ECOSYSTEMS

Effects of Long-Term Nitrogen Addition and Atmospheric Nitrogen Deposition on Carbon Accumulation in *Picea sitchensis* Plantations

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ABSTRACT

This study aimed to assess the combined effects of long-term nitrogen (N) supply and nitrogen deposition (N_{dep}) on carbon (C) accumulation within Sitka spruce [*Picea sitchensis* (Bong.) Carr.] plantations in Scotland. Six study sites established from 1970 to 1982 were periodically N-fertilized, monitored over time and commonly surveyed in 2010. Soil, aboveground biomass, and ground vegetation C stock changes were analyzed; aboveground C stocks were correlated with total additional N experienced at each site, that is, the sum of experimental N supply (N_{add}) and site-specific accumulated N_{dep} from 1900 to 2010. Results showed a positive N effect on aboveground tree C stock and no decline in tree growth was observed either during fertilization or after the latest N addition. The amount of C in litter was significantly higher in experimentally N-treated plots, whereas the amount of C in understory vegetation was higher in control plots. Pooling all the compartments (that is, understory vegetation, litter, soil, and tree biomass) the total ecosystem C content was estimated for each site, and at most sites a higher C stock was estimated for N-treated plots. Differences in aboveground C accumulation rates between treated and control plots were lower at sites with high levels of accumulated N_{dep}. Our results indicate that site-specific accumulated N_{dep} should be considered to understand tree growth responses to N fertilization.

Key words: anthropogenic nitrogen deposition; carbon stock; Sitka spruce; soil fertilization; tree growth; urea.

INTRODUCTION

Atmospheric nitrogen deposition (N_{dep}), caused by reactive nitrogen emissions via fuel combustion and intensive agricultural practice, and its deposition back to the biosphere, is considered one of the most important factors resulting from anthropogenic global environmental change (Norby 1998). At the global scale, N_{dep} is predicted to increase

nearly twofold by 2050 from its current deposition rates (Galloway and others 2004; Gruber and Galloway 2008; Schlesinger 2009). Atmospheric N_{dep} may induce long-lasting ecosystem changes over large climatic regions with profound implications for N and C biogeochemical cycles, and ultimately for the terrestrial C sink and its potential to mitigate climate change (Phoenix and others 2012).

In recent years, a beneficial role of N_{dep} on carbon uptake by nitrogen-limited forests has been proposed (Magnani and others 2007; Thomas and others 2010); however, the magnitude of the effect is still uncertain. In particular, the apparently high sensitivity of forest C sink strength to annual N input reported by Magnani and others (2007) resulted in an intense debate and the extent of carbon uptake enhancement attributed to N_{dep} has been the subject of several different estimates (De Vries and others 2008; Reay and others 2008; Sutton and others 2008; Janssens and Luyssaert 2009).

Atmospheric N_{dep} can enhance forest C uptake by different ecological mechanisms. In the long term, there can be a significant effect on soil fertility mediated by the accumulation of N in the soil, causing a change in soil organic matter stoichiometry (lower C/N ratio), that can stimulate N mineralization and N plant uptake (Aber and others 2003). In the short term, the effect could be due to direct uptake of nitrogen compounds by forest canopies that might account for a significant fraction of tree nitrogen requirements, by-passing microbial competition in the soil (Neirynck and others 2007; Sievering and others 2007). In a recent modeling exercise (Dezi and others 2010), direct canopy nitrogen uptake increased carbon storage capacity of forest ecosystems. Some experiments (Gaige and others 2007; Sheppard and others 2008; Dail and others 2009; Guerrieri and others 2011) have examined the effects of direct spraying of nitrogen onto the forest canopy, but conclusive evidence remains unavailable.

Chronic atmospheric N_{dep} can be also a threat to ecosystem function; particularly, it can lead to soil acidification and nutrient imbalances that might become relevant constraints to forest productivity (Aber and others 1998; Dail and others 2009). Increased plant biomass levels can also be associated with increased demands for water and other nutrients that ultimately limit positive N addition effects on plant growth (De Vries and others 2006). When the saturation threshold of inorganic N accumulation is exceeded, it can result in decreased productivity and biodiversity (De Schrijver and others 2008; Phoenix and others 2012), or increased tree mortality (Magill and others 2004).

In recent decades, a number of long-term N addition experiments have been carried out in forest ecosystems to explore the effects of N supply on forest growth and C pools. Re-analyzing the results of 15 long-term investigations performed in coniferous boreal forests, Hyvonen and others (2008) concluded that N addition effects on C pools in trees and soil, considered in terms of N-use efficiency (unit of C accumulated per unit of N added, kg C kg^{-1} N), were strongly dependent on soil fertility, suggesting site-specific responses.

An analysis of the combined effects of long-term N addition and atmospheric N deposition on forest carbon accumulation has not been performed so far. This is relevant in the light that even relatively low N_{dep} rates can result in important implications for ecosystem function and structure if continued for long enough. In addition, the practical forester needs to evaluate, in a specific environmental context, the utility of N fertilization as a tool to stimulate forest growth. Improved understanding of the consequences of atmospheric N deposition is fundamental to addressing its implications on forest ecosystems since cumulated N deposition (N_{cum}) can have a significant influence on plant physiological traits, such as water use efficiency, as demonstrated at the global scale recently by Leonardi and others (2012).

In the present study, we hypothesize that the interpretation of long-term N addition experiments can be improved by considering site-specific accumulated atmospheric N_{dep}. To this aim, we conducted research within six different long-term N fertilization experiments established in Sitka spruce [*Picea sitchensis* (Bong.) Carr.] plantations in Scotland. We analyzed changes over time in foliar nutrients, tree growth, soil, understory, and tree C stocks. We also related the aboveground biomass response to the total N available as a result of fertilization at each site. Finally, we interpreted the observed patterns by considering both experimental N supply and site-specific accumulated N_{dep} from 1900 to 2010.

MATERIALS AND METHODS

Experimental Forest Stands and Nitrogen Addition

Six Sitka spruce plantations in Central and North Scotland, established between 1967 and 1988 by the Forestry Commission (Forest Research, Newton, Elgin, Morayshire, UK) represented the experimental forests (Table 1). At each site, to evaluate the effects of fertilizer treatments, experimental plots

Table 1. Geographic, Environmental, and Stand Characteristics of the Experimental Sites

