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Key Points:

- Experimental whole-watershed acidification treatments reduced covariate-adjusted tree growth in three of four hardwood species examined
- For most species, tree growth responded positively to growing season water availability and spring temperatures and negatively to vapor pressure deficit
- Trees in the treated catchment were less sensitive to interannual variation in water availability than those in the control catchment

Supporting Information:

- Supporting Information S1

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Assessing Temperate Forest Growth and Climate Sensitivity in Response to a Long-Term Whole-Watershed Acidification Experiment

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Abstract Acid deposition is a major biogeochemical driver in forest ecosystems, but the impacts of long-term changes in deposition on forest productivity remain unclear. Using a combination of tree ring and forest inventory data, we examined tree growth and climate sensitivity in response to 26 years of whole-watershed ammonium sulfate ((NH₄)₂SO₄) additions at the Fernow Experimental Forest (West Virginia, USA). Linear mixed effects models revealed species-specific responses to both treatment and hydroclimate variables. When controlling for environmental covariates, growth of northern red oak (*Quercus rubra*), red maple (*Acer rubrum*), and tulip poplar (*Liriodendron tulipifera*) was greater (40%, 52%, and 42%, respectively) in the control watershed compared to the treated watershed, but there was no difference in black cherry (*Prunus serotina*). Stem growth was generally positively associated with growing season water availability and spring temperature and negatively associated with vapor pressure deficit. Sensitivity of northern red oak, red maple, and tulip poplar growth to water availability was greater in the control watershed, suggesting that acidification treatment has altered tree response to climate. Results indicate that chronic acid deposition may reduce both forest growth and climate sensitivity, with potentially significant implications for forest carbon and water cycling in deposition-affected regions.

Plain Language Summary While acidifying nitrogen and sulfur pollution has substantially declined in the eastern United States due to the Clean Air Act and its amendments, the legacy of acidification on forest ecosystems is projected to be long-lasting. However, it is often difficult to discern the effects of air pollution on forests without controlled experiments, since changes in pollution have occurred alongside other long-term environmental changes (e.g., climate change and rising atmospheric CO₂ concentrations). At the Fernow Experimental Forest in West Virginia, we examined the growth and climate sensitivity of trees in a watershed that has received 26 years of experimental acidification treatments to trees in an adjacent control watershed. Trees responded to treatment in species-specific ways, but growth of three of four examined hardwood species was greater in the control watershed. Also, trees in the acidified watershed were less sensitive to interannual variation in water availability, suggesting that forests that have experienced high levels of acid deposition respond differently to precipitation than forests that have been less impacted. However, given that trees respond to acid deposition in species-specific ways, the impacts on changes in air pollution on forests will largely depend on the species composition in a given region.

1. Introduction

Acid deposition has profoundly altered the biogeochemistry of eastern North American forests, impacting forest ecosystem health, productivity, and biodiversity (Driscoll et al., 2001; Lovett et al., 2009). In areas with poorly buffered soils, deposition of nitrogen (N), sulfur (S), and H⁺ has increased soil acidity and driven declines in the availability of essential plant nutrients (Likens et al., 1996). Although implementation of the Clean Air Act and its amendments has led to substantial declines in deposition since the 1970s (Sullivan et al., 2018), there is often a time lag between deposition reduction and soil recovery (Johnson et al., 2018). In the mid-Atlantic and southeastern United States, recovery of soil nutrients to preindustrial levels is projected to take decades to centuries (Fakhraei et al., 2016). Soil nutrients influence both above-ground and belowground tree growth (e.g., Fahey et al., 2016; Lévesque et al., 2016; Vadeboncoeur, 2010),

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suggesting that acidification-mediated changes in forest soil nutrients have the potential to alter both forest carbon uptake and response to climate. However, in the absence of controlled experiments, impacts of acid deposition on forest carbon and water balance can be difficult to separate from other long-term changes (e.g., forest succession, responses to changing climate, and increasing atmospheric CO₂). Forest ecosystem response to changes in acid deposition has key implications for the productivity of eastern U.S. temperate forests, which account for approximately two thirds of the conterminous U.S. carbon sink (Lu et al., 2015).

Acid deposition influences tree health and productivity via both direct and indirect mechanisms. As soil pH declines, phytotoxic aluminum is mobilized in the soil solution (Delhaize & Ryan, 1995; Kochian, 1995), and essential plant nutrient cations such as calcium (Ca²⁺), magnesium (Mg²⁺), and potassium (K⁺) are leached from the soil (de Vries et al., 2003; Likens et al., 1996). Soil base cation depletion, aluminum mobilization, and associated nutrient imbalances have been linked to symptoms of decline and mortality in ecologically important hardwood species including sugar maple (*Acer saccharum*; Long et al., 2009; Sullivan et al., 2013) and northern red oak (*Quercus rubra*; Demchik & Sharpe, 2000), as well as reduced productivity of eastern U.S. forests overall (Elias et al., 2009). Direct acid-induced leaching of foliar calcium and subsequent winter freezing injury appear to be the primary mechanisms by which acid deposition affects growth of red spruce (*Picea rubens*; Borer et al., 2005; DeHayes et al., 1999). Recently observed increases in the growth of red spruce in the northeastern United States and central Appalachia have been attributed to reductions in acid deposition following implementation of the Clean Air Act and its amendments (Kosiba et al., 2018; Mathias & Thomas, 2018; Wason et al., 2017). However, such recovery has not been documented in the deciduous broadleaf species that dominate eastern U.S. forests.

