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Alan F. Talhelm  
*University of Idaho*

Kurt S. Pregitzer  
*University of Idaho*

Mark E. Kubiske  
*USDA Forest Service*

Donald R. Zak  
*University of Michigan*

Courtney E. Company  
*University of Western Sydney*

*See next page for additional authors*

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**Authors**

Alan F. Talhelm, Kurt S. Pregitzer, Mark E. Kubiske, Donald R. Zak, Courtney E. Company, Andrew J. Burton, Richard E. Dickson, George R. Hendrey, J G. Isebrands, Keith F. Lewin, John Nagy, and David F. Karnosky

# Elevated carbon dioxide and ozone alter productivity and ecosystem carbon content in northern temperate forests

ALAN F. TALHELM<sup>1</sup>, KURT S. PREGITZER<sup>1</sup>, MARK E. KUBISKE<sup>2</sup>, DONALD R. ZAK<sup>3</sup>, COURTNEY E. CAMPANY<sup>4</sup>, ANDREW J. BURTON<sup>5</sup>, RICHARD E. DICKSON<sup>2</sup>, GEORGE R. HENDREY<sup>6</sup>, J. G. ISEBRANDS<sup>2,7</sup>, KEITH F. LEWIN<sup>8</sup>, JOHN NAGY<sup>8</sup> and DAVID F. KARNOSKY<sup>5†</sup>

<sup>1</sup>Department of Forest, Rangeland, & Fire Sciences, College of Natural Resources, University of Idaho, Moscow, ID 83844, USA,

<sup>2</sup>Northern Research Station, USDA Forest Service, Rhinelander, WI 54501, USA, <sup>3</sup>School of Natural Resources & Environment and Department of Ecology & Evolutionary Biology, University of Michigan, Ann Arbor, MI 48109, USA, <sup>4</sup>Hawkesbury Institute for the Environment, University of Western Sydney, Locked Bag 1797, Penrith, NSW 2751, Australia, <sup>5</sup>Ecosystem Science Center and School of Forest Resources & Environmental Science, Michigan Technological University, Houghton, MI 49931, USA, <sup>6</sup>School of Earth & Environmental Science, Queens College, City University of New York, New York, NY 11367, USA, <sup>7</sup>Environmental Forestry Consultants, LLC, E7323 State Road 54, New London, WI 54961, USA, <sup>8</sup>Environmental Sciences Department, Brookhaven National Laboratory, Upton, NY, 11973, USA

## Abstract

Three young northern temperate forest communities in the north-central United States were exposed to factorial combinations of elevated carbon dioxide (CO<sub>2</sub>) and tropospheric ozone (O<sub>3</sub>) for 11 years. Here, we report results from an extensive sampling of plant biomass and soil conducted at the conclusion of the experiment that enabled us to estimate ecosystem carbon (C) content and cumulative net primary productivity (NPP). Elevated CO<sub>2</sub> enhanced ecosystem C content by 11%, whereas elevated O<sub>3</sub> decreased ecosystem C content by 9%. There was little variation in treatment effects on C content across communities and no meaningful interactions between CO<sub>2</sub> and O<sub>3</sub>. Treatment effects on ecosystem C content resulted primarily from changes in the near-surface mineral soil and tree C, particularly differences in woody tissues. Excluding the mineral soil, cumulative NPP was a strong predictor of ecosystem C content ( $r^2 = 0.96$ ). Elevated CO<sub>2</sub> enhanced cumulative NPP by 39%, a consequence of a 28% increase in canopy nitrogen (N) content (g N m<sup>-2</sup>) and a 28% increase in N productivity (NPP/canopy N). In contrast, elevated O<sub>3</sub> lowered NPP by 10% because of a 21% decrease in canopy N, but did not impact N productivity. Consequently, as the marginal impact of canopy N on NPP ( $\Delta\text{NPP}/\Delta\text{N}$ ) decreased through time with further canopy development, the O<sub>3</sub> effect on NPP dissipated. Within the mineral soil, there was less C in the top 0.1 m of soil under elevated O<sub>3</sub> and less soil C from 0.1 to 0.2 m in depth under elevated CO<sub>2</sub>. Overall, these results suggest that elevated CO<sub>2</sub> may create a sustained increase in NPP, whereas the long-term effect of elevated O<sub>3</sub> on NPP will be smaller than expected. However, changes in soil C are not well-understood and limit our ability to predict changes in ecosystem C content.

**Keywords:** air pollution, carbon sequestration, carbon storage, elevated carbon dioxide (CO<sub>2</sub>), free-air CO<sub>2</sub> enrichment (FACE), net primary productivity (NPP), nitrogen, soil carbon

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## Introduction

Over the past 50 years, increased carbon (C) uptake by the terrestrial biosphere has slowed the rate at which carbon dioxide (CO<sub>2</sub>) has accumulated in the atmosphere (Ballantyne *et al.*, 2012). However, it is uncertain whether the terrestrial biosphere will be a sink for future anthropogenic CO<sub>2</sub> emissions (Ballantyne *et al.*, 2012). In part, this uncertainty arises because it is unclear how the anthropogenic emissions of CO<sub>2</sub>,

oxidized nitrogen (NO<sub>x</sub>), and other trace gases into the atmosphere affect forest C cycling (Nabuurs *et al.*, 2007). Changes in atmospheric composition can directly impact tree physiology (Ainsworth & Long, 2005; Wittig *et al.*, 2009), but physiological responses can be strongly influenced at the ecosystem scale by population dynamics, biogeochemical cycles, and other ecological processes (Körner, 2006), making it difficult to predict changes in forest C sequestration.

Increases in atmospheric CO<sub>2</sub> can enhance the leaf-level rate of photosynthesis, a physiological response recognized for over a century (Brown & Escombe, 1902). Even before it was certain that the atmospheric concentration of CO<sub>2</sub> was rising, this enhancement of

Correspondence: Kurt S. Pregitzer, tel. (208) 885-6442, fax 208-885-5534, e-mail: kpregitzer@uidaho.edu

†Deceased.

photosynthesis was the basis for simple models that predicted anthropogenic CO<sub>2</sub> emissions would enhance plant productivity and forest C sequestration (e.g., Hutchinson, 1948; Eriksson & Welander, 1956). Since that time, hundreds of elevated CO<sub>2</sub> experiments have been conducted and these experiments have confirmed that elevated CO<sub>2</sub> stimulates leaf-level photosynthesis (Curtis & Wang, 1998; Ainsworth & Long, 2005; Norby & Zak, 2011). However, this body of work has also demonstrated that the fate of the additional C assimilated under elevated CO<sub>2</sub> depends on the interactions between the biological and environmental factors that control terrestrial C accrual and turnover at ecosystem and landscape scales (Körner, 2006; Norby & Zak, 2011; Leuzinger & Hättenschwiler, 2013). More simply, increases in photosynthesis do not necessarily stimulate terrestrial C sequestration (Bader *et al.*, 2013; Palacio *et al.*, 2014).

The ability to understand the factors that control the long-term fate of C assimilated under elevated CO<sub>2</sub> expanded in the early 1990s with the development of free-air CO<sub>2</sub> enrichment (FACE) technology. This technology made it possible to conduct controlled experiments using replicated forest stands growing under near natural environmental conditions (Hendrey *et al.*, 1999). These FACE experiments led to a rigorous understanding of the interactions between tree physiology and environmental factors, such as water and nitrogen (N) availability, that strongly influence C accumulation within ecosystems (Norby & Zak, 2011). However, although it is clear that plant species respond individually to elevated CO<sub>2</sub> (Poorter & Navas, 2003; Kubiske *et al.*, 2007; Ali *et al.*, 2013; Smith *et al.*, 2013) and that these varying responses can mediate changes in C cycling (Bradley & Pregitzer, 2007), forest FACE experiments have largely been conducted in single species plantations (Norby & Zak, 2011). Thus, there are few observations with which to understand how community and species differences influence the long-term fate of the additional C assimilated under elevated CO<sub>2</sub> (Norby & Zak, 2011; Smith *et al.*, 2013).

