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Canopy uptake of atmospheric N deposition at a conifer forest: part I -canopy N budget, photosynthetic efficiency and net ecosystem exchange

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ABSTRACT

Global carbon cycle assessments of anthropogenic nitrogen (N) deposition influences on carbon sequestration often assume enhanced sequestration results. This assumption was evaluated at a Rocky Mountains spruce-fir forest. Forest canopy N uptake (CNU) of atmospheric N deposition was estimated by combining event wet and throughfall N fluxes with gradient measured HNO_3 and NH_3 as well as inferred (NO_x and particulate N) dry fluxes. Approximately 80% of the growing-season 3 kgN ha^{-1} total deposition is retained in canopy foliage and branches. This CNU constitutes ~1/3 of canopy growing season new N supply at this conifer forest site.

Daytime net ecosystem exchange (NEE) significantly ($P = 0.006$) and negatively (CO_2 uptake) correlated with CNU. Multiple regression indicates ~20% of daytime NEE may be attributed to CNU ($P < 0.02$); more than soil water content. A wet deposition N-amendment study (Tomaszewski and Sievering—part II), at canopy spruce branches, increased their growing-season CNU by 40–50% above ambient. Fluorometry and gas exchange results show N-amended spruce branches had greater photosynthetic efficiency and higher carboxylation rates than control and untreated branches. N-amended branches had 25% less photoinhibition, with a 5–9% greater proportion of foliar-N-in-Rubisco. The combined results provide, partly, a mechanistic explanation for the NEE dependence on CNU.

1. Introduction

Questions related to the dynamics of terrestrial carbon (C) sinks are at the core of scientific and policy debates nationally and internationally. Our ability to address the policy imperatives derived from multilateral agreements (e.g. Kyoto Protocols) will depend on improved knowledge of C-cycle variability (and future states). Quantitative understanding of the mechanisms driving C sequestration is now an imperative. For example, a large North American C sink (Pacala et al., 2001) of 0.4–0.7 PgC yr^{-1} in the conterminous US has been attributed to several factors, with eastern US forest re-growth and enhanced growth due to atmospheric N deposition being among the factors mentioned. Miller et al. (2005) have considered climatological influences on the entire terrestrial temperate northern hemisphere (18°–53 °N) C sink of 2–4 PgC yr^{-1}. They found no statistical correlation of this sink's 1992–2004 variations with surface air temperature, precipitation rate, Palmer Drought Stress Index and Multivariate ENSO Index despite there being strong correlations of the terrestrial tropics C sink (17 °S—17 °N) with these climatological variables. This leaves the large terrestrial temperate northern hemispheric C sink open to a range of explanations, including atmospheric N deposition. Estimates of the global impact of atmospheric N deposition on forest C sequestration at N-limited forests vary from as little as 0.1 PgC yr^{-1} to more than 2 PgC yr^{-1} (Schindler and Bayley, 1993; Townsend et al., 1996; Holland, 1997). These model-based estimates generally (though not always) assume C sequestration to be directly proportional to the magnitude of atmospheric N deposition. This is certainly an incorrect assumption at the minority of forests worldwide that are N-saturated.

Atmospheric N is present in both reduced and oxidized states and may be deposited directly to foliage or to underlying soils. Wet and dry deposition processes are both variable in space and time. This variability complicates our ability to quantify the impact of atmospheric N deposition on C sequestration at forests. The atmospheric N deposition contribution to total N cycling at forest ecosystems is often small compared to the total N pool size. However, its contribution to N cycling within the forest canopy, especially foliage where photosynthesis is active, may

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be substantial. For example, Schulze (2000) has shown, at European spruce stands, that 40–65% of atmospheric N deposition was retained in the canopy and that it may account for 15–40% of the annual N requirement for growth.

Earlier, Cole and Rapp (1981) showed that annual foliar N requirement for growth statistically explained the vast majority of variability in both foliar new growth and aboveground new growth at International Biosphere Program (IBP) conifer forests ($r^2 = 0.98$ and 0.95, respectively, with $P < 0.0001$). Foliar N requirement for growth—the flux of N into current year foliage—appears to have a significant influence on both foliar and aboveground growth found at conifer forests. However, foliar N concentration was a very poor explanatory variable ($r^2 = 0.14$, P of 0.34) for aboveground new growth at these same forests. In contrast, several deciduous forest data sets indicate, at the foliar scale, that light-saturated net photosynthetic capacity ($A_{\max}$) and mass-based foliar N concentration are well correlated (e.g. Reich et al., 1995). This suggests that small N fluxes into deciduous forest foliage may be contributing in a minor way (relative to foliar N concentration) to their annual growth. There are also data sets indicating that atmospheric N deposition is detrimental to forest growth and that, occasionally, reductions in photosynthesis may accompany foliar N uptake (Schulze, 2000). However, the latter outcome is likely due to plant damage caused by increased ozone that is often associated with elevated N deposition from anthropogenic sources (Harrison et al., 2000). Further statistical analysis of existing data sets, as well as new data obtained from studies such as that described here, are needed to clarify the contribution atmospheric N deposition may make to forest C sequestration.

