Forest Ecology and Management 258 (2009) 1814–1823

Contents lists available at ScienceDirect

Forest Ecology and Management

journal homepage: www.elsevier.com/locate/foreco

The impact of nitrogen deposition on carbon sequestration by European forests and heathlands

W. de Vries [a], S. Solberg [b], M. Dobbertin [c], H. Sterba [d], D. Laubhann [d], M. van Oijen [e], C. Evans [f], P. Gundersen [g], J. Kros [a], G.W.W. Wamelink [a], G.J. Reinds [a], M.A. Sutton [e,*]

[a] Alterra, Wageningen University and Research Centre, P.O. Box 47, 6700 AA Wageningen, The Netherlands
[b] Norwegian Forest and Landscape Institute, P.O. Box 115, N-1431 Ås, Norway
[c] Swiss Federal Institute for Forest, Snow and Landscape Research WSL, Switzerland
[d] BOKU-University of Natural Resources and Applied Life Sciences, Peter Jordan-Straße 82, A-1190 Vienna, Austria
[e] Centre for Ecology and Hydrology, Edinburgh Research Station, Bush Estate, Penicuik, Midlothian EH26 0QB, UK
[f] Centre for Ecology and Hydrology, Orton Building, Deiniol Road, Bangor LL57 2UP, UK
[g] Danish Centre for Forest, Landscape and Planning, KVL, Hoersholm, Denmark

ARTICLE INFO

Article history:
Received 7 February 2008
Received in revised form 13 February 2009
Accepted 25 February 2009

Keywords:
Carbon sequestration
C/N ratios
Nitrogen
Ecosystem production
Fertilizer experiments
Deposition
Forests
Heathlands
Environmental change

ABSTRACT

In this study, we present estimated ranges in carbon (C) sequestration per kg nitrogen (N) addition in above-ground biomass and in soil organic matter for forests and heathlands, based on: (i) empirical relations between spatial patterns of carbon uptake and influencing environmental factors including nitrogen deposition (forests only), (ii) ^{15}N field experiments, (iii) long-term low-dose N fertilizer experiments and (iv) results from ecosystem models. The results of the various studies are in close agreement and show that above-ground accumulation of carbon in forests is generally within the range 15–40 kg C/kg N. For heathlands, a range of 5–15 kg C/kg N has been observed based on low-dose N fertilizer experiments. The uncertainty in C sequestration per kg N addition in soils is larger than for above-ground biomass and varies on average between 5 and 35 kg C/kg N for both forests and heathlands. All together these data indicate a total carbon sequestration range of 5–75 kg C/kg N deposition for forest and heathlands, with a most common range of 20–40 kg C/kg N. Results cannot be extrapolated to systems with very high N inputs, nor to other ecosystems, such as peatlands, where the impact of N is much more variable, and may range from C sequestration to C losses.

1. Introduction

The productivity of many temperate ecosystems is nitrogen limited. Adding N via deposition thus has the potential to increase growth, and therefore to sequester CO_2 from the atmosphere. The increase in N deposition on forests may increase C sequestration by increased growth and increased accumulation of soil organic matter (SOM) through increased litter production and/or increased recalcitrance of N-enriched litter, leading to reduced long-term decomposition rates of organic matter. A range of studies has shown positive forest growth and C accumulation responses under low to moderate N additions (Vitousek and Howarth, 1991; Aber et al., 1995; Bergh et al., 1999; Franklin et al., 2003). Although fertilization by atmospheric N deposition is thus thought to have increased C storage in biomass and soils of terrestrial ecosystems (specifically of forests), estimates of the magnitude of this sink vary widely (Peterson and Melillo, 1985; Schindler and Bayley, 1993; Townsend et al., 1996; Holland et al., 1997).

Literature information suggests that the impact of nitrogen deposition on carbon sequestration in forests is highly uncertain and may vary by two orders of magnitude.

The scientific discussion on the response of carbon to nitrogen deposition has recently been intensified by a paper of Magnani et al. (2007) in Nature, who reported a strong positive relation between mean lifetime C sequestration (in terms of net ecosystem production; NEP_{av}) and N deposition, with data indicating a carbon response of approximately 725 kg C ha^{-1} $year^{-1}$ per kg N in wet deposition. Considering the ratio of wet to dry deposition mentioned by the authors, this would imply a net sequestration near 475 kg C/kg N of total deposition. A similar high carbon response to N deposition has been suggested before in modelling studies (e.g. Holland et al., 1997), by assuming that most (~80%) of the deposited N would be stored in woody biomass with a high C/N ratio (250–500). This implies a carbon response to N deposition of approximately 200–400 kg C/kg N. Using the upper limit of 400 kg C/kg N, this would suggest a global CO_2 sequestration in

* Corresponding author. Tel.: +44 131 445 4343; fax: +44 131 445 3943.
E-mail address: ms@ceh.ac.uk (M.A. Sutton).

0378-1127/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.foreco.2009.02.034

forest ecosystems up to 2.0 Pg year^{-1}, being nearly 30% of the estimated 7.1 Pg year^{-1} CO_2 release by man (5.5 fossil fuel and 1.6 from land use change and deforestation) due to N deposition. A much lower carbon response is, however, suggested by Nadelhoffer et al. (1999). By combining various tracer experiments, they showed that only a very small part of the added N (~5%) is stored in trees whereas most of the deposited N (~70%) is actually stored in soils with a much lower C/N ratio (10–30). Their data indicate a carbon response to N deposition near 50 kg C/kg N, corresponding to a global carbon sequestration of approximately 0.25 Pg year^{-1}. Currie et al. (2004), even suggest a net sequestration of approximately 5 kg C ha^{-1} year^{-1}/kg N deposition, based on model simulations for two forest types (red pine and mixed hardwoods) at Harvard Forest, USA, that best fitted decadal field data for pools and fluxes of C, N and ^{15}N.

Literature data, as described above, vary by two orders of magnitude (5–500 kg C/kg N). Reduction of this uncertainty is highly needed as it greatly influences the answer to the question whether N deposition drives global warming potential (GWP) in a positive or negative way (Sutton et al., 2007). For example, extra N deposition increases nitrous oxide emissions, which offsets the CO_2 sequestration effect. If the lowest value would be representative, it would imply that the net impact of N deposition on the sequestration of carbon is on average in the same order of magnitude as the N_2O estimate in response to N deposition, in terms of global warming potential. By contrast, if the highest value would be representative, the effect of N_2O emission would be very small compared to the extra C sequestration induced by N deposition and the CO_2 sequestration by forests could be about 1/3 to 1/2 of the estimated 7.1 Pg year^{-1} that is annually released by man, as earlier estimated by Holland et al. (1997).

The net impact of N deposition on the sequestration of carbon is not only a mere scientific issue, but also one that may have political consequences by influencing the next generation of international control protocols on N emission control. These protocols address the negative effects of nitrogen, such as eutrophication and acidification affecting biodiversity in terrestrial and aquatic ecosystems (Galloway et al., 2003). If forest carbon storage does respond very strongly to nitrogen deposition, policy makers might consider placing less focus on N emission reduction strategies given potential beneficial effects for greenhouse gas emissions and climate change. Proper information on the carbon response to N deposition is thus an important point to evaluate the trade-offs induced by excess nitrogen and the need for reducing N emissions and their effects induced by transboundary air pollution.

In this review, we present and compare estimated ranges in carbon sequestration per kg N addition in above-ground biomass and in soil organic matter for forests, with some attention to heathlands, based on four approaches:

- Correlations between spatial patterns of carbon uptake, based on measured forest growth or NEP, and influencing environmental factors including nitrogen deposition (forests only).
- Results of ^{15}N experimental studies on the fate of N, combined with C/N ratios in forest ecosystem compartments (forests only).
- Results of long-term (8–30 years) low-dose N fertilizer experiments on the C pool in biomass and soil.
- Model simulations predicting carbon response to environmental change.

In the text we use the term nitrogen use effeciciency (NUE_{eco}), defined here as carbon sequestration in the ecosystem in response to N deposition in kg C/kg N, distinguishing where relevant in tree response (NUE_{tree}) and soil response (NUE_{soil}). The results are evaluated in view of the above-mentioned dispute regarding the impact of N deposition on the terrestrial C sink in forests. Furthermore, we discuss the results in view if their applicability to other ecosystems, focusing on peatlands.

