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Carbon Sequestration in Terrestrial Ecosystems Under Elevated CO$_2$ and Temperature:

Role of Dissolved Organic versus Inorganic Nitrogen Loss

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Abstract

We used a simple model of carbon-nitrogen (C-N) interactions in terrestrial ecosystems to examine the responses to elevated CO$_2$ and to elevated CO$_2$ plus warming in ecosystems with the same total nitrogen loss but that differed in the ratio of dissolved organic nitrogen (DON) to dissolved inorganic nitrogen (DIN) loss. We postulate that DIN losses can be curtailed by higher N demand in response to elevated CO$_2$ but that DON losses cannot. We also examined simulations in which DON losses were held constant, were proportional to the amount of soil

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organic matter, were proportional to the soil C:N ratio, or were proportional to the rate of decomposition. We found that the mode of N loss made little difference to the short-term (<60 years) rate of carbon sequestration by the ecosystem, but high DON losses resulted in much lower carbon sequestration in the long term than did low DON losses. In the short term, C sequestration was fueled by an internal redistribution of N from soils to vegetation and by increases in the C:N ratio of soils and vegetation. This sequestration was about three times larger with elevated CO_2 and warming than with elevated CO_2 alone. After year 60, C sequestration is fueled by a net accumulation of N in the ecosystem and the rate of sequestration was about the same with elevated CO_2 and warming as with elevated CO_2 alone. With high DON losses, the ecosystem either sequestered C slowly after year 60 (when DON losses were constant or proportional to soil organic matter) or lost C (when DON losses were proportional to the soil C:N ratio or to decomposition). We conclude that changes in long-term C sequestration depend not only on the magnitude of N losses but on the form of those losses as well.

Introduction

Terrestrial ecosystems are thought to sequester about 25% of the carbon (C) currently emitted through fossil-fuel burning and land-use change (IPCC 2001). It is hoped that these ecosystems will continue to be a major sink for C in the future and thereby mitigate further increases in CO_2 in the atmosphere. However, productivity in terrestrial ecosystems is strongly constrained by the dynamics of the nitrogen (N) cycle (Vitousek et al. 1998) and C sequestration will likely require a net accumulation of N in these ecosystems. The input of N to ecosystems has been widely studied, especially from the perspective of atmospheric N deposition (Galloway et al. 2003, 1995, Ollinger et al. 1993) and an understanding of the controls on biological N_2

fixation is emerging (Cleveland et al. 1999, Rastetter et al. 2001, Vitousek et al. 2002).

However, surprisingly little is known about the form, magnitude, or controls of N losses from

terrestrial ecosystems (Pellerin et al. in press, McDowell 2003, Neff et al. 2003, Aber et al. 2002,

Hedin et al. 1995, Sollins and McCorrison 1981). In this paper we argue that the amount of C

sequestered in terrestrial ecosystems in response to elevated CO_2 depends on the fraction of N

losses that are in the form of dissolved organic N (DON) versus dissolved inorganic N (DIN);

because plants can curtail DIN losses as N demand increases in response to elevated CO_2, but

plants have little control over DON losses, the potential for accumulating N by limiting N losses

should be small if DON losses are high. Thus, the potential for sequestering C in response to

elevated CO_2 should be small if a large fraction of the N losses are as DON.

Modifications to the Standard Model

Our assessment of C sequestration in relation to DON losses relies upon three

modifications to what has been called "the standard model" of N accumulation in terrestrial

ecosystems (Vitousek et al. 1998). First, as suggested by Vitousek et al. (1998) and Neff et al.

(2003), the standard model needs to be modified to include DON losses. Second, the standard

model needs to be modified to accommodate an increase in N demand by both plants and

microbes in response to elevated CO_2 levels. Finally, the dynamics of DIN in the standard model

have to be modified to reflect the fact that N uptake by microorganisms, N uptake by plants, and

N losses from the ecosystems happen simultaneously rather than sequentially. These changes are

discussed in more detail below.

There are also several assumptions we have made to simplify our analysis. The first

relates to the growing evidence that plants can use organic forms of N (Schimel and Bennett

2003, Neff et al. 2003, McKane et al. 2002, Schimel and Chapin 1996, Kieland 1994, Chapin et al. 1993). We will circumvent this complication by lumping plant-available forms of DON into the DIN pool and use "DON" to refer only to unavailable forms. By lumping plant-available forms of DON into the DIN pool, we are also assuming that these forms of DON are available to soil microbes. We will further simplify our analysis by assuming that any additional DON available to microbes is retained in the ecosystems and can therefore be lumped with the soil organic N (Lispon and Monson 1998, Perakis and Hedin 2001). Thus, we assume that the DON lost from ecosystems is in a form that is unavailable to both plants and microbes. We also assume that there is no change in the ratio of NH_4 to NO_3 in soil solution so that the DIN losses can be represented as proportional to the total DIN in soil solution. Finally, we will lump gaseous N losses (e.g., denitrification) in with DIN losses.

DON losses: Until recently, DON losses from terrestrial ecosystems have been largely ignored (Goodale et al. 2000, Campbell et al. 2000) and were not incorporated into the standard model of N accumulation (Vitousek et al. 1998). Estimates that infer total N losses from stream chemistry indicate that DON losses range from less than 20% to greater than 80% of those losses (e.g., Perakis and Hedin 2002, Qualls et al. 2002, Buffam et al. 2001, Goodale et al. 2000, McHale et al. 2000).   Because of retention and processing of DON and DIN in the vadose zone, ground water, riparian areas, and streams (Kroeger 2003, Hedin et al 1998, Newbold et al. 1981, 1982), stream water chemistry probably does not faithfully reflect the chemistry of water leaving the rooting zone of upland areas. For example, Currie et al. (1996) found that DON accounted for over 97% of the N in zero-tension lysimeters at the base of the rooting zone of a previously logged New England forest, whereas Goodale et al. (2000) found that on average DON

accounted for only 67% of the N in steams draining previously logged New England forests. In a southern hardwood forest, Qualls et al. (2002) found N fluxes to be 92% DON in the B horizon, 75% in the C horizon, and 79% in the stream. In addition, none of these studies quantify the fraction of DON that might be available to either plants or microbes. Thus, although DON losses appear to be important, the relative losses of DIN versus DON from upland ecosystems are far from certain (McDowell 2003). Our purpose here is not to resolve this uncertainty but rather to assess the consequences of DIN versus DON losses on the potential for C sequestration in terrestrial ecosystems in response to elevated CO_2 concentrations.

Increased N demand in response to elevated CO_2: The standard model of N accumulation is formulated from the perspective of a single limiting resource (i.e., N) and therefore does not address the effects of other resources, like CO_2, on N dynamics. An alternate perspective is provided by the "functional equilibrium hypothesis" (Farrar and Jones 2000, Chapin et al. 1987, Bloom et al. 1985), which predicts that increased CO_2 concentrations will free plant resources currently allocated toward C acquisition and allow them to be reallocated toward the acquisition of other resources like N. This hypothesis has been corroborated in several studies on tree saplings, in which allocation to fine roots increased in response to elevated CO_2 (e.g., Tingey et al 2000, Janssens et al. 1998, Prior et al. 1997), and has also been observed in intact forest stands, although the response is weaker than in studies on saplings (Pritchard et al 2001, Matamala and Schlesinger 2000). This compensatory reallocation of internal resources should increase N-uptake potential of plants. In addition, elevated CO_2 should increase the flux of C to soils in litter and root exudates and thereby increase microbial N demand (Johnson et al. 2001, Mikan et al. 2000). These responses of plants and microbes to

elevated CO_2 should decrease soil DIN concentrations and therefore decrease DIN losses, resulting in an increase in N accumulation in the ecosystem (until enough N accumulates in the ecosystem to meet the demand through internal N recycling). However, this mechanism for accumulating N requires that N losses from the ecosystem be in a form that is available to plants and microbes and will therefore not work if N is lost as an unavailable form of DON.

Simultaneity of DIN uptake and loss processes: In the standard model of N accumulation in terrestrial ecosystems (Vitousek et al. 1998), inorganic N entering the soil by gross mineralization or deposition is assumed to satisfy microbial needs first (immobilization), then to satisfy plant needs (plant uptake), and only then is N lost from the ecosystem. This heuristic simplification arose for two reasons. First, by assuming microbes have first access to the available N, the model could be formulated directly on net N mineralization, which is readily estimated, and there would be no need to estimate gross mineralization, which is far more difficult (Nadelhoffer et al. 1985, Hart et al. 1994, Bosatta and Ågren 1995). Second, by assuming N losses only occur after plant requirements are met, the model could focus on N-breakthrough dynamics associated with N saturation (Aber et al. 1998). In an ecosystem approaching N saturation, the rate of N inputs as net mineralization plus deposition is higher than plant requirements. Nitrate therefore builds up in the soil, which results in accelerated N losses through denitrification and leaching of the highly mobile nitrate ions.