Soil-derived uptake and reallocation are the two N flux pathways generally considered responsible for most (in some cases all) of annual foliar N requirement for growth, with N fixation usually a distant third (e.g. Cole and Rapp, 1981). Yet, atmospheric deposition delivers N directly to photosynthetically active foliage, especially upper canopy and current year foliage. Consideration of this additional pathway for N flux into foliage is necessary to fully characterize the sources of foliar N requirement for growth (e.g. Ammann et al., 1995). Observations of spruce foliage growth differences (Rueth and Baron, 2002), along with atmospheric N deposition and canopy retention data (Tomaszewski et al., 2003), motivate such consideration. Rueth and Baron (2002), at sites on east and west sides of the Continental Divide, found that mean dry matter needle weight was 17% greater at spruce trees on east-side than that on west-side spruce trees (sites had similar temperature and precipitation regimes) and, also, that east-side spruce trees received about three times more atmospheric N deposition than west-side spruce trees. Further, Tomaszewski et al. (2003) showed that canopy retention at east-side spruce-fir forests exceeds 75% for both nitrate and ammonium. Additional motivation for inclusion of the atmospheric N deposition pathway in the assessment of foliar N requirement for growth comes from estimates that 10–40% of annual C sequestration occurring at a number of eastern forests may result from atmospheric N deposition (Sievering et al., 2000).

In this study, we discuss our measurements of canopy uptake of atmospheric N deposition—canopy N uptake or, simply, CNU—in the context of a spruce-fir forest canopy N budget. This budget includes the consideration of N uptake by soil-derived uptake, N fixation, and reallocation of N as well as CNU at a Rocky Mountains forest site, the Niwot Long Term Ecological Research forest (Niwot Forest). Canopy new growth mass, C, and N were also measured. Net ecosystem CO_2 exchange (NEE) and other flux data are available at this Niwot Forest AmeriFlux site. Thus, relationships among annual foliar N requirement, canopy fluxes of N, and NEE could be explored. The main objectives of this study are to:

(1) Describe the contribution that CNU makes to annual growing season N requirement for growth and to the canopy N budget at the Niwot Forest;

(2) Evaluate the observed statistical dependence of daytime NEE on CNU in the context of this canopy N budget and fluorometry/gas exchange data and

(3) Consider photosynthetic efficiency and photoinhibition (by fluorometry) and gas exchange results from a branch-applied N amendment field experiment to help identify a mechanism by which CNU may contribute to photosynthesis.

2. Materials and methods

The conifer study site is located at the Niwot Ridge AmeriFlux site in a subalpine forest ecosystem that is part of the University of Colorado's Mountain Research Station (40° 2′ N; 105°33′ W). It is ~6 km east of the Continental Divide and ~60 km west of Front Range urban centres. The research area was formerly deforested by logging but is now a ~90 year old, slowly aggrading forest, at an elevation of 3,000 m. The forest is composed of *Picea engelmannii Parry* (Engelmann spruce), *Abies lasiocarpa (Hook.) Nutt.* (subalpine fir) and *Pinus contorta Dougl.* (lodgepole pine). The months of May to October have been shown to consistently favour net CO_2 uptake by the forest (Monson et al., 2002). Thus, May–October is defined as the growing season. The canopy height is ~12 m with midsummer leaf area index of 4–4.5 $m^2\ m^{-2}$ and a canopy gap fraction of 17%; tree density is 16, 10 and 9 per 100 m^2 for spruce, fir and pine, respectively (Monson et al., 2002). Annual precipitation averages 80 cm with 35 cm occurring, almost entirely as rain, in the May–October period. The mean annual temperature is 4 °C. AmeriFlux program measurements are undertaken from a 26-m walk-up tower with CO_2, H_2O and other eddy covariance data obtained from a height of 21.5 m. Displacement height is 7.6 m and the roughness length is 1.8 m.

During May to September, episodic upslope winds bring air from Front Range metropolitan areas to the site, especially in the midday and afternoon, about once per every 3 d. This easterly flow brings high concentrations of several N compounds to the site (Fahey et al., 1986; Parrish et al., 1986). Substantially more than half of the annual atmospheric N deposition is of anthropogenic origin; modelling analyses suggest anthropogenic N sources, especially those in the Colorado Front Range may have increased N deposition by an order of magnitude (Baron et al., 2000). Together, wet and dry N deposition contribute 4–8 kgN $ha^{-1}yr^{-1}$ to this forest's annual N cycle (Sievering, 2001). Fog and cloud deposition is very rare (nonexistent during many growing seasons).

2.1. *Nitrogen measurements*

In order to describe the contribution that CNU may make to the Niwot Forest canopy N budget one must first identify whether atmospherically deposited N is taken up by canopy organisms other than forest foliar and branch elements themselves. Lichen biomass at this conifer forest canopy is no greater than 22 kg dry matter per hectare. This magnitude is minute in comparison to an eastern forest where lichen abundance was estimated to have no influence on throughfall (TF) chemistry (Friedland et al., 1991). At our site, Tomaszewski et al. (2003) had shown lichen N uptake is <0.02 kg N $ha^{-1}yr^{-1}$. This magnitude is insignificant when compared to overall N uptake by the canopy (discussed below). Canopy microorganism pool size increases due to atmospheric N deposition may occur under conditions of high (>30 kgN $ha^{-1}yr^{-1}$) N deposition (Rennenberg and Gessler, 1999). However, total N deposition at our site is only 4–8 kgN $ha^{-1}yr^{-1}$; these inputs are unlikely to cause a measurable canopy microorganism increase (H. Rennenberg, personal communication, November 2001). Canopy N uptake was, thus, not influenced by microorganism uptake.

Gaseous N emission from this forest canopy was considered. Torizzo and Sievering (2002) show, across the growing season, that net NH_3 flux at this site is negligible; similarly, J. Sparks (personal communication, 2003) showed that N_2O flux from this forest's canopy was very nearly zero. Since the forest canopy is quite dry, the likelihood that HNO_3 may be converted at dew or otherwise wetted parts of the canopy (with N being released) is very low. Gaseous N emissions from the predominantly cold and dry soils at this site are extremely small (J. Sparks, personal communication, 2003); thus, it is not included in the canopy N budget of Section 3.2.

