Whole-plant optimality predicts changes in leaf nitrogen under variable CO$_2$ and nutrient availability

Silvia Caldararu$^{1*}$, Tea Thum$^1$, Lin Yu$^1$, and Sönke Zaehle$^{1,2}$

$^1$Max Planck Institute for Biogeochemistry, Hans-Knöll Str. 10, 07745 Jena, Germany

$^2$Michael Stifel Center Jena for Data-driven and Simulation Science, Jena, Germany

$^*$Corresponding author: Silvia Caldararu, scaldra@bgc-jena.mpg.de

Summary.

- Vegetation nutrient limitation is essential for understanding ecosystem responses to global change. In particular, leaf nitrogen (N) is known to be plastic under changed nutrient limitation. However, models can often not capture these observed changes, leading to erroneous predictions of whole-ecosystem stocks and fluxes.

- We hypothesise that an optimality approach can improve representation of leaf N content compared to existing empirical approaches. Unlike previous optimality-based approaches, which adjust foliar N concentrations based on canopy carbon export, we use a maximisation criteria based on whole-plant growth and allow for a lagged response of foliar N to this maximisation criterion to account for the limited plasticity of this plant trait. We test these model variants
at a range of Free-Air CO\textsubscript{2} Enrichment (FACE) and N fertilisation experimental sites.

- We show a model solely based on canopy carbon export fails to reproduce observed patterns and predicts decreasing leaf N content with increased N availability. However, an optimal model which maximises total plant growth can correctly reproduce the observed patterns.

- The optimality model we present here is a whole-plant approach which reproduces biologically realistic changes in leaf N and can thereby improve ecosystem-level predictions under transient conditions.

1 Introduction

The response of plants to changes in environmental conditions such as climate, atmospheric CO\textsubscript{2} concentration or nutrient inputs is dependent on the plants’ ability to acclimate and adapt to the new conditions. In particular, there is strong evidence that plants can respond plasticly to changes in nutrient limitation (Bloom et al., 1985; Chapin et al., 1986), which in turn affects their response to elevated atmospheric CO\textsubscript{2} (Ainsworth & Long 2005; Long et al., 2004). However, such processes are lacking or poorly represented in terrestrial biosphere models (TBMs) (Prentice et al., 2015).
Therefore, in this study we set out to provide a better representation of plant, and specifically canopy, response to increasing nutrient limitation.

One of the main roles of nitrogen (N) in plant physiology is as a component of proteins and specifically enzymes that form part of the photosynthetic apparatus (Hawkesford et al., 2012; Evans, 1989), which is why leaves create the highest growth demand for N. Therefore, understanding leaf N physiology and its response to changing environmental drivers is essential for understanding whole-plant and ecosystem level response to N limitation.

A decrease in leaf N content has been observed over the last decades and has been previously attributed to increasing atmospheric CO$_2$ (Jonard et al., 2015; McLauchlan et al., 2010). A similar decrease has also been observed under experimentally elevated CO$_2$ (Ellsworth et al., 2004). On the other hand, increased leaf N concentration occurs in nitrogen addition experiments (e.g. Magill et al., 2004; Houle & Moore, 2008; McNulty et al., 2005; Sikström, 2002) and under high N deposition (Fleischer et al., 2013; McNeil et al., 2007). There have been a number of explanations proposed for this observed plasticity, including both plant N use and changes in the photosynthetic apparatus. Firstly, plants increase their leaf N under increased N availability to make better use of available resources. Under elevated CO$_2$, plants can achieve the same carbon assimilation rate with a
lower leaf N content (Stitt & Krapp, 1999; Drake et al., 1997). Additionally, there are other associated changes in the photosynthetic apparatus under elevated CO$_2$ which can contribute to the observed changes in leaf N (Ainsworth & Long, 2005; Rogers & Humphries, 2000). Another proposed mechanism for the observed decline in leaf N content under elevated CO$_2$ is known as the dilution effect, an accumulation of non-structural carbohydrates in the leaves which cause an overall lower N concentration, although there is little experimental evidence for this process playing an important role (Feng et al., 2015; Wujeska-Klause et al., 2019). Furthermore, the extent to which the leaf N content changes depends on the initial N limitation status of the plant (Ainsworth & Long, 2005; Stitt & Krapp, 1999).

From a modelling perspective, TBMs which include N processes represent the leaf N content either as a fixed parameter or as a flexible value which responds to plant N limitation, generally following an empirical function (Zaehle & Dalmonech, 2011; Thomas et al., 2013a,b). Meyerholt & Zaehle (2015) show that models which include a flexible C:N ratio have better predictive capability than models with a fixed ratio when compared to observations from N fertilisation experiments. However, they also found that models with flexible stoichiometry tend to overestimate the increase in foliar N under N fertilisation, thereby increasing the N costs of new tissue, leading to a lower than observed growth response. A multi-model comparison at two FACE sites has shown that models with flexible stoichiometry
can overestimate the leaf N response to elevated CO$_2$, in some cases reaching unrealistically low values (Zaehle et al., 2014). These two results together point to a need for a better representation of dynamic leaf N to capture the N dynamics of ecosystems under changing environmental conditions.

Optimality principles have long been proposed as a theoretical way of representing plant plasticity (Givnish, 1988; Mäkelä et al., 2002). The underlying hypothesis is that plants alter their physiology or morphology to maximise growth over their lifetime. The concept has been previously applied to individual processes such as photosynthesis (as the coordination hypothesis Medlyn, 1996; Maire et al., 2012; Ali et al., 2015; Smith et al., 2019), stomatal conductance (Cowan & Farquhar, 1977; Medlyn et al., 2011), biomass allocation (Mäkelä et al., 2008), nutrient content (Dewar, 1996) and phenology (Caldararu et al., 2014). Optimality has the potential to improve the representation of vegetation processes in TBMs by providing a way to include complex responses that can accurately capture observations without a large number of additional parameters. A number of studies have built basic whole-plant models centred around the optimality hypothesis (Franklin, 2007; McMurtrie et al., 2008). More recently, optimality concepts, and specifically leaf-level coordination of photosynthetic parameters, have started to be incorporated into full TBMs (e.g. Ali et al., 2015; Haverd et al., 2018). However, there are still a number of open questions about the exact mode of implementation, including which processes and
variables to optimise and the timescale of this optimisation (Dewar et al., 2009).

Here, we implement an optimality-based method for representing changes in leaf N within a newly developed TBM, QUINCY (QUantifying Interactions between terrestrial Nutrient CYcles and the climate system, Thum et al., 2019). A TBM, with its detailed representation of soil and vegetation processes, allows us to identify the drivers and assess the implications, of changes in leaf stoichiometry given the ecosystem-scale nutrient balance. In particular, the detailed representation of canopy and photosynthetic processes allows us to go beyond the basic optimality models used in the past and predict time-variant leaf N content and not simply at-equilibrium values. Instead of relying on the assumption that plant properties (here leaf N) are optimal at any timestep, we rely on a dynamic, towards equilibrium approach, in which plants adjust their properties (here leaf N) at each model timestep towards the optimal value given average environmental conditions over a predefined period. This approach reflects the known limitations to plant plasticity and thereby provides a solution to the timescale of optimisation problem introduced above. On the other hand, the use of optimality theory allows us to leverage basic processes already existent within the TBM and therefore represent complex plastic behaviour without introducing a large number of parameters in addition to those already present in the model.
In this study, we test the hypothesis that changes in leaf N concentrations can be explained by two main drivers: (1) the limitation to growth by N availability caused by an increase in leaf N content and, (2) the increase in carbon export, and thus growth, through an increase in leaf N content. The first driver is the one used by existing models to describe variations in leaf N, while the second is commonly used in optimality approaches. To explore these two drivers we use four different model setups for representing leaf N: fixed leaf N content, empirical (which includes only the N availability criteria), optimal C export (which includes only the maximum C export criteria) and optimal growth (which includes both). We test the effect of these different formulations on plant growth and foliar N responses to changes in CO$_2$ and N availability using the resulting model at two FACE sites and a range of N fertilisation sites.

2 Methods

2.1 Model description

2.1.1 The QUINCY terrestrial biosphere model

We implement the above options in a terrestrial biosphere model, QUINCY, described in detail in Thum et al. (2019). Here we briefly present the processes related to the representation of dynamic leaf N, followed by a
detailed description of the optimality implementation.

