Impacts of Human Alteration of the Nitrogen Cycle in the US on Radiative Forcing

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Abstract Nitrogen cycling processes affect radiative forcing directly through emissions of nitrous oxide (N_2O) and indirectly because emissions of nitrogen oxide (NO_x) and ammonia (NH_3) affect atmospheric concentrations of methane (CH_4), carbon dioxide (CO_2), water vapor (H_2O), ozone (O_3) and aerosols. The emissions of N_2O are mostly from agriculture and they contribute to warming on both short and long time scales. The effects of NO_x and NH_3 on CH_4, O_3, and aerosols are complex, and quantification of these effects is difficult. However, the net result on time scales of decades is likely one of cooling, which becomes less significant on longer time scales. Deposition of N onto ecosystems also affects sources and sinks of N_2O, CH_4, and CO_2, but the dominant effect is changes in carbon (C) stocks. Primary productivity in most temperate ecosystems is limited by N, so inputs from atmospheric deposition tend to stimulate plant growth and plant litter production, leading in some cases to significant C sequestration in biomass and soils. The literature reviewed here indicates a range of estimates spanning 20 – 70 kg C sequestered per kg N deposited in forests, which are the dominant C sinks. Most of the sequestration occurs in aboveground forest biomass, with less consistency and lower rates reported for C sequestration in soils. The permanency of the forest biomass sink is uncertain, but data for the fate of forest products in the US indicate that only a small fraction of enhanced forest biomass C is sequestered in long-term harvest products or in unmanaged forests. The net effect of all of these N cycle processes on radiative forcing in the US is probably a modest cooling effect for a 20-year time frame, although the uncertainty of this estimate includes zero net effect, and a modest warming for a 100-year time frame. We know that N-cycling processes are important and that biotic feedbacks to climate change are unlikely to be properly modeled or assessed without including C-N interactions. However, due to the complexity of biological processes involving C-N-climate interactions, biogeochemical models are still poorly constrained with

respect to ecosystem responses to impacts of N deposition and climate change. Only recently have N-cycling processes been incorporated into Earth system models for C-N interactions. The robustness of these models remains to be demonstrated. Much work remains for improving their representation in models used to simulate climate forcing scenarios.

Keywords Climate Change · Reactive Nitrogen

1 Introduction

Reactive nitrogen (Nr) emissions alter the climate in many ways, and the importance of the nitrogen (N) cycle in regulating climate is gaining increasing attention. Excess N in terrestrial systems can change the uptake and emission of the three most important anthropogenic greenhouse gases: carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). Many experiments have demonstrated substantial N limitations of CO_2 uptake on land. Therefore, owing to its scarcity, N is a chief player in climate change and the fate of anthropogenic CO_2 emissions. In addition, Nr is a substrate for N_2O production by nitrifying and denitrifying bacteria in soils, sediments, and water bodies. Microbial production and consumption of CH_4 is also affected by N. In the atmosphere, Nr alters atmospheric chemistry and affects the production and lifetimes of greenhouse gases such as ozone (O_3) and CH_4, and also leads to the formation of aerosols, which, in turn, affect regional and global climate. This article provides an overview on the impacts of Nr on radiative forcing, paying particular attention to the specific interaction between the N and carbon (C) cycles. We present evidence from field studies, meta-analyses, and models of biogeochemical processes within earth system models.

2 Radiative impacts of reactive nitrogen

The most direct effect of N on climate is through N_2O production, the third most important anthropogenic greenhouse gas, contributing 6% of total human-induced global warming. It has about 300 times the per-molecule warming potential of CO_2 and it is long-lived in the atmospheric (a "mean residence time" of more than 110 years) (Forster et al, 2007). The concentration of N_2O in Earth's atmosphere is derived from a variety of sources, mainly from the activity of nitrifying and denitrifying bacteria in soils, sediments, and water bodies. Globally, natural ecosystems release about 10 Tg N_2O-N yr^{-1}, and anthropogenic sources sum to about 7 Tg N_2O-N yr^{-1}, although one recent study has reported a lower natural contribution (Zhuang et al, 2012). Anthropogenic sources are dominated by the widespread use and subsequent microbial processing of fertilizer in agricultural soils (Forster et al, 2007). Atmospheric concentrations of N_2O have increased rapidly since the industrial revolution, as livestock herds increased globally and as use of synthetic-N fertilizers increased after WWII (Davidson, 2009). The natural sink for N_2O in soils is small (Syakila and Kroeze, 2011, Van Groenigen et al, 2011). The current rate of increase in the concentration of N_2O is about 0.3% yr^{-1}, equivalent to the accumulation of 4 Tg N_2O-N yr^{-1} in Earth's atmosphere. Global emissions of N_2O are likely to increase as fertilizers are used to boost agricultural productivity.

The US EPA estimates that agricultural activities in the US are directly or indirectly responsible for emissions of about 0.48 million tons of N_2O-N yr^{-1} (United States Environmental Protection Agency Office of Atmospheric Programs, 2011), which is about 80% of total US N_2O production (the remainder from energy and industrial sources) and about 10% of the global N_2O emissions from agriculture. Several mitigation options exist to reduce the emissions of N_2O from agricultural soils (Davidson et al, 2012), and are addressed in more detail in Robertson et al (this issue). Associated emissions of N_2O are estimated to negate much of the CO_2 miti-

gation effect from C sequestration in soils (e.g., Schlesinger (2010)) or from biofuel production using fertilized crops such as corn (Melillo et al, 2009).