Given the above, canopy uptake of atmospheric N deposition (CNU) by canopy foliar and branch components can be determined by subtracting throughfall (again, TF) loss of both organic and inorganic N from the sum of wet (WD) and dry (DD) N deposition (i.e. CNU = WD + DD – TF). This formulation may be applied at coniferous forests such as ours where stem flow and fog/cloud interception of N are insignificant (Sievering et al., 2001) and, also, where gaseous N emission is low. Further, it was found that the formulation CNU = WD + DD – TF may be used to evaluate relations between NEE and CNU as long as precipitation event-based WD, DD, and TF data are considered; for example, aggregated TF data (weekly, or longer) will not suffice due to large event-to-event variability. Event precipitation (WD) was collected using two (duplicate) precipitation collectors (each of 0.068 m^2 collection area). For TF collection, 22 troughs (total of 1.55 m^2 sampling area) were used as described in Tomaszewski et al. (2003). Wet deposition and TF samples were collected 2–4 hr after each precipitation event.

Litterfall samples were collected across a three year period (L. Scott-Denton, personal communication, February, 2003) using 21 litter baskets in the vicinity of the AmeriFlux tower. These data, along with intact foliar data, provide a means by which to estimate annual reallocation of N within the canopy (Fahey and Birk, 1991). The total annual N required for canopy growth (including reallocation) was estimated for the growing season using the approach described in Tomaszewski et al. (2003). A canopy N budget analysis—identifying the magnitude of respective N fluxes that contribute to the canopy N budget—can, then, be undertaken. An outcome of this analysis, by difference, is the estimation of the soil-derived N that contributes to the annual N required for canopy growth. This soil-derived N is due to the sum of N fixation and soil/root N uptake.

Dry deposition of gaseous $HNO_3{}^-$ and NH_3 were determined using the flux-gradient approach for the years 2000 and 2001. This resulted in parametrizations of HNO_3 and NH_3 dry deposition velocities (Sievering et al., 2001; Torizzo and Sievering, 2002) that were then used, along with concentrations, to estimate their dry fluxes in other years. In 2000 and 2001, HNO_3 dry deposition was found to be ≥80% of total NO_y dry deposition and that NH_3 dry deposition is of no consequence at this site (Torizzo and Sievering, 2002). Dry deposition fluxes of NO_x as well as particulate $NO_3{}^-$ and $NH_4{}^+$ were estimated using concentration data (Fahey et al., 1986; Parrish et al., 1986; Sievering, 2001; Torizzo and Sievering, 2002) along with dry deposition velocities from the literature (Wesely et al., 1982; Ruijgrok et al., 1997; Torizzo and Sievering, 2002). Although uncertainties in the resulting dry deposition fluxes of NO_x, $NO_3{}^-$ and $NH_4{}^+$ are large, they do not contribute significantly to overall dry deposition uncertainty since their mean dry flux values are quite small relative to that for HNO_3. Thus, in years beyond 2001, HNO_3 concentrations together with parameterized HNO_3 deposition velocities along with the above NO_x, $NO_3{}^-$ and $NH_4{}^+$ flux estimates are adequate to describe total dry N deposition at this site (Sievering et al., 2001). Finally, since dry N deposition is dominated by HNO_3 with zero canopy resistance to transfer (Sievering et al., 2001a), the magnitude of dry N deposition to the forest floor is insignificant. Thus, DD in the formulation for CNU above may be assumed to occur essentially only at the forest canopy.

2.2. *Laboratory analysis*

Wet deposition and TF samples were filtered with a hand pump using Whatman 0.45 μm filter paper. A 10–60 ml sample for inorganic analysis was refrigerated at ~5° C. An additional aliquot of 10–60 ml was frozen for analysis of total dissolved N using an Antek 9000 N-Analyzer (Antek Inst. Inc., Houston, TX, USA). Inorganic N in samples were determined on a Lachat Quick Chem 8000 Spectrophotometric Flow Injection Analyzer (Zellweger Analytics, Milwaukee, WI, USA; detection, NH_4^+ 0.36 μEQ/L RSD 1.00%, (NO_3^- + NO_2^-) 0.05 μEQ/L RSD 0.39%).

2.3. *Fluorometry data*

The monitoring of photosynthetic capacity using chlorophyll fluorescence techniques (fluorometry) has become fairly well standardized in the recent past (eg., Maxwell and Johnson, 2000). Generally, it is required to obtain a rapid and accurate predawn (dark-adapted) measurement of F_m, the maximum level of fluorescence, and F_o, the minimum level of fluorescence by foliar chlorophyll. Thereafter, under high-light conditions (>1000 μmol $m^{-2}s^{-1}$ at Niwot LTER forest), the maximum and minimum fluorescence levels, $F_m{}'$ and $F_o{}'$ (primes indicate in-the-light measurements) are again obtained. These parameters may, then, be used to assess photosythetic performance. Tomaszewski and Sievering (2007—part II) detail the use of fluorometry, in conjunction with CO_2 gas exchange measurements, to evaluate the contribution CNU may make to photosynthetic capacity.

Two robust foliar photosynthesis parameters obtained from fluorometry are:

(1) Potential photosynthetic efficiency, given by $(F_m - F_o)/F_m = F_v/F_m$ and obtained predawn when the maximum number of PS II reaction centres are open. (PSII is that portion of the photosynthetic apparatus that splits water into molecular oxygen, electrons, and protons.) and

(2) Maximum observed photosynthetic efficiency ($F_v{}'/F_m{}' = [F_m{}' - F_o{}']/F_m{}'$), obtained under high foliar light conditions.