	Experimental sites					
	Ardross	Craigellachie	Drumtochty	Shin 1	Shin 2	Speymouth
Latitude (N)	57°47′7″	57°31′44″	56°54′15″	57°17′30″	58°02′44″	58°10′4″
Longitude (W)	4°8′46″	3°8′14″	2°28′15″	2°47′23″	4°56′56″	4°30′33″
Altitude (m)	207	350	350	205	170	155
Soil	Peaty podzol	Podzol	Peaty podzol	Peat	Peat	Podzol
Rainfall (mm y^{-1})	950	953	810	1,200	1,700	825
Year of plantation	1988	1979	1967	1982	1987	1982
Stand age in 2010 (years)	22	31	43	28	23	28
Accumulated N_{dep} at 2010 (since 1900) (t N ha^{-1})	0.739	1.145	1.765	0.891	0.900	1.482
ESC parameters						
Accumulated temperatures (AT) (degree-days above 5°C)	1,035	1,017	900	992	1,040	1,086
Continentality (Conrad index)	4.2	5.2	5.8	4.4	4.0	5.1
DAMS (windiness index)	13.8	13.6	15.6	17.4	16.9	14.1
Moisture deficit (mm)	82	89	65	73	77	104
SMR	Moist	Slightly dry	Fresh	Wet	Wet	Fresh
SNR	Very poor	Very poor	Very poor		Extremely poor	Very poor
ESC yield index	YC 9	YC 9	YC 8/9	YC 5	YC 5	YC 9

In the UK AT can vary between <600 and >3.000 degree-days, the Conrad index between 1 and 13, the windiness index between 3 and 36, moisture deficit between <20 and >180 mm, and the yield index between 4 and 24 m^3 ha^{-1} y^{-1}. For all sites, the same provenance from Queen Charlotte Island (Canada) was planted; in Shin 2, a Sitka spruce provenance from Alaska was also tested. Site variables include productivity estimated as site yield index (m^3 ha^{-1}) derived from ecological site classification (ESC) (Pyatt and others 2001).

(area of 400–900 m^2) were defined, according to a randomized block design. Treatments consisted of soil N additions (delivered as 46% urea prills), alone or in combination with P, K, or both. Fertilization schemes and number of replicates varied among sites (Table 2). At each site, three to four plots with and without N addition were established, resulting in *N-enriched* (treated) and *not* N-*enriched* (control) plot comparisons. At sites where N was added in combination with P, K, or both, the control included the addition of P, K, or both; therefore, in all cases the effect of N addition per se was evaluated. In all cases, tree spacing was 2 × 2 m (2,500 plants per hectare) and thinnings were not performed.

Measurement of Trees and Stand Biomass

All measurements were performed within square assessment areas (185–400 m^2) delineated in the center of each plot, to avoid edge effects. In each plot, the heights and diameters (dbh) at 1.3 m of each tree (48–100 trees per plot) were measured every 1–3 years. Aboveground stand dry biomass (B_d) for each plot was estimated using existing allometric relationships based on a sample exceeding 1,000 Sitka spruce trees following Levy and others (2004); different allometric equations were used for Sitka spruce growing on peat and podzolic soils (Minunno and others 2010). Stand C stocks were calculated as 0.5 × B_d.

Concurrent with tree measurements, needle samples were detached from the upper crown of five dominant trees in each experimental plot. Needle N, P, and K concentrations were determined, following the analytical methods reported by Allen (1977).

During summer 2010 two tree cores were extracted, from orthogonal directions at 1.30 m using a Pressler corer, from the five largest trees in N-enriched and not N-enriched plots at each sampling site (with the exception of Drumtochty). Cores were sanded under a 40× stereomicroscope, and after visual cross-dating, tree ring widths (R_w) were measured to the nearest 0.01 mm using a Dendrochronograph Lega Smil 3 (Corona and others 1989). The cross-dating quality control was conducted on the R_w temporal series using the Cofecha software (Holmes 1983). However, standardization was not performed to retain the effects of fertilization and long-term stem diameter development in treatment and control plots.

Understory Vegetation and Soil

Fieldwork was conducted at all sites (with the exception of Drumtochty) during summer 2010 to

Table 2. Fertilization Treatments at Different Study Sites

Site	Nutrients added	Year of treatment	Number of replicates	Mean N dose (kg ha^{-1})[1]	Accumulated N supply (kg ha^{-1})	Accumulated P supply (kg ha^{-1})	Accumulated K supply (kg ha^{-1})	Weed control
Ardross	NPK	1994–2000–2003	4	151	453	118	108	–
Craigellachie	NP	1987–1990–1993–1996	3	160	640	63	–	+
Drumtochty A	NP	Every 2 years since 1967–1985	3	162.3	1,623	150	–	+
Drumtochty P	NP	1973–1977–1984	3	152.3	457	150	–	–
Shin 1	NP	1987–1990–1993–1996–2002	3	154	770	210	–	–
Shin 2	NPK	1993–1996–1999–2002	4	160	640	118	208	–
Speymouth	NP	1987–1990–1993–1996–1999	3	160	800	110	–	+

Two fertilization schemes were applied at Drumtochty one with periodic (P) and one with biannual (A) N additions.
N = nitrogen; P = phosphorus; K = potassium.
[1]At Shin 1, N addition was normally 150 kg ha^{-1}, but was 160 kg ha^{-1} in 1996 and 2002. At Drumtochty, N addition changed slightly over time (from a minimum of 148 to a maximum of 171 kg ha^{-1} added). At all the other sites the mean dose value correspond to the dose applied at each fertilization.

estimate C stocks in the understory vegetation and soil. In each plot, a 50 × 50 cm frame was randomly positioned along each of three to five selected lines between the trees, avoiding furrows and ridges. Six to ten points were randomly sampled in each plot along the lines. Vegetation inside the frame was totally removed; grasses and shrubs were separated from mosses, lichens, and litter. Mosses, lichens, and litter were separately sampled from a 15 × 15 cm square within the main 50 × 50 cm frame. All material was brought to the lab, oven dried at 80°C for 48 h, and weighed to the nearest mg. Total moss, lichen, and litter biomass in each 50 × 50 cm square frame was estimated from the biomass ratios measured in each 15 × 15 subsample, with the error estimate determined accounting for error propagation from sample to square frame, and from square frame to plot.

Following vegetation sampling, a soil sample was extracted from the center of each 50 × 50 cm square frame. At sites with podzolic soils, a slide-hammer corer (or an auger, internal diameters of 5.7 cm) was used down to a depth of about 30 cm at six points in each plot. After extraction, the cavity depth and soil core were measured, and the soil core was subdivided into three layers, including an upper organic horizon, and two lower 10-cm-deep layers. For sites with a peaty soil, a square cavity was excavated in the center of the frame, and a core was extracted from one exposed vertical soil wall at a depth of 20 cm, using a bulk density cylinder. Because of differences in maximum soil depth employed across soil types, our analyses of changes in soil C stocks are limited to within-sites experimental comparisons, as opposed to across sites comparisons. Soil samples were brought to the lab, oven dried at 105°C for 24 h, and weighed to the nearest mg. Bulk density of each soil sample was calculated as the ratio between dry weight and volume. Soil samples were subsequently homogenized in a mortar, and a soil subsample (1 g approximately) was weighed to the nearest 0.01 mg (W_i), placed in a muffle furnace for 5 h at 450°C, cooled in a desiccator to room temperature, and weighed again (W_e). The percentage mass loss on ignition (W_{loss}) was calculated as $(W_i \times 100)/(W_i - W_e)$. A linear transfer function converting values of W_{loss} into carbon concentration (from C/N analyser) for the same soil types was computed combining existing datasets (Zerva and Mencuccini 2005), and applied to estimate carbon concentration in each soil sample. Carbon concentration, bulk density, and soil depth were then used to estimate soil carbon content per ground area.