While not a greenhouse gas directly, nitrogen oxides (NO_x) are often a limiting factor in the production of O_3 in the troposphere (the lower atmosphere), which acts as a potent greenhouse gas (Derwent et al, 2008). Nitrogen oxide (NO) reacts with radicals that donate an oxygen atom and convert the NO to nitrogen dioxide (NO_2). In sunlight, NO_2 can give up one of its oxygen atoms as it is converted back to NO by photolysis. The extra atomic oxygen reacts with the molecular oxygen (O_2), which is abundant in the lower atmosphere, and creates O_3. In the short-term, NO_x emissions contribute to warming by enhancing tropospheric O_3 concentrations. Furthermore, the short-term increase in O_3 due to NO_x can impact climate indirectly, by damaging photosynthesis and plant CO_2 uptake by as much as 20%, leading to a reduction of atmospheric CO_2 sequestration by the plant biomass and resulting in more CO_2-driven warming (Felzer et al, 2004, Ollinger et al, 1997, Sitch et al, 2007). Carbon storage and Nr are discussed in more detail in the next section.

Another indirect effect of NO_x is through its effect on CH_4, which is the second-most important greenhouse gas, contributing 15% of total human-induced global warming. With an atmospheric lifetime of 12 years, CH_4 has roughly 27 times the per-molecule warming potential of CO_2 (Boucher et al, 2009). The largest removal process of CH_4 is oxidation by the hydroxyl radical (OH), accounting for 88% of the total sink. Emissions of NO_x can increase atmospheric OH and accordingly, decrease CH_4 concentrations (Boucher et al, 2009). An additional feedback is that the by-products of CH_4 oxidation include radicals that can convert NO to NO_2. Through this mechanism, CH_4 is also an important contributor to ozone formation (Fiore et al, 2002). Hence, in addition to increasing O_3 on daily time scales, NO_x can lead to decreases in O_3 concentration on a decadal time scale, because it causes an increase in OH radical concentration, which decreases CH_4 concentration, which decreases NO_2 formation, which decreases O_3 formation.

Because NO_x can both increase and decrease ozone production, the net result of these competing effects strongly depends on where the NO_x emissions occur (Berntsen et al, 2005, Collins et al, 2010, Fry et al, 2012, Naik et al, 2005). However, the net impact of NO_x on atmospheric chemistry is likely to be cooling, by (*i*) decreasing the CH_4 concentration, and (*ii*) decreasing O_3 formation due to lower CH_4 concentrations (Fuglestvedt et al, 2010, Wild et al, 2001). Both global, regional, and emission sector-based estimates of the impact of NO_x on CH_4 and O_3 radiative forcing are listed in Table 1.

In addition to altering radiative forcing from CH_4 and O_3, both NO_x and ammonia (NH_3) also react with other atmospheric constituents to form fine particles called aerosols. Aerosols are powerful cooling agents, both directly by scattering or absorbing light, and indirectly, by affecting cloud formation and lifetime (Forster et al, 2007). Ammonium sulfate ($(NH_4)_2SO_4$), ammonium nitrate (NH_4NO_3), and organic aerosols are especially important in these processes. Because NO_x influences the rate of oxidation in the atmosphere, it impacts the formation of sulfate and organic aerosols (Shindell et al, 2009). Ammonia (NH_3) is the most important atmospheric base, and by neutralizing sulfate and nitrate (NO_3^-), it can enhance the formation of new particles and can condense onto existing acidic particles. Both NO_x and NH_3 alter the chemical and optical properties of the aerosol (Martin et al, 2004), which influences the conversion of aerosol to cloud droplets and ice nuclei (Abbatt et al, 2006, Sorooshian et al, 2008), and alters the lifetime and brightness of clouds. The wide ranges of estimates of the effect of NH_4NO_3 on aerosol radiative forcing globally are shown in Table 2. Note that while the values presented in Table 2 are globally averaged, nearly all of the forcing from NH_4NO_3 is in the northen hemisphere. Therefore, these aerosols can have a larger impact on regional precipitation and temperature patterns.

Furthermore, O_3 and aerosols cause serious human health effects and contribute to air pollution (see Peel et al (this issue)). Interactions between the N cycle and

climate change can exacerbate air pollution problems. For example, O_3 formation is also strongly temperature sensitive (Bloomer et al, 2009), thus rising temperatures can exact a so-called "climate penalty" on the air pollution gains made by reducing NO_x emissions (Jacob and Winner, 2009, LaFranchi et al, 2011).

Ultimately, the atmosphere tends to convert NO_x and NH_3 to more water-soluble forms that are readily deposited to the Earth's surface. This is a significant source of N available to ecosystems, which influences climate forcing indirectly by altering rates of C sequestration and emissions of CH_4 and N_2O from soils. Deposition of Nr onto ecosystems changes N availability and can increase N_2O emissions and decrease uptake of atmospheric CH_4 by soil microorganisms. Natural well-drained soils (i.e., not wetlands) are an important sink for atmospheric CH_4. However, soil microbes that consume CH_4 often preferentially consume ammonium (NH_4^+), leading to reduced CH_4 consumption rates in the presence of abundant NH_4^+(Mosier et al, 1991). The effects of Nr deposition on plant growth and C storage is described in the next section.

3 N effects on carbon storage

Atmospheric deposition of Nr affects terrestrial C sinks by affecting two key processes. First, inputs of Nr from atmospheric deposition can enhance plant growth rates because of the fundamental constraint of N availability on plant productivity and CO_2 uptake into plant biomass. Second, decomposition is affected by altering Nr availability which slows decomposition of plant litter and soil organic matter in many, but not all, forest types. Excess N can also impact C cycling in coastal and marine ecosystems; this is discussed in Baron et al (this issue).