Further, one can calculate a percent photoinhibition (defined as a response to excess light energy that causes a reduction in PSII photosynthetic efficiency and, possibly, degradation of PS II reaction centres) as:

(3) Growing-season based photoinhibition: G-S Pi $= [\mathrm{max}F_v/F_m - F_v{}'/F_m{}']/[\mathrm{max}F_v/F_m]$,

where $\mathrm{max}F_v/F_m$ is the highest F_v/F_m value found during the growing season. Our consideration of G-S Pi photoinhibition is a first use for the understanding of N deposition effects on plants in natural settings.

2.4. *Net ecosystem CO_2 exchange data*

AmeriFlux CO_2 eddy flux data were provided (A. Turnipseed, personal communication, April 2003) using the approach of Goulden et al. (1996) applied to the data obtained at the 21.5 m tower height. NEE was determined by adding the beneath canopy storage of CO_2 to the half-hour averaged CO_2 eddy flux values with half-hour canopy storage values were determined using a vertical profile system of CO_2 measurements (Monson et al., 2002). Post processing of NEE data introduced a data record with some spline filling of half-hour values (A. Turnipseed, personal communication, September 2003). Half-hour NEE data were considered for which < 4 missing values were spline filled.

2.5. *Statistical methods*

Data, including the response of NEE to CNU, photosynthetic photon flux density (PPFD), and other variables were statistically analyzed using Statgraphics Plus5 (Statpoint Inc. Herndon, VA, USA) software. Regression models, using *F*-statistics and associated p values for quality of linear and nonlinear fits, were constructed (considering guidelines described in Zar (1984)) to determine the portion of NEE variability explained by each of the regression variables.

3. Results and discussion

3.1. *Canopy N uptake (CNU) calculations*

Data considered here were collected across the 2000, 2001 and 2003 growing seasons; 2002 is not included since it was the driest year on record at the Niwot LTER forest (>100 yr). Total precipitation during the May–October growing season was close to the 50 yr mean of 33 cm (σ = 8.2 cm). Sixty-two events with quality WD, DD and TF results were obtained across a majority of the growing season. Growing season extrapolated wet and dry deposition, throughfall, and CNU results are shown in Table 1 for 2000 and 2001, when HNO_3 and NH_3 dry deposition measurements were obtained.

The DD values for HNO_3 and NH_3 dry deposition were obtained using the gradient technique (Sievering, 2001; Torizzo and Sievering, 2002). The DD for NO_x as well as particulate NH_4^+ and NO_3^- were obtained using dry deposition velocities of Ruijgrok et al. (1997) and Wesely et al. (1982) and were <5% of their respective WD values. Thus, the relatively high uncertainty in the latter DD values does not contribute in any significant way to calculated CNU and its uncertainties. Table 1 also shows that the magnitude of TF is small compared to WD + DD with uptake efficiencies by the canopy being very high (~90% for ammonium and 70–80% for nitrate). These high uptake efficiencies are indicative of the fact that the Niwot LTER forest is N-limited in its growth. Uptake efficiencies for nitrate were quite variable across precipitation events, whereas variability of ammonium uptake efficiency was relatively small. High canopy ammonium uptake is often reported at coniferous forests, while substantial uptake of nitrate is less common (Parker, 1983). A study at a Rocky Mountain conifer forest similar to ours, without consideration of DD, found an ammonium uptake efficiency of ~0.70 with little uptake of nitrate (Arthur and Fahey, 1993).

Table 1. Growing season (gro.sea.—May to October) 2000 and 2001 wet (WD) and dry (DD) flux estimates for ammonium and for NO_y ($= HNO_3 + NO_x +$ particulateN) at Niwot LTER conifer forest canopy. Units are kgN ha^{-1}gro.sea.$^{-1}$. Also shown are uptake efficiency and canopy N uptake (CNU) for ammonium and nitrate as well as CNU for inorganicN (sum of NH_4^+ and NO_3^-). Numbers in parentheses are ±90% confidence intervals.

Ammonium					Nitrate					
WD	DD	TF	CNU	Uptake efficiency	WD	DD [NO_y]	TF	CNU	Uptake efficiency	Inorganic CNU
2000										
0.78	0.06	0.07	0.77	0.92	0.78	0.81	0.29	1.30	0.82	2.07
(0.05)	(0.03)	(0.01)	(0.09)	(0.12)	(0.05)	(0.29)	(0.02)	(0.35)	(0.27)	(0.45)
2001										
1.66	0.05	0.17	1.54	0.90	1.51	0.31	0.54	1.28	0.70	2.82
(0.12)	(0.03)	(0.02)	(0.13)	(0.04)	(0.11)	(0.22)	(0.03)	(0.29)	(0.20)	(0.43)

Measurements at a northeastern US forest have indicated ammonium and nitrate uptake efficiencies as high as 0.90 (Sievering et al., 2000). These high uptake efficiencies support the notion that the Niwot LTER forest is N-limited in its growth.

Both the 2000 and 2001 growing season inorganic CNU flux of 2.07 and 2.82 kgN ha^{-1} (see Table 1) indicate a substantial uptake magnitude by the canopy of atmospheric N deposition. Canopy uptake or loss of organic N was also addressed during the 2001 field season. Total dissolved N minus inorganic N in samples provided a measure of the organic N in both WD and TF collections. The canopy was found, for the growing season as a whole, to be essentially neutral with respect to organic N, given that measured WD organic N (0.93 kgN ha^{-1}) and TF organic N (0.99 kgN ha^{-1}) were statistically the same. Event WD, DD, and TF data were also collected during 2003. However, sampling was not done across the entire growing season so that 2003 values are not shown in Table 1.

