

1 **Title:** Insights into mechanisms governing forest carbon response to nitrogen deposition: a  
2 model-data comparison using observed responses to nitrogen addition

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9 **Running title:** Insights into mechanisms governing C and N interactions

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24 **Abstract**

25 In many forest ecosystems, nitrogen (N) deposition enhances plant uptake of carbon dioxide,  
26 thus reducing climate warming from fossil fuel emissions. Therefore, accurately modeling how  
27 forest carbon (C) sequestration responds to N deposition is critical for understanding how future  
28 changes in N availability will influence climate. Here, we use observations of forest C response  
29 to N inputs along N deposition gradients and at five temperate forest sites with fertilization  
30 experiments to test and improve a global biogeochemical model (CLM-CN 4.0). We show that  
31 the CLM-CN plant C growth response to N deposition was smaller than observed and the  
32 modeled response to N fertilization was larger than observed. A set of modifications to the CLM-  
33 CN improved the correspondence between model predictions and observational data (1) by  
34 increasing the aboveground C storage in response to historical N deposition (1850–2004) from  
35 14 to 34 kg C per additional kg N added through deposition and (2) by decreasing the  
36 aboveground net primary productivity response to N fertilization experiments from 91 to 57 g C  
37  $\text{m}^{-2} \text{yr}^{-1}$ . Modeled growth response to N deposition was most sensitive to altering the processes  
38 that control plant N uptake and the pathways of N loss. The response to N deposition also  
39 increased with a more closed N cycle (reduced N fixation and N gas loss) and decreased when  
40 prioritizing microbial over plant uptake of soil inorganic N. The net effect of all the  
41 modifications to the CLM-CN resulted in greater retention of N deposition and a greater role of  
42 synergy between N deposition and rising atmospheric  $\text{CO}_2$  as a mechanism governing increases  
43 in temperate forest primary production over the 20th century. Overall, testing models with both  
44 the response to gradual increases in N inputs over decades (N deposition) and N pulse additions  
45 of N over multiple years (N fertilization) allows for greater understanding of the mechanisms  
46 governing C-N coupling.

47

48 **1. Introduction**

49  
50       Reactive nitrogen (N) from fossil fuel combustion and agricultural activities influences  
51 global climate by altering atmospheric chemistry, aerosols, and greenhouse gas concentrations  
52 (Butterbach-Bahl et al., 2011; Pinder et al., 2012). However, the direction of the climate impact  
53 of reactive N primarily depends on the balance of opposing processes: positive radiative forcing  
54 from emissions of N<sub>2</sub>O, a greenhouse gas, and negative radiative forcing from altered  
55 atmospheric chemistry and carbon dioxide (CO<sub>2</sub>) storage in N-fertilized forests (Arneth et al.,  
56 2010; Pinder et al., 2012). Reactive N deposited on forest ecosystems can increase primary  
57 production if the forest is N-limited (Nadelhoffer et al., 1999; Magnani et al., 2007; Thomas et  
58 al., 2010), which results in less CO<sub>2</sub> in the atmosphere and consequently reduced climate  
59 warming. Recent estimates show that this CO<sub>2</sub> uptake broadly offsets warming from N<sub>2</sub>O  
60 emissions, indicating that the N-induced forest sink of CO<sub>2</sub> has an important role in global  
61 climate (Zaehle et al., 2011; Butterbach-Bahl et al., 2011; Pinder et al., 2012).

62       Accurately predicting how carbon (C) storage in forest ecosystems will respond to the  
63 changing deposition of reactive N is critical for developing climate change targets for reducing  
64 emissions and air pollution. Global biogeochemical models coupled to climate and atmospheric  
65 chemistry models are powerful tools for exploring this carbon- nitrogen-climate interface  
66 (Sokolov et al., 2008; Thornton et al., 2009; Yang et al., 2009; Zaehle and Friend, 2010), but it is  
67 paramount to build confidence in predictions of how C uptake and storage respond to changing  
68 N inputs.

69       Fortunately, a variety of observational and experimental data are available to test and  
70 improve the sensitivity of global biogeochemical models to changes in N deposition and the  
71 resulting C sequestration. Studies have generally shown that elevated N inputs often increase

72 plant growth and soil C sequestration (Magnani et al., 2007; Hyvonen et al., 2008; de Vries et al.,  
73 2009; Janssens et al., 2010; Thomas et al., 2010), although some ecosystems can be harmed by  
74 chronic elevated N inputs that lead to soil acidification and N saturation (Aber et al., 1998;  
75 Hogberg et al., 2006; Wallace et al., 2007). The current range of estimates quantifying the  
76 additional C sequestered per unit of N added (kg C per kg N; or dC/dN) is broad, in part due to  
77 the myriad of approaches used to quantify dC/dN. These approaches include N fertilization  
78 studies (Hyvonen et al., 2008; Liu and Greaver, 2009), where large inputs of N are added to  
79 forests over short time scales, and N deposition gradient studies (Magnani et al., 2007; de Vries  
80 et al., 2009; Thomas et al., 2010), where spatial variation in N deposition and forest growth are  
81 used to estimate the impact of gradual increases N deposition on C storage over multiple  
82 decades. Furthermore, there can be variation among studies in the C pools being measured, as  
83 some studies calculate the dC/dN of aboveground stem C (de Vries et al., 2009; Thomas et al.,  
84 2010), soil organic matter (Janssens et al., 2010), or net ecosystem production (NEP; Magnani et  
85 al., 2007; Sutton et al., 2008). Finally, the observations span regions with very different historical  
86 N deposition loads (i.e. the US compared to Western Europe). Successfully using the available  
87 data to test and improve global biogeochemical models requires directly accounting for the  
88 variation in magnitude and time-scale of N additions in the observational and experimental data.

89         Many different approaches have been used to model key processes influencing C and N  
90 cycle interactions in terrestrial ecosystems (Zaehle and Dalmonech, 2011). For example, N  
91 fixation has been modeled as a function of net primary production (NPP) (Thornton et al., 2007),  
92 evapotranspiration (Felzer 2012; Tian et al. 2011; Yang et al., 2009; Zaehle and Friend, 2010) or  
93 N demand (Gerber et al., 2010). Similarly, N uptake has been represented as a direct function of  
94 photosynthesis and C : N stoichiometric constraints on building plant tissue (Thornton et al.,

95 2002, 2007) or based on allocation of C to plant roots (Gerber et al., 2010; Raich et al. 1991;  
96 Zaehle and Friend, 2010). Soil and plant buffering of the C cycle to daily to annual changes in N  
97 availability also differs among models (Gerber et al., 2010; Zaehle and Friend, 2010). Despite  
98 these differences, global biogeochemical models predict reasonable levels of global NPP  
99 (Thornton et al., 2007; Gerber et al., 2010; Zaehle et al., 2010a), suggesting that the  
100 representation or parameterization of one C-N process may compensate for the representation or  
101 parameterization of another. Understanding how the different model structures or  
102 parameterizations influence the prediction of how ecosystems respond to N deposition requires  
103 isolating key processes that govern C and N interactions. Unfortunately, inter-model  
104 comparisons can be limited by broad differences in model structure that make it difficult to  
105 isolate particular processes that differ among models. One approach to this problem is to  
106 compare different representations of particular C and N cycle processes within the same general  
107 model, thereby obtaining a better understanding of which processes influence predictions of how  
108 terrestrial C storage and climate respond to changing N availability.

109         Here, we explored the influence of alternative approaches to modeling C and N  
110 interactions on the sensitivity of C storage to N inputs in temperate forest ecosystems. To isolate  
111 the alternative approaches, we implemented multiple alternative assumptions about C and N  
112 cycling within a single global biogeochemical land surface model (CLM- CN 4.0). We focused  
113 on five key assumptions about the N cycle in the CLM-CN 4.0: (1) the extent to which the N  
114 cycle is open, based on N fixation inputs and N losses relative to internal N cycling, (2) the  
115 buffering of the soil inorganic N pool and plant N pool, (3) the representation of plant N uptake,  
116 (4) the pathways of N loss, and (5) the canopy scaling of photosynthesis. These alternative  
117 assumptions are combined to create a modified version of the CLM-CN 4.0 that is compared to

118 observed ecosystem response to experimental N fertilization and to elevated N inputs across N  
119 deposition gradients.

120

## 121 **2.0 Methods**

122

### 123 ***2.1 Baseline model description***

124 We used the CLM-CN 4.0 as the baseline model (Lawrence et al., 2011, 2012). The  
125 CLM-CN 4.0 is the global land surface model in the Community Earth System Model (Gent et  
126 al., 2011) and includes both terrestrial biogeophysical and biogeochemical processes. Our model  
127 simulations include both the biogeophysical and biogeochemical components of the CLM-CN  
128 but we focus our model evaluation and improvement on the biogeochemical component  
129 (hereafter, referred to as the “clm4cn” model). A description of the key processes that relate to  
130 new model modifications are described below, while a more thorough description of the  
131 biogeochemical component can be found elsewhere (Thornton et al., 2002, 2007, 2009; Thornton  
132 and Rosenbloom, 2005). In the clm4cn model, the C and N cycles are coupled through litter and  
133 soil organic matter decomposition and through plant dynamics. The primary C and N coupling  
134 occurs on the 30-min time scale, as plants compete for N with microbial immobilization into soil  
135 organic matter, where N comes from a generic soil inorganic N pool (i.e.  $\text{NH}_4^+$  and  $\text{NO}_3^-$  are  
136 combined). Plant N demand is based on the N needed to match the demand set by non-N limited  
137 photosynthesis and plant tissue C : N stoichiometric constraints. If the combined N demand  
138 exceeds the available N in soil and retranslocated from sensed plant tissue, plant uptake and  
139 microbial immobilization are reduced in proportion to the available N and their relative demands.  
140 Sources of new N into the clm4cn model include N fixation and N deposition. N losses include

141 denitrification, leaching, fire, and harvest. N fixation is a saturating function of NPP derived  
142 from Cleveland et al. (1999). Fixation and deposition are both directly added to the soil inorganic  
143 N pool. The clm4cn model includes both denitrification and leaching processes; however, the  
144 vast majority of N is lost as N gas in most ecosystems in the model (Table A2). Therefore, the  
145 two loss pathways for N gases in the clm4cn model are very important: (1) a constant 1 % of net  
146 mineralization is lost as N gas and (2) the soil inorganic N that exceeds plant uptake and  
147 immobilization is denitrified at a rate of 50 % per day.