In addition to reductions in acid deposition, eastern U.S. forests are experiencing concurrent changes in other drivers of tree growth. While N deposition contributes to soil acidification, particularly on base-poor soils, evidence suggests that deposition-driven alleviation of N limitation has enhanced temperate forest productivity overall (Magnani et al., 2007), though the effects are species-specific (Horn et al., 2018; Thomas et al., 2010). As N deposition declines, some temperate forests are experiencing adverse legacy effects of excess N deposition (i.e., soil acidification and base cation depletion) while simultaneously becoming increasingly N-limited (Gilliam et al., 2019; Groffman et al., 2018). Further, future trajectories of temperate forest productivity are likely to be strongly influenced by changes in climate. Even in the relatively wet eastern United States, tree ring evidence suggests that growth of many dominant species is positively associated with growing season moisture availability, but negatively associated with vapor pressure deficit (VPD) and temperature (Helcoski et al., 2019; Jennings et al., 2016; Levesque et al., 2017; Maxwell et al., 2019). Warmer spring temperatures have been linked to increased growing season forest productivity in the eastern United States (Keenan et al., 2014; Richardson et al., 2009) but also increased N limitation (Elmore et al., 2016). Climate models project more variable precipitation (Luce et al., 2016) and greater atmospheric evaporative demand over forested regions of the eastern United States in coming decades (Dewes et al., 2017; Ficklin & Novick, 2017), but controls on temperate deciduous forest sensitivity to climate remain poorly characterized, and it remains uncertain how changes in climate, acid deposition, and soil nutrient availability will individually or interactively affect forest productivity and climate response.

Long-term acidification experiments provide unique opportunities to assess the impacts of acid deposition on forest productivity and climate sensitivity against a backdrop of other global change drivers. At the Fernow Experimental Forest in West Virginia, additions of ammonium sulfate ((NH₄)₂SO₄) have been applied to a forested watershed three times annually since 1989. The treated watershed has exhibited characteristic signs of acidification and N saturation, including reduced soil pH, higher soil water Al concentrations, soil base cation depletion, and elevated stream water nitrate compared to adjacent reference watersheds (Adams et al., 2006; Fernandez et al., 2010). However, analyses of tree growth response to acidification treatments at Fernow have been mixed or inconclusive. For example, DeWalle et al. (2006) reported that growth of black cherry (*Prunus serotina*) and tulip poplar (*Liriodendron tulipifera*) increased in the treated watershed relative to the control in the first 7 years, perhaps due to N fertilization or base cation mobilization, but growth rates declined thereafter. Jensen et al. (2014) reported species-specific differences in growth, with black cherry exhibiting significantly greater growth in the treated watershed until 2003, and tulip poplar growing consistently faster in the control watershed from 1989–2011. Notably, previous studies in these watersheds have not attempted to assess interactions between acidification treatments and forest climate response, or disentangle stand successional dynamics from treatment effects.

Here, we combine dendrochronological techniques, long-term monitoring of catchment hydrology, and repeat forest inventory data to examine productivity and climate sensitivity of trees in a forested watershed that has received experimental acidification treatments since 1989, compared to those in an adjacent control watershed. Our objectives were to (1) assess long-term effects of experimental acidification treatments on the aboveground growth of four temperate deciduous tree species and (2) examine effects of acidification on tree response to climate in a regenerating hardwood forest. We present results of linear mixed effects models (LMMs) designed to assess the contributions of treatment, climate, and individual drivers to the growth of each species and examine treatment-climate interactions. We compare species-specific responses to treatment to those at the stand scale, and discuss potential mechanisms by which acid deposition-driven changes in soil nutrients mediate tree growth and climate sensitivity.

2. Materials and Methods

2.1. Site Description

This research was conducted in Watershed 3 (WS3, treatment) and Watershed 7 (WS7, control) at the Fernow Experimental Forest in the Allegheny Highlands of West Virginia, USA (39.05°N, −79.67°W). Mean annual temperature at Fernow is 9.2°C, and average precipitation is 1,458 mm/year, distributed approximately evenly between growing and dormant seasons. WS3 and WS7 are 34.4 and 24 ha, respectively, and elevation in both watersheds ranges from ~730 to 860 m (Adams et al., 2006). Streamflow has been continuously monitored from both watersheds since the late 1950s (Edwards & Wood, 2017). Soils are thin (<1 m), acidic, loamy-skeletal, mixed, active, mesic Typic Dystrochrepts overlying quartzose sandstone, and shale (Adams et al., 2006; Fernandez et al., 2010). Between 1994 and 2015, surface soil pH dropped from 4.34 to 4.02 in the treated watershed and 4.50 to 4.39 in the control watershed (Gilliam et al., 2016, 2018). This suggests ongoing soil acidification in both watersheds, but at a faster rate in the treated watershed.