QUINCY represents fully coupled C, N and P as well as water and energy cycles (Fig. 1). The model employs a multi-layer canopy scheme, which includes a representation of photosynthesis and canopy conductance, for sunlit and shaded leaves separately within every layer. Total canopy N is distributed to each layer, with exponentially decreasing N content towards the bottom of the canopy. Variations in average leaf N content (described below) are propagated to each canopy layer. Photosynthesis is represented following the model of Kull & Kruijt (1998). Leaf N is separated into structural and photosynthetic (chlorophyll, Rubisco, and electron transport) fractions, each of which is directly related to parameters in our photosynthesis model (Kull & Kruijt 1998; Zaehle & Friend 2010). The structural N fraction is expressed as a linear function of leaf N content, decreasing with higher leaf N (Evans 1989; Friend et al. 1997). The fraction of photosynthetic N allocated to chlorophyll increases with canopy depth, while the Rubisco and electron transport components are adjusted to give a constant ratio of the respective photosynthetic parameters ($V_{cmax,25}$ and $J_{max,25}$). Maintenance respiration in the model is represented as a linear function of specific tissue N. Growth respiration is calculated as being proportional to the amount of new tissue built. Both photosynthesis and maintenance respiration acclimate to growth temperature following Friend (2010) and Atkin et al. (2014).
Nutrient uptake is calculated as a function of fine root biomass, soil inorganic nutrient content and plant demand for each specific nutrient, where the demand depends on the ratio of available to required nutrients for growth.

Photosynthesis and growth processes are separated in QUINCY by the introduction of two non-structural pools, the labile and reserve pool. Carbon assimilated through photosynthesis and nutrients taken up by the roots enter the labile pool, and are subsequently allocated to new tissue growth, transferred to the reserve compartment, or in the case of carbon, used for respiration. Growth follows a functional balance approach based on an extended pipe-theory representation, generating size-dependent allometric relationships between leaf, fine root, coarse root, and sapwood mass. Actual growth is constrained by available nutrients and tissue-specific stoichiometry. Stand-level vegetation dynamics is represented as density-dependent mortality and establishment based on the existing seedbed, as the model explicitly allocates to a reproductive pool. All pools and fluxes are representative of an average individual.

The QUINCY model includes a detailed representation of soil carbon and nutrient pools, the details of which can be found in [Thum et al. (2019)].
2.1.2 Tissue stoichiometry

The N content of leaves and fine roots at each half-hourly timestep $\Delta t$ is updated given a direction variable, $D_N$ (unitless), and a parameter representing the maximum rate of change, $\delta_N$ (day$^{-1}$), so that:

\[
N_{\text{leaf},t} = N_{\text{leaf},t-1} \times (1 + D_N \times \delta_N \times \Delta t) \quad (1a)
\]

\[
N_{\text{fine root},t} = N_{\text{fine root},t-1} \times (1 + D_N \times \delta_N \times \Delta t) \quad (1b)
\]

The $N_{\text{leaf}}$ values here refer to canopy average values of leaf N content. To conserve the mass balance, new tissue is added with a leaf N equal to that calculated above, while old tissue changes its N content towards the target C:N by recycling N through the labile pool at a timescale of 10 days. All values of leaf N content below refer to this target average leaf N value.

The C:N ratio of fine roots is represented as being directly proportional to that of leaves [Zaehle & Friend, 2010]. We consider the N content of the woody and fruit pools to be constant [Meyerholt & Zaehle, 2015]. Even though the model variants described below are centred on changes in leaf N, the associated changes in fine root N need to be taken into account at the whole plant level and will be calculated throughout. Note that as in our model the specific leaf area (SLA) remains constant, a fractional change in total leaf N results in an equal fractional change in C:N ratio and leaf N content per mass and per area.
Below, we describe in detail the four model options for calculating foliar stoichiometry, which in essence are four options for calculating the value of $D_N$. The rate of change parameter, $\delta_N$ is the same for all variants.

### 2.1.3 Variant 0: fixed leaf N

For the basic model variant, tissue stoichiometry is kept constant and the value of $D_N$ is simply set to zero. Stoichiometry parameters are prescribed, following PFT-specific values from the TRY trait data base [Kattge et al. 2011].

### 2.1.4 Variant 1: empirical leaf N

The empirical variation is based on Zaehle & Friend (2010). The leaf N concentration is modified based on the relative availabilities of labile carbon ($C_{labile}$) and nitrogen ($N_{labile}$) for growth, given the stoichiometric requirement, i.e. the C:N ratio $\chi_{CN}^{growth}$. The direction of change variable, $D_N$, is calculated as:

\[
D_N = \begin{cases} 
-(\frac{\chi_{leaf}^{CN} \times N_{labile}}{N_{labile, min} + N_{labile, max}})^{\chi_{leaf}} & \text{if } C_{labile} \leq N_{labile} \times \chi_{CN}^{growth} \\
(1 - e^{-(\frac{\chi_{leaf}^{CN} \times N_{labile}}{N_{labile, min} + N_{labile, max}})^{\chi_{leaf}}}) & \text{if } C_{labile} > N_{labile} \times \chi_{CN}^{growth} 
\end{cases}
\]

(2)

In the above, $\chi_{leaf}^{X}$ and $k_{leaf}^{X}$ are empirical shape parameters, $\chi_{CN}^{leaf}$ is the current leaf stoichiometric ratio and $\chi_{leaf, min}^{CN}$ and $\chi_{leaf, max}^{CN}$ are PFT-specific
parameters, taken from the TRY database ([Kattge et al. 2011](#)). The C:N ratio of new growth $\chi^{CN}$ is a variable calculated at every timestep given fractions allocated to each pool and current stoichiometry of all pools. Note that unlike the two optimality schemes described below, here $D_N$ can take values between -1 and 1 but the same notation has been kept for consistency.

### 2.1.5 Variant 2: optimal C export

Our first model assumption is that plants modify their leaf N to maximise net C assimilation. Increased leaf N will increase photosynthesis (gross assimilation, $A_g$) but also increase tissue maintenance respiration, resulting in a peaked relationship between net assimilation ($A_n$) and leaf nitrogen, as shown in Fig. 2. Due to the non-linear nature of N distribution in the canopy and the photosynthesis model used in QUINCY, we calculate the optimal leaf N for which net carbon export is maximised through a numerical approach, as follows.

We calculate the direction in which the leaf N content needs to be modified by increasing the current leaf N content $N_{leaf,t-1}$ by a given amount $\delta_N$ and the fine root N content by the equivalent amount to obtain two new values, $N_{leaf,\delta}$ and $N_{fine,root,\delta}$.

We then calculate the net assimilation $A_{n,\delta}$ given the new $N_{leaf,\delta}$ as:
\[ A_{n,\delta} = A_{g,\delta} - N_{\text{leaf},\delta} \times f_{\text{resp,air}} - N_{\text{wood},\delta} \times f_{\text{resp,air}} - N_{\text{fine_root},\delta} \times f_{\text{resp,soil}} - N_{\text{coarse_root},\delta} \times f_{\text{resp,soil}} \] (3)

where \( f_{\text{resp,air}} \) and \( f_{\text{resp,soil}} \) are the maintenance respiration rate per unit N given the temperature of the air and soil respectively. \( A_{g,\delta} \) is the gross canopy assimilation given the new leaf N, calculated for each canopy layer.

As the C:N ratio of wood and coarse roots is considered to be constant, the difference in \( A_n \), \( dA_n \) resulting from the change in tissue N, \( dN_{\text{leaf}} \) is equal to the change in photosynthesis with the change in N, minus the change in tissue maintenance respiration, \( dR_{\text{leaf}} \) and \( dR_{\text{fine_root}} \), with the change in N content of each specific tissue:

\[
\frac{dA_n}{dN_{\text{leaf}}} = \frac{dA_g}{dN_{\text{leaf}}} - \frac{dR_{\text{leaf}}}{dN_{\text{leaf}}} - \frac{dR_{\text{fine_root}}}{dN_{\text{fine_root}}} \] (4)

The \( A_n \) values are calculated given the average meteorological conditions over a given time period, \( \tau_N \) (30 days). Note that this approach means that plants do not reach absolute optimality but there is a rate of change in the optimal direction, with the parameter \( \delta_N \) denoting the maximum amount by which the leaf N content can change in a timestep (Table 1).

We then calculate the direction variable \( D_N \) so that the new actual leaf N increases for a positive return in \( A_n \) and decreases for a negative return:
\[ D_N = 1, \quad if \quad \frac{dA_n}{dN_{leaf}} \geq 0 \]  
\[ D_N = -1, \quad if \quad \frac{dA_n}{dN_{leaf}} < 0 \]  

The optimal direction resulting from this criterion will vary with environmental conditions (e.g. temperature, water availability), atmospheric CO\(_2\) concentration (see Fig. 2), as well as light environment in the canopy as given by direct and diffuse light levels at the top of the canopy but also the amount of leaf area (Fig. S3).