148

## 149 ***2.2. Modified model description***

150 The overall model modifications (referred to as “clm4mod”) build on recent  
151 improvements to the calculations of canopy photosynthesis in the clm4cn (Bonan et al., 2011,  
152 2012) by modifying the representations of plant N uptake, internal N cycling, N loss, and  
153 biological N fixation in mature temperate and boreal forests. The modifications within the  
154 clm4cn reflect differences between the clm4cn and other global biogeochemical models,  
155 specially models that have alternative approaches to modeling N uptake (O- CN: Zaehle and  
156 Friend, 2010; LM3V: Gerber et al., 2010), buffering of plant and soil N availability (O-CN:  
157 Zaehle and Friend, 2010; LM3V: Gerber et al., 2010) and less N fixation in temperate and boreal  
158 forests than the clm4cn (LM3V; Gerber et al., 2010). An overview of the modifications is found  
159 below while a full description is found in the Appendix A.

160

### 161 ***2.2.1 Internal N cycling***

162 Plant N uptake in the clm4mod model is based on Michaelis-Menten kinetics, where

163 the rate of N uptake depends on a maximum uptake rate per gram of fine root C, as well as the  
164 concentration of soil inorganic N in separate  $\text{NH}_4^+$  and  $\text{NO}^{-3}$  pools (see Appendix A for detailed  
165 information). Plant uptake increases with soil temperature and as the internal plant pool of N  
166 decreases relative to a maximum internal pool. The formulation of N uptake is similar to that  
167 used in other global biogeochemical models (Gerber et al., 2010; Zaehle and Friend, 2010) and  
168 differs from the clm4cn by allowing the N uptake to be decoupled from photosynthesis at short  
169 time scales (i.e. seconds to days); longer term N uptake (i.e. at days to years) remains ultimately  
170 coupled to photosynthesis. The maximum internal plant N pool is equal to one year of live plant  
171 tissue N turnover (Gerber et al., 2010). In the clm4mod model, the maximum uptake rate is  
172 assumed to be equal for  $\text{NH}_4^+$  and  $\text{NO}^{-3}$ , as implemented in other models(Gerber et al., 2010;  
173 Zaehle and Friend, 2010). N uptake occurs any time during the growing season (i.e. when leaves  
174 are present on the plant), rather than only during the day within the growing season, as  
175 implemented in the clm4cn model. Plant uptake is assumed to be less competitive for N than soil  
176 immobilization of N, in that plant uptake occurs after soil microbial immobilization demands are  
177 met; in the clm4cn, neither plant uptake nor soil immobilization of N had first access to soil  
178 inorganic N, rather they compete based on their demand for N. The clm4mod includes a plant  
179 labile N (short/medium-term storage) pool that is used to buffer the demand for N from  
180 photosynthesis from the uptake of N by roots. The buffering occurs by allowing only 2 % of the  
181 plant labile N pool to be available for combining with new photosynthate to build plant tissue.  
182 The 2 % buffering capacity leads to approximately a two-day turnover time of labile plant N.  
183 The clm4cn does not include buffering of labile plant N.

184

### 185 ***2.2.2 N losses***

186           The clm4mod model introduces a nitrification algorithm, an alternative denitrification  
187 algorithm, and a simple algorithm describing the production and hydrologic loss of dissolved  
188 organic matter, including dissolved organic N (DON). Nitrification is a function of soil  $\text{NH}_4^+$ ,  
189 soil temperature, soil water, and net mineralization based on Parton et al. (2001) with 2 % of  
190 nitrification lost as  $\text{N}_2\text{O}$  (Parton et al., 2001). Denitrification is an function of soil  $\text{NO}^{-3}$ , soil  
191 water, and soil heterotrophic activity based on Bradbury et al. (Appendix A5; Bradbury et al.,  
192 1993; Yang et al., 2009). To buffer the availability of soil inorganic N and approximate short-  
193 term cation exchange processes, 10 % of total soil  $\text{NH}_4^+$  is available for immobilization, plant  
194 uptake, nitrification, and leaching(Gerber et al., 2010), and, since  $\text{NO}^{-3}$  is highly mobile in soils,  
195 100 % of  $\text{NO}^{-3}$  is assumed to be available for soil, plant, and loss processes. Dissolved organic  
196 matter is produced at a constant proportion (2 %) of the organic matter transfer between the slow  
197 litter pool (lignin-based) and its receiving soil organic matter pool (based on Gerber et al., 2010).  
198 Both dissolved organic C and N are lost in proportion to hydrologic export, similar to leaching  
199 loss of soil inorganic N in the clm4cn model. The clm4cn does not include separate consideration  
200 of  $\text{NH}_4^+$  and  $\text{NO}^{-3}$  and cycling, nor does it include DOC and DON losses.

201

### 202 **2.2.3 N inputs**

203           In clm4mod N fixation is a function of actual evapotranspiration, based on the central  
204 relationship in the Cleveland et al. (1999) review of N fixation measurements, rather than a  
205 function of NPP, as implemented in clm4cn. In addition, symbiotic and non-symbiotic sources of  
206 N fixation are separated. Symbiotic fixation is added to the labile N pool and non-symbiotic  
207 fixation is added to the soil  $\text{NH}_4^+$  pool. Symbiotic N fixation is assumed to be zero in mid- to  
208 late-successional temperate and boreal forests (i.e. when leaf area index (LAI) > 1) and non-

209 symbiotic N fixation increases with actual evapotranspiration (Appendix A6). Both sources of  
210 fixation are assumed to occur in grasslands and tropical forests. This N fixation routine reduces  
211 the overall N inputs to mid- to late-successional extra-tropical forests (see Table A2 for the  
212 magnitude of change in N fixation). A pathway for N fertilization was also added to facilitate the  
213 simulation of N fertilization experiments.

214 N deposition in the clm4mod was equal to total N deposition in the clm4cn, except N  
215 deposition in the clm4mod was divided into  $\text{NH}_4$  and  $\text{NO}_3$  based on the N deposition input  
216 fields described in Lamarque et al. (2005).

217

#### 218 ***2.2.4 Canopy photosynthesis***

219 Finally, the clm4mod model includes changes to the canopy scaling of photosynthesis,  
220 maximum photosynthetic rates, radiative transfer, leaf photosynthesis, and stomatal conductance  
221 as described in Bonan et al. (2011) and (2012). Bonan et al. (2012) introduced a multi-layer  
222 canopy scaling approach that solved photosynthesis throughout the canopy rather than using a  
223 whole canopy approximation. The model updates in Bonan et al. (2011, 2012) decreased GPP  
224 and effectively reduced the photosynthetic potential of shaded leaves, but the impact of the  
225 changes on C-N interactions has not been investigated. The clm4mod model also includes  
226 specific values of maximum photosynthetic rate for each plant functional type from a synthesis  
227 of a plant trait database (Kattge et al., 2009), although these values differed only slightly from  
228 the values in the clm4cn model for the temperate broadleaf plant type used in this study (clm4cn,  
229 52; clm4mod, 58;  $\mu\text{mol m}^{-2} \text{s}^{-1}$ ).

230

#### 231 ***2.3 Simulations***

232 The clm4cn and clm4mod models were used to simulate forest biogeochemistry at five  
233 sites in North American broadleaf temperate deciduous forests. The five sites were chosen based  
234 on the presence of long-term forest productivity measurements (10+ years), long-term N  
235 fertilization experiments (10+ years), and are contained within the geographic boundaries of the  
236 analysis of forest inventory data by Thomas et al. (2010), which estimated how forest C storage  
237 has responded to N deposition across the northeastern US (see below). Basic descriptions of the  
238 sites are included in Table 1. Four of the five sites were in Michigan, USA, with each site  
239 receiving varying atmospheric deposition and a  $3 \text{ g N m}^{-2} \text{ yr}^{-1}$  experimental addition of N over  
240 ten years (Pregitzer et al., 2008; 1995–2005). The other site was at Harvard Forest in  
241 Massachusetts, USA (Magill et al., 2004) and included two different 14-yr fertilization additions  
242 (5 and  $15 \text{ g N m}^{-2} \text{ yr}^{-1}$ ; 1988–2002). All data used in this study for the five sites and six N  
243 fertilization experiments can be found in Magill et al. (2004) and Pregitzer et al. (2008). We  
244 simulated ecosystem response to transient N deposition and N fertilization at each of the five  
245 sites using the clm4cn and clm4mod models (Table 2). The simulations involved running each  
246 model from 1850 to 2004 at each of the five sites with different combinations of forcing data  
247 (Table 2). The baseline (control; Table 2 Sim. 1) simulation used transient N deposition ( $\text{NH}_x$   
248 and  $15 \text{ NO}_y$ ; Lamarque et al., 2005), atmospheric  $\text{CO}_2$ , land use (by harvesting vegetation  
249 biomass in the year that yields the reported stand age at the five sites), and climate. N deposition  
250 and atmospheric  $\text{CO}_2$  trends had reconstructions covering the entire simulation (1850–2004). A  
251 57-yr meteorological dataset was available to force the model (1948–2004; Qian et al., 2006).  
252 We created site-level meteorological data by using the corresponding grid cell in the global  
253 gridded data (Qian et al., 2006). We used the data from 1948–1972 for the 1850–1972  
254 simulation years, and the 1973–2004 meteorological dataset was used for the 1973–2004

255 simulation years (as in Randerson et al., 2009 and Bonan and Levis 2010). Each simulation  
256 needed initial conditions that were attained by running the model to equilibrium using 1850  
257 values for N deposition and atmospheric CO<sub>2</sub> and the 1948–1972 time-series for meteorological  
258 data. All wildfire was excluded in the spin-up and other simulations due to uncertainties using  
259 the statistical fire model at a single point location. Simulations used site-specific soil texture  
260 (Magill et al., 2004; Pregitzer et al., 2008). We refer to the equilibrium state used as initial  
261 conditions as the pre-industrial steady-state.