Human activities have also increased the abundance of tropospheric ozone (O<sub>3</sub>), a widespread regional air pollutant that can decrease photosynthesis and diminish plant growth (Wittig *et al.*, 2009; Ainsworth *et al.*, 2012). In the future, it is possible that O<sub>3</sub> will become even more abundant (Lamarque *et al.*, 2011), limiting terrestrial C sequestration across broad portions of the Earth (Felzer *et al.*, 2005; Sitch *et al.*, 2007; Ainsworth *et al.*, 2012). The impacts of O<sub>3</sub> on forest C cycling have been estimated using coupled climate-biogeochemical cycling models, which are parameterized using the physiological responses observed in seedlings and saplings (Felzer *et al.*, 2005; Sitch *et al.*, 2007; Ainsworth

*et al.*, 2012). The few ecosystem-scale forest O<sub>3</sub> experiments that have been conducted (Matyssek *et al.*, 2010a; Zak *et al.*, 2011; Díaz-de-Quijano *et al.*, 2012) have revealed that the responses of small plants cannot always be extrapolated to larger scales (Matyssek *et al.*, 2010b; Ainsworth *et al.*, 2012). It appears that, as with elevated CO<sub>2</sub>, physiological responses to O<sub>3</sub> can be modified by environmental and biological interactions (Matyssek *et al.*, 2010b; Ainsworth *et al.*, 2012). However, the understanding of these interactions is comparatively poor for O<sub>3</sub> and ecosystem-scale research on the impact of O<sub>3</sub> on forest C cycling remains a critical need (Ainsworth *et al.*, 2012). Our knowledge of the interactive effects of CO<sub>2</sub> and O<sub>3</sub> on forest C cycling is even more poorly developed (Ainsworth *et al.*, 2012).

The Aspen FACE experiment was designed to understand how ecosystem processes, particularly competition among species and genotypes, interacted with CO<sub>2</sub> and O<sub>3</sub> to influence C cycling in developing forests (Dickson *et al.*, 2000). The focal species for this experiment was trembling aspen (*Populus tremuloides* Michx.), which is the most widespread tree species in North America and a common component of forests in many regions subject to high O<sub>3</sub> exposure (Karnosky *et al.*, 2003). Aspen was grown in either mixed species (aspen-birch, aspen-maple) or mixed genotype (five clones varying in sensitivity to CO<sub>2</sub> and O<sub>3</sub>) communities, representing common forest types in the north-central United States. During the experiment, these forests were exposed to factorial combinations of elevated CO<sub>2</sub> and O<sub>3</sub> for 11 years and advanced from open-grown seedlings <0.25 m in height to closed-canopy stands that were >8 m tall.

Here, we report results from an extensive sampling of plant biomass and soil conducted at the conclusion of the Aspen FACE experiment. Our first objective was to quantify the effects of elevated CO<sub>2</sub> and/or O<sub>3</sub> on ecosystem C content. At the beginning of the experiment in 1997, we hypothesized that ecosystem C content would be enhanced by elevated CO<sub>2</sub> and decreased by elevated O<sub>3</sub>. Based on the limited information available, we further hypothesized that CO<sub>2</sub> and O<sub>3</sub> would exhibit no significant interactions, such that the two gases would have counteracting effects on ecosystem C content.

Our second objective was to quantify the cumulative input of C through net primary productivity (NPP) during the entire experiment. Although there have been previous reports on NPP at Aspen FACE (King *et al.*, 2005; Zak *et al.*, 2011), a cumulative estimate created the opportunity for a more comprehensive understanding of the treatment effects. In addition, estimating cumulative NPP allowed us to test the hypothesis that the size of major ecosystem C pools (plants and

detritus, soil C) in these developing forests was related to plant production.

Our final objective was to gain insight into the canopy characteristics that led to the differences in tree productivity ( $NPP_{tree}$ ), the dominant component of ecosystem NPP. To do this, we fit several simple stand-level models that predict productivity based on canopy development metrics (leaf area, canopy N, etc.) and canopy productivity (e.g., productivity per leaf area). This allowed us to test the hypotheses that both canopy development and canopy productivity would be stimulated by elevated  $CO_2$  and depressed by elevated  $O_3$  in these young forests (Norby & Zak, 2011; Ainsworth *et al.*, 2012). We expected that developmental effects would diminish with time as all stands approached maximum leaf area index (Körner, 2006; Norby & Zak, 2011) and  $O_3$ -tolerant trees became more dominant (Kubiske *et al.*, 2007).

## Materials and methods

Our experiment, located in Rhinelander, Wisconsin, USA, consisted of twelve 30-m diameter fumigation rings in three randomized complete blocks with factorial  $CO_2$  and  $O_3$  treatments (Dickson *et al.*, 2000). This FACE technology achieves target gas concentrations using trace gas monitors within each ring to regulate gas ( $CO_2$ ,  $O_3$ ) delivery from a system of blowers, plenums, valves, and vertical vent pipes placed around the outside of the fumigation ring (Hendrey *et al.*, 1999). Fumigation occurred during daylight hours from bud-burst to leaf-off from May 1998 to early 2009. Concentrations ranged from 40 to 55  $nl\ l^{-1}$  for elevated  $O_3$  (elevated average: 46  $nl\ l^{-1}$ , ambient average: 36  $nl\ l^{-1}$ ) and from 515 to 540  $\mu l\ l^{-1}$  for elevated  $CO_2$  (532  $\mu l\ l^{-1}$ , ambient: 369  $\mu l\ l^{-1}$ ). Soils are Alfic Haplorthods with a sandy loam Ap overlaying a sandy clay loam Bt. Prior to the experiment, the top 0.1 m of mineral soil contained  $1896 \pm 74\ g\ m^{-2}$  of C (mean  $\pm$  SE), which did not differ by treatment ( $P > 0.35$ ).

The forests were established from small trees (<25 cm tall) planted during July 1997 at 1 m spacing. Half of each ring was planted with five aspen genotypes representing a range of responsiveness to  $O_3$  or  $CO_2$  (Dickson *et al.*, 2000). The remaining two quarters of each ring were planted with paper birch (*Betula papyrifera* Marsh.) or sugar maple (*Acer saccharum* Marsh.) at equal densities with a single aspen genotype. We defined a 'core' area within each ring where gas concentrations were the most stable (aspen: 166  $m^2$ , aspen-birch: 76  $m^2$ , aspen-maple: 66  $m^2$ ; Kubiske *et al.*, 2007).

### Objective 1: Quantifying ecosystem C content

During the 2009 growing season, we sampled aboveground biomass, belowground biomass, and soil within the core area, sequentially sampling by block. Within four 0.25  $m^2$  subplots per each community section, we collected groundcover vegetation and the organic soil horizons. Mineral soil and roots

were sampled from 1 m deep pits, which were  $2 \times 5\ m$  within the aspen section and  $2 \times 3\ m$  elsewhere. All trees in the pit area were harvested and additional trees were harvested outside of the pit area so that, in each ring, at least 10 trees were harvested from the aspen section (two per genotype) and three trees of each species were harvested from the other two sections. Branches were removed from the harvested trees, then 1–2 cm thick cross-sections were cut from the main stem at heights (m) from the ground surface of 0.1, 0.25, 1.5, 2, 2.5, 3, and approximately every 1 m thereafter until the live crown was reached. Within the live crown, a cross-section was cut at the midpoint of each annual height growth increment. The mass of each harvested stem was estimated by integrating a polynomial equation for mass per length ( $g\ cm^{-1}$ ) and height of each section. We developed allometric equations for the mass of the stem, branches, and leaves. As with previous efforts at Aspen FACE (King *et al.*, 2005), we based these equations on measurements of stem diameter (1.3 m in height) conducted immediately prior to the harvest and used ANCOVA (Littell *et al.*, 2006) to determine whether a single equation could be used for a species across communities and treatments. For the C concentration analysis of stems and branches, we created a biomass-weighted sample of these two pools for each tree.

Coarse ( $\geq 2\ mm$  diameter) and herbaceous roots were sieved from the excavated soil. Fine roots (<2 mm) and soil were sampled from the walls of the soil pit by removing 10 soil cores (5 cm diameter  $\times$  10 cm length) in each 10 cm depth increment to 1 m. This sampling regime was designed to create robust estimates of how the soil C and fine root pools varied with depth. To estimate coarse root mass, we used Akaike's information criterion (AIC) to evaluate several predictive variables. These variables were aboveground tree mass, fine root mass, and leaf mass. The final model for coarse root mass ( $r^2 = 0.904$ ) used aboveground tree mass and fine root mass, with no significant interactions with treatments or communities ( $P > 0.1$ ). The dead root pool was estimated by applying the observed ratio of live to dead roots in samples collected in 2005 and 2008 to our annual estimates of fine root biomass (Pregitzer *et al.*, 2008; Zak *et al.*, 2011).