These simplifications in the standard model could be problematic for analyses of ecosystem response to elevated CO_2 because they do not allow for changes in the competitive interactions between plants and microbes for available N or for decreases in ecosystem N losses in response to increased N demand by plants and microbes. Although soil microbes clearly have

a substantial competitive advantage over plants for acquiring N, that advantage is not so overwhelming as to exclude plant N uptake until microbial N demand is fully satisfied. Nor are plants so effective at acquiring N that they can completely shut off DIN losses from the ecosystem. Thus, as CO_2 increases, plant N demand and C fluxes to the soil should increase, which should result in a shift in the relative rates of plant uptake, microbial immobilization, and ecosystem losses. To account for this shift in relative rates, the three processes have to be modeled as occurring simultaneously, a perspective that has been greatly facilitated by the development of stable isotope techniques (e.g., Perakis and Hedin 2001, Nadelhoffer et al. 1999). Our purpose here is to assess how this shift in the relative rates of plant uptake, microbial immobilization, and ecosystem losses might differ if N losses are as DON versus DIN and how that difference might affect C sequestration in response to elevated CO_2.

Analysis

Our hypothesis is easily conceptualized from the perspective of an ecosystem initially at steady state. Under this steady state, N losses exactly equal N inputs to the ecosystem so there is no net N accumulation. If an increase in CO_2 then results in higher N demand by plants and microbes, DIN losses will decline and N will accumulate in the ecosystem until enough N builds up and is recycled within the ecosystem to meet the higher N demand, at which point a new steady state is reached. However, because we assume that DON losses are in a form that is unavailable to plants and microbes, the rate of N accumulation by this mechanism cannot exceed the initial rate of DIN loss. The actual rate of N accumulation will depend upon how low the DIN concentrations can be decreased in response to increased N demand by plants and microbes. The amount of C sequestered will depend upon how the accumulated N is distributed among

ecosystem components and if there is any change in the C:N ratio of those components. To assess this potential for C sequestration we have implemented DON losses into the Multiple-Element Limitation (MEL) model (Rastetter and Shaver 1992, Rastetter et al. 1997, 2001).

Modifications to the MEL model: The MEL model was designed to examine responses of terrestrial ecosystems to changes in CO_2 and N deposition under the constraints of mass balance on both C and N and the feedbacks associated with N recycling through the soil (Table 1). The C and N cycles are coupled through the acclimation of C and N uptake rates by vegetation to maintain the ratio of C:N in plant tissues (Eqs. 6 & 21 in Table 1) and through the adjustment of microbial N immobilization and C and N assimilation efficiencies to maintain the microbial C:N ratio (Eqs. 24 - 29). The C:N ratio and growth of vegetation are constrained by simple assumptions about allometry and biomass allocation to wood (Eq. 8). The DIN pool is simultaneously available to plants, to microbes, and for losses from the ecosystem (Eqs. 15, 22, & 24).   The model and its parameterization for a northeastern US mixed forest at steady state are fully documented in Rastetter et al. (2001).

For our analysis here, we added equations to simulate both dissolved organic C (DOC) and DON losses. We assume that the DON loss rate (L_{DON}, in g N m^{-2} yr^{-1}) is proportional to the DOC loss rate (L_{DOC}, in g C m^{-2} yr^{-1}); $L_{DON} = L_{DOC}/\theta_{DOM}$, where θ_{DOM} is the C:N ratio of the dissolved organic matter lost from the ecosystem. We further assumed that this C:N ratio is constant with a value of 30 g C g^{-1} N (the average for old growth watersheds reported in Goodale et al 2000). A more dynamic representation of the C:N ratio of dissolved organic matter must wait for better empirical assessment of how those dynamics are regulated.

To explore various controls on DOC losses, we developed four alternate models (in the following equations, β_{DOC} is a constant with units that differ for the four models; respectively: 1) g C m^{-2} yr^{-1}, 2) yr^{-1}, 3) g N m^{-2} yr^{-1}, and 4) g C g^{-1} C).

1) **Constant-loss model:** $L_{DOC} = \beta_{DOC}$. This is the simplest possible model. In it we assume a constant DOC loss rate and therefore no feedbacks from other ecosystem properties.

2) **Proportional-loss model:** $L_{DOC} = \beta_{DOC} D_C$. In this model we assume DOC losses are proportional to the amount of organic matter in the soil (D_C), as was found by Neff et al. (2000) for Hawaiian soils of varying ages. This formulation provides a feedback that increases DOC losses in response to any process that increases the accumulation of soil organic C (e.g., higher litter inputs or slower decomposition rates).

3) **C:N-based model:** $L_{DOC} = \beta_{DOC} \dfrac{D_C}{D_N}$. In this model we assume DOC losses are proportional to the soil C:N ratio (D_C/D_N), as found by Aitkenhead and McDowell (2000) for a global relationship across 15 types of biome. In this formulation, losses of DOC again increase with increasing soil organic C, but decrease with increasing soil organic N. The feedbacks therefore involve the complex interactions among the inputs of C and N in litter, decomposition, and N mineralization and immobilization.

4) **R$_m$-based model:** $L_{DOC} = \beta_{DOC} R_{Cm}$. In this model we assume DOC losses are proportional to microbial respiration (R_{Cm}). Brooks et al. (1999) found a similar relationship, but for over-winter rates of soil respiration. Again DOC losses increase with higher soil organic C stocks, but also increase with elevated temperature.

We present simulations for both high (5:1) and low (1:5) ratios of DON:DIN losses. In the original parameterization with no DON losses (Rastetter et al. 2001), the steady-state DIN losses were 0.6 g N m^{-2} yr^{-1} to match the annual deposition rate. In the new model, the initial, steady-state DON and DIN losses were 0.5 and 0.1 g N m^{-2} yr^{-1}, for the high DON-loss simulations, and 0.1 and 0.5 g N m^{-2} yr^{-1}, for the low DON-loss simulations (Table 2). With a constant dissolved organic matter C:N ratio of 30 g C g^{-1} N, the DOC losses were 15 and 3 g C m^{-2} yr^{-1} in the high and low DON-loss simulations, respectively. To incorporate these additional C and N losses and maintain the assumption of an initial steady state, we had to compensate by adjusting parameters regulating the rates of other fluxes in the model (Table 2). To compensate for the additional loss of C from the soil organic matter as DOC, we decreased microbial respiration by decreasing ψ (Eq. 28), which also decreased gross N mineralization (Eqs. 25 & 29), but not enough to fully compensate for the loss of N as DON. To fully compensate for the DON losses, we increased N immobilization by increasing α_N (Eq. 24). Relative to the original parameterization, the changes in ψ and α_N were about 3% for the high DON-loss simulations and about 1% for the low DON-loss simulations. All other fluxes and all the initial C and N stocks are as reported in Rastetter et al. (2001).

Simulations: We ran four simulations with each of the four DON-loss models. In these simulations, we examined the responses with both high and low DON-losses to an instantaneous doubling of CO_2 concentration or to an instantaneous doubling of CO_2 concentration and a 4°C increase in temperature, which is the predicted temperature change for New England over the next 100 years (IPCC 2001). All simulations began at a steady state and were allowed to run to a new steady state following the change in CO_2 or CO_2 and temperature. By imposing an instantaneous change in CO_2 or CO_2 and temperature, responses that act at different time scales

tend to be segregated as the simulation proceeds, with the responses associated with fast

mechanisms appearing first and those associated with slower mechanisms emerging later. To

assess the effects of a more realistic scenario, we ran 100-year simulations with CO_2 or CO_2 and

temperature increasing linearly over the 100 years. This gradual change tends to superimpose

the fast responses onto those acting on the decade to century time scale.

Partitioning the change in Ecosystem C: To help analyze the results of our

simulations, we partitioned the predicted change in total ecosystem C among four factors relating

to the interactions between C and N (Rastetter et al. 1992): 1) the change in the total amount of N

in the ecosystem ($\Delta\mathbf{C}_{\Delta N_T}$), 2) the change in plant C:N ratio ($\Delta\mathbf{C}_{\Delta C:N_P}$), 3) the change in soil C:N

ratio ($\Delta\mathbf{C}_{\Delta C:N_S}$), and 4) the redistribution of N between plants and soil ($\Delta\mathbf{C}_{B_N \Leftrightarrow D_N}$). Rastetter et

al. (1992) quantified these four factors plus their synergistic interaction ($\Delta\mathbf{C}_{inter}$):

$$\Delta\mathbf{C}_{\Delta N_T} = (\Delta\mathbf{B_N} + \Delta\mathbf{D_N})\frac{\mathbf{B_C} + \mathbf{D_C}}{\mathbf{B_N} + \mathbf{D_N}}$$

$$\Delta\mathbf{C}_{\Delta C:N_P} = \mathbf{B_N}\Delta\left(\frac{\mathbf{B_C}}{\mathbf{B_N}}\right)$$

$$\Delta\mathbf{C}_{\Delta C:N_S} = \mathbf{D_N}\Delta\left(\frac{\mathbf{D_C}}{\mathbf{D_N}}\right)$$

$$\Delta\mathbf{C}_{B_N \Leftrightarrow D_N} = \left(\frac{\mathbf{B_C}}{\mathbf{B_N}} - \frac{\mathbf{D_C}}{\mathbf{D_N}}\right)\left(\frac{(\Delta\mathbf{B_N})\mathbf{D_N} - (\Delta\mathbf{D_N})\mathbf{B_N}}{\mathbf{B_N} + \mathbf{D_N}}\right)$$

$$\Delta\mathbf{C}_{inter} = \Delta\mathbf{B_N}\Delta\left(\frac{\mathbf{B_C}}{\mathbf{B_N}}\right) + \Delta\mathbf{D_N}\Delta\left(\frac{\mathbf{D_C}}{\mathbf{D_N}}\right)$$

where $\boldsymbol{B}_C$ and $\boldsymbol{B}_N$ are the C and N initially in plant biomass, $\boldsymbol{D}_C$ and $\boldsymbol{D}_N$ are the C and N initially

in soil detritus (litter plus soil organic matter), $\Delta\mathbf{B_N}$ and $\Delta\mathbf{D_N}$ are the changes in the plant and

soil N, and $\Delta\left(\dfrac{B_C}{B_N}\right)$ and $\Delta\left(\dfrac{D_C}{D_N}\right)$ are the changes in the plant and soil C:N ratio. The changes in

C associated with these factors are comprehensive and mutually exclusive; thus the total chance

in ecosystem C is the sum of the changes associated with each factors plus their interaction.