2.1.6 Variant 3: optimal growth

In addition to the optimal C export criterion, here we introduce an additional condition for calculating the \(D_N\) direction variable, based on the relationship between the potential N-limited growth, \(N_{growth}\), and the potential C-limited growth, \(C_{growth}\). The C-limited growth can be calculated as:

\[ C_{growth} = NPP \times \tau_{growth} + C_{labile} \]  

where \(\tau_{growth}\) is the timescale of plant growth, in this case equal to one year, and \(C_{labile}\) is the C content of the labile pool, i.e. the C available for immediate growth. Note that the NPP value here is not the same as \(A_n\) in previous equations as it also takes into account growth respiration and stor-
age fluxes into the reserve pool. The N-limited growth term is calculated
given the available N and the stoichiometry of new tissue, $\chi_{growth}^{CN}$:

$$N_{growth} = (f_{Nup} \times \tau_{growth} + N_{labile} + k_{resorb}^N \times \frac{N_{leaf}}{\tau_{leaf}}) \times \chi_{growth}^{CN} \tag{7}$$

Here, $f_{Nup}$ refers to the N uptake by the roots, $k_{resorb}^N$ is a parameter
denoting the rate of nutrient resorption from senescing leaves and $\tau_{leaf}$ is
the turnover time of the leaves. The term $\chi_{growth}^{CN}$ converts $N_{growth}$ into C
units, to allow comparison with $C_{growth}$. All parameter values can be found
in Table S1. Both C and N availability is calculated at each timestep and
$NPP$ and $f_{Nup}$ are averaged over a time period $\tau_N$, in a similar manner
to environmental drivers. The first term in each equation represents the
uptake capacity for either C or N given current environmental conditions,
while the second term represents the amount already available to the plant.
In addition to these terms, the N-limited growth $N_{growth}$, includes an ad-
ditional flux, the nutrients reabsorbed before leaf shedding. All variables
are averaged over a period of $\tau_N$, as above.

Given the availability of C and N, the direction variable $D_N$ is then
calculated as:

$$D_N = -1, \text{if } C_{growth} > N_{growth} \tag{8a}$$

$$See \text{ eq. 5, if } C_{growth} \leq N_{growth} \tag{8b}$$
so that the leaf N content decreases if there is not sufficient N available for growth and defaults to the optimal C export variant if there is.

2.2 Optimality approach

For the purpose of the optimal C export and optimal growth model variants, we apply an optimality criteria which states that plants aim to maximise the canopy carbon export and growth respectively given moving averages over a period of $\tau_N$ (30 days) of all environmental drivers as well as relevant model state variables, such as NPP and N uptake (Eq. 6 and 7). The optimality criteria is applied at every timestep, resulting in a continuously changing values of the direction variable $D_N$ and therefore a continuously changing leaf N value. The $\tau_N$ parameter is meant to buffer sub-daily and abrupt changes in driving variables. The short timestep ensures that, although the approach only calculates the direction of change, the change in leaf N is smooth.

We use a numerical approach to solve the optimality problem for two reasons. The first is that, while using a TBM with an explicitly layered canopy and complex representation of photosynthesis produces more realistic predictions, it also means that the problem is non-linear and has no analytic solution. The second reason is that one of the central concepts of our approach is that we are not solving for the leaf N values that gives the actual maximum C export or growth at any point in time but rather assume
that plants tend towards equilibrium, given physiological and biochemical constraints to their rate of change.

2.3 Site description

We test all model variants at two Free Air CO$_2$ Enrichment (FACE) sites, the Duke Forest (hereafter Duke) and Oak Ridge National Laboratory (hereafter ORNL) experimental sites as well as at the Harvard Forest N addition experiment.

The Duke FACE experiment ([McCarthy et al., 2007]) was carried out in a loblolly pine (*Pinus taeda*) plantation (35.9°N, 79.08°W). The experiment began in August 1996 and consisted of three plots with ambient and three plots with elevated (+200 ppm) CO$_2$ concentrations, paired according to soil N availability. The Oak Ridge FACE experiment ([Norby et al., 2002]) was set up in a sweetgum (*Liquidambar styraciflua L.*) plantation (35.9°N, 84.33°W) and begun in April 1998. It included five experimental plots, of which three were at ambient and two at elevated CO$_2$ (average of 547 ppm) concentrations. For both sites we use annual measurements of leaf N content, leaf biomass and net primary productivity for model evaluation ([Finzi et al., 2007; Walker et al., 2014; Zaehle et al., 2014]). At both sites, the NPP is derived from direct and indirect measurements of leaf, fine root and wood growth.

The chronic nitrogen addition experiment ([Magill et al., 2004]) located...
at Harvard Forest (42.5°N, 72.16°W) is part of the Harvard Forest Long-term Ecological Research (LTER) site. The experiment consists of four plots each in a red pine (\textit{(Pinus resinosa)} and a hardwood stand (dominated by black and red oak, \textit{Quercus velutina} Lam.; \textit{Q. rubra} L.)). Of the four plots in each stand, we used the control (no N addition), as well as the low N addition (5 gN m\(^{-2}\) yr\(^{-1}\)), and high N addition (15 gN m\(^{-2}\) yr\(^{-1}\)) treatments, excluding the nitrogen and sulphur treatment as this was shown to not be significantly different from the low N treatment and is also outside the scope of this paper. Nutrient additions started in spring of 1988 and were made in equal doses at 4-week intervals throughout the growing season, with the last application taking place in 2004. For model evaluation, we use woody biomass increment from Magill \textit{et al.} (2004) and annual leaf N content from the Harvard Forest Data Archive (Frey S, 2018). For the detailed analysis we show only results from the hardwood stand, as observations for the pine stand shows a decrease in biomass with N addition, as this is a result of processes not relevant to the current analysis.

In addition to this detailed analysis, we also run the model at 16 additional N fertilisation sites located in Northern Europe and North America, including evergreen needleleaf and broadleaf deciduous forests. A number of these sites include plots with different levels of N addition, resulting in 23 separate experimental plots. The rate of N addition varies widely (1.7 - 15 gN m\(^{-2}\) year\(^{-1}\)) as does the duration of the experiment (1 - 40 years).
We extract all biomass and leaf N response data from the respective experiment papers where available. See Table S2 for details for each site. For the purpose of this analysis, we exclude sites with a pronounced decrease in biomass under fertilisation, namely Asa, Norrilden N3 and Harvard pine.

2.4 Model protocol and input data

The QUINCY model requires as input half-hourly meteorological drivers (short- and longwave radiation, air temperature, precipitation (as rain and snowfall), air pressure, humidity and wind velocity), as well as atmospheric CO$_2$ concentration and nitrogen and phosphorus deposition rates. Additional model inputs are vegetation type, and soil physical and chemical parameters (texture, bulk density, rooting and soil depth, as well as inorganic soil P content). Although the model has the option to include fully coupled N and P cycles, for the purpose of this paper we have used the CN-only model, where soil solute inorganic P availability is kept at a constant, non-limiting level.

The daily meteorological data for 1901 to 2015 was extracted from the CRUNCEP dataset, version 7, ([Viovy] 2016), and disaggregated to the half-hourly model timestep using the statistical weather generator described in ([Zaehle & Friend] 2010). The annually changing CO$_2$ atmospheric concentration was taken from [Le Quéré et al.] (2018), and the time series of N deposition for each site from [Lamarque et al.] (2010) and [Lamarque et al.].
In addition, for the two FACE sites, the local meteorological data as well as the CO$_2$ concentrations for the duration of the experiments were used (Walker et al., 2014).

The soil and vegetation biogeochemical pools are brought to quasi-equilibrium through a 500 year model spinup, using forcing data from the period 1901-1930. We then run the model with transient climate and CO$_2$ concentrations for the years 1901-2015 for all sites and treatments. Each site is harvested according to the harvest date available in the literature. After harvest, all biomass is retained in the system as litter, with the exception of the harvested fraction of the woody biomass (set to 80 %), which is removed. A side effect of harvesting is that as the four model variants have different growth rates, the sites can be at slightly different stages of succession at the start of each experiment.

For the N addition experiments, we introduce the additional N to the system to either or both the NH$_4$ or NO$_3$ soil inorganic pool depending on the chemical form of the fertiliser, in the quantity and with the timing described for each respective experiment.

As well as the above fully transient runs, we run the model to equilibrium for a range of soil N availabilities, to explore the behaviour of all model variants at equilibrium without the added behaviour of transient runs or experimental additions. For this purpose, we perform model runs using the meteorology and initial conditions of the control Harvard hardwood site.
We run the model for 500 years with a fixed atmospheric CO₂ concentration for a mature forest. We vary the available soil N level by varying the rate of biological N fixation (BNF). BNF in QUINCY is represented as an asymbiotic processes with a fixed, temperature-dependent rate given a soil mineral N threshold, so that effectively varying the rate of BNF results in varying levels of soil mineral N without affecting other model processes. We perform two sets of runs, with the CO₂ concentrations set to 380 ppm and 550 ppm respectively.

2.5 Model parameterisation

We use the QUINCY model with its default parameterisation, without calibrating the parameters to any of the sites used in this study. Table 1 shows the parameters that differ between each of the four model variants and their values for the two PFTS used here, broadleaf deciduous forest and needleleaf evergreen forest. Table S1 lists other model parameters relevant to the four leaf N variants. For a full list of parameters see Thum et al. (2019).