3.1 N effects on plant growth rates

It is well established that net primary production (NPP) is limited by N availability in many terrestrial ecosystems (LeBauer and Treseder, 2008), due to the fact that ex-

perimental or fertilizer N additions typically increase C capture and storage. A meta-analysis of 126 N addition experiments evaluated N limitation of aboveground net primary productivity (ANPP) in terrestrial ecosystems by comparing above-ground plant growth in fertilized to control plots (LeBauer and Treseder, 2008). ANPP was calculated by multiple methods, including allometric biomass increment plus litter-fall, basal area increment, diameter increment, annual litterfall, and allometric volume increment. The results showed that most ecosystems are N limited with an average 29% growth response to N additions. The response was significant within temperate forests, tropical forests, temperate grasslands, tropical grasslands, wetlands, and tundra, but not deserts (LeBauer and Treseder, 2008). The majority of these estimates were based on data from forest ecosystems in northern latitudes, whereas tropical areas and other ecosystem types were not well represented (LeBauer and Treseder, 2008).

While increasing N availability can stimulate plant growth, estimates of this stimulation show greater variation. For example, in a recent synthesis by Butterbach-Bahl et al (2011), the average increase in above ground C sequestration per unit of N addition is 25 kg C kg^{-1} N (Table 3). For eastern US forests, Thomas et al (2010) estimated an above-ground sink of 61 kg C kg^{-1} N. The magnitude of growth stimulation is likely greatest in regions of moderate Nr deposition and slower or even leading to enhanced mortality in regions of highest Nr deposition, due to nutrient imbalances or acidification (Aber et al, 1998). At present, most US ecosystems are probably in the former category, although some high elevation ecosystems in the eastern US may be in the latter category (Pardo et al, 2011). Finally, some ecosystems are also limited by phosphorous (P). When both N and P are enhanced, the impact of N can be substantially larger (Elser et al, 2007, Harpole et al, 2011).

The addition of N has also been shown to increase foliar N concentration (Xia and Wan, 2008), which often results in higher photosynthetic rates, but not at high levels of chronic N addition (Bauer et al, 2004). The de-coupling of a photosynthetic-

N relationship was observed in numerous chronic N-addition studies, mainly because the excess N was invested in amino acids rather than enzymes and proteins associated with the photosynthetic process (Bauer et al, 2004). Foliar N may also increase the albedo of the canopy, enhancing the reflectivity of the Earth's surface, and hence contribute to cooling (Hollinger et al, 2010, Ollinger et al, 2008).

It is important to note that the potential for N addition to increase above-ground C biomass is limited in part because only a small portion of added N is actually taken up by vegetation, and thus only a small portion of N contributes to C capture by trees (Nadelhoffer et al, 1999). Recovery in tree biomass (e.g., foliage, woody tissue, and fine roots) of N that was experimentally added to forests has been estimated to range between 7 to 16% (Nadelhoffer et al, 2004) and 0 to 45% (Schlesinger, 2009). Nitrogen may be immobilized in the soil, leached out before biological assimilation, or, upon the addition of N, another factor may become limiting to growth (e.g., water or other nutrients).

3.2 N effects on carbon storage in soils

While N deposition may stimulate productivity and facilitate significant C storage aboveground (LeBauer and Treseder, 2008, Xia and Wan, 2008), similar trends have not been as clearly observed in soils. With greater productivity, N addition generally increases aboveground litter inputs (LeBauer and Treseder, 2008, Liu and Greaver, 2010, Xia and Wan, 2008), and improves the chemical quality of that litter (i.e., lower lignin: N ratios and greater labile C inputs to surface soils; (Berg and Laskowski, 2006). In contrast, N addition decreases fine root production, root respiration (Janssens et al, 2010), and mycorrhizal abundance (Treseder, 2004). Although these patterns are not consistent across meta-analyses (Liu and Greaver, 2010), they support the idea that higher plant productivity associated with N deposition shifts

litter production aboveground as plant investment for nutrient acquisition declines (Aerts and Chapin, 2000).

The biochemistry of litter inputs, and especially litter lignin content, influences the effect of N addition on soil C storage. For example, Waldrop et al (2004) report significant soil C losses with N addition in a sugar maple forest delivering high quality litter, and significant soil C gains with N addition in a nearby oak-dominated forest with lower quality litter. Similarly, root lignin content affects soil C storage in grassland ecosystems receiving elevated CO_2 and N addition (Dijkstra et al, 2004). Concurrently, N addition is also known to influence changes in plant species composition (Clark and Tilman, 2008). The extent to which climate, N addition, and their interactions may drive changes in species composition that simultaneously alter the quantity and quality of litter inputs have been little explored in the literature (but see Aerts and Bobbink (1999)).

Nitrogen deposition elicits a host of microbial responses that influence organic matter decomposition and, ultimately, influence soil C storage. Microbial responses to N addition include: changes in relative enzyme activity, microbial substrate use, and microbial community composition (Cusack et al, 2011, Sinsabaugh and Moorhead, 1994). Notably, N addition accelerates the decomposition of high quality (low lignin) litter by stimulating cellulose degradation, which is typically N limited (Berg and Matzner, 1997, Carreiro et al, 2000, Fog, 1988, Frey et al, 2004, Saiya-Cork et al, 2002, Sinsabaugh et al, 2002). In contrast, N addition significantly slows decomposition of low quality (high lignin) litter because of decreases in phenol oxidase activity, which reduces rates of lignin degradation (Fog, 1988, Hammel, 1997, Sinsabaugh et al, 2002). This divergent pattern based on litter quality has significant implications for soil C storage in systems receiving N deposition. In some systems, decreases in phenol oxidase activity are attributed to declines in fungal biomass, declining fungal: bacterial ratios, and a reduction of Basidiomycetes, or white rot fungi (Carreiro et al, 2000, Fog, 1988, Frey et al, 2004, Saiya-Cork et al, 2002, Sinsabaugh et al,