2. Nitrogen stimulated carbon sequestration in forested ecosystems

2.1. Empirical correlations between carbon uptake and nitrogen deposition

One approach to assess impacts of N deposition on carbon sequestration is to assess correlations between spatial patterns of carbon uptake, based on measured NEP or forest growth, and nitrogen deposition. In doing this, the impact of other influencing environmental factors on growth have to be included as well.

2.1.1. Studies in forest chronosequences

Magnani et al. (2007) reported a strong positive relation the NEP of representative forest chronosequences and the wet deposition of nitrogen, based on interpolated precipitation chemistry measurements for the period 1978–1994. Their data indicate a response of approximately 725 kg C ha^{-1} year^{-1} per kg N in wet deposition in the range between 4.9 and 9.8 kg N ha^{-1} year^{-1}. According to the authors, the maximum measured annual N wet deposition level of 9.8 kg N ha^{-1} year^{-1} is equivalent to a total deposition of 15 kg N ha^{-1} year^{-1}, implying an assumed total to wet deposition ratio near 1.5. Dividing the mentioned value of 725 with this ratio leads to a value of NUE_{eco} near 475 kg C/kg N. As demonstrated by Sutton et al. (2008), the assumed ratio of wet N deposition to total N deposition can vary from 2 to 7, the ratio generally increasing with N deposition (Simpson et al., 2006). Sutton et al. (2008) plotted the measured NEP_{av} data of Magnani et al. (2007) against both modelled wet N deposition and total N deposition data, using the EMEP model, leading to 177 kg C sequestration per kg total N deposition (R^2 = 0.88). Applying $N_{dep(tot)}$ for 1990, being more consistent with the period used by Magnani et al. (2007), gave NUE_{eco} is 126 kg C/kg N (R^2 = 0.87). When using dry deposition estimates according to the NEGTAP methodology (UK National Expert Group on Transboundary Air Pollution, 2001, Defra, London), NUE_{eco} reduced even further to 91 kg C/kg N. Using the re-interpreted results of Magnani et al. (2007) by Sutton et al. (2008) imply a carbon response to N deposition (NUE_{eco}) in the range between 91 and 177 kg C/kg N.

The above-mentioned range is still likely too high, owing to the fact that other factors that co-varied with N deposition may have contributed to the increasing NEP.

Sutton et al. (2008) showed, however, that these values were influenced by climatic differences across Europe, given that temperature and nitrogen deposition are positively correlated between the sites. By excluding water limited sites and accounting for an interaction between growing degree days above 5 °C and NEP_{av}, they found the data from Magnani et al. (2007) were consistent with a response of 68 kg C/kg N. Even the value of 68 kg C/kg N may be larger than the real nitrogen response. Magnani et al. (2007) combined their results for all tree species and their related site characteristics, such as site fertility and stand density, into one relationship. Site fertility may however correlate to N emission and deposition, as human activities and N emissions are expected to be most intense in regions having fertile soils, which would further reduce the N response. Overall, this analysis highlights the need for caution in interpreting spatial datasets where many factors co-vary, and it indicates a much smaller C:N response than suggested by Magnani et al. (2007).

Table 1
Multivariate regression results at stand level indicating the relative change in stem volume growth per unit change in influencing factor. Values for N deposition are given in bold. *Note*: a hyphen implies that the effect was statistically insignificant ($p > 0.05$) (after Solberg et al., 2009).

Tree species	Site prod[a]	Age[b]	SDI[c]	N_{dep}[d]	Drought[e]	Temp change[f]
All plots						
Norway spruce	0.054	−0.005	–	0.020[g]	–	0.524
Scots pine	–	−0.017	–	**0.010**	−0.0032	–
Sensitive plots						
Norway spruce	0.039	−0.004	–	**0.022**	–	0.32
Scots pine	–	−0.017	0.001	**0.013**	−0.002	–

[a] *Site prod* is a variable for site productivity (m^3 ha^{-1} $year^{-1}$) derived from selected European site index curves, with input variables being age and top height.
[b] *Age* is stand age (year).
[c] *SDI* = stand density index (indexed number of trees/ha).
[d] N_{dep} is total N deposition (kg ha^{-1} $year^{-1}$).
[e] *Drought* is a variable describing drought given as a relative value (in %) to the normal (30 years mean) drought stress at each site.
[f] *Temp change* is the temperature difference during the growing period compared with the 30-year average temperature (°C).
[g] Results from a simple linear regression gave a value of 0.020, but in the multivariate analysis, the coefficient was not significant at $p < 0.05$.

2.1.2. Studies at intensively monitored forest plots

The influence of site and environmental factors on measured forest growth data was investigated at stand level by Solberg et al. (2009) and at tree level by Laubhann et al. (2009) for nearly 400 intensively monitored forest plots in Europe, including Norway spruce, Scots pine, common beech, European oak and sessile oak. There was a large overlap in plots, but the number of plots differed due to different criteria for using plots, such as even aged stands in the stand level study and at least 20 measured tree individuals per plot with a basal area higher than 10% in the tree level study.

In the study by Solberg et al. (2009), the influence of nitrogen and acid deposition was considered at stand level by using the deposition during the growth period (1993–2000), while the impacts of temperature, precipitation and drought were addressed by taking the deviation of these climatic parameters in the growth period (1993–2000) from the 30-year mean. They accounted for site factors influencing measured tree growth, including site productivity, stand age and stand density, all at stand level. Relative stand growth, calculated as actual growth as a percentage of expected growth, that was derived from site productivity, stand age and a stand density index (SDI), was then correlated to the deposition and climatic factors. The statistical models included (i) simple regressions with a multivariate analysis-of-co-variance and (ii) a multivariate analysis, where actual growth was regressed against the site and stand factors and the growth affecting factors simultaneously. The analysis-of-co-variance model was done in a backward stepwise way, where the model was reduced step-by-step by removing non-significant effects, and applying sequential (Type I) *F*-tests.

Results of the multivariate analyses at stand level are shown in Table 1. The regression results indicate the relative change in stem volume growth per unit change in influencing factor. For example, a value of 0.01 for N deposition (see values in bold in Table 1) implies an increase of stem growth of 1.0% per kg N deposition The results indicated roughly a 1% increase in site productivity in response to a fertilizing effect of N deposition of 1 kg of N ha^{-1} $year^{-1}$ for Scots pine and 2% for Norway spruce. More information on the approach used and results obtained is given in Solberg et al. (2009). Similar results were obtained for a study with nationwide datasets for Norway (Solberg et al., 2004), although higher growth responses to N deposition (up to 4%) have also been observed in selected Swiss observation plots (Braun et al., 1999). For beech and oak, the response was not significant.

These responses for pine and spruce were recalculated in terms of C sequestration, by taking the product of the measured mean annual volume increment times the mean wood density times the estimated growth increase (the modelled regression slope of the relative growth residuals against N deposition, being around 1% for pine and 2% for spruce), assuming a C content of 50%. In converting volume increment to tree C changes, the increase in branches and woody roots is accounting for. The means of the models were 38 and 76 kg of additionally produced wood for pine and spruce, respectively, converting to 19–38 kg C/kg N (NUE_{tree}). Stronger responses were seen for N sensitive sites (high soil C/N ratio), having roughly a 1.3 and 2.2% increase in growth for pine and spruce, respectively, in response to a fertilizing effect of 1 kg N ha^{-1} $year^{-1}$.

Laubhann et al. (2009) applied a regression analysis at tree level, with measured basal area increment of each individual tree as responding factor and tree size (tree diameter at breast height, dbh), tree competition (basal area of larger trees, BAL, and stand density index), site factors (e.g. soil C/N ratio, 30-year average temperature) and environmental factors (temperature change compared to long-term average, nitrogen and sulphur deposition) as influencing parameters. The multivariate regression analysis at tree level was carried out by using tree size and tree competition variables on tree level and site factors and environmental factors on plot level, including plot as a random effect and applying the restricted maximum likelihood (REML) method for parameter estimation.

Results of the multivariate analyses at individual tree level, shown in Table 2, indicated a 1.2–1.5% increase in basal area

Table 2
Multivariate regression results at tree level indicating the relative change in basal area increment per unit change in influencing factor. Values for N deposition are given in bold. *Note*: Implies that the effect was statistically insignificant ($p > 0.05$) (after Laubhann et al., 2009).