Both the optimal C export and optimal growth introduce two new, PFT-independent parameters, in addition to those present in the standard QUINCY model, δ_N and τ_N. In comparison, the empirical model requires two PFT-specific parameters for leaf CN ratio bounds, two empirical parameters that drive the shape of the curve and the two parameters it has
in common with the optimal variants (Table 1).

To test the model stability to variations in parameters, we perform a
parameter sensitivity analysis, detailed in Section S1 of the Supplementary
material 1.

3 Results

3.1 Model predictions at equilibrium

Figure 3 shows model NPP and canopy average foliar N content at equilib-
rium under a gradient of soil N availability for all four model variants, for
ambient CO$_2$ (380 ppm, (a) and (b)) and the relative response under ele-
vated CO$_2$ (550 ppm, (c) and (d)). This explores the theoretical response
of the model at equilibrium, without transient climate or CO$_2$ concentra-
tions. This provides a prediction of a similar type to what previous optimal
models have included, but derived from a fully prognostic carbon-nitrogen
cycle model. It is also a clearer way of explaining the model runs at ex-
perimental sites below and testing the theoretical assumptions behind each
model variant (Fig. 2).

Both the empirical and optimal growth model options predict an in-
crease in leaf N content with increasing N availability, as expected, while
the optimal C export model predicts a slight decrease, as it mainly re-
ponds to variations in LAI rather than plant N status. Generally, under
lower N availability, the model predicts a lower LAI, meaning that there are no deeper, more shaded, canopy layers which would have higher respiration values compared to photosynthesis, thereby shifting the NPP response curve (Fig. S3 (a) and (b)) and increasing the optimal leaf N concentration. As there is a slight increase in LAI predicted by all model variants with an increase in soil N (Fig. 3(c)), there is a resulting slight increase in leaf N predicted by the optimal C export variant with increased N availability. This kind of response is also observed under increased CO₂. At higher soil N values, when plants are not N-limited, the optimal C export and optimal growth versions produce similar predictions, as is expected from the assumptions of these two variants (Fig. 2 Eq. 8). All models with flexible leaf N show a higher NPP than the fixed variant at high soil N, however the empirical and optimal C export versions show lower NPP at low N availability. The optimal C export variant shows a leaf N response that does not match observations or our process understanding, namely the decrease in leaf N content with increased N availability (Fig. 3(a)), and therefore a lower overall growth (Fig. 3(b)), demonstrating that a canopy C export only optimal approach does not produce physiologically realistic predictions. The optimal growth variant results in the highest NPP for most of the soil N range, as expected from the optimal criteria that maximises growth. At high soil N however, the optimal C export variant predicts a slightly higher NPP, as its higher N demand caused by the high
leaf N content, can be met by the available soil N.

In terms of predictions under elevated CO$_2$ (Fig. 3 (d) - (f)), both the empirical and optimal growth versions show a decrease in leaf N, strongest at low N availability, while the optimal C export shows only a very small change. The optimal C export model also shows an overall less pronounced response to elevated CO$_2$ than the empirical and optimal growth versions.

### 3.2 Model response to elevated CO$_2$

We show timeseries from fully transient model runs at the two FACE sites, including observed and predicted NPP and leaf N content (Fig. 4) as well as whole ecosystem responses as a mean of the first three years and the last three years of each experiment (Fig. 5). In terms of leaf N content, all model variants are able to predict the observed ambient values reasonably at both FACE sites, although the empirical option shows slightly higher values for Duke (Fig. 4 (a) and (b)). For the ORNL site, the optimal C export model overestimates observed values.

Both the empirical and optimal growth model variants can correctly predict the direction of change in leaf N throughout the experiment under elevated CO$_2$ for both sites (Fig. 4 (c) and (d)), while the optimal C export fails to show a significant change in leaf N concentrations under elevated CO$_2$ (-1.3 % and 1.4 % for Duke and ORNL, respectively at the end of each experiment, Fig. 5 (e) and (d)). For the Duke FACE both
The empirical and optimal growth variants predict a decrease in leaf N; the observations also show a decrease (Fig. 4(c)), although not so pronounced and increasing slightly at the end of the experiment. While both model variants overestimate the magnitude of the change, the optimal growth does so to a lesser degree (-24.1% empirical and -14.3% optimal growth, Fig. 5(c)). In the case of the ORNL site, the optimal growth variant gives the prediction closest to observations (observed -13.6%, empirical -6.1%, optimal growth -15.8%, Fig. 5(d)).

All model variants predict similar NPP at ambient CO₂, generally underestimating observed values at both sites (Fig. 4(e) and (f)). The optimal C export variant predicts an even lower NPP at ORNL, caused by the predicted high leaf N value, which leads to a higher growth demand for N and therefore a lower resulting growth.

Observations at both the Duke and ORNL sites show a positive NPP response to elevated CO₂ throughout the experiment (Fig. 4(g) and (h)), although for ORNL this response decreases significantly towards the end of the experiment. The first notable feature is that all model variants are capable of predicting this positive response at the start of the experiment (Fig. 5(a) and (b)), although this is overestimated for all variants in the case of Duke. However, by the end of the experiment (Fig. 5(c) and (d)), only the empirical and optimal growth are able to predict the sustained positive response for the Duke site (observed 30.6%, empirical 19.5%,
optimal growth 17.4 %). All models predict a lower NPP response than
expected for the entire duration of the ORNL experiment (observed 12.0
%, empirical 4.5 %, optimal growth 6.3 % at the end of the experiment,
Fig. 5(d)).

All model variants predict a lower response in total canopy C at both
sites (Fig. 5(c) and (d)), indicating a missing shift in biomass allocation.
While there is an observed positive response in N uptake at both sites,
all model variants predict a lower response, including a decrease in uptake
in the case of the empirical variant at Duke and the optimal growth at
ORNL at the end of the experiment (Fig. 5(c) and (d)). Overall, we find
that the optimal C export variant does not reproduce observed responses
to elevated CO₂ in either leaf N content or NPP, while the empirical and
optimal growth variants both reproduce the direction of response correctly
throughout both experiments, but the optimal growth captures better the
magnitude of the observations.

3.3 Model response to N addition

In the case of the Harvard Forest N addition experiment (Fig. 7 and
timeseries in Fig. 6), the observations indicate an increase in leaf N content
with N addition (low N 3.3 %, high N 21.0 %), as do the empirical variant
(low N 50.9 %, high N 64.0 %) and optimal growth variant (low N 34.3
% and high N 45.4 %), even though they both overestimate this response
(Fig. 7(c) and (d)). Notably, the optimal C export variant does not predict an increase in leaf N (low N -4.4 %, high N -4.3 % at the end of the experiment), as it responds to changes in LAI and not N availability, and the model predicts a low change in LAI and therefore a corresponding low response in leaf N (Fig. 7(c) and (d)). This is expected from the model assumptions (Fig. 2) and discussed for the at-equilibrium simulations. The peak in the leaf N response of the optimal growth variant (Fig. 6) is caused by a decrease in the control leaf N (but not the treatments), which coincides with the 2001-2002 drought; as there is a gap in the data in this period it is difficult to assess whether this reflects a real plant response.

All model variants with flexible leaf N content predict a positive current wood increment (CWI) response both at the start and end of the experiment (Fig. 6(c) and (d)), although they all overestimate the response in the case of low N addition at the end of the experiment (observed 11.8 %, empirical 31.0 %, optimal growth 26.2 %, Fig. 7(c)). However, they show a better fit for the high N addition (observed 29.9 %, empirical 30.9 %, optimal growth 26.0 %, Fig. 7(d)). All variants underestimate the magnitude of observed CWI for the control plot (Fig. 6(d)).

To test the generality of the findings at the Harvard Forest site, we run our model for a selection of forest N fertilisation sites (Fig. 8). The magnitude of the predicted growth response for both the empirical and optimal growth model variants is linked to the average ambient temperature of the
site, with a stronger response at colder sites (Fig. 8(a) and (b)). This is because the soil N availability, and therefore plant N limitation status is strongly dependent on temperature. The temperature dependency of plant response to N addition is present in reality, however this relation between observations and temperature is less evident than in the case of the model. This is caused by other confounding factors that drive local N availability in reality at each site, such as soil type and N deposition rates, factors which are not necessarily present in the model. The empirical variant largely reproduces observed biomass responses for warmer sites but over-estimates responses for the colder sites (Fig. 8(a)). On the other hand, the optimal growth variant has a tendency to underestimate the biomass response, especially for the higher observed responses (Fig. 8(a)). It is worth noting that the site with a very high observed biomass response (127 %) is one of the very northern sites, with very low annual average temperatures. Both variants show a better fit for the biomass response for broadleaved deciduous forests (normalised root mean squared error, NRMSE = 0.49 empirical and 0.55 optimal growth) than for the needleleaved evergreen (NRMSE = 1.88 empirical and 1.82 optimal growth). For the leaf N content observations (Fig. 8(c) and (d)), both model variants perform similarly, with the empirical one tending to predict a higher leaf N response. Unlike the biomass response there is no relation between ambient temperature and leaf N in either the model or the data. The optimal growth variant shows
a better fit for the broadleaved site leaf N response than the empirical, although it is worth noting that the only deciduous site with leaf N measurements is Harvard Forest (discussed in detail above) so it is difficult to generalise this finding. For the evergreen forest sites, both the empirical and optimal growth variants perform similarly (NRMSE of 0.95 and 1.10 respectively). The two data points with very high leaf N response, which cannot be reproduced by either variant, are both the same site with different N addition levels and as McNulty et al. (2005) note, the observed values are unusually high.