262 We isolated the influence of transient N deposition on C cycling at each site by repeating  
263 the baseline simulation described above except for holding N deposition constant at 1850 values  
264 (Table 2 Sim. 2). We also tested whether C cycle sensitivity was different for larger inputs of N  
265 deposition than included in the baseline simulation (Table 2 Sim. 3). The higher N deposition  
266 simulation used N deposition values found in Western Europe and allowed us to explore why N  
267 deposition gradients in North America (Thomas et al. 2010) yielded steeper C responses than in  
268 Western Europe (de Vries et al. 2009). The high N deposition simulation used a N deposition  
269 trajectory from 1850 to 2004 with 1995–2004 mean deposition levels at the five sites of 2.2 g N  
270 m<sup>-2</sup> yr<sup>-1</sup> (Table 1: EU deposition gradient) rather than the actual N deposition rates (1995- 2004)  
271 at these sites that ranged from 0.68–1.18 g N m<sup>-2</sup> yr<sup>-1</sup> (Table 1: US deposition gradient).

272 Because the NPP response to rising atmospheric CO<sub>2</sub> is constrained by N availability, we  
273 tested whether the sensitivity of NPP to rising atmospheric CO<sub>2</sub> changed as a result of the  
274 modifications to clm4cn. To test this sensitivity we performed two additional simulations at the  
275 five sites using the clm4cn and clm4mod: a simulation that held both N deposition and  
276 atmospheric CO<sub>2</sub> constant at 1850 levels (Table 2 Sim. 4) and a simulation that held CO<sub>2</sub>  
277 constant at 1850 levels but included 1850–2004 N deposition levels (Table 2 Sim. 5)

278 Finally, we simulated the six N fertilization experiments (Table 1; four sites with one  
279 experiment each and one site with two experiments) by adding N fertilization to the baseline  
280 simulation to best approximate the field experiments by specifying the start year, duration,  
281 magnitude and intra-annual distribution of N application (Table 2 Sim. 6). To test whether the N  
282 fertilization treatments completely relieved N limitation, we simulated the six N fertilization  
283 experiments again, this time adding a very large dose of enough additional N to the fertilized  
284 treatment to meet any remaining N demand by plants and microbial immobilization ('Non-  
285 nitrogen limited' treatment).

286

287

#### 288 ***2.4 N fertilization and deposition analysis and observations***

289 The model comparison to observations focused on NPP, net ecosystem productivity  
290 (NEP), and annual aboveground net C increment (ACI) in plants because these were the  
291 variables measured in the N fertilization experiments and across the N deposition gradients. The  
292 model response to N fertilization was assessed by comparing mean annual aboveground NPP  
293 (ANPP) in the control and N-fertilized simulations to ANPP data from the corresponding control  
294 and fertilized treatments in the field data. In both the models and observations, ANPP  
295 corresponded to the sum of mean leaf and stem productivity over the course of the observational  
296 data. We also report ACI for the fertilization experiments (the change of aboveground C stock  
297 between years). ACI differs from NPP and ANPP in that ACI does not include the production  
298 and turnover of wood or leaves within a year that are included in NPP measurements. However,  
299 ACI includes the losses of C from mortality that are not included in NPP measurements. We  
300 report the ACI response to fertilization by dividing by the N added ( $dC_{ACI}/dN_{fertilization}$ ).

301 We compared the model response to N deposition to observations from the literature. We  
302 calculated average ACI between 1994 and 2004 in the simulations with and without transient N  
303 deposition (both including transient CO<sub>2</sub>). The ACI response to N deposition was expressed as  
304 the ACI difference divided by the difference in mean N deposition over the same time period  
305 ( $dC_{ACI}/dN_{deposition}$ ). We also report the difference in NEP divided by the difference in N  
306 deposition ( $dC_{NEP}/dN_{deposition}$ ). We compared the above metrics of N deposition response to the  
307 corresponding metrics reported in analyses listed in Table 3.

308 We also assessed the contribution of N retention to the N deposition response in the  
309 clm4cn and clm4mod model. The total N deposition retained between 1970 and 2004 and  
310 between 2000 and 2004 was calculated to determine the long- and short-term retention of N  
311 deposition, respectively. Over each time period, we calculated total deposition retained in the  
312 ecosystem and the fate of N deposition into soil organic matter (including litter and coarse  
313 woody debris) and vegetation.

314 In addition, we assessed the relative contribution of CO<sub>2</sub> fertilization to how NPP  
315 responds to N deposition for each site using the method developed by Churkina et al. (2009) and  
316 Zaehle et al. (2010b). That is, we isolated the pure N deposition (i.e. N deposition enhancement  
317 without an interaction with CO<sub>2</sub> fertilization), the pure CO<sub>2</sub> fertilization, and the synergistic  
318 effect of CO<sub>2</sub> fertilization and N deposition on NPP by calculating the mean NPP (1994–2004) in  
319 the simulations with (1) N deposition and atmospheric CO<sub>2</sub> at pre-industrial levels (Table 2 Sim.  
320 4), (2) only transient N deposition (CO<sub>2</sub> at pre-industrial levels; Table 2 Sim. 5), (3) only  
321 transient CO<sub>2</sub> (N deposition at pre-industrial levels; Table 2 Sim. 2), and (4) both transient N  
322 deposition and CO<sub>2</sub> (i.e. control simulation described above; Table 2 Sim 1). The pure N  
323 deposition response was the difference in NPP between (1) and (2), while the pure CO<sub>2</sub>

324 fertilization response was the difference between (1) and (3). The additional NPP needed to  
325 reach the difference between (1) and (4) was the synergy between N deposition and CO<sub>2</sub>  
326 fertilization.

327

## 328 ***2.5 Sensitivity Analysis to model structure***

329 We explored the sensitivity of the N deposition response ( $dC_{ACI}/dN_{deposition}$ ) to the  
330 mechanisms and processes that differ between the clm4cn and clm4mod models. We created  
331 intermediate models that sequentially step through the 15 model modifications that were  
332 included in the shift from the clm4cn to the clm4mod (Table 4; Fig. 1). The sensitivity analysis  
333 was sequential in that each intermediate model in Table 4 includes all the modifications in the  
334 intermediate models listed above it. To quantify the influence of each process on the N  
335 deposition response, we ran simulations with and without transient N deposition using each of  
336 the intermediate models at a single site (Harvard Forest). The change in  $dC_{ACI}/dN_{deposition}$   
337 associated with each modification was used to quantify the sensitivity of N deposition response  
338 to assumptions about the structure of the C and N cycles.

339 In most of the modifications, we were interested in the influence of the model structure  
340 on the N deposition response in the absence of altered baseline productivity (the pre-industrial  
341 steady-state; see Sect. 2.3). We manually adjusted the denitrification parameter to maintain the  
342 same baseline production so that Model 4 had the same baseline production (NPP and total C  
343 stocks) as Model 3 and Models 5–14 had the same baseline production as Model 15. To maintain  
344 the same baseline production, we altered the proportion excess soil inorganic N (Models 4–10;  
345 Table 4) or NO<sub>3</sub><sup>-</sup> (Models 11–15; Table 4) that is denitrified. A detailed description of the  
346 intermediate models can be found in Appendix B.

347

## 348 **3.0 Results**

349

### 350 ***3.1 Model response to N fertilization experiments***

351 The clm4mod model better predicted mean ANPP measured at the control plots at  
352 the five field sites (i.e. no N additional fertilization added) than the clm4cn model. The mean  
353 observed ANPP across the five sites was  $464 \pm 36 \text{ g C m}^{-2} \text{ yr}^{-1}$  (1 S.E. across sites), while the  
354 mean in the clm4mod and clm4cn model was  $411 \pm 28$  and  $352 \pm 50 \text{ g C m}^{-2} \text{ yr}^{-1}$  (1S.E.),  
355 respectively (Table 5). Both models predicted lower ANPP than observed in the most productive  
356 sites (MI-B, MI-C and MI-D; Fig. 2). Neither model was consistently higher or lower than  
357 observed ANPP in the two least productive sites (HF and MI-A; Fig. 2a).

358 The clm4mod also better predicted both the mean ANPP in the N fertilized simulations  
359 and the increase in ANPP over the control simulation than the clm4cn. The fertilized ANPP in  
360 the clm4mod model ( $474 \pm 8 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) compared better to the observed fertilized ANPP in the  
361 six experiments at the five sites ( $504 \pm 40 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) than the clm4cn model ( $420 \pm 41 \text{ g C m}^{-2}$   
362  $\text{yr}^{-1}$ ; Table 5). The mean increase in ANPP in the fertilization experiments was similar between  
363 the observations ( $55 \pm 8 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) and the clm4mod ( $57 \pm 18 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) with the clm4cn  
364 fertilization response 65% higher ( $91 \pm 19 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) than the observed response. At individual  
365 sites, the clm4mod corresponded substantially better to observations from the two N fertilization  
366 experiments at Harvard Forest than the clm4cn (Fig. 2c). At the MI-A site, the clm4cn performed  
367 better compared to the observations than the clm4mod (Fig. 2c). Both the fertilization responses  
368 in the clm4cn and clm4mod models were within the bounds of uncertainty in the observations at  
369 MI-B, MI-C, and MI-D (Fig. 2c).

370 Despite differences in ANPP response to N fertilization, both the clm4cn and 20  
371 clm4mod models predicted similar aboveground C increments per unit N fertilizer added  
372 ( $dC_{ACI}/dN_{fertilizer}$ ; clm4cn:  $10.7 \pm 1.3 \text{ kgC}(\text{kgN})^{-1}$ ; clm4mod:  $10.6 \pm 4.5 \text{ kgC}(\text{kgN})^{-1}$ ), that was on  
373 average 1.5 times larger than the observed  $dC_{ACI}/dN_{fertilizer}$  ( $4.0 \pm 2.7 \text{ kgC}(\text{kgN})^{-1}$ ; Table 3).  
374 However, the site-to-site variability in the clm4mod  $dC_{ACI}/dN_{fertilizer}$  was larger than the  
375 variability in clm4cn, leading to overlapping uncertainty with the observations. The mean annual  
376 net ecosystem production response to N fertilization ( $dC_{NEP}/dN_{fertilizer}$ ) across all five sites was  
377  $27.9 \pm 2.8$  and  $23.2 \pm 9.4 \text{ kgC}(\text{kgN})^{-1}$  for the clm4cn and clm4mod models, respectively (Table  
378 3). The clm4cn compared better to the observed  $dC_{NEP}/dN_{fertilizer}$  at the six fertilization  
379 experiments ( $31 \pm 10 \text{ kgC}(\text{kgN})^{-1}$ ; Table 3), although both models were contained in the  
380 observational uncertainty. Additionally, both net ecosystem production responses were within  
381 the uncertainty ( $24 \pm 8.7 \text{ kgC}(\text{kgN})^{-1}$ ) reported by Lui and Greaver (2009) in a meta-analysis of  
382 forest NEP response to N fertilization.