Tree species	BAL[a]	SDI	C/N soil[b]	N_{dep}	Temp[c]	Temp change
Norway spruce	−0.39	−0.00056	−0.023	**0.013**	–	–
Scots pine	−0.29	−0.00066	–	**0.015**	0.053	–
Common beech	−0.16	–	–	**0.012**[d]	–	0.064
Oak	−0.38	−0.00062	–	**0.013**	0.080	–

[a] *BAL* is basal area of larger trees (m^2 ha^{-1})
[b] *C/N soil* is the C/N ratio of the mineral topsoil (0–30 cm).
[c] *Temp* is 30-year average temperature (°C).
[d] For common beech, the effect was almost significant at $p = 0.05$ ($p = 0.77$).

increment (coefficients varying between 0.012 and 0.015 relative increase), depending on tree species in response to a fertilizing N deposition effect of 1 kg N ha^{-1} $year^{-1}$. In this case, the response was significant for all included tree species. Relating an increase in basal area increment to an increase in carbon fixation in stem wood is not trivial. Laubhann et al. (2009) first plotted volume increment against basal area increment, limited to those trees where both dbh and height measurements were available, to confirm that volume increment was proportionally related to basal area increment. Referring to a total carbon uptake for European forests of 1729 kg C ha^{-1} $year^{-1}$ by De Vries et al. (2006), they then estimated the average response in terms of C sequestration between 20.7 and 25.8 kg C/kg N (1.2–1.5% of 1729), depending on tree species composition. In summary, the results of both studies indicate a response of trees between approximately 20 and 40 kg C/kg N on the basis of this wide European growth dataset in the period 1993–2000 (see also De Vries et al., 2008).

2.2. The fate of N in forest ecosystems and carbon to nitrogen ratios in various ecosystem compartments

The potential C fixation response to elevated N deposition is restricted by the C–N stoichiometry of the forest ecosystem compartments. Net ecosystem productivity may be defined as the net rate of C accumulation in ecosystems (Woodwell and Whittaker, 1968), which can either take place in the vegetation or the soil. Since C and N accumulate together in organic matter, the longer-term average accumulation of C per unit N in a compartment cannot exceed the C–N stoichiometry as described in Table 3. Because of the different C/N ratios, much more N is required to lock up C in soils, and also in foliage and roots, than in woody biomass. This aspect is the rationale behind using information on the fate of N in the soil to assess the related carbon sequestration.

A nitrogen use effeciency near 500 kg C/kg N, as mentioned by Holland et al. (1997) and Magnani et al. (2007) would imply that all deposited nitrogen ends up in stem wood. This is unreasonable since systems that are N-limited invest primarily in roots, not in wood (Brouwer, 1983; Cannell and Dewar, 1994). The implausibility of near total N uptake by stem wood follows also from the expected N leaching rates at elevated N deposition. Below a total N deposition of 10 kg N ha^{-1} $year^{-1}$, N leaching is not expected, as illustrated by N budgets for hundreds of forest sites across Europe and North America (MacDonald et al., 2002; Gundersen et al., 2006; De Vries et al., 2007). However, in the range between 15 and 25 kg N ha^{-1} $year^{-1}$, which is the likely range for total N deposition at the plots of Magnani et al. (2007), the N leaching rate varies generally between 10 and 50% of the N input (De Vries et al., 2007). An elevated leaching is also indicated by spring water concentrations of 2–3 mg N l^{-1} in the Hainich forest area (Günther, 2001), being a site in the chronosequence with a total N deposition near 25 kg N ha^{-1} $year^{-1}$.

The fate of new N entering forest ecosystems can also be traced by applying ^{15}N to the forest floor of sites and tracking where the ^{15}N went. Melin et al. (1983) thus studied the distribution and quantitative recovery of isotopically labelled fertilizer N in a strongly N-limited Scots pine stand in central Sweden. The fertilizer application rate was 100 kg ammonium nitrate-N per hectare, with either ammonium or nitrate being enriched with ^{15}N. On average, 79% of the supplied N was recovered in the ecosystem, the not recovered part presumably been lost by leaching. They found that between 12 and 28% of the supplied N was recovered in trees, half of which in the needle biomass and half in stem wood. In the above-ground parts of the shrub layer 3–13% of applied N was recovered. The recovery figure in the soil varied between 37 and 59%. Assuming an average C/N ratio in stem wood of 500 and an average soil C/N ratio of 30 (see below), this leads to an NUE_{tree} of 30–70 kg C/kg N and an NUE_{soil} of 11–18 kg C/kg N.

Nadelhoffer et al. (1999) carried out ^{15}N labelled tracer experiments in nine temperate forests during a 3-year period. Results indicated an average N retention fraction in stem wood of 0.05 only, while the largest part of the added N (70%) accumulated in the soil (Nadelhoffer et al., 1999). Assuming an average C/N ratio in stem wood of 500, which stays constant with N deposition, these authors estimated NUE_{tree} to be 25 kg C/kg N. Assuming an average soil C/N ratio of 30, they estimated a below-ground accumulation NUE_{soil} of 21 kg C/kg N. De Vries et al. (2006) used the approach by Nadelhoffer et al. (1999) in a spatially explicit on more than 6000 Level I plots, using site specific soil C/N ratios and assuming N retention fractions in stem wood that depend upon N deposition and increase up to 0.1. The upper value is closer to the results by Melin et al. (1983), who observed a value of 0.06–0.14 for N-limited forest stands. Using this approach, they found a European wide average NUE_{tree} and NUE_{soil} of 33 and 15 kg C/kg N, respectively.

The above-ground carbon sequestration may have been underestimated by Nadelhoffer et al. (1999), since the authors neglected the effect of direct foliar uptake (Jenkinson et al., 1999; Sievering, 1999). Recently, it was shown that canopy uptake of N directly stimulated NEE at a low deposition (4–8 kg N ha^{-1} $year^{-1}$) site in USA (Sievering et al., 2007). However, experimental evidence shows that the effect is likely to be small. In a large-scale experiment, adding 18–20 kg N ha^{-1} $year^{-1}$ to the canopy in mist using a helicopter, Gaige et al. (2007) found that 70% of added N appears retained or volatilized from the canopy, and suggested a possible 5% recovery in leaves. Furthermore, above-ground nitrogen uptake in foliage is not equal to the nitrogen retention in stem wood. Changes in N content in stem wood will reflect changes in total N uptake, consisting of both below-ground root uptake and above-ground foliar uptake, which both are affected by N deposition. More above-ground N uptake implies less N input to the soil and most likely less root N uptake, thus compensating for the above-ground effect.

2.3. Experimental N fertilization results

Hyvönen et al. (2008) investigated the impact of long-term nitrogen addition on carbon stocks in trees and soils in northern

Table 3
Data on C/N ratio in forest ecosystem compartments.

C accumulation—NEP	Compartment	C/N ratio typical conifer[a]	Probable max C/N including also broadleaf species
NEP vegetation	Wood and bark	325	500
	Canopy	65	100
	Roots incl. stump	65	100
	Total vegetation	150	250
NEP soil	Organic layer	33	45
	Mineral soil	29	35
	Total soil	30	40

[a] C/N ratios from a 80 years Norway spruce plantation at Klosterhede, Denmark (Gundersen, 1998).

Europe (Sweden and Finland). They quantified the effects of fertilizer N on C stocks in trees (stems, stumps, branches, needles, and coarse roots) and soils (organic layer +0–10 cm mineral soil) by analysing data from 15 long-term (14–30 years) experiments in *Picea abies* and *Pinus sylvestris* stands in Sweden and Finland. Low application rates (30–50 kg N ha^{-1} $year^{-1}$) were always more efficient per unit of N than high application rates (50–200 kg N ha^{-1} $year^{-1}$). Addition of a cumulative amount of N of 600–1800 kg N ha^{-1} resulted in a mean NUE_{tree} of 25 kg C/kg N and NUE_{soil} of 11 kg C/kg N. The NUE_{tree} strongly depended on soil N status and increased from close to zero at a C/N ratio of 25 in the humus layer, up to 40 kg C/kg N at a soil C/N ratio of 35. The NUE_{tree} decreased again to about 20 kg C/kg N at a soil C/N ratio of 50, when N only was added. In contrast, addition of NPK resulted in higher N-use efficiencies, also at N rich sites (soil C/N ratio of less than 25), reflecting a limitation of P and K for tree growth at these sites. Soil organic carbon (SOC) sequestration in response to N input (NUE_{soil}) was, on average, 3–4 times lower than tree C sequestration.