To implement these equations, we simply ran the MEL model to predict values for plant

and soil C and N stocks (i.e., B_C, B_N, D_C, and D_N), calculated the changes in the N stocks and

C:N ratios from their initial values, and plugged these values into the equations. In this way, the

cumulative effect of each factor can be assessed at any time during the simulations.

Results and Discussion

Terrestrial ecosystems display a wide range of DON versus DIN losses across ecological

and biophysical gradients, and in response to anthropogenic factors. Our simulations indicate

these N loss variations can have important consequences for ecosystem carbon accumulation

under changing CO_2 and temperature regimes. After 1000 years of simulated exposure to

doubled CO_2 alone and doubled CO_2 with 4^oC warming, we found that ecosystems with high

DON losses accumulated 1.5 to 4 times less carbon than ecosystems with low DON losses (Table

3). Steady-state differences in carbon accumulation were even greater, with greatest effect

observed under elevated CO2 and temperature (Fig. 1).

The divergent patterns of C accumulation observed in our simulations arise from

differences in the accumulation and internal cycling of N, and involve several mechanisms acting

at different time scales. In our earlier analysis of responses to elevated CO_2, but without DON

losses (Rastetter et al. 1997), we identified four distinct mechanisms acting on different time

scales: 1) the nearly instantaneous response of the photosynthetic enzyme system, 2) the 1-10

year, within-plant acclimation to reestablish a balance in the C versus N uptake rates, 3) the 50-

80-year redistribution of N between soils and plants, and 4) the long-term accumulation of N. For the analysis presented here, the first three of these mechanisms can be lumped into a single category of within-ecosystem mechanisms. Our results indicate that these within-ecosystem mechanisms dominate the dynamics during the first 60 years of the simulations and that the slow accumulation of N by the ecosystem dominates the dynamics thereafter.

Responses during the first 60 years: During the first 60 years of the simulations with doubled CO_2 concentration alone, the behaviors of the models are almost identical; there is very little difference in the amount or rate of C sequestration between simulations with high versus low DON losses or among the four DON-loss models (Fig. 1). During this period, differences in N accumulation are too small to have a noticeable effect on C sequestration relative to the within-ecosystem mechanisms for C sequestration. The major mechanism of C sequestration during this early response is a redistribution of N from soils to vegetation (Fig. 2, Table 3). When the CO_2 concentration is increased, plants compete more effectively against soil microbes for available N so there is a net loss of N from soils and a concurrent net N gain by plants (Fig. 1). Because the C:N ratio of soils is about 25 and that of plants is about 143 (initial C:N values), this redistribution of N results in a net increase in the amount of C stored per unit N in the ecosystem. Although there is a net loss of N from the soil during these first 60 years, soil C actually increases because of higher plant productivity and litterfall. The soil C:N ratio therefore increases and also contributes to the amount of C stored per unit N in the ecosystem (Fig. 2). Elevated CO_2 increased plant production and biomass, which in a mature, closed-canopy forest results in a larger increase in woody tissues, with a high C:N ratio, than in leaves and fine roots (i.e., active tissues), with a much lower C:N ratio. Thus, increases in the plant C:N ratio also contributed slightly to an increase the C stored per unit N in the ecosystem.

When the ecosystem is warmed in addition to increasing CO_2, the results are again nearly identical during the first 60 years between simulations with high versus low DON losses and among the four DON-loss models (Fig. 1). However, N redistribution from soils to plants is enhanced because of direct stimulation of N mineralization by warming (Fig.3). Thus the amount of N redistributed and the resulting C accumulation are larger under both elevated CO_2 and higher temperatures than elevated CO_2 alone. Warming also stimulated microbial respiration, resulting in a short-lived (<10 years) decrease in soil C that was quickly compensated for by higher litter inputs. Within about 30 years, these litter inputs increased the soil C:N ratio above its initial value, which contributed to an increase in the C stored per unit N in the ecosystem. The combined effects of elevated CO_2 and of warming-induced increases in N availability stimulated plant production and biomass more than elevated CO_2 alone. Thus, with a combination of elevated CO_2 and warming, increases in woody tissues and the consequent increase in plant C:N ratio contributed significantly to an increase the C stored per unit N in the ecosystem (Fig. 2). By year 60, the simulations with both elevated CO_2 and warming sequestered about 2.5 times as much C as with only elevated CO_2.

Responses after year 60 with low DON losses: With low DON losses, the amount of C stored after year 60 does not differ much among the four DON-loss models in either the simulations with elevated CO_2 alone or with elevated CO_2 and warming. This concordance among models results from the low cumulative losses of DON relative to the other N fluxes in the models. The differences among DON-loss models are therefore not strongly manifested in the rates of N accumulation. Sequestration of C continues for the duration of all low-DON-loss simulations, although at a rate that is only about 17% of that during the first 60 years (Fig. 1,

Table 3). Most of the C accumulation after year 60 is associated with the net accumulation of N by the ecosystem (Fig. 2, Table 3).

For the low-DON-loss simulations, the amount of C accumulated between years 60 and 1000 is about the same with elevated CO_2 alone as it is with elevated CO_2 and warming (~ 16 kg C m^{-2}; Fig. 1, Table 3). In both sets of simulations, increases in plant and microbial N demand draw down DIN concentrations. However, there is a diminishing return of increases in N demand on DIN concentration as the DIN concentrations decline. Thus, although N demand differs between the two sets of simulations, DIN concentrations decrease to about the same level and place similar limits on plant productivity. Loss rates of DIN are therefore about the same and the net rates of N accumulation comparable. Because N accumulation in the ecosystem is the major mechanism of C sequestration after year 60 (~ 70% of total, Fig. 2), the rates of C accumulation are also about the same in the elevated CO_2 alone and elevated CO_2 and temperature simulations.   However, in response to elevated CO_2 alone, the low-DON-loss ecosystems accumulate an additional ~5 kg C m^{-2} after year 1000 before eventually approaching a steady state (~2500 years); in response to both elevated CO_2 and temperature, the ecosystems sequester less than 1 kg C m^{-2} after year 1000.

Changes in plant and soil C:N ratios contribute substantially less to C sequestration than does N accumulation after year 60 in the low-DON-loss simulations (Fig. 2, Table 3). Plant C:N ratios increase because of an increase in wood relative to leaves and roots, but the associated contribution to C sequestration is less than 5% of the total change between years 60 and 1000. The C:N ratio of soils remains almost constant in the elevated CO_2 simulations and actually declines back toward its initial value in the elevated CO_2 plus warming simulations. Thus

changes in the soil C:N ratio contribute little or negatively (-1.1 to 1.5% of the total) to C sequestration after year 60 (Fig. 2).

After year 60 in these low-DON-loss simulations, the rate of N accumulation in plants per unit N already in plants is only slightly higher than the rate of N accumulation in soils per unit N already in soil (Fig. 1). In our four-factor analysis, this slight difference in the relative rates of N accumulation is reflected in the small increase in C accumulation attributable to N redistribution after year 60 (15 to 20% of the total, Fig. 2). This nearly equal relative rate of N accumulation in plants and soils indicates that the competitive balance between plants and microbes has been almost reestablished after year 60.

The synergistic interaction among the four factors in our analysis also contributes to C sequestration after year 60 of the low-DON-loss simulations (7 to 10% of the total, Fig. 2). This interactive effect results mostly from the net accumulation of N in ecosystem components with C:N ratios that had increased during the first 60 years of the simulation. Thus, new N accumulating in the ecosystem has an enhanced effect on C sequestration because of the higher C:N ratios.

Responses after year 60 with High DON losses: Averaged over all four DON-loss models, the ecosystems with high DON losses sequestered about 1.9 kg C m^{-2} between years 60 and 1000 under elevated CO_2 alone (Fig. 1). This C accumulation amounts to about 38% of the C sequestered in the first 60 years and only about 12% of the C sequestered between years 60 and 1000 by the ecosystems with low DON losses. Before reaching a steady state, the high-DON-loss ecosystems accumulate on average an additional ~6 kg C m^{-2}, but this average increase is due almost entirely to C accumulation in the constant-loss model (Fig. 1, see

discussion below). The major mechanism for C sequestration is again N accumulation in the ecosystem (Table 3). However, because most of the N losses were as unavailable DON, N accumulated at only 12% of the rate in the ecosystems with low DON losses. The within-ecosystem mechanisms of C sequestration (changes in plant and soil C:N ratio and N redistribution between plants and soils) cumulatively contributed only 30% of the C storage between years 60 and 1000, compared to 96% of the C storage during the first 60 years. All the mechanisms of C sequestration differed substantially among the DON-loss models (Fig. 2) and are discussed further below.