A complete uncertainty analysis is beyond the scope of the paper, however, we have included a parameter sensitivity study for two of the sites used in this study, Duke FACE and Harvard hardwood high N, for the empirical and optimal growth variants (Fig. S1 and S2), which shows that model predictions are stable and robust. Model uncertainty in absolute values of leaf N content, NPP and biomass production is similar for both variants. The uncertainty in the response under elevated CO₂ is largely reduced for the optimal growth variant, although for the N addition site both variants show a high uncertainty.
4 Discussion

4.1 Model performance

In the current paper, we test the hypothesis that changes in leaf nitrogen content can be represented in a terrestrial biosphere model using the optimality principle, by assuming that leaf N changes dynamically in order to maximise plant growth, from the perspective of both net canopy C export and whole plant nutrient status. We show that an optimal scheme which focuses solely on the plant C balance can not represent plant responses at the two FACE and one N fertilisation site we used. Additionally, the empirical formulation, while performing better than the optimal C export variant under both elevated CO$_2$ and N fertilisation, overestimates the magnitude of changes in leaf N (Fig. 4 and 6).

All four model variants underestimate total biomass growth, as either NPP for the two FACE sites (Fig. 4) or current wood increment for the Harvard forest site (Fig. 6). The predicted absolute biomass values are strongly dependent on initial conditions and model spinup, specifically on the soil nutrient status. In the current paper we use the default QUINCY soil model and we have not attempted to improve its representation, therefore the present model biases are intrinsic to its structure, but common to all leaf N model variants. The soil initial condition and process representation is also why the model, while representing the direction of change in
leaf N under N addition, largely fails to represent the temporal trend (Fig. 6(b) and (c)).

It is important to note that the model has not been calibrated to any of the sites used in this study. In fact, one of the advantages of the optimality approach is that the property considered optimal, in our case leaf N content, becomes an emergent property of the model. This means that optimal models are more general and portable across sites and ecosystems.

4.2 Implications of the optimality approach

The majority of previous studies implement optimality as an at-equilibrium process, meaning that plants have the capacity to reach the optimum at any given time. This is done either in a model that only predicts steady-state vegetation processes or, when incorporated into a TBM (e.g. Ali et al. 2015), the plant property being optimised is set to its optimum value at each timestep. In the current study, we choose a 'towards equilibrium' approach, in that the optimality criterion only gives the direction of change and the magnitude of the actual change is limited to a given amount, meant to mirror physiological and ecological limitations to plant plasticity (Val ladares et al. 2007). Therefore, this optimality approach can be used to simulate transient plant acclimation under variable environmental conditions at the timescale that a TBM is usually run at, whilst at the same time maintaining biologically realistic change rates of leaf N concentrations.
Another novel aspect of the optimality scheme we present here is the inclusion of a whole-plant optimality criterion, in contrast with existing studies which have focused on canopy C assimilation or export (e.g. Dewar, 1996; Ali et al., 2015; Smith et al., 2019). We show that a carbon only optimality criterion can not represent observed plant responses at the three main experimental sites we used. As shown in Fig. 3, the optimal C export variant varies the leaf N content in response to changes in LAI and environmental conditions that drive variation in photosynthesis, rather than directly changes in N availability. In particular, it predicts an increase or no change in leaf N under elevated CO\(_2\), as the photosynthesis rate per unit N increases, but respiration remains constant, a behaviour we see both in theory (Fig. 2) and in at-equilibrium and transient simulations. This has implications for productivity estimates and therefore for predictions under future conditions, as seen at both FACE sites (Fig. 4) in this study, where the optimal C export variant cannot predict the positive response of NPP over the duration of the experiment as well as at the N addition site (Fig. 3), where it predicts a too high growth response. Our results highlight the importance of including whole-plant responses in order to correctly represent growth, especially when taking into account nutrient limitation.

A previous model inter-comparison at FACE sites (Zaehle et al., 2014) has shown that models that include an empirical variation in leaf N content
tend to overestimate the decrease in leaf N, something which this study also
shows, specifically at the Duke site (Fig. 4(c)). The optimal growth model
also predicts a too strong response in leaf N, both at the Duke FACE and
the Harvard N addition sites but to a lesser extent. From the theoretical,
at-equilibrium results shown in Fig. 3 we can see that the differences in
response to elevated CO₂ between the empirical and the optimal growth
vary with soil N availability so that it is possible that the mismatch be-
tween the observations and the optimal growth scheme is caused by the
wrong initial soil conditions, as is also indicated by the too low predicted
NPP and wood growth at both the FACE and the Harvard sites. It is im-
portant to note that the empirical functional form used here includes upper
and lower bounds to leaf N variation derived from observations, while the
optimal formulation emerges from the processes and interactions already
included in the model. This means that the optimal approach has less em-
pirical, PFT-specific parameters and relies only on our understanding of
plant physiological processes. The optimal growth variant allows for plas-
tic short-term response to environmental conditions, without the need for
a change in PFT and is therefore a method more generally applicable in
space and time.

Beyond the choice of optimal model there is of course also the choice of
empirical formulation. Here, we use the formulation of [Zaehle & Friend
2010], as implemented in QUINCY [Thum et al. 2019]. It aims to con-
strain leaf N values within observed ranges and responds to whole-plant nutrient limitation. Other TBMs use similar response functions, making it a good benchmark for existing representations of leaf N versus optimal representations.

### 4.3 Extending the optimality approach to other model processes

The optimality approach we present here focuses solely on the representation of dynamic leaf N content, while keeping the baseline model structure of QUINCY as described in Thum et al. (2019). Dynamic variation in leaf N content clearly plays a key role in model predictions of ecosystem productivity, as shown here, a number of other key canopy processes do so also, as discussed below. Optimality concepts have been proposed as a way to represent many of these processes and future development of our model can incorporate such representations.

Changes in atmospheric CO₂ have been shown to lead to changes in the photosynthetic apparatus, a process which is often represented mathematically as a shift in the allocation of photosynthetic N between the different components to maximise C assimilation at the leaf level, a representation sometimes referred to as the coordination hypothesis (Medlyn, 1996; Ali et al., 2015; Smith et al., 2019). The inclusion of this hypothesis within
our optimal leaf N model would modify the shape of the NPP response to leaf N content (Fig. 2) thereby changing the optimal leaf N under given conditions, especially for the optimal C export model variant potentially leading to a more pronounced response under elevated CO₂.

Here, we assume that carbon export and plant nutrient status are the only drivers of changes in leaf N. While we can consider this assumption to hold at longer than annual timescales, at seasonal scales two more processes come into play: leaf ageing and N distribution with depth in the canopy. The QUINCY model assumes a constant C:N ratio throughout a leaf’s lifetime, but in reality leaf N decreases with leaf age (Reich et al. 1991, Kitajima et al. 1997). This process is particularly important in evergreen species, and has been shown to lead to changes in ecosystem productivity in tropical forests (Caldararu et al. 2012, Wu et al. 2016). Combining an age-based decline in leaf N with our optimality model would provide further constraints on the flexibility of leaf N values, potentially reducing the too-large response currently predicted.

We represent the distribution of N in the canopy as an exponentially decreasing function with depth (and therefore light level), with parameters based on observations (Zaehle & Friend 2010), which is consistent with numerous observational studies (e.g. Meir et al. 2002, Kull & Niinemets 1998). However, this distribution is time invariant, although we would expect it to vary with changes in the light environment in time and space.
Previous studies (e.g., Dewar et al., 2012) have used the optimality approach to also predict the within-canopy N distribution albeit at steady state and it should therefore be possible in future versions of our model to include this further flexible process.

Leaf N content is known to co-vary with specific leaf area (SLA) (e.g., Wright et al., 2004), a process which we have not included in the current version of QUINCY. While the co-variation is an established fact, the actual causes of variation in SLA are complex, including structural and biochemical drivers (Poorter et al., 2009). Evans & Poorter (2001) present an optimality-based model which explores the trade-offs between N allocation and changes in leaf structure and SLA, which can be a way forward for a flexible and realistic way of representing variations in SLA. A flexible SLA value in QUINCY would affect the response of the C export variant as this depends strongly on LAI (Fig. 3), which will vary strongly with changes in SLA.