The forest on WS3 was clear cut in 1969–1970 before being allowed to regenerate naturally; WS7 was clear cut and maintained barren with herbicides between 1963 and 1969 and has regenerated naturally from 1969 to present. Forests on both watersheds are mixed-mesophytic, dominated by black cherry (*Prunus serotina*), northern red oak (*Quercus rubra*), tulip poplar (*Liriodendron tulipifera*), red maple (*Acer rubrum*), and sweet birch (*Betula lenta*). Forest inventory data, including DBH and canopy position classification of all trees greater than 2.54 cm diameter within 25 0.1-ha plots in each watershed, were collected by the Forest Service in 1990 (1991 in WS7), 1996, 1999, 2003, 2009, and 2018.

Acidification treatments in the treatment watershed have consisted of three aerial applications (March, July, and November) of ammonium sulfate fertilizer annually since 1989, totaling an additional 40.6 kg N ha^{−1} and 35.4 kg S ha^{−1} per year (Adams et al., 2006). At the onset of the acidification experiment, this represented approximately double the bulk N and S throughfall inputs measured at Fernow. Experimental ammonium sulfate inputs to the treated watershed at Fernow have remained the same even as background deposition has declined in recent decades. For reference, in 2015 background total deposition of N and S at the National Atmospheric Deposition Program (NADP) monitoring station in Parsons, WV (4.4 km from the study watersheds) was 3.1 and 4.0 kg ha^{−1}, respectively (Figure S1 in the supporting information; National Atmospheric Deposition Program, 2018).

2.2. Field Sampling

Our field sampling design was based on the goal of collecting increment cores from at least 15 black cherry (*Prunus serotina*), northern red oak (*Quercus rubra*), red maple (*Acer rubrum*), and tulip poplar (*Liriodendron tulipifera*) individuals in the treatment and control watersheds. Tree cores were collected in plots spaced ~50 m apart along transects running from stream edge to ridge in order to capture elevation and aspect gradients within each watershed (Figure 1). At each plot, beginning at magnetic north and moving clockwise for odd-numbered points and counterclockwise for even-numbered points, we first searched within 10 m for trees of the target species at least 15 cm in DBH, selecting the first satisfactory tree encountered for sampling. If no satisfactory trees were found within 10 m, we repeated the procedure with a radius of 20 m. Trees were rejected for sampling if they had defects in the bottom 2 m that indicated a likelihood of center rot, or if they had a severe lean or a visible history of major crown damage (more than a third of the crown removed in a single event). Trees were also rejected if they were the subject of ongoing research

Figure 1. Locations of tree core and soil sampling plots and forest inventory plots. WS3 (treatment) has received aerial applications of ammonium sulfate since 1989.

(e.g., trees within permanent plots or with dendrometer bands). Two cores were collected from each tree using a 5.3-mm-diameter increment borer. Cores were taken on opposite sides of the bole, perpendicular to the slope to avoid reaction wood. Because of our minimum diameter requirement, trees sampled tended to be canopy dominant or codominant (with the exception of shade-tolerant red maple), potentially biasing our results toward larger trees in these stands (Nehrbass-Ahles et al., 2014). As a result, tree growth results more closely reflect the “optimal” response of trees to treatment and climate (Jennings et al., 2016).

After scraping away the litter layer, two 15-cm soil samples were also collected within the 20-m plot radius. Soil cores were frozen until they could be processed for C and N analyses.

2.3. Tree Core and Soil Sample Processing

Increment cores were air-dried, hand-sanded, skeleton-plotted, and visually cross dated using standard dendrochronological procedures (Speer, 2012). Tree rings were measured using a sliding scale micrometer (Velmex Measuring System, Velmex, Inc., Bloomfield, NY) and MeasureJ2X software (VoorTech Consulting, Holderness, NH). Cross dating was statistically validated using COFECHA (Holmes, 1983). For each tree, the core with the greater correlation with the master series for that species/watershed was selected for subsequent analyses. Raw ring widths were converted to basal area increment (BAI) using the R package dplR (Bunn, 2008). BAI allows for a comparison of growth rates across trees of different sizes by accounting for the often-negative correlation between measured ring width and diameter, while preserving both high- and low-frequency variability in the tree growth (Martin-Benito et al., 2011). However, while BAI is a useful proxy for estimating annual stem growth increment, converting ring widths to BAI assumes uniform cross-sectional growth, even though tree stems are not perfectly circular.

Frozen soil samples were thawed and then sieved to 2 mm. Subsamples of oven-dried soil were ground and analyzed for total C and N concentrations using a Carlo Erba Elemental Analyzer (CE Instruments, Ltd., Wigam, UK). Soil ammonium and nitrate were extracted from 10-g subsamples of field-moist soil in 50 ml of 2.0 M KCl, which was shaken for 1 hr, allowed to settle, then filtered through Whatman Grade 1 filter

paper. Filtered extracts were analyzed for $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ using a QuickChem 8,500 Series Flow Injection Analyzer (Lachat Instruments, Loveland, CO).