The ammonium growing season WD magnitudes for 2000 and 2001 bracket the longer-term mean ammonium (1.18 ± 0.39 kgN ha^{-1}) growing season WD; this is also true for nitrate (mean $= 1.13 \pm 0.31$ kgN ha^{-1}). The parametrization of HNO_3 DD is only dependent on meteorological data. Since it dominates the entire DD at this site, this parametrization allows for the estimation of growing season DD beyond 2000 and 2001. Thus, it is appropriate to incorporate mean 2000, 2001, and 2003 WD and DD data in the canopy N budget of the next section. Discounting any net organic N uptake by (or gaseous N loss [Section 2.1] from) the canopy, results in CNU across three growing seasons (2000, 2001, 2003) of ~2.5 kgN ha^{-1}, that is likely representative of longer-term magnitudes for CNU at this site.

3.2. Canopy N budget

Given that our site's canopy mass is, on an annual basis, within 3–5% of steady state (Tomaszewski et al., 2003), the N required for canopy new growth may be equated to the sum of the annual N lost in litterfall plus reallocation of N to new growth. Further, annual litterfall N loss from the canopy can be equated to the sum of the sources of new N to the canopy: CNU; N made available by way of soil processing and root uptake and N fixation. Perez and Bayley (2003) have shown, for Rocky Mountain Engelmann spruce forests, that N fixation is quite small, ≤0.3 kgN $ha^{-1}yr^{-1}$, and almost entirely occurs by way of the soil. Soil/root N uptake and transfer to the canopy can be estimated by difference, once subtracting N fixation. Finally, canopy N requirement may be partitioned into four component sources: (1) CNU; (2) reallocation; (3) soil/root uptake and (4) N fixation. Figure 1 shows these flux pathways and the resultant Niwot Forest's canopy N budget.

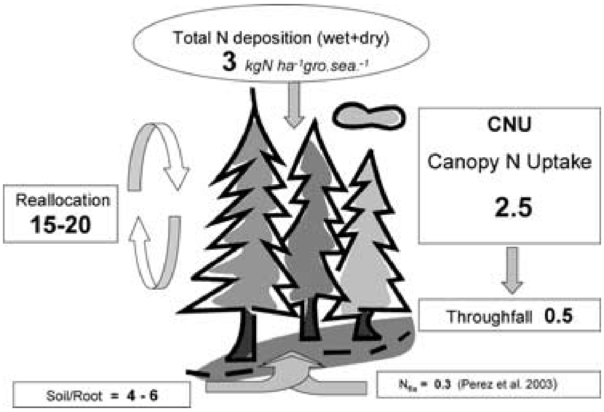

Fig. 1. The canopy N budget for the Niwot LTER forest. Growing season (gro.sea.) canopy N flux magnitudes (see text calculations) are based on three growing seasons' data. All units are in kgN ha^{-1}(gro.sea.)$^{-1}$

The maximum observed N in new growth foliage determines the annual foliar N requirement. Tomaszewski et al., (2003) showed that foliar N requirement in 2000 was 19 kgN ha^{-1} and, for 2001, 22 kgN ha^{-1}. Thereafter, a value for 2003 was found to be 23 kgN ha^{-1}. The N required for branch growth was found, in 2001, to be 4 kgN ha^{-1} and is expected to differ very little from year to year. Given the uncertainty inherent in such determinations, a mean of 25 kgN ha^{-1} will be assumed as most applicable for the N required for canopy (foliar + branch) growth. These growing season canopy N requirement data show that the large majority of annual aboveground N requirement is due to current-year foliage.

Litterfall collections across three years (L. Scott-Denton, personal communication, February, 2003) plus analysis for foliar and branch N in this litterfall indicate that 7–8 kgN ha^{-1} yr^{-1} was lost as part of canopy N litter. This may be subtracted from the 25 kgN ha^{-1} canopy N requirement to estimate the magnitude of reallocated N as 17–18 kgN ha^{-1}. Once uncertainties are considered, 60–80%, or 15–20 kgN ha^{-1}, of the canopy N required during any one growing season may be attributed to reallocation. The literature indicates that the portion of canopy N requirement provided by reallocation varies greatly across species and study sites; mature coniferous forests range from about 30% to as much as 80% (Cole and Rapp, 1981; Birk and Vitousek, 1986; Friedland et al., 1991). The large portion of canopy N requirement met by reallocation at our site strongly supports earlier data (e.g. Sievering, 2001) that shows this conifer forest to be N-limited in its growth. Other recent support comes from a study of stream chemistry at the tundra above and the watershed including our forest site (Hood et al., 2003). High inorganic N concentrations were found in stream water as it entered the subalpine forest area but these concentrations dropped below detection quite rapidly within the forest.

From the viewpoint of new N sources to canopy N requirement, the remainder of 7–8 kgN ha^{-1} must be met by the sum of CNU and soil/root uptake. Given that CNU in 2000–2003 growing seasons was ~2.5 kgN ha^{-1}, the soil/root pathway (including N fixation) appears to be only twice that of CNU, ~5 kgN ha^{-1}. Thus, one may expect CNU to be an important contributor to canopy N demand in any one year at this spruce-fir forest.

Of further interest are decadal timescale considerations of the canopy N budget. Over a ten year, or longer, timescale the soil/root pathway becomes a second recycling pathway, in addition to canopy reallocation. This, except for the N provided by N fixation ($\leq$0.3 kgN ha^{-1}yr^{-1}). Thus, on decadal and longer timescales, the CNU of ~2.5 kgN ha^{-1} per growing season provides about ten times more new, available N to this forest canopy than N fixation. In fact, the small growing season throughfall N flux (0.5 kgN ha^{-1}—see Fig. 1) annually supplies more new N to the forest floor for possible soil/root uptake than does N fixation.

The above canopy N budget results are shown in Fig. 1. This figure illustrates the significant perturbation of the natural N cycle that prevails at this forest given that a majority of the wet plus dry N deposition is anthropogenic in origin. Sources in the Colorado Front Range may have increased growing season N deposition by an order of magnitude (Baron et al., 2000). It is likely that >80% of the new, available N at this forest is derived from anthropogenic sources. Further, the large majority of this anthropogenic new N is directly applied to and retained by the canopy (i.e. CNU).