383

### 384 ***3.2 Model response to historical N deposition***

385 The clm4mod model had a 144 % larger response of aboveground C increment to N  
386 deposition ( $dC_{ACI}/dN_{deposition}$ ) than the clm4cn model (Table 3). The  $dC_{ACI}/dN_{deposition}$   
387 was  $14.0$  and  $34.2 \text{ kgC}(\text{kgN})^{-1}$  in the clm4cn and clm4mod models, respectively (Table 3). Both  
388 models predicted lower responses than reported for aboveground C increment across the  
389 Northeastern US ( $50 \text{ kgC}(\text{kgN})^{-1}$ ; adjusted from Thomas et al., 2010 for higher rates of N  
390 deposition); however, the bias was substantially reduced in the clm4mod model (Table 3).

391 In the simulations with higher rates of N deposition (1995–2004 mean=  $15.2 \text{ g N m}^{-2}$   
392  $\text{yr}^{-1}$ ), the aboveground C increment response to N deposition ( $dC_{ACI}/dN_{deposition}$ ) decreased in

393 both models (Table 3). The simulations with a large increase in N deposition were designed to  
394 test model sensitivity to N deposition levels larger than typically found in the US but similar to  
395 levels found in parts of Western Europe. Comparing the model results to observations from  
396 Western Europe, the mean  $dC_{ACI}/dN_{deposition}$  across all sites in the clm4mod model (26.2  
397  $kgC(kgN)^{-1}$ ) was contained within the range reported from inventory measurements of European  
398 forests across a N deposition gradient (Table 2; 15–40  $kgC(kgN)^{-1}$ ; DeVries et al., 2009), while  
399 the mean clm4cn model result was lower than the reported range (11.8  $kgC(kgN)^{-1}$ ).

400 For the NEP response to N, adding the belowground vegetation and soil response  
401 increased average  $dC_{NEP}/dN_{deposition}$  across the five sites to 30.0 and 74.1  $kg C (kgN)^{-1}$  in the  
402 clm4cn and clm4mod models, respectively (Table 3). The  $dC_{NEP}/dN_{deposition}$  using the EU N  
403 deposition values for the clm4mod model (49.5 $kgC(kgN)^{-1}$ ) was within the range of values  
404 recalculated for European forest by Sutton et al. (2008; 50–75  $kgC(kgN)^{-1}$ ) using eddy flux  
405 observations reported by Magnani et al. (2007). The mean  $dC_{NEP}/dN_{deposition}$  using the EU N  
406 deposition from the clm4cn model (24.5  $kgC(kgN)^{-1}$ ) was 50% less than the lower bound  
407 reported in Sutton et al. (2008).

408

### 409 ***3.3 Mechanisms explaining the increased responsiveness of the modified model to N***

#### 410 ***deposition and fertilization***

411 The mean retention of N deposition within ecosystem was larger in clm4mod than  
412 clm4cn, mirroring the  $dC_{ACI}/dN_{deposition}$  response. Across all five sites, the mean retention of N  
413 deposition was higher in the clm4mod model than the clm4cn model (Fig. 3). In both the clm4cn  
414 and clm4mod, ecosystem N retention (Fig. 3; sum of N recovery in vegetation and soil) was  
415 lower when integrated over a 34-yr period (1970–2004: clm4cn = 51 %; clm4mod = 81 %) than

416 a 4-yr period (2000–2004: clm4cn = 55 %, clm4mod = 94 %). The fate of N retained in the  
417 ecosystem was predominately in soil organic matter in both models and at both time scales (Fig.  
418 3). However, the proportion of N deposition retained in both vegetation and soil was higher in  
419 the clm4mod than the clm4cn model (Fig. 3) with particularly large increases in N retained in  
420 soil. The response to N deposition ( $dC_{ACI}/dN_{deposition}$ ) was most sensitive to the use of a  
421 Michaelis-Menten approach to plant N uptake in the clm4mod that describes N up- take as a  
422 function of soil inorganic N and fine root C, rather than the approach in clm4cn that represents N  
423 uptake as a function of photosynthesis and the N required by stoichiometric constraints (Model  
424 6; Table 3; Fig. 4). Implementing the Michaelis-Menten approach increased the  $dC_{ACI}/dN_{deposition}$   
425 by  $27 \text{ kgC}(\text{kgN})^{-1}$ . The next largest sensitivity was associated with the pathway of N loss (Model  
426 10; Table 4; Fig. 4); the removal of the N gas loss process simulated in clm4cn as 1 % of net N  
427 mineralization decreased the  $dC_{ACI}/dN_{deposition}$  by  $15 \text{ kgC}(\text{kgN})^{-1}$  (Fig. 4). Because we increased  
428 the proportion of excess N that was denitrified to maintain the same total N loss at pre-industrial  
429 steady-state, the change in  $dC_{ACI}/dN_{deposition}$  was associated with modifying the particular  
430 pathways of N loss, rather than being associated with a modification to the total pre-industrial N  
431 export.

432 The implementation of a less open N cycle with lower N input and outputs increased  
433  $dC_{ACI}/dN_{deposition}$  by  $6 \text{ kgC}(\text{kgN})^{-1}$  (Model 4; Table 4; Fig. 4).  $dC_{ACI}/dN_{deposition}$  increased by 8  
434  $\text{kgC}(\text{kgN})^{-1}$  in the simulation that isolated the influence of reduced denitrification on the  
435 response to N deposition (Model 5; Table 4; Fig. 4). This simulation had the same pre-industrial  
436 primary productivity as the simulation with Michaelis- Menten plant N uptake (Model 6) but  
437 without the Michelis-Menten uptake mechanism (same model structure as Model 4 but with a  
438 lower denitrification parameter). In contrast, allowing microbial immobilization to have first

439 access to soil inorganic N (Model 8; Fig. 1; Fig. 4) and implementing a representation of  
440 nitrification with separated  $\text{NH}_4^+$  and  $\text{NO}_3^-$  pools (Model 11; Table 4; Fig. 4) decreased  
441  $dC_{\text{ACI}}/dN_{\text{deposition}}$  by 7 and 5  $\text{kgC}(\text{kgN})^{-1}$ , respectively. All other processes described in Table 4  
442 had little impact on  $dC_{\text{ACI}}/dN_{\text{deposition}}$  ( $< 3 \text{ kgC}(\text{kgN})^{-1}$ ).

443

### 444 **3.4. Synergy between N deposition and atmospheric $\text{CO}_2$**

445 Averaged across all five sites, the clm4mod and clm4cn models predicted similar  
446 increases in NPP over pre-industrial NPP, attributed to the increase in both N deposition and  
447 atmospheric  $\text{CO}_2$  over the period from 1850 to 2004 (clm4cn =  $84.1 \text{ gCm}^{-2} \text{ yr}^{-1}$ ; clm4mod =  
448  $87.2 \text{ gCm}^{-2} \text{ yr}^{-1}$ ; 1994–2004; Fig. 5a). However, the relative contribution of N deposition and  
449  $\text{CO}_2$  fertilization differed strongly between the two models (Fig. 5b). The increase in NPP  
450 predicted by the clm4cn model was comprised of a pure N deposition response (46 %) and a pure  
451  $\text{CO}_2$  fertilization response (57 %) that were largely independent of each other, as the synergy  
452 only explained 7% of the NPP change (Fig. 5b). In contrast, the pure  $\text{CO}_2$  fertilization response  
453 in the clm4mod model was small (12%) while the majority of the NPP increase was explained by  
454 a pure N deposition response (58 %) and a synergy between N deposition and rising  $\text{CO}_2$  (30 %;  
455 Fig. 5b).

456

## 457 **4.0 Discussion**

458 Overall, our modifications to the CLM-CN 4.0 substantially improved predictions of C  
459 storage in response to historical N deposition. The modifications in the clm4mod model  
460 increased the aboveground C increment response to historical N deposition by 144 % compared  
461 to the clm4cn model and compared more closely with observations across N deposition gradients

462 in the Northeastern US and Western Europe. Modifications to the representation of plant N  
463 uptake and pathways of N losses were the most important mechanisms that were responsible for  
464 the increased aboveground C increment response to N deposition in the clm4mod model. The net  
465 effect of the modifications increased ecosystem retention of N deposition and increased synergy  
466 between N deposition and fertilization from elevated atmospheric CO<sub>2</sub>.

467

#### 468 ***4.1 Response to historical N deposition***

469         The response to N deposition was most sensitive to the implementation of a Michaelis-  
470 Menten approach to plant N uptake (Model 6; Fig. 4). By adding the Michaelis-Menten uptake,  
471 plant N uptake depended on the fine root C stock, the soil inorganic N concentration,  
472 temperature, and N status of the plant rather than the N demand set by photosynthesis, allocation,  
473 and C : N ratios of the allocated tissues. The Michaelis-Menten uptake increased the N  
474 deposition response by increasing N retention (2000–2004) by 9%, likely due to greater ability of  
475 plants to compete against N loss processes. The increased N retention was also associated with a  
476 lower parameter value for the proportion of excess N lost as N gas in the model with Michaelis-  
477 Menten uptake (Model 6; Fig. 4). The lower parameter was necessary to yield the same steady-  
478 state N gas loss at pre-industrial conditions in the Michaelis-Menten uptake simulation because  
479 this approach to N uptake requires larger stocks of soil inorganic N to retain the same N uptake  
480 rate as a model without Michaelis-Menten uptake. The lower N gas loss parameter likely  
481 allowed more N deposition to be retained in the ecosystem in the transient simulations.  
482 Michaelis-Menten uptake also increased soil inorganic N stocks necessary to yield the same N  
483 uptake as the clm4cn model. This reduced the competition between plants and immobilization,  
484 thus increasing the total assimilation into N soil organic matter. A range of N uptake approaches

485 are used in ecosystem and global biogeochemical models suggesting the choice of how N uptake  
486 is represented can strongly influence the sensitivity to N deposition in the model.

