Biogeosciences Discussion BG_2019-335

"Carbon / nitrogen interactions in European forests and semi-natural vegetation. Part II: Untangling climatic, edaphic, management and nitrogen deposition effects on carbon sequestration potentials" by Chris R. Flechard et al.

Point by point reply to Referees' comments

We are thankful to both referees for their interest in our study, for their constructive comments, sharp insights and challenging questions, which have helped improve the manuscript. For clarity's sake, we provide our point-by-point **responses to each comment in blue**, and provide **the effective changes to the manuscript text in green**.

Anonymous Referee #1

General remarks:

Magnani et al. (2007) reported very large responses of forest carbon sequestration to nitrogen deposition. Several authors rapidly pointed out that the response proposed was way above previous estimates and direct observations in N addition studies. This apparent discrepancy has been discussed at length for more than a decade now, but there is still a need for a more stringent analysis of how dC responds to dN. The effort made in this manuscript is, therefore, most interesting and commendable.

However, this model analysis is very complex. Many hours of careful reading is needed to get an insight into how the model is constructed and how it handles the critical assumptions involved. A reader will also need to read the companion paper. Most readers will still be left with many queries. This is not uncommon in the case of modelling papers. Vital assumptions are deeply embedded and not clearly visible although the outcome is constrained by the assumptions. In fact, trust in the many reputed authors, rather than the apparent quality of the manuscript, drove me to read it once again. Could these complex matters and their analysis be made more understandable and transparent, respectively? I am not sure how, but would like to ask the authors to do their utmost. Hopefully, the comments below will be helpful when revising the ms.

When we submitted this paper (Part II) and its companion paper (Part I) as components of the same study, a more detailed description of the BASFOR model, and of the way it was implemented in this study, were included in Part I. We only originally provided a cursory description of the model in Part II, but we now recognize that, with Part II being much more model-oriented than Part I (which is more measurements-oriented), it made sense to move the detailed description to Part II, while keeping a short outline in Paper I and refer there to Part II for details.

We have done this in the revised papers. The detailed model description was inserted in Section 2.2.1 of the Part II paper, alongside with the relevant supplementary information also moved to the Part II Supplement (see Figs. S1, S2). In addition, we remind the Reviewer that, as stated on line 198, we have provided a link to the online BASFOR model description at https://github.com/MarcelVanOijen/BASFOR. This link provides the complete Fortran code of the model, which makes it completely transparent with respect to model structure, embedded assumptions and data flows between the model parts.

In addition to the model description imported from Paper I, we have added the following paragraph to summarize key underlying model assumptions specifically regarding the coupling of C and N in vegetation and soil processes. We explain that new vegetation growth proceeds with a prescribed N/C ratio, unless when N is limiting, which accelerates senescence and triggers changes in allocation from leaves to roots. The coupling of C and N in trees impacts the C/N ratio in soil organic matter through litterfall and its subsequent decomposition. The following text was added towards the end of Section 2.2.1 'General description of the BASFOR ecosystem model':

'…In BASFOR, the C and N cycles are coupled in both trees and soil. The model assumes that new growth of any organ proceeds with a prescribed N/C ratio, which is species-specific but generally higher for leaves and roots than for stems and branches. If the nitrogen demand for growth cannot be met by supply from the soil, some of the foliar nitrogen is recycled until leaves approach a minimum N/C ratio when leaf senescence will be accelerated. The calculation of foliar senescence accounts for a vertical profile of nitrogen content, such that the lowest leaves have the lowest N-C ratio and senesce first. Nitrogen deficiency, as measured by foliar nitrogen content, not only increases leaf senescence, but also decreases GPP and shifts allocation from leaves to roots. Given that foliar N content is variable in BASFOR, the litter that is produced from leaf fall also has a variable N/C ratio. When the litter decomposes and is transformed, the N/C ratio of the new soil organic matter will therefore vary too in response to the ratio in the litter. Except for woody plant parts, the C and N couplings in BASFOR vegetation and soil are based on the same generic ecophysiological assumptions as those explained in detail for grassland model BASGRA (Höglind et al. 2020)…'

Additional reference:

Höglind, M., Cameron, D., Persson, T., Huang, X. and Van Oijen, M.: BASGRA_N: a model for grassland productivity, quality and greenhouse gas balance, Ecol. Model., 417, 108925, https://doi.org/10.1016/j.ecolmodel.2019.108925, 2020.

Further, to ease the reader into the flow of the paper, to better introduce the modelling background and framework that underpin the C and N budget calculations of Fig. 2-5, and to illustrate graphically the temporal dimension of all the different forest sites of this study in relation to changing N_r deposition and increasing atmospheric CO_2 through the 20th century, we have added the new Figure 1 to the main body of the paper. This figure is shown hereafter as Fig. R1 and depicts the modelled (baseline) time course of GPP, NEP, N deposition and N losses for all study sites over the period 1900-2010. In the text, we have added a short paragraph to describe this new Fig. 1 at the start of Results – 3.1, just before the description of the Sankey plots for C and N budgets:

'*3.1 Short term (5-yr) versus lifetime C and N budgets from ecosystem modelling*

The time course of modelled (baseline) GPP, NEP and total leaching and gaseous N losses is shown in Fig. 1 for all forests sites over the 20th century and until 2010, forced by climate, increasing atmospheric CO_2 and by the assumed time course of N_r deposition over this period (Fig. 1A). For each stand, regardless of its age and establishment date, an initial phase of around 20-25 years occurs, during which GPP increases sharply from zero to a potential value attained upon canopy closure (Fig. 1B), while NEP switches from a net C source to a net C sink after about 10 years (Fig. 1D). Initially N_r losses are very large (typically of the order of 10 g (N) m^{-2} yr^{-1}), then decrease rapidly to pseudo steady-state levels when GPP and tree N uptake reach their potential.

After this initial phase, modelled GPP increases steadily in response to increasing N_{dep} and atmospheric CO_2, but only for the older stands established before around 1960, i.e. those stands that reach canopy closure well before the 1980's, when N_r deposition is assumed to start declining. Thereafter, modelled GPP ceases to increase, except for the recently established stands that have not yet reached canopy closure. The stabilization of GPP for mature trees at the end of the 20th century in the model is likely a consequence of the effects of decreasing N_{dep} and increasing CO_2 cancelling each other out to a large extent. In parallel, modelled total N losses start to decrease after the 1980's, even for sites long past canopy closure (Fig. 1E-F), but this mostly applies to stands subject to the largest N_{dep} levels, i.e. where the historical high N_{dep} of the 1980's, added to the internal N supply, were well in excess of growth requirements in the model.

These temporal interactions of differently-aged stands with changing N_{dep} and CO_2 over their lifetimes therefore impact C and N budget simulations made over different time horizons. Modelled C and N budgets are represented schematically in Fig. 2 and Fig. 3, respectively, as «Sankey» diagrams …'

We believe all these changes haved helped make '*…the analysis … more understandable and transparent…*', as requested by the Referee. The added text and the new Figure 1 also introduce the topic of downward trends in N leaching since the end of the 20th century, raised in another comment by Referee 1, which are then further discussed in the paper (see below).

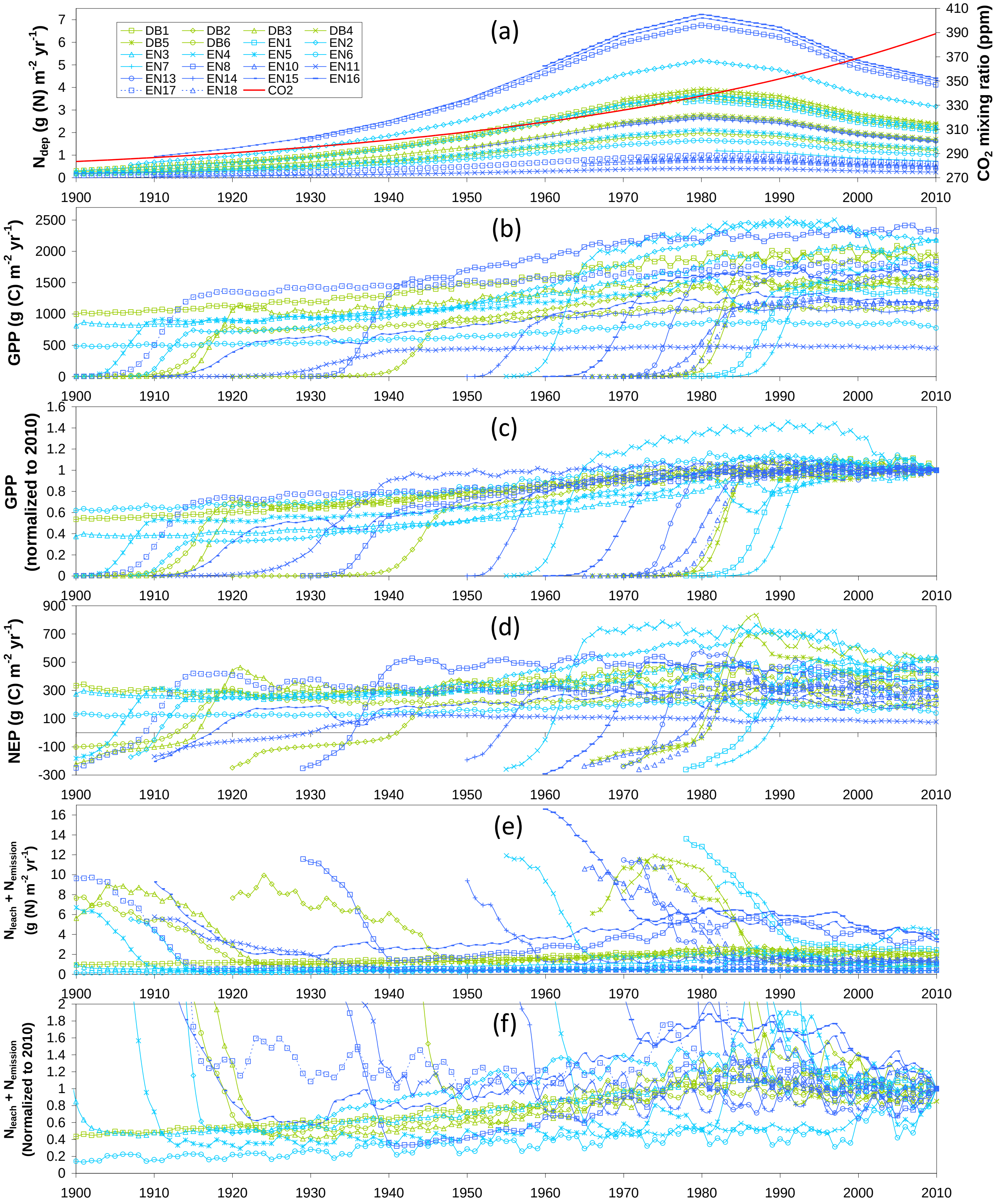

Figure R1 *(= new Fig. 1 of revised paper)*. Time courses of (a) assumed atmospheric N_r deposition and CO_2 mixing ratio, and baseline model simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching and gaseous emissions, and (f) total N losses normalized to 2010.

The treatment of the relations between Ndep and the internal forest N cycle is pivotal. A step ahead here is the use of local data on Ndep where possible and not just regional estimates. As regards the internal N cycle, the authors do not appear (e.g., lines 264-265) to handle that organic N sources (chiefly amino acids and peptides) are used by plants and probably dominate in less fertile systems, especially boreal forests. Inorganic N sources become dominant when the N supply is large relative to the biological demand.

The referee is right in pointing out that we do not consider organic N sources as part of the nitrogen supply and uptake by trees. The BASFOR model does not account for dissolved organic N (DON) forms with a relatively low molecular weight (e.g. amino acids and small peptides), that may be taken up by roots alongside dissolved inorganic N (DIN, mainly ammonium and nitrate). Proteins and other N-containing molecules from the litter layers, roots and soil organic matter (with slow or fast turnover) are decomposed, in the model pools, into mineral N, CO_2 and H_2O, but the large number of intermediate organic degradation products are not explicitly simulated. We therefore cannot, through modelling, address the Referee’s question about the importance of organic N uptake in boreal forests and elsewhere.

However, the role of organic N is perhaps less clear than the Referee suggests, as far as the dominance of organic over inorganic N uptake in less fertile systems and boreal forests is concerned. There is no doubt that roots can take up some forms of bio-available organic N, and that in acidic soils containing large amounts of organic matter, soluble N is dominated by organic forms, with amino acids making up to 10-20% of the total DON pool, and correspondingly low DIN concentrations (Jones and Kielland, 2002, Soil Biology and Biogeochemistry 34, 209-219). But the ecological significance of this N acquisition pathway remains controversial (Moreau et al., 2019, Functional Ecology 33:540–552). It appears that DON has very fast turnover to ammonium (Jones and Kielland, 2002), so that ultimately the trees take up much DIN as well, despite the high concentrations of DON in the soil. It is also important that a large fraction of DON is originally lost from plant roots, so that its uptake compensates this N-leak, rather than leading to an important pathway of net N-acquisition by plants (Jones et al., 2005, Soil Biol Biochem 37, 413-423). Further, many forms of DON are by-products of microbial breakdown of humic substances, that are not necessarily bio-available (Warren, 2014, Plant Soil, 375, 1–19).

We nonetheless agree that the revised manuscript should make it clear that i) the current state of knowledge is that soil plant N nutrition relies on both dissolved organic and inorganic N forms, albeit with uncertain partitioning, and ii) our ecosystem model does not consider organic N supply and uptake by trees. We have therefore made the following additions to the BASFOR description of N cycling that was transferred from the Part I paper:

‘…Dissolved inorganic nitrogen (DIN) *is taken up by the trees from the soil, and nitrogen returns to the soil with senescence of leaves, branches and roots, and also when trees are pruned or thinned. Part of the N from senescing leaves is re-used for growth. The availability of mineral nitrogen is a Michaelis-Menten function of the mineral nitrogen pool and is proportional to root biomass.* The model does not include a dissolved organic nitrogen (DON) pool and therefore does not account for the possible uptake of bio-available DON forms (e.g. amino acids, peptides) by trees. *Transformation between the four soil nitrogen pools are similar to those of the carbon pools, with mineral nitrogen as the loss term*…’

Further, in the discussion on challenges and limitations in the modelling study, line 583, we have mentioned the uncertainty in N_{supply} related to the non-inclusion of DON supply and uptake:

‘*iv) Nitrogen deposition likely contributes a minor fraction (on average 20% according to the model) of total ecosystem N supply (heavily dominated by soil organic N mineralization), except for the very high deposition sites (up to 40%).* The fraction of N_{dep}/N_{supply} may even be smaller considering the pool of DON (not included in BASFOR), from which bio-available organic N forms may be taken up by trees in significant quantities in non-fertile, acidic organic soils (Jones and Kielland, 2002; Warren, 2014; Moreau et al., 2019). *Thus, in many cases the Ndep fertilisation effect may be marginal and difficult to detect, because it may be smaller than typical measurement uncertainties and noise in C and N budgets.*’

Additional references:

Jones, D.L. and Kielland, K.: Soil amino acid turnover dominates the nitrogen flux in permafrost-dominated taiga forest soils, Soil Biology Biochem., 34, 209–219, https://doi.org/10.1016/S0038-0717(01)00175-4, 2002.

Moreau, D., Bardgett, R.D., Finlay, R.D., Jones, D.L. and Philippot, L.: A plant perspective on nitrogen cycling in the rhizosphere, Funct. Ecol., 33, 540–552, https://doi.org/10.1111/1365-2435.13303, 2019.

Warren, C.R.: Organic N molecules in the soil solution: What is known, what is unknown and the path forwards, Plant Soil, 375, 1–19, https://doi.org/10.1007/s11104-013-1939-y, 2014.

The authors may also reflect on the trends of decreasing leaching of inorganic N from forests in NE USA and N Europe. What in their models could drive this phenomenon? It could be related to higher tree growth (more C) in response to management or environmental change? How should C be coupled to N?

In this paper we set out to tackle the issue of how regional (spatial) differences in climate (and also soil and forest age structure) influence the response of C sequestration to N deposition. However, we did not specifically seek to address the issue of how forest N losses have responded over the last 2-3 decades to changes in management, climate or other environmental factors. We unfortunately do not possess the depth of historical data (neither long-term DIN leaching nor NPP data) necessary to investigate this empirically at our sites. Others have reported long term changes in N or nitrate losses/export over the last few decades, as pointed out by the Referee (e.g. Verstraeten et al., 2012, Atmospheric Environment 62: 50-63; Goodale et al., 2003, Ecosystems 6:75–86; Bernal et al., 2012, PNAS, 109, 9, 3406–3411)

From a modelling viewpoint, the main factors in BASFOR likely to reduce DIN leaching over the last 2-3 decades are the assumed decreasing trends in total N_{dep} since the 1980's (see Fig. R1-A below), and the increasing trends of GPP and NPP (and increasing N uptake) for the forest stands still at the aggrading stage or in response to increasing atmospheric CO_2 (Fig. R1-A). We did not include a changing climate in our simulations over the 20th century (though inter-annual meteorological variability is accounted for). The forests included in the study range from ~25 to >150 years old. This means that the younger stands (<40 years) mostly experienced decreasing N_{dep} (since ~1980), and sharply increasing NPP as part of their initial growth phase (before reaching peak LAI and canopy closure). By contrast, the oldest stands experienced increasing N_{dep} and increasing NPP during a large part of the 20th century, and then decreasing N_{dep} after ~1980 and stabilized NPP over the last 30 years. The age of the forest therefore influences, in model simulations, the extent to which decreasing N_{dep} after 1980 would translate into reduced N leaching. But the level of N_{dep} in itself is important: for the high N_{dep} locations, a more important reduction in N losses may be expected, following the decrease in N_{dep}, than at N-limited sites, as suggested in Fig. R1E-F.

We have described these patterns in the new sub-section added to 3.1 together with the new Figure 1 (= Fig. R1 introduced above). To further address the referee's question as to what could drive observed recent downward trends in leaching in the model, we have made three additional model scenario runs besides the baseline simulation of Fig. R1. The corresponding figures are shown below (Fig. R2, R3, R4) and were added to the paper's supplement as Figures S9, S10 and S11. These additional model runs test the effects of increasing N_{dep} or CO_2 separately on forest productivity and N losses. In Fig. R2, the CO_2 mixing ratio is kept constant at 310 ppm (~ the mean value over the period 1900-2010); in Fig. R3, CO_2 increases exponentially as in the baseline run, but N_{dep} is constant at 1.5 g (N) m^{-2} yr^{-1} at all sites; in Fig. R4, N_{dep} is constant at 3.0 g (N) m^{-2} yr^{-1} at all sites. These additional model sensitivity runs and supplementary figures are referred to in the discussion of the revised version, in the following added text as part of *Section 4.2 Limitations and uncertainties in the approach for quantifying the dC/dN response*:, starting line 587:

'*…smaller than typical measurement uncertainties and noise in C and N budgets*.

A further limitation to our estimates of the dC/dN response, based on the analysis of the spatial (inter-site) variability in C and N fluxes, is that these forests are not in steady state with respect to N_r deposition and ambient CO_2. Some stands have been affected by, and may be slowly recovering from, excess N_r deposition in the second half of the 20th century; while the more remote sites may always have been N-limited. Figure 1 showed that the modelled GPP of the older forests increased through most of the 20th century, but stabilized when N_{dep} started to decrease after the 1980's, while total N losses also declined over the last 2-3 decades. This is consistent with observations of decreasing N (nitrate) leaching at long term study sites in N-E USA (Goodale et al., 2003; Bernal et al., 2012) and N Europe (Verstraeten et al., 2012; Johnson et al., 2018; Schmitz et al., 2019).

In our model analysis, the declining trend in N_r deposition appears to be the primary driver for the modelled reduced N losses since the 1980's. This can be inferred from model input-sensitivity scenario runs shown in Fig. S9-S11 of the Supplement. In Fig. S9, a constant CO_2 mixing ratio of 310 ppm (i.e. the mean value over the period 1900-2010), used instead of the exponential increase since the 19th century, does not greatly alter overall productivity patterns, nor the decreasing trend in N losses over the period 1980-2010 (Fig S9E-F), compared with the baseline run (Fig. 1). By contrast, in scenarios shown in Fig. S10-S11, the assumed constant N_{dep} levels at all sites of 1.5 and 3.0 g (N) m^{-2} yr^{-1}, respectively, together with the exponential CO_2 increase, remove the decreasing trends in N_r losses over the period 1980-2010. Meanwhile, in constant N_{dep} scenarios the increase in GPP over the whole period is fairly monotonous, in response to a steadily increasing CO_2 (Fig. S10B-C), without the inflexion point around 1980 simulated in the baseline run (Fig 1B-D). In real-life stands, however, decadal decreases in N losses or exports have been observed without any significant reductions in N_{dep} (Goodale et al., 2003). Other potential factors such as increased denitrification, longer growing season, plant N accumulation, changes in soil hydrological properties or temperature, historical disturbances may also play a role (Bernal et al., 2012). Many such factors are not considered in our model, and neither is long term climate change.

The EC-based flux data suggest that the Ndep response of forest productivity…'

Additional references

Bernal, S., Hedin, L.O., Likens, G.E., Gerber, S. and Buso, D.C.: Complex response of the forest nitrogen cycle to climate change, P. Natl. Acad. Sci. USA, 109(9), 3406–3411, https://doi.org/10.1073/pnas.1121448109, 2012.

Goodale, C.L., Aber, J.D. and Vitousek, P.M.: An unexpected nitrate decline in New Hampshire streams, Ecosystems, 6, 75–86, https://doi.org/10.1007/s10021-002-0219-0, 2003.

Johnson, J., Graf Pannatier, E., Carnicelli, S., Cecchini, G., Clarke, N., Cools, N., Hansen, K., Meesenburg, H., Nieminen, T.M., Pihl-Karlsson, G., Titeux, H., Vanguelova, E., Verstraeten, A., Vesterdal, L., Waldner, P. and Jonard, M.: The response of soil solution chemistry in European forests to decreasing acid deposition, Glob. Change Biol., 24, 3603–3619, https://doi.org/10.1111/gcb.14156, 2018.

Schmitz, A., Sanders, T.G.M., Bolte, A., Bussotti, F., Dirnböck, T., Johnson, J., Peñuelas, J., Pollastrini, M., Prescher, A.-K., Sardans, J., Verstraeten, A. and de Vries, W.: Responses of forest ecosystems in Europe to decreasing nitrogen deposition, Environ. Pollut., 244, 980 –994, https://doi.org/10.1016/j.envpol.2018.09.101, 2019.

Verstraeten, A., Neirynck, J., Genouw, G., Cools, N., Roskams, P. and Hens, M.: Impact of declining atmospheric deposition on forest soil solution chemistry in Flanders, Belgium, Atmos. Environ., 62, 50–63, https://doi.org/10.1016/j.atmosenv.2012.08.017, 2012.

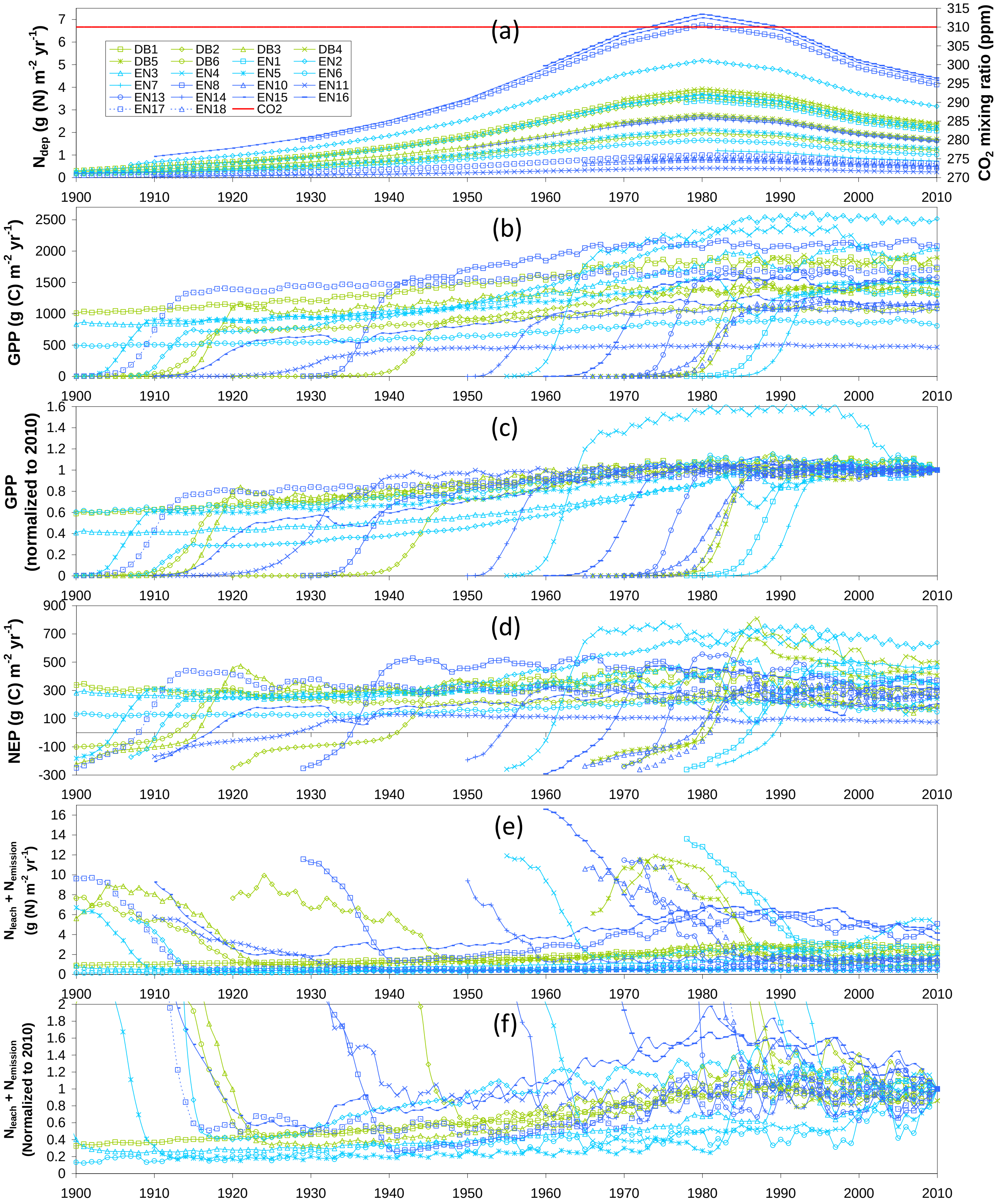

Figure R2. *(= additional Fig. S9 in revised Supplement)*. Alternative model scenario using a constant CO_2 mixing ratio of 310 ppm through the entire modelling period (a), showing simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching and gaseous emissions, and (f) total N losses normalized to 2010.