Our results highlight a number of key discrepancies in model predictions. All model variants fail to represent the observed increase in both nitrogen uptake and leaf biomass at the two FACE sites (Fig. 5), pointing to a need for dynamic biomass allocation. Plant flexible biomass allocation in response to nutrient and water availability is a well documented process (Poorter & Nagel, 2000, Hermans et al., 2006) and one that has previously been shown to impact models’ predictive capability under ele-
vated CO$_2$ (De Kauwe et al., 2014). In the current study, for clarity and to avoid complex model feedbacks, we have chosen to keep biomass allocation as a purely allometric process, independent of plant nutrient demand. The current model version has the option to include an empirical dynamic biomass allocation, following Zaehle & Friend (2010), but more process-based ways of representing this are also available including a scheme for optimal allocation (Mäkelä et al., 2008; Franklin, 2007). Such approaches would represent an increase in belowground allocation in response to nutrient and water limitation. A representation of dynamic allocation would lead to more N being available to plants, through an increase in overall N uptake, which would lead to a less pronounced decrease in leaf N as predicted by both the empirical and optimal growth variants, specifically at the Duke FACE site, and therefore a more accurate prediction. Similarly, under N addition, a flexible allocation scheme would reduce the fraction of biomass allocated to roots and therefore reduce N uptake, leading to a less pronounced increase in leaf N content, which is too high for both the empirical and optimal growth variants. This in turn would lead to lower C assimilation rates and therefore a reduction in the growth response, which is currently too high for all model variants.
4.4 Concluding remarks

In conclusion, we show that a whole plant optimality approach incorporated into a TBM can reproduce observed NPP and leaf N content responses for both elevated CO$_2$ and N addition experiments and that an optimal approach which considers carbon export only cannot reproduce these responses. While both an empirical and a whole-plant optimality approach capture observations, with their own particular biases, we argue for the use of the optimality approach as being more rooted in physiological and ecological concepts. Our study shows how optimality concepts can be implemented in terrestrial biosphere models, making use of existing variables and parameters, to reproduce plant plastic responses in a biologically realistic manner.

Acknowledgements

This work was supported by the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (QUINCY; grant no. 647204). LY was supported by the framework of Priority Program SPP 1685 “Ecosystem Nutrition: Forest Strategies for Limited Phosphorus Resources” of the German Research Foundation (DFG), grant No.ZA 763/2-1. We are grateful to our scientific programmer, Dr. Jan Engel, for technical assistance in developing the code.
Author contributions

SC and SZ designed the study and performed the analyses. SC, TT, LY and SZ developed the model. All authors contributed to writing the manuscript.

Data availability

The FACE experiment data is freely available through the FACE Data Management System [https://facedata.ornl.gov/](https://facedata.ornl.gov/) The Harvard Forest chronic nitrogen addition experiment foliar N data is freely available from the Harvard Forest Data Archive [http://harvardforest.fas.harvard.edu:8080/exist/apps/datasets/showData.html?id=hf008](http://harvardforest.fas.harvard.edu:8080/exist/apps/datasets/showData.html?id=hf008). Compiled literature data on biomass and leaf N responses at N fertilisation experimental sites can be found as Supplementary material 2.

Code availability

The scientific part of the code is available under a GPL v3 licence. The scientific code of QUINCY relies on software infrastructure from the MPI-ESM environment, which is subject to the MPI-M-Software-License-Agreement in its most recent form [http://www.mpimet.mpg.de/en/science/models/license](http://www.mpimet.mpg.de/en/science/models/license). The source code is available online [https://git.bgc-jena.mpg.de/quincy/quincy-model-releases](https://git.bgc-jena.mpg.de/quincy/quincy-model-releases), but its access is restricted to registered users. Readers interested in running the model should request a username and password from the corresponding authors or via the git-
repository. Model users are strongly encouraged to follow the fair-use policy stated on [https://www.bgc-jena.mpg.de/bgi/index.php/Projects/QUINCYModel](https://www.bgc-jena.mpg.de/bgi/index.php/Projects/QUINCYModel).

References

Ainsworth EA, Long SP, 2005. What have we learned from 15 years of free-air CO2 enrichment (FACE)? a meta-analytic review of the responses of photosynthesis, canopy properties and plant production to rising CO2. The New phytologist 165(2): 351–371. doi:10.1111/j.1469-8137.2004.01224.x.

Ali AA, et al., 2015. A global scale mechanistic model of the photosynthetic capacity. Geoscientific Model Development Discussions 8: 6217–6266.

Atkin OK, Meir P, Turnbull MH, 2014. Improving representation of leaf respiration in large-scale predictive climate-vegetation models. The New phytologist 202: 743–748. doi:10.1111/nph.12686.

Bloom AJ, Chapin FS, Mooney HA, 1985. Resource limitation in Plants-An economic analogy. Annual review of ecology and systematics 16(1): 363–392. doi:10.1146/annurev.es.16.110185.002051.

Caldararu S, Palmer PI, Purves DW, 2012. Inferring Amazon leaf demography from satellite observations of leaf area index. Biogeosciences
Caldararu S, Purves DW, Palmer PI, 2014. Phenology as a strategy for carbon optimality: a global model. *Biogeosciences* **11**(3): 763–778. doi:10.5194/bg-11-763-2014.

Chapin FS, Vitousek PM, Van Cleve K, 1986. The nature of nutrient limitation in plant communities. *The American naturalist* **127**(1): 48–58. doi:10.1086/284466.

Cowan IR, Farquhar GD, 1977. Stomatal function in relation to leaf metabolism and environment. In *Symposia of the Society for Experimental Biology*, 471–505.

De Kauwe MG, et al., 2014. Where does the carbon go? a model-data intercomparison of vegetation carbon allocation and turnover processes at two temperate forest free-air CO2 enrichment sites. *The New phytologist* **203**(3): 883–899. doi:10.1111/nph.12847.

Dewar RC, 1996. The correlation between plant growth and intercepted radiation: An interpretation in terms of optimal plant nitrogen content. *Annals of botany* **78**(1): 125–136. doi:10.1006/anbo.1996.0104.

Dewar RC, Franklin O, Makela A, McMurtrie RE, Valentine HT, 2009. Optimal function explains forest responses to global change. *Biog-
Dewar RC, Tarvainen L, Parker K, Wallin G, McMurtrie RE, 2012. Why does leaf nitrogen decline within tree canopies less rapidly than light? An explanation from optimization subject to a lower bound on leaf mass per area. *Tree physiology* 32(5): 520–534. doi: 10.1093/treephys/tps044.

Drake BG, Gonzalez-Meler MA, Long SP, 1997. More efficient plants: A consequence of rising atmospheric CO2? *Annual review of plant physiology and plant molecular biology* 48: 609–639. doi: 10.1146/annurev.arplant.48.1.609.

Ellsworth DS, Reich PB, Naumburg ES, Koch GW, Kubiske ME, Smith SD, 2004. Photosynthesis, carboxylation and leaf nitrogen responses of 16 species to elevated pCO2 across four free-air CO2 enrichment experiments in forest, grassland and desert. *Global change biology* 10(12): 2121–2138. doi:10.1111/j.1365-2486.2004.00867.x.

Evans JR, 1989. Photosynthesis and nitrogen relationships in leaves of C3 plants. *Oecologia* 78(1): 9–19. doi:10.1007/BF00377192.

Evans JR, Poorter H, 2001. Photosynthetic acclimation of plants to growth irradiance: the relative importance of specific leaf area and nitrogen partitioning in maximizing carbon gain. *Plant, cell & environment* 24(8): 755–767. doi:10.1046/j.1365-3040.2001.00724.x.
Feng Z, Rütting T, Pleijel H, Wallin G, Reich PB, Kammann CI, Newton PCD, Kobayashi K, Luo Y, Uddling J, 2015. Constraints to nitrogen acquisition of terrestrial plants under elevated CO2. *Global change biology* 21(8): 3152–3168. doi:10.1111/gcb.12938.

Finzi AC, *et al.*, 2007. Increases in nitrogen uptake rather than nitrogen-use efficiency support higher rates of temperate forest productivity under elevated CO2. *Proceedings of the National Academy of Sciences of the United States of America* 104(35): 14014–14019. doi:10.1073/pnas.0706518104.

Fleischer K, *et al.*, 2013. The contribution of nitrogen deposition to the photosynthetic capacity of forests. *Global biogeochemical cycles* 27(1): 187–199. doi:10.1002/gbc.20026.

Franklin O, 2007. Optimal nitrogen allocation controls tree responses to elevated CO2. *The New phytologist* 174(4): 811–822. doi:10.1111/j.1469-8137.2007.02063.x.

Frey S OS, 2018. Chronic nitrogen amendment experiment at harvard forest since 1988. harvard forest data archive: HF008. doi:10.6073/pasta/91b1f285a44d385cc84242b52653396d.