2.4. Climate, Deposition, and Streamflow Data

Precipitation data were collected from rain gauges within the treatment and control watersheds (Edwards & Wood, 2017). Temperature data were collected at a meteorological station at the top of Fernow Watershed 4 (adjacent to the treated watershed). Daily maximum vapor pressure deficit (VPD_{max}) data were acquired from PRISM (2018) and converted to monthly averages. Nitrogen and S deposition data were collected at the National Atmospheric Deposition Program (NADP) monitoring station in Parsons, WV (National Atmospheric Deposition Program, 2018). Growing season evapotranspiration (ET) was calculated by differencing precipitation and stream discharge, assuming changes in catchment storage to be negligible. Hydrograph separation was performed to partition streamflow into baseflow and quickflow components (Hewlett & Hibbert, 1967). Catchment wetting (W) was calculated by differencing precipitation and quickflow and determined to be a better estimate of plant available water than precipitation alone. This approach was originally developed by Horton (1933) and was more recently reintroduced (Troch et al., 2009; Voepel et al., 2011). Total growing season (June–August) W, mean March and April temperatures, and mean growing season maximum daily VPD (VPD_{max}) were used as predictors of tree growth in statistical analyses.

2.5. Statistical Analyses

Total soil C, N, C:N ratios, NO_3^- , and NH_4^+ from the treatment and control catchments were compared using nonparametric Mann-Whitney U tests. Mann-Kendall tests were used to examine temporal trends in hydroclimate variables (temperature, precipitation, catchment wetting, and ET) during two time periods: 1957–2015, coinciding with the instrumental record for both watersheds at Fernow, and 1990–2015, coinciding with the ammonium sulfate treatment period. Trends in wet deposition of SO_4^{2-} , NO_3^- , and NH_4^+ were also examined between 1990 and 2015. Preliminary analyses using Kendall's rank correlation (τ) were performed to examine relationships between BAI of each species and climate and deposition variables, and correlations of potential environmental drivers with each other. These analyses indicated that atmospheric CO_2 , SO_4^{2-} deposition, and NO_3^- deposition were highly collinear, and because they increased/decreased monotonically, were also strongly correlated with tree age. To avoid collinearity issues, CO_2 and background deposition variables were excluded as predictors in subsequent analyses, which focus on the effects of acidification treatment and hydroclimate variables that have a higher degree of interannual variability.

LMs were used to determine environmental controls on tree growth while accounting for the hierarchical structure of our data (growth rings in trees, trees in plots, and plots in watersheds) and also temporal autocorrelation in BAI chronologies (e.g., Lévesque et al., 2016; Martin-Benito et al., 2011). BAI data were right-skewed and were square-root-transformed to achieve normality for use as the response variable in LMs. For each species, fixed effects in the LMs included ammonium sulfate treatment, tree development (age and canopy class), hydroclimate variables (VPD_{max} , W, and mean March/April temperatures), local plot factors (solar radiation, topographic wetness index, soil nutrients), and also interactions between acidification and tree response to climate. A conceptual equation for the fixed structure of the “beyond optimal” model (Zurr et al., 2009) is expressed as

$$\sqrt{\text{BAI}} = \beta_0 + \beta_1(\text{Treatment}) + \beta_2(\text{Climate}) + \beta_3(\text{Development}) + \beta_4(\text{Local}) + \varepsilon,$$

where β_0 represents the intercept, β_{1-4} represent regression parameters describing effects of treatment, hydroclimate variability, tree development, and local factors, and ε is the residual term. Tree ID was specified as a random effect in order to isolate growth differences attributable to fixed effects and those due to individual variability among trees. An AR(1,0) structure was used to account for residual autocorrelation. Models with random intercepts and random slopes and intercepts were compared using Akaike Information Criterion (AIC). The difference in AIC between random intercepts and random slopes and intercepts models was <4 , and thus, models were determined to be sufficiently similar to justify the use of more parsimonious random intercepts models. LMs were fitted using the R Version 3.4.4 (R Core Team, 2018) and the package nlme (Pinheiro et al., 2017). Optimal models for each species were determined using a stepwise selection method, with F tests to assess significance of model terms (i.e., Zurr et al., 2009). Because we were specifically interested in the effects of acidification treatment, we included treatment in each model, regardless

of whether or not it improved model fit. Residual normality was verified using histograms and quantile-quantile plots.

Variance explained by fixed effects (marginal r^2) and total variance explained by fixed and random effects (conditional r^2) were computed using the MuMIn R package (Barton, 2018). Importance of individual predictors was determined from LMM output using partial regression coefficients for the relationships between BAI and standardized predictor variables. To test for evidence of a treatment effect, we calculated estimated marginal means (EMMs), which describe the effects of acidification treatment while controlling for fixed covariates, using the emmeans package in R (Lenth et al., 2017). Reported effect sizes for tree growth in treated and control watersheds are thus back-transformed, covariate-adjusted EMMs. For each species, t tests were used to compare EMMs between trees in WS3 and WS7, with significance of $\alpha = 0.05$.