2002). However, fungal declines with N addition are not ubiquitous in studies reporting changes in microbial community structure (Nemergut et al, 2008, Ramirez et al, 2010, Saiya-Cork et al, 2002). The effects of N addition on shifts in microbial community structure and function and their influence on litter decomposition are mediated by substrate quality. As a result of these changes in microbial community structure and function, rates of litter decomposition generally slow with N deposition, although the consistency of these findings is influenced by ambient levels of N deposition and initial litter chemistry (Fog, 1988, Hobbie, 2005, Janssens et al, 2010, Knorr et al, 2005). Notably, rates of N-addition more than 5 kg ha^{-1} yr^{-1} slow litter decomposition, whereas rates of N deposition less than 5 kg ha^{-1} yr^{-1} may actually accelerate leaf litter decomposition (Knorr et al, 2005).

Additionally, N deposition may affect dissolved organic carbon (DOC) export from soil C (Liu and Greaver, 2010). Across multiple spatial scales, increasing N availability increases DOC export from soils (Aitkenhead and McDowell, 2000). Mechanisms to explain these patterns are still unresolved, but generally increased DOC losses result from the combination of higher aboveground litterfall, decreased microbial lignin degradation, and soil acidification (Evans et al, 2008, Findlay, 2005, Monteith et al, 2007, Sinsabaugh et al, 2004). Although the acceleration of DOC losses by N-addition may have little impact on ecosystem C storage (Aitkenhead and McDowell, 2000), these DOC and Nr inputs have significant consequences for aquatic ecosystems.

When combined with observations of higher aboveground productivity and litterfall, one might expect significantly greater soil C storage in systems exposed to N addition, but reported rates of accumulation of C in soils are generally modest. Butterbach-Bahl et al (2011) estimate that 15 kg C are sequestered per kg N deposition in forest soils (Table 3). However, meta-analyses show conflicting results for accumulation of soil C with N-addtion (Janssens et al, 2010, Liu and Greaver, 2010, Nave et al, 2009). Some of the variation of soil C accumulation reported in these

meta-analyses could result from variation in regional / ecosystem response to N addition, or the type, duration, and intensity of N additions.

3.3 N effects on total ecosystem carbon storage

It is important to consider both the above and belowground C pools in terrestrial ecosystems to understand N effects on total ecosystem C sequestration. Various approaches, such as modeling, inventory, and static accounting, have been used to estimate the N-induced C sink for different ecosystems (Holland et al, 1997, Liu and Greaver, 2009, Magnani et al, 2007, Thomas et al, 2010). The effect of N on net C flux (both above and below ground pools) differs among ecosystems. In general N addition to grasslands and wetlands does not increase C storage; however N stimulates more C storage in forests (Liu and Greaver, 2009). In grasslands and wetlands N stimulation of ANPP is offset by other C losses in the system. For example, Bragazza et al (2006) investigated peatlands across a gradient of N deposition levels and found higher atmospheric N deposition resulted in higher C loss by increasing heterotrophic respiration and DOC leaching. Similarly, Mack et al (2004) found N fertilization stimulated soil organic carbon (SOC) decomposition more than plant production in a tundra ecosystem, leading to a net loss of ecosystem C. Among terrestrial ecosystems, the response of forests to N availability has been most intensively studied, but more data are needed to better characterize other types of terrestrial ecosystems.

In forests, a wide range of values have been reported for how much additional C is expected to be sequestered per unit of N added. Magnani et al (2007) published a very high estimate of 725 kg C accumulated per kg N added (dC/dN) to boreal and temperate forests. However, this estimate was quickly contested as biologically implausible by Sutton et al (2008) who reanalyzed the original data and suggested that 68 dC/dN was more accurate. Since then, attention has been drawn to the basic sto-

ichiometry constraints for C sequestration by N at the ecosystem scale (Schlesinger et al, 2011).

Several studies have evaluated dC/dN ratios in US forests and a meta-analysis examined the effect of N fertilization on ecosystem C content (EC), defined as the sum of C content of vegetation, forest floor and soil (Liu and Greaver, 2010). To avoid possible confounded variability caused by site conditions, this meta-analysis only included studies where control and treatment sites experienced the same climatic, soil and vegetation conditions. Studies on N effects along a deposition gradient were not included. Results show that while there was a great deal of variation in response, overall N addition increased EC by 6% for US forest ecosystems. This study did not find any correlation between the amount of N addition and the response magnitudes of EC. On average, forest ecosystems sequestered 24.5 ± 8.7 kg C ha^{-1} yr^{-1} per kg N ha^{-1} yr^{-1} (Liu and Greaver, 2009). Using a different approach, Thomas et al (2010) examined tree growth rates over an N deposition gradient in US Northeastern forests. Their results indicate that enhancement of above-ground C storage averaged 61 kg C ha^{-1} yr^{-1} per kg increase in N deposition. When calculating a dC/dN response ratio using values of N deposition, it is very important to consider how N deposition is calculated and whether all relevant chemical species are included. In Thomas et al (2010), N deposition was calculated using estimates of wet NO_3^-, wet NH_4^+, dry HNO_3 gas and particulate NH_4^+ and NO_3^-; it did not, however, include other forms of N deposition, such as dry NH_3, NO and NO_2, or organic N. Because all forms of N deposition were not used in the calculation, above ground dC/dN is likely to be over estimated compared to N-addition studies. In addition, when a biometric relationship is applied that assumes below-ground tree biomass represents roughly 20% of above-ground biomass, then enhancement of total tree C would increase to 73 kg C ha^{-1} yr^{-1} per kg increase in N deposition. This approach assumes dC/dN in belowground biomass is the same as above ground biomass, which is often not the case (Table 3),

and does not include other soil pools that affect dC/dN. These reasons may partially explain why the Thomas et al (2010) estimate is larger than the N addition studies.