Atmospheric N Deposition

Nitrogen deposition (N_{dep}) from 1970 to 2010 was estimated for the UK at a spatial resolution of 5×5 km^2 with the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME, Vieno and others 2010). Data are available for 1970, 1980, and 1990 (that is, at decadal intervals) and then for 1995, 2000, and 2005 (5-year intervals), a prediction for 2020 is reported. Annual deposition values were linearly interpolated between the modeled years. The FRAME model can be run at resolutions of 1×1, 5×5, and 50×50 km^2. Model validations using data from national monitoring networks was carried out for ammonia (Hallsworth and others 2010), gas and aerosol concentrations of N compounds and wet N deposition (Dore and others 2007), and gaseous NO_2 (Dore and others 2012). Moreover, good agreement was found by comparing FRAME model predictions with the observations-derived UK deposition (Smith and others 2000) for the years 1990–2005 (Matejko and others 2009). In general, the use of FRAME at 5×5 km^2 resolution gives reasonable correlations with observed data (slopes between 0.75 and 0.97 with the exclusion of HNO_3) and high values of R^2 (between 0.58 and 0.92), suggesting that FRAME is able to represent accurately the spatial distribution of gas and particle concentration and deposition fluxes over the UK (Dore and others 2007).

To reconstruct total N_{dep} prior to 1970, the TM4 atmospheric transport model output from the EU RETRO project (Schultz and others 2007) was used. The TM4 results have been extrapolated back in time using decadal emission estimates from the EDGAR-HYDE dataset (van Aardenne and others 2001). The FRAME model runs were derived from the National Atmospheric Emissions Inventory estimates.

The TM4 model is unlikely to give precise deposition values for a specific UK site due to the highly spatial variability of N_{dep} in the UK (Dore and others 2012). The FRAME model with a resolution of 5×5 km^2 represents the spatial variability of N_{dep} in UK better than the TM4 model with a resolution of $0.5° \times 0.5°$. Therefore, the historical TM4 depositions data were corrected to match the FRAME model for the overlapping years and this relationship was used to rescale the TM4 historical data prior to 1970. The FRAME model also calculates land-use specific deposition rates for forest, moorland, and grassland. Therefore to correct the TM4 data, the appropriate land-use was used to account for changes in forest coverage at any specific sites. Finally, accumulated N_{dep} from 1900 to 2010 were estimated (Table 1).

Site Productivity Estimates

To compare environmental conditions and productivity levels across sites, the suitability of individual stands to produce timber was predicted on the basis of six ecological site classification (ESC) factors (Pyatt and Suarez 1997; Pyatt and others 2001). Four climatic factors (i) accumulated temperature, (ii) moisture deficit, (iii) windiness (by Detailed Aspect Method Scoring; DAMS), (iv) and continentality (Conrad index) and two soil quality factors (v) soil moisture regime (SMR), and (vi) soil nutrient regime (SNR), were evaluated.

ESC–DSS (Ray 2001) calculates the climatic indices from user input of grid reference and elevation of the site, and site yield index is modified by accumulated temperature and the most constraining site factor. The site yield index was calculated for all plots assuming no nutrient addition and no thinning (Table 1).

Statistical Analyses

Mean differences between treatments and blocks were assessed by two-way ANOVAs (one-way ANOVA if blocks were not significant, repeated-measures ANOVA on nutrient time series), within the framework of general linear models, using the GLM procedure available in the SPSS statistical package, version 17 (SPSS Inc. 2008).

RESULTS

Atmospheric N Deposition

Atmospheric N_{dep} (Figure 1), as simulated by the FRAME model, shows marked differences
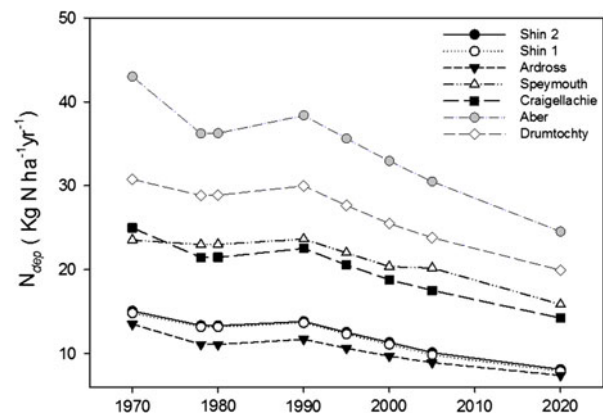

Figure 1. Temporal variation of atmospheric nitrogen deposition (N_{dep}) at the study sites as calculated by the FRAME model. The deposition values at Aber (in Wales) are also given because it was the site of a past N manipulation experiment in a comparable Sitka spruce plantation (for example, Emmett and others 1998).

among sites, with a general decreasing trend over the last 30 years, likely due to the stabilization of NO_x emissions after the implementation of air pollution policies (Lamarque and others 2010).

Aboveground Carbon Stock and Foliar Nutrient Concentration

Above ground carbon stock (C_{ag}) showed distinct trends for control and treated plots throughout stand development, with the treatment group displaying higher C_{ag} rates of increase and higher final C_{ag} values at all sites, with the exception of Drumtochty, where the largest C_{ag} values did not differ between control and treated plots for the whole duration of the observations (Figure 2). Estimates of inherent site productivity suggest that two main levels of inherent site productivity occurred across the sites, which differed primarily because of soil type (that is, podzol > peat) and without other significant climatic constraints (Table 1).

Long-term patterns of radial growth showed clear differences in ring width (R_w) values between treated and control plots for most of the period while by 2010, after fertilizations had ceased, no significant differences were observed any longer (data not shown). At all sites, with the exception of Craigellachie, accumulated differences in stem radial growth between control and treated plots (Figure 3) revealed an initial increase followed by a tendency to plateau, which was particularly evident in Ardross and Speymouth, where fertilizations had ceased in 2003 and 1999, respectively.

Foliar N concentration was significantly different between treatments (T) and years (Y) at all sites, and the T × Y interaction was also significant; in several cases, significant differences in foliar P and K concentration were not detected, whereas at most sites N/P and N/K ratios exhibited significant differences between treatments (Table 3).

Temporal variations in N and P foliar concentrations and N/P and N/K ratios (Figure 4) exhibited the following trends: (i) N foliar concentrations were generally higher in treated plots, and displayed some tendency to decrease with time at Drumtochty, Shin 1, and Shin 2; (ii) P foliar concentration was often indistinguishable between treatments, whereas control plots showed a slightly higher trend compared to treated plots during the last experimental period at Shin 2A and Drumtochty, (iii) N/P and N/K ratios were often higher in treated plots and showed a tendency to decrease with time at most sites.

