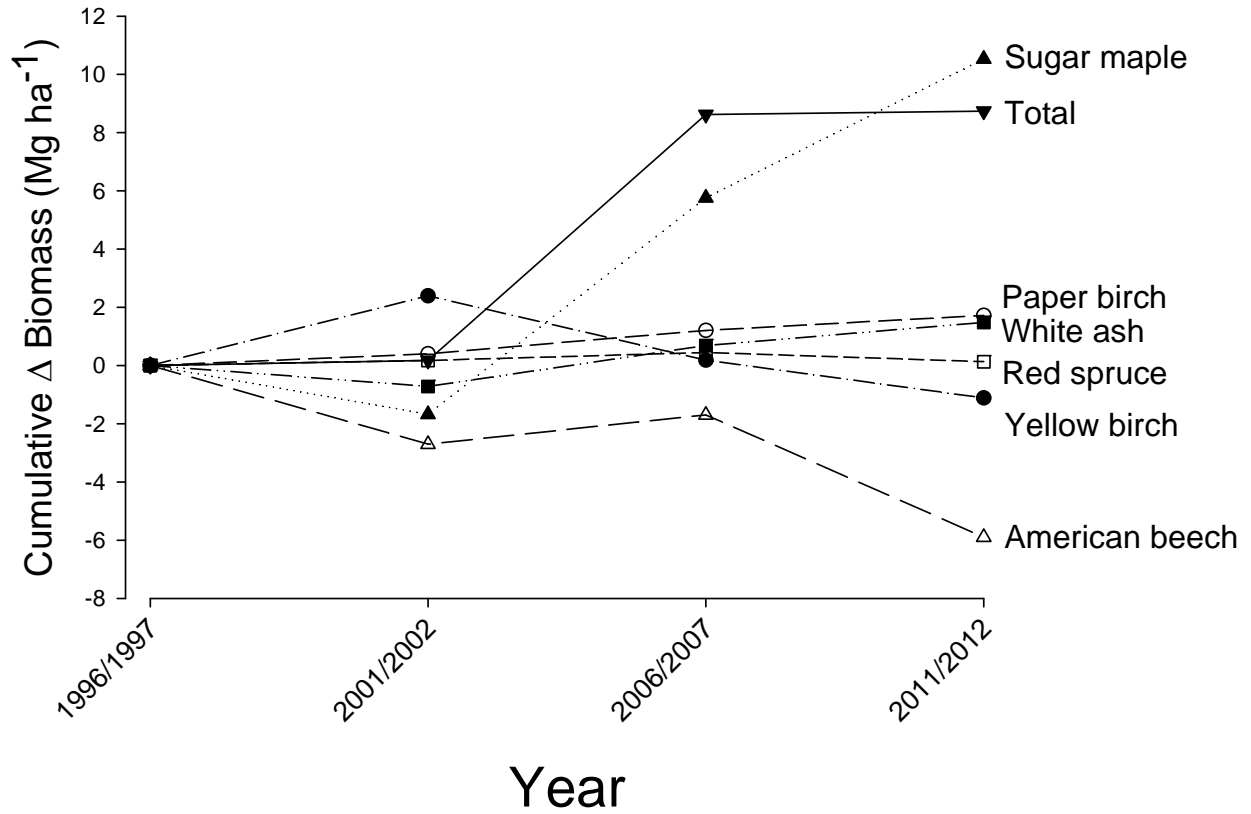


Figure S2. Cumulative change in tree biomass by species during the last 15 years at HBEF.

Temporal trends in WS6 were calculated as the baseline (0-line). Positive values represent increases in WS1 relative to WS6. Negative values represent decreases in WS1 relative to WS6.