Under elevated CO_2 and temperature and high DON losses, the four DON-loss models diverged substantially (Figs. 1 & 2). On average, the ecosystems sequestered only about 1 kg C m^{-2} between years 60 and 1000 or about 7% of the C sequestered during the first 60 years and 6% of the C sequestered in the ecosystems with low DON losses (Fig. 1). After year 1000 the models continued to diverge with changes in C storage ranging from an additional gain of ~ 18 kg C m^{-2} to a loss of ~14 kg C m^{-2} before reaching a steady state (Fig. 1, see discussion below). These ecosystems only accumulated N at 1% the rate of the ecosystems with low DON losses and at 10% the rate that they accumulated N under elevated CO_2 alone (Table 3). The major mechanisms of C sequestration were an increase in soil C:N ratio (59% of the total) and a redistribution of N from soils to plants (36% of the total). The mechanisms of C sequestration again differed among DON-loss models (Fig. 2) and are discussed further below.

Because of the explicit linkages between DOC and DON in the various model structures, simulations with higher DON loss also exhibit higher DOC loss. Thus, the ecosystems with high DON losses lost between 12 to 15 kg m^{-2} 1000 yr^{-1} more C as DOC than did the ecosystems with low DON losses. These differences are about the same magnitude as the differences in total C

sequestration between the two sets of simulations. However, the differences in DOC loss do not account for differences in C sequestration under elevated CO_2 or elevated CO_2 and temperature. Indeed, these differences DOC loss were present at the initial steady state, prior to imposed changes in CO_2 or temperature. In addition, higher DOC losses in the high DON loss simulations were exactly compensated for by lower microbial respiration (see section on "**Modifications to the MEL model**" above). To test the importance of direct C losses as DOC, we reran all the simulations with a DOC:DON ratio of 15 rather than 30. Thus, relative to the original simulations there was the same DON loss but only half the DOC loss. After 1000 years, there was less than a 1.2% difference in the resulting C and N stocks for the R_m-based model, and less than 0.3% difference for the other three models. In contrast, the differences in N retention between the low and high DON-loss simulations resulted in differences in NPP of between 87 and 159 kg C m^{-2} 1000 yr^{-1} or about ten times the net difference in C sequestration. Clearly the direct C losses as DOC exert only a marginal influence on ecosystem C sequestration relative to the effects of DON losses.

   Comparison of Individual DON loss-models: Patterns of ecosystem N and C accumulation were virtually identical for the four DON loss models in the initial 60 years, the period when internal N redistribution dominated ecosystem dynamics.   However, DON-loss model behaviors diverged sharply between years 60 and 1000, particularly with high DON losses and the combination of elevated CO_2 plus warming.

  By year 1000, the constant-loss model accumulated substantially less C in response to elevated CO_2 alone in the high-DON-loss simulations than in the low-DON-loss simulations (about 5 vs. 17 kg C m^{-2}). However, at steady state, C sequestration was similar in high- versus

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low DON-loss simulations because of similar N accumulation. Similar steady-state N pools develop because both simulations have the same constant rate of DON loss, and total N losses (DON plus DIN) must equal the total (constant) N inputs to the ecosystem. As a result, N eventually accumulates in both simulations until the DIN losses equal the initial DIN losses. This condition is met when soil DIN concentrations build up to their initial values, which were identical for the two simulations. Thus, both the low- and high-DON-loss simulations converge on the same soil fertility at steady state, which in turn results in virtually identical steady state values for vegetation biomass and soil organic matter (Fig 2). However, it takes about five times as long to accumulate enough N and approach that steady state with high DON losses than with low DON losses.

In response to elevated CO_2 alone, the other three models accumulate less than a tenth the C between years 60 and 1000 with high DON losses as with low DON losses (<1.7 vs. >15 kg C m^{-2}; Fig. 1). The steady state C and N stocks in these other three models are also much lower with high DON losses than with low DON losses, and are only slightly higher than the stocks after 1000 years. As with the constant-loss model, these models also have to accumulate enough N for the DON plus DIN losses to equal N inputs at steady state. However, unlike the constant-loss model, DON losses in these other models increase as soil C increases (proportional-loss model), as the soil C:N ratio increases (C:N-based model), or as soil respiration increases (R_m-based model).   Because all three of these ecosystem characteristics increase in the simulations, the models accumulate N 2.5 to 6 times slower than the constant-loss model (Table 3). The higher rates of DON loss, and consequent slower rates of N accumulation in the other three models, are indirectly associated with the increase in productivity during the first 60 years. Higher productivity increases litterfall and soil C, which increases DON losses in the

proportional-loss model. The higher soil C stocks and a net decline in soil N (Fig. 2) increase

soil C:N ratios, which increases DON losses in the C:N-based model. The higher soil C stocks

and lower C assimilation efficiency associated with higher soil C:N ratios increase the absolute

rate of microbial respiration, which increases DON losses in the R_m-based model.

The accumulation of N can also have secondary effects on the within-ecosystem

mechanisms for C sequestration. Because the competition between plants and microbes for N

shifts toward plants when CO_2 increases, the relative rate of N accumulation increases more in

plants than in soils, especially before year 60. In the constant-loss model with high DON losses,

this redistribution of N persists after year 60, but at a much slower rate. Because there was no

appreciable net accumulation of ecosystem N in any of the other models between years 60 and

1000, and because the competitive balance between plants and microbes had become

reestablished, the redistribution of N did not contribute to C sequestration after year 60 in these

simulations. The C:N ratios of both plants and soils remained almost constant after year 60 in all

four models and therefore also did not contribute to C sequestration.

With both elevated CO_2 and warming, the four models diverged substantially after year

60 in the high-DON-loss simulations (Figs. 1 & 2, Table 3). The constant-loss and proportional-

loss models sequestered C in the ecosystem and gained N in both plants and soils, but at about a

quarter of the rate in the low-DON-loss simulations (Fig. 1). The C:N-based and R_m-based

models lost C from the ecosystem and N from the plants and soils. The primary mechanism for

these patterns is a net accumulation of ecosystem N after year 60 in the constant-loss and

proportional-loss models and a net N loss after year 60 in the C:N-based and R_m-based models

(Fig. 2). Secondarily, in the simulations that gained N (constant-loss and proportional-loss

models), the persistent competitive advantage of plants favored N redistribution from soils to

plants, which increased C sequestration (Fig. 2). In the simulations that lost N (C:N-based and R_m-based models), the relative redistribution of N was from plants to soils because the net loss of N limits production and the decline in plant biomass precedes the decline in soil organic matter. Thus, although the redistribution of N during the first 60 years resulted in a net accumulation of C, the loss of N eventually overwhelms the dynamics, causing the ecosystem to rebound and lose C. The C:N ratios of both plants and soils again remained almost constant after year 60 in all four models.

These differences among models in the high DON-loss simulations arise because of differences in the sensitivities of the DON-loss mechanisms to warming. In the constant-loss model, DON losses are constant and therefore not sensitive to warming. Thus, after year 60, N continues to accumulate in the constant-loss model with both elevated CO_2 and warming for the same reasons and at about the same rate as with elevated CO_2 alone (Table 3). In the proportional-loss model, DON losses are proportional to soil C stocks, which initially decline because of higher respiration when the ecosystem is first warmed but then recover as litter inputs increase. The DON losses eventually increase as soil C stocks build, but by only about 2/3s as much as in the simulation with elevated CO_2 alone. Therefore, N accumulation is higher in the proportional-loss model with elevated CO_2 and warming than with elevated CO_2 alone. In the C:N-based model, DON loss is proportional to the soil C:N ratio, which increases during the first 60 years of the simulation because of increased litter inputs and a net loss of N to the plants. The higher net N mineralization and higher litter inputs with warming result in even higher soil C:N ratios with elevated CO_2 and warming than with elevated CO_2 alone. Therefore, the C:N-based model loses N faster with elevated CO_2 and warming than with elevated CO_2 alone and there is a net loss of N from the ecosystem. In the R_m-based model, DON losses are proportional to

microbial respiration, which increases with warming and with higher soil C stocks. Therefore, the R_m-based model loses N faster with elevated CO_2 and warming than with elevated CO_2 alone and there is a net loss of N form the ecosystem.

Responses to gradual increases in CO_2 or CO_2 and warming: In the simulations with a gradual doubling of CO_2 or gradual doubling of CO_2 and a 4^oC warming over 100 years, the relatively fast responses associated with within-ecosystem mechanisms (primarily N redistribution) were superimposed on the slower responses associated with N accumulation by the ecosystem. Thus the responses that dominated during the first 60 years under an instantaneous increase in CO_2 or CO_2 and temperature were spread out over the 100-year simulations and had not fully developed by the end of the simulations (Fig. 3, Table 4). However, the instantaneous and gradual-change simulations eventually reach identical steady states.