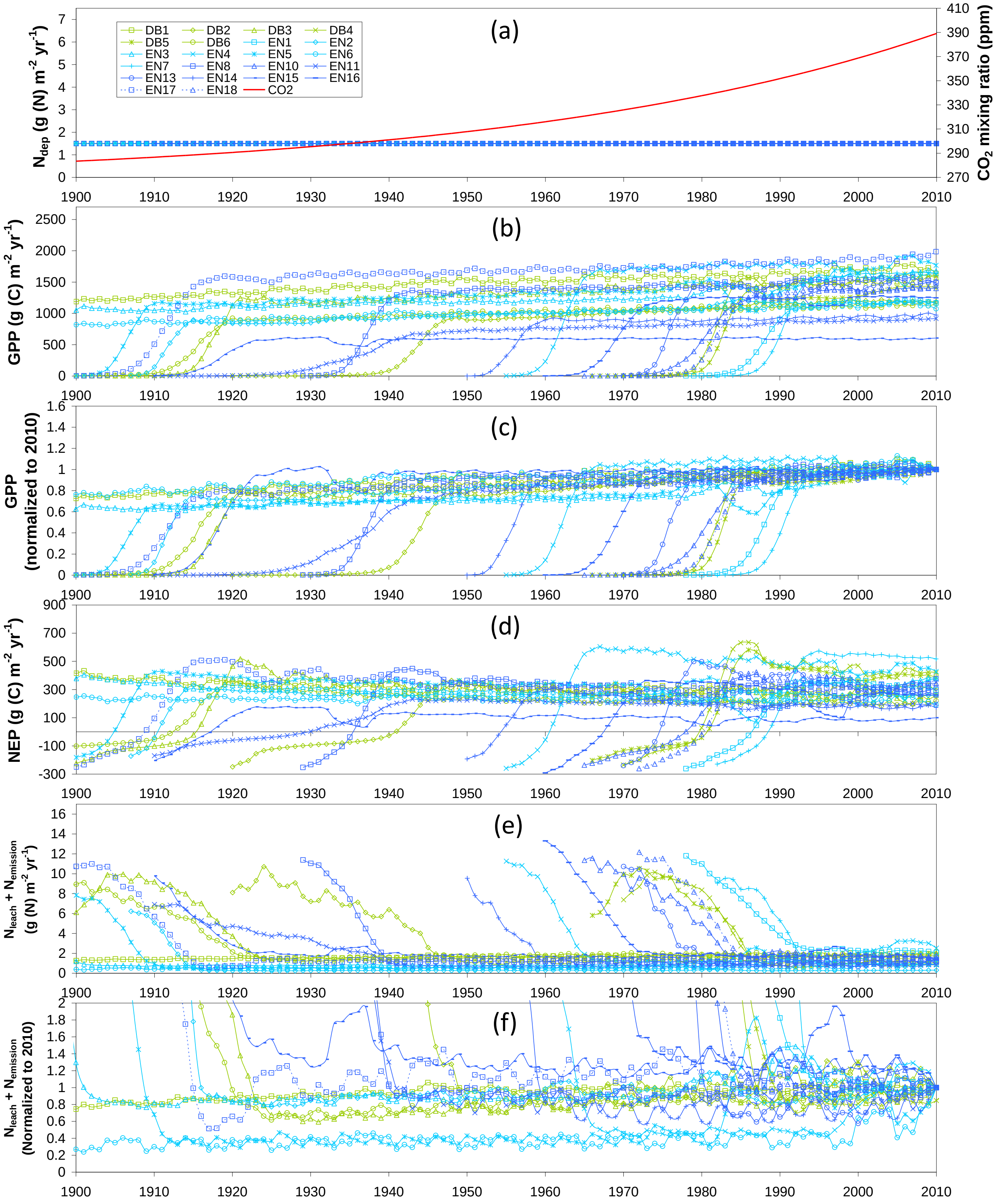

Figure R3. *(= additional Fig. S10 in revised Supplement)*. Alternative model scenario using a constant N_{dep} level of 1.5 g (N) m^{-2} yr^{-1} at all sites through the entire modelling period (a), showing simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching and gaseous emissions, and (f) total N losses normalized to 2010.

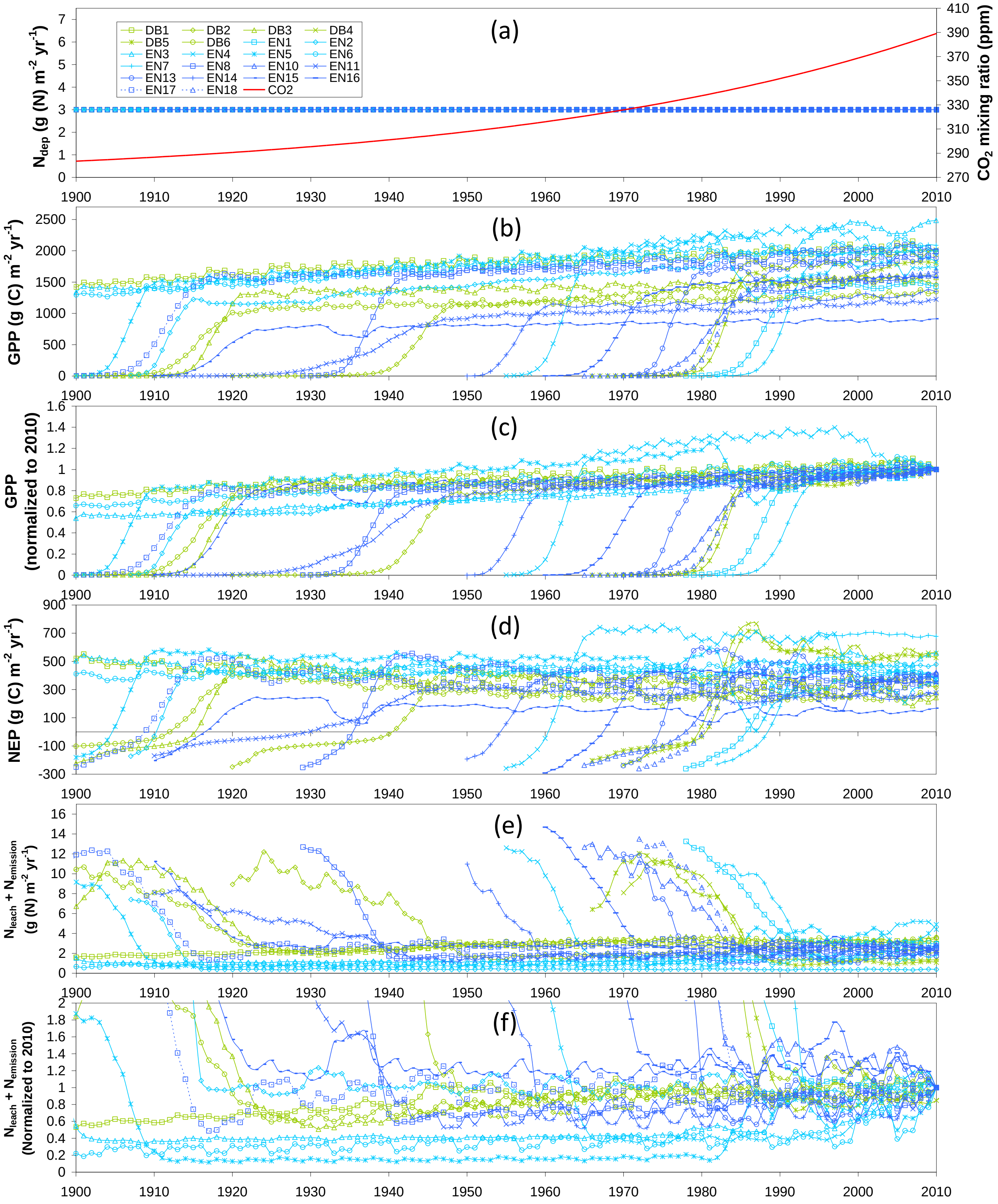

Figure R4. *(= additional Fig. S11 in revised Supplement)*. Alternative model scenario using a constant N_{dep} level of 3.0 g (N) m^{-2} yr^{-1} at all sites through the entire modelling period (a), showing simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching and gaseous emissions, and (f) total N losses normalized to 2010.

On p. 15 potential net effects of N supply on C sequestration efficiency are discussed. The authors mention that C sequestration in a high C/N component like wood would be one explanation (used also by Magnani et al.) among the many complex and nonlinear interactions between N and C. Is it at all possible and in line with findings in N-15 tracer studies that the majority of the N added goes into wood? The answer from many studies appears to be no (e.g., Nadelhoffer et al. 1999).

In the text p. 13-14 (lines 551-556) the argument we made was that the $dNEP/dN_{dep}$ response should logically be steeper in forests than in short, non-woody semi-natural (SN) vegetation, since a large fraction of C is stored in high C/N components (wood) in forests, versus SOM with a much lower C/N ratio in non-woody SN ecosystems. By that statement, we had not actually meant to extrapolate to nitrogen storage and to imply that "…*the majority of the N added goes into wood*…", as suggested by the Referee. Nevertheless, the most recent synthesis of ^{15}N tracer experiments by Du and de Vries (Environ. Pollut., 242, 1476–1487, 2018) does indeed suggest that tree biomass is the primary sink for the added nitrogen in both boreal and temperate forests (about 70%), with the remaining 30% retained in soil. For carbon, our forest ecosystem model suggests that up to 60-80% of NECB is sequestered in above-ground biomass (branches and stems), which would be consistent with the partitioning of the N sink. Since we do not have a model for our non-woody SN sites, we cannot however provide a comparison between the two types of vegetation (F *versus* non-woody SN) of the modelled fractions of C and N stored above and below ground.

Regarding other studies on the fate of added N, in the introduction to the paper we did originally provide some background and references, l 100-106 on p.3, which we have now expanded and revised with a contrasting view on the contribution of tree and soil carbon sequestration in response to N deposition:

'…The questions of the allocation and fate of both the assimilated carbon (Franklin et al., 2012) and deposited nitrogen (Nadelhoffer et al., 1999; Templer et al., 2012; Du and de Vries, 2018) appear to be crucial. It has been suggested that N_r deposition plays a significant role *in promoting the carbon sink strength only if N is stored in woody tissues with high C/N ratios (>200–500) and long turnover times, as opposed to soil organic matter (SOM) with C/N ratios that are an order of magnitude smaller (de Vries et al., 2008). Nadelhoffer et al. (1999) argued on the basis of a review of ^{15}N tracer experiments that soil, rather than tree biomass, was the primary sink for the added nitrogen in temperate forests*. However, based on a recent synthesis of ^{15}N tracer field experiments (only including measurements of ^{15}N recovery after > 1 year of ^{15}N addition), Du and de Vries (2018) estimated that tree biomass was the primary sink for the added nitrogen in both boreal and temperate forests (about 70%), with the remaining 30% retained in soil. At *sites with elevated N inputs, increasingly large fractions are lost as nitrate (NO_3^-) leaching. Lovett et al. (2013) found in north-eastern US forests that added N increased C and N stocks and the C/N ratio in the forest floor, but did not increase woody biomass or aboveground NPP.*"

Additional references:

Du, E. and de Vries, W.: Nitrogen-induced new net primary production and carbon sequestration in global forests, Environ. Pollut., 242, 1476–1487, https://doi.org/10.1016/j.envpol.2018.08.041, 2018.

Franklin, O., Johansson, J., Dewar, R.C., Dieckmann, U., McMurtrie, R.E., Brännström, Å and Dybzinski, R.: Modeling carbon allocation in trees: a search for principles, Tree Physiol., 32, 648–666, https://doi.org/10.1093/treephys/tpr138, 2012.

Templer, P.H., Mack, M.C., Chapin, F.S. III, Christenson, L.M., Compton, J.E., Crook, H.D., Currie, W.S., Curtis, C.J., Dail, D.B., D'Antonio, C.M., Emmett, B.A., Epstein, H.E., Goodale, C.L., Gundersen, P., Hobbie, S.E., Holland, K., Hooper, D.U., Hungate, B.A., Lamontagne, S., Nadelhoffer, K.J., Osenberg, C.W., Perakis, S.S., Schleppi, P., Schimel, J., Schmidt, I.K., Sommerkorn, M., Spoelstra, J., Tietema, A., Wessel, W.W. and Zak, D.R.: Sinks for nitrogen inputs in terrestrial ecosystems: A meta-analysis of ^{15}N tracer field studies, Ecology, 93, 1816–829, https://doi.org/10.1890/11-1146.1, 2012.

This calls for an analysis of the physiological processes in which interactions between the cycles of N and C are particularly important. Modelling is necessary, there is no doubt about that, but it needs to make best use of all the data available including recent findings. These are many, but the authors could perhaps consider some, which describe non-linear biological controls (e.g., Kallokoski et al. 2013, Tree Physiol. 33, 1145- , show that wood cell formation is similar in N-limited and N-fertilized trees during early summer, but then cease in the former but continue in the latter, and Högberg et al. 2010, New Phytol. 187, 485- , show that tree belowground C allocation is greatly reduced by additions of N; such relations may be interconnected).

We have acknowledged the limitations in modelling in our study in several places in the discussion, with respect to mechanisms that are not included (e.g. l. 580-582, l.594-596, l.659-661, etc), though at this point we are unable to make such changes in the model. We did mention originally (l. 720-723) that '…*excess Nr deposition reduces soil – especially heterotrophic – respiration in many temperate forests*…' through, amongst other things (Janssens et al., 2010) , '… *a decrease in below-ground C allocation and the resulting root respiration, permitted by a lesser need to develop the rooting system when more N is available (see also Alberti et al., 2015)*…'. But we have made it clearer in Section 4.2.1 in the revised version('*Limitations and uncertainties*…') that this and other important mechanisms, such as mentioned above by the Referee, affect non-linear C/N relations but are not included in the model, and therefore represent a significant limitation to our analysis. A fifth item (v) was added in section 4.2, line 587:

'v) Non-linear biological controls that affect C/N relations but are not explicitly considered in the model. For example, BASFOR does consider that N addition can reduce below-ground C allocation (e.g. Högberg et al., 2010), resulting in decreased soil R_{aut} and R_{het} (Janssens et al., 2010), but does not account for the possible consequences of a stimulation of wood cell formation from mid-summer onwards and a delay in the cessation of tracheid production in late season (Kalliokoski et al., 2013).'

Additional References:

Högberg, M.N., Briones, M.J.I, Keel, S.G., Metcalfe, D.B., Campbell, C., Midwood, A.J., Thornton, B., Hurry, V., Linder, S., Näsholm, T. and and Högberg, P.: Quantification of effects of season and nitrogen, supply on tree below-ground carbon transfer to ectomycorrhizal fungi and other soil organisms in a boreal pine forest, New Phytol., 187, 485–493, https://doi.org/10.1111/j.1469-8137.2010.03274.x, 2010.

Kalliokoski, T., Mäkinen, H., Jyske, T., Nöjd, P. and Linder, S.: Effects of nutrient optimization on intra-annual wood formation in Norway spruce, Tree Physiol., 33, 1145–1155, https://doi.org/10.1093/treephys/tpt078, 2013.

I would suggest that the authors rethink and reword a part of the reporting of results (lines 387-393). Firstly, forests 30-60 years are not young, especially not in Central Europe, in the sense that they have a low demand for N because of a low biomass as stated by the authors. Older forests may have a larger biomass for sure, but this is because of their trunks, tissues with much less biological activity and N demand than foliage and fine roots. On the contrary, 30-60 yrs-old forest most probably have fully closed canopies and a very high demand for N.

Secondly, the idea of such forests leaching more N because of less canopy interception of water (and hence greater runoff), is also unlikely. Check with hydrologists if they see more runoff from forests 30-60 yrs-old than from older forests! Moreover, foresters would describe forests < 30 years old as young; in the context of rotational forestry in Europe 30-60 yrs-old forest are middle-aged.

We agree that the adjective '*young*' was not appropriate to describe the 30-60 yr age class, even if we in fact mostly used the comparative '*young**er***', by opposition to 'mature' (>80 yrs). We have shown in our additional figure R1 (see above) that, in the model, the initial phase of rapidly increasing GPP and fast decreasing N losses lasts around 20 years, therefore we agree that for trees aged 30-60 the NUPE or N_{loss} fractions, calculated over a recent 5-yr period, are no longer affected by that initial growth phase characterized by large N losses. Nevertheless, if NUPE or N_{loss} fractions are calculated over the whole period since the forest was established ('lifetime'), then the initial ~20-yr phase has a greater weight in the 30-60 yr-old forests lifetime calculation than in the >80 yr-old stands. We have therefore rephrased this paragraph in the following way:

Line 383 '…By contrast, the analogous term for nitrogen, the N_{upt} fraction of total N_{supply}, is a much more variable term, both between sites of the network and between the 5-yr and lifetime simulations (Fig. 3, S6–S8). Modelled lifetime CSE and NUPE values are compared in Fig. 4 with the 5-yr values, as a function of stand age, indicating that (i) the older forests of the network (age range ~80–190 yrs) tend to have larger NUPE than younger or middle aged forests (~30–60 yrs), but (ii) the difference in NUPE between the two age groups is much clearer if NUPE is calculated over the whole period since planting (lifetime). As shown in *(the new)* Fig. 1 *(= Fig. R1 above)*, BASFOR predicts large N losses in young stands (<20-25 years), in which lower N demand by a smaller living biomass, combined in the early years with enhanced N_{miner} from higher soil temperature (canopy not yet closed) and with a larger drainage rate (smaller canopy interception of incident rainfall), all lead to larger NMIN losses. The 22 forests sites of this study were past this juvenile stage, but observation (ii) is a mathematical consequence of high N losses during the forest's early years having a larger impact on lifetime calculations in middle-aged than mature forests. NUPE tends to reach 70-80% on average after 100 years and is smaller calculated from lifetime than from a 5-yr thinning-free period. For forests younger than 60 years, lifetime NUPE is only around 60%.'

More specific comments:

Lines 73-74: Shouldn0t this be phrased the other way around: ". . . with no further C uptake response at high Ndep levels (Ndep > 2.2-3 g m-2 yr-1) followed by large N losses by leaching and gaseous emissions."

We are not sure if the Referee means '*followed*' in a temporal sense. The absence of productivity response to N_{dep} above a certain threshold is derived in our study from a spatial analysis (a comparison between sites), not from time series. To reduce the ambiguity on causality in the sentence, we have rephrased in the following way:

'…patterns of gross primary and net ecosystem productivity versus N_{dep} were non-linear, with no further growth responses at high N_{dep} levels (N_{dep} > 2.5–3 g (N) m^{-2} yr^{-1}) but accompanied by increasingly large ecosystem N losses by leaching and gaseous emissions.'

Line 140: add fires and insect attacks here.

'…Severe storms, fire outbreaks and insect infestations may have a similar effect'

Line 145: in some regions, e.g. in N. Europe, there are many N-fertilizer experiments.

We have added the following two references after '*…manipulation plots…*' in the sentence:

Nohrstedt, H.-Ö.: Response of coniferous forest ecosystems on mineral soils to nutrient additions: a review of Swedish experiences, Scand. J. Forest Res., 16, 555–573, https://doi.org/10.1080/02827580152699385, 2001.

Saarsalmi, A. and Mälkönen, E.: Forest fertilization research in Finland: a literature review, Scand. J. Forest Res., 16, 514–535, https://doi.org/10.1080/02827580152699358, 2001.

Line 216: you write even, but maybe mean seven?

No. We mean this (and have corrected thus) :

'…baseline BASFOR runs were produced for all 31 forest sites of the network, **including also those stands** for which the model was not calibrated…'

Line 266-268: the use of another definition of NUE is widespread; I understand that you want to use an acronym, but it is unfortunate to use one that commonly has another meaning.

We agree it is best to avoid confusion, and we have therefore changed from NUE (N use efficiency) to NUPE (N uptake efficiency) in the text, tables and figures. We have changed the sentence lines 272-273, to further dissociate NUPE from NUE:

'…Note that i) NUPE is a different concept from the nitrogen use efficiency (NUE), often defined as the amount of biomass produced per unit of N taken up from the soil, or the ratio NPP/N_{upt} (e.g. Finzi et al., 2007), and ii) …'

May I also suggest that you use Nmob = N mobilized, rather than Nmin, which means that you overlook organic N compounds as N sources.

Since the model does not account for dissolved organic N pools and uptake by trees, we believe it is preferable to stick to NMIN, which is explicit, rather than use NMOB which might imply otherwise.

Line 373: do you have clear evidence that Nmin does not change over time? Other authors discuss N oligotrophication and report that runoff of mineral N from forest decreases.

The soil mineral N concentration and leaching/export do change over time in the model, as shown and discussed above in Fig. R1 in our response to the Referee's comments on observed long term decreasing trends in forest N losses. Our sentence on line 373 ('*Since there is no significant long term (multi-annual) change in NMIN*') was indeed slightly misleading; by this we did not mean that mineral N concentrations and loss fluxes were stable over multi-decadal time scales. Rather, since NMIN is the soil inorganic N pool (g (N) m^{-2}), mineral N is transient and does not accumulate in the model because it leaves the root zone and effectively disappears (except for the fraction that is taken up by trees). There are inter-annual changes in NMIN, but the rate of change of the mineral N pool dNMIN/dt (g N m^{-2} yr^{-1}) is insignificant compared with the annual rates of N_{dep}, N_{upt}, N_{leach} and N_{miner}. We have therefore rephrased this sentence:

'…*Since the modelled long term (multi-annual) changes in the transient NMIN pool are negligible compared with the magnitudes of the N input and output fluxes, the dNMIN/dt term is not represented as an arrow in the budget plots, and the total mineral N_{supply} … is basically balanced by N uptake… and losses…*'

Line 402: it is interesting to learn in which direction the non-linearity develops.

In Fig. 3 of the paper we presented the differences in modelled CSE between sites, which were plotted versus the age of the different sites. Since many other factors differentiate our forest sites apart from age, the CSE trends versus stand age could have been affected by co-varying factors. In Fig. R5-A, shown below, modelled CSE is plotted for each site as a function of time elapsed since the stands were established, and similar trends are reproduced as in Fig. 3 of the paper, i.e. a decrease in modelled CSE from 25-35% in the age class 30-60 yrs down to around 20-25% for the stands older than 100 yrs.

The non-linearity we mention on line 402 is related to the increase with age of the R_{het}/GPP ratio, shown in Fig. R5-A. We have added Fig. R5 to the supplement (as Fig. S12) and rephrase our sentence on lines 400-402 of the manuscript:

'…*in the model, R_{eco} in 30 to 60-yr old stands represents a smaller fraction of GPP than in mature stands*. From Eq. (1) it can readily be shown that CSE = 1 - R_{aut}/GPP - R_{het}/GPP, which is roughly equivalent to 0.5 – R_{het}/GPP, since in the model R_{aut} is constant and approximately 0.5 for all species. By contrast, BASFOR predicts that the R_{het}/GPP ratio increases steadily with age at each site, after the initial establishment phase (Fig. S12-A). This induces a decline in modelled CSE from 25-35% in the age class 30-60 yrs down to around 20-25% for the older forests (Fig. S12-B). This also implies a non-linearity developing over time of GPP versus soil and litter layers C pools, since R_{het} is assumed to a linear function of fast and slow C pools in litter layers and SOM. *Lifetime CSE values are slightly smaller…*'

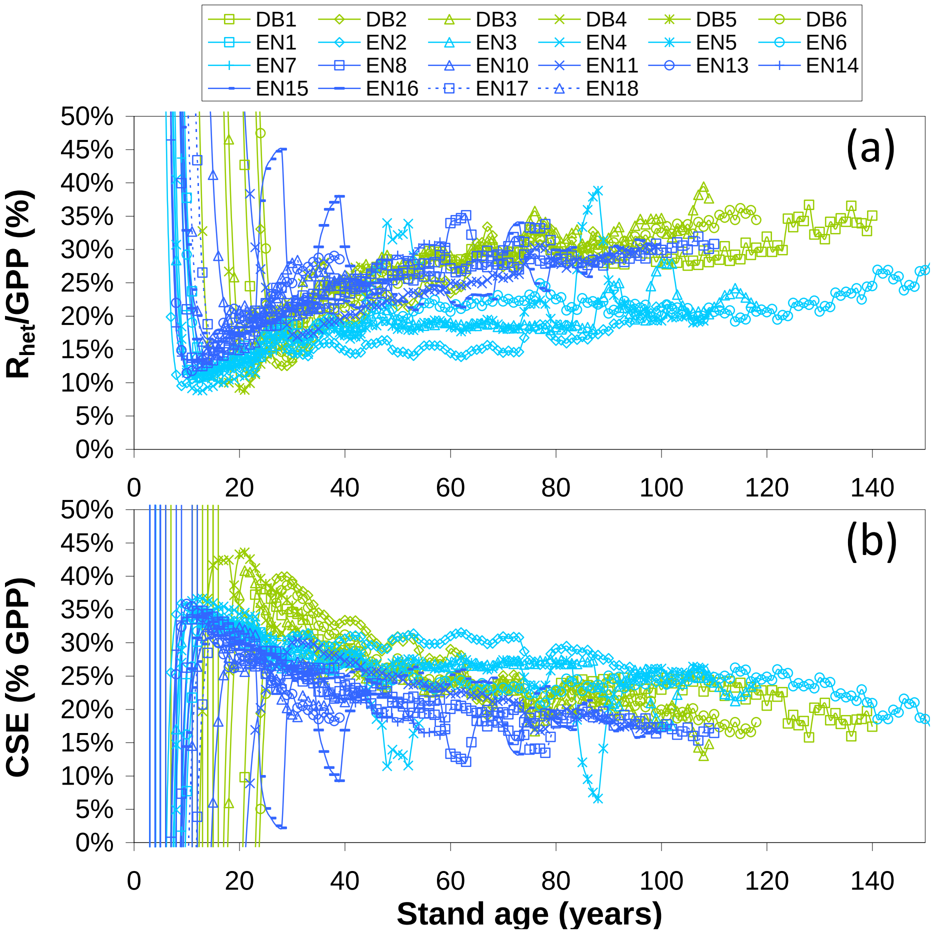

Figure R5. *(= additional Fig. S12 in revised Supplement)*. Modelled time courses for all forests of the study of (a) the ratio of heterotrophic respiration (R_{het}) to gross primary productivity (GPP) and (b) the carbon sequestration efficiency (CSE = NEP/GPP). Short term excursions are related to thinning events.

Line 525: BASFOR may be mechanistic, but the vital interactions are not discussed and clarified in the description of the model.

We have included a more thorough description of the model (imported from the Part I paper), as indicated above in response to one of the Referee's general remarks.

Line 595: it is unclear if internal N supply is a component of soil fertility.

Internal N supply reflects the overall ability of soil microorganisms to mineralize dead organic matter and deliver plant available N. Both abundance and diversity of microbial and fungal communities are required to optimize SOM mineralization, but these depend on many factors that are not treated explicitly in the model, which uses empirically optimized SOM mineralization potentials. We have therefore added 'internal N supply' to the list on line 595, in the sense that site-specific limitations to SOM mineralization are not explicitly accounted for in the model.

Line 616: more thorough discussions about optimal allocation theory, especially C-N interactions, are found in, e.g., Franklin et al. (2012, Tree Physiol. 32,648-).

A substantial discussion of optimal allocation theory is well beyond the scope of this paper. However we agree it makes sense to include a reference to Franklin et al. (2012), as well as Du and de Vries (2018) and Templer et al. 2012), in the introduction, line 100:

'…The questions of the allocation and fate of both the assimilated carbon (Franklin et al., 2012) and deposited nitrogen (Nadelhoffer et al., 1999; Templer et al., 2012; Du and de Vries, 2018) appear to be crucial. It has been suggested that N_r deposition plays a significant role…'

Additional references:

Du, E. and de Vries, W.: Nitrogen-induced new net primary production and carbon sequestration in global forests, Environ. Pollut., 242, 1476–1487, https://doi.org/10.1016/j.envpol.2018.08.041, 2018.