Högberg et al. (2006) reported effects of a long-term (30 years) N fertilization experiment, with annual N loading, on tree growth and soil chemistry in an unpolluted boreal forest. Ammonium nitrate was added to replicated 0.09 ha plots at two doses, of 34 and 68 kg N ha^{-1} $year^{-1}$, respectively. A third treatment of 108 kg N ha^{-1} $year^{-1}$ was terminated after 20 years, allowing assessment of recovery during the following 10 years. Tree growth initially responded positively to all N treatments, but the longer-term response depended on the rate of N addition, with no gain for the highest treatment and a gain of 100 m^3 ha^{-1} stem wood in excess of the control for the lowest N treatment. Assuming a tree wood density of 500 kg m^{-3} and a C content of 50%, this implies a net C gain of 25,000 kg C at an accumulated N input of 1020 kg (30 years $\times$ 34 kg N ha^{-1} $year^{-1}$) implying an NUE_{tree} of 25 kg C/kg N. Adding higher doses of N up to 108 kg N ha^{-1} $year^{-1}$ caused reduced growth again, likely due to acidification and losses of base cations from the mineral soil.

Pregitzer et al. (2007) experimentally simulated chronic N deposition by adding 30 kg N ha^{-1} $year^{-1}$ to four different northern hardwood forests in Michigan since 1994. Each year they measured tree growth and in 2004 they also examined the soil C content to a depth of 70 cm. Comparison of the control treatment with the N deposition treatment after a decade of experimentation (a total N input of 300 kg ha^{-1}) showed that C storage in woody biomass increased on average with 5000 kg C ha^{-1}, being equal to an NUE_{tree} of 17 kg C/kg N. The increase in soil organic matter (0–10 cm) was 6900 kg C ha^{-1}, being equal to an NUE_{soil} of 23 kg C/kg N. The total average ecosystem C response was thus 40 kg C/kg N. The soil C sequestration is relatively high compared to the Nordic sites mentioned before and may be affected by one experimental site where the amount of reported C accumulation is very high: 20,000 kg C ha^{-1} for 300 kg total N input during 10 years, leading to a soil C/N sequestration of 67.

In summary, experimental long-term (10–30 years) N fertilization results in Sweden, Finland and the USA have shown growth increases of nitrogen-limited forests, but responses are limited to an average of 20–25 kg carbon in the trees and 10–25 kg in soil per kilogram of nitrogen added to the ecosystem at rates of nitrogen addition comparable to (high) N deposition levels (below 50 kg N ha^{-1} $year^{-1}$) (see also Högberg, 2007).

2.4. Model simulations

As with ^{15}N addition and N fertilization experiments, simulations with process-based models also indicate an ecosystem response ranging mostly between 10 and 50 kg C/kg N. For example, Rehfuess et al. (1999) presented simulation results of five process-based models on two forest sites showing a variation in NUE_{eco} of 15–25 kg C/kg N depending on the model used. Levy et al. (2004) presented results of a Monte Carlo approach of three ecosystem models, CENTURY, BGC and Hybrid applied to a coniferous forest ecosystem in southern Sweden (the experimental site at Skogaby) for a period of 100 years. The change in total C content of the ecosystem with the cumulative change in N deposition over 100 years (NUE_{eco}) was estimated at 20.1 kg C/kg N with a standard deviation of 13.8 kg C/kg N. Variability in parameters accounted for 92% of the total uncertainty in this ratio. Only 8% was attributable to differences between models even though the models differed greatly in structure and parameterization. The most sensitive parameters were those which controlled the allocation of assimilates between fine roots, leaves, and stem. In all models, an increase in allocation to fine roots lead to a strong reduction in NIC_{eco} since fine roots have a very high turnover rate, and extra carbon allocated there is soon lost through mortality and decomposition.

In an analysis with a complex forest growth model (EFM), parameterized for Norway spruce and Scots pine, and tested against measurements from 22 forest locations across Europe, Milne and Van Oijen (2005) showed that the main driver of increased forest growth in the 20th century has been increased nitrogen deposition, rather than increased CO_2 concentration or climate change. The EFM model was also used to predict the effects of future environmental change, and suggested that climate change and CO_2 concentration may become the dominant environmental drivers for forest carbon exchange. The impact of N deposition was studied by repeating simulations with different values of N deposition, which gave on average a change in NEP of 41 kg C/kg N deposition (numbers derived from Milne and Van Oijen, 2005). Recently, four different models (EFM, EFIMOD, FinnFor and Q) were applied to the same 22 forest stands across Europe to examine to what extent changes from 1920 to 2080 in nitrogen deposition, atmospheric CO_2 concentration and six different weather variables have affected and will affect forest growth of pine and spruce forests across Europe (Van Oijen et al., 2008). As with the EFM model analysis by Milne and Van Oijen (2005), all models identified increasing nitrogen deposition as the major cause of observed changes in European forest growth during the 20th century, while future changes in forest growth are more likely to be caused by increasing atmospheric CO_2 concentration and, especially in northern latitudes, by increasing temperature (Van Oijen et al., 2008). The paper presents information on the net primary productivity (NPP) divided by total N uptake, but unfortunately, the paper does not permit calculation of NUE_{eco}.

Sutton et al. (2008) reported simulated NEP responses to total N deposition using the process-based biogeochemistry models EFM, CENTURY and BGC applied to: (i) the same coniferous forest as simulated by Levy et al. (2004) through an entire rotation (100 years) and (ii) the 22 forest stands across Europe of the EU RECOGNITION project used by Van Oijen et al. (2008). In the first simulation, Sutton et al. (2008) using the same inputs Levy et al. (2004) but using smaller values of total N deposition. For a total N deposition range of 6–26 kg N ha^{-1} $year^{-1}$, the modelled NUE_{eco} by EFM, CENTURY and BCG were 75, 58 and 43 kg C/kg N, respectively. In the modelled N deposition range, a N saturation effect was revealed by EFM and CENTURY, implying a lower C response at elevated N deposition. In the second simulation, total N deposition at each of the RECOGNITION forest stands was calculated as the mean over the lifetime of each stand (33–125 years) and the evaluation was limited to the EFM model. For these stands, an average NUE_{eco} of 54 kg C/kg N was simulated (Sutton et al., 2008).

Wamelink et al. (2009a) evaluated the impact of N deposition on forest growth by applying the succession model SUMO2 to Dutch forests, using a spatial resolution of 250 m $\times$ 250 m grid

Table 4
Carbon sequestration of European forest in 2070 compared to a reference run for the N deposition per latitude class (after Wamelink et al., 2009b).

Latitude	ΔCseq (kg C ha^{-1} year^{-1})			NUE or ΔCseq/ΔN (kg C/kg N)		
	Tree	Soil	Total	Tree	Soil	Ecosystem
40–50	−32	−35	−67	3.6	5.0	8.6
50–60	−25	−31	−56	3.1	4.2	7.3
60–70	−13	−13	−26	12	11	24

cells (109,374 and 38,707 cells for coniferous and deciduous forests, respectively). They simulated an increase in average net carbon sequestration in living biomass, litter and dead wood from 0 to 1.1 ton ha^{-1} year^{-1} for coniferous forest and from 0.4 to 2.2 ton ha^{-1} year^{-1} for deciduous forest between the lowest (5 kg N ha^{-1} year^{-1}) and the highest nitrogen deposition level (70 kg N ha^{-1} year^{-1}). The average simulated increase was 20–30 kg C/kgN.

In another study, Wamelink et al. (2009b) used the SUMO2 model, combined with the soil model SMART2 and the hydrological model WATBAL to predict the effects of a change in CO_2 concentration, climatic parameters (temperature and precipitation) and nitrogen deposition on carbon sequestration in 166 intensively monitored forest plots in Europe. The predicted effects of a change in the investigated environmental variables on soil carbon sequestration were generally lower than on carbon sequestration by the trees (especially the response to changes in climate and CO_2) but the magnitude was similar. In the study, future nitrogen deposition was assumed to decrease, causing a decrease in carbon accumulation in both trees and soil all over Europe compared to a reference run in which N deposition was assumed constant. The change in carbon sequestration was largest in the Southern countries and smallest in the Nordic countries, in accordance with the change in N deposition. However, when expressed per kg N change the effect was largest in the Nordic countries, as shown in Table 4. On average, the ratio in C sequestration per kg N deposition was comparable for trees and soil and decreased from an average NUE_{eco} near 25 kg C/kg N in the Nordic countries to a value near 8 kg C/kg N in Central and Southern Europe. These low results are in line with model simulation by Currie et al. (2004), who found a carbon sequestration of 5 kg C/kg N. In these areas, values ranged mostly between 1 and 20 kg C/kg N for both above- and below-ground biomass with some values going up to 30 kg C/kg N. The comparatively low C response to N deposition calculated by Wamelink et al. (2009b), also compared to earlier results by Wamelink et al. (2009a), indicate that the carbon response to a decrease in nitrogen deposition may differ from increasing nitrogen deposition. Apparently, the N-pool does not decrease as rapidly in response to decreased N deposition as it was built up due to elevated N deposition, implying a slow decrease in N availability for the vegetation and thus a limited effect on NPP and carbon sequestration.