3. Results

3.1. Deposition, Hydroclimate, and Soils

Annual background wet deposition of sulfate declined 83% between 1990 and 2015, while nitrate wet deposition declined 55%. There was no significant temporal trend in wet ammonium deposition (Figure S1 and Table S1). Between 1957 and 2015, mean June–August temperature increased by an average of 0.02°C per year, mean April temperature also increased 0.02°C per year, and there was a significant positive trend in June–August ET in both watersheds (Table S1). However, there were no clear trends in growing season precipitation or VPD_{max} during this time period. Due to substantial interannual variability in temperature, precipitation, and VPD, there were no significant trends in any examined hydroclimate variables between 1990 and 2015.

The soil C:N ratio was 10% greater in samples from the treated catchment ($p = 0.02$; Figure 2a), but total soil N did not differ between watersheds (Figure 2b). NO_3^- -N in the upper 15 cm of soil was 375% greater in the treated catchment compared to the control ($p < 0.0001$; Figure 2c). NH_4^+ -N did not differ between the treatment and control watersheds (Figure 2d).

3.2. Environmental Controls on BAI

Catchment wetting, a proxy for plant-available water, was the most important hydroclimate predictor of BAI in red maple, northern red oak, and tulip poplar across years. There was a significant interaction between catchment wetting and treatment in these three species, with trees in the control watershed more sensitive to water availability than those in the treated watershed (Table 1 and Figures S3–S5). Black cherry, red maple, and tulip poplar responded negatively to VPD_{max} in both watersheds, and the effect did not differ with treatment. To a lesser extent, mean April and March temperatures contributed positively to BAI for all species examined except red maple (Table 1 and Figures S2–S5).

Canopy class was an important predictor of BAI for all species—dominant and codominant trees had greater growth rates than subcanopy trees (Table 1). Tree age was a significant predictor of BAI for northern red oak and tulip poplar, as annual stem growth of both of these species has generally increased over time (Figure 3 and Table 1). Soil NO_3^- and NH_4^+ were generally not important predictors of tree growth across plots. However, there were notable differences in within-species responses to soil N content between the treatment and control watersheds. Soil NO_3^- was positively associated with red maple growth in the control watershed, but there was no relationship in the treated watershed. Soil NH_4^+ was negatively associated with black cherry growth in the control watershed, but there was no relationship in the treated watershed. While soil NO_3^- was not a significant predictor of northern red oak growth ($p > 0.05$), its inclusion marginally improved model fit (Table 1).

As hypothesized, acidification treatments influenced tree growth in species-specific ways. Comparisons of EMMs revealed that acidification treatment reduced BAI by 40% in northern red oak ($p = 0.047$), 52% in red maple ($p = 0.002$), and 42% in tulip poplar ($p = 0.004$), but there was no difference in black cherry growth (Figure 4).

Fixed effects in the LMMs explained between 20% and 53% of the total variance in BAI, as indicated by the marginal r^2 values, while fixed and random effects combined explained between 26% and 74% of variance, as indicated by the conditional r^2 values (Table 1). The considerable increase in explained variance when

Figure 2. Comparisons of (a) soil C:N ratios, (b) total N, (c) NO_3 -N, and (d) NH_4 -N between treatment and control watersheds at Fernow. Boxes represent upper and lower quartiles of each distribution, inner lines represent median values, and dots represent values greater than 1.5 times the interquartile range. Mean values were compared using nonparametric Mann-Whitney U tests (* $P < 0.05$, *** $P < 0.001$).

random effects are included indicates a relatively high degree of among-tree variation in BAI in a given year for black cherry, northern red oak, and tulip poplar.

3.3. Forest Inventory Data

Between 1990 and 2018 (the most recent forest inventory year), the basal area of the four focal species increased from 61% to 79% of total stand basal area in the treated watershed and from 41 to 65% of total stand basal area in the control (Figure 5). Black cherry was the dominant species in the treated watershed, increasing from 38% to 52% of total stand basal area during the 1990–2018 study period. Black cherry increased from 15–22% of stand basal area in the control watershed over the entire study period. In the most recent inventory measurement period (2009–2018), the fraction of black cherry declined in the treated watershed (53.6% to 51.9%) and increased only slightly in the control watershed (21.6% to 22.2%). Tulip poplar was the dominant species in the control watershed, comprising 18–35% of basal area. Red maple held a relatively constant fraction of basal area throughout the study period in both watersheds, making up 12–14% of stand basal area in the treated watershed and ~8% of stand basal area in the control watershed. Northern red oak was the least important species of the four in each watershed, accounting for <8% of basal area in the treated watershed and ~1% of basal area in the control. Total stand basal area was greater in the control watershed throughout the duration of the study period (Figure 6). However, the basal area of the average tree in each watershed (regardless of species) was similar in each inventory year until 2018, when tree size in the control watershed was 16% greater (Figure 6).