Franklin, O., Johansson, J., Dewar, R.C., Dieckmann, U., McMurtrie, R.E., Brännström, Å and Dybzinski, R.: Modeling carbon allocation in trees: a search for principles, Tree Physiol., 32, 648–666, https://doi.org/10.1093/treephys/tpr138, 2012.

Templer, P.H., Mack, M.C., Chapin, F.S. III, Christenson, L.M., Compton, J.E., Crook, H.D., Currie, W.S., Curtis, C.J., Dail, D.B., D'Antonio, C.M., Emmett, B.A., Epstein, H.E., Goodale, C.L., Gundersen, P., Hobbie, S.E., Holland, K., Hooper, D.U., Hungate, B.A., Lamontagne, S., Nadelhoffer, K.J., Osenberg, C.W., Perakis, S.S., Schleppi, P., Schimel, J., Schmidt, I.K., Sommerkorn, M., Spoelstra, J., Tietema, A., Wessel, W.W. and Zak, D.R.: Sinks for nitrogen inputs in terrestrial ecosystems: A meta-analysis of ^{15}N tracer field studies, Ecology, 93, 1816–829, https://doi.org/10.1890/11-1146.1, 2012.

Line 716: It is OK to cite Fog here, but why not cite authors, which discuss similar phenomena in forests (like Berg & Matzner 1997 Environ. Rev. 5, 1-).

We have added a reference to Berg and Matzner here. Additional reference:

Berg, B. and Matzner, E.: Effect of N deposition on decomposition of plant litter and soil organic matter in forest systems, Environ. Rev., 5, 1-25, https://doi.org/10.1139/a96-017, 1997.

Lines 743-744: below-ground autotrophic respiration does not exactly follow photosynthesis, but is also affected by seasonality in C below-ground allocation (Högberg et al. 2010 New Phytol. 187, 485-).

This sentence was rephrased as follows:

'…Since the below-ground autotrophic (root and rhizosphere) respiration component is regulated to a large extent by photosynthetic activity (Collalti and Prentice, 2019), as well as seasonality in below-ground C allocation (Högberg et al., 2010), and contributes a large part of Rsoil on an annual basis…'

Line 780: what is the difference between fertility and nutrient availability in this context?

Fertility includes, but is not limited to, the pool of nutrients available in the soil. Legout et al. (2014) phrase it this way: 'The definition of the chemical fertility of forest ecosystems should not be limited to the pool of plant available nutrients in the soil but must also integrate the cycling and recycling of nutrients characteristic of biogeochemical cycling'

Legout, A., Hansson, K., Van der Heijden, G., Laclau, J.-P., Augusto, L. and Ranger, J.: Fertilité chimique des sols forestiers: concepts de base (in French), Revue forestière française, 4–2014, 413–424, https://doi.org/10.4267/2042/56556, English translation available at http://mycor.nancy.inra.fr/ARBRE/wp-content/uploads/2015/02/SP_4_Chemical-fertility-offorest-soils-basic-concepts.pdf, 2014.

All texts to Figures and Tables should be self-explanatory. Thus, acronyms should thus always be explained in these texts.

We have reviewed the text in each figure caption and provided explanations where required.

Figures 3-6 and 8: these take some time to comprehend. A reader will need some guidance. And the text in the boxes are difficult to read and understand.

We have provided more explicit captions for these figures.

Figures 9 & 10: the texts by the symbols are difficult to read as sometimes they come on top of each other.

We have moved the symbols to avoid text overlapping in these two figures.

Anonymous Referee #2

Summary: Flechard et al. use a "meta-modelling" analysis of forest C fluxes and balance to examine the response of these processes to N deposition for ~30 sites (22 forests). They confirm that estimates of C gain from N deposition are smaller if environmental drivers are considered first.

General comments:

Overall, the analysis seems generally reasonable, and broadly supports the past re-analysis of forest C gain from N deposition by Sutton et al. (2008), which showed a much smaller C gain than imputed from the widely-critiqued Magnani et al. (2007) study. This text seems a bit long for that main take-home, with a data set only a bit larger – though analyzed in greater detail that that earlier dataset. It would be nice to have somewhat more focus in parsing this overall NEP response (i.e., more GPP vs less Reco or Rh?) beyond the surprisingly large reported GPP response.

Extensive discussion space is used on C sequestration efficiency (CSE =NEP/GPP), though it's not apparent quite what this adds over more in-depth examination of the individual C flux responses that go into this ratio. In particular, discussion of mechanistic explanations for the N effects on GPP and Rh (or Rsoil) would seem to be more directly related here – i.e., to explain saturation of the GPP response (discussed reasonably), or suppression of decomposition as a substantial portion of the overall dC/dN response. The direct effects of N on decomposition process appears largely restricted to the last page of the Discussion, and they merit much greater attention earlier and throughout the manuscript.

The reason we discuss CSE variability extensively is that we use various estimates of mean CSE to provide the step from the calculated $dGPP/dN_{dep}$ (effect on gross assimilation) to $dNEP/dN_{dep}$ (effect on C sequestration). In a nutshell, this is key to understanding how the paper works. It is clear that nitrogen addition can impact both assimilation (C gain) and respiration (C loss), and we choose to treat the two steps separately: 1) the assimilation (GPP) step by BASFOR meta-modelling, because we are confident that the model is reasonably well calibrated and constrained for GPP; and 2) the respiration (R_{eco}) step appears to be less well understood in terms of its ecological controls, and CSE is a useful normalized indicator or proxy to describe the fraction of C assimilated that is not lost by respiration. Discussing R_{eco} in more detail would be less handy because it scales with GPP.

The flux data show a much wider range of measurement-based CSE values than does the model, and therefore either A- the measurements are imperfect (measurement uncertainties are discussed in the Part I paper), and/or B- the model is imperfect as it does not reproduce the natural variability of observations (as discussed in both Part I and II of the study). Of course we know that both A- and B- are true to some extent; but the combination of both measurements and model(s) helps close knowledge gaps.

Once a $dGPP/dN_{dep}$ value was estimated by model-based normalization for non-nitrogen effects (step 1 of the approach), we had to rely (for step 2) on various mean estimates of CSE to translate the response to N_{dep} of gross photosynthesis into a response of net ecosystem productivity or C sequestration. This was because the variability in CSE_{obs} was not fully reproduced by the model, and faced with the uncertainty in individual CSE values, the reasonable approach was to test various mean CSE values and examine the plausibility of the different results (as shown in Table 2). Hence the extended discussion on the potential ecological controls of CSE, which do include a discussion of N effects on R_h. Therefore we do think that we have in effect provided *'…discussion of mechanistic explanations for the N effects on GPP and Rh (or Rsoil)…'* in the paper. An explicit outline of our approach was given on lines 587-597, which we believe summarizes the above arguments, and fully answers the Referee's question about why it is necessary to discuss CSE and its variability:

'…The EC-based flux data suggest that the N_{dep} response of forest productivity is clearer at the gross photosynthesis level, in patterns of (normalized) GPP differences among sites, than at the NEP level, where very large differences in CSE among sites lead to a de-coupling of N_{dep} and NEP. The response of GPP to N_{dep} appeared to be reasonably well constrained by both EC flux measurements and BASFOR modelling, which is why we chose to normalize GPP, not NEP.

The significantly better model performance obtained for GPP than for R_{eco} and NEP (Fig. 6 in Flechard et al., 2020) likely reveals a relatively poor understanding and mathematical representation of R_{eco} (especially for the soil heterotrophic and autotrophic components), and the factors controlling their variability among sites. The large unexplained variability in CSE and C sequestration potentials may also involve other limiting factors that could not be accounted for in our measurement/model analysis, since they are not treated in BASFOR. Such factors may be related to soil fertility, ecosystem health, tree mortality, insect or wind damages in the previous decade, incorrect assumptions on historical forest thinning, all affecting general productivity patterns…'

The reported response of a saturation of the growth response to N dep, coinciding with an increase in N losses, fits exactly within expectations of N saturation theory (e.g., Aber et al. 1989, 1998, BioScience), which deserves more explicit recognition and discussion.

The Referee is right; these seminal papers by Aber et al. are referred to in the revised paper. The following text was added to the introduction, line 106:

'*…but did not increase woody biomass or aboveground NPP*. In fact, Aber et al. (1989) even predicted 30 years ago that the last stage of nitrogen saturation in forests, following long term exposure to excess N_r deposition, would be characterized by reduced NPP or possibly tree death, even if during the early or intermediate stages the addition of N could boost productivity with no visible negative ecosystem impact beyond NO_3^- leaching. In that initial theory, Aber et al. (1989) suggested that plant uptake was the main N sink and led to increased photosynthesis and tree growth, while N was recycled through litter and humus to the available pool; this fertilization mechanism would saturate quickly, resulting in nitrate mobility. However, observations of large rates of soil nitrogen retention gradually led to the hypothesis that pools of dissolved organic carbon in soils allowed free-living microbial communities to compete with plants for N uptake. A revision of that theory by Aber et al. (1998) hypothesized the important role of mycorrhizal assimilation and root exudation as a process of N immobilization, and suggested that the process of nitrogen saturation involved soil microbial communities becoming bacterial dominated, rather than fungal or mycorrhizal dominated in pristine soils…'

Text added to introduction, line 154:

'*…implying that the net dC/dN response was likely non-linear,* in line with an overview of dC/dN response results from various approaches (De Vries et al., 2014a), possibly due to the onset of N saturation as predicted by Aber et al. (1989), and associated with enhanced acidification and increase sensitivity to drought, frost and disseases (De Vries et al., 2014b).'

Reference added to discussion, lines 549-550:

'…the highly non-linear response depends on current and historical Ndep exposure levels, and on the degree of N saturation (Aber et al., 1989, 1998)*, although other factors…'*

Additional references:

Aber, J.D., Nadelhoffer, K.J., Steudler, P. and Melillo, J.M.: Nitrogen Saturation in Northern Forest Ecosystems: Excess nitrogen from fossil fuel combustion may stress the biosphere, BioScience, 39, 6, 378-386, https://doi.org/10.2307/1311067, 1989.

Aber, J., McDowell, W., Nadelhoffer, K., Magill, A., Berntson, G., Kamakea, M., McNulty, S., Currie, W., Rustad, L. and Fernandez, I.: Nitrogen Saturation in Temperate Forest Ecosystems, BioScience, 48, 11, 921–934, https://doi.org/10.2307/1313296, 1998.

De Vries, W., Dobbertin, M.H., Solberg, S., van Dobben, H. and Schaub, M.: Impacts of acid deposition, ozone exposure and weather conditions on forest ecosystems in Europe: an overview, Plant Soil, 380, 1–45, https://doi.org/10.1007/s11104-014-2056-2, 2014b.

A less central suggestion: The authors state the importance of detailed site-level N deposition values over estimated modeled ones. While believable, this point would be supported more substantively by showing it directly, e.g., by comparing estimated v measured N deposition values, and quantitatively comparing dC/dN results for these two types of N deposition estimates.

In the Part I companion paper of this study, we provided extensive comparison and discussion of the N_r deposition levels based on our in-situ measurements versus modelled deposition values from the EMEP chemical transport model (CTM). We believe it is unnecessary to repeat this comparison in the present paper (Part II) and we have referred to Part I for more details in Methods (Section 2.1). As to a comparison of dC/dN results based on N_{dep} estimates from in-situ measurements versus CTM outputs, they would scale according to the slope (0.74) of the linear regression between the two N_{dep} estimates (since the intercept is negligeable) (see Fig. R6 below). Since N_{dep} is lower by around 25% in EMEP results versus the in situ estimates, the dC/dN estimates obtained on the basis of EMEP N_{dep} data would be approximately 33% larger than those obtained on the basis of in situ data, used in the paper.

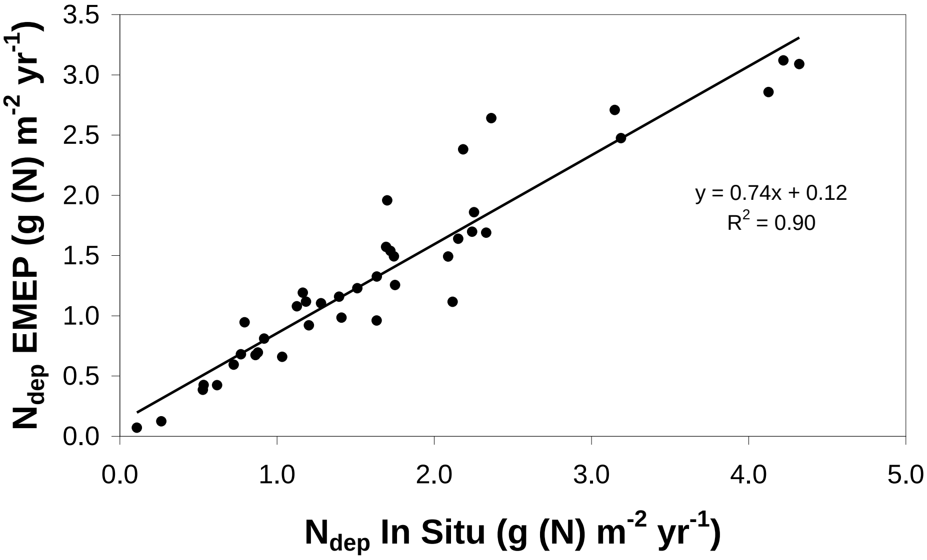

Figure R6. Comparison of total N_{dep} rates between in-situ measurement-based estimates and EMEP CTM outputs for the forest sites of this study.

Overall, the manuscript might be revised to reduce sometimes redundant-seeming extensive discussion of CSE, and provide greater and more direct emphasis on its novel insights (beyond Sutton et al. 2008, or classic N saturation theory). These points are addressed in the detailed comments below.

Detailed comments:

Abstract

Line 65 – somewhere in the abstract, specify the number of sites included in this analysis

This was added to lines 65-66:

'…in combination with eddy covariance CO2 exchange fluxes from a Europe-wide network of 22 forest flux towers…'

Line 67-71 – The reduction of dC/dN from considering factors other than N deposition was for GPP, not NEP, right? that should be clear in the abstract, which describes this response in terms of C sequestration, which generally aligns more closely with NEP. Similarly, be clear about which C cycle term yielded the 40-50 gC/gN response.

The sentence on lines 67-69 was rephrased thus:

‘…The response of forest net ecosystem productivity to nitrogen deposition ($dNEP/dN_{dep}$) was estimated after accounting for the effects on gross primary productivity (GPP) of the co-correlates by means of a meta-modelling standardization procedure, which resulted in a reduction by a factor of about 2 of the uncorrected, apparent $dGPP/dN_{dep}$ value…’

Line 71: dC/dN was changed to $dNEP/dN_{dep}$

Line 74 – text indicates that the dC/dN response saturates above 2.5-3.0 gN/m2/yr “due to” leaching and other losses.. but the latter don’t appear to be measured here? If this attribution is from the model analyses, do indicate that as a modeled result.

Leaching and other (gaseous) N losses were measured at some sites, and the results were shown and discussed in the companion (Part I) paper. The model also indicated increased N loss fractions at the upper end of the N_{dep} range. However, we agree that the causality implied by ‘due to’ was inappropriate in this sentence, and, in response to a comment by the other Referee (#1), we have modified the sentence in the following way:

‘…patterns of gross primary and net ecosystem productivity versus N_{dep} were non-linear, with no further growth responses at high N_{dep} levels (N_{dep} > 2.5–3 g (N) m^{-2} yr^{-1}) but accompanied by increasingly large ecosystem N losses by leaching and gaseous emissions.’

Main text:

Line 81-93. The cited references provide examples of experimental studies that indicate little or no increase in C sequestration from N addition, and might be presented as some of the conflicting evidence for a universal N-deposition induced C sink, rather than challenging the entire notion of this phenomenon in its entirety

We believe that we present references for both sides of the argument (A- nitrogen deposition is a major driver of C sequestration, versus B- nitrogen deposition affects C sequestration very little, versus C- anything in between) in a balanced way, which shows that the experimental evidence is conflicting . But it appears that some authors do question the notion entirely, e.g. Nadelhoffer et al. (Nature, 398, 1999) write ‘…*that elevated nitrogen deposition is unlikely to be a major contributor to the putative CO_2 sink in forested northern temperature regions.*’

Line 92. Is the Dezi et al. 2010 reference a model-based analysis of dC/dN? If so, clarify that it’s different from the empirical approaches of the other studies

We made it clear this was a modelling study:

‘…to 121 (in a model-based analysis by Dezi et al., 2010), …’

Line 93-97. In this review of dC/dN values – and throughout the manuscript – be clear as to which values pertain to which C pool (i.e., tree, soil, or whole ecosystem) or which specific C cycle processes.

The C pools considered were specified for each reference:

‘…ranging from 61– 98 for above-ground biomass increment in US forests (Thomas et al., 2010), 35–65 for above-ground biomass and soil organic matter (Erisman et al., 2011; Butterbach-Bahl and Gundersen, 2011), 16–33 for the whole ecosystem (Liu and Greaver, 2009), 5–75 (mid-range 20–40) for the whole ecosystem in European forests and heathlands (de Vries et al., 2009), and down to 13–14 for aboveground woody biomass in temperate and boreal forests (Schulte-Uebbing and de Vries, 2018), and 10–70 for the whole ecosystem for forests globally, increasing from tropical, to temperate to boreal forests (de Vries et al., 2014a; Du and de Vries, 2018).’

Line 107 – 120. It’s not wholly apparent why this review of basic N balance processes is needed? That is, nearly every forest N budget shows that new N deposition supplies only a small fraction of plant annual N demand compared to internal N recycling; the key (missing?) point is that the value of N deposition is that it may be acquired directly or at little energetic cost to plants, and can accumulate over time.

We agree, this is a key point that should be mentioned. We have added the following sentence on line 120:

Importantly, unlike other ecosystem mechanisms for acquiring N from the environment (resorption from senescing leaves, biological N_2 fixation, mobilization and uptake of N from soil solution or from SOM), the nitrogen supplied from atmospheric deposition comes at little or zero energetic cost (Shi et al., 2016), especially if absorbed directly at leaf level (Nair et al., 2016).

Additional references:

Shi, M., Fisher, J.B., Brzostek, E.R. and Phillips, R.P.: Carbon cost of plant nitrogen acquisition: global carbon cycle impact from an improved plant nitrogen cycle in the Community Land Model, Glob. Change Biol., 22, 1299–1314, https://doi.org/10.1111/gcb.13131, 2016.

Nair, R.K.F., Perks, M.P., Weatherall, A., Baggs, E.M. and Mencuccini, M.: Does canopy nitrogen uptake enhance carbon sequestration by trees?, Glob. Change Biol., 22, 875–888, https://doi.org/10.1111/gcb.13096, 2016.

Line 121. Add specific citations to this sentence critiquing "some previous estimates" for failing to account for factors other than N deposition.

We have added a reference to Magnani et al. (2007) on line 122.

Line 122-130. This section seems somewhat oversimplified in pitching its novelty: N deposition can, but does not necessarily covary with gradients of other environmental variables; this covariation often depends on the geographic region selected.

We agree that N deposition does not necessarily co-vary spatially with other environmental variables, and that is not what we meant. We meant that if there is co-variation then it must be factored out (we wrote, line 125: '…***if*** *Nr deposition is co-correlated with any of these other drivers*..').

Line 126, we have changed 'is usually' to 'can be' in '…*as ~~is usually~~ the case in spatial gradient survey analyses across a wide geographic domain…*'

The Magnani et al. (2007) simple regression analysis indeed failed to consider this covariation, but its problems seemed very effectively addressed by the Sutton et al. (2008) reanalysis, in demonstrating the need to consider variation in factors besides N deposition. Is the goal in this manuscript to do a similar analysis of tower-based C balance measurements in greater depth than that one? Other gradient analyses have also considered N deposition along with other environmental drivers, sometimes also considering nonlinear responses (e.g., Solberg et al. 2009, Thomas et al. 2010).

We are of course well aware that other gradient studies, such as these cited by the Referee, have investigated the multiple controls (including the potential covariation with N_{dep}) of forest productivity. We recognized in the Discussion (4.1 and 4.2) that i) our flux network dataset was much smaller compared with studies based on long term growth monitoring plots and large-scale forest inventories, and therefore unsuited to large-scale multiple regression-type analyses such as provided by e.g. Solberg et al. (2009); and ii) our final estimates of $dNEP/dN_{dep}$ were not significantly different from recent reviews. We have added references to such studies on line 143:

'…Altogether, these complex interactions mean that it is far from a simple task to untangle the N_r deposition effect on ecosystem C sequestration from the impacts of climatic, edaphic and management factors, when analysing data from diverse monitoring sites situated over a large geographic area (Laubhann et al., 2009; Solberg et al., 2009; Thomas et al., 2010).'

Additional References:

Laubhann, D., Sterba, H., Reinds, G.J. and de Vries, W.: The impact of atmospheric deposition and climate on forest growth in European monitoring plots: An empirical tree growth model, Forest Ecol. Manag., 258, 1751–1761, https://doi.org/10.1016/j.foreco.2008.09.050, 2009.

Solberg, S., Dobbertin, M., Reinds, G.J., Andreassen, K., Lange, H., Garcia Fernandez, P., Hildingsson, A. and de Vries, W.: Analyses of the impact of changes in atmospheric deposition and climate on forest growth in European monitoring plots: A stand growth approach, Forest Ecol. Manag., 258, 1735–1750, https://doi.org/10.1016/j.foreco.2008.09.057, 2009.

Nevertheless, the use of flux tower data in our study provides reliable measurement-based estimates of NEP, or NECB if there are no disturbances, which provide more direct measures of net ecosystem-scale C storage, compared with inventory-based studies, which must rely on critical assumptions regarding below-ground biomass and SOM balance to derive NECB estimates. There is value in both approaches, and they complement each other. What our study lacks in terms of site number and statistical power, is compensated by high temporal resolution data for assimilation and respiration fluxes at two dozen sites. This has enabled us to calibrate our forest ecosystem model using a multiple constraint Bayesian procedure, yielding generic model parameters for each tree class (Cameron et al., 2018). We did not adjust model parameters ‘manually’ for each site in this study, but allowed the Bayesian calibration algorithm to optimize all parameters at once, constrained by an a priori range (probability density function) for each parameter, and by all available calibration data (C and N fluxes, C stocks, tree heights and diameters, LAI, etc) with appropriate uncertainty estimates on the measurements.

The re-analysis by Sutton et al. (2008) of the Magnani et al. (2007) flux tower dataset did show convincingly that covariation of climate and N_{dep} needed to be considered, and we have acknowledged (line 535) that our analysis came to the same conclusion. However we wish to stress a few key methodological differences, which give our study the novelty value questioned by the Referee:

- Our approach is a model-enhanced data analysis, by which measurement-based GPP is standardized into GPP* by the model. This is by contrast to the purely model-based (data-free) analysis used with the Edinburgh Forest Model (EFM) in the Sutton et al. (2008) paper. Disentangling the effects of N_{dep} vs. climate was then done simply by running the EFM n times (i.e. once for each of the n forest sites) and calculating a multiple linear regression of the lifetime average NEP vs. {lifetime average N_{dep} + Temperature + Precipitation}.
- The role of soils was not analysed in Sutton et al. (2008), i.e. the EFM was run with site-specific soil data, but soil properties did not appear in the multiple regression.
- The meta-modelling approach of the present paper involved running BASFOR n^2 times at each site (i.e. n^3 in total for all n sites) for the climate and soil normalization runs, by swapping climate data, or soil physical properties, between sites.
- This means that the 2008 analysis was a non-mechanistic analysis that could – fundamentally – never rule out any of the correlated variables as being the main driver of variation in NEP, as we point out in 2.2.4; while the new analysis uses the strength of mechanistic modelling by actually doing an input-sensitivity analysis.
- The EFM had not at the time undergone any thorough, multiple contraint calibration (in contrast to BASFOR). EFM results for individual runs were not compared to data: all that was compared to data was the regression line of productivity vs. N_{dep}.
- Because the new (BASFOR) modelling is grounded in real data, the new paper can identify a key role for differences between sites in level of N-saturation, which was not possible before. The new analysis in terms of Carbon Sequestration Efficiency clarifies the important implications of the results.

To summarize, the novelty of the study (including both Parts I and II) lies in the methods employed (detailed measurements of dynamic C and N fluxes at each site; model-enhanced analysis and untangling of the flux data and inter-relationships), rather than in the key end results themselves (non-linear response of C sequestration to N_{dep}, decline of forest productivity at N-saturated sites), which admittedly have long been known. However, with the development and standardization of flux tower networks in Europe (ICOS) or worldwide (FLUXNET), we can envisage further such studies, at many more sites, using measurement-model fusion techniques.

Line 175-176. Elaborate on what exactly is meant by “soil C pools rely on various assumptions or empirical models for their estimation.” Assumed and modeled soil C can often vary markedly from measured values; how well do these assumptions work?

By “soil C pools” we did not mean specifically SOM pools, but more generally below-ground C pools, including therefore also fine and coarse roots. Below-ground C pools are much more difficult to evaluate on the basis of measurements than above-ground stocks. SOM stocks are evaluated on the basis of soil cores, but often there is large spatial heterogeneity in soil depth, in vertical horizon structure, etc, and therefore we have to rely on the assumption that the spatial samping scheme is statistically representative of the whole ecosystem (or the flux tower footprint). The root C pools are evaluated on the basis of allometric relationships to the above-ground vegetation, which effectively are empirical models of the ratio of below-ground to above-ground tree C stocks. In short, we are not dealing with measurements per se, but with measurement-derived quantities, whose uncertainties need to be recognized and quantified for model evaluation and calibration. We have changed ‘*soil carbon pools*’ to ‘below-ground C pools’ in the sentence, and add ‘… (e.g. flux partitioning procedure to derive GPP from NEE; allometric relations for tree and root C stocks; spatial representativeness of soil core sampling for SOM).’ after *‘…on the basis of measured data*’, line 176.

Line 184. Specify the minimum and/or mean number of years of EC data are used to compute the C fluxes of interest here.

The mean number of years of EC data was 5 (site-specific details provided in Table S6 of the companion Part I paper).

‘…the C datasets include multi-annual (on average, 5-year) mean estimates of NEP, GPP and R_{eco}…’

Line 202-204. Does this model’s soil dynamics allow it to represent the inhibitory effect of N deposition on soil decomposition? If not, this point should receive explicit attention in the Methods and/or Discussion, on how this effect of N was considered in this analysis.

Indeed the model does not contain any such mechanism, and we have made this clear in the discussion on the effect of N addition on soil respiration, line 728:

‘…Of the five afore-mentioned mechanisms potentially involved in the suppression of soil respiration by N addition, only the first one (control by N availability of the root/shoot allocation ratio) is functional in BASFOR, and therefore our simulations do not include the other inhibitory effects of excess N on mycorrhizal, fungal and bacterial respiration.’

Line 207-208. It’s certainly difficult to reliably simulate N loss fluxes to DON and N2, and it’s correspondingly understandable that this set of model-based estimates would not include them. However, when measured, these fluxes can dominate ecosystem N loss fluxes and should thus receive more attention as to the uncertainties introduced by their omission in these calculations.

We fully agree that these fluxes can be significant, or even dominant, as discussed in the companion Part I paper, which focused on uncertainties in the C and N fluxes derived from observations. We wrote explicitly in the present paper (lines 273-276) that these fluxes are not considered in BASFOR, but unfortunately there are not enough data to address the Referee’s question about uncertainties from a modelling viewpoint. We have added the following sentence to the conclusion, line 785:

‘…*N_r losses, especially as NO_3^- leaching*. Further sources of uncertainty in our forest ecosystem model involve missing – but possibly large – terms of the N cycle, such as N_2 fixation, N_2 loss by denitrification, DON uptake by trees and DON leaching.’