Despite the variation, the various model results are generally quite consistent and most of them show an average variation in NUE_{eco} between 15 and 40 kg C/kg N depending on the model used and the forest compartment considered (only trees or trees and soil), with an overall range between 5 and 75 kg C/kg N.

3. Nitrogen stimulated carbon sequestration in heathlands

In general, there are very few studies allowing a quantitative assessment of the carbon response per kg N addition for heathlands. Here we summarize results for several UK ecosystems based on long-term (ca. 10–15 years) low to medium dose N fertilizer experiments and modelling (heathlands). Evidence of carbon accumulation in response to N addition is available for three UK heathlands N manipulation sites on mineral sandy (podzolic) soils. The first site, Ruabon, is an upland (470 m) heath in North Wales dominated by heather (*Calluna vulgaris*) on a well-drained ironpan stagnopodzol. The manipulation experiment, established in 1989, includes a control treatment plus three N addition treatments of 40, 80 and 120 kg N ha^{-1} year^{-1}, added monthly as finely sprinkled NH_4NO_3 solution. Research at the site has focused on the N dynamics of the system (Pilkington et al., 2005a,b) and impacts of N on plant vitality (Carroll et al., 1999). Effects of N on C accumulation were examined by Evans et al. (2006), based on measured pool changes after 11 years of N addition, which allowed C/N sequestration ratios to be calculated for both vegetation and surface organic soils (Table 5). The system has shown remarkably clear responses to N addition, including increases in above-ground biomass and litter, and consequent increases in both vegetation and soil C and N storage. Measured below-ground C/N sequestration ratios (i.e. NUE_{soil}) declined (as in the Scandinavian forest N addition experiments described above) from 34 to 20 kg C/kg N, coinciding with a decrease in organic soil C/N ratio from 33.4 to 30.7. The resulting NUE_{soil} are thus comparable to the C/N ratio of the soil itself. Modelled NUE_{soil}, based on an application of the MAGIC model to all treatments, was 28 kg C/kg N.

The second site, Budworth, is a lowland heath located in Northwest England dominated by heather (*C. vulgaris*, with small amounts of *Deschampsia flexuosa*) on humo-ferric podzol soils. Treatments at Budworth began in 1996, with an experimental design similar to that at Ruabon, and NH_4NO_3 additions of 20, 60 and 120 kg N ha^{-1} year^{-1}. Unlike the strongly N-retaining Ruabon site, a significant proportion of N addition at Budworth is lost from the site as inorganic N leaching.

At Budworth, no measurements of soil C pools were available to calculate a NUE_{soil}, but application of the MAGIC model (Evans et al. (2006) best reproduced observed C/N changes with simulated NUE_{soil} being 21 kg C/kg N.

The third site studied is Thursley, a lowland heathland on humo-ferric podzol in southeast England. Plots were subjected to 7.7 and 15.4 kg N ha^{-1} year^{-1}doses of NH_4NO_3 solution from 1989 to 1997 (Power et al., 1998). Soil sampling and destructive vegetation harvesting show accumulation of N and C in above-

Table 5
Estimated soil carbon sequestration per kg nitrogen addition and soil C/N ratios at the Ruabon heathland N manipulation site (after Evans et al., 2006).

N input in 11 years (kg ha^{-1})	Soil C pool (kg ha^{-1})	Change in soil C pool compared to ambient (kg ha^{-1})	NUE or ΔCseq/ΔN (kg C/kg N)	C/N soil (kg C/kg N)
0	105,360	–	–	33.4
440	120,360	15,000	34.1	31.9
880	125,640	20,280	23.0	31.2
1320	131,880	26,520	20.1	30.7

Table 6
Estimated ranges in carbon sequestration per kg nitrogen addition in above and below-ground biomass in heathlands at sites in the UK.

Heathland site	Carbon sequestration (kg C/kg N)		Approach	Author
	Above ground	Below ground		
Ruabon (upland heath)	15	34	Observed, 40 kg N ha^{-1} $year^{-1}$ addition	Evans et al. (2006)
	5	23	Observed, 80 kg N ha^{-1} $year^{-1}$ addition	
	9	20	Observed, 120 kg N ha^{-1} $year^{-1}$ addition	
	–	28	Simulated	Evans et al. (2006)
Budworth (sandy soil)	–	21	Simulated	Evans et al. (2006)
Thursley	7	−12[a]	Observed, 8 kg N ha^{-1} $year^{-1}$ addition	Evans (personal communication)
	9	33	Observed, 15 kg N ha^{-1} $year^{-1}$ addition	
	–	32	Simulated	

[a] The decrease in the Thurley low treatment at low N deposition may have been an artefact of the sampling and analysis. Litter C certainly increased in this treatment, but because of one low %C measurement there appears to have been a decrease in the C pool of the A horizon, which is much harder to define. The treatments at Thursley are also much smaller than at Ruabon, so we are looking for more subtle changes.

ground biomass under both treatments but with a fairly low C/N sequestration ratio (Table 6; Evans, personal communication). Below-ground data show C and N accumulation in litter under both treatments, and in the underlying A horizon of the soil in the higher N treatment, but an apparent loss of C from the A horizon (and therefore the combined litter plus soil) in the lower N treatment. Since this result stems from a single low A horizon %C measurement, and changes are relatively subtle due to the low level of N addition, it should be treated with caution. Under the higher N treatment, substantial below-ground C accumulation was recorded, equating to a measured NUE_{soil} of 33 kg C/kg N (MAGIC simulated value 32 kg C/kg N).

These heathland studies, based on both experimental and modelling data, show similar results for below-ground soil C accumulation in response to N input as obtained for forests by Nadelhoffer et al. (1999) and De Vries et al. (2006), with NUE_{soil} ranging from 20 to 35 kg C/kg N. This suggests that long-term below-ground C accumulation due to N deposition in unforested heathland ecosystems may be similar to that for forests. However, as would be expected given the smaller above-ground biomass, above-ground C sequestration is very much lower, with NUE_{plant} varying from 7 to 15 kg C/kg N (Table 6). Furthermore, unlike managed forests, where tree removal causes a continuous C sink, in non-forest ecosystems the net C sequestration is ultimately negligible with the exception of from managed ecosystems, such as mown grasslands or heathlands with sod cutting.

4. Discussion and conclusions

A summary of the derived estimates is shown in Table 7. With one exception, the figures are in close agreement and show that the range in above-ground accumulation of carbon in forests is 15–40 kg C/kg N. For heathlands and moorlands, values are lower. A range of 5–15 kg C/kg N has been observed based on low-dose N fertilizer experiments, with the caveat that at high rates of N deposition peatlands may actually lose existing carbon stocks. The uncertainty in carbon sequestration per kg N addition in soils is larger than for above-ground biomass although the results are also quite consistent and vary on average between 5 and 35 kg C/kg N for both forests and heathlands, being in the same order of magnitude as above-ground accumulation in forests. These figures indicate a total carbon sequestration in a range of 5–75 kg C/kg N for forests, with a most probable range between 20 and 40 kgC/kgN, considering the results of empirical relations derived from field data and N fertilizer experiments (see discussion on uncertainties below). For heathlands on mineral soils, the overall range is 20–50 kg C/kg N deposition. The only response that is clearly higher is the result by Magnani et al. (2007), re-interpreted by Sutton et al. (2008), unless the lower value of 68 kg C/kg N is used, including (i) a proper total N deposition estimate and (ii) accounting for the influence of climatic differences across Europe that co-varied with N deposition.