487         The N deposition response was also sensitive to the pathway of N loss (Model 10; Fig. 4).  
488 Broadly there are two pathways of gas and leaching N loss used in ecosystem and global  
489 biogeochemical models: turnover-dependent and concentration-dependent pathways (Menge,  
490 2011). Turnover dependent pathways refer to N losses that are proportional to the turnover of an  
491 internal nitrogen pool (i.e. litter N, soil organic N, or vegetation N). Concentration-dependent  
492 pathways refer to N losses that are proportional to the concentration of soil inorganic N and  
493 typically only occur under periods of N saturation. As an example of a turnover-dependent  
494 pathway, the clm4cn has a pathway of N gas loss that is proportional to the net N mineralization  
495 associated with soil organic matter turnover (1 % of net N mineralization). The sensitivity  
496 analysis found that substituting concentration- dependent for a turnover-dependent gas loss  
497 pathway decreased the plant C response to N deposition; the substitution occurred through  
498 removing the N gas loss associated with net mineralization and compensating the reduced N loss  
499 by increasing denitrification of excess soil inorganic N, a concentration-dependent pathway  
500 (Model 10; Fig. 4). Similarly, DON leaching in the clm4mod is another example of a turnover-  
501 dependent loss pathway (Menge, 2011) because it is proportional to the decomposition of litter.  
502 We found that by including DON leaching and reducing the denitrification of  $\text{NO}_3^-$  (Model 14;  
503 Fig. 4), the plant C response to N deposition response increased. Overall it is clear that models  
504 with a greater role of turnover-dependent N losses had a greater plant C response to N deposition  
505 than models with N loss dominated concentration- dependent pathways; We hypothesize that  
506 concentration-dependent pathways are able to more quickly lose N deposition when N

507 availability is elevated, while turnover-dependent pathways increase N retention by requiring N  
508 deposition to cycle through the plants and soil before being lost.

509 Another important change to the CLM-CN that was responsible for increasing the  
510  $dC_{ACI}/dN_{deposition}$  response in the clm4mod model was the implementation of a less open N cycle  
511 (Model 4; Fig. 4). N cycles range from being open to closed depending on the importance of the  
512 inputs and outputs of N relative to the internal N cycling fluxes. For example, at steady state in  
513 global biogeochemical models, all N fixation inputs are balanced by N losses from the  
514 ecosystem. As such, assuming equal stocks of N, models with larger N inputs will have larger  
515 outputs at steady state, resulting in faster and a more open N cycle. The degree of openness of  
516 the N cycle in an ecosystem is not explicitly specified in ecosystem models; rather, it is  
517 controlled by the balance of N inputs to outputs. The magnitude and mechanisms governing N  
518 fixation and N losses vary widely among global biogeochemical models, likely indicating that  
519 the degree of openness of the N cycle also varies among models. Indeed, in comparison to  
520 another global biogeochemical model, in the extra-tropics the clm4cn model reports both a  
521 greater role of N availability in the C cycle and greater N fixation (Thornton et al., 2007) than the  
522 LM3V model (Gerber et al., 2010). The LM3V model uses a demand- driven approach to N  
523 fixation that recognizes that N fixation is limited in closed-canopy temperate and boreal forests  
524 (Crews, 1999) rather than the relationship between NPP and N fixation, derived from Cleveland  
525 et al. (1999), used in the clm4cn.

526 The net effect of all the modifications (Models 2–15) was to increase the  $dC_{ACI}/dN_{deposition}$   
527 response to N deposition by increasing the retention of N deposition within the ecosystem, as  
528 greater long-term N retention increased the availability of N to plants and allowed them to  
529 respond to rising atmospheric CO<sub>2</sub>. Retention of N deposition within the five simulated forests at

530 the 4 to 30 yr time horizon in the clm4mod model was between 81 and 95 %, and only 51 to 55  
531 % in the clm4cn model. The higher N retention rate in the clm4mod model than the clm4cn  
532 model better matches observations from field tracer experiments in which isotopically labeled N  
533 ( $^{15}\text{N}$ ) was added to forests and total isotope recovery was used to measure N retention  
534 (Nadelhoffer et al., 1999, 2004; Templer et al., 2012). At the Harvard Forest site, retention of  
535 added  $^{15}\text{N}$  in two experiments after 7 yr ranged from 88 to 100%, which compares well to  
536 clm4mod (Nadelhoffer et al., 2004; treatments with no additional N fertilization added). In  
537 contrast, a  $^{15}\text{N}$  tracer experiment at one of the Michigan sites (MI-B) only recovered 17.5% of  
538 added  $^{15}\text{N}$ , which is substantially lower than both the clm4cn and clm4mod models (Zak et al.,  
539 2004). This tracer study differed markedly from most others. That is, a meta-analysis of  $^{15}\text{N}$   
540 experiments found that approximately 78 % of added N was recovered in 11 temperate  
541 deciduous broadleaf forest ecosystems, and 75 % was recovered across all forests (Templer et  
542 al., 2012). A similar analysis on nine sites, many of which were included in the meta-analysis  
543 conducted by Templer et al. (2012), found 90 % recovery of  $^{15}\text{N}$  after 1–3 yr of addition  
544 (Nadelhoffer et al., 1999). Overall, on the balance of evidence, the increased N retention in the  
545 clm4mod better reflects the  $^{15}\text{N}$  retention observed in most temperate forest tracer studies.

546 The net effect of all the modifications also increased synergy between N deposition and  
547 atmospheric  $\text{CO}_2$  in the clm4mod. Surprisingly, when averaged across all five sites, the increase  
548 in NPP resulting from N deposition was similar between the two models in the simulations when  
549 rising atmospheric  $\text{CO}_2$  was not included (Fig. 5b). However, when rising atmospheric  $\text{CO}_2$  was  
550 included, the synergy between N deposition and  $\text{CO}_2$  led to a larger total response to N  
551 deposition in the clm4mod model than clm4cn model. This increase in synergy was due to the  
552 greater retention of N deposition in the clm4mod than the clm4cn model. The reduction of N

553 limitation resulting from the additional ecosystem N allowed an enhancement of photosynthesis  
554 by the next most limiting resource in the model, CO<sub>2</sub>, whereas the clm4cn model remained N  
555 limited. Consequently, the clm4mod model exhibited CO<sub>2</sub> fertilization in the simulation with  
556 both rising CO<sub>2</sub> and N deposition, enabled in part by rising N deposition. Overall, the increased  
557 enhancement of NPP due to synergy between N deposition and CO<sub>2</sub> from 7 % in clm4cn to 30 %  
558 in clm4mod led to better correspondence with other studies: a 25 % synergistic effect measured  
559 in field-based CO<sub>2</sub> fertilization experiment of needle leaf pine forest (Oren et al., 2001) and a 28  
560 % synergistic effect reported by a global modeling analysis using BIOME-BGC model  
561 (Churkina et al., 2009). It is important to note that the sites used in the analysis were not in arid  
562 environments where soil moisture can regulate N limitation and CO<sub>2</sub> fertilization (Felzer et. al  
563 2011). Future studies can expand the analysis beyond temperate deciduous forests and explore  
564 how the set of model modifications influences C-N interactions in a range of ecosystem types.

565

#### 566 ***4.2 Response to N fertilization***

567 Overall, the clm4mod either improved or had no impact on the comparison to  
568 observations from N fertilization experiments, depending on the metric used in the model  
569 evaluation. In this study, we used three metrics to compare model predictions from clm4cn and  
570 clm4mod to N fertilization experiments, with each metric testing different aspects of model  
571 representation of N limitation. The first metric, the increase of ANPP in response to N  
572 fertilization, tested the productivity response, particularly wood and leaf production, to N  
573 fertilization. The ANPP response metric showed that, on average, the clm4mod corresponded  
574 better to observations than the clm4cn, with particular improvements at the Harvard Forest site.  
575 The improved correspondence at the Harvard Forest site was attributable to both a decrease in

576 potential ANPP when N was not limiting (see discussion below) and an increase in the ANPP of  
577 the control treatment.

578         However, the ANPP increase metric did not include changes in mortality that were  
579 included in the second metric,  $dC_{ACI}/dN_{fertilization}$  (i.e. the change in standing stock of  
580 aboveground C between years). The model modifications did not have an impact on the mean  
581  $dC_{ACI}/dN_{fertilization}$  response to N fertilization and both the clm4mod and clm4cn predicted larger  
582  $dC_{ACI}/dN_{fertilization}$  than observed. The two key differences between the ANPP responses and  
583  $dC_{ACI}/dN_{fertilization}$  to fertilization were: one, increased mortality from N fertilization in the field  
584 studies may not decrease ANPP but will decrease  $dC/dN$ , and two, increased foliar production in  
585 the models increases ANPP without directly increasing  $dC/dN$ . Neither model included  
586 mechanisms through which elevated N inputs could increase tree mortality and tissue turnover,  
587 and both models predicted an increase in foliar productivity not found in the fertilization  
588 experiments (Magill et al., 2004; Pregitzer et al., 2008).

589         The productivity of the N fertilized treatment alone is the third metric describing how  
590 productivity responded to N fertilization. The ANPP in the field fertilized plots can be viewed  
591 as an approximation of the N unlimited productivity, assuming the fertilization level was high  
592 enough to meet plant demand and low enough to prevent negative effects of soil acidification. If  
593 so, the measure of N unlimited productivity is a metric that does not test the model response to N  
594 per se; rather, it tests the representation of the next most limiting resource in the models.

595         Averaged across all six fertilization experiments, the clm4mod model did improve predictions of  
596 ANPP in the fertilized treatment. Higher ANPP in the fertilization treatments in the clm4mod  
597 model than the clm4cn model was surprising because the clm4mod model included changes to  
598 the CLM-CN 4.0, described in Bonan et al. (2012), that decreased canopy level photosynthesis.

599 Therefore, including the modifications that lowered photosynthesis should have decreased the  
600 simulated productivity when N limitation was relieved. However, a key difference between the  
601 clm4cn and clm4mod models was that the simulated N fertilization experiments relieved N  
602 limitation in the clm4mod simulations while it did not in the clm4cn simulations, potentially due  
603 to the high ecosystem retention of N in the clm4mod. Table 4 shows that the N unlimited ANPP  
604 in the clm4mod did not differ from the ANPP in the fertilization simulations, while the N  
605 unlimited ANPP in the clm4cn was 77 % greater than the ANPP in the fertilization simulations.  
606 The low ecosystem retention of N in the clm4cn maintained N limitation even at fertilization  
607 levels over double net N mineralization rates.