Line 246 / Section 2.2.3 – why focus so much text here and the Discussion on the ratio of C sequestration _efficiency_, CSE, (NEP / GPP and similar), rather than NEP itself and its component parts (i.e., increased GPP? Suppressed Reco or Rh)? The Introduction provides no context or central questions for focusing on questions concerning the CSE ratio, and so this emphasis seems somewhat unexpected and extraneous here, and the lengthy text in the Discussion (~line 600-700)

We have addressed, in our opening paragraph and response to the Referee’s general comments (see above), the rationale for the focus on the CSE term in this study. We have explained how the paper focuses first on the response of assimilation (GPP) to N_{dep} and other factors (through meta-modelling), and second on the factors (including N_{dep}) controlling respiration or R_{eco}, through CSE by proxy. However, we agree that the CSE concept, which is central for the discussion but is not introduced until Section 2.2.3, should be mentioned in the introduction. We have done this at the end of the first paragraph, line 87, and referred to the companion Part I paper, in which the concept was first introduced:

‘…actually sequestered in the ecosystem. Indeed, it is possible to view this ratio of NECB to GPP as the efficiency of the long term retention in the system of the assimilated C, in other words a carbon sequestration efficiency (CSE = NECB/GPP) (Flechard et al., 2020).’

Line 265 – late text (line 273) indicate N fixation wasn’t considered, so be consistent with that point here

We are not sure what the Referee means. N_2 fixation is not considered in the model (as stated line 273), and therefore it follows we cannot account for N_2 fixation in our definition of NUPE in Eq. 6-8.

Line 314-316 – identify what is “the broad pattern of GPP vs N dep. in Flechard et al. (2019).”

We have added a brief description of the pattern in this sentence:

‘…*relationships reported in Flechard et al. (2020)*, i.e. a non-linear increase and eventual saturation of GPP as N_{dep} increases beyond a critical threshold, *did not show any marked difference*…’

Line 373-374 – in this N balance (N mineralisation + N dep – N plant uptake – N leach – N emissions), what about accumulation of N in soils or soil organic matter? Often a very large if not the largest sink.

There are several interesting N balances that could be examined. In the paper (lines 369-378), we focus on the balance of NMIN, i.e. inorganic N in the soils, and no other N-balance. One could also be interested in Nsoil (=NMIN + NLITT + NSOMF + NSOMS), which the Referee is referring to, or also Nsys (= Nsoil + Ntree).

We choose to study the NMIN balance because mineral N is the only source of N available to the trees in our model (BASFOR does not consider DON supply and uptake, as explained above in reply to Referee #1), and our objective was to understand how increased N-availability to trees from N-deposition affects growth, C storage and other GHG fluxes. The only processes that affect that NMIN-balance, in the model, are the ones that we mention, i.e. increases in the NMIN pool from Nminer and Ndep, and decreases from Nupt, Nleach, Nemission. We ignore adsorption of NMIN to soil particles and the reverse (release from soil particles back to the free NMIN pool) because those processes are assumed to be small and in a stable mutual balance, i.e. the amount of adsorbed (unavailable) mineral N is not likely to change much over the lifetime of the trees, as soil pH etc. do not fluctuate much. It thus seems reasonable to not include those in the model.

The referee is suggesting to study the long-term accumulation of N as a chemical part of soil organic matter (e.g. when tree senescence adds more organic matter than is mineralized, so soil organic N increases over time). This is indeed interesting in itself and calculated by BASFOR as the net change over time in NLITT+NSOMF+NSOMS.

However, we believe that this is not a separate process of direct relevance to the balance of NMIN and to the central question of the study (what is the impact of N_{dep} on C sequestration). This soil organic N balance reflects the shift of organic matter from plants to soil, minus mineralization of that SOM, in contrast to mineralization itself which directly contributes to N availability and the supply of N to trees that enables C sequestration.

We have added a sentence line 369, before the description of the N budgets, to clarify this point:

'For nitrogen, by contrast to carbon, the focus of the budget diagrams is not on changes over time of the total ecosystem (tree + soil, organic + mineral) N pools. Rather, we examine in Fig. 2 and S4–S6 the extent to which N_r deposition contributes to the mineral N pool (NMIN), which in the model is considered to be the only source of N available to the trees and therefore acts as a control of C assimilation and ultimately sequestration. In these diagrams for NMIN, *the largest (horizontal) arrows indicate the modelled internal ecosystem N cycling terms…'*

Line 457 – suggest "at the low_er_ N dep sites. . ." That is, 1.0 g/N/m2 is often considered elevated.

Agreed, we have changed to 'at the lower N_{dep} sites'.

Line 466 – clarify which "this set" is meant – higher or lower N dep group?

The answer to the Referee's question is provided on the following line (467), where we specify that the regressions were *'…either calculated over the whole range of 22 sites, or for a subset of 18 sites that excludes the four highest deposition sites (>2.5 g (N) m^{-2} yr^{-1})'*. (data are provided in Table 2 in italics or bold characters, respectively)

Line 468-485. These dC-GPP/dN values are simply _enormous_ and correspondingly difficult to fathom – even when reduced from 425 to 234 gC/gN! How do these compare to empirical NPP values? Presumably a 50% GPP to NPP efficiency would yield something like 212 to 117 gC/gN, still well beyond empirical NPP responses. How /why is it so large compared to eventual dC-NEP/dN response?

There are several arguments to consider in response to the Referee's question.

1- We show in the paper that it is misleading to use GPP_{obs} directly in a regression versus N_{dep} alone, that observation-based GPP must be standardized for climate, soil and age, and that only GPP* should be used for the purpose of calculating a response to N_{dep}. Therefore the figure of 425 g (C) g^{-1} (N), cited above by the Referee and taken from Table 2, is precisely that: an uncorrected, overestimated response, that we show in Table 2 only for the purpose of comparison. The only correct number to consider here is the one derived from GPP*, obtained when GPP_{obs} is corrected for climate, soil and age, i.e. 234 g (C) g^{-1} (N) (see Table 2, as explained on lines 471-472).
2- From this $dGPP^*/dN_{dep}$ slope, the Referee is right in saying that the model would assume a theoretical reduction of ~50% from GPP to NPP to account for autotrophic respiration, thus $dNPP/dN_{dep}$ would be of the order of 117 g (C) g^{-1} (N). But NPP is still not NEP; there is C allocation to exudation and mycorrhizae, which ends up being respired too, and heterotrophic respiration of SOM from free-living microbes (R_{het}). Eddy covariance flux towers tell us that ecosystem respiration (R_{eco}) is around 70-90% of GPP, more precisely 75% on average in the case of our forest dataset, i.e. NEP is around 25% of GPP (CSE~25%) on average (though with large variability, as pointed out in the paper) *(See also Figure R5 above, provided in our response to Referee #1)*. This means that R_{het} removes around 25% of GPP (on average) from NPP; i.e., from the above $dNPP/dN_{dep}$ estimate of 117 g (C) g^{-1} (N), about 0.25*234 is further removed to account for R_{het}. The resulting $dNEP/dN_{dep}$ from this back-of-the-envelope calculation is 50-60 g (C) g^{-1} (N), and comparable in magnitude to what we show in Table 2 from the proper analysis.
3- A ratio of the order of 0.5 for NEP/NPP is also suggested by the analysis by Du and De Vries (Environmental Pollution, 242, 1476-1487, 2018), who estimate that the part of global forest NPP that is supported by external N inputs is 3.48 Pg C yr^{-1} (see their Table 7), while the corresponding figure for the net global forest biome C sink (NEP) is 1.83 Pg C yr^{-1} (see their Table 9), i.e. a NEP/NPP ratio of 0.53.

4- If the ratios of NPP/GPP and NEP/GPP are both fairly constrained (by the literature and by flux towers, respectively) and mutually consistent in our analysis, and if the final dNEP/dN_{dep} responses are considered plausible by the Referee, then the dNPP/dN_{dep} response should be, too. Our model was calibrated (see Bayesian calibration paper by Cameron et al., 2018, to which we refer in both papers) using multiple constraints, including measured NEE and evapotranspiration (eddy covariance at all sites), soil heterotrophic respiration (where available), carbon stocks in above- and below ground tree pools, in soil organic matter, also tree heights and diameters at breast height (at different dates wherever available), leaf area index, soil water content, and soil N emissions. We believe that this multiple constraint approach should ensure that NPP is not massively over-estimated, based on the model's asumptions.

The Referee argues that the NPP response is much (unreasonably) larger than empirical responses, but empirical NPP estimates are commonly made through the proxy of biomass production (BP), whereby BP is assumed to constitute the largest fraction of NPP. However, field measurements in forests show substantial variation in the BP/GPP ratio (e.g. Vicca et al., Ecology Letters, 15: 520–526, 2012). Also, some studies have considered only above-ground (not total) NPP responses to N_{dep}, which would make the number smaller. In our study we chose to focus on the responses of GPP and NEP to N_{dep}, because we have measurement-based values for both terms, but not the response of NPP, due to the above uncertainties and because no reliable NPP measurements were available.

Line 495-502. These dC-NEP/dN values (~40-60 gC/gN) seem more consistent with empirical responses: to what extent are these values due to modeled plant vs soil C sequestration? Is the soil C sink from additional litterfall inputs or from suppressed decomposition?

This question is addressed in Fig. 5, which shows the fractions of the net ecosystem carbon balance (NECB) that are stored in above-ground tree compartments (CLBS: carbon in leaves, branches and stems), in roots (CR), litter (CLITT) and in soil organic matter (CSOM). The results are discussed on lines 408-418; clearly the model allocates most of the C storage to above-ground tree parts (woody biomass), but over a lifetime the fraction stored in wood vs. SOM depends on the age of the forest.

Line 535. Yes, the results here seem to confirm that of Sutton et al. (2008). How does this analysis provide additional insights beyond that one?

We have addressed this question in detail previously, in response to another comment by the Referee (see above).

Line 537. Provide citations for observations of N losses. Thresholds of 0.8 – 1.0 g N/m2/yr for N leaching have been reported commonly (e.g., MacDonald et al. 2003, Global Change Biology, and similar).

We have modified this sentence on line 537:

'… *Observations and model simulations both indicate that the N_{loss} fraction of N_{supply} increases with N_{dep}*, consistent with widespread observations of increasing NO_3^- leaching above N_{dep} thresholds as low as 1.0 g (N) m^{-2} yr^{-1} in European forests (Dise and Wright, 1995; De Vries et al, 2007; Dise et al., 2009), and exacerbated by large C/N ratios (> 25) in the organic horizons (Gundersen et al., 1998; MacDonald et al., 2002). Higher thresholds for N_{dep} around 2.5 g (N) m^{-2} yr^{-1} (Dise and Wright, 1995; Van der Salm et al., 2007) typically indicate advanced saturation stages. *Thus, at many sites but especially those with N_{dep} > 1.5–2 g (N) m^{-2} yr^{-1}*,…'

Additional references:

De Vries, W., van der Salm, C., Reinds, G.J. and Erisman, J.W.: Element fluxes through European forest ecosystems and 1205 their relationships with stand and site characteristics, Environ. Pollut., 148, 501–513, https://doi.org/10.1016/j.envpol.2006.12.001, 2007.

Dise, N.B. and Wright, R.F.: Nitrogen leaching from European forests in relation to nitrogen deposition, Forest Ecol. Manag., 71, 153–161, https://doi.org/10.1016/0378-1127(94)06092-W, 1995.

Dise, N.B., Rothwell, J.J., Gauci, V., van der Salm, C. and de Vries, W.: Predicting dissolved inorganic nitrogen leaching in European forests using two independent databases, Sci. Total Environ., 1225 407, 1798–1808, https://doi.org/10.1016/j.scitotenv.2008.11.003, 2009.

Gundersen, P., Callesen, I. and de Vries, W.: Nitrate leaching in forest soils is related to forest floor C/N ratios, Environ. Pollut., 102, 403–407, https://doi.org/10.1016/B978-0-08-043201-4.50058-7 , 1998.

MacDonald, J.A., Dise, N.B., Matzner, E., Armbruster, M., Gundersen, P., Forsuis, M.: Nitrogen input together with ecosystem nitrogen enrichment predict nitrate leaching from European forests, Glob. Change Biol., 8, 1028–1033, https://doi.org/10.1046/j.1365-2486.2002.00532.x, 2002.

Van der Salm, C., de Vries, W., Reinds, G.J. and Dise, N.B.: N leaching across European forests: Derivation and validation of empirical relationships using data from intensive monitoring plots, Forest Ecol. Manag., 238, 81–91, https://doi.org/10.1016/j.foreco.2006.09.092, 2007.

Line 540. These responses are exactly as expected – i.e., The saturating response of ecosystem NPP to N deposition, and corresponding increase in N losses, are standard predictions of classic N saturation theory as originally proposed (e.g., Aber et al. 1989 & 1998, BioScience). Discuss how this work provides an advance over that prior set of expectations.

Our results are consistent with (or not significantly different from) previously published thresholds for early and advanced N saturation. In that sense the end results are not new. However, to our knowledge, this may be the first time such results have been published based on eddy covariance GPP and NEP datasets, complemented by in-situ N flux (deposition, emission, leaching) measurements. As mentioned earlier, the multiplication of flux towers worldwide holds much promise for generalized coupled C/N studies, but this can only be achieved if these eddy covariance tower sites are also equipped to quantify nitrogen inputs and losses to the same degree of accuracy as for CO_2.

A reference to Aber et al. (1989, 1998) was added to line 542:

'…beyond which growth and C sequestration were not further increased or even reversed, as predicted in classical N saturation theory by Aber et al. (1989, 1998).'

Line 560-570. This paragraph states that the detailed, more-precise N deposition measurements improve calculation of dC/dN responses.. and while plausible, it should first be demonstrated how these estimates compare with the simpler alternative.

We have addressed this question in detail previously, in response to another comment by the Referee (see above, Fig. R6).

Line 580-582. This conclusion on Reco vs N dep does not seem to have been discussed in the Results?

The variability of R_{eco} and CSE was discussed extensively in the companion (Part I) paper, and we agree that we need to add a reference to Flechard et al. (2020) on line 580. However, note that at this stage we do not draw any '*conclusion on R_{eco} vs N_{dep}*', as the Referee suggests, but merely point to the large unexplained variance in CSE and R_{eco}.

Line 585. Per above, the annual N input is small relative to annual N demand. But its accumulation over time can support a much larger fraction of N demand.

That is a valid point. We have added the following sentence to line 586:

‘…measurement uncertainties and noise in C and N budgets. Conversely, the effect may be delayed and may manifest even after N_r deposition levels have decreased, as the past N accumulation in soil may support later growth through enhanced N supply.’

Line ~590. This would seem one of several places to mention the effect of N on decomposition

This short section (lines 587-597) was written to introduce the later discussion items on potential drivers of the CSE (4.3), including nutrient limitation, N saturation, forest history and effects of N on soil respiration. This is why these items are mentioned here very briefly, as a preamble to the sections that follow (e.g. the effect of N on decomposition is discussed in 4.3.3). We have added the following sentence to line 597 to clarify the transition:

‘…*all affecting general productivity patterns*. Since the observed variability in CSE is key to understanding and quantifying the real-world NEP response to N_{dep} (beyond the relatively well constrained response of GPP in the model world), we explore some of the main issues in the following sections.’

Line 598 – 708. It’s not apparent why so much emphasis is placed on carbon sequestration efficiency (CSE = NEP / GPP) rather than the component C fluxes (GPP, NPP, NEP, Rh). It seems somewhat redundant with these other responses, and a direct outcome of individual responses. What additional insights does it provide?

The CSE indicator (=NEP/GPP) was introduced in the companion (Part I) paper to make it apparent that:

1. Flux tower eddy covariance data indicate a very large range of the ratio $C_{sequestered}/C_{assimilated}$, which our ecosystem model did not reproduce, and which we believe most ecosystem models would fail to reproduce;
2. The unexplained variability in CSE could indicate large measurement uncertainties; this was discussed in some detail in the Part I paper;
3. And / or our mechanistic understanding (and therefore ecosystem models) is incomplete;
4. We believe both 2 and 3 cannot be ruled out, but by considering both measurement and model uncertainties side by side, we may further our understanding;
5. CSE is useful for interpreting differences in R_{eco} between sites, because R_{eco} scales with GPP, and we have a very large variability of climates (and soils) at the European scale in this dataset; therefore a normalized indicator can be compared, while absolute values cannot.

We have explained this approach in the present paper, in Methods (2.2.3, 2.2.4) and reiterated in Results (3.3, 3.4 and especially 3.5), and discussion (4.2). As discussed above in our response to an earlier comment by the Referee, it seems logical to proceed in two stages: i) response of GPP, then ii) response of NEP via the proxy of a mean CSE, since the model does not allow us fundamentally to understand differences in R_{het} and R_{aut} between sites.

Line 649. A large soil C stock doesn’t necessarily indicate higher heterotrophic respiration responses – and can result from the opposite situation (i.e., lower Rhet allows more soil C to accumulate).

We agree with the Referee, this was an over-simplification and indeed misleading. We have rephrased thus:

‘…The EN4, EN6, EN17 sites had the three largest soil organic contents (SOC, Fig. 9A), which may either have induced larger rates of heterotrophic respiration, or may instead indicate low-fertility wet soils where both assimilation and respiration are suppressed. However, EN4 has also been reported…’

Line 652. The history of N and S deposition at this site (EN8) indeed might be important. What about considering cumulative N deposition across the range of sites?

Unfortunately not all (in fact very few) measurement sites possess the historical depth of N_{dep} (and also N leaching) measurements that are available at EN8. In a way we have considered cumulative N_{dep} at all sites through the assumed historical curve of N_{dep} as an input to the model (see Fig. R1 above, which was added to the paper), but the temporal trends are model-derived and identical for all sites, though scaled for actual N_{dep} measurements made around 2005-2010.

We believe our inclusion of Fig. R1 in the paper, and the accompanying description text (see above), in response to a comment by Referee #1, have clarified the temporal aspects of the modelling study, including the way historical N_{dep} was handled.

Line 709 onward. This seems quite late for a first substantive mention of the direct effects of extra N on decomposition and belowground processes, often shown to be of comparable magnitude as many aboveground responses (e.g., Janssens et al. 2010, Frey et al. 2014).

Our paper's ultimate objective was to derive $dNEP/dN_{dep}$ estimates using a data-model fusion approach. The discussion of N effects on below-ground respiratory processes, although fully relevant and contributing to the overall discussion, was not the central theme. The occurrence of this specific discussion topic in the last part of the paper made sense with respect to the logical flow of the paper:

1. Describe model simulations for the C and N cycles (short-term and lifetime), as the foundation for the scenarios / input-sensitivity simulations used in 2:
2. Describe method and results for the model-based normalization of the GPP response to N_{dep}
3. Step from $dGPP/dN_{dep}$ to $dNEP/dN_{dep}$, using the CSE proxy
4. Discuss limitations and uncertainties of the approach
5. Discuss ecological controls of CSE variability

In addition: does the modelling approach consider these processes, or would it miss them?

We repeat here the response we made earlier to a similar question by the Referee:

************ Indeed the model does not contain any such mechanism, and we have made this clear in the discussion on the effect of N addition on soil respiration, line 728:*

*'…Of the five afore-mentioned mechanisms potentially involved in the suppression of soil respiration by N addition, only the first one (control by N availability of the root/shoot allocation ratio) is functional in BASFOR, and therefore our simulations do not include the other inhibitory effects of excess N on mycorrhizal, fungal and bacterial respiration.' ************

Line 714. Add citation(s) for this "traditional theory of role of N..."

We have added this reference to Alexander (1977):

Alexander, M.: Introduction to soil microbiology, 2nd ed., John Wiley and Sons, London, 467pp., 1977.

Line 737-742. This content seems more appropriate to the Methods, as a general data synthesis activity part of this study. Similarly line 743-763 seem more appropriate to Results.

The data were collected as part of the data synthesis activity and described in Methods (section 2.3.2, Table S7) of the companion (Part I) paper, to which we refer on line 746 of the present paper. We don't think it is necessary to repeat this information in Methods of the present paper. The short description on lines 743-763 does not actually describe results of the present study, but supplementary information that comes in support of our analysis and therefore fits better in the discussion section.

Line 775. NEP dC/dN of 40-50 is on the lower end of inventory? Many inventory-based assessments seem to show something in this range, with ~20-25 for trees and 20-25 gC/gN for soil.

We agree with the Referee, and have changed the wording of this sentence accordingly:

'…and comparable with current estimates obtained from inventory data and deposition rates…

Carbon/nitrogen interactions in European forests and semi-natural vegetation. Part II: Untangling climatic, edaphic, management and nitrogen deposition effects on carbon sequestration potentials

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Abstract. The effects of atmospheric nitrogen deposition (N_{dep}) on carbon (C) sequestration in forests have often been assessed by relating differences in productivity to spatial variations of N_{dep} across a large geographic domain. These correlations generally suffer from covariation of other confounding variables related to climate and other growth-limiting factors, as well as large uncertainties in total (dry + wet) reactive nitrogen (N_r) deposition. We propose a methodology for untangling the effects of N_{dep} from those of meteorological variables, soil water retention capacity and stand age, using a mechanistic forest growth model in combination with eddy covariance CO_2 exchange fluxes from a Europe-wide network of 22 forest flux towers. Total N_r deposition rates were estimated from local measurements as far as possible. The forest data were compared with data from natural or semi-natural, non-woody vegetation sites.

The response of forest net ecosystem productivity to nitrogen deposition ($dNEP/dN_{dep}$) was estimated after accounting for the effects on gross primary productivity (GPP) of the co-correlates by means of a meta-modelling standardization procedure, which resulted in a reduction by a factor of about 2 of the uncorrected, apparent $dGPP/dN_{dep}$ value. This model-enhanced analysis of the C and N_{dep} flux observations at the scale of the European network suggests a mean overall $dNEP/dN_{dep}$ response of forest lifetime C sequestration to N_{dep} of the order of 40–50 g (C) g^{-1} (N), which is slightly larger but not significantly different from the range of estimates published in the most recent reviews. Importantly, patterns of gross primary and net ecosystem productivity versus N_{dep} were non-linear, with no further growth responses at high N_{dep} levels (N_{dep} > 2.5–3 g (N) m^{-2} yr^{-1}) but accompanied by increasingly large ecosystem N losses by leaching and gaseous emissions. The reduced increase in productivity per unit N deposited at high N_{dep} levels implies that the forecast increased N_r emissions and increased N_{dep} levels in large areas of Asia may not positively impact the continent's forest CO_2 sink. The large level of unexplained variability in observed carbon sequestration efficiency (CSE) across sites further adds to the uncertainty in the dC/dN response.

Commenté [c1]: Sentence re-phrased for more clarity

1 Introduction

Atmospheric reactive nitrogen (N_r) deposition (N_{dep}) has often been suggested to be a major driver of the large forest carbon (C) sink observed in the Northern Hemisphere (Reay et al., 2008; Ciais et al., 2013), but this view has been challenged, both in temperate (Nadelhoffer et al., 1999; Lovett et al., 2013) and in boreal regions (Gundale et al., 2014). In principle, there is a general consensus that N limitation significantly reduces net primary productivity (NPP) (LeBauer and Treseder, 2008; Zaehle and Dalmonech, 2011; Finzi et al., 2007). However, the measure of carbon sequestration is not the NPP, but the long term net ecosystem carbon balance (NECB; Chapin et al., 2006) or the net biome productivity at a large spatial scale (NBP; Schulze et al., 2010), whereby heterotrophic respiration (R_{het}) and all other C losses, including exported wood products and other disturbances over a forest lifetime, reduce the fraction of photosynthesized C (gross primary production, GPP) that is actually sequestered in the ecosystem. Indeed, it is possible to view this ratio of NECB to GPP as the efficiency of the long term retention in the system of the assimilated C, in other words a carbon sequestration efficiency (CSE = NECB/GPP) (Flechard et al., 2020).

Commenté [CF2]: Introduced CSE concept early in the paper

There is considerable debate as to the magnitude of the "fertilisation" role that atmospheric N_r deposition may play on forest carbon balance, as illustrated by the controversy over the study by Magnani et al. (2007) and subsequent comments by Högberg (2007), De Schrijver et al. (2008), Sutton et al. (2008), and others. Estimates of the dC/dN response (mass C stored in the ecosystem per mass atmospheric N deposited) vary across these studies over an order of magnitude, from 30–70 g (C) g^{-1} (N) (de Vries et al., 2008; Sutton et al., 2008; Högberg, 2012), to 121 (in a model-based analysis by Dezi et al., 2010), to 200–725 (Magnani et al., 2007, 2008). Recent reviews have suggested mean dC/dN responses generally well below 100 g (C) g^{-1} (N), ranging from 61–98 for above-ground biomass increment in US forests (Thomas et al., 2010), 35–65 for above-ground biomass and soil organic matter (Erisman et al., 2011; Butterbach-Bahl and Gundersen, 2011), 16–33 for the whole ecosystem (Liu and Greaver, 2009), 5–75 (mid-range 20–40) for the whole ecosystem in European forests and heathlands (de Vries et al., 2009), and down to 13–14 for aboveground woody biomass in temperate and boreal forests (Schulte-Uebbing and de Vries, 2018), and 10–70 for the whole ecosystem for forests globally, increasing from tropical, to temperate, to boreal forests (de Vries et al., 2014a; Du and de Vries, 2018).

Commenté [CF3]: Specified ecosystem compartments for which the dC/dN responses were evaluated in these publications

A better understanding of processes controlling the dC/dN response is key to predicting the magnitude of the forest C sink under global change in response to changing patterns of reactive nitrogen (N_r) emissions and deposition (Fowler et al., 2015). The questions of the allocation and fate of both the assimilated carbon (Franklin et al., 2012) and deposited nitrogen (Nadelhoffer et al., 1999; Templer et al., 2012; Du and de Vries, 2018) appear to be crucial. It has been suggested that N_r deposition plays a significant role in promoting the carbon sink strength only if N is stored in woody tissues with high C/N ratios (>200–500) and long turnover times, as opposed to soil organic matter (SOM) with C/N ratios that are an order of magnitude smaller (de Vries et al., 2008). Nadelhoffer et al. (1999) argued on the basis of a review of ^{15}N tracer experiments that soil, rather than tree biomass, was the primary sink for the added nitrogen in temperate forests. However, based on a recent synthesis of ^{15}N tracer field experiments (only including measurements of ^{15}N recovery after > 1 year of ^{15}N addition), Du and de Vries (2018) estimated that tree biomass was the primary sink for the added nitrogen in both boreal and temperate forests (about 70%), with the remaining 30% retained in soil. At sites with elevated N inputs, increasingly large fractions are lost as nitrate (NO_3^-) leaching. Lovett et al. (2013) found in north-eastern US forests that added N increased C and N stocks and the C/N ratio in the forest floor, but did not increase woody biomass or aboveground NPP.

Commenté [CF4]: Expanded contrasting views on the contribution of tree and soil carbon sequestration in response to N deposition.
Added references on allocation theory.