Tree canopy class data collected during inventory sampling between 1996 and 2018 reveals that while both stands have thinned overall, the proportion of canopy dominant or codominant black cherry, northern red oak, and tulip poplar has increased in both watersheds (Figure S6). Red maple was most common as an understory species in both watersheds during the study period. While black cherry remains an important

Table 1

Results of LMMs Examining Effects of Acidification Treatment, Canopy Class, Soil Nutrients, and Hydroclimate on BAI of Focal Species

Species	Marginal r^2	Conditional r^2	Fixed effects	Df	Estimate	P
Black cherry	0.2	0.5	Treatment	29	0	0.97
			Treatment:Soil NH ₄	29	6.2	0.02
			Soil NH ₄	29	1.5	0.22
			Spring temperature	873	20.2	<0.0001
			VPD	873	86.3	<0.0001
			Canopy class	29	4.1	0.03
Northern red oak	0.47	0.63	Treatment	24	1.2	0.27
			Treatment:Wetting	721	17.1	<0.0001
			Wetting	721	57.2	<0.0001
			Spring temperature	721	11.9	0.0006
			Soil NO ₃	24	3.8	0.06
			Age	721	77.8	<0.0001
Red maple	0.27	0.27	Canopy class	24	9.3	0.001
			Treatment	26	8.1	0.002
			Treatment:Wetting	767	13.9	0.0002
			Treatment:Soil NO ₃	26	6.7	0.015
			Wetting	767	131.2	<0.0001
			Soil NO ₃	26	0.3	0.56
Tulip poplar	0.52	0.73	VPD	767	39.1	<0.0001
			Canopy class	26	13.8	0.001
			Treatment	29	10.5	0.003
			Treatment:Wetting	818	18.3	<0.0001
			Wetting	818	81.5	<0.0001
			VPD	818	43.7	<0.0001
			Spring temperature	818	26.4	<0.0001
			Age	818	24.7	<0.0001
			Canopy class	29	27.1	<0.0001

Note. Bold text indicates $P < 0.05$ for acidification treatment and its interactions. The response variable is square-root-transformed BAI of each species. Treatment:Variable indicates an interaction between treatment and a given predictor variable.

canopy tree in both watersheds, it nearly disappeared from the understory by 2018 (Figure S6), consistent with its role as a shade-intolerant, early successional species (USDA, 2006).

4. Discussion

4.1. Treatment Effects on Tree Growth

Whole-watershed ammonium sulfate treatments reduced EMM growth of northern red oak, red maple, and tulip poplar relative to an adjacent control watershed, but did not affect the growth of black cherry (Figure 4). The observed species-specific responses to treatment are generally consistent with previous tree ring studies at Fernow, which reported greater tulip poplar growth in the control catchment (Jensen et al., 2014) and comparable growth of black cherry after the first years of treatment (DeWalle et al., 2006). Our findings are also consistent with studies showing that black cherry tends to be acid-tolerant (Long et al., 2009) and fast-growing, but shorter-lived than other cooccurring hardwood species (Auclair & Cottam, 1971). After rapid growth in the first two decades of stand development, black cherry growth has declined in both watersheds since ~2004 (Figure 3). We found 42% greater tulip poplar BAI in the control watershed compared to the treated watershed between 1990 and 2015. This contrasts with recent studies reporting that declines in atmospheric N and S deposition were not associated with tulip poplar growth (Levesque et al., 2017; Maxwell et al., 2019), but we note that experimental additions of ammonium sulfate at Fernow contribute substantially greater N and S loads than eastern U.S. forests have received via background deposition.

Growth of red maple was 52% greater in the control watershed. Adverse effects of acid deposition and soil base depletion on sugar maple (*Acer saccharum*) have been examined extensively (e.g., Juice et al., 2006; Sullivan et al., 2013), but less is known about the sensitivity of red maple to deposition. Our results are

Figure 3. Mean basal area increment chronologies for each focal species: (a) black cherry, (b) northern red oak, (c) red maple, and (d) tulip poplar. Shaded regions represent 95% confidence intervals for each year, and vertical dotted lines mark the beginning of acidification treatments (1989).

consistent with evidence that S deposition is negatively associated with red maple growth (Horn et al., 2018), and the interactive effect between treatment and soil NO_3^- (positive BAI response in the control watershed, no relationship in the treated watershed; Figure S4), suggests that NO_3^- concentrations in the treated watershed have exceeded levels favorable to red maple growth.

While EMMs revealed significantly greater growth of northern red oak in the control catchment (Figure 4), the effect was weaker than that observed in red maple and tulip poplar. The modest negative effect of ammonium sulfate treatment on northern red oak BAI may be a result of negative effects of S-driven acidification (Demchik & Sharpe, 2000; Elias et al., 2009) outweighing positive effects of N fertilization (Horn et al., 2018; Thomas et al., 2010).