As in the first 60 years of the instantaneous-change simulations, the redistribution of N from soils to vegetation dominated the response during the 100 years of the gradual-change simulations and the effect of this mechanism on C sequestration was higher with both elevated CO_2 and warming than with elevated CO_2 alone (Fig. 3, Table 4). Increases in plant and soil C:N ratios contributed less to C sequestration, but in amounts proportionately equivalent to their contributions in the instantaneous-change simulations. With low DON losses, ecosystem N accumulated during the first 100 years in the gradual-change simulations at about 68 to 75% the rate observed in the instantaneous-change simulations, primarily because N demands track the gradual increase in CO_2. With high DON losses, N gains and losses were small during the first 100 years of all the simulations, and the dynamics in the gradual-change simulations generally lagged behind those in the instantaneous-change simulations by about two decades.

Conclusions

The major conclusion from our analysis is that DON losses can have significant effects on the long-term sequestration of C in terrestrial ecosystems in response to elevated CO_2 and warming. Because our simulations consider forms of DON that are not available to plants or microbes (e.g., Neff et al. 2003), DON losses cannot be attenuated when elevated CO_2 concentrations increase ecosystem N demand, whereas DIN losses can. Thus, high rates of DON relative to DIN loss result in lower rates of ecosystem net N accumulation, and hence lower C sequestration, in response to elevated CO_2. This mechanism for constraining C sequestration is similar to the mechanism proposed by Hedin et al. (1995) and Vitousek et al. (1998) for the persistence of N limitation late into succession.

What general characteristics of the C and N cycles promote C sequestration in our simulations? Potential C sequestration is greatest when the ratio of DON:DIN is small, when there is a high N throughput by the ecosystem, when there is a large and prolonged depletion of DIN in response to elevated N demand, and when there is a large amount of C stored per unit N in the ecosystem (i.e., a high ecosystem C:N ratio). The first three characteristics determine the rate of N accumulation in response to elevated CO_2 and the last determines the C return associated with that N accumulation. Each of these characteristics varies widely among ecosystems (e.g., DIN versus DON, Perakis and Hedin 2002, Goodale et al. 2000; rates of N throughput, Aber et al. 1998, Likens et al 1977; DIN depletion in response to elevated N demand, Finzi and Schlesinger 2003, Hungate et al. 1997; and variable ecosystem C:N ratios, Vitousek et al. 1988) and has potential to affect C sequestration. Although our simulations primarily highlight the important yet overlooked role of variations in DON versus DIN losses on

C sequestration, we expect that many other factors, including disturbance regimes and nutrient limitation patterns, are also likely to be important. For example, in simulations of clear cutting, high DIN losses promoted even greater N losses during the first 10-15 years of recovery as plant N uptake reestablishes (data not shown). However, after about 60 years of recovery under elevated CO2, N demand lowers DIN concentration and the high-DIN-loss ecosystems retain more N than the high-DON-loss ecosystems, thus increasing C sequestration (see Hungate et al. 2003 for a more thorough assessment of N losses and disturbance). Our analysis can also be extended to consider losses of dissolved organic forms of phosphorus, which may be particularly important in P-limited tropical ecosystems (Hedin et al. 2003), or in temperate regions where accelerated N deposition has relaxed N limitation.

Despite the potentially large consequences of DON losses on N accumulation and C sequestration, our analysis indicates that these effects are slow to emerge and may take millennia to fully develop. Within ecosystem mechanisms, such as N redistribution and changes in plant and soil C:N ratios (e.g., Hooker and Compton 2003), dominated short-term C sequestration in response to elevated CO2 and temperature. The long-term impact of ecosystem N accumulation on C sequestration will remain difficult to evaluate empirically, independent of effects from shorter-term mechanisms, yet such dynamics can be captured through ecosystem simulation models. Moreover, because these mechanisms operate at different time scales, short-term observations cannot be used to assess long-term responses (Rastetter 1996).

The degree to which the long-term effects of DON losses are masked by faster within-ecosystem responses depends on their relative strength at any particular time. From our analysis, within-ecosystem responses are rapid, and are dominated by the redistribution of N from soils to vegetation   The magnitude of this effect is proportional to the difference between the C:N ratios

of vegetation and soil. This difference is large if the vegetation is woody, suggesting that the effects of DON losses will be more strongly masked, and for a longer duration, in woody ecosystems. In addition, increases in plant and soil C:N ratios can contribute to the within-ecosystem responses and help mask the effects of DON losses. The effects of DON losses would therefore be more effectively masked by within-ecosystem N cycling processes in ecosystems that become more woody in response to elevated CO_2 and warming (e.g., Sturm et al. 2001).

Our analysis also indicates that the way that DON losses are regulated can affect long-term C sequestration, provided that DON losses are large relative to DIN losses. Although ecological factors such as N inputs, disturbance, climate, and succession are thought to influence broad patterns of N loss (Sollins and McCorison 1980, Hedin et al. 1995, Goodale et al. 2000, Perakis and Hedin 2002), we lack information on the mechanisms that regulate relative DIN versus DON losses from ecosystems. Variation in the availability and remineralization of different DON forms to DIN (Qualls and Haines 1992) as well as direct incorporation of DIN into DON (Perakis and Hedin 2001, Dail et al. 2001), provide further complexity that was not captured by our simulations. Our simulations indicate that different mechanisms of DON regulation can yield similar responses to some perturbations but not others (e.g., the similarity among models in the responses to elevated CO_2 versus their divergence in response to elevated CO_2 and temperature). Yet despite such uncertainty, results from our four DON-loss models consistently indicate that as DON losses increase in response to the changing environment (e.g., warming), the effects of high DON losses will be exacerbated (i.e., C sequestration decreased).

Losses of DON are accompanied by a direct loss of C as DOC. We assumed a constant C:N ratio for these two losses, but compensated for the difference in C loss between low- versus high-DON-loss simulations by adjusting microbial respiration. Our analysis indicates that the

direct losses of C as DOC do not play a major role in terrestrial C sequestration. Thus, differences in the C:N ratio of dissolved organic matter leaching out of the ecosystems should have only a minor influence on C sequestration. Far more important is the direct loss of N as DON and its consequent effects on fertility and plant production. The differences in productivity between our low- and high-DON-loss simulations were an order of magnitude greater than the differences in the cumulative losses of C as DOC. Nevertheless, even small changes in DOC leaching may signal large changes in terrestrial C storage (e.g., Freeman et al. 2001).

On a regional or global scale, the fate DON lost from the ecosystem also has ramifications for C sequestration. The initial C:N ratio of our simulated ecosystem (plants plus soils) is 52. Therefore if the DON is later transformed and entrained into the N cycle of an ecosystem with a higher C:N ratio (e.g., a system with a larger woody component), then it would contribute to C sequestration. On the other hand, if it is buried as is (C:N ~ 30) or gets entrained into an ecosystem with a lower C:N ratio (e.g., a planktonic ecosystem), then the loss of that DON would decrease C sequestration.

Because of the very long time constants associated with the effects of DON losses on the responses of terrestrial ecosystems to elevated CO_2 and warming, the results of our analysis will be very difficult to test (Rastetter 1996). Furthermore, it will be difficult to find pairs of ecosystems that differ in their relative rates of DON versus DIN loss, but do not differ in some other fundamental way that affects responses to elevated CO_2 and warming. The best potential for testing our ideas in a timely manner would be to experimentally manipulate ecosystems where the masking effects of within-ecosystem responses are likely to be small relative to the effects of DON losses to determine if there is a trend toward high C sequestration with low DON losses relative to DIN losses. Thus the manipulations should be on ecosystems where the C:N

ratio of vegetation is low (i.e., close to the C:N ratio of soils so that the redistribution of N has a smaller effect), where the vegetation is unlikely to increase in woodiness (i.e., to avoid the masking effects of increasing C:N ratios), and where the total throughput of DON plus DIN is high (i.e., a high potential to sequester N). Marshes might be ideal candidates if they are demonstrably N limited.

Our aim in this paper has been to examine how considering the relative magnitudes of DON versus DIN losses might influence assessments of potential C sequestration in terrestrial ecosystems. Our conclusions are that it is vital to quantify these fluxes at least in regards to evaluations of the long-term potential for C sequestration. Because the mechanism underlying the DON-DIN effect is the attenuation of DIN losses by plants and microbes, it is important to quantify these fluxes as they leave the rooting zone before transformation along hydrologic flowpaths through deep soils, groundwaters, and riparian zones to surface waters. Furthermore, it is important to distinguish between DON that is available to plants and microbes and that that is not. Clearly DON in soils is comprised of a wide range of compounds with a continuum of availabilities to both plants and microbes, and our analysis examines responses at the extremes of this continuum. Although our basic conclusions would not be altered with a more realistic representation of DON availability, we recognize that predictive assessments of the long-term potential for C sequestration in response to elevated CO_2 and climate change requires improved understanding and quantification of losses of DIN and all forms of DON.

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Table 1: Model equations, variables, and parameters.