Understory Vegetation, Litter, and Soil

At all sites, total (grasses and shrubs, mosses, and lichens) understory vegetation carbon stock was significantly higher in control than treated plots (Table 4). On the contrary, litter C stock was higher in treated plots at all sites with the exception of Craigellachie (Table 4). However, vegetation plus litter C stock was higher in control plots at all sites, with the exception of Speymouth (Figure 5A). Soil C stocks did not exhibit any general trend, with no significant differences observed among treatments (Table 4; Figure 5B).
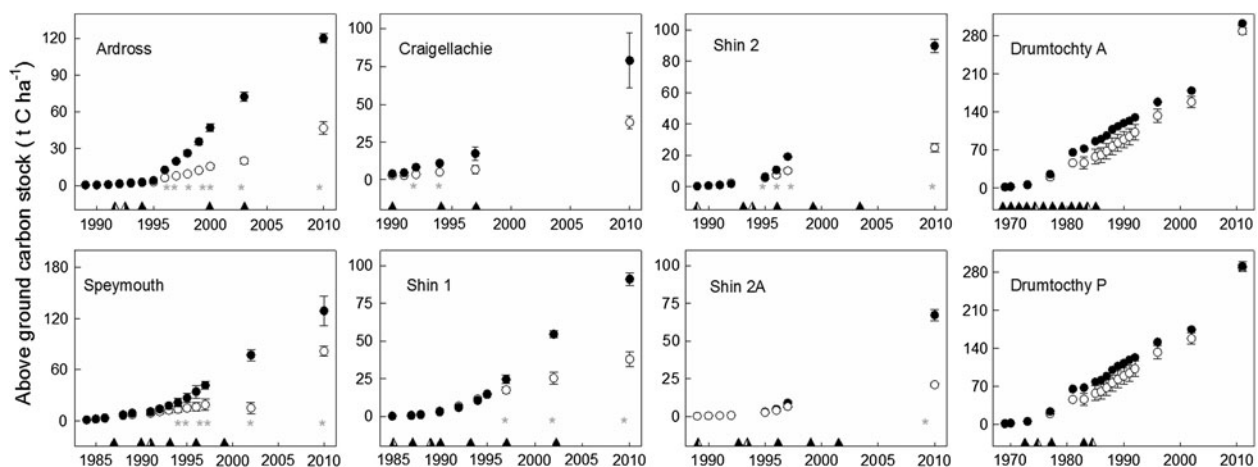

Figure 2. Variations over time in tree aboveground C stocks at study sites. *Each panel* shows comparisons between treated (*black circles*) and control plots (*white circles*). *Each symbol* represents the average of 3–4 replicates per site (see Table 2 for experimental set up); *asterisks* indicate significant differences ($P < 0.05$) between treatments, as determined by ANOVA, *vertical bars* represent ±one standard error. *Triangles* on X-axis show the years of N ▲, P ◮, and K ◿ fertilizations: whilst N was added only to treated plots, P and K were applied in both treated and control plots.

Pooling all the compartments at 2010, total ecosystem C content was estimated for each site and at most sites higher C stocks were estimated in treated plots (Figure 5C).

Temporal Responses of Aboveground C Stock to N Addition and N Deposition

The temporal effects of N supply on aboveground C stocks (C_{ag}) were evaluated across study sites. For each site, the accumulated differences in C_{ag} between treated and control plots (ΔC_{cum}) at a given year were plotted against the corresponding accumulated amount of N supplied (ΔN_{cum}) for the same year (Figure 6). An approximately linear relationship between ΔC_{cum} and ΔN_{cum} was observed for many sites, with the exception of Craigellachie and Drumtochty, where a less aligned response was revealed. All fertilization experiments were terminated between 1984 and 2003 (Table 2). Therefore, subsequent surveys (including the one in 2010) could be used to gage the effect of stopping
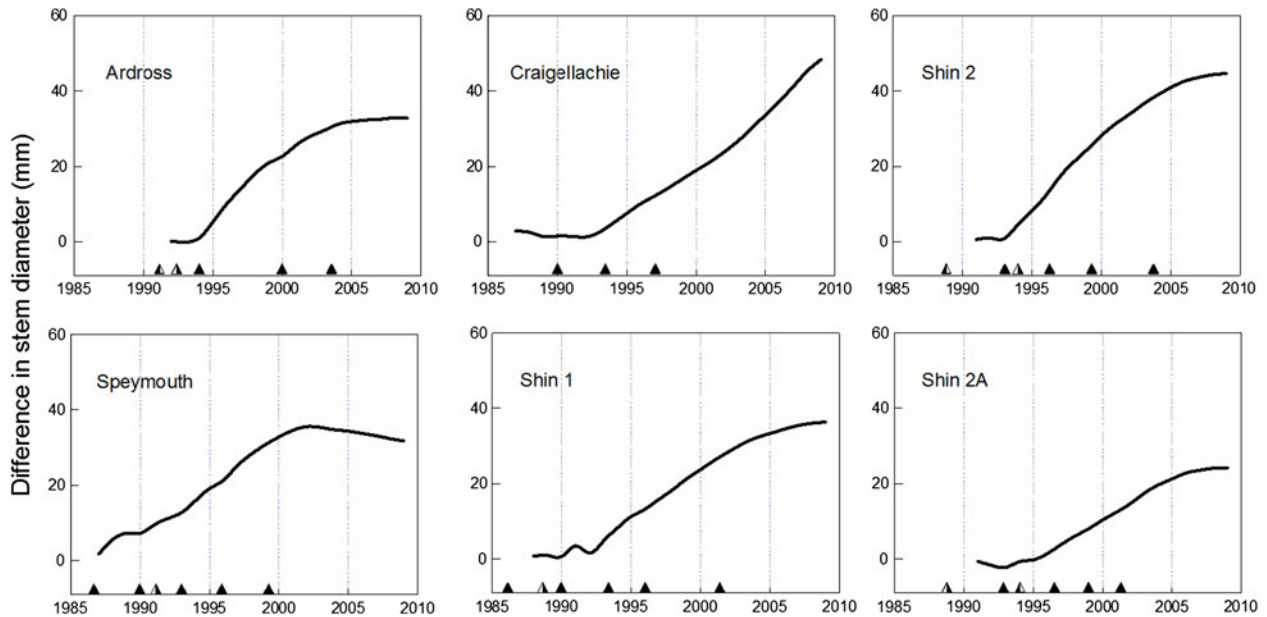

Figure 3. Mean accumulated stem diameter differences at breast height between the five largest trees of the treated and control plots at the study sites. *Triangles* on X-axis show the years of N ▲, P /\, and K /\ fertilizations: whilst N was added only to treated plots, P and K were applied in both treated and control plots.