The pools of dead wood and dead coarse roots were estimated by combining our annual observations of tree mortality with decay rates observed in this region. Tree mortality was assessed late in each growing season and trees were considered dead if no live foliage was observed. Branch loss occurred rapidly in aspen after mortality (Vanderwel *et al.*, 2006). Consequently, we divided branch mass into orders based on the fractions present in the 2009 harvest, subtracted indeterminate-, first-, and second-order branches in the second year, then an additional order in subsequent years, amounting to a loss of 15–30% of branch mass per year. We assumed dropped branches were included within our samples of the organic soil horizons. We split coarse roots into the root crown (directly below the stem) and the noncrown roots (King *et al.*, 2005) and assumed the noncrown fraction of the coarse roots would appear in the dead root pool (see the preceding paragraph). For wood decay, we used a rate constant (0.09) estimated for aspen (Gough *et al.*, 2007). We assumed root crowns decayed

at a rate of 0.15 yr<sup>-1</sup> (Fahey *et al.*, 1988; Fahey & Arthur, 1994).

### Objective 2: Quantifying NPP

Previous NPP estimates at Aspen FACE included only the first 6 years (1998–2003; King *et al.*, 2005) or last 3 years of the experiment (2006–2008; Zak *et al.*, 2011), had been constructed using different allometric equations and assumptions, and excluded some small components of NPP. Consequently, estimating cumulative NPP was not a simple combination of earlier analyses. We considered NPP to include fine roots (<1 mm diameter), small roots (1–2 mm diameter), coarse roots (>2 mm diameter), stem, branches, leaves, groundcover vegetation, and other plant litter (e.g., reproductive litter). We did not estimate NPP for the partial year of 2009 during the harvest because there was ambiguity in dividing annual productivity estimates for ecosystem components that likely exhibit seasonal dynamics (e.g., fine roots, reproductive litter, etc.).

The production of wood (branches and stem) and coarse roots (>2 mm diameter) was estimated as the annual change in biomass. Allometric equations were based on stem diameter, which was measured annually (1997–2008) in September/October. Prior to 2003, biomass estimates for wood and coarse roots were derived from species-specific allometric equations developed from trees harvested in 2000 and 2002 (King *et al.*, 2005). From 2003 to 2008, the biomass estimates for the stem, branches, and coarse roots of each tree were derived from a combination of the 2000/2002 allometric equations and the 2009 allometric equations. In this 6 year period, the 2009 equations were applied to trees exceeding the maximum diameter of the trees harvested in 2000/2002 and the 2000/2002 equations were applied to trees smaller than the minimum diameter harvested in 2009. Within the range of diameter overlap between the 2000/2002 and 2009 harvests, we employed both sets of equations and increased the contribution of the 2009 equations linearly from 0% in 2002 to 100% in 2009.

In contrast with the 2009 harvest, the 2000/2002 harvests defined the stem and branches as a single pool (wood). To assign the proper C concentration values to this wood, we needed to calculate the contribution of branches to total wood mass. For 1998–2002, we assigned wood biomass a branch fraction of 100% in 1997 and linearly decreased this fraction each year to the level calculated by applying the 2009 allometric equations to the 2003 stem diameter data. For 2003–2008, we used the 2009 allometric equations to calculate the contribution of branches to total wood mass. We derived tree-level estimates of coarse root biomass from the stand-level 2009 equations by assuming that each tree's contribution to the stand-level coarse root pool was proportionally equal to its contribution to stand-level aboveground biomass.

Our techniques for estimating fine root production differed slightly for 1998–2001 compared to 2002–2008. For 2002–2008, we relied on previously published data derived from repeated root sampling and minirhizotron observations (2002–2005: Pregitzer *et al.*, 2008; 2006–2008: Zak *et al.*, 2011). Prior to 2002, we used an allometric approach to estimate root mass (King

*et al.*, 2005) and estimated productivity as the amount of root growth needed to match the annual increase in biomass given the rate of root mortality observed in the 2002–2005 minirhizotron data (Pregitzer *et al.*, 2008). Estimates for the production small roots (1–2 mm diameter) were conducted similarly, but in the absence of direct observations we assumed that small roots had a life span three times longer than fine roots (Matala *et al.*, 2003). Fine and small root production estimates are only for roots within the top 25 cm of the soil, but this encompassed most root production in these young forests. At the end of the study, 71.1 ± 1.2% of all fine roots were contained within the top 30 cm of soil and further, root turnover decreases with depth (Joslin *et al.*, 2006).

For leaf production, we relied on previously published data from litter trap collections that occurred from 2002 to 2008 (Tallhelm *et al.*, 2012). For leaf production prior to 2002, we matched the litter trap data in 2002 with allometric estimates of leaf mass for each species in create a correction factor to account for differences in mass due to processes such as retranslocation and indeterminate growth. We then applied this correction factor to allometric estimates of leaf mass from 1998 to 2001. The C concentration of leaf litter was measured annually from 2002 to 2008 (Tallhelm *et al.*, 2012). We applied the 2002 C concentration data to our 1998–2001 estimates of leaf mass. The litter traps were also used to estimate the production of other plant litter (e.g., bud scales, unidentifiable fragments, etc.) from 2002 to 2008. We calculated the ratio of 'other' litter to leaf litter within each community in 2002 and then applied this ratio to the 1998–2001 leaf production data in order to estimate the production of this material in the years before litter trap deployment. We assumed this material had the same C concentration as the leaf litter.

The aboveground portion of the groundcover vegetation was sampled in 2004 (Bandeff *et al.*, 2006) and 2009. Prior to 2000, this vegetation was controlled by repeated herbicide treatments (Bandeff *et al.*, 2006). We assumed that the aboveground groundcover mass increased linearly from zero in 1999 to the observed 2004 values. We also used linear interpolation during 2005–2008, although there were only small differences in groundcover mass when comparing the 2004 and 2009 samples (C.E. Company, K.S. Pregitzer, unpublished data). For this vegetation, we assumed that annual aboveground production was equivalent to aboveground biomass. Nontree (herb) roots were separated from tree roots and quantified in the samples used to calculate tree fine root turnover in 2002–2008. Root mass prior to 2002 was estimated assuming the root to shoot ratio was unchanged. We assumed these roots had the same production dynamics as tree fine roots. The biomass of aboveground and belowground parts was converted to C content based on samples collected in 2009.

### Objective 3: Identifying characteristics important for $NPP_{tree}$

We evaluated several canopy attribute stand productivity models using cumulative data (1998–2008). We tested (i) the N Productivity Model (Ågren, 1983), which describes increasing  $NPP_{tree}$  with canopy N but a diminishing marginal increase as

foliar biomass accumulates; (ii) the Reich model (Reich, 2012), which predicts productivity based upon stand leaf area index (LAI,  $m^2 m^{-2}$ ), foliar N concentration, and their interaction (LAI  $\times$  N); and (iii) a model developed from remote sensing (Smith *et al.*, 2002) that predicts a base rate of productivity (an intercept) and greater rates of productivity as foliar N concentration increases. We also tested versions of the N Productivity Model that used LAI or canopy N as the independent variables explaining the diminishing return of canopy N. Furthermore, because the decline in N productivity is predominately caused by reductions in light availability (Ågren, 1983), which decreases exponentially (Binkley *et al.*, 2013), we included variants in which N productivity declined exponentially rather than linearly. We fit these models with the SAS (Version 9.1.3, SAS Institute, Cary, NC, USA) MIXED procedure (Littell *et al.*, 2006) and used the ANCOVA output to identify which experimental factors were likely to improve model fit. We then iteratively added experimental factors to the models, first adding the factors identified as significant within the ANCOVA and then testing additional factors. We used corrected AIC (AICc) for model selection. The MIXED procedure cannot accommodate exponential models, so for these models we parameterized and tested effects on the N productivity decline using log-transformed N productivity data.