In fact, Aber et al. (1989) even predicted 30 years ago that the last stage of nitrogen saturation in forests, following long term exposure to excess N_r deposition, would be characterized by reduced NPP or possibly tree death, even if during the early or intermediate stages the addition of N could boost productivity with no visible negative ecosystem impact beyond NO_3^- leaching. In that initial theory, Aber et al. (1989) suggested that plant uptake was the main N sink and led to increased photosynthesis and tree growth, while N was recycled through litter and humus to the available pool; this fertilization mechanism would saturate quickly, resulting in nitrate mobility. However, observations of large rates of soil nitrogen retention gradually led to the hypothesis that pools of dissolved organic carbon in soils allowed free-living microbial communities to compete with plants for N uptake. A revision of that theory by Aber et al. (1998) hypothesized the important role of mycorrhizal assimilation and root exudation as a process of N immobilization, and suggested that the process of nitrogen saturation involved soil microbial communities becoming bacterial dominated, rather than fungal or mycorrhizal dominated in pristine soils.

Commenté [c5]: Added a paragraph on classical N saturation theory by Aber et al

Atmospheric N_r deposition is rarely the dominant source of N supply for forests and semi-natural vegetation. Ecosystem internal turnover (e.g. leaf fall and subsequent decomposition of leaf litter) and mineralization of SOM provide annually larger amounts of mineral N than N_{dep} (although ultimately, over pedogenic time scales much of the N contained in SOM is of atmospheric origin). In addition, resorption mechanisms help conserve within the tree the externally acquired N (and other nutrients), whereby N is re-translocated from senescing leaves to other growing parts of the tree, prior to leaf shedding, with resorption efficiencies of potentially up to 70% and larger at N-poor sites than at N-saturated sites (Vergutz et al., 2012; Wang et al., 2013). Biological N_2 fixation can also be significant in forests (Vitousek et al., 2002). Högberg (2012) showed for eleven European forest sites that N_r deposition was a relatively small fraction (13–14% on average) of the total N supply, which was dominated by SOM mineralization (up to 15–20 g (N) m^{-2} yr^{-1}). He further argued that there may be a correlation between soil fertility (of which the natural N supply by mineralization is an indicator) and N_r deposition, since historically human populations have tended to develop settlements in areas of favourable edaphic conditions, in which over time agriculture, industry and population intensified, leading to increased emissions and deposition. Thus, an apparent effect of ambient N_{dep} on current net ecosystem productivity (NEP) levels could also be related to the legacy of more than a century of N_r deposition on a modified internal ecosystem cycle. Importantly, unlike other ecosystem mechanisms for acquiring N from the environment (resorption from senescing leaves, biological N_2 fixation, mobilization and uptake of N from soil solution or from SOM), the nitrogen supplied from atmospheric deposition comes at little or zero energetic cost (Shi et al., 2016), especially if absorbed directly at leaf level (Nair et al., 2016).

Commenté [CF6]: Added sentence on energy cost for acquiring nitrogen from the environment

Some previous estimates of forest dC/dN response obtained by meta-analyses of NEP or NECB across a geographic gradient did not account for the major drivers of plant growth apart from nitrogen (e.g. Magnani et al., 2007). These include climate (precipitation, temperature, photosynthetically active radiation), soil physical and chemical properties (e.g. soil drainage, depth, and water holding capacity, nutrients, pH), site history and land use. Using univariate statistics such as simple regressions of NECB as a function of N_r deposition is flawed if N_r deposition is co-correlated with any of these other drivers (Fleischer et al., 2013), as can be the case in spatial gradient survey analyses across a wide geographic domain. This is because all of the variability in ecosystem C sequestration across the physical space is only allowed to be explained by one factor, N_r deposition. For example, Sutton et al. (2008) showed (using forest ecosystem modelling) that the apparently large dC/dN slope in the dataset of Magnani et al. (2007) was reduced by a factor of 2–3 when accounting for climatic differences between sites, i.e. when co-varying limitations in (photosynthetic) energy and water were factored out.

Commenté [CF7]: Inidicates that N deposition can vary with climate, but that is not necessarily the case

Similarly, ignoring the growth stage (forest age) and the effects of management (thinning) in the analysis introduces additional uncertainty in the estimated dC/dN response. Contrasting C cycling patterns and different N use efficiencies are expected between young and mature forests. Nutrient demand is highest in the early stages of forest development (especially pole stage); a recently planted forest becomes a net C sink only after a few decades, while at maturity NPP and NEP may or may not decrease, depending on a shift in the balance between autotrophic and heterotrophic respiration (R_{aut} and R_{het}, respectively) and GPP (Odum, 1969; Besnard et al., 2018). Thinning can initially increase ecosystem respiration by increasing litter and SOM stocks and reducing NPP in the short term, and some biomass can be exported (tree trunks), but the ultimate effect after a year or two is to boost forest growth as thinning indirectly increases nutrient availability at the tree level by reducing plant–plant competition. Thus, the frequency and intensity of thinning will also affect long-term or lifetime NECB. Severe storms, fire outbreaks and insect infestations may have a similar effect.

Altogether, these complex interactions mean that it is far from a simple task to untangle the N_r deposition effect on ecosystem C sequestration from the impacts of climatic, edaphic and management factors, when analysing data from diverse monitoring sites situated over a large geographic area (Laubhann et al., 2009; Solberg et al., 2009; Thomas et al., 2010). This is in contrast to fertilisation experiments, where the N effect can be quantified with all other variables being equal between manipulation plots (Nohrstedt, 2001; Saarsalmi and Mälkönen, 2001), although their results are only valid for the conditions at the specific location where the experiment has been performed (Schulte-Uebbing and de Vries, 2018).

Commenté [CF8]: References added

There are also potentially large uncertainties in the C and N flux measurements or model estimates used to calculate a dC/dN response. In the companion paper (Flechard et al., 2020), we presented – and discussed uncertainties in – plausible estimates of C and N budgets of 40 forests and natural or semi-natural ecosystems covering the main climatic zones of Europe (from Mediterranean to temperate to boreal, from oceanic to continental), investigated as part of the CarboEurope Integrated Project (CEIP, 2004–2008) and the parallel NitroEurope Integrated Project (NEU, 2006–2011). The NEP budgets were based on multi-annual eddy covariance (EC) datasets following well-established protocols, and in order to better constrain the N budgets, specific local measurements of dry and wet N_r deposition were made. Nitrogen losses by leaching and gaseous emissions were estimated by a combination of measurements and modelling. The data showed that observation-based GPP and NEP peaked at sites with N_{dep} of the order of 2–2.5 g (N) m^{-2} yr^{-1}, but decreased above that, and that increasingly large N_r losses occurred at larger N_{dep} levels, implying that the net dC/dN response was likely non-linear, in line with an overview of dC/dN response results from various approaches (De Vries et al., 2014a), possibly due to the onset of N saturation as predicted by Aber et al. (1989), and associated with enhanced acidification and increase sensitivity to drought, frost and disseases (De Vries et al., 2014b). The data also showed that at the scale of the CEIP-NEU flux tower networks, nitrogen deposition was not independent of climate, but peaked in mid-range for both mean annual temperature and precipitation, which geographically corresponds to mid-latitude Central-Western Europe, where climate is most conducive to forest productivity and growth.

In the present paper, we further the analysis of the same CEIP-NEU observational datasets through forest ecosystem modelling, with the objective of isolating the N_r deposition impact on forest productivity and C sequestration potential from the parallel effects of climate, soil water retention, and forest age and management. A mechanistic modelling framework, driven by environmental forcings, inputs, growth limitations, internal cycling and losses, was required to untangle the relationships in measurement data, because the observed dependence of N_r deposition on climate, combined with the large diversity but limited number of flux observation sites, restricted the applicability and validity of multivariate statistical methods. We describe a methodology to derive, through meta-modelling, standardization factors for observation-based forest productivity metrics, in order to factor out the part of variance that was caused by influences other than N_r deposition (climate, soil, stand age). Further, we examine patterns of C and N use efficiencies both at the decadal time scale of flux towers and over the lifetime of forests.

2 Materials and methods

2.1 Carbon and nitrogen datasets from flux tower sites

Ecosystem-scale carbon fluxes and atmospheric nitrogen deposition data were estimated within the CEIP and NEU networks at 31 European forests (six deciduous broadleaf forests, DBF; 18 coniferous evergreen needleleaf forests, ENF, of which seven spruce-dominated and eleven pine-dominated; two mixed needleleaf/broadleaf forests, MF; five Mediterranean evergreen broadleaf forests, EBF), and nine short natural or semi-natural (SN) vegetation sites (wetlands, peatlands, unimproved and upland grasslands) (Table S1). In the following we often adopted the terminology «*observation-based*» rather than simply «*measured*», to reflect the fact many variables such as e.g. GPP or below-ground C pools rely on various assumptions or even empirical models for their estimation on the basis of measured data (e.g. flux partitioning procedure to derive GPP from NEE; allometric relations for tree and root C stocks; spatial representativeness of soil core sampling for SOM). For convenience in this paper, we use the following sign convention for CO_2 fluxes: GPP and R_{eco} are both positive, while NEP is positive for a net sink (a C gain from an ecosystem perspective) and negative for a net source.

Commenté [CF9]: Provided examples of what these critical assumptions can be

The general characteristics of the observation sites (coordinates, dominant vegetation, forest stand age and height, temperature and precipitation, N_{dep}, inter-annual mean C fluxes) are provided in Table S1 of the Supplement. The sites, measurement methods and data sources were described in more detail in the companion paper (Flechard et al., 2020); for additional information on vegetation, soils, C and N flux results and budgets, and their variability and uncertainties across the network, the reader is referred to that paper and the accompanying supplement. Briefly, the C datasets include multi-annual (on average, 5-year) mean estimates of NEP, GPP and R_{eco} (total ecosystem respiration) based on 10–20 Hz EC measurements, post-processing, spectral and other corrections, flux partitioning and empirical gap-filling (e.g. Lee et al., 2004; Aubinet et al., 2000; Falge et al., 2001; Reichstein et al., 2005; Lasslop et al., 2010). The fully analysed, validated, gap-filled and partitioned inter-annual mean CO_2 fluxes (NEP, GPP, R_{eco}), as well as the meteorological data used as ecosystem model inputs (Sect. 2.2), were retrieved from the European Fluxes Database Cluster (2012) and the NEU (2013) database. Dry deposition of reactive nitrogen was estimated by measuring at each site ambient concentrations of the dominant gas-phase (NH_3, HNO_3, NO_2) and aerosol phase (NH_4^+, NO_3^-) N_r concentrations (data available from the NitroEurope database; NEU, 2013), and applying four different inferential models to the concentration and micro-meteorological data, as described in Flechard et al. (2011). Wet deposition was measured using bulk precipitation samplers (NEU, 2013, with additional data retrieved from national monitoring networks and from the EMEP chemical transport model (Simpson et al., 2012).

2.2 Modelling of forest carbon and nitrogen fluxes and pools

2.2.1 General description of the BASFOR ecosystem model

Commenté [c10]: Added detailed description of BASFOR model and implementation

The BASic FORest (BASFOR) model is a process-based, deterministic forest ecosystem model, which simulates the growth and biogeochemistry (C, N and water cycles) of temperate deciduous and coniferous stands at a daily time step (van Oijen et al., 2005; Cameron et al., 2013, 2018). Model code and documentation are available on GitHub (BASFOR, 2016). Interactions with the atmospheric and soil environments are simulated in some detail, including the role of management (thinning or pruning). BASFOR is a one-dimensional model, i.e. no horizontal heterogeneity of the forest is captured, and BASFOR does not simulate some variables which are important in forest production, such as wood quality or pests and diseases.

Nine state variables for the trees describe i) C pools: leaves, branches, stems, roots, reserves (CL, CB, CS, or collectively CLBS, CR, CRES; kg (C) m^{-2}); ii) N pool in leaves (NL; kg (N) m^{-2}); and iii) Stand density (SD, trees m^{-2}), tree phenology (only for deciduous trees): accumulated chill days (chillday; d) and accumulated thermal time (Tsum; °C d). Seven state variables for the soil can be divided into three categories, according to the three biogeochemical cycles being simulated: i) C pools in litter layers of the forest floor (CLITT), soil organic matter (SOM) with fast turn-over (CSOMF), SOM with slow turn-over (CSOMS) (kg (C) m^{-2}); ii) N pools as for C but also including mineral N (NLITT, NSOMF, NSOMS, NMIN; kg (N) m^{-2}); and iii) the water pool: amount of water to the depth of soil explored by the roots (WA; kg H_2O m^{-2} = mm) (see Table 1).

Carbon enters the system via photosynthesis, calculated as the product of photosynthetically active radiation (PAR) absorption by the plant canopy and light use efficiency (LUE). The leaf and branch pools are subject to senescence, causing carbon flows to litter. Roots are also subject to senescence, causing a flow to fast-decomposing soil organic matter. Litter carbon decomposes to fast-decomposing soil organic matter plus respiration. Fast-decomposing soil organic matter decomposes to slow-decomposing soil organic matter plus respiration. Finally, the slow organic carbon pool decomposes very slowly to CO_2. Nitrogen enters the system in mineral form through atmospheric deposition. Nitrogen leaves the system through leaching and through emission of N_2O and NO from the soil to the atmosphere. N_2 losses from denitrification and biological N_2 fixation are not simulated. Dissolved inorganic nitrogen (DIN) is taken up by the trees from the soil, and nitrogen returns to the soil with senescence of leaves, branches and roots, and also when trees are pruned or thinned. Part of the N from senescing leaves is re-used for growth. The availability of mineral nitrogen is a Michaelis-Menten function of the mineral nitrogen pool and is proportional to root biomass. The model does not include a dissolved organic nitrogen (DON) pool and therefore does not account for the possible uptake of bio-available DON forms (e.g. amino acids, peptides) by trees. Transformation between the four soil nitrogen pools are similar to those of the carbon pools, with mineral nitrogen as the loss term. Water is added to the soil by precipitation and lost through transpiration, evaporation, and drainage. Evaporation and transpiration are calculated using the Penman equation, as functions of the radiation intercepted by soil and vegetation layer, and atmospheric temperature, humidity and wind speed. Drainage of ground water results from water infiltration exceeding field capacity of the soil.

Commenté [CF11]: Add mention that DON is not considered explicitly for N supply and uptake in the model

In BASFOR, the C and N cycles are coupled in both trees and soil. The model assumes that new growth of any organ proceeds with a prescribed N/C ratio, which is species-specific but generally higher for leaves and roots than for stems and branches. If the nitrogen demand for growth cannot be met by supply from the soil, some of the foliar nitrogen is recycled until leaves approach a minimum N/C ratio when leaf senescence will be accelerated. The calculation of foliar senescence accounts for a vertical profile of nitrogen content, such that the lowest leaves have the lowest N-C ratio and senesce first. Nitrogen deficiency, as measured by foliar nitrogen content, not only increases leaf senescence, but also decreases GPP and shifts allocation from leaves to roots. Given that foliar N content is variable in BASFOR, the litter that is produced from leaf fall also has a variable N/C ratio. When the litter decomposes and is transformed, the N/C ratio of the new soil organic matter will therefore vary too in response to the ratio in the litter. Except for woody plant parts, the C and N couplings in BASFOR

Commenté [CF12]: Additional specific paragraph on the key assumptions in the model regarding the coupling of C and N in trees and soil.

vegetation and soil are based on the same generic ecophysiological assumptions as those explained in detail for grassland model BASGRA (Höglind et al. 2020).

The major inputs to the model are daily time series of weather variables (global radiation, air temperature, precipitation, wind speed and relative humidity). The last two of these are used in the calculation of potential rates of evaporation and transpiration. Soil properties, such as parameters of water retention (field capacity, wilting point, soil depth) are provided as constants. Further, the model requires time series indicating at which days the stand was thinned or pruned. The model outputs include, amongst others, the state variable for trees and soil as well as evapotranspiration (ET), groundwater recharge, canopy height (H), leaf area index (LAI), diameter at breast height (DBH), GPP, R_{eco} and R_{soil}, NEP, N mineralisation, N leaching, NO and N_2O emissions (Table 1).

{Insert Table 1 here}

2.2.2 Model implementation and calibration

BASFOR simulations of forest growth and C, N and H_2O fluxes were made for all CEIP-NEU forest sites from planting (spanning the interval 1860-2002), until the end of the NEU project (2011). At a few sites, natural regeneration occurred, but for modelling purposes a planting date was assigned based on the age of the trees. Meteorological data measured at each site over several years since the establishment of the flux towers (typically 5-10 yr) were replicated backwards in time in order to generate a time series of model inputs for the whole period since planting. Assumptions were made that inter-annual meteorological variability was sufficiently covered in the span of available measurements and that the impact of climate change since planting was small and could be neglected.

The atmospheric CO_2 mixing ratio was provided as an exponential function of calendar year, fitted to Mauna Loa data since the beginning of records in 1958 (NOAA, 2014) and extrapolated backwards to around 1860-1900 for the oldest forests included in this study. The global CO_2 mixing ratio driving the model thus increased from around 290 ppm in 1900, to 315 ppm in 1958, to 390 ppm in 2010 (Fig. 1). Similarly, atmospheric N_r deposition was a key input to the model and was forced to vary over the lifetimes of the planted forests; N_{dep} was assumed to rise from pan-European levels well below 0.5 g (N) m^{-2} yr^{-1} at the turn of the 20th century, to increase sharply after World War II to reach an all-time peak around 1980, and to decrease subsequently from peak values by about one third until 2005-2010, at which point the NEU N_{dep} estimates were obtained. We assumed that all sites of the European network followed the same relative time course of N_{dep} over the course of the 20th century, taken from van Oijen et al. (2008), but scaled for each site using the NEU N_{dep} estimates (Supplement Fig. S1).

Forest management was included as an input to the model in the form of a prescribed time course of stand density and thinning from planting to the present date. Tree density was known at all sites around the time of the CEIP-NEU projects (Table S2 in Flechard et al., 2020), but information on thinning history since planting (dates and fractions removed) was much sparser. A record of the last thinning event was available at only one third of all sites, and a knowledge of the initial (planting) density and a reasonably complete record of all thinning events were available at only a few sites. For the purposes of BASFOR modelling, we attempted to recreate a plausible density and thinning history over the lifetime of the stands. The guiding principle was that after the age of 20 years one could expect a decadal thinning of the order of 20%, following Cameron et al. (2013), while the initial reduction was 40% during the first 20 years. In the absence of an actual record of planting density (observed range: 1400-15000 trees ha^{-1}), a default initial value of 4500 trees ha^{-1} was assumed (for around two thirds of the sites). The general principles of this default scheme were then applied to fit the available density and thinning data for each site, preserving all actual data in the time series while filling in the gaps by plausible interpolation. The density time courses thus obtained, underlying all subsequent model runs, are shown in Fig. S2.

The model was calibrated through a multiple site Bayesian calibration (BC) procedure, applied to three groups of plant functional types (PFT), based on C/N/H_2O flux and pool data from the CEIP-NEU databases (see Cameron et al., 2018). A total of 22 sites were calibrated, including deciduous broadleaf forests (DB1-6), evergreen needleleaf forests ENF-spruce

(EN1-7), and ENF-pine (EN8-18). The model parameters were calibrated generically within each PFT group, i.e. they were not optimized or adjusted individually for each observation site. In the companion paper (Flechard et al., 2020), baseline BASFOR runs were produced for all 31 forest sites of the network, including also those stands for which the model was not calibrated, such as Mediterranean evergreen broadleaf (EB1 through EB5) and mixed deciduous/coniferous (MF1, MF2), to test the predictive capacity of the model beyond its calibration range (see Fig. 6 in Flechard et al., 2020). However, for the analyses and scenarios presented hereafter, these seven uncalibrated sites were removed from the dataset, as were two additional sites: EN9 and EN12 (EN9 because this agrosilvopastoral ecosystem called «dehesa» has a very low tree density (70 trees ha^{-1}; Tables S1-S2 in the Supplement to Flechard et al., 2020) and is otherwise essentially dry grassland for much of the surface area, which BASFOR cannot simulate; EN12 because this was a very young plantation at the time of the measurements, also with a very large fraction of measured NEP from non-woody biomass). All the conclusions from BASFOR meta-modelling are drawn from the remaining 22 deciduous, pine and spruce stands (sites highlighted in Table S1).

2.2.3 Modelling time frames

In the companion paper (Flechard et al., 2020), C and N budgets were estimated primarily on the basis of ecosystem measurements and for the time horizon of the CEIP and NEU projects (2004–2010). In this paper, BASFOR simulations of the C and N budgets for the 22 forest sites were considered both i) over the most recent 5-year period (around the time of CEIP-NEU) which did not include any thinning event and started at least 3 years after the last thinning event (referred to hereafter as «5-yr»); and ii) over the whole time span since forest establishment, referred to here as «lifetime», which ranged from 30 to 190 years across the network and reflected the age of the stand at the time of the CEIP-NEU projects. Note that the term «lifetime» in this context was not used to represent the expected age of senescence or harvest.

On the one hand, the short term (5-yr) simulations were made to evaluate cases where no disturbance by management impacted fluxes and pools over a recent period, whatever the age of the stands at the time of the C and N flux measurements (ca 2000–2010). On the other hand, the lifetime simulations represent the time-integrated flux and pool history since planting, which reflects the long-term C sequestration (NECB) potential, controlled by the cumulative impact of management (thinning), increasing atmospheric CO_2 mixing ratio, and changing N_r deposition over the last few decades. Thinning modifies the canopy structure and therefore light, water and nutrient availability for the trees, reduces the LAI momentarily, and in theory the left-over additional organic residues (branches and leaves) could increase heterotrophic respiration and affect the NEP. However, the impact of the disturbance on NEP and R_{eco} is expected to be small and short-lived (Granier et al., 2008), and a 3-year wait after the last thinning event appears to be reasonable for the modelling. The 5-yr data should in theory reflect the C/N flux observations, although there were a few recorded thinning events during the CEIP-NEU measurement period, and the thinning sequences used as inputs to the model were reconstructed and thus not necessarily accurate (Fig S2).

2.2.4 Modelled carbon sequestration efficiency (CSE) and nitrogen uptake efficiency (NUPE)

Commenté [c13]: Changed NUE to NUPE everywhere in the paper, text and figures

For both C and N, we define modelled indicators of ecosystem retention efficiency relative to a potential input level, which corresponds to the total C or N supply, calculated over both 5-yr (no thinning) and lifetime horizons to contrast short-term and long-term patterns. For C sequestration, the relevant terms are the temporal changes in carbon stocks in leaves, branches and stems (CLBS), roots (CR), soil organic matter (CSOM), and litter layers (CLITT), and the C export of woody biomass (CEXP), relative to the available incoming C from gross photosynthesis (GPP). We thus define the carbon sequestration efficiency (CSE) as the ratio of either modelled 5-yr NEP, or modelled lifetime NECB, to modelled GPP in a given environment, constrained by climate, nitrogen availability and other factors included in the BASFOR model:

$$CSE_{5-yr}\,(no\ thinning) = {}^{NEP_{5-yr}}\!\big/_{GPP_{5-yr}} \tag{1}$$

$$CSE_{lifetime} = {}^{NECB_{lifetime}}\!\big/_{GPP_{lifetime}} \tag{2}$$

with $$NECB = \frac{d(CLBS+CR+CSOM+CLITT)}{dt} \qquad (3)$$

$$NECB_{5-yr}\,(no\ thinning) = NEP_{5-yr} \qquad (4)$$

$$NECB_{lifetime} = NEP_{lifetime} - CEXP_{thinning} \qquad (5)$$

The modelled $CSE_{5\text{-}yr}$ can be contrasted with observation based CSE_{obs} (= NEP_{obs} / GPP_{obs}) derived from flux tower data over a similar, relatively short time period compared with a forest rotation (see Flechard et al., 2020). By extension, the $CSE_{lifetime}$ indicator quantifies the efficiency of C sequestration processes by a managed forest system, reflecting not only biological and ecophysiological mechanisms, but also the long term impact of human management through thinning frequency and severity. For the N budget we define, by analogy to CSE, the N uptake efficiency (NUPE) as the ratio of N immobilized in the forest system to the available mineral N, i.e. the ratio of tree N uptake (N_{upt}) to the total N_{supply} from internal SOM mineralization and N cycling processes (N_{miner}) and from external sources such as atmospheric N deposition (N_{dep}):

Commenté [CF14]: Changed NUE to NUPE

$$NUPE = {N_{upt}}/{N_{supply}} \qquad (6)$$

with $$N_{supply} = N_{miner} + N_{dep} \qquad (7)$$

$$N_{supply} \approx N_{upt} + N_{leach} + N_{emission} \qquad (8)$$

The fraction of N_{supply} not taken up in biomass and lost to the environment (N_{loss}) comprises dissolved inorganic N leaching (N_{leach}) and gaseous NO and N_2O emissions ($N_{emission}$):

$$N_{loss} = {(N_{leach} + N_{emission})}/{N_{supply}} \qquad (9)$$

Note that i) NUPE is a different concept from the nitrogen use efficiency (NUE), often defined as the amount of biomass produced per unit of N taken up from the soil, or the ratio NPP/N_{upt} (e.g. Finzi et al., 2007), and ii) biological N_2 fixation, as well as N loss by total denitrification, are not accounted for in the current BASFOR version; also, leaching of dissolved organic N and C (DON, DOC) and dissolved inorganic C (DIC) is not included either, all of which potentially impact budget calculations.

2.2.5 Meta-modelling as a tool to standardize EC-based productivity data

One purpose of BASFOR modelling in this study was to gain knowledge on patterns of C and N fluxes, pools and internal cycling that were not, or could not be, evaluated solely on the basis of the available measurements (for example, SOM mineralization and soil N transfer; retranslocation processes at canopy level; patterns over the lifetime of a stand). The model results were used to complement the flux tower observations to better constrain elemental budgets and assess potential and limitations of C sequestration at the European forest sites considered here. Additionally, we used meta-modelling as an alternative to multivariate statistics (e.g. stepwise multiple regression, mixed non-linear models, residual analysis) to isolate the importance of N_r deposition from other drivers of productivity. This follows from the observations by Flechard et al. (2020) that i) N_r deposition and climate were not independent in the dataset, and that ii) due to the large diversity of sites, the limited size of the dataset, and incomplete information on other important drivers (e.g. stand age, soil type, management), regression analyses were unable to untangle these climatic and other inter-relationships from the influence of N_r deposition. BASFOR (or any other mechanistic model) is useful in this context, not so much to predict absolute fluxes and stocks, but to investigate the relative importance of drivers, which is done by assessing changes in simulated quantities when model inputs are modified. Meta-modeling involves building and using surrogate models that can approximate results from more complicated simulation models; in this case we derived simplified relationships linking forest productivity to the impact of major drivers, which were then used to harmonize observations from different sites. For example, running BASFOR for a given site using meteorological input data from another site, or indeed from all other sites of the network, provides insight into the impact of climate on GPP or NEP, all other factors (soil, vegetation structure and age, N_r deposition) being equal. Within the boundaries of the network of 22 selected sites, this sensitivity analysis provides *relative* information as to which of the 22 meteorological datasets is most, or least, favourable to growth for this particular site. This can be repeated for all sites

(22*22 climate «scenario» simulations). It can also be done for soil physical properties that affect the soil water holding capacity (texture, porosity, rooting depth), in which case the result is a *relative* ranking within the network of the different soils for their capacity to sustain an adequate water supply for tree growth. The procedure for the normalization of data between sites is described hereafter.