Table 3. Significance Levels for Treatment Effects (T); Year Effects (Y), and the Interaction (T × Y) on Foliar Nutrient Concentration of Nitrogen (N), Phosphorus (P), Potassium (K), N/P, and N/K Ratios from a One-Way Repeated-Measures ANOVA

Site	N			P			K			N/P			N/K		
	T	Y	T × Y	T	Y	T × Y	T	Y	T × Y	T	Years	T × Y	T	Y	T × Y
Ardross	***	***	***	*	***	***	ns	***	***	**	***	**	***	***	***
Craigellachie	**	***	***	ns	***	ns	ns	***	*	*	**	ns	*	***	***
Drumtochty P	**	***	*	ns	*	ns	ns	***	ns	*	***	**	**	***	**
Drumtochty A	***	***	***	ns	ns	ns	*	***	ns	***	***	ns	***	***	***
Shin 1	*	***	**	ns	***	***	ns	***	**	ns	***	ns	**	***	***
Shin 2	*	***	***	ns	***	ns	ns	***	ns	**	***	*	*	***	*
Shin 2 Alaska	**	***	***	**	***	***	*	***	***	***	***	***	*	***	***
Speymouth	***	***	***	*	***	***	**	**	**	***	***	***	ns	ns	ns

*Significance at * $P < 0.05$, ** $P < 0.01$, and *** $P < 0.001$, respectively. At Shin 2, a Sitka spruce provenance from Alaska was also tested.*
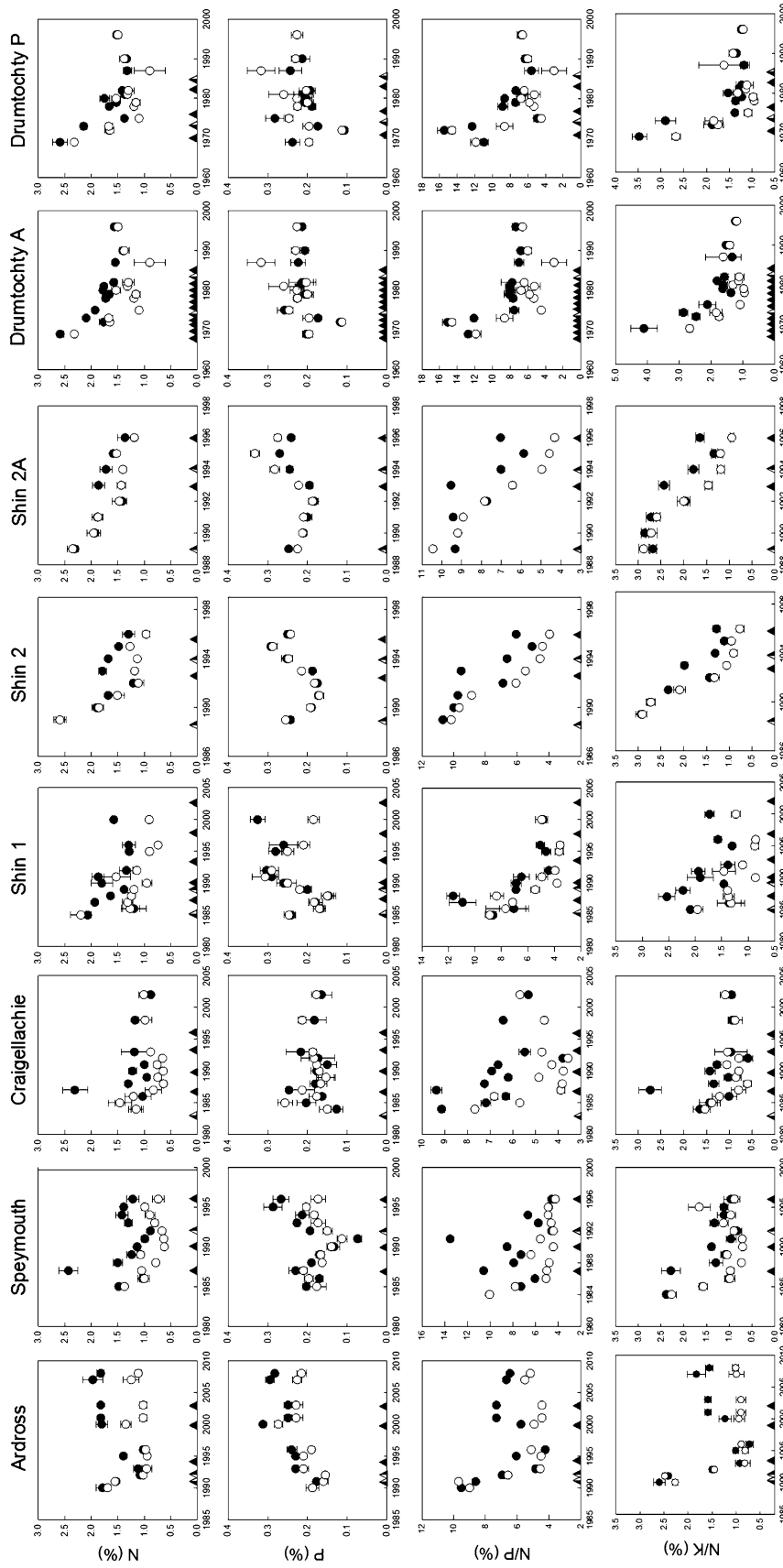*ns = not significant.*

Figure 4. *Y*-axis shows the temporal variation in foliar nitrogen (N) and phosphorus (P) percentage nutrient concentration and N/P and N/K molar ratios at each study site over the experimental periods; *X*-axis shows the temporal line. *Each symbol* indicates the average value in treated (*empty circles*) and control plots (*filled circles*) of 15/20 replicates per treatment at each site. *Vertical bars* represent ±one standard error. *Triangles* on *X*-axis show the years of N ▲, P ▲, and K △ fertilizations: whilst N was added only to treated plots, P and K were applied in both treated and control plots.

treatment on aboveground C accumulation rates, from 7 to 26 years after the end of the treatments. To achieve this, we compared $\Delta C_{\text{cum}(i)}$ versus $\Delta C_{\text{cum}(i+5)}$ (where i is the last N addition year, Figure 7). All experiments showed continued differential accumulation of C in the treated plots 5 years after stopping the treatments, with the slope of the linear relationship not being significantly different from 1, and the intercept of the relationship being significantly greater than 0 (Figure 7).

Finally, the site-specific accumulated atmospheric nitrogen deposition (N_{dep}) was included in

the analysis of the effects of experimental nitrogen addition (N_{exp}) on C_{ag}. First, the relationship between total N entering the system ($N_{\text{ent}} = N_{\text{exp}} + N_{\text{dep}}$) and ΔC_{cum} was reconstructed for the entire experimental period. ΔC_{cum} increased steadily with N_{ent}, with the exception of Drumtochty and Speymouth (data not shown). Successively, the values of ΔC_{cum} were plotted with respect to the controls (set to zero) against the total N_{ent}, that is, $N_{\text{exp}} + N_{\text{dep}}$ for treated plots and N_{dep} for control plots (Figure 8). In this graph, each experiment is represented by two points, the one at $\Delta C_{\text{cum}} = 0$

Table 4. Carbon Stocks in 2010 (Mean ± Standard Error, Number of Observations in Brackets) in Soil, Litter, and Understory Vegetation in the Lines Between Trees of Control and Treated Plots of Six N Addition Experiments