(1) $\dfrac{dE_N}{dt} = R_{Ne} + R_{Nm} - L_{Ne} - U_{Nm} - U_{Nv}$

(2) $\dfrac{dB_C}{dt} = U_{Cv} - R_{Cv} - L_{Cv}$ (3) $\dfrac{dB_N}{dt} = U_{Nv} - L_{Nv}$

(4) $\dfrac{dD_C}{dt} = L_{Cv} - R_{Cm}$ (5) $\dfrac{dD_N}{dt} = L_{Nv} + U_{Nm} - R_{Nm}$

(6) $\dfrac{dV_C}{dt} = -aAV_C$: if $A > 0$ (7) $\dfrac{dV_N}{dt} = -\dfrac{dV_C}{dt}$

$\qquad = -aAV_N$: if $A < 0$

(8) $B_A = \dfrac{B_{Amax}\, c\, B_C}{B_{Amax} + c\, B_C}$ (9) $q = \dfrac{B_C q_{min} q_{max}}{B_A q_{max} + (B_C - B_A) q_{min}}$

(10) $S_C = b_C V_C B_A$ (11) $S_N = b_N V_N B_A$

(12) $G_C = \dfrac{g_C\, C_a}{(k_C + C_a)}$ (13) $G_N = \dfrac{g_N\, N}{(k_N + N)}$

(14) $U_{Cv} = G_C\, I_0 \left(1 - e^{-k_I S_C}\right)$ (15) $U_{Nv} = G_N \left(1 - e^{-k_S S_N}\right)$

(16) $r_{Nup} = \dfrac{G_C b_C k_I}{G_N b_N k_S} e^{k_S S_N - k_I S_C}$ (17) $U_{Nfix} = \dfrac{g_{Nfix} S_N \left(r_{Nup} - r_{Nfix}\right)}{k_{Nfix} + \left(r_{Nup} - r_{Nfix}\right)}$: if $r_{Nup} > r_{Nfix}$

$\qquad\qquad\qquad = 0$: otherwise

(18) $L_{Cv} = m_A B_A + m_W \left(B_C - B_A\right)$ (19) $L_{Nv} = \dfrac{q\, B_N}{B_C}\left[\dfrac{m_A}{q_A} B_A + \dfrac{m_W}{q_W}\left(B_C - B_A\right)\right]$

(20) $R_{Cv} = r_A B_A + r_W \left(B_C - B_A\right)$ (21) $A = \ln\left(\dfrac{B_C}{q\, B_N}\right) + h\left(\dfrac{1}{B_C}\dfrac{dB_C}{dt} - \dfrac{1}{B_N}\dfrac{dB_N}{dt}\right)$

(22) $L_{Ne} = \beta_{Ne} E_N$ (23) $\theta = \dfrac{\varepsilon_N \phi}{\varepsilon_C}$

(24) $U_{Nm} = \dfrac{\psi\, \alpha_N\, D_C^2\, N}{\theta\, D_N\left(\kappa_{Nm} + N\right)}$ (25) $M_N = \psi\, D_N + U_{Nm}$

(26) $\Lambda_C = \dfrac{\varepsilon_C \theta M_N}{\psi\, D_C + \theta M_N}$ (27) $\Lambda_N = \dfrac{\varepsilon_N\, \psi\, D_C}{\psi\, D_C + \theta M_N}$

(28) $R_{Cm} = \psi\, D_C (1 - \Lambda_C)$ (29) $R_{Nm} = M_N (1 - \Lambda_N)$

(30) Constant-loss model: $L_{DOC} = \beta_{DOC}$

Proportional-loss model: $L_{DOC} = \beta_{DOC} D_C$

C:N-based model: $L_{DOC} = \beta_{DOC} \dfrac{D_C}{D_N}$

R_m-based model: $L_{DOC} = \beta_{DOC} R_{Cm}$

(31) $L_{DON} = L_{DOC} / \theta_{DOM}$

A	acclimation potential
a	acclimation rate (yr^{-1})
B_A	active-tissue C in plants (g C m^{-2})
B_{Amax}	maximum B_A (g C m^{-2})
B_C	C in plant biomass (g C m^{-2})
b_C	leaf area per unit C (m^2 g^{-1} C)
B_N	N in plant biomass (g N m^{-2})
b_N	root length per unit C (m g^{-1} C)
c	allometric parameter
C_a	CO_2 concentration (ml m^{-3})
D_C	C in soil detritus (g C m^{-2})
D_N	N in soil detritus (g N m^{-2})
g_C	canopy-level quantum yield (g C MJ^{-1})
G_C	substrate-corrected g_C (g C m^{-2} yr^{-1})
g_N	maximum plant N uptake (g N m^{-2} yr^{-1})
G_N	substrate-corrected g_N (g N m^{-2} yr^{-1})
g_{Nfix}	N fixation constant (g^2 N g^{-1} C m^{-2} yr^{-1})
h	acclimation damping (yr)
I_0	above-canopy irradiance (MJ m^{-2} yr^{-1})
k_C	1/2-saturation for CO_2 (ml m^{-3})
k_I	light extinction (m^2 m^{-2})
k_N	plant 1/2-saturation for N (g N m^{-2})
k_{Nfix}	1/2-saturation fixation cost (g C g^{-1} N)
k_S	soil-resource extinction (m^2 m^{-1})
L_{CD}	organic-C leaching (g C m^{-2} yr^{-1})
L_{Cv}	litter C loss (g C m^{-2} yr^{-1})
L_{DOC}	DOC loss (g C m^{-2} yr^{-1})
L_{DON}	DON loss (g N m^{-2} yr^{-1})
L_{ND}	organic-N leaching (g N m^{-2} yr^{-1})
L_{Ne}	inorganic-N leaching (g N m^{-2} yr^{-1})
L_{Nv}	litter N loss (g N m^{-2} yr^{-1})
m_A	active-tissue-litter loss rate (yr^{-1})
m_W	woody-tissue-litter loss rate (yr^{-1})
N	inorganic N (g N m^{-2})
q	plant optimal C:N (g C g^{-1} N)
q_A	active-tissues-litter C:N (g C g^{-1} N)
q_{max}	maximum plant C:N (g C g^{-1} N)
q_{min}	minimum plant C:N (g C g^{-1} N)
q_W	woody-tissue-litter C:N (g C g^{-1} N)
r_A	active-tissue respiration rate (yr^{-1})
R_{Cm}	microbial respiration (g C m^{-2} yr^{-1})
R_{Cv}	plant respiration (g C m^{-2} yr^{-1})
R_{Ne}	external N supply (g N m^{-2} yr^{-1})
R_{Nm}	N mineralization (g N m^{-2} yr^{-1})
r_{Nfix}	cost of N fixation (g C g^{-1} N)
r_{Nup}	cost of N uptake (g C g^{-1} N)

r_W woody-tissue respiration rate (yr^{-1})

S_C leaf area ($m^2\ m^{-2}$)

S_N root length ($m\ m^{-2}$)

t time (yr)

U_{Cv} gross photosynthesis ($g\ C\ m^{-2}\ yr^{-1}$)

U_{Nm} microbial N uptake ($g\ N\ m^{-2}\ yr^{-1}$)

U_{Nv} plant N uptake ($g\ N\ m^{-2}\ yr^{-1}$)

V_C plant C-uptake effort

V_N plant N-uptake effort

α_N microbial N-uptake ($g\ N\ g^{-1}\ C$)

β_{CD} organic-C loss (yr^{-1})

β_{Ne} inorganic-N loss rate (yr^{-1})

β_{DOC} DOC loss rate (varies, see text)

ε_C maximum microbial C efficiency

ε_N maximum microbial N efficiency

η leached organic matter C:N ($g\ C\ g^{-1}\ N$)

θ C:N microbial consumption ($g\ C\ g^{-1}\ N$)

θ_{DOM} C:N dissolved organic matter ($g\ C\ g^{-1}\ N$)

κ_{Nm} microbial N 1/2-saturation ($g\ N\ m^{-2}$)

Λ_C microbial C efficiency

Λ_N microbial N efficiency

M_N microbial N consumption ($g\ N\ m^{-2}\ yr^{-1}$)

ϕ C:N microbial byproducts ($g\ C\ g^{-1}\ N$)

ψ decomposition rate (yr^{-1})

Table 2: Initial steady state fluxes and associated parameter values for processes altered in this application of the MEL model. Original Value is the value reported in Rastetter et al. (2001). A separate value is given for β_{DOC} for each of the four equations used to simulate DOC loss (M1-constant-loss model, M2-proportional-loss model, M3-C:N-based model, and M4-R_m-based model).