Additional nitrogen affects C uptake primarily through releasing N limitations at the leaf level for photosynthesis (Wortman et al., 2012; Fleischer et al., 2013), which scales up to GPP at the ecosystem level. Other major factors affecting carbon uptake are related to climate (photosynthetically active radiation, temperature, precipitation), soil (for example water holding capacity) or growth stage (tree age). In the following section, we postulate that observation-based gross primary productivity (GPP_{obs}), which represents an actuation of all limitations in the real world, can be transformed through meta-modelling into a standardized potential value (GPP*) for a given set of environmental conditions (climate, soil, age), common to all sites, thereby enabling comparisons between sites. We define GPP* as GPP_{obs} being modulated by one or several dimensionless factors (f_X):

$$GPP^{*} = GPP_{obs} \times f_{CLIM} \times f_{SOIL} \times f_{AGE} \qquad (10)$$

where the standardization factors f_{CLIM}, f_{SOIL} and f_{AGE} are derived from BASFOR model simulations corresponding to the CEIP-NEU time interval around 2005–2010, as described below. The factors involved in Eq. (10) address commonly considered drivers, but not nitrogen, which is later assessed on the basis of GPP*, rather than GPP_{obs}. Other potentially important limitations such as non-N nutrients, soil fertility, air pollution (O_3), poor ecosystem health, soil acidification, etc., are not treated in BASFOR, and cannot be quantified here. Further, the broad patterns of the GPP *vs.* N_{dep} relationships reported in Flechard et al. (2020), i.e. a non-linear increase and eventual saturation of GPP as N_{dep} increases beyond a critical threshold, did not show any marked difference between the three forest PFT (deciduous, pine, spruce), possibly because the datasets were not large enough and fairly heterogeneous. Thus, although PFT may be expected to influence C/N interactions, we did not seek to standardize GPP with an additional f_{PFT} factor.

To determine the f_{CLIM} and f_{SOIL} factors, the model was run multiple times with all climate and soil scenarios for the *n* (=22) sites, a scenario being defined as using model input data or parameters from another site. Specifically, for f_{CLIM}, the model weather inputs at each site were substituted in turn by the climate data (daily air temperature, global radiation, rainfall, wind speed and relative humidity) from all other sites; and for f_{SOIL}, the field capacity and wilting point parameters (Φ_{FC}, Φ_{WP}) and soil depth that determine the soil water holding capacity at each site (SWHC = (Φ_{FC} - Φ_{WP}) x soil depth), were substituted in turn by parameters from all other sites. Values of f_{CLIM} and f_{SOIL} were calculated for each site in several steps, starting with the calculation of the ratios of modelled GPP from the scenarios to the baseline value GPP_{base} such that:

$$X(i,j) = GPP(i,j)/GPP_{base}(i) \qquad (11)$$

where i (1..*n*) denotes the site being modelled and j (1..*n*) denotes the climate data set (j_{CLIM}) or soil parameter set (j_{SOIL}) used in the scenario being simulated (see Table S2 for the calculation matrices). The value of the X (i, j) ratio indicates whether the j^{th} scenario is more (> 1) or less (< 1) favourable to GPP for the i^{th} forest site.

For each site, the aim of the f_{CLIM} factor (and similar reasoning for f_{SOIL}) (Eq. (10)) is to quantify the extent to which GPP differs from a standard GPP* value that would occur if all sites were placed under the same climatic conditions. Rather than choose the climate of one particular site to normalize to, which could bias the analysis, we normalise GPP to the equivalent of a «mean» climate, by averaging BASFOR results over all (22) climate scenarios (Eq. (14)–(15)). However, since each of the scenarios has a different mean impact across all sites ($\overline{X(j)}$, Eq. (12)), we first normalize X(i,j) to $\overline{X(j)}$ value within each j^{th} scenario (Eq. (13)):

$$\overline{X(j)} = {}^{1}/_{n} \sum_{i=1}^{n} X(i,j) \qquad (12)$$

$$X_{norm}(i,j) = {}^{X(i,j)}\Big/_{\overline{X(j)}} \qquad (13)$$

The normalization of X (i, j) to X_{norm} (i, j) ensures that the relative impacts of each scenario on all *n* sites can be compared between scenarios. The final step is the averaging for each site of X_{norm} (i, j) values from all scenarios (either j_{CLIM} or j_{SOIL}) into the overall f_{CLIM} or f_{SOIL} values:

$$f_{CLIM}(i) = \overline{X_{norm}(i)} = {}^{1}\!/_{n} \sum_{j_{CLIM}=1}^{n} X_{norm}(i, j_{CLIM}) \tag{14}$$

or

$$f_{SOIL}(i) = \overline{X_{norm}(i)} = {}^{1}\!/_{n} \sum_{j_{SOIL}=1}^{n} X_{norm}(i, j_{SOIL}) \tag{15}$$

The factors f_{AGE} were determined by first normalizing modelled GPP (base run) to the value predicted at age 80, for every year of the simulated GPP time series at those *m* (=12) mature sites where stand age exceeded 80. The age of 80 was chosen since this was the mean stand age of the whole network. The following ratios were thus calculated:

$$Y(k, yr) = GPP_{base}(k, yr) / GPP_{base}(k, 80) \tag{16}$$

where k (1..*m*) denotes the mature forest site being modelled. A mean temporal curve for f_{AGE} (normalized to 80 years) was calculated, to be used subsequently for all sites, after the following:

$$f_{AGE}(yr) = \left({}^{1}\!/_{m} \sum_{k=1}^{m} Y(k, yr)\right)^{-1} \tag{17}$$

3 Results

3.1 Short term (5-yr) versus lifetime C and N budgets from ecosystem modelling

The time course of modelled (baseline) GPP, NEP and total leaching and gaseous N losses is shown in Fig. 1 for all forest sites over the 20th century and until 2010, forced by climate, increasing atmospheric CO_2 and by the assumed time course of N_r deposition over this period (Fig. 1a). For each stand, regardless of its age and establishment date, an initial phase of around 20-25 years occurs, during which GPP increases sharply from zero to a potential value attained upon canopy closure (Fig. 1b), while NEP switches from a net C source to a net C sink after about 10 years (Fig. 1d). Initially N_r losses are very large (typically of the order of 10 g (N) m^{-2} yr^{-1}), then decrease rapidly to pseudo steady-state levels when GPP and tree N uptake reach their potential.

Commenté [CF15]: Added these 2 paragraphs to decribe the new Figure 1, to introduce the budget calculations made in the following figures

After this initial phase, modelled GPP increases steadily in response to increasing N_{dep} and atmospheric CO_2, but only for the older stands established before around 1960, i.e. those stands that reach canopy closure well before the 1980's, when N_r deposition is assumed to start declining. Thereafter, modelled GPP ceases to increase, except for the recently established stands that have not yet reached canopy closure. The stabilization of GPP for mature trees at the end of the 20th century in the model is likely a consequence of the effects of decreasing N_{dep} and increasing CO_2 cancelling each other out to a large extent. In parallel, modelled total N losses start to decrease after the 1980's, even for sites long past canopy closure (Fig. 1e-f), but this mostly applies to stands subject to the largest N_{dep} levels, i.e. where the historical high N_{dep} of the 1980's, added to the internal N supply, were well in excess of growth requirements in the model.

{Insert Fig. 1 here}

These temporal interactions of differently-aged stands with changing N_{dep} and CO_2 over their lifetimes therefore impact C and N budget simulations made over different time horizons. Modelled C and N budgets are represented schematically in Fig. 2 and Fig. 3, respectively, as «Sankey» diagrams (Matlab «*drawSankey.m*» function; Spelling, 2009) for three example forest sites (DB5, EN3, EN16), and in Fig. S3–S8 of the Supplement for all sites of the study. Each diagram represents the input, output and internal flows in the ecosystem, with arrow width within each diagram being proportional to flow. For carbon (Fig. 2 and S3–S5), the largest (horizontal) arrows indicate exchange fluxes with the atmosphere (GPP, R_{eco}), while the smaller (vertical) arrows indicate gains (green) or losses (red) in internal ecosystem C pools (CSOM, CBS, CR, CL, CLITT), as well as any exported wood products (CEXP, orange). NEP is the balance of the two horizontal arrows, as well as the balance of all vertical arrows.

In the 5-yr simulations with no thinning occurring (Fig. 2-left; Fig. S3), NEP is equal to NECB, which is the sum of ecosystem C pool changes over time (= C sequestration if positive). By contrast, in the lifetime (since planting) simulations

(Fig. 2-center; Fig. S4), the long-term impact of thinning is shown by the additional orange lateral arrow for C exported as woody biomass (CEXP). In this case, C sequestration or NECB no longer equals NEP, the difference being CEXP, the C contained in exported stems from thinned trees. By contrast, in the model, upon thinning the C from leaves, branches and roots join the litter layers or soil pools and is ultimately respired or sequestered. To compare between sites with different productivity levels, the lifetime data are also normalized as a percentage of GPP (Fig. 2-right; Fig. S5). The clear differences between 5-yr and lifetime C-budget simulations were: i) systematically larger GPP in recent 5-yr horizon (combined effects of age as well as CO_2 and N_{dep} changes over time); ii) C storage in branches and stems (CBS) dominated in both cases, but CBS fractions were larger in the 5-yr horizon; iii) larger relative storage in soil organic matter (CSOM) when calculated over lifetime.

For nitrogen, by contrast to carbon, the focus of the budget diagrams is not on changes over time of the total ecosystem (tree + soil, organic + mineral) N pools. Rather, we examine in Fig. 3 and S6–S8 the extent to which N_r deposition contributes to the mineral N pool (NMIN), which in the model is considered to be the only source of N available to the trees and therefore acts as a control of C assimilation and ultimately sequestration. In these diagrams for NMIN, the largest (horizontal) arrows indicate the modelled internal ecosystem N cycling terms (N_{miner} from SOM mineralisation, N_{upt} uptake by trees) and the secondary (vertical) arrows represent external exchange (inputs and losses) fluxes as N_{dep}, N_{leach} and $N_{emission}$ (unit: g (N) m^{-2} yr^{-1}). The variable NMIN describes the transient soil inorganic N pool in the soil solution and adsorbed on the soil matrix (NMIN = NO_3^-+ NH_4^+; units g (N) m^{-2}). Since the modelled long term (multi-annual) changes in the transient NMIN pool are negligible compared with the magnitudes of the N input and output fluxes, the dNMIN/dt term is not represented as an arrow in the budget plots, and the total mineral N_{supply} (defined as N_{miner} + N_{dep}) is basically balanced by N uptake (N_{upt}) and losses (N_{leach} + $N_{emission}$) (Eq. (8)). Modelled N budgets were calculated for a 5-yr time horizon (Fig. 3-left; Fig. S6) and for the whole time period since the forest was established (lifetime, Fig. 3-center; Fig. S7). Lifetime data were also normalized as a percentage of N_{supply} (Fig. 3-right; Fig. S8). The clear differences between 5-yr and lifetime N-budget simulations are: i) N_{loss} and especially N_{leach} were significantly larger over the stand lifetime since planting; ii) N_{upt} was a larger fraction of total N_{supply} over the recent 5-yr period.

Commenté [CF16]: Made it clear that we're dealing with mineral nitrogen budgets, not with long term ecosystem total N retention

Commenté [CF17]: Clarified meaning of negligible long term change in NMIN

{Insert Fig. 2 here}

{Insert Fig. 3 here}

3.2 Contrasted efficiencies of carbon sequestration and nitrogen uptake

Commenté [CF18]: Changed title

Collectively, the changes in the ecosystem C pools, especially the increases in stems and branches (CBS), roots (CR) and soil organic matter (CSOM) represent roughly 20–30% of GPP for both 5-yr and lifetime simulations (Fig. 2, S3–S5). By contrast, the analogous term for nitrogen, the N_{upt} fraction of total N_{supply}, is a much more variable term, both between sites of the network and between the 5-yr and lifetime simulations (Fig. 3, S6–S8). Modelled lifetime CSE and NUPE values are compared in Fig. 4 with the 5-yr values, as a function of stand age, indicating that (i) the older forests of the network (age range ~80–190 yrs) tend to have larger NUPE than younger or middle aged forests (~30–60 yrs), but (ii) the difference in NUPE between the two age groups is much clearer if NUPE is calculated over the whole period since planting (lifetime). As shown in Fig. 1, BASFOR predicts large N losses in young stands (<20-25 years), in which lower N demand by a smaller living biomass, combined in the early years with enhanced N_{miner} from higher soil temperature (canopy not yet closed) and with a larger drainage rate (smaller canopy interception of incident rainfall), all lead to larger NMIN losses. The 22 forests sites of this study were past this juvenile stage, but observation (ii) is a mathematical consequence of high N losses during the forest's early years having a larger impact on lifetime calculations in middle-aged than mature forests. NUPE tends to reach 70-80% on average after 100 years and is smaller calculated from lifetime than from a 5-yr thinning-free period. For forests younger than 60 years, lifetime NUPE is only around 60%.

Commenté [CF19]: NUE changed to NUPE throughout

Revised description of results for NUPE, especially for the 30-60 yr age class

Modelled carbon sequestration efficiency is less affected than NUPE by forest age (CSE range ~15–30%) (Fig. 4). There is a tendency for 5-yr (thinning-free) CSE to decrease from ~30% to ~20% between the ages of 30 and 190 years. This means that, in the model, R_{eco} in 30 to 60-yr old stands represents a smaller fraction of GPP than in mature stands. From Eq. (1) it can readily be shown that CSE = 1 - R_{aut}/GPP - R_{het}/GPP, which is roughly equivalent to 0.5 – R_{het}/GPP, since in the model R_{aut} is constant and approximately 0.5 for all species. By contrast, BASFOR predicts that the R_{het}/GPP ratio increases steadily with age at each site, after the initial establishment phase (Fig. S12a). This induces a decline in modelled CSE from 25-35% in the age class 30-60 yrs down to around 20-25% for the older forests (Fig. S12b). This also implies a non-linearity developing over time of GPP versus soil and litter layers C pools, since R_{het} is assumed to a linear function of fast and slow C pools in litter layers and SOM. Lifetime CSE values are slightly smaller than 5-yr values: the difference corresponds to cumulative CEXP over time, but the trend with age is weaker than for 5-yr CSE. The relatively narrow range of modelled 5-yr CSE values (20–30%) is in sharp contrast to the much wider range of observation-based CSE_{obs} values (from -9% to 61%), likely reflecting some limitations of the model and possibly also measurement uncertainties, as discussed in Flechard et al. (2020).

Commenté [CF20]: Clarify non linearity issue using additional Supplement Fig S12

{Insert Fig. 4 here}

Beyond the overall capacity of the forest to retain assimilated C (as quantified by CSE), the modelled fate of sequestered C, the simulated ultimate destination of the C sink, is also a function of forest age and of the time horizon considered (Fig. 5). The fraction of NECB sequestered in above-ground biomass (CLBS) over a recent 5-yr horizon is on average around 80% (*versus* around 10% each for CR and CSOM) and not clearly linked to forest age, i.e. the model does not simulate any slowing down with age of the annual growth of above-ground biomass. Calculated over lifetimes, the dominant ultimate destination of sequestered C remains CLBS. However, this fraction is smaller (50–60%) in old-growth forests than in younger stands (60–80%), since a larger cumulative fraction of above-ground biomass (timber) will have been removed (CEXP) by a lifetime of thinnings in a mature forest, while the cumulative gain in CSOM is not repeatedly depleted, but on the contrary enhanced, by thinnings (since the model assumes bole removal only, not total tree harvest). Modelled annual C storage to the rooting system clearly declines with age and is an increasingly marginal term over time (although the absolute CR stock itself keeps increasing over time, except when thinning transfers C from roots to SOM).

{Insert Fig. 5 here}

3.3 Standardization of observation-based GPP through meta-modelling

The purpose of meta-modelling was to standardize observation-based GPP_{obs} into GPP* through model-derived factors that separate out the effects of climate, soil and age between monitoring sites (Eq. (10)), so that the importance of N_r can be isolated. The sensitivity of modelled GPP to climate and soil physical properties was tested through various model input and parameter scenarios, allowing standardization factors f_{CLIM} and f_{SOIL} to be calculated as described in Methods (Eq. (11)–(15)) and Table S2 in the Supplement. The resulting distributions of all simulations for all sites were represented in Fig. 6 as «violin» plots (Matlab «*distributionPlot*.m» function; Dorn, 2008) for the climate-only and soil-only scenarios (n^2 = 484 simulations each), and also combined climate*soil scenarios (n^3 = 10648 simulations). For each site, the scenarios explore the modelled response of ecosystem C dynamics to a range of climate and soil forcings different from their own. The size and position of the violin distribution indicate, respectively, the degrees of sensitivity to- and limitation by- climate, soil, or both; a site is especially limited by either factor (relative to the other sites of the network) when the baseline/default run (GPP_{base}) is located in the lower part of the distribution.

Similarly, to account for the effect of tree age, the f_{AGE} factor was calculated following Eq. (17), whereby the time series for the ratio of modelled GPP_{base}(yr) to GPP_{base}(80) (Eq. (16)) followed broadly similar patterns for the different sites (Fig. 7), with values mostly in the range 0.6–0.8 at around age 40, crossing unity at 80 and levelling off around 1.2–1.4 after a century. Some of the older sites (e.g. EN2, EN6, EN15) showed a peak followed by a slight decrease in modelled GPP, but not at the same age. This was due to the peak in N_{dep} in the early 1980's in Europe (Fig. S1), with the N_{dep} peak occurring at different ontogenetic stages in the differently aged stands. By calculating a mean f_{AGE} factor across sites the peak N_{dep} effect was smoothed out (Fig. 7). Thus, for a younger forest, the multiplication of GPP_{obs} by f_{AGE} (>1) simulated the larger GPP* that one could expect for the same site at 80 yr; conversely, the GPP* a mature forest (>100 yr) would be reduced compared with GPP_{obs}.

{Insert Fig. 6 here}

{Insert Fig. 7 here}

The combined modelled effects of climate, soil, and stand age on GPP are summarized in Fig. 8. Values for both f_{CLIM} and f_{SOIL} are mostly in the range 0.7–1.5, and are predictably negatively correlated to mean annual temperature (MAT) and soil water holding capacity (SWHC), respectively (Fig. 8a). A value well above 1 implies that GPP_{obs} for one site lies below the value one might have observed if climate or SWHC had been similar to the average of all other sites of the network. In other words this particular site was significantly limited by climate, SWHC, or both, relative to the other sites. Conversely, a value below 1 means that GPP at the site was particularly favoured by weather and soil. Climate or soil conditions at some sites have therefore the potential to restrict GPP by around one third, while other climates or soil conditions may enhance GPP by around one third, compared with the average conditions of the whole network. Applying the f_{CLIM}, f_{SOIL} and f_{AGE} multipliers to GPP_{obs} (Eq. (10)) provides a level playing field (GPP*) for later comparing sites with respect to N_r deposition, but also increases the scatter and noise in the relationship of GPP* to N_{dep}, particularly with the introduction of f_{AGE} (Fig. 8b).

{Insert Fig. 8 here}

3.4 Response of gross primary productivity to N_r deposition

The standardized forest GPP* values, i.e. GPP*(f_{CLIM}), GPP*(f_{CLIM} x f_{SOIL}) and GPP*(f_{CLIM} x f_{SOIL} x f_{AGE}), show in the N_{dep} range 0–1 g (N) m^{-2} yr^{-1} a much less steep relationship to N_{dep} than the original GPP_{obs} (Fig. 8b). This supports the hypothesis that GPP at the lower N_{dep} sites is also limited by climate and/or soil water availability. In Fig. 8b, 2nd-order polynomials are fitted to the data to reflect the strong non-linearity present in GPP_{obs}, driven especially by the 4 highest N_{dep} sites (>2.5 g (N) m^{-2} yr^{-1} at EN2, EN8, EN15 and EN16). The non-linearity (magnitude of the 2nd-order coefficient) is reduced by the introduction of f_{CLIM} and f_{SOIL}, while f_{AGE} has a small residual impact on the shape of the regression. Due to this non-linear behaviour, the $dGPP/dN_{dep}$ responses decrease with N_{dep} for the observation-based GPP, but less so for the standardized GPP* estimates (Fig. 8c). Values of $dGPP_{obs}/dN_{dep}$ (calculated for each N_{dep} level by the slope of the tangent line to the quadratic fits of Fig. 8b) range from around 800 g (C) g^{-1} (N) at the lowest N_{dep} level down to negative values at the highest N_{dep} sites; for the standardized GPP* accounting for all climate, soil and age effects, this range is much narrower, from around 350 down to near 0 g (C) g^{-1} (N).

Average $dGPP/dN_{dep}$ figures that are representative of this set of forest sites are given in the upper part of Table 2, either calculated over the whole range of 22 sites, or for a subset of 18 sites that excludes the four highest deposition sites (>2.5 g (N) m^{-2} yr^{-1}). If all modelled sites are considered, the mean $dGPP/dN_{dep}$ regression slopes are smaller (190–260 g (C) g^{-1} (N)), being influenced by the reductions in GPP at very high N_{dep} levels, possibly induced by the negative side effects of N saturation. If these four sites are excluded, the mean $dGPP/dN_{dep}$ is larger (234–425 g (C) g^{-1} (N)), reflecting the fact that healthier, N-limited forests are more responsive to N additions. In this subset of 18 sites, the effects of climate, soil and stand age account for approximately half of GPP (the mean $dGPP/dN_{dep}$ response changes from 425 to 234 g (C) g^{-1} (N)). For comparison, Table 2 also provides the values of $dGPP_{obs}/dN_{dep}$ obtained directly through simple linear regression for all forest sites and for the semi-natural vegetation sites, with values of the same order (432 and 504 g (C) g^{-1} (N), respectively) if the high N deposition sites (N_{dep} > 2.5 g (N) m^{-2} yr^{-1}) are removed.

As a further comparison, an additional BASFOR modelling experiment is shown in Fig. 9a, in which GPP at all sites is simulated in a range of N_{dep} scenarios (0, 0.1, 0.2, 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4 and 4.5 g (N) m^{-2} yr^{-1}, constant over lifetime) to substitute for the actual N_{dep} levels of each site. Around half the sites show a steadily increasing (modelled) GPP as N_{dep} increases over the whole range 0–4.5 g (N) m^{-2} yr^{-1}, with broadly similar slopes between sites; while the other half levels off and reaches a plateau at various N_{dep} thresholds, indicating that beyond a certain level N_{dep} is no longer limiting, according to the model. For comparison with the dC/dN responses calculated previously for GPP_{obs} and GPP* in Fig. 8b-c and Table 2, we derive a mean modelled $dGPP/dN_{dep}$ response from a linear regression of Fig. 9a data over the range 0–2.5 g (N) m^{-2} yr^{-1} (i.e. excluding the highest deposition levels). This yields a mean $dGPP/dN_{dep}$ slope across all sites of 297 (273–322) g (C) g^{-1} (N) for the N_{dep} model experiment, only marginally larger than the three GPP* average slopes of Table 2. Note that in Fig. 8b, the response of GPP* to N_{dep} is calculated *between sites* of the network, while in Fig. 9a the GPP to N_{dep} response is calculated *within each site* from the model scenarios, then averaged across all sites.

{Insert Fig. 9 here}

3.5 Response of net ecosystem productivity to N_r deposition

Similarly to GPP, the NEP and NECB responses to N_{dep} cannot be reliably inferred directly from EC-flux network data given the large variability between sites in climate, soil type, age and other constraints to photosynthesis and ecosystem respiration. However, plausible estimates can be obtained by applying a range of mean CSE indicators (as defined previously) to project the normalized GPP* responses to N_{dep} (Table 2). Carbon sequestration efficiencies for forests are confined to a narrow range (17–31% of GPP, average μ=22%, standard deviation σ=4%) in model simulations over 5-yr (no thinning) time horizons (Fig. 4); they vary considerably more in EC-based observations (range -9 to 61%, σ=17%), but with a similar mean (μ=25%). CSE metrics express the GPP fraction not being respired (R_{eco}) or exported (CEXP) out of the ecosystem. Multiplied by the $dGPP/dN_{dep}$ slope they provide estimates of the net ecosystem C gain per unit N deposited (Table 2).

Short-term (5-yr) mean estimates for NEP responses, based on average CSE from both observations (CSE_{obs}) and modelling ($CSE_{5\text{-}yr}$), and accounting for GPP climate/soil/age normalization, range from 41 to 47 g (C) g^{-1} (N), averaged over all sites, or 51 to 57 g (C) g^{-1} (N) removing the four highest N_{dep} sites (middle part of Table 2). Predictably, lifetime estimates for $dNECB/dN_{dep}$ responses are about 20% smaller, on the order of 34–42 g (C) g^{-1} (N). For comparison, the mean 5-yr $dNEP/dN_{dep}$ obtained directly by BASFOR modelling of N_{dep} scenarios for all sites (Fig. 9b) was larger (76 ± 7 g (C) g^{-1} (N)) than the measurement-based, model-corrected estimates of Table 2.

If the forest NEP response to N_{dep} is calculated directly through simple linear or quadratic regression of NEP_{obs} *vs.* N_{dep} (bottom part of Table 2), therefore not including any standardization of the data, the dC/dN slope is much larger (178–224 g (C) g^{-1} (N)) within the N_{dep} range 0–2.5 g (N) m^{-2} yr^{-1}. If all forest sites are considered (including N-saturated sites with N_{dep} up to 4.3 g (N) m^{-2} yr^{-1}), the dC/dN slope is much smaller (71–108 g (C) g^{-1} (N)), but this only reflects the reduced NEP observed at those elevated N_{dep} sites (see Fig. 4c in Flechard et al., 2020), with altogether very large scatter and very small R^2. Equivalent figures for (not standardized) semi-natural NEP *vs.* N_{dep} appear to be significantly smaller (34–89 g (C) g^{-1} (N)) than in forests.

If the meta-modelling standardization procedure for climate, soil and age is attempted (for comparison only) directly on NEP, as opposed to the preferred procedure using GPP (Eq. (10)–(17)), the simulated f_{CLIM}, f_{SOIL} and f_{AGE} reduce the NEP response to N_{dep} by only 18%, from 178 down to 146 g (C) g^{-1} (N) (bottom part of Table 2), while the equivalent reduction for GPP was 45%. The resulting figure (112–146 g (C) g^{-1} (N)) is likely much over-estimated, around factor of 2–3 larger than those obtained through the stepwise method using CSE * $dGPP/dN_{dep}$. Standardization factors derived from BASFOR meta-modelling are more reliable for GPP than for NEP, since model performance is significantly better for GPP than for R_{eco} and hence NEP (Fig. 6 in Flechard et al., 2020).

{Insert Table 2 here}

4 Discussion

4.1 A moderate non-linear response of forest productivity to N_r deposition

The C sequestration response to N_{dep} in European forests was derived using a combination of flux tower-based C and N exchange data and process-based modelling, while a number of previous studies have been based on forest inventory methods and stem growth rates (e.g. de Vries et al.2009; Etzold et al., 2014). The main differences with previous meta-analyses that were also based on EC-flux datasets (e.g. Magnani et al., 2007; Fleischer et al., 2013; Fernández-Martínez et al., 2014, 2017), were that i) we derived total N_{dep} from local measurements of the wet and dry fractions as opposed to regional/global CTM outputs; ii) we untangled the N_{dep} effect from climatic, soil and other influences by means of a mechanistic model, not through statistical methods; and iii) in Flechard et al. (2020) we estimated ecosystem-level N, C and GHG budgets calculated through a combination of local measurements, mechanistic and empirical models, and database and literature data mining.