Site	Carbon stock (Mg ha^{-1})									n
	Soil			Litter			Understory vegetation			
	Control	Treated	P	Control	Treated	P	Control	Treated	P	
Ardross	193 ± 16	187 ± 9	0.826	0.8 ± 0.1	1.7 ± 0.2	0.002	1.1 ± 0.1	0.0 ± 0.0	0.000	(24)
Craigellachie	268 ± 40	305 ± 41	0.218	1.0 ± 0.2	0.9 ± 0.1	0.667	2.6 ± 0.3	0.9 ± 0.1	0.005	(30)
Shin 1	357 ± 10	352 ± 11	0.856	0.8 ± 0.1	1.6 ± 0.2	0.005	1.6 ± 0.8	0.8 ± 0.2	0.015	(12)
Shin 2	509 ± 4	547 ± 6	0.213	0.8 ± 0.1	2.0 ± 0.1	0.000	3.1 ± 0.4	0.2 ± 0.1	0.009	(12)
Shin 2 Alaska	308 ± 4	259 ± 7	0.113	1.4 ± 0.1	2.9 ± 0.5	0.029	5.0 ± 0.3	2.6 ± 0.7	0.017	(12)
Speymouth	132 ± 15	173 ± 26	0.173	0.8 ± 0.0	1.4 ± 0.0	0.000	0.9 ± 0.1	0.4 ± 0.1	0.009	(18)

P values are from two-way ANOVA (treated vs control plots). At Shin 2, a Sitka spruce provenance from Alaska was also tested.
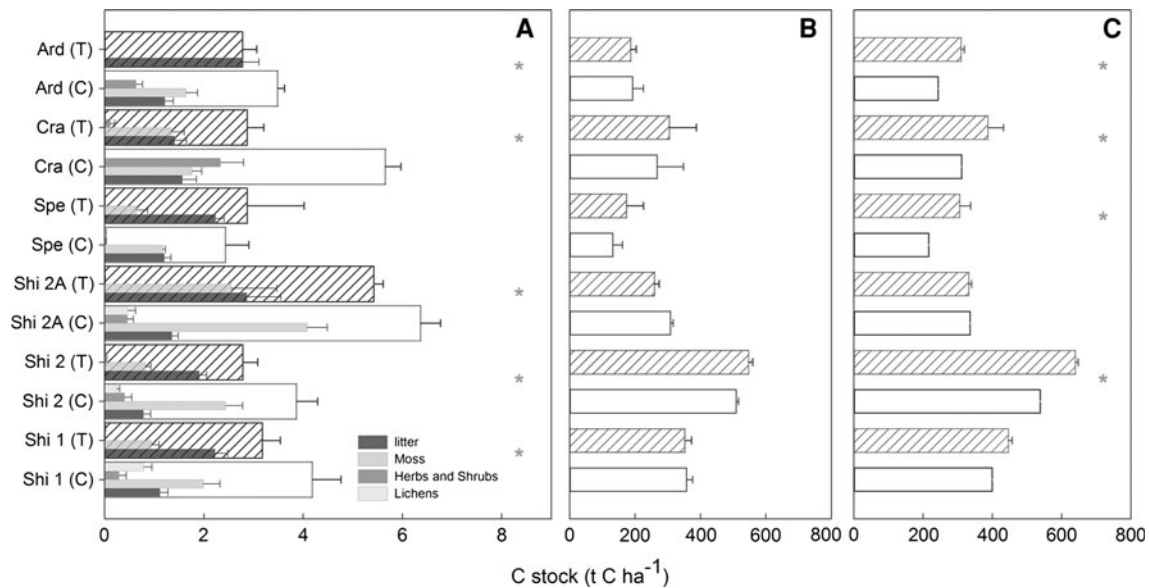

Figure 5. Carbon stock in control (C) and treated (T) plots of Sitka spruce plantations at different experimental sites. Data are referred to the summer of 2010. *Panels* show the carbon stock separately estimated for: **A** litter, understory vegetation, and sum (the widest bars); **B** soil; **C** total ecosystem (including soil, litter and vegetation, and tree biomass). *Lines* represent ±standard error. *Asterisks* are referred to the common symbols in panels and indicate significant differences ($P < 0.05$) between treated and control plots at each site.
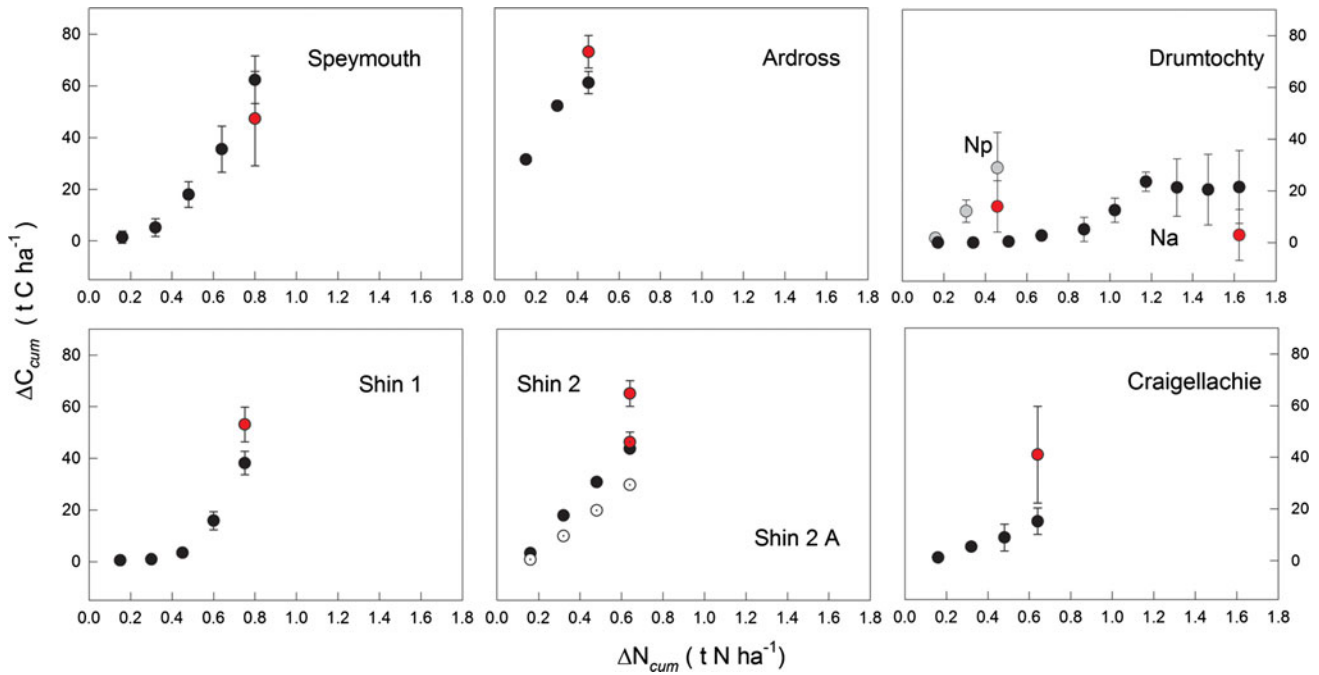
Figure 6. Accumulated aboveground carbon stock differences between treated and control plots (ΔC_{cum}) of Sitka spruce up to a given year, and the corresponding accumulated nitrogen supplied experimentally (ΔN_{cum}). The *red points* correspond to 2010 measurements; *vertical bars* represent ±one standard error. At Drumtochty, two separate experiments were carried out, one with periodic (Np) and one with biannual (Na) N additions. Shin 2A is a second experiment at Shin 2, where a provenance from Alaska was tested (Color figure online).
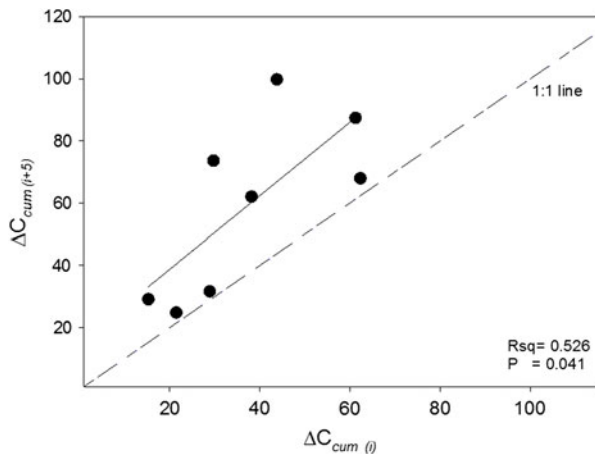
Figure 7. Linear relationship between $\Delta C_{cum(i)}$ (where i is the year of the last N addition), and $\Delta C_{cum(i+5)}$ (that is, 5 years after the end of the fertilization experiment) superimposed on a 1:1 line.