Given all the above, correlations of CNU with NEE, and with other flux, concentration, and meteorological data obtained from the nearby AmeriFlux tower were explored. (All N-flux data of Fig. 1 were obtained within the footprint of the AmeriFlux tower.)

3.3. Net ecosystem CO_2 exchange and canopy N uptake (NEE versus CNU)

NEE strongly depends on solar radiation at the Niwot LTER forest. Monson et al. (2002) have shown that a nonlinear dependence of NEE on PPFD with an $r^2 \approx 0.7$–0.8 prevails during the growing season. The observed half-hourly NEE values reach a mean of -8 to -9 μmol m^{-2}s^{-1} (negative is forest uptake) under high PPFD conditions.

This strong PPFD influence on NEE must be accounted for when considering the possible dependence of NEE on CNU. Any correlation of NEE and CNU variability may also depend on the timescale over which one considers the canopy uptake of atmospheric N deposition. A response of NEE to short-term variability in dry deposition alone may be of importance. On the other hand, this response may not be noticeable until a sequence of CNU events (i.e. over several wet deposition intervals) have been aggregated. Our data indicate that NEE response times in relation to CNU may be optimally separated by each precipitation event (Sievering, 2003). This suggests that canopy wetness provides for the mobilization of N dry deposition that occurs during the antecedent dry period prior to a wet deposition event. This mobilized N, along with the N in wet deposition, is then made available to canopy foliage for use in photosynthesis (Tomaszewski, 2006). This view of the manner by which CNU may contribute to C uptake and sequestration is in line with a number of European and US studies. Horn et al. (1989) found that only 2% of the foliar N required for growth at a German spruce forest was derived from direct gaseous uptake by stomata but that 24% was derived from uptake in the liquid phase. Harrison et al. (2000) have reviewed several similar studies and conclude: (1) foliar uptake from the liquid phase is much more important than that from the gaseous phase; (2) under these circumstances, uptake of NH_4^+ is much faster than NO_3^- uptake; (3) NH_4^+ is more readily available to participate in photosynthesis and (4) CNU may substantially contribute to N requirement for growth at a number of European forest sites. This last point supports the notion that the Niwot Forest results may be applicable to other conifer forest sites. Further, this point suggests that it may be CNU, and not N deposition alone, that shows a significant correlation with NEE. In fact, a comparison of the correlation of total N deposition versus NEE with that of CNU versus NEE showed that the latter is significant (with a P value of 0.006, see below) but the former is not ($P > 0.05$).

A total of 26 CNU events with quality NEE, PPFD, and also WD, DD, and TF for NH_4^+, NO_3^- and organic N data were obtained in 2001. Nighttime periods were removed from this

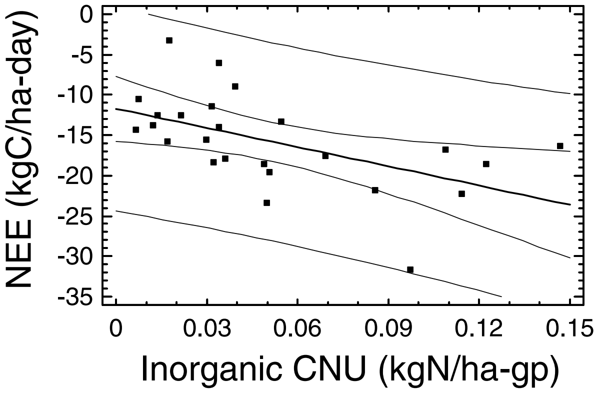

Fig. 2. Daily mean daytime net ecosystem CO_2 exchange (NEE) versus inorganic canopy nitrogen uptake of atmospheric deposition (inorganic CNU) based on a growth period (gp) timescale = antecedent dry period + ensuing wet deposition time. The linear regression line is NEE $= -11.7 - 79.4 \bullet$ (inorganic CNU); $r^2 = 0.27$ ($P = 0.006$). Also shown are the 98% confidence and prediction bands; prediction band shows no data point is an outlier ($P < 0.01$). The standard assumptions of regression analysis—normality, independence, and homoscedasticity—were all met at $P < 0.01$.

Table 2. Linear regressions of daily mean daytime NEE (kgC $ha^{-1}d^{-1}$) versus components of canopy nitrogen uptake (CNU), kgN $ha^{-1}gp^{-1}$, where gp (growth period) = antecedent dry period + ensuing wet deposition time. Inorganic CNU is the sum NH_4^+ and NO_3^-.

Model	P	r^2
NEE vs. inorganic CNU	0.006	0.27
NEE vs. total dissolved N-CNU	0.34	0.03
NEE vs. NH_4^+ CNU	0.02	0.22
NEE vs. NO_3^- CNU	0.09	0.07

Table 3. Multiple regression results for daily mean daytime NEE (kgC $ha^{-1}d^{-1}$) versus variables with added-last-$P < 0.025$. Soil water content is considered a surrogate measure for water availability.

Model variable	Added-last-P	Partial r
PPFD	0.0005	0.79
Inorganic CNU	0.018	0.44
Soil water content	0.021	0.39

analysis since the influence of CNU on daytime photosynthesis is the focus here. Further, there was no correlation evident between nighttime NEE and CNU values ($r^2 = 0.04$, $P = 0.55$).