Flux	Associated Parameter	Original Value	High DON-loss Value	Low-DON-loss value	Units
DOC loss (L_{DOC})		0	15	3	g C m^{-2} yr^{-1}
	β_{DOC} (M1)	-	15	3	g C m^{-2} yr^{-1}
	β_{DOC} (M2)	-	0.00115	0.000231	yr^{-1}
	β_{DOC} (M3)	-	0.601	0.120	g N m^{-2} yr^{-1}
	β_{DOC} (M4)	-	0.0277	0.00542	g C g^{-1} C
DON loss (L_{DON})		0	0.5	0.1	g N m^{-2} yr^{-1}
	θ_{DON}	-	30	30	g N g^{-1} C
DIN loss (L_{Ne})		0.6	0.1	0.5	g N m^{-2} yr^{-1}
	β_{Ne}	0.231	0.0385	0.192	yr^{-1}
Microbial respiration (R_{Cm})		556	541	553	g C m^{-2} yr^{-1}
	ψ	0.0508	0.0495	0.0505	yr^{-1}
Gross N mineralization (R_{Nm})		60.75	60.42	60.70	g N m^{-2} yr^{-1}
N immobilization (U_{Nm})		47.25	47.42	47.30	g N m^{-2} yr^{-1}
	α_N	0.0126	0.0130	0.0127	g N g^{-1} C

Table 3: Simulated changes in ecosystem C 60 and 1000 years after an instantaneous doubling of CO_2 concentration or an instantaneous doubling of CO_2 concentration and a 4°C increase in temperature. The simulations are for 4 DON-loss models that differ in the way DON losses are controlled and for both low and high initial DON loss rates. The total chance in ecosystem C (ΔC_{total}) has been partitioned among factors associated with: 1) changes in total ecosystem N ($\Delta C_{\Delta N_T}$), 2) changes in plant C:N ratios ($\Delta C_{\Delta C:N_P}$), 3) changes in soil C:N ratios ($\Delta C_{\Delta C:N_S}$), 4) a redistribution of N between plants and soils ($\Delta C_{B_N \Leftrightarrow D_N}$), and 5) the interactions of the changes in total N, C:N ratios, and N redistribution (ΔC_{inter}).

2 X CO_2 only										
DON-loss Model	Average of all models at year 60		Constant-loss model at year 1000		Proportional-loss model at year 1000		C:N-based model at year 1000		R_m-based model at year 1000	
DON loss	Low	High	Low	High	Low	High	Low	High	Low	High
$\Delta C_{\Delta N_T}$	0.97	0.21	12.85	3.43	11.95	1.33	12.44	0.62	11.88	0.72
$\Delta C_{\Delta C:N_P}$	0.52	0.45	1.24	0.71	1.21	0.54	1.23	0.47	1.21	0.48
$\Delta C_{\Delta C:N_S}$	1.42	1.42	1.65	1.65	1.68	1.56	1.66	1.52	1.69	1.53
$\Delta C_{B_N \Leftrightarrow D_N}$	3.40	3.05	6.15	3.89	6.06	3.29	6.10	3.07	6.05	3.10
ΔC_{inter}	0.07	0.02	1.32	0.28	1.23	0.09	1.28	0.04	1.22	0.05
ΔC_{total}	6.39	5.15	23.21	9.96	22.13	6.81	22.71	5.72	22.05	5.88

2 X CO_2 and +4 $^\circ$C										
DON-loss Model	Average of all models at year 60		Constant-loss model at year 1000		Proportional-loss model at year 1000		C:N-based model at year 1000		R_m-based model at year 1000	
DON loss	Low	High	Low	High	Low	High	Low	High	Low	High
$\Delta C_{\Delta N_T}$	0.45	0.08	12.06	3.70	11.69	2.65	11.73	-1.71	10.89	-3.81
$\Delta C_{\Delta C:N_P}$	1.16	1.15	1.59	1.34	1.59	1.29	1.59	1.03	1.57	0.86
$\Delta C_{\Delta C:N_S}$	2.22	2.31	1.95	2.92	2.02	2.96	2.01	2.93	2.16	2.81
$\Delta C_{B_N \Leftrightarrow D_N}$	10.86	10.71	13.98	12.55	14.03	12.20	14.01	10.30	14.09	9.22
ΔC_{inter}	0.34	0.28	2.00	0.78	1.97	0.60	1.97	-0.10	1.89	-0.39
ΔC_{total}	15.02	14.53	31.58	21.29	31.30	19.70	31.31	12.45	30.60	8.69

Table 4: Simulated changes in ecosystem C after 100 years with CO_2 concentration doubling linearly over the 100 years or with CO_2 concentration doubling and temperature increasing by 4^oC over the 100 years. The simulations are for 4 DON-loss models that differ in the way DON losses are controlled and for both low and high initial DON loss rates. The total chance in ecosystem C (ΔC_{total}) has been partitioned among factors associated with: 1) changes in total ecosystem N ($\Delta C_{\Delta N_T}$), 2) changes in plant C:N ratios ($\Delta C_{\Delta C:N_P}$), 3) changes in soil C:N ratios ($\Delta C_{\Delta C:N_S}$), 4) a redistribution of N between plants and soils ($\Delta C_{B_N \Leftrightarrow D_N}$), and 5) the interactions of the changes in total N, C:N ratios, and N redistribution (ΔC_{inter}).

2 X CO_2 only								
Model	Constant-loss		Proportional-loss		C:N-based		R_m-based	
DON loss	Low	High	Low	High	Low	High	Low	High
$\Delta C_{\Delta N_T}$	1.24	0.34	1.23	0.27	1.22	0.20	1.22	0.22
$\Delta C_{\Delta C:N_P}$	0.55	0.45	0.55	0.45	0.55	0.44	0.55	0.44
$\Delta C_{\Delta C:N_S}$	1.38	1.38	1.38	1.38	1.38	1.38	1.38	1.38
$\Delta C_{B_N \Leftrightarrow D_N}$	3.33	2.93	3.33	2.93	3.33	2.92	3.33	2.92
ΔC_{inter}	0.09	0.02	0.09	0.02	0.09	0.02	0.09	0.02
ΔC_{total}	6.59	5.12	6.58	5.05	6.57	4.97	6.57	4.98
2 X CO_2 and +4 ^oC								
Model	Constant-loss		Proportional-loss		C:N-based		R_m-based	
DON loss	Low	High	Low	High	Low	High	Low	High
$\Delta C_{\Delta N_T}$	0.83	0.26	0.84	0.31	0.81	0.15	0.76	-0.10
$\Delta C_{\Delta C:N_P}$	1.04	1.01	1.04	1.01	1.03	1.01	1.03	1.00
$\Delta C_{\Delta C:N_S}$	1.67	1.80	1.67	1.79	1.68	1.81	1.69	1.85
$\Delta C_{B_N \Leftrightarrow D_N}$	9.38	9.23	9.38	9.23	9.39	9.24	9.40	9.26
ΔC_{inter}	0.34	0.26	0.34	0.27	0.34	0.25	0.34	0.22
ΔC_{total}	13.27	12.56	13.28	12.61	13.25	12.46	13.21	12.23

Figure 1: Responses of the MEL model to an instantaneous doubling in CO_2 concentration (left column) or an instantaneous doubling in CO_2 concentration plus 4^oC warming (right column). Results are for simulations with low or high DON losses (as indicated in upper two panels). Line types indicate the results from 4 DON-loss models (as indicated in upper right panel) that differ in how DOC and DON losses are controlled: 1) Constant-loss model, 2) proportional-loss model, 3) C:N-based model, and 4) R_m-based model. The results at year 1000 for vegetation and soil N follow the same sequence from top to bottom as the results for total C in the upper panels. The final steady state is indicated on the right of each panel following the break in the x-axis.

Figure 2: Partitioning of the cumulative change in ecosystem C following a doubling of CO_2 concentration (left column) or following a doubling of CO_2 concentration and 4^oC warming (right column) among four factors. Shown are the cumulative changes in C associated with total changes in ecosystem N (Δ total N), with changes in plant C:N ratios (Δ plant C:N), with changes in soil C:N ratios (Δ plant C:N), with the redistribution of N between plants and soils (N redistribution), or with the interaction among these four factors (interaction) for the proportional-loss model with low and high DON losses and for the R_m-based model with high DON losses. The changes in ecosystem C associated with these factors and their interaction sum to the total change in ecosystem C shown in Figure 1 for the respective models. The final steady state is indicated on the right of each panel following the break in the x-axis.

Figure 3: Partitioning of the cumulative change in ecosystem C during a gradual doubling of CO_2 concentration over 100 years (left column) or during a gradual doubling of CO_2 concentration and 4^oC warming over 100 years (right column) among four factors. Shown are the cumulative

changes in C associated with total changes in ecosystem N (Δ total N), with changes in plant C:N

ratios (Δ plant C:N), with changes in soil C:N ratios (Δ plant C:N), with the redistribution of N

between plants and soils (N redistribution), or with the interaction among these four factors