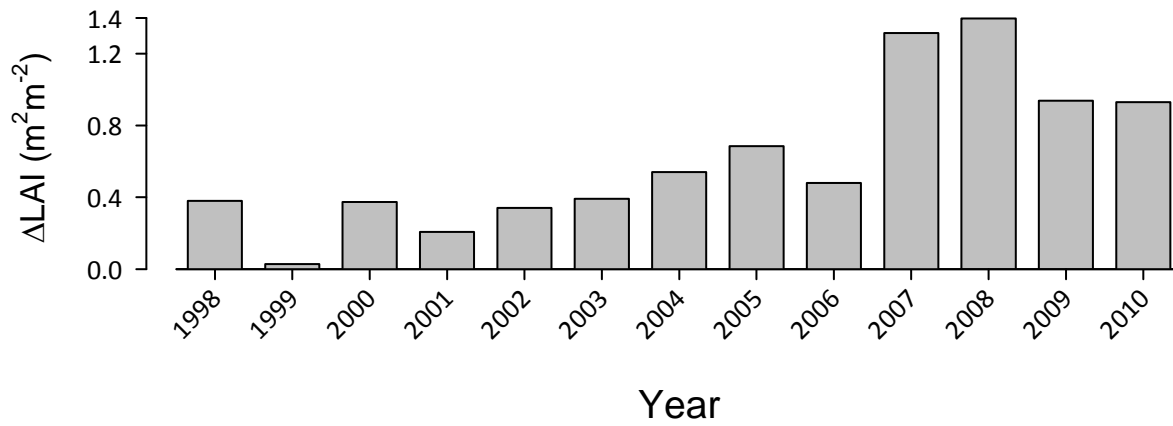


Figure S3. Annual differences in sugar maple leaf area index. Positive values represent increases in WS1 relative to WS6. Negative values represent decreases in WS1 relative to WS6.

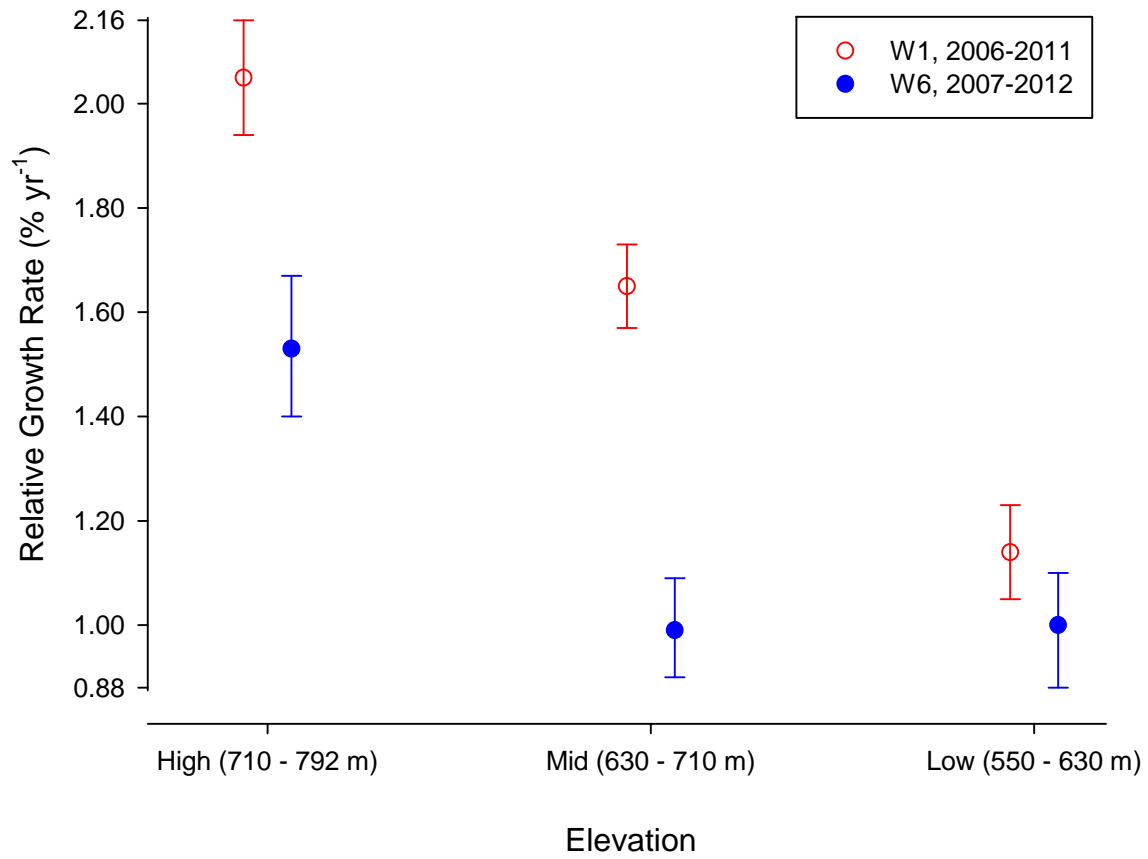


Figure S4. Relative growth rate of sugar maple by elevation band at HBEF. Values represent median growth rates during the most recent five-year interval. Error bars are 95% credibility intervals.

Table S1. Forest composition in the experimental watersheds at HBEF at the start of the experiment (1996 for WS1; 1997 for WS6). Only trees ≥ 10 cm dbh included. Means (with standard errors in parentheses) are reported.

	WS6	WS1
Density (trees ha ⁻¹)	537 (15)	568 (16)
Basal area (m ² ha ⁻¹)	26.0 (0.4)	26.6 (0.4)
Relative Dominance (% Basal area)		
Sugar maple	34.2	35.0
American beech	32.2	26.0
Yellow birch	19.9	19.1
White ash	1.2	6.4
Paper birch	6.6	4.2
Red spruce	2.5	4.5
Balsam fir	2.4	1.4