Friend AD, 2010. Terrestrial plant production and climate change. *Journal of experimental botany* 61(5): 1293–1309. doi:10.1093/jxb/erq019.
Friend AD, Stevens AK, Knox RG, Cannell MGR, 1997. A process-based, terrestrial biosphere model of ecosystem dynamics (hybrid v3.0). *Ecological modelling* **95(2-3)**: 249–287.

Givnish T, 1988. Adaptation to sun and shade - a whole-plant perspective. *Australian journal of plant physiology* **15(1-2)**: 63–92.

Haverd V, Smith B, Nieradzik L, Briggs PR, Woodgate W, Trudinger CM, Canadell JG, Cuntz M, 2018. A new version of the CABLE land surface model (subversion revision r4601) incorporating land use and land cover change, woody vegetation demography, and a novel optimisation-based approach to plant coordination of photosynthesis. *Geoscientific Model Development* **11(7)**: 2995–3026. doi:10.5194/gmd-11-2995-2018.

Hawkesford M, Horst W, Kichey T, Lambers H, Schjoerring J, Mueller IS, White P, 2012. Chapter 6 - functions of macronutrients. In Marschner P, editor, *Marschner’s Mineral Nutrition of Higher Plants (Third Edition)*, 135–189. Academic Press, San Diego, third edition edition. doi:10.1016/B978-0-12-384905-2.00006-6.

Hermans C, Hammond JP, White PJ, Verbruggen N, 2006. How do plants respond to nutrient shortage by biomass allocation? *Trends in plant science* **11(12)**: 610–617. doi:10.1016/j.tplants.2006.10.007.
Houle D, Moore JD, 2008. Soil solution, foliar concentrations and tree
growth response to 3-year of ammonium-nitrate addition in two boreal
forests of québec, canada. Forest ecology and management 255(7): 2049–
2060. doi:10.1016/j.foreco.2007.10.056.

Jonard M, et al., 2015. Tree mineral nutrition is deteriorating in europe.
Global change biology 21(1): 418–430. doi:10.1111/gcb.12657.

Kattge J, Diaz S, Lavorel S, Prentice IC, others, 2011. TRY–a global
database of plant traits. Global change biology.

Kitajima K, Mulkey SS, Wright SJ, 1997. Decline of photosynthetic
capacity with leaf age in relation to leaf longevities for five tropical
canopy tree species. American journal of botany 84(5): 702–708.

Kull O, Kruijt B, 1998. Leaf photosynthetic light response: a mechanis-
tic model for scaling photosynthesis to leaves and canopies. Functional
ecology 12(5): 767–777. doi:10.1046/j.1365-2435.1998.00257.x.

Kull O, Niinemets U, 1998. Distribution of leaf photosynthetic proper-
ties in tree canopies: comparison of species with different shade tolerance.
Functional ecology 12(3): 472–479. doi:10.1046/j.1365-2435.1998.00214.

Lamarque JF, Kyle GP, Meinshausen M, Riahi K, Smith SJ, van
Vuuren DP, Conley AJ, Vitt F, 2011. Global and regional evolution
of short-lived radiatively-active gases and aerosols in the representative concentration pathways. *Climatic change* **109**(1): 191. doi:10.1007/s10584-011-0155-0.

**Lamarque JF, et al., 2010.** Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. *Atmospheric Chemistry and Physics* **10**(15): 7017–7039. doi:10.5194/acp-10-7017-2010.

**Le Quéré C, et al., 2018.** Global carbon budget 2017. *Earth System Science Data* **10**(1): 405–448. doi:10.5194/essd-10-405-2018.

**Long SP, Ainsworth EA, Rogers A, Ort DR, 2004.** Rising atmospheric carbon dioxide: plants FACE the future. *Annual review of plant biology* **55**: 591–628. doi:10.1146/annurev.arplant.55.031903.141610.

**Magill AH, Aber JD, Currie WS, Nadelhoffer KJ, Martin ME, McDowell WH, Melillo JM, Steudler P, 2004.** Ecosystem response to 15 years of chronic nitrogen additions at the harvard forest LTER, massachusetts, USA. *Forest ecology and management* **196**(1): 7–28. doi:10.1016/j.foreco.2004.03.033.

**Maire V, Martre P, Kattge J, Gastal F, Esser G, Fontaine S, Soussana JF, 2012.** The coordination of leaf photosynthesis links
C and N fluxes in C3 plant species. *PloS one* **7**(6): e38345. doi: 10.1371/journal.pone.0038345.

Mäkelä A, Givnish TJ, Berninger F, Buckley TN, Farquhar GD, Hari P, 2002. Challenges and opportunities of the optimality approach in plant ecology. *Silva Fennica* **36**(3): 605–614.

Mäkelä A, Valentine HT, Helmisaari HS, 2008. Optimal co-allocation of carbon and nitrogen in a forest stand at steady state. *The New phytologist* **180**(1): 114–123. doi:10.1111/j.1469-8137.2008.02558.x.

McCarthy HR, Oren R, Finzi AC, Ellsworth DS, Kim HS, Johnsen KH, Millar B, 2007. Temporal dynamics and spatial variability in the enhancement of canopy leaf area under elevated atmospheric CO2. *Global change biology* **13**(12): 2479–2497.

McLauchlan KK, Ferguson CJ, Wilson IE, Ocheltree TW, Craine JM, 2010. Thirteen decades of foliar isotopes indicate declining nitrogen availability in central north american grasslands. *The New phytologist* **187**(4): 1135–1145. doi:10.1111/j.1469-8137.2010.03322.x.

McMurtrie RE, Norby RJ, Medlyn BE, Dewar RC, Pepper DA, Reich PB, Barton CVM, 2008. Why is plant-growth response to elevated CO2 amplified when water is limiting, but reduced when nitrogen
is limiting? a growth-optimisation hypothesis. *Functional plant biology*: FPB 35(6): 521–534. doi:10.1071/FP08128.

McNeil BE, Read JM, Driscoll CT, 2007. Foliar nitrogen responses to elevated atmospheric nitrogen deposition in nine temperate forest canopy species. Environmental science & technology 41(15): 5191–5197.

McNulty SG, Boggs J, Aber JD, Rustad L, Magill A, 2005. Red spruce ecosystem level changes following 14 years of chronic N fertilization. Forest ecology and management 219(2): 279–291. doi: 10.1016/j.foreco.2005.09.004.

Medlyn BE, 1996. The optimal allocation of nitrogen within the C3 photosynthetic system at elevated CO2. *Functional plant biology*: FPB 23(5): 593–603. doi:10.1071/pp9960593.

Medlyn BE, Duursma RA, Eamus D, Ellsworth DS, Prentice IC, Barton CVM, Crous KY, De Angelis P, Freeman M, Wingate L, 2011. Reconciling the optimal and empirical approaches to modelling stomatal conductance. Global change biology 17(6): 2134–2144.

Medlyn BE, et al., 2015. Using ecosystem experiments to improve vegetation models. Nature climate change 5: 528. doi:10.1038/nclimate2621.

Meir P, Kruijt B, Broadmeadow M, Barbosa E, Kull O, Carswell F, Nobre A, Jarvis PG, 2002. Acclimation of photosynthetic capacity
to irradiance in tree canopies in relation to leaf nitrogen concentration and leaf mass per unit area. *Plant, cell & environment* **25**(3): 343–357. doi:10.1046/j.0016-8025.2001.00811.x.

Meyerholt J, Zaehle S, 2015. The role of stoichiometric flexibility in modelling forest ecosystem responses to nitrogen fertilization. *The New phytologist* **208**(4): 1042–1055. doi:10.1111/nph.13547.

Norby RJ, et al., 2002. Net primary productivity of a CO$_2$-Enriched deciduous forest and the implications for carbon storage. *Ecological applications: a publication of the Ecological Society of America* **12**(5): 1261–1266. doi:10.2307/3099969.

Poorter H, Nagel O, 2000. The role of biomass allocation in the growth response of plants to different levels of light, CO2, nutrients and water: a quantitative review. *Functional plant biology: FPB* **27**(12): 1191–1191. doi:10.1071/pp99173\_co.

Poorter H, Niinemets Ü, Poorter L, Wright IJ, others, 2009. Causes and consequences of variation in leaf mass per area (LMA): a meta-analysis. *New* .

Prentice IC, Liang X, Medlyn BE, Wang YP, 2015. Reliable, robust and realistic: the three r’s of next-generation land-surface modelling.
Atmospheric Chemistry and Physics 15(10): 5987–6005. doi:10.5194/acp-15-5987-2015.

Reich PB, Walters MB, Ellsworth DS, 1991. Leaf age and season influence the relationships between leaf nitrogen, leaf mass per area and photosynthesis in maple and oak trees. Plant, cell & environment 14(3): 251–259. doi:10.1111/j.1365-3040.1991.tb01499.x.