Figure 8. Accumulated aboveground carbon stock differences between treated and control plots (ΔC_{cum}) at the end of the experiments (2010) (normalized based on control plot values set to zero) versus the total N entering the system (N_{ent}).

represents the control, and is shifted to the right on the X-axis by a value equal to the cumulative N_{dep} for that site through 2010. The second point of each pair represents the treated plot, and is further shifted to the right of the control by an amount equivalent to the cumulative N_{exp} added to the
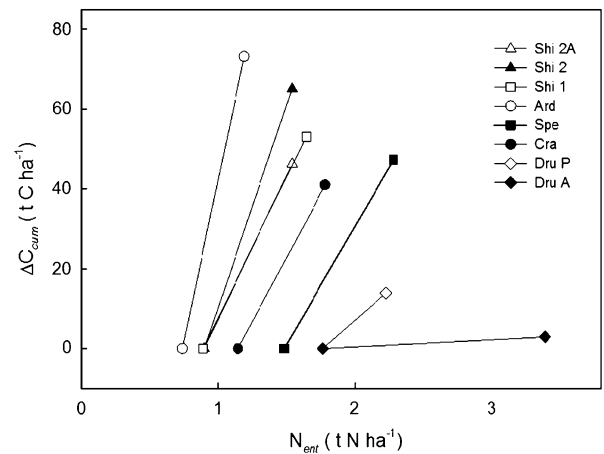
system during the experiment. This way of plotting the data allows us to easily rank all the experiments according to the prevailing levels of accumulated N_{dep} at each site. In particular, the slope of the relationship between N_{ent} and ΔC_{cum} (Figure 8) reflects the response to the experimental N addition,
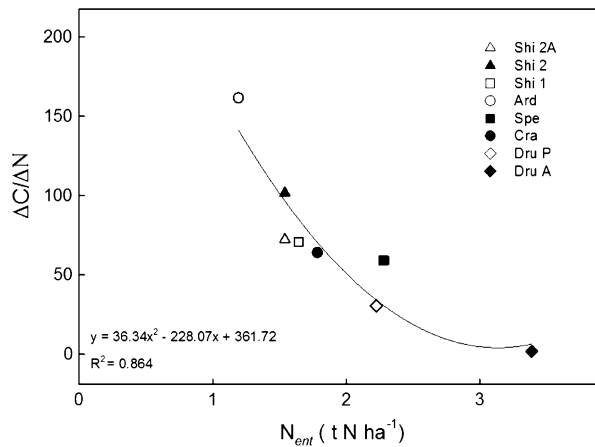
Figure 9. Relationship between site-specific $\Delta C/\Delta N$ (t C ha^{-1} y^{-1}/t N ha^{-1} y^{-1}, calculated as mean annual value from the beginning of the experiment to 2010) and total nitrogen entering the system (N_{ent}, t N ha^{-1} y^{-1}); N_{ent} represents the sum of N deposition and N supplied to treated plots at each site, ΔC represents the aboveground carbon stock difference between treated and control plots, and ΔN represents the difference between the amounts of N supplied to treated and control plots at each site.

whereas the x-intercepts reflect the prevailing level of site-specific N_{cum}. In our experiments, the magnitude of $\Delta C/\Delta N$ (that is, the Y-axis values of Figure 9 that represent the slope of the lines reported in Figure 8) declines with increased N_{ent} (which represents the amount of accumulated N deposition and addition of fertilizer N) approaching zero at Drumtochty.

Discussion

Aboveground Carbon Stocks and Foliar Nutrient Concentrations

A positive N addition effect on tree carbon stocks was generally recorded in our experiments, in accordance with previous evidence from several studies on the effects of N addition in forest stands (for example, Högberg and others 2006; Hyvonen and others 2008). In other studies, no effects (Christ and others 1995; Magill and others 1996; Emmett and others 1995a), or even negative effects (McNulty and others 1996) on tree growth were observed. In some cases (that is, Brooks and Coulombe 2009), the positive effect of N addition on tree growth was complicated by changes in stand density, due to thinnings or natural mortality that resulted in an increase in the available area for plant growth. It is worth noting that at our sites,

with the exception of Craigellachie, where some limited mortality was observed, no change in stand density occurred over the experimental period.

During our experiments, a decline in tree growth was not observed either during fertilization or following the last N addition. As further evidence, Figure 7 suggests that biomass continued to accumulate at a faster rate in treated plots in the 5 years following the end of the experiment, in accordance with the results reported by Petterson (1994).

Foliar nutrient analyses have been widely used in forest research as indicators of soil and plant nutrient status, and as powerful tools to identify if fertilization is required. Results (Figure 4) indicated that sites where N addition led to a significant increase in N foliar concentration also showed an increased biomass accumulation (for example, Speymouth and Ardross), in agreement with Ingerslev and others (2001). Furthermore, at these same sites, foliar P concentration was higher in treated plots even though control plots received the same P level. These observations suggest a positive N addition effect on P metabolism by plants. However, this was not a general trend; for example, at some sites (for example, Drumtochty) a decrease in P foliar concentration in treated plots was observed. Correspondingly, this site did not show a positive effect of N addition on tree growth. Braun and others (2010) reported similar results from a 15-year-long N addition experiment on *Picea abies* and *Fagus sylvatica*, with a reduction in P foliar concentration, and a parallel decrease in stem increments. In other cases (Emmett and others 1995b; Wright and Tietema 1995; Beier and others 1998), no change in foliar nutrient concentrations following fertilization were detected.