In recent national-scale study examining growth and survival of North American tree species in response to N and S deposition, Horn et al. (2018) found that N deposition was positively associated with growth of red maple, northern red oak, and tulip poplar, and reported a threshold response for black cherry (increasing growth until $>15 \text{ kg ha}^{-1} \text{ N}$, but decreasing at higher levels). Given evidence that all four species examined in this study tend to respond positively to N deposition, the negative response to treatment in three of four species may reflect adverse acidifying effects of S, including Ca deficiency (Battles et al., 2014). Phosphorus (P) is often a colimiting nutrient in temperate tree species subjected to elevated N (Goswami et al., 2018; Vadeboncoeur, 2010), and evidence of P limitation in understory plants has been reported in Fernow WS3 (Gress et al., 2007). Future work examining soil and plant stoichiometry in these watersheds may contribute to a better mechanistic understanding of treatment effects on above and belowground tree growth.

Although our tree ring evidence suggests negative tree growth responses to treatment, stand basal area (on a per hectare basis) was greater in the treated watershed throughout the entire study period (Figure 6). This may be attributable to the abundance of black cherry, which was insensitive to treatment, in the treated watershed (Figure 5). While black cherry remains a major component of the upper canopy in both

Figure 4. Estimated marginal means (BAI adjusted for fixed covariates) derived from LMMs for (a) black cherry, (b) northern red oak, (c) red maple, and (d) tulip poplar in the treatment and control watersheds during the treatment period (1989–2015). Error bars represent 95% confidence intervals (* $P < 0.05$, ** $P < 0.01$).

watersheds, it has become less prevalent in the understory and will likely become less important in this forest as succession progresses, barring major gap-generating disturbances (Figure S6). While total stand basal area was greater in the treated watershed, the average tree in the control watershed was larger than that in the treated watershed in the 2018 inventory survey (Figure 6). However, the observed species-specific responses to treatment, and the differences in species composition between the two watersheds (both before and during the treatment period), make it difficult to assess forest response to treatment at the stand level. The increasing proportion of canopy dominant and codominant black cherry, tulip poplar, and northern red oak over time (Figure S6) suggests that stands in both watersheds are in the stem exclusion phase of stand development (Oliver & Larson, 1996). Continued monitoring of forest structure, growth, and species composition will offer insights into the long-term impacts of chronic acidification on forest growth and successional trajectories in these watersheds.

4.2. Tree Response to Hydroclimate Variability

Growing season catchment wetting was the strongest climatic predictor of BAI in all species except black cherry. Positive growth response to precipitation has been observed in broadleaf species throughout the eastern North American forest biome, even where growing season precipitation is generally considered to be abundant (Elliott et al., 2015; Helcoski et al., 2019; Levesque et al., 2017). Physiologically, this may reflect the importance of water availability in photosynthetic carbon assimilation (Lawlor & Cornic, 2002) and/or hydraulically-driven cell expansion during tree ring formation (Rathgeber et al., 2016; Zweifel et al., 2006). To a lesser extent, March and April temperatures were positively associated with BAI in all species except red maple. This is consistent with findings that extended growing season length driven by warm spring temperatures has been associated with greater growing season BAI (Elmore et al., 2016; Mathias & Thomas, 2018) and forest net primary production (Keenan et al., 2014; Ouimette et al., 2018) in temperate regions.

Figure 5. Percent of total stand basal area of the four focal species in each watershed between 1990 and 2018. The four species increased from 61–79% of total stand basal area in WS3 (a) and 41–65% of total stand basal area in WS7 (b) during the study period.

Growing season VPD has been found to be an important limitation on tree growth and carbon uptake in mesic forests (Jennings et al., 2016; Sulman et al., 2016). In this study, VPD was a negative control on BAI for all species except northern red oak. This likely reflects differences in xylem anatomy and hydraulic behavior between northern red oak and the other species examined. Ring-porous species such as northern red

Figure 6. (a) Whole-watershed basal area, based on 25 0.1-ha inventory plots in each watershed. Dotted lines represent linear interpolations stand basal area between inventory years. (b) Mean basal area of the average tree in each watershed each inventory year. Error bars represent 95% confidence intervals. In each figure 1990 data for the control watershed are linearly extrapolated backward, since the first forest inventory data collection in this watershed occurred in 1991.

oak tend to exhibit anisohydric behavior, maintaining high rates of stomatal conductance (and thus carbon assimilation) when VPD is high, despite the risk of hydraulic failure (e.g., Yi et al., 2017). In contrast, diffuse-porous tulip poplar and red maple exhibited negative growth response to VPD. These species tend to exhibit isohydric stomatal behavior, closely regulating stomatal conductance to maintain near-constant leaf water potential regardless of water status (Roman et al., 2015). Black cherry, which is semi-ring porous (Elliott et al., 2017), also responded negatively to growing season VPD. The observed negative response to VPD in the majority of species studied suggests that projected future increases in VPD (Ficklin & Novick, 2017) could result in reduced productivity in mixed-mesophytic forests with species compositions similar to those at Fernow.