Rogers A, Humphries SW, 2000. A mechanistic evaluation of photosynthetic acclimation at elevated CO2. Global change biology 6(8): 1005–1011. doi:10.1046/j.1365-2486.2000.00375.x.

Sikström U, 2002. Effects of liming and fertilization (n, PK) on stem growth, crown transparency, and needle element concentrations of picea abies stands in southwestern sweden. Canadian journal of forest research. Journal canadien de la recherche forestiere 32(10): 1717–1727. doi: 10.1139/x02-094.

Smith NG, et al., 2019. Global photosynthetic capacity is optimized to the environment. Ecology letters 22(3): 506–517. doi:10.1111/ele.13210.

Stitt M, Krapp A, 1999. The interaction between elevated carbon dioxide and nitrogen nutrition: the physiological and molecular background. Plant, cell & environment.
Thomas RQ, Bonan GB, Goodale CL, 2013a. Insights into mechanisms governing forest carbon response to nitrogen deposition: a model–data comparison using observed responses to nitrogen addition. Biogeo-}

sciences.

Thomas RQ, Zaehle S, Templer PH, Goodale CL, 2013b. Global patterns of nitrogen limitation: confronting two global biogeochemical models with observations. Global change biology 19(10): 2986–2998. doi:10.1111/gcb.12281.

Thum T, Caldararu S, Engel J, Kern M, Pallandt M, Schnur R, Yu L, Zaehle S, 2019. A new terrestrial biosphere model with coupled carbon, nitrogen, and phosphorus cycles (QUINCY v1.0; revision 1772). Geoscientific Model Development Discussions 2019: 1–38. doi:10.5194/gmd-2019-49.

Valladares F, Gianoli E, Gómez JM, 2007. Ecological limits to plant phenotypic plasticity. The New phytologist 176(4): 749–763. doi:10.1111/j.1469-8137.2007.02275.x.

Viovy N, 2016. CRUNCEP data set.

Walker AP, et al., 2014. Comprehensive ecosystem model-data synthesis using multiple data sets at two temperate forest free-air CO 2 enrichment experiments: Model performance at ambient CO 2 concentration:
FACE MODEL-DATA SYNTHESIS. *Journal of Geophysical Research: Biogeosciences* **119**(5): 937–964. doi:10.1002/2013JG002553.

**Wright IJ, et al., 2004.** The worldwide leaf economics spectrum. *Nature* **428**(6985): 821–827. doi:10.1038/nature02403.

**Wu J, et al., 2016.** Leaf development and demography explain photosynthetic seasonality in amazon evergreen forests. *Science* **351**(6276): 972–976. doi:10.1126/science.aad5068.

**Wujeska-Klause A, Crous KY, Ghannoum O, Ellsworth DS, 2019.** Lower photorespiration in elevated CO2 reduces leaf N concentrations in mature eucalyptus trees in the field. *Global change biology* doi:10.1111/gcb.14555.

**Zaehle S, Dalmorech D, 2011.** Carbon–nitrogen interactions on land at global scales: current understanding in modelling climate biosphere feedbacks. *Current Opinion in Environmental Sustainability* **3**(5): 311–320. doi:10.1016/j.cosust.2011.08.008.

**Zaehle S, Friend AD, 2010.** Carbon and nitrogen cycle dynamics in the O-CN land surface model: 1. model description, site-scale evaluation, and sensitivity to parameter estimates. *Global biogeochemical cycles* **24**(1): Gb1005. doi:10.1029/2009GB003521.
Zaehle S, et al., 2014. Evaluation of 11 terrestrial carbon–nitrogen cycle models against observations from two temperate Free-Air CO2 enrichment studies. *The New phytologist* 202(3): 803–822. doi: 10.1111/nph.12697.
Figure 1: Schematic representation of the QUINCY baseline model structure. Ellipses: biogeochemical pools and other state variables; rectangles: biogeochemical processes; tetraeders: model input; solid green lines: carbon fluxes; solid dark red lines: nitrogen and phosphorus fluxes; solid black lines: carbon, nitrogen and phosphorus fluxes; dotted blue lines: effects (Thum et al., 2019).
Figure 2: Theoretical representation of optimal leaf N. Black lines show the variation in net C assimilation with leaf N and blue lines show the C-equivalent plant growth given an amount of N available in the soil. Both  $A_n$ and $NPP$ represent whole-canopy values. $A_n$, net assimilation refers to gross assimilation minus canopy maintenance respiration, while $NPP$ refers to whole-plant net primary productivity available for growth. Leaf N values are canopy averages. Triangles show the leaf N for which maximum NPP occurs, equivalent to the optimal C export variant. Black circles show leaf N for which C uptake is equal to N uptake, equivalent to the optimal growth variant.
Figure 3: At-equilibrium whole-canopy leaf N content (a), NPP (b) and maximum annual LAI (c) over a range of available soil mineral N, as well as the response to elevated CO$_2$ (550 ppm) relative to the predictions at 380 ppm ((d), (e) and (f)) for all model variants. Leaf N values are canopy averages. Leaf N values predicted by the optimal C export variant show a very strong positive response to elevated CO$_2$ at very low availability and have been omitted for plot clarity. Soil N values refer to total mineral N to a depth of 1 m. All simulation are done using the meteorology and initial conditions of the control Harvard hardwood site, a temperate broadleaf deciduous forest.
Figure 4: Model predictions at the Duke (left) and ORNL (right) FACE sites for absolute leaf N concentration ((a) and (b)), relative response of leaf N under elevated CO$_2$ ((c) and (d)), absolute NPP values ((e) and (f)) and relative response of NPP ((g) and (h)).
Figure 5: Ecosystem response to elevated CO$_2$ at the Duke and ORNL FACE sites for the first 3 years ((a) and (b)) and last three years ((c) and (d)) for all model variants. Variables shown are net primary productivity (NPP), plant N uptake ($fN_{\text{up}}$), nitrogen use efficiency (NUE), leaf N concentration (N$_\%$), maximum annual total canopy C (C$_{\text{leaf}}$). All variables are shown as relative responses %.
Figure 6: Model predictions at the Harvard long term N addition experimental site for absolute leaf N content values ((a)), relative response of leaf N under low ((b)) and high ((c)) N addition levels, as well as absolute current wood increment (CWI) ((d)) and relative response of CWI ((e) and (f)).
Figure 7: Ecosystem response to N addition at the Harvard hardwood site for the first 3 years ((a) and (b)) and last three years ((c) and (d)) for all model variants. Variables shown are current wood increment (CWI), plant N uptake ($fN_{up}$), nitrogen use efficiency (NUE), leaf N concentration ($N_\%$), maximum annual total canopy C ($C_{leaf}$). All variables are shown as relative responses %.
Figure 8: Biomass ((a) and (b)) and leaf N ((c) and (d)) response for a set of forest N addition sites as predicted by the empirical (left) and optimal growth (right) model variants, grouped by PFT (BS - broadleaved seasonal, open symbols, NE - needleleaved evergreen, closed symbols) and average annual temperature (mean annual temperature ≤ 5°C, blue circles and mean annual temperature > 5°C, red triangles).
Table 1: Model parameters introduced for each of the four variants of representing leaf N content. Parameters which are part of the baseline QUINCY model and are relevant for the processes described here can be found in Table S1. BS = broadleaf deciduous, NE = needleleaf evergreen

| Symbol       | Description                                    | Value BS | Value NE | Unit            | Citation                  |
|--------------|------------------------------------------------|----------|----------|-----------------|---------------------------|
| $\chi_{CN}$  | Foliar C:N                                     | 22.5     | 39.7     | $\text{molC/molN}$ | Kattge et al. (2011)      |
| $\lambda_{leaf}$ | Shape parameter in leaf stoichiometry nutrient response | 2.0       | 2.0       | -               | Zachle & Friend (2010)    |
| $k_{leaf}$   | Shape parameter in leaf stoichiometry nutrient response | 8.0       | 8.0       | -               | Zachle & Friend (2010)    |
| $\chi_{CN,min}$ | Minimum foliar C:N                             | 14.0     | 24.0     | $\text{molC/molN}$ | Kattge et al. (2011)      |
| $\chi_{CN,max}$ | Maximum foliar C:N                             | 38.7     | 64.9     | $\text{molC/molN}$ | Kattge et al. (2011)      |
| $\delta_N$   | Maximum rate of change in foliar N content     | 0.0048   | 0.0048   | $\text{day}^{-1}$ | This study                |
| $\tau_N$     | Response timescale for changes in foliar N content | 30        | 30       | days            | This study                |

| Symbol       | Description                                    | Value BS | Value NE | Unit            | Citation                  |
|--------------|------------------------------------------------|----------|----------|-----------------|---------------------------|
| $\delta_N$   | Maximum rate of change in foliar N content     | 0.0048   | 0.0048   | $\text{day}^{-1}$ | This study                |
| $\tau_N$     | Response timescale for changes in foliar N content | 30        | 30       | days            | This study                |