Our most plausible estimates of the dC/dN response of net productivity over the lifetime of a forest are of the order of 40–50 g (C) g^{-1} (N) on average over the network of sites included in the study (Table 2). Such values are broadly in line with the recent reviews by Erisman et al. (2011) and by Butterbach-Bahl et al. (2011) (range 35–65 g (C) g^{-1} (N)), but slightly larger than estimates given in a number of other studies (e.g. Liu and Greaver, 2009; de Vries et al., 2009, 2014a). Given the considerable uncertainty attached to these numbers (Table 2), they cannot be considered significantly different from any of those earlier studies. The meta-modelling-based approach we describe for normalizing forest productivity data to account for differences in climate, soil and age among sites, reduces the net productivity response to N_{dep} by roughly 50%, which is of the same order as the results (factor of 2–3 reduction) of a similar climate normalization exercise by Sutton et al. (2008). This means that not accounting for inter-site differences would have led to an over-estimation of the dC/dN slope by a factor of 2.

Observations and model simulations both indicate that the N_{loss} fraction of N_{supply} increases with N_{dep}, consistent with widespread observations of increasing NO_3^- leaching above N_{dep} thresholds as low as 1.0 g (N) m^{-2} yr^{-1} in European forests (Dise and Wright, 1995; De Vries et al, 2007; Dise et al., 2009), and exacerbated by large C/N ratios (> 25) in the organic horizons (Gundersen et al., 1998; MacDonald et al., 2002). Higher thresholds for N_{dep} around 2.5 g (N) m^{-2} yr^{-1} (Dise and Wright, 1995; Van der Salm et al., 2007) typically indicate advanced saturation stages.

Commenté [CF21]: Added references to publications on NO_3^- leaching thresholds

Thus, at many sites but especially those with $N_{dep} > 1.5–2$ g (N) m^{-2} yr^{-1}, N availability is not limiting forest growth. In such cases it becomes meaningless to try to quantify a N fertilisation effect. Indeed, despite large uncertainties in measured data and in model-derived normalization factors, the non-linear trend is robust, with dC/dN values tending to zero in N-saturated forests (>2.5–3 g (N) m^{-2} yr^{-1}). In their review paper De Vries et al. (2014a) gave a range of N_{dep} levels varying between 1.5–3 g (N) m^{-2} yr^{-1} beyond which growth and C sequestration were not further increased or even reversed, as predicted in classical N saturation theory by Aber et al. (1989, 1998). These findings suggest that in areas of the world where N_{dep} levels are larger than 2.5–3 g (N) m^{-2} yr^{-1}, which now occur increasingly in Asia, specifically in parts of China, Japan, Indonesia, and India (Schwede et al., 2018), the forecast increased N_r emissions and increased N_{dep} levels may thus not have a positive impact on the continent's land based CO_2 sink. Data treatment and selection in our dataset (e.g. removal of N-saturated forests) strongly impacted the plausible range of dC/dN responses (Table 2) derived from the original data. The non-linearity of ecosystem productivity relationships to N_{dep} (Butterbach-Bahl and Gundersen, 2011; Etzold et al., 2014) limits the usefulness and significance of simple linear approaches. These data suggest that there is no single dC/dN figure applicable to all ecosystems, that the highly non-linear response depends on current and historical N_{dep} exposure levels, and on the degree of N saturation (Aber et al., 1989, 1998), although other factors than N, discussed later, may also be involved.

For the short semi-natural vegetation sites, included in the study as a non-fertilised, non-woody contrast to forests, the apparent impact of N_{dep} on GPP_{obs} was of the same order as in forests, but likely much smaller than in forests when considering NEP_{obs} (Table 2). This is in principle consistent with the hypothesis (de Vries et al., 2009) that the ecosystem dC/dN response may be larger in forests due to the large C/N ratio (200–500) of above-ground biomass (stems and branches), where much of the C storage occurs (up to 60–80% according to BASFOR, Fig. 5); whereas in semi-natural ecosystems C storage in SOM dominates, with a much lower C/N ratio (10–40). However, this comparison of semi-natural *versus* forests is based on NEP_{obs} that was not normalized for inter-site climatic and edaphic differences, since no single model was available to carry out a meta-modelling standardization for all the different semi-natural ecosystem types (peatland, moorland, fen, grassland), and therefore these values must be regarded as highly uncertain.

4.2 Limitations and uncertainties in the approach for quantifying the dC/dN response

Monitoring atmospheric gas-phase and aerosol N_r contributed to reducing the large uncertainty in total N_r deposition at individual sites, because dry deposition dominates over wet deposition in most forests (Flechard et al., 2020), except at sites a long way from sources of atmospheric pollution, and because the uncertainty in dry deposition and its modelling is much larger (Flechard et al., 2011; Simpson et al., 2014). However, despite the considerable effort involved in coordinating the continental-scale measurement network (Tang et al., 2009), the number of forest sites in this study (31) was relatively small compared with other studies based on ICP (de Vries et al., 2009; ICP, 2019) or other forest growth databases, or global-scale FLUXNET data (hundreds of sites worldwide; see Burba, 2019). Thus, the gain in precision of N_{dep} estimates from local measurements was offset by the smaller population sample size. Nonetheless this study does show the added value of the N_r concentration monitoring exercise and the need to repeat and extend such initiatives.

Understanding, quantifying and reducing all uncertainties leading up to dC/dN estimates are key issues to explore. Apart from measurement uncertainties in N_r deposition and losses, and in the C balance based on EC measurements, analysed in the companion paper, the major difficulties that arose when assessing the response to N_{dep} of forest productivity included:

i) The heterogeneity of the population of forests, climates and soils in the network, and the large number of potential drivers relative to the limited number of sites, hindered the use of a straightforward, regression-based analysis of observational data without a preliminary (model-based) harmonization;

ii) The model-based normalization procedure for GPP, used to factor out differences in climate, soil and age among sites, significantly amplified the noise in C/N relationships, an indication that the generalized modelled effects may not apply to all individual sites and that other important ecological determinants affecting forest productivity are missing in the BASFOR model;

iii) The EC measurement-based ratio of R_{eco} to GPP (=1-CSE) was very variable among forests (Flechard et al., 2020) and this high variability cannot be explained or simulated by the ecosystem model we used, i.e. more complex model parameterizations of R_{aut} and R_{het} may be required to better represent the diversity of situations and processes;

iv) Nitrogen deposition likely contributes a minor fraction (on average 20% according to the model) of total ecosystem N supply (heavily dominated by soil organic N mineralization), except for the very high deposition sites (up to 40%). The fraction of N_{dep}/N_{supply} may even be smaller considering the pool of DON (not included in BASFOR), from which bio-available organic N forms may be taken up by trees in significant quantities in non-fertile, acidic organic soils (Jones and Kielland, 2002; Warren, 2014; Moreau et al., 2019). Thus, in many cases the N_{dep} fertilisation effect may be marginal and difficult to detect, because it may be smaller than typical measurement uncertainties and noise in C and N budgets. Conversely, the effect may be delayed and may manifest even after N_r deposition levels have decreased, as the past N accumulation in soil may support later growth through enhanced N supply.

Commenté [CF22]: Reference to uncertainty in N_{supply} associated with uncertain DON availability

v) Non-linear biological controls that affect C/N relations but are not explicitly considered in the model. For example, BASFOR does consider that N addition can reduce below-ground C allocation (e.g. Högberg et al., 2010), resulting in decreased soil R_{aut} and R_{het} (Janssens et al., 2010), but does not account for the possible consequences of a stimulation of wood cell formation from mid-summer onwards and a delay in the cessation of tracheid production in late season (Kalliokoski et al., 2013).

Commenté [CF23]: Acknowledge model limitations regarding non-linear biological controls

A further limitation to our estimates of the dC/dN response, based on the analysis of the spatial (inter-site) variability in C and N fluxes, is that these forests are not in steady state with respect to N_r deposition and ambient CO_2. Some stands have been affected by, and may be slowly recovering from, excess N_r deposition in the second half of the 20th century; while the more remote sites may always have been N-limited. Figure 1 showed that the modelled GPP of the older forests increased through most of the 20th century, but stabilized when N_{dep} started to decrease after the 1980's, while total N losses also declined over the last 2-3 decades. This is consistent with observations of decreasing N (nitrate) leaching at long term study sites in N-E USA (Goodale et al., 2003; Bernal et al., 2012) and N Europe (Verstraeten et al., 2012; Johnson et al., 2018; Schmitz et al., 2019).

In our model analysis, the declining trend in N_r deposition appears to be the primary driver for the modelled reduced N losses since the 1980's. This can be inferred from model input-sensitivity scenario runs shown in Fig. S9-S11 of the Supplement. In Fig. S9, a constant CO_2 mixing ratio of 310 ppm (i.e. the mean value over the period 1900-2010), used instead of the exponential increase since the 19th century, does not greatly alter overall productivity patterns, nor the decreasing trend in N losses over the period 1980-2010 (Fig S9e-f), compared with the baseline run (Fig. 1). By contrast, in scenarios shown in Fig. S10-S11, the assumed constant N_{dep} levels at all sites of 1.5 and 3.0 g (N) m^{-2} yr^{-1}, respectively, together with the exponential CO_2 increase, remove the decreasing trends in N_r losses over the period 1980-2010. Meanwhile, in constant N_{dep} scenarios the increase in GPP over the whole period is fairly monotonous, in response to a steadily increasing CO_2 (Fig. S10b-c), without the inflexion point around 1980 simulated in the baseline run (Fig 1b-d). In real-life stands, however, decadal decreases in N losses or exports have been observed without any significant reductions in N_{dep} (Goodale et al., 2003). Other potential factors such as increased denitrification, longer growing season, plant N accumulation, changes in soil hydrological properties or temperature, historical disturbances, may also play a role (Bernal et al., 2012). Many such factors are not considered in our model, and neither is long term climate change.

Commenté [CF24]: Added discussion item on declining N losses observed in long term measurements and how this compares with model analysis

The EC-based flux data suggest that the N_{dep} response of forest productivity is clearer at the gross photosynthesis level, in patterns of (normalized) GPP differences among sites, than at the NEP level, where very large differences in CSE among sites lead to a de-coupling of N_{dep} and NEP. The response of GPP to N_{dep} appeared to be reasonably well constrained by both EC flux measurements and BASFOR modelling, which is why we chose to normalize GPP, not NEP. The significantly better model performance obtained for GPP than for R_{eco} and NEP (Fig. 6 in Flechard et al., 2020) likely reveals a relatively poor understanding and mathematical representation of R_{eco} (especially for the soil heterotrophic and autotrophic components)$_t$, and the factors controlling their variability among sites. The large unexplained variability in CSE and C sequestration potentials may also involve other limiting factors that could not be accounted for in our measurement/model analysis, since they are not treated in BASFOR. Such factors may be related to soil fertility, internal N supply, ecosystem health, tree mortality, insect or wind damages in the previous decade, incorrect assumptions on historical forest thinning, all affecting general productivity patterns. Since the observed variability in CSE is key to understanding and quantifying the real-world NEP response to N_{dep} (beyond the relatively well constrained response of GPP in the model world), we explore some of the main issues in the following sections.

Commenté [CF25]: Added transition sentence to introduce later sections

4.3 What drives the large variability in carbon sequestration efficiency?

Carbon sequestration efficiency metrics are directly and negatively related to the ratio of R_{eco} to GPP, expressing the likelihood that one C atom fixed by photosynthesis will be sequestered in the ecosystem. Earlier FLUXNET-based statistical meta-analyses have demonstrated that although R_{eco} is strongly dependent on temperature on synoptic or seasonal scales (Mahecha et al., 2010; Migliavacca et al., 2011), GPP is the key determinant of spatial variations in R_{eco} (Janssens et al., 2001; Migliavacca et al., 2011; Chen et al., 2015), and further, that the fraction of GPP that is respired by the ecosystem is highly variable (Fernández-Martínez et al., 2014), and more variable than in current model representations. We have used three different CSE indicators, averaged across all sites, to derive a NEP/N_{dep} response from model-standardized GPP* data (Table 2). Values of CSE_{obs} varied over a large range among sites (-9 to 61%, Fig. 10). Some of the variability might be due to measurement errors, but small (<10%) or large (>40%) CSE_{obs} values could also genuinely reflect the influence or the absence of ecological limitations related to nutrient availability or vegetation health.

4.3.1 From nutrient limitation to nitrogen saturation

Can nutrient limitation (nitrogen or otherwise) impact ecosystem carbon sequestration efficiency? Soil fertility has been suggested to be a strong driver at least of the forest biomass production efficiency (BPE), defined by Vicca et al. (2012) as the ratio of biomass production to GPP, with BPE increasing in their global dataset of 49 forests from 42% to 58% in soils with low- to high-nutrient availability, respectively. The study by Fernández-Martínez et al. (2014) of 92 forest sites around the globe reported a large variability in CSE (=NEP/GPP calculated from FLUXNET flux data), which they suggest is strongly driven by ecosystem nutrient availability (ENA), with CSE levels below 10% in nutrient-poor forests and above 30% in nutrient-rich forests. The range of CSE values derived from flux measurements in our study (CSE_{obs} in Table 2) was similarly large, even though all of our sites were European and our dataset size was one third of theirs (N=31, of which 26 sites in common with Fernández-Martínez et al., 2014). We did not attempt in this study to characterize a general indicator of ENA beyond total N_r deposition; but if we use the high, medium or low (H, M, L) scores of ENA attributed to each site through factor analysis of nutrient indicators by Fernández-Martínez et al. (2014), we find that the H group (7 sites) has a mean CSE_{obs} of 32% (range 16–48%), the M group is slightly higher (7 sites, mean 39%, range 21–61%), while the L group has indeed a significantly smaller mean CSE_{obs} of 14% (12 sites, range -9 to 38 %). Interestingly, the mean N_{dep} levels for each group are H = 1.5 (range 0.5–2.3) g (N) m^{-2} yr^{-1} , M = 2.1 (range 1.1–4.2) g (N) m^{-2} yr^{-1} and L = 1.3 (range 0.3–4.1) g (N) m^{-2} yr^{-1}, i.e. the highest mean CSE_{obs} of the three groups is found in the group with the highest mean N_{dep} (M).

The nutrients and other indicators of fertility considered by Fernández-Martínez et al. (2014) included, in addition to N, P, soil pH, C/N ratios and cation exchange capacity, as well as soil texture and soil type. However, very few sites were fully documented (see their Supplement Table S1), data were often qualitative, and other key nutrients were not included in the analysis (K, Mg and other cations; S also has been suggested to have become a limiting factor for forest growth following emission reductions, see Fernández-Martínez et al., 2017). The extent to which the overall fertility indicator quantified by ENA was driven by nitrogen in the Fernández-Martínez et al. (2014) factor analysis is not evident. At sites where other nutrients are limiting, the response to N additions would be small or negligible regardless of whether N itself is limiting. This places severe constraints on the interpretation of productivity data in response to N_{dep}, since most current models, which do not account for other nutrient limitations, cannot be called upon to normalize for differences between sites.

The impact of the fertility classification on CSE of the sites included in Fernández-Martínez et al. (2014) was questioned by Kutsch and Kolari (2015) on the basis of unequal quality of the EC-flux datasets found in FLUXNET and other databases. By excluding complex terrain sites (and young forests) from the Fernández-Martínez et al. (2014) dataset, Kutsch and Kolari (2015) calculated a much reduced variability in CSE, with a «reasonable» mean value of 15% (range 0–30%), and suggesting a much lower influence of nutrient status than claimed by Fernández-Martínez et al. (2014). In their reply, Fernández-Martínez et al. (2015) re-analyzed the same subset of sites selected by Kutsch and Kolari (2015), but using the same generalized linear model as used in their original analysis of the whole dataset, as opposed to the linear model used by Kutsch and Kolari (2015). Fernández-Martínez et al. (2015) then maintained that the findings of the original study were still valid for the restricted dataset, i.e. that the nutrient status had a significant influence on CSE.

The smaller European dataset of our study poses a similar dilemma. The much wider variation in CSE_{obs} than modelled $CSE_{5\text{-}yr}$ may both point to possible measurement issues if CSE_{obs} values (especially the larger ones) are considered ecologically implausible, and/or inform on important ecological processes that are not accounted for in the model. Among the forests in our study that seemed particularly inefficient (CSE_{obs} <10%) at retaining photosynthesized carbon (EN4, EN6, EN8, EN11, EN17, EB5), all were classified as L (low ENA) in Fernández-Martínez et al. (2014) and two (EN6, EN11) were even net C sources (R_{eco} > GPP). The EN4, EN6, EN17 sites had the three largest soil organic contents (SOC, Fig. 10a), which may either have induced larger rates of heterotrophic respiration, or may instead indicate low-fertility wet soils where both assimilation and respiration are suppressed. However, EN4 has also been reported as having unrealistically large ecosystem respiration rates (Anthoni et al., 2004). The EN8 site (mature pine-dominated forest in Belgium) was very unlikely to be Nor S-limited, having been under the high deposition footprint of Antwerp petrochemical harbour and local intensive agriculture for decades, even if emissions have declined over the last 20 years (Neirynck et al., 2007, 2011). However, the comparatively low LAI, GPP and CSE (Fig. 4 in Flechard et al., 2020) at this site are likely not independent of the historical, N- and S-induced soil acidification, which has worsened the already low P and Mg availabilities (Janssens et al., 1999), and from which the forest is only slowly recovering (Neirynck et al., 2002; Holmberg et al., 2018). This site is actually an excellent example to illustrate the complex web of biogeochemical and ecological interactions, which further complicate the quantification of the (single-factor) N_{dep} impact on C fluxes. By not accounting for the low Mg and P availabilities and the poor ecosystem health, the BASFOR model massively over-estimated GPP, R_{eco} and NEP at EN8 (Fig. 6 in Flechard et al., 2020). In fact, based on prior knowledge of this site's acidification history, and since such mechanisms and impacts are not mathematically represented in BASFOR, EN8 was from the start discarded from the calibration dataset for the Bayesian procedure (Cameron et al., 2018). The four lowest CSE_{obs} values were found at sites with topsoil pH < 4 (Fig. 10c), although other forests growing on acidic soils had reasonably large CSE_{obs} ratios.

The large variability in CSE_{obs} cannot be explained by any single edaphic factor (Fig. 10a-c), more likely by a combination of many factors that may include N_{dep} (Fig. 10e). As noted previously, C flux measurements at all four forest sites with N_{dep} > 2.5 g (N) m^{-2} yr^{-1} (EN2, EN8, EN15, EN16) indicated lower productivity estimates than those in the intermediate N_{dep} range, or at least smaller than might have been expected from a linear N fertilisation effect (Fig. 4 in Flechard et al., 2020). EN2 (spruce forest in southern Germany) is also well-documented as an N-saturated spruce forest with large total N losses (~3 g (N) m^{-2} yr^{-1}) as NO, N_2O and NO_3^-- (Kreutzer et al., 2009), but its productivity and CSE are not affected to the same extent as EN8. Not all the difference is necessarily attributable to the deleterious impacts of excess N_r deposition, as suggested by the GPP normalization exercise (Fig. 8). For example, EN15 and EN16, planted on sandy soils, appear from meta-modelling to suffer from water stress comparatively more than the average of all sites (Fig. 6-Soil), if indicators of soil water retention based on estimates of soil depth, field capacity and wilting point can be considered reliable.

{Insert Fig. 10 here}

4.3.2 Forest age

Forest age is expected to affect photosynthesis (GPP), growth (NPP), carbon sequestration (NEP) and CSE for many reasons. A traditional view of the effect of stand age on forest NPP (Odum, 1969) postulated that R_{aut} increases with age and eventually nearly balances a stabilized GPP, such that NPP approaches zero upon reaching a dynamic steady state. Revisiting the paradigm, Tang et al. (2014) found that NPP did decrease with age (> 100 yr) in boreal and temperate forests, but the reason was that both GPP and R_{aut} declined, with the reduction in forest growth being primarily driven by GPP, which decreased more rapidly with age than R_{aut} after 100 years. However, the ratio NPP/GPP remained approximately constant within each biome.

The effect of age on NEP and CSE is even more complex since this involves not only changing successional patterns of GPP and R_{aut}, but also of R_{het} over a stand rotation of typically one century or more, which is much longer than the longest available flux datasets. Therefore age effects are often studied by comparing differently aged forest sites across the world, which introduces many additional factors of variation, including differences in water availability, soil fertility, or even tree species, genera, or PFTs. Forest and tree ages should in theory be normalized to account for species-specific ontogeny patterns, i.e. the age of 80 years may be relatively young for some species, and quite old for others, and therefore population dynamics may be very different for the same age. Nevertheless, forest age has been suggested to be a dominant factor controlling the spatial and temporal variability in forest NEP at the global scale, compared with abiotic factors such as climate, soil characteristics and nutrient availability (Besnard et al., 2018). In that study, the multivariate statistical model of NEP, using data from 126 forest eddy-covariance flux sites worldwide, postulated a non-linear empirical relationship of NEP to age, adapted from Amiro et al. (2010), whereby NEP was negative (a net C source) for only a few years after forest establishment, then increased sharply above 0 (a net C sink), stabilized after around 30 years and remained at that level thereafter for mature forests (> 100 years). This model, therefore, did not assume any significant reduction in forest net productivity after maturiy, up to 300 years, consistent with several synthesis studies that have reported significant NEP of centuries-old forest stands (Buchmann and Schulze, 1999; Kolari et al., 2004; Luyssaert et al., 2008).

By analogy, our approach for accounting for the age effect was based on the modelled time course of GPP (Eq. (16)-(17)), which in the BASFOR model tended to stabilize after 100 years, and subsequently using a mean CSE that did not depend on stand age. However, the variability in CSE_{obs} appeared to be much larger in mature forests (>80 years) than in the younger stands (Fig. 10d). For the younger forests (<60 years, all sites probably still in an aggrading phase), the CSE_{obs} values were in a narrow band of 15–30% and were well represented by model simulations, with the exceptions of EN1, EB3 at around 50% and of EN4 being near 0% (all three locations being high elevation sites with complex terrain and potential EC measurement issues, see Flechard et al., 2020). By contrast, values for mature forests were either below 15% or above 30%. For some cold sites such as EN6 and EN11, growing in low nutrient environments (e.g. peat at EN6) with high SOC (Fig. 10a) and/or high soil C/N ratio (Fig. 10b) and low soil pH (Fig. 10c), or for the N-saturated and acidified EN8 site, the low CSE is not necessarily linked to age. Aging, senescence and acidification may at some point curb sequestration efficiency in older forests, but even excluding the complex terrain sites, there remain a good number of productive mature sites with CSE_{obs} in the range 30–40%, which questions the Odum (1969) paradigm of declining net productivity and C equilibirum in old forests.

4.3.3 Does nitrogen deposition impact soil respiration?

The overall net effect of N_r deposition on carbon sequestration must include not only productivity gains, but also indirect, positive or negative impacts on soil C losses, which all affect CSE. Carbon sequestration efficiency reflects the combined magnitudes of soil heterotrophic (R_{het}) and autotrophic (R_{aut}, both below- and above-ground) respiration components, relative to GPP. We postulated that the primary effect of N_{dep} and N_{supply} is on GPP, but potential side effects of N_{dep} or N additions on ecosystem and soil carbon cycling have been postulated. The traditional theory of the role of N on microbial decomposition of SOM was that, above a certain C/N threshold value, the lack of N inhibits microbial activity compared with lower C/N ratios (Alexander, 1977). However, reviews by Fog (1988) and Berg and Matzner (1997) found that microbial activity was often unaffected, or even negatively affected, by the addition of N to low-N decomposing organic material. The negative effects were mostly found for recalcitrant organic matter (high lignin content) with a high C/N ratio (e.g. wood or straw); while N addition to easily degradable organic matter with a low C/N ratio (e.g. leaf litter with low lignin content) actually boosted microbial activity. The meta-analysis by Janssens et al. (2010) of N manipulation experiments in forests suggests that excess N_r deposition reduces soil – especially heterotrophic – respiration in many temperate forests. They argue that the mechanisms include i) a decrease in below-ground C allocation and the resulting root respiration, permitted by a lesser need to develop the rooting system when more N is available (see also Alberti et al., 2015); ii) a reduction in the activity, diversity and biomass of rhizospheric mycorrhizal communities (see also Treseder, 2008); iii) a reduction in the priming effect, the stimulation of SOM decomposition by saprotrophic organisms through root and mycorrhizal release of energy-rich organic compounds; iv) N-induced shifts in saprotrophic microbial communities, leading to reduced saprotrophic respiration; and v) increased chemical stabilization of SOM into more recalcitrant compounds. The authors point out that in N-saturated forests different processes and adverse effects are at play (e.g. base cation leaching and soil acidification). Of the five afore-mentioned mechanisms potentially involved in the suppression of soil respiration by N addition, only the first one (control by N availability of the root/shoot allocation ratio) is functional in BASFOR, and therefore our simulations do not include the other inhibitory effects of excess N on mycorrhizal, fungal and bacterial respiration.

Commenté [CF26]: Reference added

Commenté [CF27]: We make it explicit which of these mechanisms are included, or not, in BASFOR

An important implication of the negative impact of N_r on soil respiration is that the nitrogen fertilisation effect on gross photosynthesis would be roughly doubled, in terms of C sequestration, by the concomitant decrease in soil respiration. In their meta-analyses of N addition experiments in forests and comparison of sites exposed to low *vs* elevated N_{dep}, Janssens et al. (2010) show that both R_{het} and soil carbon efflux (SCE), a proxy for total R_{soil} (= R_{het} + $R_{aut,soil}$), tend to decline with N addition, be it through fertilisation or atmospheric deposition, although the effect is far from universal. The negative N_{dep}

response of R_{het} was much more pronounced for SOM than for leaf litter, and stronger at highly productive sites than at less productive sites. The negative impact on SCE was mostly found at sites where N was not limiting for photosynthesis. When N is strongly limiting, and in young forests, N_r deposition may well favour SOM decomposition.

To examine the potential impact of N_{dep} on R_{soil}, we compiled the soil respiration data available from the literature and databases for the collection of forest sites in our study, which covers the whole N limitation to N saturation spectrum. Sites ranged from highly N-limited boreal systems, where an N addition might trigger enhanced tree growth, increased microbial biomass and heterotrophic respiration, to N-saturated, acidified systems (EN2, EN8, possibly also EN15, EN16), in which poor ecosystem and soil health may lead to different ecological responses than those of the below-ground carbon cycling scheme in Janssens et al. (2010).

Since the below-ground autotrophic (root and rhizosphere) respiration component is regulated to a large extent by photosynthetic activity (Collalti and Prentice, 2019), as well as seasonality in below-ground C allocation (Högberg et al., 2010), and contributes a large part of R_{soil} on an annual basis (Korhonen et al., 2009), the relationship of R_{soil} to N_{dep} is examined by first normalizing to GPP (Fig. 11a), yielding a soil respiration metric that is comparable between sites (for R_{soil} data, see Table S7 in the Supplement to Flechard et al., 2020). Similarly, the ratio R_{soil}/R_{eco} shows the relative contribution of below-ground to total (ecosystem) respiration (Fig. 11c). Note that caution is needed when considering both R_{soil}/GPP and R_{soil}/R_{eco} ratios, since significant uncertainty may arise from i) methodological flaws in comparing chamber *versus* eddy covariance measurements (e.g. considerations over tower footprint, spatial heterogeneity and representativeness of soil collars), ii) uncertainty in deriving GPP and R_{eco} estimates from EC-NEE measurements, and iii) different time spans for the EC and soil chamber measurements, affected by inter-annual flux variability. Thus, values of R_{soil}/R_{eco} above unity (Fig. 11c), although physically non-sensical, do not necessarily imply large measurement errors, but possibly also that there may be no spatial or temporal coherence in EC and chamber data (Luyssaert et al., 2009).

Commenté [CF28]: Added a mention of the seasonality in C allocation to this sentence.