608 One important caveat when using N fertilization studies to evaluate C-N interactions in  
609 models is accounting for potential harmful affects of fertilization-induced inorganic N leaching  
610 on base cation exchange and forest health (i.e., Hogberg et al. 2007; Wallace et al. 2007). The  
611 clm4mod simulates greater inorganic N leaching than the clm4cn (Table A2). This increased N  
612 leached would likely lead to greater cation depletion and acidification. However, neither clm4cn  
613 nor clm4mod simulates other element cycles, such as calcium and aluminum, nor links between  
614 soil solution chemistry and growth or mortality needed to mechanistically represent the negative  
615 impacts of N leaching on forest productivity and tree survival. While some N fertilization  
616 studies across the Northeast US have reported harmful impacts of N fertilization (e.g. Wallace et  
617 al. 2007; Magill et al. 2004 – Pine site; McNulty et al. 2005), all of the studies used in this  
618 analysis yielded neutral or positive effects of fertilization on ANPP and C accumulation (Magill  
619 et al. 2004; Pregitzer et al. 2008).

620

### 621 ***4.3 Implications***

622 The set of model simulations presented in this study also provide insight into the  
623 observational data. The reported  $dC/dN$  was lower for the fertilization experiments than for the  
624  $dC/dN$  from N deposition gradients (Table 3). Furthermore, there was a lower reported  $dC/dN$  in  
625 gradient studies in Europe than in the US (Table 3). Despite these disparities, we show that the  
626 reported  $dC/dN$  data are reconcilable if the differences in the magnitude and time-scale of N  
627 additions are considered. The clm4mod model simulations overlapped or were near the  
628 uncertainty bounds in the observations across the different times scales and magnitudes of N  
629 additions. The N deposition gradient studies measured the response to lower N inputs over a  
630 longer period of time (decades to a century), while the N fertilization experiments measured the  
631 response to higher inputs over a shorter time scale (years to decades). This indicates that the  
632 differences in ecosystem N use efficiencies reported for different fertilization studies and N  
633 deposition gradients may be explained by differences in the magnitude and time scale of N  
634 addition.

635 Overall, the substantial increase in C storage response to N deposition that occurred as a  
636 result of modifications to the CLM-CN 4.0 model resulted in a better comparison to observations  
637 of temperate broadleaf forest growth across N deposition gradients and to N fertilization  
638 experiments. The improved sensitivity to N inputs was driven primary by altering the  
639 mechanisms governing plant N uptake and the pathways of N loss. At the global scale, the  
640 modifications to CLM-CN presented are likely to improve the model correspondence to the  
641 globally distributed set of N fertilization experiments,  $^{15}N$  tracer studies, and small catchment N  
642 budgets that have been previously used to benchmark global biogeochemical models (Thomas,  
643 2013). Furthermore, we show that due to non-linearity in ecosystem response to N addition,  
644 testing models with both the response to gradual increases in N inputs over decades (N

645 deposition) and N pulse additions of N over multiple years (N fertilization) allows for greater  
646 understanding of the mechanisms governing C-N coupling.

647

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657 data cited in this study.

658 **Appendix A**

659 *clm4mod model description*

660 The clm4mod model includes changes to the canopy scaling of photosynthesis, maximum  
661 photosynthetic rates, radiative transfer, leaf photosynthesis, and stomatal conductance described  
662 in Bonan et al. (2012) and (2011). Additional modifications are described below.

663

664 *AI Nitrogen uptake by plants*

665 Plant uptake of inorganic nitrogen (N) in CLM-CN 4.0 (“clm4cn”) is based on the N required to  
666 match the demand set by N unlimited photosynthesis (i.e. potential gross primary productivity)  
667 and plant tissue C:N stoichiometric constraints. In the clm4cn model, N uptake is independent of  
668 allocation to fine root mass. The clm4mod model represents N uptake as a function of fine root  
669 carbon (C) mass ( $C_{\text{root}}$ ), soil inorganic N availability ( $\text{NH}_{4,\text{av}} + \text{NO}_{3,\text{av}}$ ), soil temperature ( $f(T)$ ),  
670 and plant demand for N ( $f(N_{\text{labile}})$ ).

671

$$U_{N,\text{plant}} = V_{n,\text{max}} \frac{\text{NH}_{4,\text{av}} + \text{NO}_{3,\text{av}}}{(\text{NH}_{4,\text{av}} + \text{NO}_{3,\text{av}}) + K_{\text{min}}} C_{\text{root}} f(N_{\text{labile}}) f(T)$$

672

673 where  $f(N_{\text{labile}})$  down regulates the uptake capacity based on the stock of labile N in the plant. As  
674  $N_{\text{labile}}$  approaches one year’s worth of N turnover of live tissue ( $x$ ; leaves, fine roots, and live  
675 wood), the maximum uptake capacity decreases. The down regulation function is based on Aber  
676 et al. (1997).

677

$$f(N_{\text{labile}}) = \sqrt{1 - \frac{N_{\text{labile}}}{x}}$$

678

679 The temperature function  $f(T)$ , is the same function governing soil decomposition, nitrification  
680 (see below), and denitrification (see below).  $\text{NH}_{4,\text{av}}^+$   $\text{NO}_{3,\text{av}}$  are the concentrations of ammonium  
681 and nitrate that are available for plant uptake (see below).  $V_{\text{nmax}}$  is the maximum uptake capacity  
682 at 25 °C when N demand was large (i.e.  $f(\text{N}_{\text{labile}})=1$ ).  $K_{\text{min}}$  is the half saturation constant for plant  
683 uptake.

684 The availability of N within the plant for growth is buffered so that a proportion (2 %) of  
685 the labile N pool is available to build plant tissue.

686

### 687 *A2 Fine root turnover*

688 In the clm4cn model, the turnover of fine root C is linked to the turnover of leaf C. In the  
689 clm4mod version, the root turnover is an explicit parameter and is decoupled from leaf turnover.  
690 Root turnover occurs throughout the year, rather than only when leaves senesce. Root turnover is  
691 maintained at the same rate as the clm4cn model for all species ( $1.0\text{yr}^{-1}$ ), except boreal and  
692 temperate needleleaf species, which has a turnover of  $0.3\text{ yr}^{-1}$  based on White et al. (2000).  
693 Decoupling fine root turnover from leaf turnover allows fine roots to be present throughout the  
694 year and permits a fast response of plant N uptake in the spring.

695

### 696 *A3 Soil N dynamics*

697 The clm4mod version of the model includes soil inorganic pools of  $\text{NH}_4^+$  and  $\text{NO}_3^-$ , rather than  
698 a single inorganic N pool, as implemented in the clm4cn model. The  $\text{NH}_4^+$  pool is buffered to  
699 represent an exchangeable pool and a pool in solution that is available for plants, immobilization,  
700 nitrification, and leaching.

$$NH_{4,av} = b_{NH_4}NH_4$$

701  
 702 where  $b_{NH_4}$  is assumed to be 10 % of total soil  $NH_4^+$  (Gerber et al., 2010). Constant buffering  
 703 capacity is a first approximation for a more complex process of resorption/desorption. Future  
 704 model development could parameterize non-linearity into the buffering capacity that is a function  
 705 of the total soil organic matter and the bulk density of the soil.  $NO_3^-$  is assumed to have no  
 706 buffering capacity in the soil therefore  
 707  $NO_{3,av}=NO_3^-$ .

708  
 709 ***A4 Internal N cycling***

710 The clm4mod model assumes that microbes have priority for soil inorganic N to meet the  
 711 immobilization demand. Plant uptake and immobilization of N is divided between  $NH_4^+$  and  
 712  $NO_3^-$  in proportion to the availability of each N species ( $NH_{4,av}$ ;  $NO_{3,av}$ ). The conversion of  $NH_4^+$   
 713 to  $NO_3^-$  (nitrification) is represented as function of net N mineralization,  $NH_4^+$  availability,  
 714 temperature, and water availability based on Parton et al. (2001). Nitrifiers are assumed to be less  
 715 competitive for  $NH_4^+$  than plants and immobilization into soil organic matter

$$N_{nitr} = K_{nitr1}N_{mineralization} + K_{nitr2}NH_{av,nitr}f(T)f(W)$$

717  
 718 where the  $NH_{4,av,nitr}$  is the  $NH_4^+$  available for nitrification

$$NH_{4,av,nitr} = NH_{4,av} - U_{NH_4,plant} - U_{NH_4,soil}$$

720

721  $K_{nitr1}$ ,  $K_{nitr2}$ ,  $f(T)$ , and  $f(W)$  are the proportion of net mineralization that is nitrified, maximum  
 722 nitrification rate ( $s^{-1}$ ) based on available  $NH_{4,av,nitr}$ , temperature modifier, and water availability  
 723 modifier, respectively.  $f(T)$  and  $f(W)$  are the same temperature and water functions that  
 724 modified decomposition and plant N uptake (Thornton et al., 2007, 2009). The clm4mod model  
 725 ignores the effect of pH on nitrification (Parton et al., 1996). A proportion (0.02) of the  
 726 nitrification is lost to  $N_2O$  and not converted to  $NO_3^-$  (Parton et al., 2001).

727 ***A5 Inorganic N loss***

728 In the clm4mod model, the leaching of  $NH_4^+$  is a function of the soil water drainage and  
 729  $NH_{4,av}$ , minus the  $NH_4^+$  uptake by plants, immobilization, and nitrifiers during the model time  
 730 step. Likewise, the leaching of  $NO_3^-$  is a function of the soil water drainage and  $NO_3^-$ , minus the  
 731  $NO_3^-$  taken up by plants and immobilization during the model timestep.

732 The loss of  $NO_3^-$  through denitrification is modeled as a function of the available nitrate  
 733  $NO_{3,av}$  (minus nitrate uptake by plants, immobilization, and leaching), the ratio of soil water to  
 734 saturated soil water, and total respiration from soil organic matter decomposition (a proxy for  
 735 microbial activity and oxygen composition;  $CO_{2,soil}$ ). The representation is described in  
 736 Bradbury et al. (1993) and Yang et al. (2010).

737

$$N_{denit} = DCO_{2,soil}NO_{3,av}\frac{W}{W_s}$$

738

739 where  $D$  is the denitrification rate per g of  $CO_2$  respiration of soil organic matter, the  $W$  is soil  
 740 water in the top five soil layers and  $W_s$  is water holding capacity at saturation.