It is worth remembering that at all our sites P fertilization was also carried out during the first few years after planting. Our experimental design allowed us to isolate the N effect per se, because our control plots also received the same P (and in some cases, K) applications. This however needs to be kept in mind in the interpretation of our results, because they were obtained under conditions of likely reduced P and K limitation.

Understory Vegetation, Litter, and Soil

We recorded a clear reduction in understory vegetation biomass, consistent with Makipa (1995) and Malkonen (1990) as a result of N addition. As understory vegetation decreased, litter C stock increased in treated plots relative to control ones. Generally, N fertilization could lead per se to a decline in ground flora species composition, and

subsequent biodiversity loss (Phoenix and others 2012; De Schrijver and others 2008). Indeed, Thomas and others (1999) analyzing understory plant diversity response to nitrogen fertilization and thinning, found an approximately 61% decrease in species diversity following urea fertilization. However, our observations are more likely due to the increase in tree canopy cover in treated plots which may have shaded out the understory (Siefert 2005) and contributed to the increase in litter production.

In contrast with evidence (Blevins and others 1983; McAndrew and Malhi 1992) reporting an increased soil organic C following N addition, we did not observe differences in soil characteristics (that is, soil bulk density, soil C stock) attributable to N application (Table 4; Figure 5). This is most likely due to the inherently high spatial variability and high C content of our organic soils, which makes it very difficult to detect any experimental difference. The recorded increase in litter C stocks in the treatment plots could be due both to increased tree growth and litter production (Figure 2) and to a reduction in litter decomposition caused by increased N availability (for example, Janssens and others 2010).

Temporal Responses of Aboveground C Stocks to N Addition and N Deposition

Pooling all the ecosystem C stocks, a variable but positive N fertilization effect was evident across all sites (Figure 5), largely due to the increase of aboveground biomass. Previous studies (Melin and others 1983; Pregitzer and others 2008; Hyvonen and others 2008) reported positive responses on both above and belowground C stocks to N addition. Photosynthesis and tree growth increase with soil N availability, and therefore plant uptake could represent the main sink for added N (Aber and others 1989). However, excess N is recycled through litter and humus, and this could lead to an increase in nitrate losses via leaching (Malkonen 1990; Petterson 1994; Magill and others 2004).

We did not find a clear relationship between increased ΔC_{cum} and number of fertilization events. Nonetheless, all treatments exhibited a positive response in terms of biomass accumulation (Figure 6). Consistent with Högberg and others (2006), a higher response was elicited for low N application rates, that is, Ardross (0.42 t N ha^{-1}), relative to intermediate, that is, Craigellachie (0.62 t N ha^{-1}), or high, that is, Drumtochty (1.62 t N ha^{-1}). Regular biannual fertilizer applications, that is, Drumtochty A, did not seem to increase stem growth compared

with stands periodically fertilized (that is, Drumtochty P).

A clearer interpretation of tree biomass C responses to N fertilization is possible if accumulated atmospheric nitrogen deposition (N_{dep}) is assessed together with experimental N addition. Our results (Figure 8) show a clear decline in C response to the experimental N addition when the amount of N entering the system increases as a result of atmospheric deposition.

This is a very relevant point in the context of the debate on the magnitude of C response to N deposition (Nadelhoffer and others 2004; Högberg and others 2006; De Vries and others 2006, 2009; Magnani and others 2007; Sutton and others 2008).

High annual N_{dep} and accumulated N_{dep} levels at the Drumtochty site (Figure 1) might be responsible for the lack of a significant difference in C_{ag} stock between treated and control plots at this site (Figure 5), which also shows the highest carbon stock accumulation in control plots (Figure 2) and the lowest $\Delta C_{cum}/N_{ent}$ (Figure 8) and $\Delta C/\Delta N$ (Figure 9) values. It is worth noticing that inherent site differences cannot explain this result, based on the environmental variables and the yield index calculated with ESC (Table 1). We also considered that a possible age-related factor could have overshadowed the fertilization effects at Drumtochty site, but the long-term observation (Figure 2) did not show any significant differences between C_{ag} in fertilized and control plots during the whole experiment. Accumulated N_{dep} is the most likely explanation for the differential tree growth response, in particular we argue that the high levels of N_{dep} (Figure 1) may have acted by promoting tree growth in the short term and by enhancing the inherent site-specific fertility in the long term.

Our results suggest that trees growing on N-rich sites can be less efficient at using added N. The absence of significant differences in tree growth, as well as in several other plant physiological parameters, observed in other N addition experiments (for example, at Aber in Wales, see Emmett and others 1995a, b, 1998), may therefore be explained by the fact that they were performed at sites where historic N_{dep} levels are much higher than in Scotland (Figure 1). Values of $\Delta C/\Delta N$ (the instantaneous response of the ecosystem C cycle to an unit addition of N to the system), reported in Figure 9 seem to be on the higher end of prior reports (see De Vries and others 2009, for review) for N_{ent} less than 2 ton ha^{-1}. However, note these values come from experiments performed on young stands that experienced also P and/or K fertilization, possibly

leading to positive nutrient interaction effects on aboveground C accumulation.

The novel information from Figure 9 is long-term N addition experiments might be better interpreted by considering the total N entering the system, including site-specific accumulated atmospheric N_{dep}. Therefore, we suggest the use of accumulated N_{dep} as an important variable to ascertain tree growth responses to N fertilization. Much of the research carried out on the effects of N deposition took place around the highly urbanized areas of Central Europe, where the research centers were located. Our results suggest that the lack of tree responses to N manipulation observed in those experiments was at least partly caused by the high levels of N availability of those soils caused by high N_{dep}. Carbon sequestration in forests of more remote areas may still be increasing as a result of lower but biologically significant rates of chronic N deposition.

CONCLUSIONS

Positive N addition effects on tree carbon stock were generally recorded, and a decline in tree growth was not observed during N fertilization or several years after the final N addition. At all sites, carbon stocks in the understory vegetation (grasses and shrubs, mosses, and lichens) were significantly higher in control plots than in treated (N-enriched) plots. However, the amount of carbon stock in the litter was significantly higher in treated plots. Pooling all components (that is, understory vegetation, litter, soil, and tree biomass) for the common year 2010, the total ecosystem C content could be estimated for each site, and in most of the experiments considered a higher C stock was estimated for the N-treated plots. Understanding changes in tree C over the experimental period is improved if accumulated atmospheric nitrogen deposition (N_{dep}) is assessed together with experimental N addition, and we suggest site-specific accumulated N_{dep} levels as an integral variable to consider to understand tree growth responses to N fertilization. Overall, our stands were in their juvenile expansion phase during the period of the experiments described here. It remains to be seen whether the responses to N addition found here would change in older stands.

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