The range presented above should also be interpreted with some care due to limitations of the various approaches. The approach to combine ^{15}N tracer additions and C/N ratios of forest ecosystem compartments for example assumes no change in the C/N ratio of the systems due to N addition and this is questionable. After addition of ^{15}N, e.g. bacteria become labelled, but their biomass C does not increase because they are C-limited. Furthermore, the response of field data may not be applicable in very high N deposition ranges. In general, terrestrial ecosystems will only respond to elevated N inputs if they are N-limited. Since nitrogen often is the limiting nutrient in forests, nitrogen deposition does generally increase wood production and accumulation of soil organic matter, thus increasing C sequestration into the forest. With increasing N-enrichment, N immobilization will decrease (N leaching will increase) and C/N will decline, and consequently less C will be sequestered per unit N deposition. This effect is likely to occur in high N deposition areas. In such areas, the N fertilization may not even be beneficial anymore but it may lead to adverse growth effects due to impacts of nitrogen induced eutrophication and acidification on forest health. The nitrogen saturation hypothesis (Aber et al., 1989, 1998) predicts that the final stages of N saturation lead to tree decline and even death. Some studies also report decreased growth in response to high N-loads (e.g. Magill et al., 1997, 2004). In the case of additions of 150 kgN ha^{-1} $year^{-1}$ for 15 years, high tree mortality was induced (Magill et al., 2004). Furthermore, Boxman et al. (1998) observed a growth improvement in a highly N-saturated Scots pine stand in which the N input to the forest floor was reduced from $\approx$60 to <5 kg N ha^{-1} $year^{-1}$ by means of a transparent roof and application of clean, artificial throughfall water.

The uncertainty in soil carbon response to N deposition may also be larger than the range presented in this paper. Magill et al. (2004) for example found no significant change in soil carbon after 15 years of N addition to Harvard Forest. N additions to forest soils have also been found to lower the C/N ratio without causing major changes in the total amount of soil carbon (Neilsen et al., 1992; Harding and Jokela, 1994). Even increases in soil respiration, implying short-term carbon loss, in response to N fertilization have been reported (Brumme and Beese, 1992; Gallardo and Schlesinger, 1994; Bowden et al., 2004). In a recent review paper, Reay et al. (2008) reports a range of 0–23 kg C/kg N for forest soils, being a bit wider than our range of 5–23 kg C/kg N for forest soils (Table 4).

It is finally important to realize that the above-mentioned ranges in carbon sequestration in forests and heathlands in response to nitrogen deposition cannot be extrapolated to other

Table 7
Estimated ranges in carbon sequestration per kg nitrogen addition in above and below-ground biomass in forest at various scales.

Approach	Carbon sequestration (kg C/kg N)			Scale of application	Author
	Above ground	Below ground	Total		
Forests					
Empirical field data					
Correlation between NEP and total N deposition	–	–	68–177	Chronosequences (5) in boreal and temperate forests of Eurasia and North America	Magnani et al. (2007) as re-evaluated by Sutton et al. (2008)
Correlation between the average growth increase of nearly 400 intensive monitoring plots and N deposition in a multivariate analysis	15–38	–	–	Nearly 400 intensive forest monitoring plots	Solberg et al. (2009) and Laubhann et al. (2009)
^{15}N experimental data					
Extrapolation of ^{15}N experimental data with average C/N ratios of forest ecosystem compartments	30–70	11–18	41–88	One forest site in Sweden	Melin et al. (1983)
Extrapolation of ^{15}N experimental data with average C/N ratios of forest ecosystem compartments	25	21	46	Generic average	Nadelhoffer et al. (1999)
Extrapolation of ^{15}N experimental data with site specific data at 6000 plots in Europe	33	15	48	European average	De Vries et al. (2006)
Results of fertilizer experiments					
Average results from 30-year low-dose (34 kg N ha^{-1} $year^{-1}$) fertilizer experiments	25	–	–	Forest plot in Sweden	Högberg et al. (2006)
Average results from 14 to 30 years fertilizer experiments	25	11	36	Two forest plots in Sweden and Finland	Hyvönen et al. (2008)
Average results from 10 years chronic N addition (30 kg N ha^{-1} $year^{-1}$) experiments	17	23	40	Four forest plots in the USA	Pregitzer et al. (2007)
Results of model simulations					
Range in results of three process-based models	–	–	10–30	One forest site in Sweden	Levy et al. (2004)
Range in results of three process-based models	–	–	43–75	One forest site in Sweden	Sutton et al. (2008)
Range in results of five process-based models	15–25	–	–	Two forest plots in UK	Rehfuess et al. (1999)
Average result of the process-based model EFM	–	–	41–54	22 forest plots in Europe	Milne and Van Oijen (2005) and Sutton et al. (2008)
Range in results of the process-based model SUMO	20–30	–	–	Dutch forests	Wamelink et al. (2009a)
Range in average results per latitude of the process-based model chain SMART2-SUMO2	3–12	5–11	7–24	166 forest plots in Europe	Wamelink et al. (2009b)
Heathlands					
Results from 5- to 11-year N fertilizer experiments at 20–120 kg N ha^{-1} $year^{-1}$	5–15	20–34	25–49	2 heathland plots	Evans et al. (2006) and Evans (personal communication)
Model simulations for the N fertilizer experiment sites	–	21–32	–	3 heathland plots	Evans et al. (2006) and Evans (personal communication)

systems, which show a range of responses to N additions. Examples are enhanced C sequestration in an Arctic wet sedge system (Johnson et al., 2000); no overall change in C storage in an alpine meadow system (Neff et al., 2002) and decreased C sequestration (C loss) in European peat bogs (Bragazza et al., 2006) and Alaskan tundra (Mack et al., 2004). Specifically the response of peatlands to N deposition may vary, but there are currently very few studies available on the effect of N deposition on the accumulation or release of C in peatlands. Bragazza et al. (2006) show increased C loss from peat under elevated N deposition due to increased litter decomposition rates (elevated microbial activity) rather than plant species change. In situations of very high nitrogen deposition, peat C/N ratios may decrease substantially, potentially increasing the rates of heterotrophic respiration. Combined with the species changes, substantial degradation of peatland may then occur, leading to major C losses. Mack et al. (2004) presented results of a long-term (over 20 years) fertilization experiment in Alaskan tundra, showing that increased nutrient availability had a larger effect on decomposition than on plant production. Above-ground carbon storage increased due to accumulation of woody shrub biomass and litter, but this was offset by a larger decrease of C in belowground pools, resulting in a net loss of almost 20,000 kg C ha^{-1} over 20 years. Mack et al. (2004) presented various hypothesis for the effect, focusing on nutrient stimulation of decomposer activity, such as shifts in microbial community composition from fungal to bacterial dominance in response to decreased C:N ratios.

In principle, there may be several offsetting effects of N deposition on C sequestration response by peatlands. At low N deposition, additional atmospheric nitrogen deposition may stimulate net primary productivity as in other systems. However, at medium rates of N deposition (e.g. above critical loads), N-induced eutrophication leads to vegetation change, particularly a shift from species producing recalcitrant litter to those producing more decomposable material and most notably to the loss of peat-forming species such as *Sphagnum*, with their replacement by grasses and mosses (e.g. Berendse et al., 2001). This may reduce or even reverse the positive effect of N deposition on C sequestration, putting existing peatland carbon stocks at risk.

Much more knowledge is needed on the actual N deposition values that apply to these hypothesized phases. Current empirical critical loads for ombrotrophic peatlands (related to species changes and especially a decline in *Sphagnum* cover) are set at 5–10 kg N ha^{-1} $year^{-1}$ (Bobbink et al., 2003). But it is unclear at what value potential gains in higher plant GPP are more than offset by changes in heterotophic respiration, linked to losses of peat-forming plant species. For example, Bragazza et al. (2006) found a shift from positive to negative impacts on C sequestration in peatlands when atmospheric nitrogen deposition exceeded 10 kg N ha^{-1} $year^{-1}$. On the other hand, one of the heathland sites above, Ruabon, is an organo-mineral soil (~20 cm peat layer) that has shown increased C stocks after a decade of inputs at 120 kg N ha^{-1} $year^{-1}$ in the highest treatment, despite some decrease in C/N. It thus remains an open question whether and to what extent atmospheric nitrogen is currently leading to an overall increase or decrease in the carbon stored in European peatlands.

Acknowledgements

The authors gratefully acknowledge funding for this review by the European Commission DG Agriculture within the context of Forest Focus, under the project "carbon sequestration" and the DG Research, 6th Framework Programme under the NitroEurope Integrated Project (017841).

References

Aber, J.D., Nadelhoffer, K.J., Steudler, P., Melillo, J.M., 1989. Nitrogen saturation in northern forest ecosystems. Bioscience 39, 378–386.

Aber, J.D., Magill, A., McNulty, S.G., Boone, R.D., Nadelhoffer, K.J., Downs, M., Hallett, R., 1995. Forest biogeochemistry and primary production altered by nitrogen saturation. Water, Air, and Soil Pollution 85, 1665–1670.

Aber, J.D., McDowell, W., Nadelhoffer, K., Magill, A., Berntsen, G., Kamakea, M., McNulty, S., Currie, W., Rustad, L., Fernandez, I., 1998. Nitrogen saturation in temperate forest ecosystems: hypothesis revisited. Bioscience 48, 921–934.