741

742 ***A6 Biological N fixation***

743 The biological N fixation in temperate and boreal forests is modified to better represent  
 744 observations that N fixing tree species are largely absent from mid- to late- successional forests,  
 745 but can be present in earlier successional forests (Crews, 1999). Based on biome specific data on  
 746 N fixation and evapotranspiration in Cleveland et al. (1999), annual non-symbiotic fixation is a  
 747 function of annual evapotranspiration and occurs in all ecosystems:

$$N_{fix,nonsym} = 0.0006AAET + 0.0117$$

748  
 749 where AAET is annual evapotranspiration of the previous year. Non-symbiotic N fixation  
 750 is added to the soil  $NH_4^+$  pool.

751 Symbiotic N fixation is function of the plant functional type, proportion of grid cell  
 752 occupied by the plant function type, leaf area index, and annual evapotranspiration. Symbolic N  
 753 fixed is determined by subtracting the non-symbiotic relationship described above from the  
 754 relationship between total N fixation and evapotranspiration in Cleveland et al. (1999: central  
 755 relationship; Figure A1). In grasslands and tropical ecosystems, symbiotic N fixation is added to  
 756 the plant labile N pool. In temperate and boreal forests with leaf area index  $< 1$ , symbiotic N  
 757 fixation is also added to the plant labile N pool, as they are assumed to be early successional and  
 758 contain some N fixing plants. No symbiotic N fixation is added to the plant labile N pool in  
 759 temperate and boreal forest with leaf area index  $\geq 1$ . The overall relationship describing  
 760 symbiotic N fixation is

$$N_{fix,sym} = \begin{cases} w_{pft}(0.0018AAET - 0.0289) & \text{temperate or boreal trees and LAI} < 1 \\ 0 & \text{temperate or boreal trees and LAI} \geq 1 \\ w_{pft}(0.0018AAET - 0.0289) & \text{all other pfts} \end{cases}$$

762

763 where  $w_{pft}$  is the proportion of the grid cell occupied by the plant functional type. N fixation is  
764 constrained to be  $\geq 0 \text{ g N m}^{-2} \text{ yr}^{-1}$

765

#### 766 ***A7 Dissolved organic matter dynamics***

767 Dissolved organic N losses can be important for maintaining N limitation (Menge, 2011),  
768 especially in ecosystems with low anthropogenic N inputs (Hedin et al., 1995). A simple  
769 representation of the production and leaching of dissolved organic matter is added to the CLM-  
770 CN. As presented in Gerber et al. (2010), DOM production is assumed to be a fraction of the  
771 turnover of the structural litter pool. In the clm4mod model, DOM production is parameterized to  
772 be 1.5% ( $p_{dom}$ ) of the transfer from the lignin-based litter pool (Litter 3) to its receiving soil  
773 organic matter pool (SOM 3). So that DOM production does not alter the decomposition rate of  
774 the litter pool, the C : N ratio of DOM must equal that of receiving SOM pool. Therefore, the C :  
775 N of DOM is set to be 10 : 1 ( $CN_{DOM}$ ). Future research should focus on improving the  
776 representation of DOM production so that the C : N better match observations (i.e. more C  
777 produced for the same N production). However, increasing the C : N ratio of the DOM while  
778 maintaining the same production of DON will likely have little effect on N cycle. All DOM  
779 produced is assumed to be unavailable for plant uptake and immobilization. The leaching of  
780 DOM is based on the water drainage and the total soil water.

781 **Appendix B. Model descriptions for intermediary models used in sensitivity analysis**

782 The series of intermediary models using to the sensitivity analysis are described below:

783

784 **Model 1 (*clm4cn*):** Model 1 is the same as the *clm4cn* model described in the main text.

785

786 **Model 2 (*Model 1 + Multi-layer canopy*):** Model 2 adds modifications to the canopy  
787 scaling of photosynthesis using a multi-layer approach with updated maximum photosynthetic  
788 rates, radiative transfer, leaf photosynthesis, stomatal conductance described in Bonan et al.  
789 (2012) and (2011). Model 2 uses Kattge et al. (2009) values for  $V_{cmax}$  (see main text).

790

791 **Model 3 (*Model 2 + soil buffering*):** Model 3 adds a soil buffering parameter to Model  
792 2. The soil buffering assumes that 19 % of the generic soil inorganic N pool is available for plant  
793 uptake, immobilization into soil organic matter, leaching, and denitrification. The parameter (19  
794 %) was chosen to represent a distribution of the generic soil inorganic N pool into 90%  $NH_4^+$  and  
795 10%  $NO_3^-$  with the same buffering parameters used in the *clm4mod* model. By assuming 10 %  
796 of N is  $NO_3^-$ , the model is consistent with the preexisting parameterization of N available for  
797 leaching losses in Models 1 and 2.

798

799 **Model 4 (*Model 3 + less open N cycle*):** Model 4 creates a less open N cycle by reducing N  
800 fixation (see Appendix A6). To isolate the impact of a less open N cycle rather from the  
801 sensitivity to N reduced N inputs, we decreased the proportion of excess soil inorganic N that  
802 was loss as denitrification (from  $0.50 \text{ day}^{-1}$  to  $0.0475 \text{ day}^{-1}$ ) to maintain the same pre-industrial  
803 productivity as Model 3.

804

805 **Model 5 (Model 4 + reduced denitrification):** Model 5 reduces the proportion of excess N that is  
806 lost through denitrification ( $0.013 \text{ day}^{-1}$ ), setting the pre-industrial steady state equal to  
807 clm4mod.

808

809 **Model 6 (Model 5 + Michaelis-Menten approach to plant N uptake):** Model 5 adds the  
810 Michaelis-Menten approach (described in Appendix A1) for simulating plant N uptake. The  
811 parameters used are listed in Table A1. N uptake is only allowed to occur when vegetation is  
812 photosynthesizing, thus matching the diurnal cycle of N uptake found in Models 1–5. To  
813 maintain the same pre-industrial steady state as model clm4mod, the proportion of excess N that  
814 is denitrified was set to  $0.0011 \text{ day}^{-1}$ .

815

816

817 **Model 7 (Model 6 + N uptake throughout day):** Model 7 is equivalent to Model 6 except that N  
818 uptake is allowed to occur throughout the day when leaves are present. To maintain the same  
819 pre-industrial steady state as clm4mod, the proportion of excess N that is denitrified was set to  
820  $0.00165 \text{ day}^{-1}$ .

821

822 **Model 8 (Model 7 + priority to microbial immobilization):** In Models 1–7 competition between  
823 plant N uptake and microbial N uptake was based on the relative N demand by the two  
824 processes. Soil inorganic N is distributed based on the relative demand. In the Model 8,  
825 microbial immobilization is given first priority to the soil inorganic N required meet its demand

826 for N. To maintain the same pre-industrial steady state as clm4mod, the proportion of excess N  
827 that is denitrified was set to  $0.0022 \text{ day}^{-1}$ .

828  
829 **Model 9 (Model 8 + temperate and soil water limitation of denitrification):** In Models 1–8, the  
830 proportion of excess N that is denitrified depended only available N. Model 9 limits  
831 denitrification based on soil water and soil temperature (see Appendix A5). To maintain the  
832 same pre-industrial steady state as clm4mod, the proportion of excess N that is denitrified was  
833 set to  $0.0011 \text{ day}^{-1}$ .

834  
835  
836 **Model 10 (Model 9 + removed N gas loss that is proportion to net N mineralization):** Model  
837 10 removes the turnover-dependent pathway of N loss by setting the proportion of net N  
838 mineralization lost as N gas to zero. To maintain the same pre-industrial steady state as  
839 clm4mod, the proportion of excess N that is denitrified, a concentration-dependent N loss  
840 pathway, was set to  $0.029 \text{ day}^{-1}$ .

841  
842 **Model 11 (Model 10 +  $\text{NH}_4^+$  pool,  $\text{NO}_3^-$  pool, and nitrification):** Model 11 separates  
843 the generic soil inorganic N pool in Models 1–10 into  $\text{NH}_4^+$  and  $\text{NO}_3^-$  pools. Model 11 also adds  
844 the nitrification process described in Appendix A4. To maintain the same pre-industrial steady  
845 state as clm4mod, the proportion of  $\text{NO}_3^-$  that is denitrified was set to  $0.17 \text{ day}^{-1}$ .

846  
847 **Model 12 (Model 11 + Denitrification a function of heterotrophic respiration):** Model replaces  
848 the temperature limitation of denitrification with a parameter that sets denitrification as a

849 proportion of heterotrophic respiration (see Appendix A5). The proportion is set to  $0.097 \text{ (g C)}^{-1}$   
850 to maintain the same pre-industrial steady state as clm4mod.

851  
852 **Model 13 (Model 12 +  $N_2O$  loss that is proportion to nitrification):** Model 13 adds a  $N_2O$  loss  
853 process where  $N_2O$  production is a proportion (0.2) of nitrification (see Appendix A4). To  
854 maintain the same pre-industrial steady state as clm4mod, the parameter describing the  
855 denitrification rate per g of soil respiration  $0.077(\text{gC})^{-1}$ .

856  
857 **Model 14 (Model 13 + DON leaching):** Model 14 adds the production and leaching of dissolved  
858 organic C and N (see Appendix A6). To maintain the same pre-industrial steady state as  
859 clm4mod, the parameter describing the denitrification rate per g of soil  
860 respiration was set to  $0.05(\text{gC})^{-1}$ .

861  
862 **Model 15 (Model 14 + plant N buffering, clm4mod):** Model 15 includes all the  
863 changes described for the clm4mod model (main text and Appendix A). It adds to Model 14 a  
864 plant labile N pool that buffers the availability of N to meet the N demand set by the  
865 photosynthetic rate, tissue allocation, and tissue C : N ratios (see Appendix A1).

866

867

868

869

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## Tables

Table 1: Descriptions of sites used in model simulations.