Butterbach-Bahl et al (2011) recently synthesized and reviewed published dC/dN ratios from studies conducted in Europe and North America (not including Liu and Greaver (2009) or Thomas et al (2010)) and found that average total C sequestration was 41 kg C per kg N addition in forests. Although more research needs to be done to further refine estimates of dC/dN in forests, considering the studies summarized in Table 3 and their caveats, the range of values reported in the literature are between 20 – 70 kg C ha^{-1} yr^{-1} per kg N ha^{-1} yr^{-1}. Key uncertainties in the sensitivity of ecosystem C sequestration response to N addition include the form and manner of N input, succession status of the forest and prior land-use history (Butterbach-Bahl et al, 2011).

Three factors could decrease rates of dC/dN reported for a given forest: N saturation status, stand age, and availability of other essential nutrients. First, N will increase NPP of an N-limited system; however N addition beyond a certain point may lead to decreases in NPP (Aber et al, 1998). Second, several studies have shown that NPP declines with stand age (Gower, 2003, Ryan et al, 2004), which could reduce the potential response to N addition. Furthermore, as NPP decreases due to age, so too will dC/dN. The relative effect of saturation and stand age is varied – a flux study found evidence of nitrogen enhanced productivity even in an old growth (200 to 300 years old) forest (Luyssaert et al, 2007).

4 Biogeochemical models: C-N interactions, C storage, and N gas emissions

4.1 Modeling N effects on C sequestration

As climate models evolve into models of the behavior of the entire Earth system, they have expanded beyond their hydrometeorological heritage to include biogeochemical cycles and atmospheric chemistry. Early global climate models focused solely on

atmospheric physics; later models incorporated the C cycle in order to include feedback with atmospheric CO_2. Coupled C cycle-climate models include terrestrial and marine C fluxes so that changes in atmospheric CO_2 concentration are simulated in response to anthropogenic CO_2 emissions (Denman et al, 2007, Friedlingstein et al, 2006). In these models, rising atmospheric CO_2 concentration increases land C uptake by stimulating plant productivity, and this "CO_2 fertilization" is a negative feedback to higher atmospheric CO_2 concentration (the concentration-C feedback). Land C loss through ecosystem respiration increases with warming in a positive climate feedback (the climate-C feedback). Additionally, warming can enhance productivity (negative feedback) in cold regions, but decrease productivity (positive feedback) in warm regions, where greater evaporative demand dries soil. These predictions for the terrestrial C cycle are found in Earth system models that do not include C-N biogeochemistry. In recent years, some Earth system models have added some representation of the N cycle as a crucial regulator of C-cycle dynamics and aspects of atmospheric chemistry, but much work is needed to properly incorporate representation of N cycling processes in climate models. Global biogeochemical models of the terrestrial C and N cycles for the Earth build upon a rich heritage of terrestrial ecosystem models (Bonan, 2008). They simulate C and N flows among various vegetation and soil components, N inputs for atmospheric deposition and biological N fixation, and N losses from denitrification and leaching.

Carbon cycle-climate model simulations of future climate change predict that nitrogen has an important effect on future carbon uptake (Sokolov et al, 2008, Thornton et al, 2009, Zaehle et al, 2010). Limited mineral N availability restricts the increase in plant productivity from rising CO_2 concentration. Conversely, warming increases decomposition of organic material and N mineralization, stimulating plant productivity. These findings are generally consistent with results from free-air CO_2 enrichment experiments and soil warming experiments, though few models have been directly compared with experimental manipulations (Melillo et al, 2011).

As mentioned earlier, because N availability restricts plant productivity in many ecosystems; N addition from atmospheric N deposition can enhance C storage. Initial studies of the effect of anthropogenic N deposition on the C cycle reported that the additional N in the system increased global terrestrial C storage from as much as 0.6-1.5 Pg C yr^{-1} (Holland and Lamarque, 1997, Townsend et al, 1997) to as little as 0.25 Pg C yr^{-1} (Nadelhoffer et al, 1999). More recent model simulations support a C sink of about 0.2 Pg C yr^{-1} (Bonan and Levis, 2010, Jain et al, 2009, Thornton et al, 2009, Zaehle et al, 2011). These models differ in important ecological and biogeochemical processes (e.g., how N affects plant productivity; below-ground C-N dynamics; and denitrification) that determine the amount of N in the system available for plant use and the magnitude of the C sink. Model comparison with results from N-deposition gradient analyses (Thomas et al, 2010) or N-enrichment experiments (Liu and Greaver, 2009) are needed to evaluate the model simulations and to identify deficiencies in model parameterizations. Estimates of N-enhanced C storage, whether derived from observational studies or from models, require knowledge of N deposition rates. These rates can differ greatly among studies in the magnitude and spatial distribution of the deposition, which makes comparison among studies difficult.