Leaf area index estimates used published data (2002–2008; Talhelm *et al.*, 2012) and data generated by applying 2002 specific leaf area ( $m^2 g^{-1}$ ) to prior leaf production estimates

(1998–2001). Samples for leaf N concentration were taken in 2001, 2004 (Zak *et al.*, 2007), 2007, and 2009 (Zak *et al.*, 2011). We used the 2001 values for 1998–2001 and used linear interpolation between other samplings.

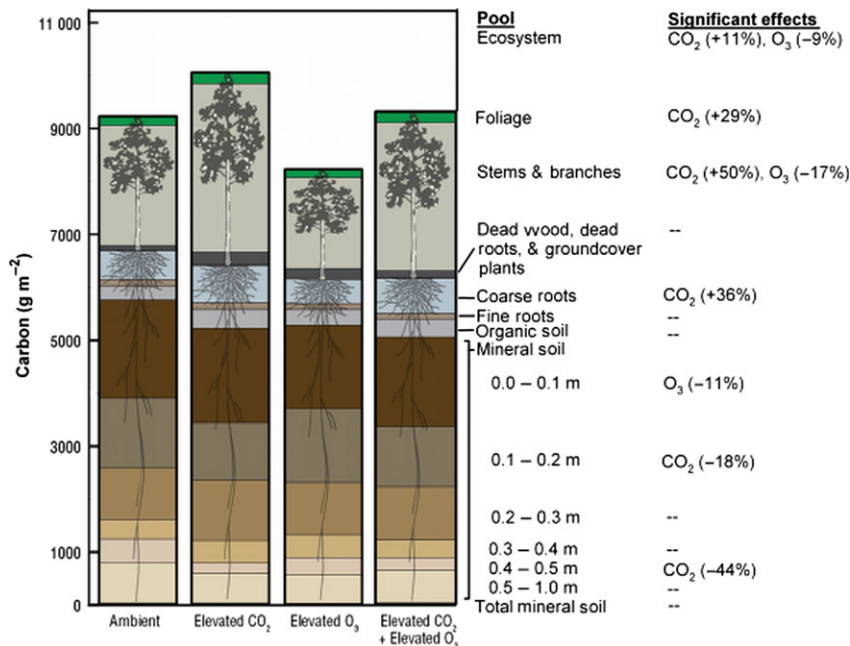
*Statistics*

Analyses were conducted as a randomized complete block design with a split-plot (repeated measures where necessary) using the SAS MIXED procedure.

**Results**

*Objective 1: Quantifying ecosystem C content*

After 11 years, the two treatments had opposite and nearly equal effects on ecosystem C content (Fig. 1): elevated CO<sub>2</sub> increased ecosystem C content by 11%, whereas elevated O<sub>3</sub> decreased ecosystem C content by 9%. Total ecosystem C content and all individual C pools, aside from foliar C and groundcover plant C, responded similarly to the treatments across communities (Table S1). There were also no significant interactions between CO<sub>2</sub> and O<sub>3</sub> for any of the largest C pools (Table S1). Total ecosystem C content in the interaction



**Fig. 1** Ecosystem carbon content after 11 years of fumigation at the Aspen FACE experiment. Data are averaged across the three forest community types and include soil to 1 m in depth. The height of each bar segment represents mean size of each pool and the total bar height represents ecosystem C content for each treatment. For simplicity, soil C below 0.5 m in depth is grouped into a single pool because there were no significant treatment effects. Significant ( $P \leq 0.05$ ) effects of the treatment gases and the size of these effects (%) are shown to the right of the figure. Pools without significant treatment effects are denoted with '--'. With the exception of two small pools (foliage, groundcover plants), there were no significant treatment  $\times$  community interactions. More detailed results can be found in Tables S1 and S2.

treatment (elevated CO<sub>2</sub> and O<sub>3</sub>) did not significantly differ from that under current ambient conditions (Fig. 1). The treatment effects on ecosystem C content resulted from differences in tree biomass, particularly woody tissues (branches, stem, and coarse roots), and lower C content in the near-surface mineral soil (Fig. 1). For tree C, the negative effect of elevated O<sub>3</sub> was smaller (−15%) than the positive effect of elevated CO<sub>2</sub> (+44%). Changes in woody tissue C accounted for 96% of the increase in tree C under elevated CO<sub>2</sub> and 98% of the decrease in tree C under elevated O<sub>3</sub> (Fig. 1, Table S1).

Leaves, fine roots, and groundcover plants together represented only 3.5% of ecosystem C (Fig. 1). While elevated CO<sub>2</sub> significantly increased fine root biomass in previous analyses (King *et al.*, 2001; Pregitzer *et al.*, 2008; Zak *et al.*, 2011), this stimulation shrank from +44% in 1999 (King *et al.*, 2001) to an average of +12% in 2006–2008 (Zak *et al.*, 2011) and was not significant at the end of the experiment (Fig. 1). Similarly, a positive O<sub>3</sub> effect on fine root biomass in the aspen community in 2002 and 2005 (Pregitzer *et al.*, 2008) was not apparent at the end of the experiment. Elevated O<sub>3</sub> shifted the distribution of fine roots toward the soil surface (O<sub>3</sub> × depth:  $P = 0.041$ ; data not shown), with slightly increased fine root C in the top 0.2 m of soil (+7 g m<sup>−2</sup>) and decreased fine root C elsewhere, particularly at 0.5–0.7 m in depth (−3 g m<sup>−2</sup>). For leaf C, the overall responses to CO<sub>2</sub> and O<sub>3</sub> were similar in magnitude to the respectively positive and negative effects of these gases on litter trap-based estimates of leaf production during the last 7 years of the experiment (Talhelm *et al.*, 2012), but less consistent. Unlike these previous estimates, the CO<sub>2</sub> effect on leaf C was significant only in the two mixed species communities (CO<sub>2</sub> × Community:  $P = 0.001$ ) and there was no significant O<sub>3</sub> effect (Table S1). Groundcover plant C had a complex response to the treatments (CO<sub>2</sub> × O<sub>3</sub> × community:  $P = 0.004$ , Table S1), which was likely a consequence of differences in light availability (Bandeff *et al.*, 2006).

We also assessed tree C content at the species-level, though we did not include fine roots because they were not identified by species. For total tree C content (branches, stems, coarse roots, and leaves), the two species within the aspen-birch community responded similarly to the treatments (Treatment × Species:  $P > 0.7$ ) and the proportion of community tree C represented by aspen was not influenced by CO<sub>2</sub> or O<sub>3</sub> ( $44 \pm 4\%$  aspen;  $P > 0.69$ ). However, there was not a uniform treatment response within the aspen-maple community: elevated CO<sub>2</sub> increased aspen tree C by 76% and decreased maple tree C by 32% (CO<sub>2</sub> × species:  $P < 0.001$ ), while elevated O<sub>3</sub> decreased aspen tree C by 22% and changed maple tree C by <1% (O<sub>3</sub> × species:  $P < 0.001$ ). The fraction of community tree C that was

maple C decreased by 10% under elevated CO<sub>2</sub> ( $P = 0.002$ ). In interpreting the treatment effects on maple, the competitive status of this species should be noted: the shade intolerant and fast growing aspen represented 87% ( $\pm 2\%$ ) of tree C within this community and was taller throughout the experiment than the shade tolerant and slow growing maple (Figure S1). In comparison, height differences were not significant between aspen and birch until the final full year of the experiment (Figure S1). The species-level treatment effects in tree C content we observed are similar to those observed in analyses of tree N content (Zak *et al.*, 2012) and leaf production (Talhelm *et al.*, 2012). The one notable difference was that these previous analyses did not observe an O<sub>3</sub> × species interaction in the aspen-maple community. However, the O<sub>3</sub> × species interaction for leaf C in this community was also not significant in our analysis ( $P = 0.481$ ).