Site	Harvard Forest	Michigan gradient A	Michigan gradient B	Michigan gradient C	Michigan gradient D
Code	HF	MI-A	MI-B	MI-C	MI-D
Latitude (N)	42° 30' N	46° 52'	45° 33'	44° 23'	43° 40'
Longitude (W)	72° 10' W	88° 53'	84° 51'	85° 50'	86° 09'
Stand Age (years) <sup>^</sup>	52	97	91	92	96
MAT* (°C)	7.1	4.7	6.0	6.9	7.6
MAP <sup>#</sup> (mm)	1120	873	871	888	812
Aboveground carbon (g C m <sup>-2</sup> ) <sup>^</sup>	6597	14,700	14,150	15,800	14,450
Vegetation type	temperate deciduous broadleaf				
Years of experiment	1988-2002	1995-2004	1995-2004	1995-2004	1995-2004
Nitrogen fertilization rate (g N m <sup>-2</sup> yr <sup>-1</sup> )	5 & 15	3	3	3	3
Nitrogen deposition (g N m <sup>-2</sup> yr <sup>-1</sup> )	0.8	0.68	0.91	1.17	1.18
Citation	Magill et al. 2004	Pregitzer et al. 2008			

\* Mean annual temperature

# Mean annual precipitation

<sup>^</sup> Age and carbon stocks in the year 1994

Table 2. Description of simulations and years of driver data used in each simulation

Simulation <sup>1</sup>	N deposition	N fertilization	CO <sub>2</sub> concentration
(1) Control (baseline)	1850-2004	None	1850-2004
(2) Hold N deposition	1850	None	1850-2004
(3) Elevated N deposition; EU N deposition gradient	1850-2004; values more similar to EU deposition levels	None	1850-2004
(4) Hold N deposition and CO <sub>2</sub>	1850	None	1850
(5) Hold CO <sub>2</sub>	1850-2004	None	1850
(6) N fertilization	1850-2004	Yrs. of field study, rate of study (see Table 1)	1850-2004
(7) Non-N limited	1850-2004	Yrs. of field study, rate necessary to match N demand	1850-2004

<sup>1</sup> All simulations included transient land-use with a harvest at the year that yields the observed stand age in 1994 (see Table 1)

Table 3. Model simulations and observations of the vegetation and ecosystem carbon increment response to nitrogen deposition and fertilization. Model uncertainty estimates correspond to variation among sites (1 S.E.).

Response metric	Carbon measurement	Nitrogen input	clm4cn model	clm4mod model	Observations	Citation
$dC_{ACI}/dN_{deposition}$	Aboveground carbon increment	U.S. deposition gradient	14.0±1.1	34.2±3.0	50 (41-66)	Thomas et al. 2010*
$dC_{ACI}/dN_{deposition}$	Aboveground carbon increment	E.U. deposition gradient	11.8±0.8 <sup>#</sup>	26.2±2.6 <sup>#</sup>	15-40	DeVries et al. 2009
$dC_{ACI}/dN_{fertilization}$	Aboveground carbon increment	Fertilization experiments	10.7±1.3	10.6±4.5	4±2.7	Magill et al. 2004, Pregitzer et al. 2008
$dC_{NEP}/dN_{deposition}$	Net ecosystem production	U.S. deposition gradient	30.0±1.8	74.1±8.17	no data	
$dC_{NEP}/dN_{deposition}$	Net ecosystem production	E.U. deposition gradient	24.5±1.5 <sup>#</sup>	49.5±4.4 <sup>#</sup>	50-75	Sutton et al. 2008
$dC_{NEP}/dN_{fertilization}$	Net ecosystem production	Fertilization experiments	27.9±2.8	23.2±9.4	31 ± 10	Magill et al. 2004, Pregitzer et al. 2008

\* The reported 61  $dC/dN$  was divided by a factor of 1.23 to account for unmeasured nitrogen deposition. See Pinder et al. (2012) for more information.

<sup>#</sup> These model simulations used higher N deposition (1994-2004 mean = 2.2 g N m<sup>-2</sup> yr<sup>-1</sup>) to approximate nitrogen deposition in regions that have experience higher historical N deposition loads.

Table 4. Description of model modifications. Letters (a–l) refer to the processes shown in Fig. 1.

Model	Model modifications
Model 1	Same as clm4cn
Model 2	Decreased plant N uptake (d) by reducing the canopy photosynthesis (Bonan et al., 2011, 2012)
Model 3	Buffered the soil inorganic N available for immobilization (c) and plant N uptake (d).
*Model 4	Introduced a less open N cycle by decreasing denitrification (f) and N fixation (h). Maintains same pre-industrial steady-state as model 3.
*Model 5	Decreased denitrification (f) to increase productivity to match pre-industrial steady state of the models 5–15. Isolates the influence of higher baseline (pre-industrial steady-state) primary productivity on the N deposition response.
*Model 6	Replaced (d) with a Michaelis-Menten approach to modeling plant N uptake. N uptake only occurs when the plants are photosynthesizing. Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
Model 7	Same as model 6 except plant N uptake also occurs at night when plants are not photosynthesizing. Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
*Model 8	Altered immobilization (c) and plant N uptake (d) by allowing immobilization to have first access to soil inorganic N. Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
Model 9	Added temperature and soil water limitations to denitrification (f). Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
*Model 10	Removed the N gas loss (g) that was proportional to net N mineralization (b–c). Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
*Model 11	Added $\text{NH}_4^+$ and $\text{NO}_3^-$ pools; added nitrification (j). Adjusted (f) to maintain same pre-industrial steady-state as models 5–14.
Model 12	Replaced temperature limitation of denitrification (f). Now proportional to SON & litter N respiration. Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
Model 13	Added $\text{N}_2\text{O}$ loss (k) that is proportional to nitrification (i). Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
Model 14	Added dissolved organic N leaching (l). Proportional to the turnover of lignin litter pool. Adjusted (f) to maintain same pre-industrial steady-state as models 5–15.
Model 15 (clm4mod)	Added internal plant N pool to buffer plant N demand. Pre-industrial steady-state for the models 5–14 are calibrated to match model 15. Model 15 is equivalent to clm4mod.

\* Denotes the model modifications that had the largest impact on the N deposition response ( $> 3 \text{ dC}_{\text{ACI}}/\text{dN}_{\text{deposition}}$ ). See Fig. 4.

Table 5. Mean annual net primary productivity ( $\text{g C m}^{-2} \text{ yr}^{-1}$ ) in the six nitrogen fertilization experiments at five temperate broadleaf deciduous forests. The field observations are compared to simulations from the clm4cn model, and clm4mod model. Error estimates correspond to variation among sites (1 S.E.).

	Observations	clm4cn	clm4mod
Control ANPP (n = 5)	464 ± 36	352 ± 50	411 ± 28
Fertilized ANPP (n = 6)	504 ± 40	420 ± 41	474 ± 8
$\Delta$ ANPP (n = 6) *	55 ± 8	91 ± 19	57 ± 18
Non-nitrogen limited ANPP (n = 6)	Not measured	742 ± 10	474 ± 8

\*The mean fertilization responses for the observations and model simulations were different than the difference between the mean control and mean fertilization ANPP because two experiments at Harvard Forest shared the same control treatment

Table A1. New parameters introduced to the CLM-CN 4.0.

Parameter	Value	Units	Description	Reference
$V_{nmax}$	$2.7 \cdot 10^{-8}$	$g\ N\ g\ C^{-1}\ sec^{-1}$	Maximum N uptake per unit fine root C at 25C	none
$K_{min}$	1	$gN\ m^{-2}$	Half saturation constant for plant nitrogen uptake	Yang et al. 2010
x	1	proportion	Proportion of 1 years worth of live tissue nitrogen turnover	Gerber et al. 2010
$b_{NH4}$	0.10	proportion	Proportion of soil $NH_4^+$ available for plant uptake, immobilization and loss processed	Gerber et al. 2010
$b_{NO3}$	1	proportion	Proportion of soil $NO_3^-$ available for plant uptake, immobilization and loss processed	Gerber et al. 2010
$b_{nlabile}$	0.02	proportion	Proportion of plant labile nitrogen available to build tissue per 30 minute time step	none
$K_{nitr1}$	0.2	proportion	Proportion of net mineralization that is nitrified	Parton et al. 2000
$K_{nitr2}$	0.1	$day^{-1}$	Maximum proportion of available $NH_4^+$ nitrified	Parton et al. 2000
$K_{n2o}$	0.02	proportion	Proportion of nitrification lost as $N_2O$	Parton et al. 2000
D	0.05	$g\ C^{-1}$	Maximum denitrification rate per g of soil respiration	Bradberry et al. 1993
$p_{DOM}$	0.015	proportion	Proportion of litter mass transferred from litter 3 pool to soil 3 pool that produces dissolved organic carbon and nitrogen	Gerber et al. 2010

Table A2. Mean simulated pre-industrial nitrogen fluxes ( $\text{g N m}^{-2} \text{ yr}^{-1}$ ) average across all five sites in Table 1 ( $\pm 1$  S. E.).

Flux	clm4cn	clm4mod
Nitrogen fixation	$1.3 \pm 0.1$	$0.26 \pm 0.01$
Nitrogen deposition	$0.15 \pm 0.01$	$0.15 \pm 0.01$
Nitrogen gas loss	$1.4 \pm 0.1$	$0.26 \pm 0.01$
Mineral nitrogen leaching	$0 \pm 0$	$0.07 \pm 0.01$
DON leaching	NA	$0.1 \pm 0.004$
Plant nitrogen uptake	$6.2 \pm 0.67$	$6.9 \pm 0.29$
Net nitrogen mineralization	$6.1 \pm 0.66$	$6.5 \pm 0.25$
Nitrification	NA	$3.6 \pm 0.06$

## Figure captions.

Figure 1. Overview of the clm4cn and clm4mod model structure.

Figure 2. Mean annual aboveground net primary production (ANPP) for the five temperate deciduous forests in Table 1. The measured values, clm4cn model predictions, and clm4mod model predictions are shown for (a) the control (non-fertilized) plots and (b) the nitrogen fertilized plots, along with (c) the differences between the control and fertilized treatments. The model simulations include transient nitrogen deposition, atmospheric CO<sub>2</sub>, and land-use. Error bars represent the S. E. reported in Pregitzer et al. (2008).

Figure 3. Model predictions of ecosystem retention of nitrogen deposition, partitioned among soil organic matter and vegetation averaged across all five sites for two time periods: 1970–2004 and 2000–2004. Error bars represent variation among sites (1 S.E.).

Figure 4. The sensitivity of the nitrogen deposition response to key changes in model structure for a single site (Harvard Forest). The figure shows the nitrogen deposition response, expressed as an annual aboveground carbon increment ( $dC_{ACI}/dN_{deposition}$ ) for the clm4cn model (Model 1), clm4mod (Model 15), and intermediary models (Model 2–14). Each model builds on the modifications in the previous models and the difference in  $dC_{ACI}/dN_{deposition}$  between a model and the previous model is shown in black. See Table 3 for a description of the mechanisms isolated in each model.

Figure 5. Model predictions of net primary production (NPP) response to rising nitrogen