(interaction) for the proportional-loss model with low and high DON losses and for the R_m-based
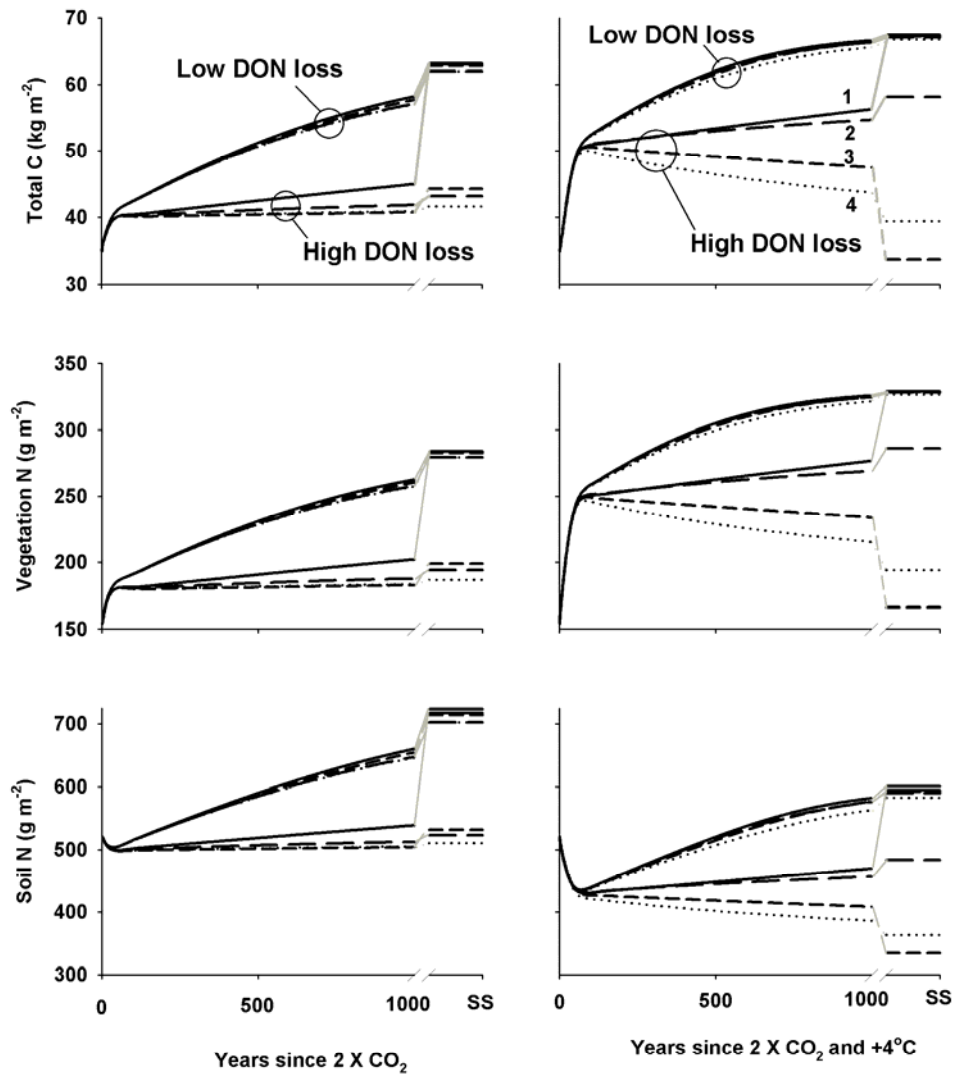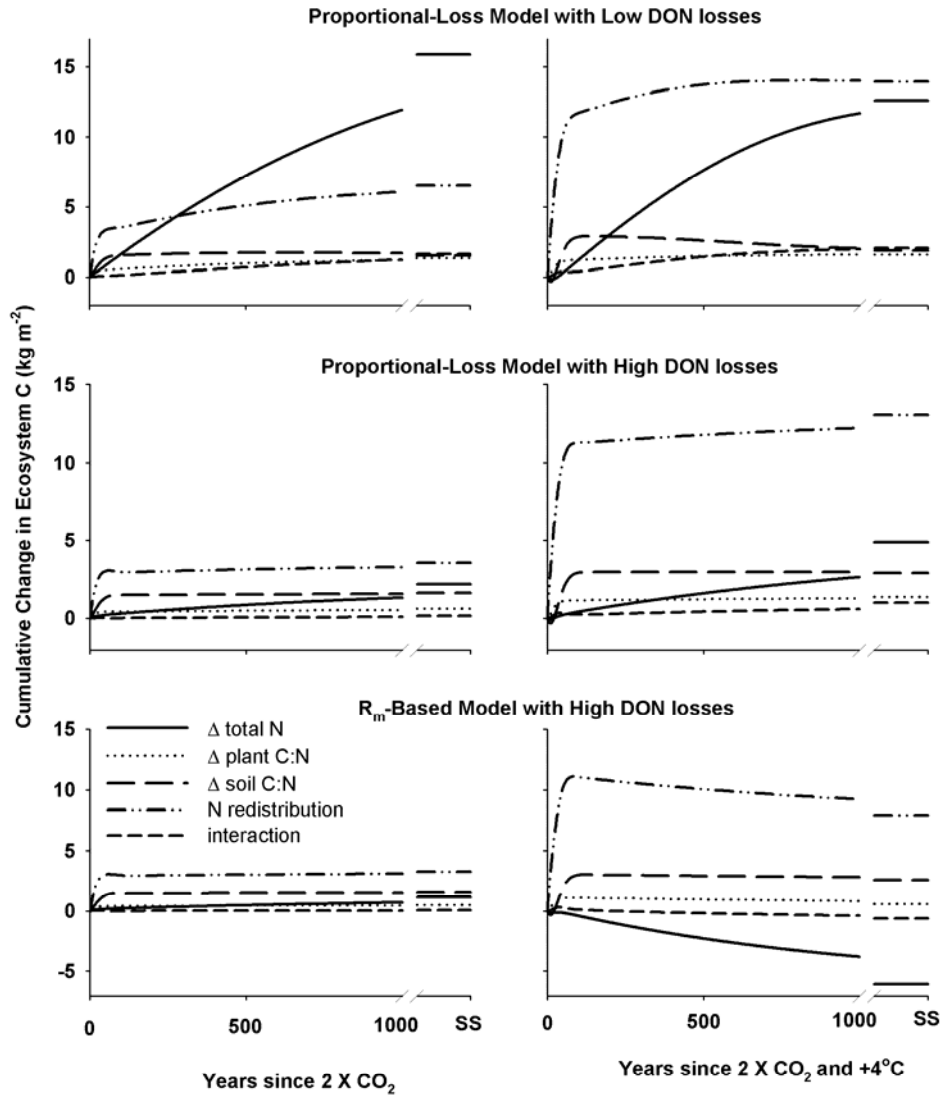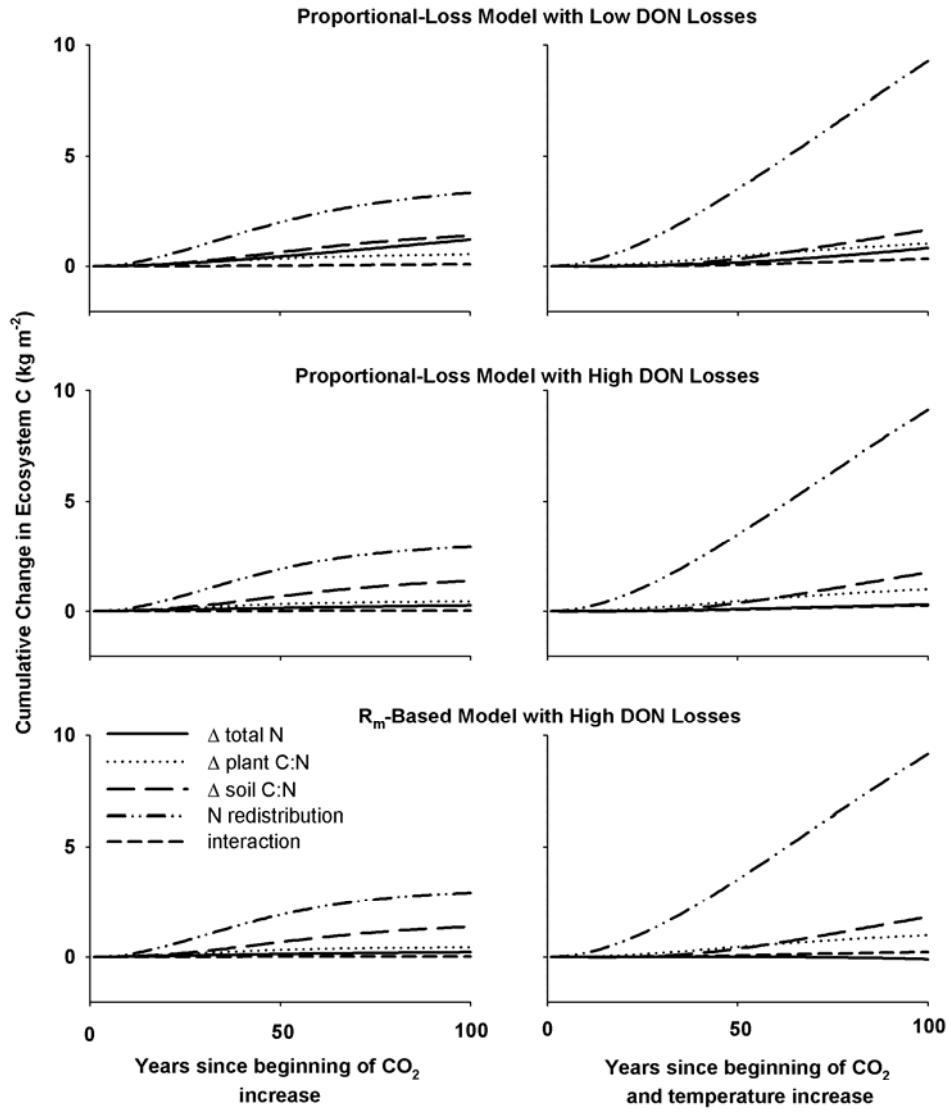
model with high DON losses.

Figure 1

Figure 2

Figure 3