Berendse, F., van Breemen, N., Rydin, H., Buttler, A., Heijmans, M., Hoosbeek, M.R., Lee, J.A., Mitchell, E., Saarinen, T., Vasander, H., Wallen, B., 2001. Raised atmospheric CO_2 levels and increased N deposition cause shifts in plant species composition and production in *Sphagnum bogs*. Global Change Biology 7, 591–598.

Bergh, J., Linder, S., Lundmark, T., Elfving, B., 1999. The effect of water and nutrient availability on the productivity of Norway spruce in northern and southern Sweden. Forest Ecology and Management 119, 51–62.

Bobbink, R., Ashmore, M., Braun, S., Flückiger, W., van den Wyngaert, I.J.J., 2003. Empirical nitrogen critical loads for natural and semi-natural ecosystems: 2002 update. In: Achermann, B., Bobbink, R. (Eds.), Empirical Critical Loads for Nitrogen. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne, pp. 43–170.

Bowden, R.D., Davidson, E., Savage, K., Arabia, C., Steudler, P., 2004. Chronic nitrogen additions reduce total soil respiration and microbial respiration in temperate forest soils at the Harvard Forest. Forest Ecology and Management 196, 43–56.

Boxman, A.W., van der Ven, P.J.M., Roelofs, J.G.M., 1998. Ecosystem recovery after a decrease in nitrogen input to a Scots pine stand at Ysselsteyn, the Netherlands. Forest Ecology and Management 101, 155–163.

Bragazza, L., Freeman, C., Jones, T., Rydin, H., Limpens, J., Fenner, N., Ellis, T., Gerdol, R., Hajek, M., Hajek, T., Iacumin, P., Kutnar, L., Tahvanainen, T., Toberman, H., 2006. Atmospheric nitrogen deposition promotes carbon loss from peat bogs. Proceedings of the National Academy of Sciences 103, 19386–19389.

Braun, S., Rihm, B., Schindler, C., Flückiger, W., 1999. Growth of mature beech in relation to ozone and nitrogen deposition: an epidemiological approach. Water, Air, and Soil Pollution 116, 357–364.

Brouwer, R., 1983. Functional equilibrium: sense or nonsense. Netherlands Journal of Agricultural Science 31, 335–348.

Brumme, R., Beese, F., 1992. Effects of liming and nitrogen fertilization on emissions of CO_2 and N_2O from a temperate forest. Journal of Geophysical Research 97, 851–858.

Cannell, M.G.R., Dewar, R.C., 1994. Carbon allocation in trees: a review of concepts for modelling. Advances in Ecological Research 20, 59–104.

Carroll, J.A., Caporn, S.J.M., Cawley, L., Read, D.J., Lee, J.A., 1999. The effect of increased deposition of atmospheric nitrogen on *Calluna vulgaris* in upland Britain. New Phytologist 141, 423–431.

Currie, W.S., Nadelhoffer, K.J., Aber, J.D., 2004. Redistributions of 15^N highlight turnover and replenishment of mineral soil organic N as a long-term control on forest C balance. Forest Ecology and Management 196, 109–127.

De Vries, W., Reinds, G.J., Gundersen, P., Sterba, H., 2006. The impact of nitrogen deposition on carbon sequestration in European forests and forest soils. Global Change Biology 12, 1151–1173.

De Vries, W., van der Salm, C., Reinds, G.J., Erisman, J.W., 2007. Element fluxes through European forest ecosystems and their relationships with stand and site characteristics. Environmental Pollution 148, 501–513.

De Vries, W., Solberg, S., Dobbertin, M., Sterba, H., Laubhann, D., Reinds, G.J., Nabuurs, G.J., Gundersen, P., Sutton, M.A., 2008. Ecologically implausible carbon response? Nature 451, E1–E3.

Evans, C.D., Caporn, S.J.M., Carroll, J.A., Pilkington, M.G., Wilson, D.B., Ray, N., Cresswell, N., 2006. Modelling nitrogen saturation and carbon accumulation in heathland soils under elevated nitrogen deposition. Environmental Pollution 143, 468–478.

Franklin, O., Högberg, P., Ekblad, A., Ågren, G.I., 2003. Pine forest floor carbon accumulation in response to N and PK additions—bomb ^{14}C modelling and respiration studies. Ecosystems 6, 644–658.

Gaige, E., Dail, D.B., Hollinger, D.Y., Davidson, E.A., Fernandez, I.F., Sievering, H., White, A., Halteman, W., 2007. Changes in canopy processes following whole-forest canopy nitrogen fertilization of a mature spruce-hemlock forest. Ecosystems 10, 1133–1147.

Gallardo, A., Schlesinger, W.H., 1994. Factors limiting microbial biomass in the mineral soil and forest floor of a warm-temperate forest. Soil Biology & Biochemistry 26, 1409–1415.

Galloway, J.N., Aber, J.D., Erisman, J.W., Seitzinger, S.P., Howarth, R.W., Cowling, E.B., Cosby, B.J., 2003. The Nitrogen Cascade. Bioscience 53, 341–356.

Gundersen, P., 1998. Effects of enhanced nitrogen deposition in a spruce forest at Klosterhede, Denmark, examined by moderate NH_4NO_3 addition. Forest Ecology and Management 101, 251–268.

Gundersen, P., Schmidt, I.K., Raulund-Rasmussen, K., 2006. Leaching of nitrate from temperate forests—effects of air pollution and forest management. Environmental Reviews 14, 1–57.

Günther, B., 2001. Wasserqualität an den Quellen im Bereich der Wald- und Hauptmessstationen. In: Forstliches Umweltmonitoring in Thüringen, Mitteilungen der Landesanstalt für Wald und Forstwirtschaft, Heft 19/2001, pp. 103–124.

Harding, R.B., Jokela, E.J., 1994. Long-term effects of forest fertilization on site organic matter and nutrients. Soil Science Society of America Journal 58, 216–221.

Högberg, P., 2007. Nitrogen impacts on forest carbon. Nature 447, 781–782.

Högberg, P., Fan, H., Quist, M., Binkley, D., Tamm, C.O., 2006. Tree growth and soil acidification in response to 30 years of experimental nitrogen loading on boreal forest. Global Change Biology 12, 489–499.

Holland, E.A., Braswell, B.H., Lamarque, J.F., Townsend, A.R., Sulzman, J., Müller, J.F., Dentener, F., Brasseur, G., Levy II, H., Penner, J.E., Roelofs, G.J., 1997. Variations in the predicted spatial distribution of atmospheric nitrogen deposition and their impact on carbon uptake by terrestial ecosystems. Journal of Geophysical Research 102, 15849–15866.

Hyvönen, R., Persson, T., Andersson, S., Olsson, B., Ågren, G.I., Linder, S., 2008. Impact of long-term nitrogen addition on carbon stocks in trees and soils in northern Europe. Biogeochemistry 89, 121–137.

Jenkinson, D.S., Goulding, K., Powlson, D.S., 1999. Reply to nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. Nature 400, 629.

Johnson, L.C., Shaver, G.R., Cades, D.B., Rastetter, E., Nadelhoffer, K., Giblin, A., Laundre, J., Stanley, A., 2000. Plant carbon–nutrient interactions control CO_2 exchange in Alaskan wet sedge tundra ecosystems. Ecology 81, 453–469.

Laubhann, D., Sterba, H., Reinds, G.J., de Vries, W., 2009. The impact of atmospheric deposition and climate on forest growth in European monitoring plots: an individual tree growth model. Forest Ecology and Management 258, 1751–1761.

Levy, P.E., Wendler, R., Van Oijen, M., Cannell, M.G.R., Millard, P., 2004. The effect of nitrogen enrichment on the carbon sink in coniferous forests: uncertainty and sensitivity analyses of three ecosystem models. Water, Air, and Soil Pollution: Focus 4, 67–74.

MacDonald, J.A., Dise, N.B., Matzner, E., Armbruster, M., Gundersen, P., Forsius, M., 2002. Nitrogen input together with ecosystem nitrogen enrichment predict nitrate leaching from European forests. Global Change Biology 8, 1028–1033.

Mack, M.C., Schuur, E.A.G., Bret-Harte, M.S., Shaver, G.R., Chapin III, F.S., 2004. Ecosystem carbon storage in arctic tundra reduced by long-term nutrient fertilization. Nature 431, 440–443.