Neither CO<sub>2</sub> nor O<sub>3</sub> affected the total amount of C in the top 1 m of mineral soil. However, each gas significantly decreased mineral soil C content in one of the two depth increments nearest to the surface: soil C within the top 0.1 m of mineral soil was lower under elevated O<sub>3</sub>, whereas soil C from 0.1 to 0.2 m in depth was lower under elevated CO<sub>2</sub> (Fig. 1; Table S2). These portions of the soil contained more C than any other individual soil depth increments (Table S2). Soil C was also lower under elevated CO<sub>2</sub> at 0.4–0.5 m in depth (Fig. 1); but there were no additional treatment effects on soil C. The observed differences in soil C were in apparent contrast with a previous analysis of the top 0.2 m of mineral soil, wherein the only significant treatment effect was that soil C in the aspen community accumulated more slowly under elevated CO<sub>2</sub> (Talhelm *et al.*, 2009). However, analyzing the top 0.2 m of soil as a single increment produced a result that was consistent with the earlier analysis (CO<sub>2</sub> effect in the aspen community:  $P = 0.085$ ).

### Objective 2: Quantifying NPP

Cumulatively, NPP increased by 39% under elevated CO<sub>2</sub> ( $P < 0.001$ ), decreased by 10% under elevated O<sub>3</sub> ( $P = 0.026$ ), and varied by more than 27% across communities ( $P < 0.001$ ). Interactions were not significant between treatments ( $P = 0.661$ ) or between the treatments and communities ( $P > 0.65$ ). Overall, tree productivity (NPP<sub>tree</sub>) comprised 95% of cumulative NPP. Treatment effects on cumulative NPP<sub>tree</sub> (Table S3) were slightly larger than those for overall NPP, with a 42% increase in NPP<sub>tree</sub> under elevated CO<sub>2</sub> and an 11% decrease in NPP<sub>tree</sub> under elevated O<sub>3</sub>. There were no significant interactions between the treatments or between the treatments and communities for NPP<sub>tree</sub>.

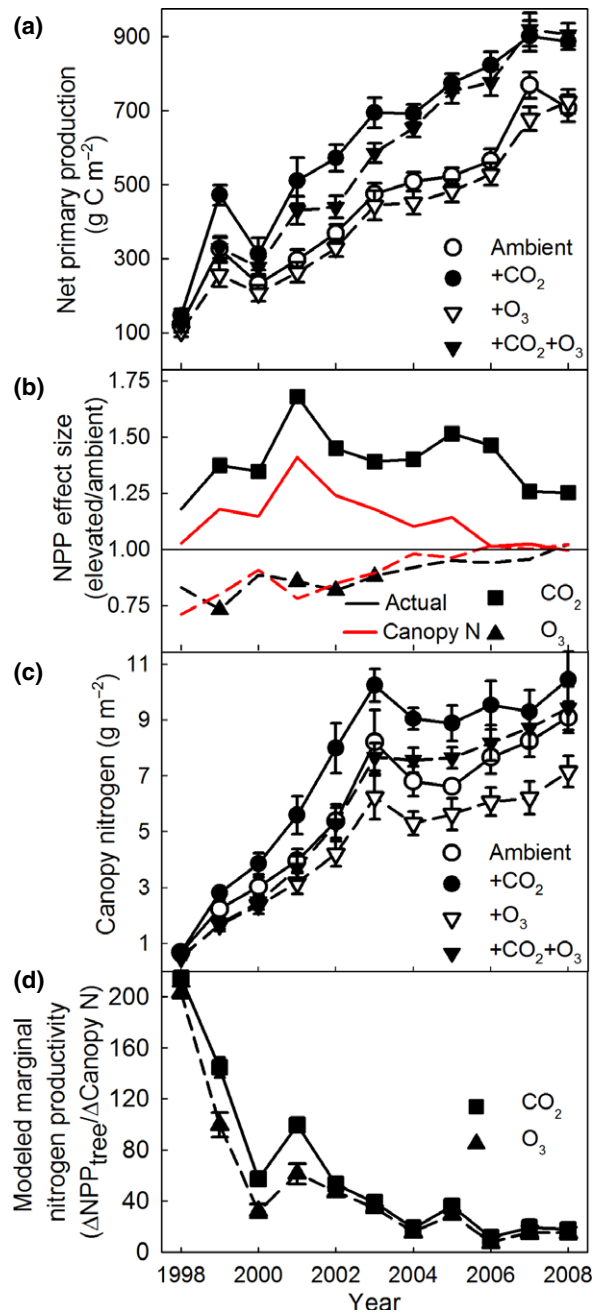


Annual NPP increased greatly during the experiment across all treatments (Fig. 2a), with the exception of three individual years: 2000, 2004, and 2008. A previous analysis found that summer photosynthetic photon flux (PPF) strongly influenced tree growth and that 2000 and 2004 had comparatively low summer PPF (Kubiske *et al.*, 2006). This analysis has not yet been extended to 2008. As noted in the analyses of NPP during the first 6 years and final 3 years of the experiment (King *et al.*, 2005; Zak *et al.*, 2011), the treatment effects on NPP were dynamic, particularly for O<sub>3</sub> (Fig. 2a). From our comprehensive analysis, it is clear that the O<sub>3</sub> effect on NPP gradually disappeared during the final 7 years of the experiment (dashed black line in Fig. 2b). Specifically, the O<sub>3</sub> effect on NPP<sub>tree</sub> declined from a peak of -95 g m<sup>-2</sup> in 2002 (*P* = 0.002) to -17 g m<sup>-2</sup> in 2008 (*P* = 0.554; linear *r*<sup>2</sup> = 0.66, *P* = 0.026). This diminishing impact of elevated O<sub>3</sub> occurred despite persistent negative effects of elevated O<sub>3</sub> on canopy N (Zak *et al.*, 2011; Talhelm *et al.*, 2012; Fig. 2c, *P* < 0.05 in 1999–2008), which only changed from -1.9 g m<sup>-2</sup> in 2002 to -1.5 g m<sup>-2</sup> in 2008. Over a similar time period, the absolute effect of elevated CO<sub>2</sub> on NPP<sub>tree</sub> was fairly consistent, changing from +189 g m<sup>-2</sup> in 2001 to +200 g m<sup>-2</sup> in 2008 and peaking at +261 g m<sup>-2</sup> in 2005 (linear *r*<sup>2</sup> = 0.24, *P* = 0.223). However, the relative effect of elevated CO<sub>2</sub> on NPP<sub>tree</sub> declined linearly from +68% in 2001 to +25% in 2008 (*r*<sup>2</sup> = 0.58, *P* = 0.029). Elevated CO<sub>2</sub> increased canopy N (Zak *et al.*, 2011; Talhelm *et al.*, 2012; Fig. 2c), an effect that did not consistently change in absolute or relative terms between 2001 and 2008 (linear *r*<sup>2</sup> < 0.28, *P* > 0.19).

Excluding C in the mineral soil, much of which existed prior to the experiment (Talhelm *et al.*, 2009), variation in ecosystem C content exhibited a strong positive relationship with cumulative NPP (Fig. 3). This relationship was not affected by CO<sub>2</sub> or O<sub>3</sub>, except in the aspen community, wherein exposure to elevated O<sub>3</sub> resulted in less C content than expected given the estimated amount of cumulative NPP. Although elevated O<sub>3</sub> decreased both soil C within the top 0.1 m of the mineral soil and cumulative NPP, there was not a clear link between soil C at this depth and forest productivity: regression relationships with cumulative estimates of NPP (Figure S2), total plant litter, aboveground litter, and fine root mortality were not significant (*P* > 0.25). In comparison, mineral soil C at 0.1–0.2 m in depth was negatively related to cumulative NPP (*P* = 0.005, Figure S2).