A complete understanding of the effects of increased N deposition on terrestrial C storage and radiative forcing requires a multi-disciplinary integration of biogeochemical processes with biogeophysical processes (i.e., energy and water fluxes), and with changes in ecosystem structure and community composition arising from stand dynamics. For example, a more productive forest with higher leaf area index resulting from enhanced N deposition is likely to decrease surface albedo, warming climate with a positive radiative forcing and increasing evapotranspiration (Bonan, 2008). Increased evapotranspiration locally cools temperature, but can warm global temperature through increased atmospheric water vapor. The net effect of changes in C storage, surface albedo, and evapotranspiration on radiative forcing is largely unknown for forest ecosystems, and initial estimates of the forcing are quite speculative

(Bonan, 2008). Another possible biogeophysical forcing is manifested through the effect of foliar N on leaf-, stem-, and canopy-level traits that alter the overall plant reflectance. Canopy N concentration is strongly and positively correlated with canopy albedo, suggesting a possibly significant biogeophysical role of N in the climate system through solar radiation absorption and canopy energy exchange (Hollinger et al, 2010, Ollinger et al, 2008). The long-term sustainability of the N-enhanced C sink is unclear, and carbon uptake may saturate with future levels of N deposition. The future potential of C storage in terrestrial ecosystems depends on trajectories of climate change and land use, which alter community composition and ecosystem structure. Redistribution of plant species in response to climate change alters patterns of C storage, N uptake, and N mineralization (Metcalfe et al, 2011, Pastor and Post, 1988). Enhanced C storage in forest ecosystems arising from atmospheric N deposition becomes less important in a warmer climate where droughts and wildfire are more common. Trajectories of land use (e.g., deforestation, reforestation, and afforestation) driven by socioeconomic needs and policy implementation will also come into play and have competing biogeophysical and biogeochemical impacts on climate. These changes in community composition and ecosystem structure are largely ignored in the current generation of Earth system models, which build on biogeochemical models rather than models of vegetation dynamics.

5 Modeling N effects on N_2O emissions and other radiative forcing

The atmospheric chemistry models included in Earth system models allow for additional biogeochemical land-atmosphere interactions such as surface N-gas emission and atmospheric N deposition (Lamarque et al, 2011). With the addition of N-gas emissions, the models provide surface N fluxes to atmospheric chemistry models, and can be used to quantify the net radiative forcing due to Nr. This forcing includes the effect of N on terrestrial C storage, the direct radiative forcing from N_2O emis-

sions, and Nr in the atmosphere and its effects on CH_4, tropospheric and stratospheric O_3, and secondary aerosols.

Nitrogen losses associated with nitrification and denitrification are poorly represented in the biogeochemical component of Earth system models and present a large uncertainty in global simulations of climate-N interactions (Schlesinger, 2009). Dinitrogen gas (N_2) loss during denitrification is a large term in the global terrestrial N budget (Galloway et al, 2004, Houlton and Bai, 2009, Schlesinger, 2009), but there is a high degree of uncertainty regarding the amount of N_2 lost to the atmosphere (see Houlton et al (this issue)). A better understanding and further quantification of ecosystem N_2 flux is needed given that this is the best possible outcome for minimizing environmental impacts from excess N. The DayCENT (Del Grosso et al, 2000) and DeNitrification-DeComposition (DNDC) (Li et al, 2000) models are two commonly used approaches to represent nitrification, denitrification, and associated N-gas emissions. In addition, the Environmental Policy Integrated Climate (EPIC) (Williams et al, 1996) and Agricultural Policy Environmental EXtender (APEX) (Gassman et al, 2009) simulate these processes for agricultural lands under a range of farming conditions and activities. These models have been evaluated for a wide range of environmental conditions, ecosystem types, and N inputs (Olander and Haugen-Kozyra, 2011), but are mostly applied at the site or regional scale. Global terrestrial biogeochemical models for use with Earth system models may not explicitly simulate denitrification and instead include it as a generic N loss term (Gerber et al, 2010, Melillo et al, 1993, Wang et al, 2010). Furthermore, some of the current global models represent denitrification as a fraction of mineralization or mineral soil N (Thornton et al, 2009, Yang et al, 2009). Zaehle et al (2010) developed an advanced process-oriented formulation of nitrification, denitrification, and N-gas emissions based on the DNDC model structure, which observed a likely contribution of N addition to C sequestration in forest ecosystems and concurred with ecosystem field studies. Houlton and Bai (2009) used a mass-balance approach constrained

by observations of ^{15}N:^{14}N isotope ratios to estimate NO, N_2O, and N_2 emissions globally and regionally. However, the complexity of trace gas biogeochemistry, the fine-scale spatial heterogeneity of trace gas production, and anthropogenic alterations from agricultural practices makes modeling N-gas emissions an uncertain aspect of global Earth system model simulations.

In addition, a key aspect of climate-N interactions not currently considered by Earth system models is the effect of anthropogenic N on radiative forcing mediated through changes in atmospheric chemistry. Secondary atmospheric aerosols resulting from emissions of NO_x and NH_3 provide a negative radiative forcing that cools climate. None of the currently available Earth system models are able to fully assess these effects, in part because the current generation of global terrestrial C-N biogeochemical models used with Earth system models does not represent N-gas emissions and the anthropogenic and environmental drivers of these emissions.

6 Net effects of C-N interactions on radiative forcing

Reactive N has numerous effects on climate, including N_2O emissions, indirect effects on O_3, CH_4, and aerosols, and C sequestration. To compare these impacts, the effects must be converted to a common metric. A recent effort in Europe has led to a continental assessment of the contribution of European emissions of Nr to instantaneous radiative forcing, expressed as W m^{-2} (Butterbach-Bahl et al, 2011). Because aerosols have a large effect on short-term radiative forcing, it was found that the net effect of cooling from aerosols and C sequestration outweighed the warming effect of N_2O emissions across Europe. However, the pathways by which Nr impacts climate change do not have the same lifetime – aerosols last for only a few weeks, CH_4 on the order of a decade, and N_2O and CO_2 persist for more than a century. While radiative forcing is a measure of the instantaneous climate change impact, the long-term climate effects depend heavily on atmospheric longevity (Penner et al, 2010).