Correlations of daytime NEE (PPFD > 10 μmol m^{-2} s^{-1}) versus CNU were conducted on hourly (dry deposition only), on multihourly (dry deposition only), and on antecedent dry period + ensuing wet deposition timescales. CNU, based on hourly and multihourly dry deposition timescales, had insignificant correlations versus NEE ($P > 0.36$) despite the large variability in N dry deposition at our forest (the site is often, during summer months, impacted by mountain upslope, high N concentration episodes with a factor of 3–8 greater N dry deposition occurring during these episodes (Sievering, 2001).

Figure 2 shows the relationship of daily mean daytime NEE versus inorganic CNU for the [antecedent dry period + ensuing wet deposition] timescale. This [antecedent dry period + ensuing wet deposition] period will be called a 'growth period' or simply gp. Note, in Fig. 2, that all daytime NEE values associated with gp periods are negative, that is, CO_2 uptake by the forest, or short-term forest growth periods (thus, gp). The mean gp duration was 73 hr. The best linear relationship between daytime NEE and inorganic CNU is (see Table 2):

$$\text{daytimeNEE, kgC/ha-day} = -11.7 - 79.4 \bullet (\text{inorganicCNU, kgN/ha-gp}) \quad (1)$$

with $r^2 = 0.27$ ($P = 0.006$).

Table 2 also shows that total dissolved N-CNU is not a significant independent variable, as expected since the canopy was found to be neutral with respect to organic N. Further, ammonium and nitrate, separately (Table 2), are not as significant as is inorganic CNU (sum of both).

The dependence of daytime NEE on CNU must, however, be considered in the context of other independent variables, especially PPFD. Stepwise multiple regression analysis, using $P < 0.025$ as the acceptance criterion of a variable and considering over 20 variables, provided a model with PPFD strongly contributing (as found by Monson et al., 2002) and with both inorganic CNU and soil water content as significant explanatory variables, in this order. (Hourly soil water content data, at 25 cm depth, were available [Niwot LTER Prog. data] within the forest and only 0.7 km removed from the AmeriFlux tower; soil water content is considered a surrogate measure for water availability.) Table 3 presents a summary of the multiple regression results.

It is notable that inorganic CNU enters the multiple regression ahead of soil water content. This suggests that canopy uptake of atmospheric N deposition may have a greater influence on daytime NEE variability than does water availability at this conifer forest. Since most of the inorganic CNU is delivered with precipitation, it is important to separate the effects of these two factors. Greater precipitation contributes to higher soil moisture levels that, in turn, allow for greater stomatal conductance and, thereby, may increase photosynthesis (Kozlowski et al., 1991). Increases in daytime NEE may also result. Modelling studies have shown forest carbon uptake may be reduced during periods of low soil water content (e.g. Ollinger et al., 1998). However, precipitation amount did not enter into the daytime NEE multiple regression, indicating the effect of CNU significantly outweighs that of precipitation on NEE variability.

The portion of total daytime NEE variability explained by inorganic CNU in the above multiple regression model is about 20% ($P < 0.02$). This lower r^2 value is more appropriate than the ~27% obtained from single regression. Yet, this 20% indicates that a substantial fraction of daytime NEE may be attributed to canopy uptake of atmospheric inorganic N deposition at this conifer forest. AmeriFlux program data (A. Turnipseed, personal communication, May 2004) indicate that the forest canopy's daytime NEE is more than 25 times that of the forest understory. Thus, throughfall N and N deposited directly to the understory in canopy gap areas has a negligible influence on daytime NEE.

Consideration of the 2000 and 2003 growing season CNU data together with that of 2001 also indicated increasing NEE with increasing CNU, but without significance ($P = 0.09$). If only CNU events with >0.06 kgN $\mathrm{ha}^{-1}\mathrm{gp}^{-1}$ are considered, P becomes <0.05 for the combined 3-yr NEE versus CNU data and the inorganic CNU regression coefficient becomes a larger negative value than that in eq. (1). The large P of 0.09, considering the entire 2000, 2001 and 2003 data, may be due to the fact that 2000 and 2003 dry deposition N fluxes are based on parametrizations (Torizzo and Sievering, 2002) as opposed to the more directly obtained (gradient technique; Sievering et al., 2001) dry deposition HNO_3 and NH_3 fluxes for the 2001 growing season.

The statistical results above indicate that the flux of atmospherically deposited N into foliage at this conifer forest does very likely influence NEE measurements. Further, these results suggest a short timescale (mean gp = 73 hr) for this influence; on the order of a few days. It is possible that such a relatively fast response to CNU is, partly, due to the quite rapid foliar uptake of atmospherically deposited N into the amino acid pool of spruce needles, only 4 ± 2 hr delay time, at the Niwot Forest (Calanni et al., 1999). Studies using ^{15}N-labelling at eastern US coniferous forest sites also indicate relatively rapid uptake of atmospherically deposited N by the foliar amino acid pool (e.g. Vose and Swank, 1990; Boyce et al., 1996).

3.4. *Supporting chlorophyll fluorescence and gas exchange data*

The accompanying paper by Tomaszewski and Sievering (2007—part II, section 3.1 and Table 1) presents the photosynthetic capacity (by fluorometry) and the CO_2 gas exchange (by infrared gas analysis) data resulting from a spruce branch wet deposition N-amendment study in the Niwot Forest canopy during the 2004 growing season. The methods section of that paper notes this branch applied N increased 2004 growing season wet deposition by 1.4 kgN ha^{-1}; that is, by 60% above ambient wet deposition (2.4 kgN ha^{-1}). Given that growing season dry N deposition is (on average) ~30% of wet deposition at the Niwot Forest (Tomaszewski et al., 2003), the N-amendment of 1.4 kgN ha^{-1} increased total (wet plus dry) deposition by 40–50% above total ambient N deposition for the N-amended spruce trees. Fluorometry data were obtained under high light, as well as predawn, conditions and provided data on potential photosynthetic efficiency (F_v/F_m—predawn), maximum observed photosynthetic efficiency (F_v'/F_m'—high light; occurred 1000–1100), and growing season photoinhibition (G-S Pi; see Section 2.3-3). Since the amended N was applied in a DI water solution with a similar ion matrix as that of mean precipitation at the site (except for higher NH_4^+ and NO_3^- concentrations) both control (same water solution as for N-amendments, except no NH_4^+ and no NO_3^- in solution) and background branches (no spray applied at all) were part of the experimental design.