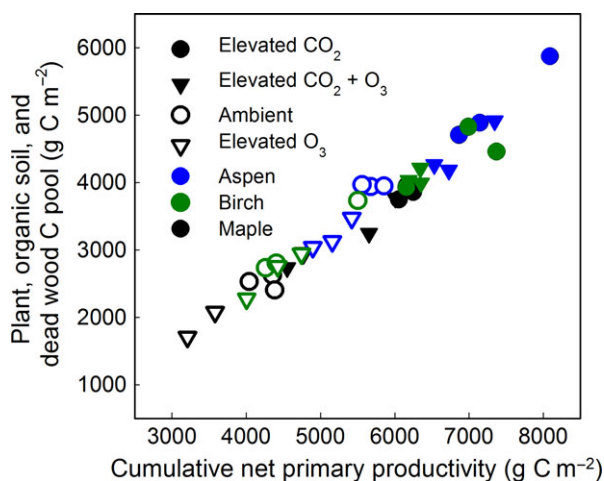
*Objective 3: Identifying characteristics important for NPP<sub>tree</sub>*

In the selected model for NPP<sub>tree</sub> (Table S4), stands with more cumulative canopy N (g foliar N m<sup>-2</sup> of ground



**Fig. 2** (a) NPP, (b) actual NPP effect sizes and effects modeled from canopy N differences, (c) canopy N, and (d) marginal N productivity  $[(NPP_{tree(elevated)} - NPP_{tree(ambient)}) / (Canopy N_{(elevated)} - Canopy N_{(ambient)})]$ . In (b), black lines show actual NPP effect sizes (elevated/ambient, 1 = no effect) and red lines show effect sizes modeled from canopy N differences; black symbols shown in (b) only when actual NPP effects are significant (*P* < 0.05). In (b), groundcover plants are assumed to be unresponsive to modeled changes in NPP<sub>tree</sub>. Bars are ±1SE.

area) had greater cumulative NPP<sub>tree</sub> (Fig. 4a), but N productivity (NPP<sub>tree</sub> per canopy N) decreased as canopy N accrued (Fig. 4b; *sensu* Ågren, 1983). The

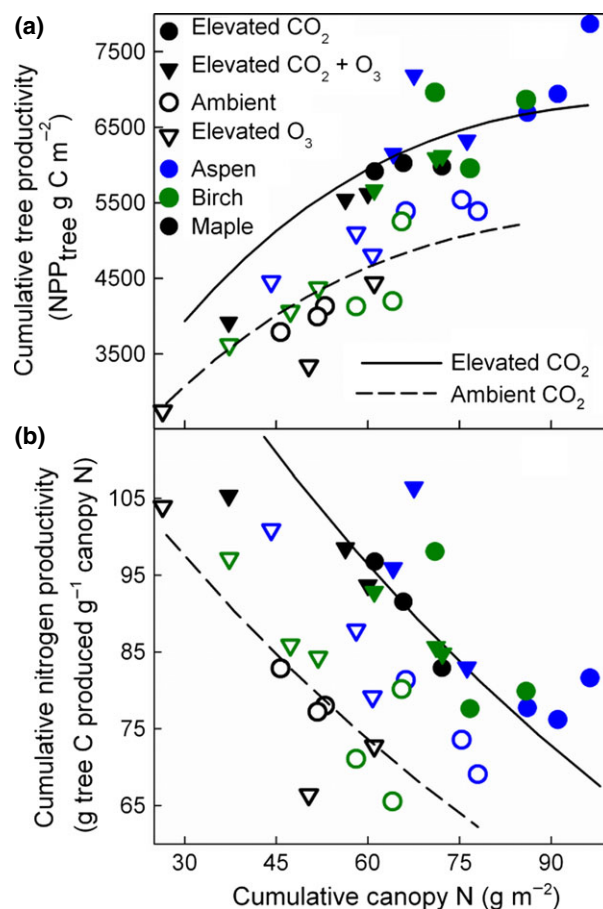


**Fig. 3** The relationship between cumulative NPP (through 2008) and C stored within the plant, dead wood and roots, and soil organic horizon pools at the conclusion of the experiment (2009). The overall regression fit is  $r^2 = 0.96$ , although the amount of ecosystem C relative to NPP is smaller under elevated O<sub>3</sub> in the aspen community (O<sub>3</sub> × Community:  $P = 0.003$ ).

decrease in N productivity with canopy N accrual was exponential in the selected model (Table S5,  $r^2 = 0.93$ ). However, there was not a substantial difference in model fit or AICc between the selected exponential model and one with a linear decline in N productivity with canopy N accrual ( $r^2 = 0.92$ , Table S4). Cumulative canopy N, leaf area ( $\text{m}^2 \text{m}^{-2}$ ), and canopy leaf mass ( $\text{g m}^{-2}$ ) were correlated with each other ( $n = 36$ ,  $r > 0.80$ ,  $P < 0.001$ ; Table S3). Likewise, annual canopy N (Fig. 2c), leaf area (Figure S3), and canopy leaf mass (Talhelm *et al.*, 2012) responded similarly to the treatments through time. However, canopy N was the best predictor of  $\text{NPP}_{\text{tree}}$  (Table S4).

Neither CO<sub>2</sub> nor O<sub>3</sub> affected the rate at which N productivity decreased with canopy N accrual (i.e. slopes in Fig. 4b were not different:  $P > 0.25$ ). Cumulative  $\text{NPP}_{\text{tree}}$  was greater under elevated CO<sub>2</sub> because of increases in both canopy N content (+28%,  $P < 0.001$ ) and the maximum rate of N productivity (N productivity<sub>max</sub>, the  $y$ -intercept in Fig. 4b; +28%,  $P < 0.001$ ). Communities also differed in both of these traits ( $P < 0.035$ ). In contrast, the negative effect of elevated O<sub>3</sub> on cumulative  $\text{NPP}_{\text{tree}}$  resulted from decreased canopy N (−21%,  $P < 0.001$ ), as there was no meaningful impact on cumulative N productivity<sub>max</sub> (−2%,  $P = 0.659$ ).

To understand the annual treatment effects on NPP, we parameterized the selected cumulative  $\text{NPP}_{\text{tree}}$  model ( $\text{Canopy N} \times e^{[x \times \text{Canopy N}]}$ ) with annual data, again using AICc for model selection. The annual



**Fig. 4** Cumulative canopy N in relation to (a) cumulative tree productivity and (b) N productivity, with lines displayed representing mixed model estimates of these relationships (community effects not shown for simplicity). Slopes in (b) do not differ, but intercepts differ by community ( $P = 0.031$ ) and between ambient CO<sub>2</sub> and elevated CO<sub>2</sub> ( $P < 0.001$ ). Ozone effects on the slopes and intercepts were not significant ( $P > 0.25$ ). The simplified model in (a) has a fit of  $r^2 = 0.87$ . Community effects in the full model (Table S5;  $r^2 = 0.93$ ) shift the lines vertically.

models differed from the cumulative model in several years: O<sub>3</sub> affected N productivity<sub>max</sub> (positive effect: 1998, 2000; negative effect: 2006) and the rate at which N productivity declined (faster declines: 1999, 2000), and there was no community effect on N productivity<sub>max</sub> in 2003.

Two further analyses provided additional insight into the annual treatment effects on NPP. In both analyses, we isolated the influence of canopy N on NPP by applying the annual  $\text{NPP}_{\text{tree}}$  model for the elevated CO<sub>2</sub> or O<sub>3</sub> treatments to the matching ambient stands (18 pairs at the ring-section level). In the first analysis (red lines in Fig. 2b), the modeled effects on NPP created by the differences in canopy N between ambient and elevated O<sub>3</sub> (dashed red line) closely matched the observed O<sub>3</sub> effects on NPP (dashed black line) in terms

of both annual effect size ( $r = 0.82$ ,  $P = 0.002$ ) and cumulative effect on NPP (modeled NPP effect of  $-12\%$  vs. actual effect of  $-10\%$ ). For elevated  $\text{CO}_2$ , the modeled effects on NPP created by the differences in canopy N (solid red line) were strongly correlated with the observed effects on NPP (solid black line;  $r = 0.80$ ,  $P = 0.003$ ), but the modeled effects underestimated the total  $\text{CO}_2$  effect on an annual and cumulative basis (modeled cumulative NPP effect of  $+14\%$  vs. actual effect of  $+39\%$ ). In the second analysis (Fig. 2d), we created annual estimates of the marginal increase in  $\text{NPP}_{\text{tree}}$  caused by additional canopy N ( $\Delta\text{NPP}_{\text{tree}}/\Delta\text{N}$ ). Here, marginal N productivity decreased by more than a factor of 10 during the experiment, meaning that differences in canopy N created by elevated  $\text{CO}_2$  or  $\text{O}_3$  had gradually smaller impacts on NPP.