Magill, A.H., Aber, J.D., Hendricks, J.J., Bowden, R.D., Melillo, J.M., Steudler, P.A., 1997. Biogeochemical response of forest ecosystems to simulated chronic nitrogen deposition. Ecological Applications 7, 402–415.

Magill, A.H., Aber, J.D., Currie, W., Nadelhoffer, K., Martin, M., McDowell, W.H., Melillo, J.M., Steudler, P., 2004. Ecosystem response to 15 years of chronic nitrogen additions at the Harvard Forest LTER, Massachusetts, USA. Forest Ecology and Management 196, 7–28.

Magnani, F., Mencuccini, M., Borghetti, M., Berbigier, P., Berninger, F., Delzon, S., Grelle, A., Hari, P., Jarvis, P.G., Kolari, P., Kowalski, A.S., Lankreijer, H., Law, B.E., Lindroth, A., Loustau, D., Manca, G., Moncrieff, J.B., Rayment, M., Tedeschi, V., Valentini, R., Grace, J., 2007. The human footprint in the carbon cycle of temperate and boreal forests. Nature 447, 848–850.

Melin, J., Nommik, H., Lohm, U., Flower-Ellis, J., 1983. Fertilizer nitrogen budget in a Scots pine ecosystem attained by using root-isolated plots and ^{15}N tracer technique. Plant and Soil 74, 249–263.

Milne, R., Van Oijen, M., 2005. A comparison of two modelling studies of environmental effects on forest carbon stocks across Europe. Annals of Forest Science 62, 911–923.

Nadelhoffer, K.J., Emmett, B.A., Gundersen, P., Kjønaas, O.J., Koopmans, C.J., Schleppi, P., Tietema, A., Wright, R.F., 1999. Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. Nature 398, 145–148.

Neff, J.C., Townsend, A.R., Gleixner, G., Lehman, S.J., Turnbull, J., Bowman, W.D., 2002. Variable effects of nitrogen additions on the stability and turnover of soil carbon. Nature 419, 915–917.

Neilsen, W.A., Pataczek, W., Lynch, T., Pyrke, R., 1992. Growth response of *Pinus radiata* to multiple applications of nitrogen fertilizer and evaluation of the quantity of added nitrogen remaining in the forest system. Plant and Soil 144, 207–217.

Peterson, B.J., Melillo, J.M., 1985. The potential storage of carbon caused by eutrophication of the biosphere. Tellus 37B, 117–127.

Pilkington, M.G., Caporn, S.J.M., Carroll, J.A., Cresswell, N., Lee, J.A., Emmett, B.A., Johnson, D., 2005a. Effects of increased deposition of atmospheric nitrogen on an upland *Calluna moor*: N and P transformations. Environmental Pollution 135, 469–480.

Pilkington, M.G., Caporn, S.J.M., Carroll, J.A., Cresswell, N., Lee, J.A., Ashenden, T.W., Brittain, S.A., Reynolds, B., Emmett, B.A., 2005b. Effects of increased deposition of atmospheric nitrogen on an upland moor: leaching of N species and soil solution chemistry. Environmental Pollution 135, 29–40.

Power, S.A., Ashmore, M.R., Cousins, D.A., 1998. Impacts and fate of experimentally enhanced nitrogen deposition on a British lowland heath. Environmental Pollution 102, 27–34.

Pregitzer, K.S., Burton, A.J., Zak, D.R., Talhelm, A.F., 2007. Simulated chronic nitrogen deposition increases carbon storage in Northern Temperate forests. Global Change Biology 14, 1–12.

Reay, D.S., Dentener, F., Smith, P., Grace, J., Feely, R.A., 2008. Global nitrogen deposition and carbon sinks. Nature Geoscience 1, 430–437.

Rehfuess, K.-E., Ågren, G.I., Andersson, F., Cannell, M.G.R., Friend, A., Hunter, I., Kahle, H.-P., Prietzel, J., Spiecker, H., 1999. Relationships between recent changes of growth and nutrition of Norway spruce, Scots pine and European beech forests in Europe-RECOGNITION. Working Paper 19. European Forest Institute, Joensuu, Finland.

Schindler, D.W., Bayley, S.E., 1993. The biosphere as an increasing sink for atmospheric carbon: estimates from increased nitrogen deposition. Global Biogeochemical Cycles 7, 717–733.

Sievering, H., 1999. Reply to nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. Nature 400, 629–630.

Sievering, H., Tomaszewski, T., Torizzo, J., 2007. Canopy uptake of atmospheric N deposition at a conifer forest: Part I—canopy N budget, photosynthetic efficiency and net ecosystem exchange. Tellus 59B, 483–492.

Simpson, D., Butterbach-Bahl, K., Fagerli, H., Kesik, M., Skiba, U., Tang, S., 2006. Deposition and emissions of reactive nitrogen over European forests: a modelling study. Atmospheric Environment 40, 5712–5726.

Solberg, S., Andreassen, K., Clarke, N., Torseth, K., Tveito, O.E., Strand, G.H., Tomter, S., 2004. The possible influence of nitrogen and acid deposition on forest growth in Norway. Forest Ecology and Management 192, 241–249.

Solberg, S., Dobbertin, M., Reinds, G.J., Lange, H., Andreassen, K., Fernandez, P.G., Hildingsson, A., de Vries, W., 2009. Analyses of the impact of changes in atmospheric deposition and climate on forest growth in European monitoring plots: a stand growth approach. Forest Ecology and Management 258, 1735–1750.

Sutton, M.A., Simpson, D., Levy, P.E., Smith, R.I., Reis, S., van Oijen, M., de Vries, W., 2008. Uncertainties in the relationship between atmospheric nitrogen deposition and forest carbon sequestration. Global Change Biology 14, 2057–2063.

Sutton, M.A., Nemitz, E., Erisman, J.W., Beier, C., Butterbach-Bahl, K., Cellier, P., de Vries, W., Cotrufo, F., Skiba, U., Di Marco, C., Jones, S., Laville, P., Soussana, J.F., Loubet, B., Twigg, M., Famulari, D., Whitehead, J., Gallagher, M.W., Neftel, A., Flechard, C.R., Herrmann, B., Calanca, P.L., Schjoerring, J.K., Daemmgen, U., Horvath, L., Tang, Y.S., Emmett, B.A., Tietema, A., Peñuelas, J., Kesik, M., Brueggemann, N., Pilegaard, K., Vesala, T., Campbell, C.L., Olesen, J.E., Dragosits, U., Theobald, M.R., Levy, P., Mobbs, D.C., Milne, R., Viovy, N., Vuichard, N., Smith, J.U., Smith, P., Bergamaschi, P., Fowler, D., Reis, S., 2007. Challenges in quantifying biosphere–atmosphere exchange of nitrogen species. Environmental Pollution 150, 125–139.

Townsend, A.R., Braswell, B.H., Holland, E.A., Penner, J.E., 1996. Spatial and temporal patterns in terrestrial carbon storage due to deposition of fossil fuel nitrogen. Ecological Applications 6, 806–814.

Van Oijen, M., Ågren, G.I., Chertov, O., Kellomäki, S., Komarov, A., Mobbs, D.C., Murray, M.B., 2008. Evaluation of past and future changes in European forest growth by means of four process-based models—Chapter 4.4. In: Kahle, H.P., Karjalainen, T., Schuck, A., Ågren, G.I., Kellomäki, S., Mellert, K., Prietzel, J., Rehfuess, K.E., Spiecker, H. (Eds), Causes and Consequences of Forest Growth Trends in Europe—Results of the RECOGNITION Project. European Forest Institute Research Report 21. Brill, Leiden, pp. 183–199.

Vitousek, P.M., Howarth, R.W., 1991. Nitrogen limitation on land and in the sea: how can it occur? Biogeochemistry 13, 87–115.

Wamelink, G.W.W., van Dobben, H.F., Mol-Dijkstra, J.P., Schouwenberg, E.P.A.G., Kros, J., de Vries, W., Berendse, F., 2009a. Effect of nitrogen deposition reduction on biodiversity and carbon sequestration. Forest Ecology and Management 258, 1774–1779.

Wamelink, G.W.W., Wieggers, H.J.J., Reinds, G.J., Kros, J., Mol-Dijkstra, J.P., van Oijen, M., de Vries, W., 2009b. Modelling impacts of changes in carbon dioxide concentration, climate and nitrogen deposition on NPP and carbon sequestration of intensive forest monitoring plots in Europe. Forest Ecology and Management 258, 1794–1805.

Woodwell, G.M., Whittaker, R.H., 1968. Primary production in terrestrial ecosystems. American Zoologist 8, 19–30.