4.3. Treatment—Hydroclimate Interactions

Growth of northern red oak, red maple, and tulip poplar was more sensitive to water availability in the control watershed compared to the treated watershed (Table 1 and Figures S3–S5), suggesting that acidification treatment has altered tree physiological response to climate. While the proximate cause of reduced climate sensitivity is unknown, it may be related to reductions in tree root growth in the treated catchment. Gaines et al. (2015) determined that the majority of growing season water uptake for hardwood species in a Pennsylvania forest occurred at less than 60-cm depth, indicating that deciduous species in this region are reliant on relatively shallow soil water. Evidence from root surveys conducted in 1991 and 2013 at Fernow suggests that root density in the organic and upper mineral layers of the soil has declined relative to the control catchment during the experimental period (Adams & Peterjohn, 2016; Peterjohn, 2013; Figure S7). Elevated soil Al^{3+} , which has been observed in WS3 at Fernow (Burnham et al., 2017), has been shown to reduce root growth (Delhaize & Ryan, 1995), impacting plant water and nutrient uptake (Kochian, 1995). In another study at Fernow, Carrara et al. (2018) observed 25% lower mineral soil (0–15 cm) root biomass in WS3, attributed to reduced belowground C allocation in response to elevated soil N. Whether driven by Al toxicity or changes in allocation, reduced root growth could influence tree response to water availability. Acidification and/or nutrient effects on root growth warrant further examination, as rooting depth and distribution are important determinants of ecosystem productivity and drought response (Jackson et al., 1996), and they are important parameterizations in ecohydrologic models (Fan et al., 2017).

While foliar exposure to atmospheric pollution has been shown to affect tree stomatal function, and thus response to climate (Mansfield, 1998), we believe observed differences in tree growth and moisture sensitivity are primarily mediated via belowground effects. This is because ammonium sulfate is aerially applied in solid form three times annually, with two of the applications occurring outside the growing season (Adams et al., 2006). We found substantially higher NO_3^- in the treated watershed, consistent with findings from Burnham et al. (2017), although there was no difference in total N or NH_4^+ (Figure 2). Gilliam et al. (2018) found that net nitrification rates did not differ between the control and treatment watersheds at Fernow, implying that reduced plant uptake of NO_3^- in the treated watershed, rather than elevated nitrification, is responsible for the observed difference in NO_3^- in the upper 15 cm of soil. Soil C:N ratios were higher in the treated watershed (Figure 2), a result consistent with slower decomposition rates reported in N addition and soil acidification experiments (Frey et al., 2014; Hobbie, 2008; Marinos & Bernhardt, 2018). While we considered the possibility that differences in soil organic material between watersheds could affect soil water retention, the treatment and control watersheds have exhibited similar hydrologic dynamics during the treatment period (Figure S8), suggesting that trees in the treatment and control watersheds have had similar access to available soil water.

Direct soil acidification effects may have also influenced tree climate response in the treated watershed. While we did not measure soil pH, soils in the treated watershed have acidified to a greater extent than those in the control watershed during the study period (Gilliam et al., 2016, 2018). Soil acidity has been found to diminish the capacity of red maple and white oak (*Quercus alba*) saplings to adjust coordination of hydraulic traits (xylem anatomy and leaf water relations) in response to N and P manipulations (Medeiros et al., 2016). Further, low soil pH has also been linked to reduced root hydraulic conductivity in paper birch (*Betula papyrifera*; Kamaluddin & Zwiazek, 2004). We cannot discount the possibility that, in addition to differences in root density, lower soil pH may play a role in the observed differences in tree climate sensitivity between the treated and control watersheds. Future studies examining interactions between soil pH, nutrients, and tree ecophysiology may improve our ability to predict tree response to climate deposition-affected forests.

5. Summary and Implications

While the Clean Air Act and its amendments have resulted in substantial declines in N and S pollutant loads in central Appalachia since 1989, acid deposition continues to be a globally important biogeochemical driver. Ammonium sulfate treatments to Fernow WS3 contribute 40.6 kg S ha⁻¹ and 35.4 kg N ha⁻¹ per year. Placing this in context, Yu et al. (2016) report that the average SO₄²⁻ and NO₃⁻ deposition over natural and agricultural systems in China between 2009 and 2014 was 116 kg S ha⁻¹ and 33 kg N ha⁻¹ per year, respectively. Thus, the Fernow Watershed Acidification Study has potential to offer insights into the impacts of acid deposition on temperate forest productivity at levels similar to or below globally realistic pollutant loads.

We found that ammonium sulfate addition reduced tree growth in the majority of hardwood species examined, and that this effect may be mediated by differences in tree sensitivity to water availability. Growing season water availability and spring temperatures were positively associated with tree growth, while VPD was generally a negative control on growth. Together, results suggest that elevated acidic N and S deposition is a negative control on the productivity of canopy trees at Fernow, and acidification-mediated changes to soil chemistry may alter tree response to climate. This may have important implications for forest productivity, hydrology, and vegetation-climate feedbacks in regions that are impacted by acid deposition. However, the extent to which acid deposition influences overall forest productivity likely depends on species-specific responses to deposition.

Data Availability Statement

Tree ring, forest inventory, soil nutrient, hydroclimate, and atmospheric deposition data used to generate the results presented in this manuscript are available in an open-access UVA Dataverse repository (10.18130/V3/FRZYXV).

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