## Discussion

### Objective 1: Quantifying ecosystem C content

At the decadal time-scale of our experiment, changes in total ecosystem C content (Fig. 1) were consistent with our *a priori* hypotheses: C content was enhanced by elevated  $\text{CO}_2$  and decreased by elevated  $\text{O}_3$ . In the interaction treatment (elevated  $\text{CO}_2$  and  $\text{O}_3$ ), the gases had counteracting influences on total ecosystem C content (Fig. 1). Aside from maple, the effects of  $\text{CO}_2$  and  $\text{O}_3$  on tree and ecosystem C content were consistent across species and communities. Although maple responded negatively to  $\text{CO}_2$  and was unaffected by  $\text{O}_3$  (Kubiske *et al.*, 2007; Talhelm *et al.*, 2012; Zak *et al.*, 2012), the unique treatment effects on this species were mediated by competition from aspen, the faster growing and more dominant species in this community. A similar result was observed at the Bangor FACE experiment, wherein the slower growing *Fagus sylvatica* did not respond to elevated  $\text{CO}_2$  when living in competition with two faster growing species (Smith *et al.*, 2013). Qualitatively, our overall findings match the broadly hypothesized effects of  $\text{CO}_2$  and  $\text{O}_3$  on forest C content (Eriksson & Welander, 1956; Sitch *et al.*, 2007; Norby & Zak, 2011; Ainsworth *et al.*, 2012). However, some key elements of the C cycle contributed to these overall results in unexpected ways.

Elevated  $\text{CO}_2$  caused an increase in ecosystem C content that was similar to that of other young temperate forests exposed to elevated  $\text{CO}_2$  in FACE experiments. In each of these forests, elevated  $\text{CO}_2$  increased tree C content (Norby *et al.*, 2004; Liberloo *et al.*, 2009; McCarthy *et al.*, 2010; Hoosbeek *et al.*, 2011; Smith *et al.*, 2013). While the accumulation of additional tree C was nearly always in woody tissues, additional tree growth at Oak Ridge FACE occurred almost exclusively as increased

fine root biomass (Norby *et al.*, 2004). Furthermore, mature temperate trees in Switzerland showed almost no growth response to 8 years of exposure to elevated  $\text{CO}_2$  (Bader *et al.*, 2013). The Oak Ridge FACE experiment (Iversen *et al.*, 2012) was also unique among these forest FACE experiments as the only site in which elevated  $\text{CO}_2$  increased soil C content (Hoosbeek & Scarascia-Mugnozza, 2009; Hoosbeek *et al.*, 2011; Phillips *et al.*, 2012). Besides our study, an open-top chamber experiment in a Florida scrub-oak forest (Langley *et al.*, 2009) was the only other forest experiment wherein elevated  $\text{CO}_2$  had long-term negative effects on soil C.

We are aware of two other free-air forest  $\text{O}_3$  fumigation experiments (Matyssek *et al.*, 2010a,b; Diaz-de-Quijano *et al.*, 2012); elevated  $\text{O}_3$  decreased tree biomass in both experiments, but neither experiment reported soil C measurements. In general, there are very few other experimental observations of the effect of  $\text{O}_3$  on soil C with which to compare our results (Talhelm *et al.*, 2009). However, coupled climate-biogeochemical cycling models predict that elevated  $\text{O}_3$  will decrease soil C (Ren *et al.*, 2007; Sitch *et al.*, 2007), as it did within the top 0.1 m of mineral soil in our study (Fig. 1). The sensitivity of woody biomass to  $\text{O}_3$  was also consistent with previous research: In a temperate forest productivity model, woody biomass was more responsive to  $\text{O}_3$  than leaves or roots (Ollinger *et al.*, 1997), a prediction supported by a meta-analysis of  $\text{O}_3$  effects on tree productivity (Wittig *et al.*, 2009).

### Objective 2: Quantifying NPP

Given our previous observations of NPP (King *et al.*, 2005; Zak *et al.*, 2011), it was unsurprising that elevated  $\text{CO}_2$  had a larger effect than elevated  $\text{O}_3$  on cumulative NPP. In the first 6 years of the experiment, elevated  $\text{CO}_2$  stimulated NPP and elevated  $\text{O}_3$  diminished NPP (King *et al.*, 2005), but only elevated  $\text{CO}_2$  caused significant changes in NPP during the final 3 years (Zak *et al.*, 2011). Variation in NPP had clear consequences for ecosystem C content: there was a strong positive relationship between ecosystem C content excluding the mineral soil and cumulative NPP (Fig. 3) and a negative relationship between cumulative NPP and mineral soil C at 0.1–0.2 m in depth (Figure S2). The amount of C in pools other than the mineral soil was lower than expected based upon cumulative NPP under elevated  $\text{O}_3$  in the aspen community (Fig. 3). This effect is consistent with an earlier increase in the production of fine roots, which are ephemeral, under elevated  $\text{O}_3$  in this community (Pregitzer *et al.*, 2008). Aside from this community  $\times \text{O}_3$  interaction, it is notable that none of the other previously observed treatment effects on fine root, groundcover, and leaf production (King *et al.*,

2005; Bandeff *et al.*, 2006; Pregitzer *et al.*, 2008; Talhelm *et al.*, 2012, Table S3) influenced the relationship between cumulative NPP and the quantity of C in pools other than the mineral soil because these tissues represented approximately 40% of cumulative NPP.

There is indirect evidence that elevated CO<sub>2</sub> stimulated both total heterotrophic respiration ( $R_h$ ) and the relative fraction of NPP respired by heterotrophs. The increase in cumulative NPP under elevated CO<sub>2</sub> included a 32% increase in leaf litter production (Table S3) and a 30% increase in fine root litter (A.F. Talhelm, K.S. Pregitzer, unpublished data), but the rate of C accumulation within the mineral and organic soil was similar or slower under elevated CO<sub>2</sub> than under ambient conditions (Talhelm *et al.*, 2009; Zak *et al.*, 2011). Increased  $R_h$  is consistent with repeated observations of greater activity of two important litter-degrading extracellular enzymes (cellobiohydrolase and *N*-acetylglucosaminidase) in the soil under elevated CO<sub>2</sub> (Edwards & Zak, 2011). Increased  $R_h$  under elevated CO<sub>2</sub> also has been observed at the Duke Forest FACE experiment (Hamilton *et al.*, 2002; Drake *et al.*, 2011) and attributed to faster turnover of root-derived C (Phillips *et al.*, 2012). These observations indicate that elevated CO<sub>2</sub> stimulates forest C cycling as a whole, including autotrophic and heterotrophic components.

The responses of soil C and  $R_h$  have important implications for the long-term effects of elevated CO<sub>2</sub> on forest biogeochemistry. It has been hypothesized that the stimulation of NPP by elevated CO<sub>2</sub> will be progressively limited by decreased N availability, a consequence of N sequestration in accumulating organic matter (Luo *et al.*, 2004). This process has not occurred in our experiment (Zak *et al.*, 2011). Instead, elevated CO<sub>2</sub> created positive feedbacks in organic matter cycling, sustaining increased canopy N (Fig. 2c) and increased tree N content (Zak *et al.*, 2012). In fact, the negative relationship between soil C at 0.1–0.2 m in depth and cumulative NPP (Figure S2) provides some indication that the opposite of progressive N limitation occurred (Zak *et al.*, 1993), wherein elevated CO<sub>2</sub> has primed the mineralization of C and N in the soil (Phillips *et al.*, 2012). Within the top 0.5 m of mineral soil, there was a strong positive correlation between the C and N pools within each increment ( $n = 36$ ,  $r > 0.95$ ); in depth increments where soil C pools were smaller under elevated CO<sub>2</sub> (Fig. 1), soil N pools were also smaller (A.F. Talhelm, K.S. Pregitzer, unpublished data). The negative relationship between soil C and cumulative NPP, together with lack of any significant relationship between NPP and soil C within the top 0.1 m (Figure S2) and the infrequency with which elevated CO<sub>2</sub> has increased soil C in other FACE experi-

ments (Iversen *et al.*, 2012), suggests that soil C accumulation cannot be simplistically linked to NPP (Phillips *et al.*, 2012).