Fluorometry results for F_v'/F_m' and G-S Pi differed for N-amended shoots versus both control and background spruce shoots. For old growth (>1 yr old) shoots, F_v'/F_m' was 11–12% greater ($P < 0.05$) and G-S Pi was ~25% less ($P < 0.05$) for N-amendment shoots. Similar results were found for new growth shoots, although not with significance ($P > 0.05$; greater variability is inherent in new growth fluorometry and, also, gas exchange data). Thus, wet N deposition in N-amendment sprays provided for more photosynthetic efficiency and reduced photoinhibition in spruce branches' photosynthetic apparatus at the Niwot Forest.

Although not significant, at the end of the 2004 growing season area-based N content (gN m^{-2}) for the N-amended branches' old growth needles was 8% greater than that for the mean of these same needles on control and background branches. This lends credence to the results from fluorometry. Gas exchange data provided by the portable IRGA showed that the maximum rate of carboxylation (V_{cmax}) was 14–15% greater ($P < 0.05$) for the N-amended old growth spruce shoots versus control and background old growth shoots with, again, similar results for new growth shoots (Tables 2 and 3 of Tomaszewski and Sievering, 2007—part II).

Combined, the fluorometry and gas exchange data show that canopy uptake of atmospherically deposited N improved the photosynthetic capacity of spruce shoots and, thereby, increased the rate of assimilation and carboxylation of CO_2. Both the fluorometry and gas exchange results provide substantial physical supporting evidence for the statistical results that show increasing daytime NEE with increasing CNU, eq. (1). Further, these results indicate that CO_2 uptake may be expected to increase with CNU at N-limited conifer forests such as the Niwot Forest (although further experimentation is warranted).

Consideration of the process-based gas exchange model for C_3 plants of Niinemets and Tenhunen (1997) provides further supporting evidence. The proportion of foliar-N-in-Rubisco, P_r, may be estimated using their model:

$$P_r = V_{cmax}/(6.25 \bullet N_a \bullet V_{cr}), \tag{2}$$

where V_{cr}, the maximum rate of RUBP carboxylation per unit Rubisco, may be estimated using temperature dependent parameters in Appendix A of Niinemets and Tenhunen (1997). Given that V_{cmax} and N_a are the two inputs to eq. (2) that differ for N-amended versus control and background shoots, the data of

Table 2 of Tomaszewski and Sievering 2007—part II) allow for P_r to be estimated for old growth shoots. The P_r is (considering uncertainty) at least 5%, to as much as 9%, greater for the N-amended shoots than for the mean P_r of control and background shoots. It is likely that the improved N-amended spruce shoots' photosynthetic capacity is due to this greater proportion of foliar-N-in-Rubisco. Further, the G-S Pi results indicate the improved N-amended shoots' photosynthetic capacity is, at least partly, due to greater photosynthetic efficiency and possibly less PS II reaction centre degradation. Amended N assimilation, along with incorporation into the photosynthetic apparatus, likely accounts for the enhanced photosynthetic efficiency and capacity found in the N-amended shoots. Thus, the combined fluorometry, gas exchange, and P_r estimation provide a measure of mechanistic explanation for the NEE dependence on CNU, eq. (1).

4. Conclusions

The results shown in Tables 2 and 3 as well as Fig. 2 indicate that forest CO_2 uptake increases with increments in canopy uptake of atmospheric inorganic N deposition at the Niwot Forest. The large majority of this canopy N uptake—CNU—occurs in the foliage since 80–90% of the forest canopy's N required for growth is in the foliage and also since canopy lichen and microbes do not contribute in any significant way to CNU. This foliar N flux contributes to forest CO_2 uptake (daily mean daytime NEE versus inorganic CNU, Fig. 2). Based on 3-day time-step, multiple regression analysis, ~20% of the variability in growing season, daytime NEE is explained by CNU and it appears to contribute more to NEE than precipitation magnitude or soil water content.

Nitrogen measurements at this site indicate the forest is N-limited in its growth. This N limitation, likely, contributes to the observed dependence of daytime NEE on inorganic CNU fluxes. A strong statistical dependence of CO_2 uptake was found on short timescales of ~3 d, despite relatively small increments of atmospheric N deposition. This conclusion is supported by fluorometry data showing that greater photosynthetic efficiency and less photoinhibition prevailed for N-amended spruce branches, relative to control and untreated branches. These N-amended branches also had greater V_{cmax} and a greater proportion of foliar-N-in-Rubisco. The mean CNU flux of $\sim$0.06 kgN ha^{-1} over these ~3-d periods is less than occurs at many European and US conifer forests (Johnson and Lindberg, 1992; Thimonier et al., 2005). Extrapolation of the results found at the Niwot Forest to other conifer forests (that are not N-saturated) suggests that increased CO_2 uptake, derived from CNU, may equal several hundred kgC $ha^{-1}yr^{-1}$ for some European and U.S. forest sites. Consideration of four eastern U.S. forests for which CNU data are available (Sievering et al., 2000) indicates that 250–1350 kgC ha^{-1} additional CO_2 uptake likely results from the enhanced NEE that is driven by CNU. Thus, a substantial percentage of growing season net NEE, a greater percentage than at the Niwot Forest, may result from canopy N uptake at these eastern U.S. forests.

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