An alternative approach is the global temperature potential (GTP), which is a measure of the change in global temperature, after a fixed number of years, due to a 1 kg pulse of emissions. The GTP can be calculated on a 20-year basis, to identify Nr impacts likely to change the rate of climate change in the coming decades, as well as a 100-year basis, to understand the long-term magnitude of climate change. To compare across compounds, the GTP is normalized by the change in temperature due to a pulse of CO_2 and expressed in common units of kg CO_2 equivalence (CO_2e).

The climate change impact of US Nr, on a global temperature potential basis, is presented in Figure 1. Each bar represents the climate change impact, in units of Tg CO_2e, due to US Nr emissions, via the processes listed on the left. The length of the bar denotes the range of uncertainty as estimated by a synthesis of the relevant literature. The impacts from changes in O_3, CH_4, and aerosols were calculated as the product of US emissions and the GTP of those compounds as calculated by Fuglestvedt et al (2010). For the change in greenhouse gas fluxes due to N deposition, the dC/dN values were multiplied by the anthropogenic N deposition calculated by the Community Multiscale Air Quality model (CMAQ; Appel et al (2010)) to each landcover type. A range of 24 – 65 was used for the dC/dN value for forests. The lower value of this range (24) is from Liu and Greaver (2009) and the upper end of this range is from Thomas et al (2010). The value 65 results from adjusting the Thomas et al (2010) value upwards to account for below ground biomass and soil C, and downwards, to account for incomplete measurement of N. For other land cover types, the ranges reported in Liu and Greaver (2009) were used. The permanence of enhanced CO_2 uptake on a 20-year and 100-year timescale was estimated using forestry management data (Heath et al, 2011). The details of these calculations are described in Pinder et al (2012).

The relative impact of each aspect of Nr depends strongly on the time frame of interest. On the left side of Figure 1, the impacts are compared on a 20-year basis. Here, the change in O_3, CH_4, and aerosol concentrations due to NO_x contribute sub-

515 stantially to climate change. But on a 100-year basis, these processes are negligible.
516 Emissions of NO_x in the US contribute to cooling on a 20-year basis, but have a very
517 little effect on a 100-year basis. Overall, the cooling effects (i.e., C sequestration en-
518 hanced by N deposition, increased lifetime of CH_4, and greater aerosol burden) are
519 slightly larger than the warming effect of N_2O on a 20-year time frame. The error
520 terms on these estimates are large, and the range of uncertainty includes the possi-
521 bility that the net effect is negligible. But on a 100-year basis, the net impact of Nr
522 appears to be one of warming. Putting these estimates into a broader perspective, the
523 modest warming effect US Nr shown in Figure 1 is equivalent to less than 10% of the
524 warming effect of US emissions of CO_2 derived from fossil fuel combustion.

525 While the net radiative forcing from the alternation of the N cycle in the US may
526 be relatively small, there are many offsetting impacts that occur over different time-
527 scales. The long atmospheric half-life of N_2O and uncertainties regarding the per-
528 manence of C sequestration mean that there is a risk that the long-term net warming
529 effects may be underestimated. Moreover, the profound effect that excess Nr has on
530 ecosystem processes and biodiversity suggests that assumptions about future radiative
531 forcing of C-N interactions played out in changing terrestrial and aquatic ecosystems
532 must be considered with caution. Despite these uncertainties, we can conclude with
533 confidence that C-N interactions do have important climatic effects that should be
534 included in future measurement and modeling efforts to improve understanding of
535 biological feedbacks to climate change and global change processes.

536 7 Research needs

537 Improved quantification of the effects of excess Nr on radiative forcing will require
538 improvements in our understanding of atmospheric chemical processes, rates of to-
539 tal N deposition, responses of ecosystems to N deposition, and integration of these
540 processes into Earth system models. We identify a number of research needs below:

1. The complex nonlinear atmospheric chemistry involving NO, NO_2, O_3, OH, and CH_4 and how it will change with climate and changing sources and sinks requires more research attention to determine impacts at times scales from days to decades.
2. The effects of the chemical composition of aerosols on radiative forcing and cloud formation are not well known.
3. Improvements are needed in spatially explicit modeling and measurements of all forms of N deposition. Estimates of deposition of organic-N are particularly uncertain.
4. Variation in dC/dN responses of ecosystems and the factors that control them are poorly understood for both aboveground and belowground processes. Comparisons between model simulations and results from N addition enrichment studies, gradient analyses, and other field data are needed to validate and identify deficiencies in parameters of both empirical and process-based models.
5. Biogeochemical models need improvements to better constrain and reduce uncertainty of estimates of N losses associated with nitrification and denitrification, especially losses of N_2 from denitrification.
6. Earth system models need improved representation of C-N-P interactions in ecosystems and their feedbacks to climate change. This includes feedbacks between vegetation, water vapor, and albedo. Most Earth system models also do not yet include the effects of anthropogenic N on radiative forcing mediated through changes in atmospheric chemistry.