The effects of elevated O<sub>3</sub> on C content within the mineral and organic soil were similar to those of elevated CO<sub>2</sub> (negative or neutral; Fig. 1, Talhelm *et al.*, 2009; Zak *et al.*, 2011). Unlike elevated CO<sub>2</sub>, elevated O<sub>3</sub> had little effect (+3%) on cumulative plant litter production (leaves, groundcover plants, fine roots, etc.). Here, smaller leaf production (Talhelm *et al.*, 2012; Table S3) was offset by greater fine root litter production (Pregitzer *et al.*, 2008) and groundcover plant growth (Bandeff *et al.*, 2006; Table S1). The combination of decreased surface mineral soil C (Fig. 1) and unchanged plant litter inputs suggests that elevated O<sub>3</sub> had a modest positive effect on  $R_h$ . Among litter-degrading enzymes, cellobiohydrolase activity was not consistently affected by elevated O<sub>3</sub>, but elevated O<sub>3</sub> was associated with higher *N*-acetylglucosaminidase activity within the soil Ap horizon (Edwards & Zak, 2011). We are unaware of any previous observations of  $R_h$  under elevated O<sub>3</sub>.

### Objective 3: Identifying characteristics important for $NPP_{tree}$

The AICc-selected model for cumulative  $NPP_{tree}$  was similar to the Nitrogen Productivity Model (Ågren, 1983); both models predict differences in NPP based upon canopy N and N productivity ( $NPP_{tree}/\text{canopy N}$ ), but there were two differences. First, canopy N was the predictor of N productivity in the selected model instead of leaf mass (Fig. 4b). Second, an exponential, rather than linear, function described the decline in N productivity as canopy N accumulated. This type of decline matches the widely observed pattern of canopy light absorption, wherein marginal gains in light absorption from additional foliage decrease exponentially during canopy development (Binkley *et al.*, 2013). Our model selection contrasts with a recent cross-site analysis, in which the combination of leaf area and leaf N concentration was the best predictor of aboveground forest productivity (Reich, 2012). However, a comparatively narrow range of leaf traits and environmental conditions was present in our experiment.

The decrease in canopy N under elevated O<sub>3</sub> (Fig. 2c) could be a consequence of a shift in C allocation. Elevated O<sub>3</sub> consistently limited leaf production (Talhelm *et al.*, 2012), whereas O<sub>3</sub> effects on fine root biomass were often positive or neutral (Pregitzer *et al.*, 2008). A shift in allocation belowground also occurred with the free-air O<sub>3</sub> fumigation of a spruce-beech forest (Matyssek *et al.*, 2010a; Nikolova *et al.*, 2010), a response attributed to O<sub>3</sub> impacts on cytokinin hormones. However, such a shift in allocation may not be

universal: a meta-analysis dominated by chamber experiments did not find a consistent effect of O<sub>3</sub> on tree root : shoot ratios (Wittig *et al.*, 2009).

Because NPP<sub>tree</sub> was a function of canopy N, the disappearance of the O<sub>3</sub> effect on annual NPP (Fig. 2b) despite the consistent negative effect on canopy N (Fig. 2c) might seem to indicate a weakening physiological impact of O<sub>3</sub>. However, this conclusion was not supported by our analyses. First, when parameterizing the canopy productivity model for individual years, O<sub>3</sub> was not a consistent influence. Second, the modeled O<sub>3</sub> effects on NPP based solely on canopy N differences corresponded well with the observed NPP effects (Fig. 2b). Finally, the marginal impact of canopy N on NPP declined steadily during the experiment (Fig. 2d). Together, these results suggest that rather than a weakening physiological impact of O<sub>3</sub>, decreases in canopy N caused large declines in NPP early in the experiment, but the importance of this effect diminished as canopy development increased. The weakening effect of O<sub>3</sub> on NPP (Fig. 2b) implies that long-term forest productivity may be surprisingly insensitive to O<sub>3</sub>.

The relative stimulation of NPP by elevated CO<sub>2</sub> peaked several years into experiment, then declined to approximately +25% during the final 2 years (Fig. 2b). Elevated CO<sub>2</sub> experiments frequently report similar declines in the relative stimulation of NPP with time, with this decline attributed to increasing N limitation, shifts in species composition, or ontogenetic effects (Leuzinger *et al.*, 2011). Nitrogen availability has limited the stimulation of NPP in other FACE studies in young temperate forests (e.g., McCarthy *et al.*, 2010; Norby *et al.*, 2010), but there was no evidence of this at Aspen FACE (Zak *et al.*, 2011). Likewise, the only observed effect on species composition favored aspen, the more productive species. However, there is evidence that stand ontogeny influenced the response to elevated CO<sub>2</sub>. As with elevated O<sub>3</sub>, differences in canopy N content created by elevated CO<sub>2</sub> (Fig. 2c) had large effects on NPP early in the experiment (solid red line in Fig. 2b). With time, canopy development increased and the marginal impact of canopy N on NPP declined (Fig. 2d). Thus, during the last several years of the experiment, increased N productivity (Fig. 4b) was the dominant reason NPP remained higher under elevated CO<sub>2</sub> (Fig. 2a). This implies that if the experiment had not ended, the positive effects of elevated CO<sub>2</sub> on NPP and the C content of pools other than the mineral soil (Fig. 3) would have continued until the stands were harvested or limitations were imposed by other factors. Mature forests have shown little response to elevated CO<sub>2</sub> (Bader *et al.*, 2013) and appear to be more limited by nutrient and hydraulic constraints than C uptake (Körner, 2003; Palacio *et al.*,

2014). Elevated CO<sub>2</sub> could have accelerated the eventual onset of these limitations at Aspen FACE, but this was not apparent during our experiment. Although elevated CO<sub>2</sub> increased tree height (Figure S1), it also increased leaf and canopy conductance (Uddling *et al.*, 2009). Likewise, the additional plant N accumulated under elevated CO<sub>2</sub> at the end of the experiment (Zak *et al.*, 2012) was equivalent to only 1% of total soil N (A.F. Talhelm, K.S. Pregitzer, unpublished data).

### Implications

The value of each FACE experiment is not that it is uniquely predictive of future terrestrial C cycling, but that it creates insight into the mechanisms that control forest C cycling at broader scales (Norby & Zak, 2011). Although elevated CO<sub>2</sub> and elevated O<sub>3</sub> had counteracting effects on ecosystem C content, this particular result should not be extrapolated in time or space. Differences in NPP were explained by two mechanisms: the accrual of canopy N (Fig. 2c) and the rate of N productivity (NPP/canopy N; Fig. 4). Elevated CO<sub>2</sub> enhanced NPP because it increased both canopy N and N productivity, while elevated O<sub>3</sub> only decreased canopy N. With time, increasing canopy development diminished the impact of canopy N differences on NPP, but changes in N productivity continued to be important. Consequently, the O<sub>3</sub> effect on NPP was eliminated, but the CO<sub>2</sub> effect persisted (Fig. 2). This implies that O<sub>3</sub> may not have decreased annual NPP into the future, lessening its cumulative impact. For instance, if recent (2006–2008) rates of NPP were sustained through a 30-year harvest rotation that is common for aspen in this region (Perala, 1977), the O<sub>3</sub> effect on cumulative NPP would have been only –4%. However, we cannot state that O<sub>3</sub> would have a limited impact on C cycling at this time scale because we have not identified a mechanism linking O<sub>3</sub> effects on soil C to plant productivity. At a larger spatial scale, the influences of CO<sub>2</sub> and O<sub>3</sub> on forest C sequestration depend on forest management, stand turnover, and interaction of these factors with CO<sub>2</sub> and O<sub>3</sub> at a landscape level (Körner, 2006).

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## Supporting Information

Additional Supporting Information may be found in the online version of this article:

**Figure S1.** Treatment and species differences in mean tree height through time.

**Figure S2.** Mineral soil carbon pools in (a) the top 0.1 m of soil and (b) at 0.1–0.2 m in depth, relative to cumulative NPP.

**Figure S3.** Leaf area index (LAI) through time.

**Table S1.** Carbon pool sizes for each community and treatment.

**Table S2.** Mineral soil C pool sizes by depth for each community and treatment.

**Table S3.** Cumulative values of NPP<sub>tree</sub> and canopy leaf properties.

**Table S4.** Corrected Akaike's Information Criteria (AICc) used for selection of the NPP<sub>tree</sub> model.

**Table S5.** Parameter estimates and *P* values for the selected NPP<sub>tree</sub> model (see Table S3).