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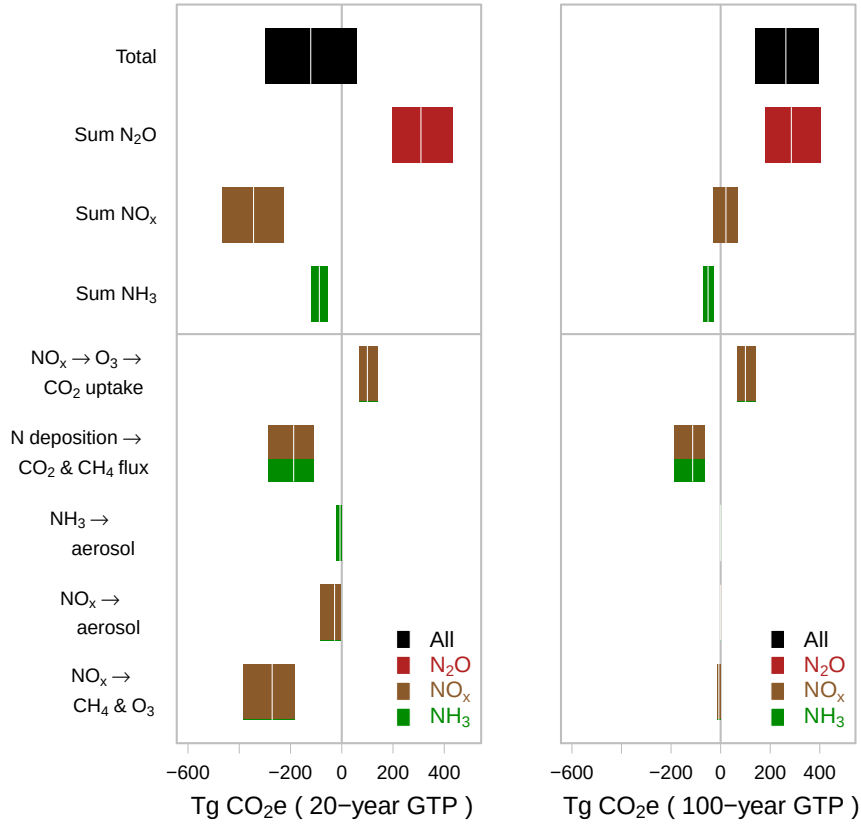

Fig. 1 The climate change impacts of US reactive nitrogen emissions, by chemical species, in common units of equivalent Tg of CO_2 (Tg CO_2e) on a 20-year and 100-year global temperature potential (GTP) basis. The width of the bar denotes the uncertainty range; the white line is the best-estimate; and the color shading shows the relative contribution of NO_x and NH_3 emissions to nitrogen deposition (adapted from Pinder et al (2012)).

Table 1 Change in ozone and methane radiative forcing (mW m^{-2}) due to reactive nitrogen (per Tg N), as calculated in global, regional, and source-specific sensitivity studies.

Source	region / sector	$NO_x \rightarrow$ ozone	$NO_x \rightarrow$ methane
Derwent et al (2008)	global	+1.0	-2.4
Naik et al (2005)	North America	+0.088	-1.7
Fry et al (2012)	North America	+2.2	-2.7
Berntsen et al (2005)	Europe	+2.0	-1.9
Wild et al (2001)	mid-latitudes	+1.1	-1.9
West et al (2007)	anthropogenic	+2.9	-3.7
Stevenson et al (2004)	aircraft	+1.5	-13.8
Khler et al (2008)	aircraft	+28	-28
Eyring et al (2007)	shipping	+1.3	-4.5
Endresen et al (2003)	shipping	+3.8	-7.7
Fuglestvedt et al (2008)	shipping	+5.3	-7.6

Table 2 Change ammonium nitrate (NH_4NO_3) radiative forcing (W m^{-2}) due to global anthropogenic emissions, as calculated in global climate modeling studies.

Source	(W m^{-2})	Type of radiative forcing
Forster et al (2007) (Table 2.13)	-0.10 $\pm$ 0.10	NH_4NO_3 aerosol direct effect
Adams et al (2001)	-0.19	NH_4NO_3 aerosol direct effect
Liao and Seinfeld (2005)	-0.16	NH_4NO_3 aerosol direct effect
Bauer et al (2007)	-0.06	NH_4NO_3 aerosol direct effect
Myhre et al (2009)	-0.023	NH_4NO_3 aerosol direct effect
Shindell et al (2009)	-0.11	NH_4NO_3 aerosol direct effect
Xu and Penner (2012)	-0.12	NH_4NO_3 aerosol direct effect
Xu and Penner (2012)	-0.09	effect of nitric acid gas and NH_4NO_3 aerosol on cloud droplets

Table 3 Current mean estimates of dC/dN ratio for forest ecosystems in North America

Approach	Carbon sequestration, kg C yr^{-1} (kg N yr^{-1})$^{-1}$			Scale of application	Authors
	Above ground	Below ground	Total		
Empirical field data; correlation between NEP and total N deposition[1]	-	-	68 – 177	Chronosequences in boreal and temperate forests of Eurasia and North America	Magnani et al (2007) as re-evaluated by Sutton et al (2008)[3]
Meta-analysis of 9 U.S. studies measuring the effects of N addition on total ecosystem carbon (EC); only included studies of which control and treatment sites experienced the same climatic, soil and vegetation conditions	-	-	24.5	U.S. forests	Liu and Greaver (2009)[3]
Modeled values of N stimulation of above ground C accumulation based on measurements of tree growth along an N deposition gradient[2]; below ground values calculated using a biometric relationship	61	12	73	24 common tree species occurring in Northeastern forest in the US	Thomas et al (2010)[3]
Synthesis of 14 forest studies (conducted from 1983-2010) including observed measurements and modeled values	25	15	41	Mostly European sites, several North American sites	Butterbach-Bahl et al (2011)

[1] N deposition values from the EMEP model for the year 2000
[2] N deposition values did not include several chemical species found in dry N deposition and organic N deposition
[3] These studies were not included in the value reported for Butterbach-Bahl et al (2011)