Either ignoring such outliers, or judging that a measurement bias by soil chambers affects all sites the same way (e.g. systematic over-estimation of soil respiration in low turbulence conditions when using static chambers, Brændholt et al., 2017), we may argue that the apparent decrease of both chamber/EC ratios R_{soil}/GPP and R_{soil}/R_{eco} with N_{dep} (Fig. 11a, 11c) has some reality, even if their absolute values are biased. Soil CO_2 efflux tends to be a larger fraction of GPP (>0.5) at the smaller N_{dep} rates (<1.5 g (N) m^{-2} yr^{-1}) than at sites with larger N_{dep}, where this fraction is more often in the range 0.4–0.5. It is also noteworthy that the largest R_{soil}/GPP ratios (EN5, EN17) are found at sites with very large SOC compared with the other sites (Fig. 11b). The R_{het}/R_{soil} ratio also tends to decrease with N_{dep} (Fig. 11e), and although measured by different methods at the different sites, this is arguably a more robust metric than chamber/EC respiration ratios, because the differential respiration measurements on control and treatment plots (root exclusion, trenching, girdling) are made on the same spatial and temporal scales.

Many other factors that impact soil respiration (age, soil pH, microbial abundance and diversity, etc.) are not considered here and beyond the scope of this paper. In view of these uncertainties, if the assessment within this restricted dataset does not provide a full and incontrovertible proof of the negative impact of N_r deposition on soil respiration, it at least is not in open contradiction to the prevailing paradigm that both below-ground autotrophic and heterotrophic respiration are expected to decrease as N_r deposition increases. However, the decreasing trends observed in Fig. 11a, 11c, 11e are largely driven by these few high N_{dep} sites (>3g (N) m^{-2} yr^{-1}) in which the negative effects of N saturation and acidification very likely outweigh the benefits of reduced soil respiration in terms of C sequestration.

{Insert Fig. 11 here}

5 Conclusion

The magnitude of the mean N_r deposition-induced fertilisation effect on forest C sequestration, derived here from eddy covariance flux data from a diverse range of European forest sites, is of the order of 40–50 g (C) g^{-1} (N), and comparable with current estimates obtained from inventory data and deposition rates from continental-scale deposition modelling used in the most recent studies and reviews. The range of dC/dN values is a consequence of where in the ecosystem the N_r-induced carbon sequestration takes place, whether there are N_r losses and how other environmental conditions affect growth. However, this mean dC/dN response should be taken with caution for several reasons. First, uncertainties in our dC/dN estimates are large, partly because of the relatively small number of sites (31) and their large diversity in terms of age, species, climate, soils, and possibly fertility and nutrient availability. Second, adopting a mean overall dC/dN response universally and regardless of the context may be misleading due to the clear non-linearity in the relationship between forest productivity and the level of N_r deposition, i.e. the magnitude of the response changes with the N status of the ecosystem. Beyond a N_r deposition threshold of 1–2 g (N) m^{-2} yr^{-1} the productivity gain per unit N_r deposited from the atmosphere starts to decrease significantly. Above 2.5 g (N) m^{-2} yr^{-1}, productivity actually decreases with further N_r deposition additions, and this is accompanied by increasingly large ecosystem N_r losses, especially as NO_3^- leaching. Further sources of uncertainty in our forest ecosystem model involve missing – but possibly large – terms of the N cycle, such as N_2 fixation, N_2 loss by denitrification, DON uptake by trees and DON leaching.

Ecosystem meta-modelling was required to factor out the effects of climate, soil water retention and age on forest productivity, a necessary step before estimating a generalised response of C storage to N_r deposition. Neglecting these effects would lead to a large over-estimation (factor of 2) of the dC/dN effect in this European dataset and possibly also in other datasets worldwide. After factoring out the effects of climate, soil water retention and forest age in the present dataset, only part of the non-linearity was removed and there was still a decline in the dC/dN response with increasing N_{dep}. One possible interpretation is that the remaining non-linearity may be regarded as an indicator of the impact of increasing severity of N saturation on ecosystem functioning and forest growth. However, the results also show that the large inter-site variability in carbon sequestration efficiency, here defined at the ecosystem scale and observed in flux data, cannot be entirely explained by the processes represented in model we used. This is likely due in part to an incomplete understanding and over-simplified model representation of plant carbon relations, soil heterotrophic and autotrophic respiration, the response to nitrogen deposition of physiological processes such as stomatal conductance and water-use efficiency, and possibly also because other nutrient limitations were insufficiently documented at the monitoring sites and not accounted for in the model.

Code and data availability

The data used in this study are publicly available from online databases and from the literature as described in the Materials and Methods section.

The codes of models and other software used in this study are publicly available online as described in the Materials and Methods section.

Author Contributions

CRF, MvO, DRC, WdV, MAS, AI conceived the paper; CRF performed the data analyses, ran model simulations and wrote the text; MvO, DRC wrote and provided the BASFOR model code and performed Bayesian calibration; MAS, EN, UMS, KBB, WdV conceived or designed the NEU study; AI, NB, IAJ, JN, LM, AV, DL, ALeg, KZ, MAub, MAur, BHC, JD, WE, RJ, WLK, ALoh, BL, GM, VM, JO, MJS, TV, CV, KBB, UMS provided eddy covariance and/or other field data, or contributed to data collection from external databases and literature; MvO, DRC, WdV, AI, MAS, NB, NBD, IAJ, JN, LM, AV, DL, ALeg, KZ, AJF, RJ, AN, EN, UMS contributed substantially to discussions and revisions.

Competing Interests

The authors declare that they have no conflict of interest

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Figures and Tables

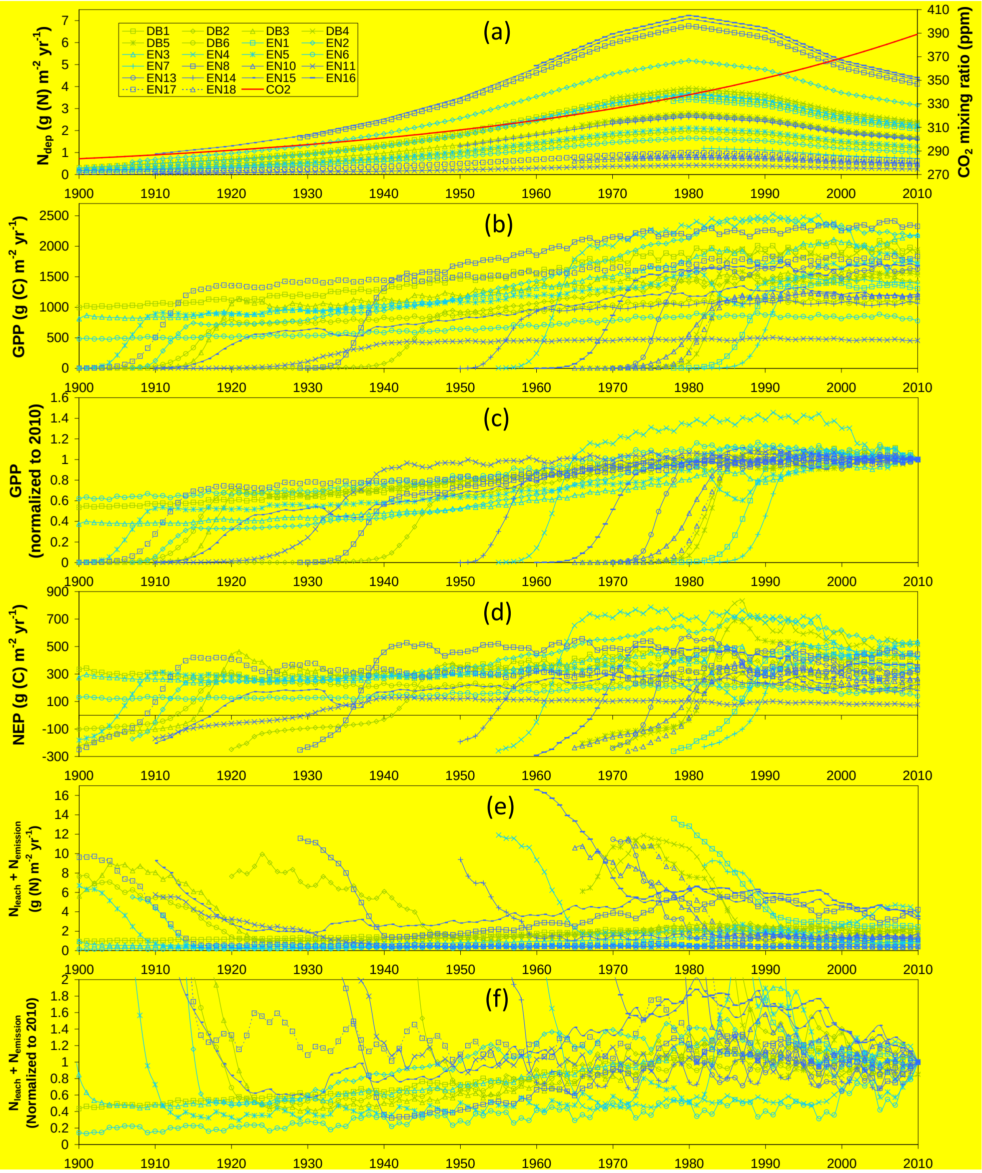


Figure 1. Time courses for 22 forest study sites (DB: deciduous broadleaf; EN: evergreen needleleaf) of (a) assumed atmospheric N_r deposition (N_{dep}) and CO_2 mixing ratio, and baseline model simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching (N_{leach}) and gaseous emissions ($N_{emission}$), and (f) total N losses normalized to 2010.

Commenté [CF29]: Added figure to show historical dimension. All other figures were re-numbered.

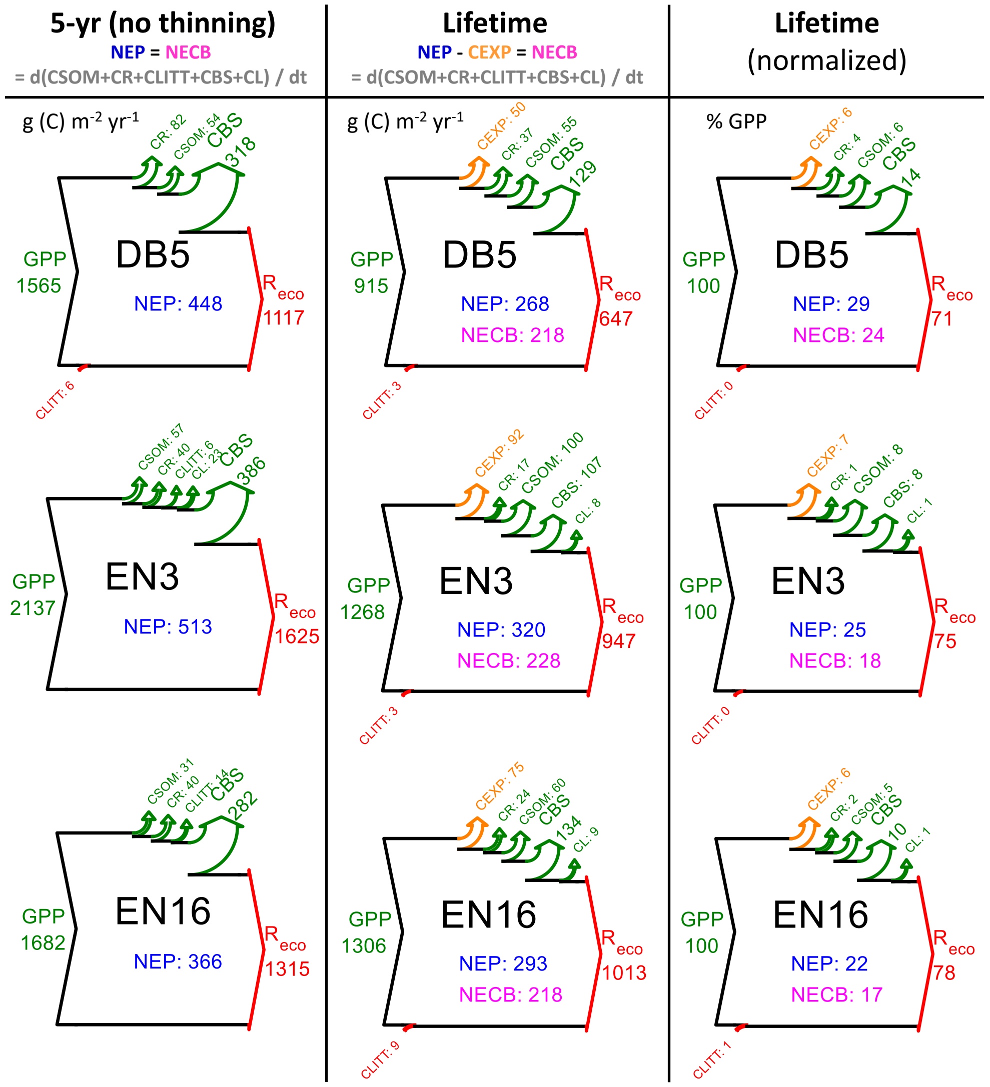


Figure 2. Modelled (BASFOR) budgets and partitioning of gross primary productivity (GPP), ecosystem respiration (R_{eco}), net ecosystem productivity (NEP), net ecosystem carbon balance (NECB), at three example forest sites (DB5: 45-yr old *Fagus sylvatica*; EN3: 120-yr old *Picea abies*; EN16: 51-yr old *Pseudotsuga menziesii*), and associated modelled changes in C pools in soil organic matter (CSOM), roots (CR), litter layers (CLITT), branches and stems (CBS) and leaves (CL) (units: g (C) m^{-2} yr^{-1} left and center; normalized to % lifetime GPP on the right). Simulations were run either over the most recent 5-year period which did not include any thinning event («5-yr» in the text), or over the whole time period since the forest was established («lifetime»). Green indicates ecosystem C gain (photosynthesis and C pool increase); red denotes ecosystem C loss (respiration and C pool decrease); the orange arrows indicate C export through thinning (CEXP). The NECB percentage value (right) corresponds to the lifetime carbon sequestration efficiency (CSE). The sizes of the Sankey plots are not proportional to the C fluxes of the different study sites.

Commenté [CF30]: Acronyms made explicit.

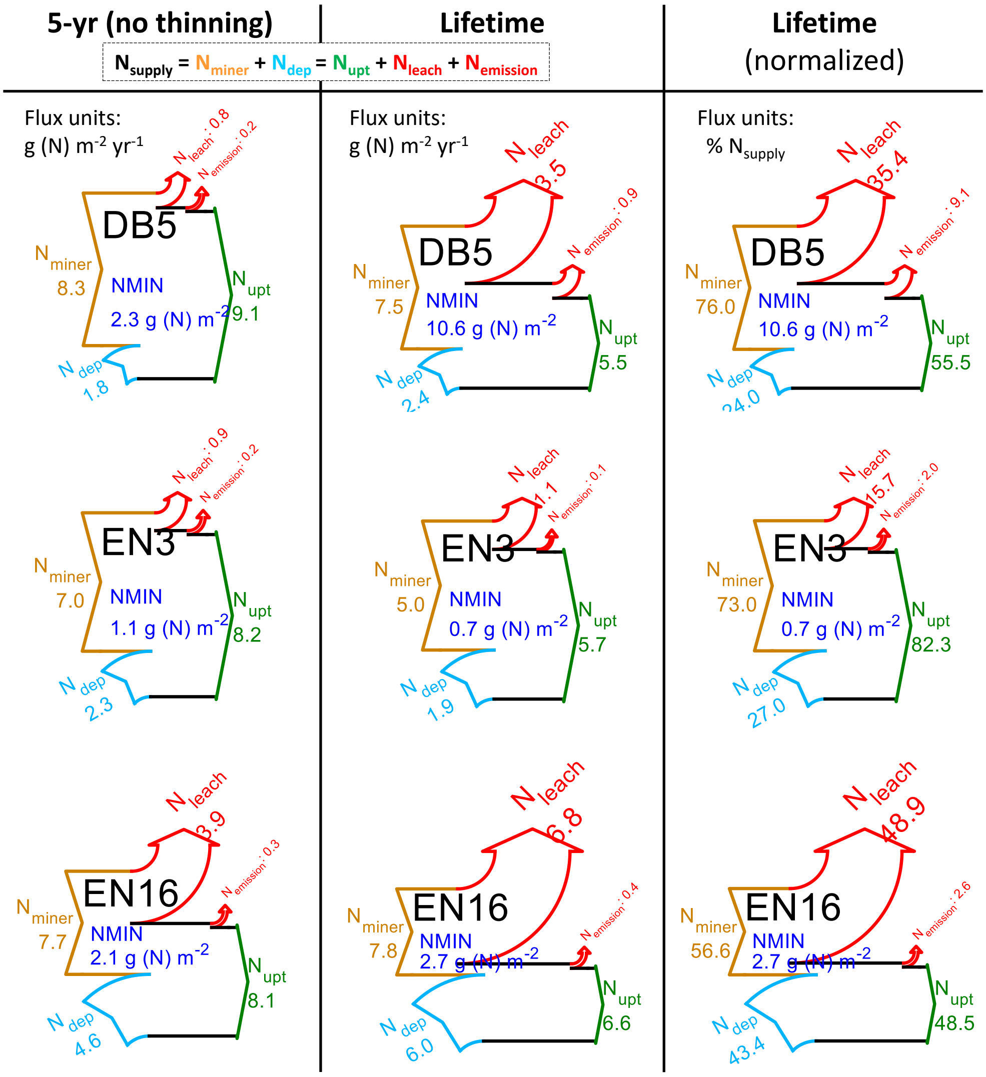


Figure 3. Modelled (BASFOR) inorganic nitrogen budgets at three example forest sites (DB5: 45-yr old *Fagus sylvatica*; EN3: 120-yr old *Picea abies*; EN16: 51-yr old *Pseudotsuga menziesii*). Simulations were run either over the most recent 5-year period which did not include any thinning event («5-yr» in the text), or over the whole time period since the forest was established («lifetime»). The data show ecosystem SOM mineralisation (N_{miner}) and atmospheric N_r deposition (N_{dep}), balanced by vegetation uptake (N_{upt}) and the sum of losses as dissolved N (N_{leach}) and gaseous NO + N_2O ($N_{emission}$) (units: g (N) m^{-2} yr^{-1} left and center; % of lifetime N_{supply} on the right, with N_{supply} defined as N_{miner} + N_{dep}). NMIN indicates the mean size of the soil inorganic N pool (g (N) m^{-2}) over the modelling period. The N uptake percentage value (right) corresponds to the lifetime nitrogen uptake efficiency (NUPE). The sizes of the Sankey plots are not proportional to the N fluxes of the different study sites.

Commenté [CF31]: Acronyms made explicit.

Commenté [CF32]: Changed NUE to NUPE

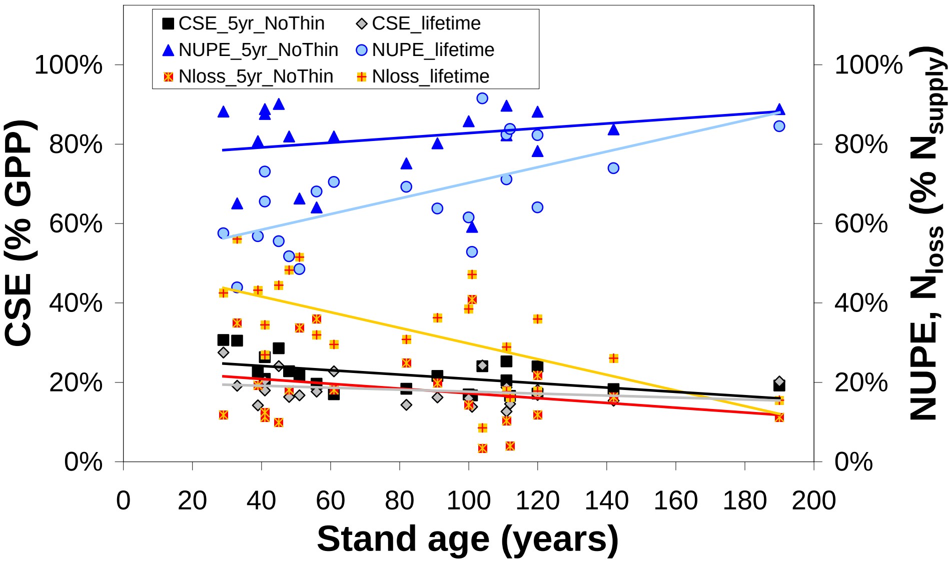


Figure 4. Influence of forest stand age on modelled (BASFOR) C sequestration efficiency (CSE, expressed as % gross primary productivity GPP), N uptake efficiency (NUPE) and the N_{loss} fraction (expressed as % N_{supply}). Each data point represents one of 22 modelled forest sites. CSE and NUPE values are calculated either i) over the most recent 5-yr period including no thinning event around the time frame of the CEIP-NEU integrated projects, or ii) over the whole lifetime of the stands (including all thinning events). See Eq. (1)-(9) for definitions and calculations of the indicators.

Commenté [CF33]: Changed NUE to NUPE

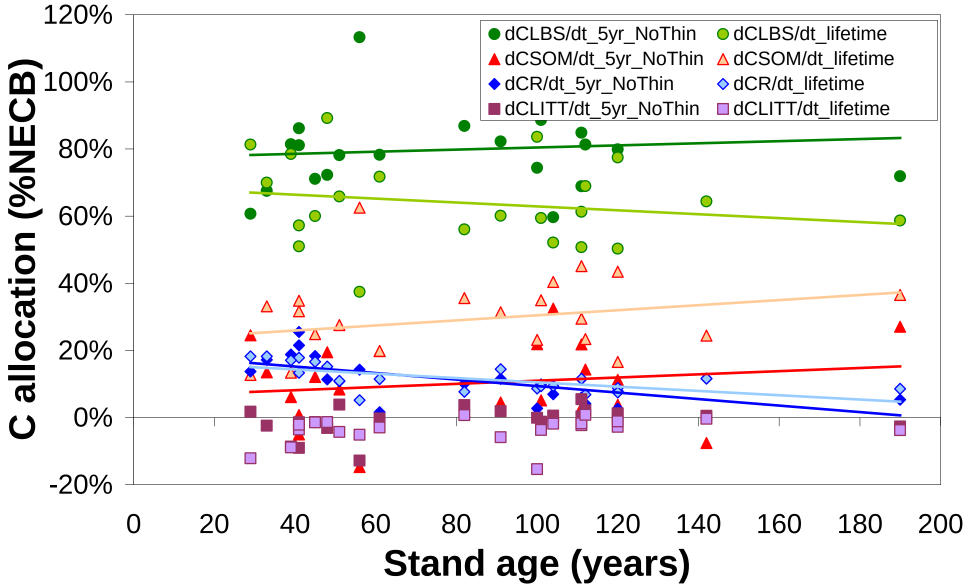


Figure 5. Modelled (BASFOR) ultimate allocation of sequestered C (expressed as % net ecosystem carbon balance NECB) into ecosystem pools in soil organic matter (CSOM), roots (CR), litter layers (CLITT), leaves, branches and stems (CLBS). Each data point represents one of 22 modelled forest sites, plotted as a function of stand age. At each site, the net ecosystem carbon balance equals the sum of all individual storage (or loss) terms, i.e. NECB = dCLBS/dt + dCSOM/dt + dCR/dt + dCLITT/dt, shown here as fractions of the total to indicate the relative importance of the different ecosystem sinks. Values are calculated either i) over the most recent 5-year period including no thinning event around the time frame of the CEIP-NEU integrated projects, or ii) over the whole lifetime of the stands (including all thinning events).

Commenté [CF34]: Provided additional explanations to help reading of figure

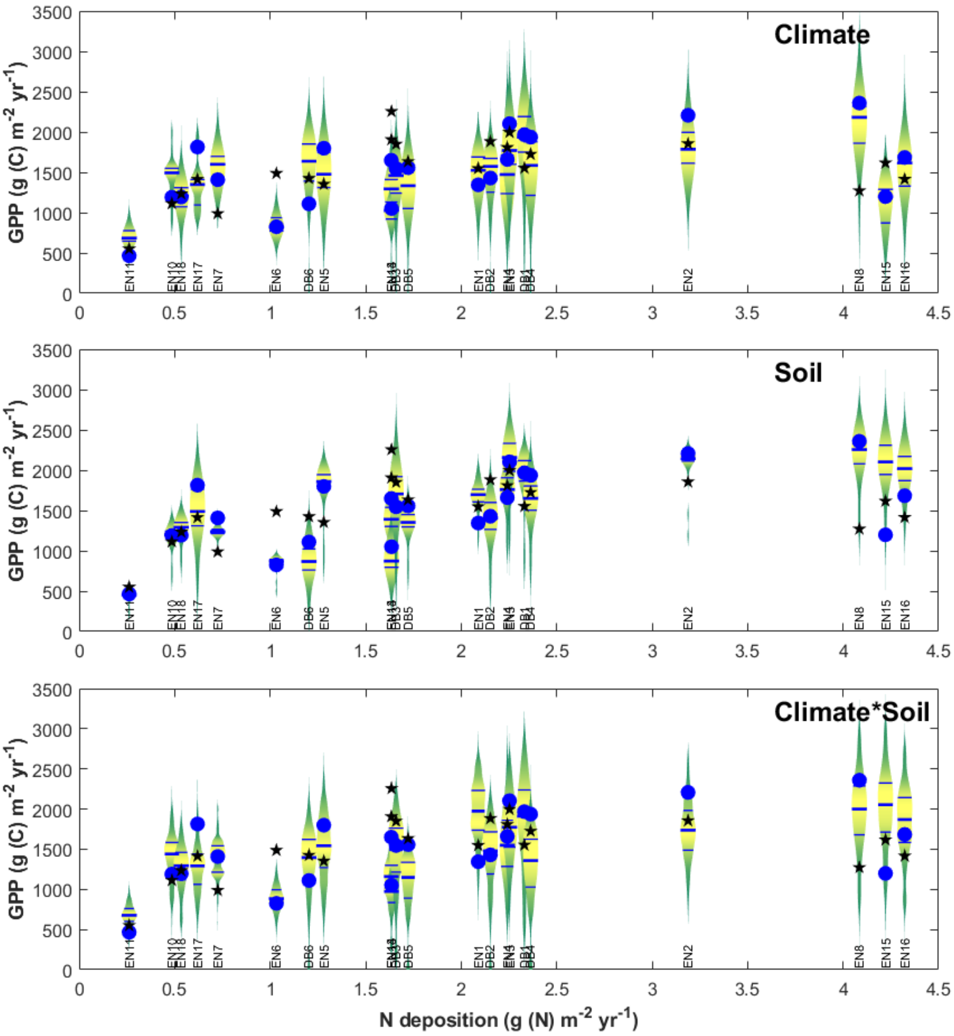


Figure 6. Input sensitivity study for gross primary productivity (GPP) modelled at each forest monitoring site for different soil/climate scenarios (vertical «violin» plots), compared with model base runs GPP$_{base}$ (blue circles) and EC-derived GPP$_{obs}$ (black stars). The data are displayed as a function of N$_r$ deposition over the CEIP-NEU measurement periods, for n=22 deciduous broadleaf (DB) and coniferous evergreen needleleaf (EN) forest ecosystems. For each site, the violin plot shows the range and distribution (median, quartiles) of GPP modelled at the site using climate and/or soil input data from all 22 sites, showing the sensitivity to model inputs other than N deposition. See text for details.

Commenté [CF35]: Provided additional explanations to help reading of figure

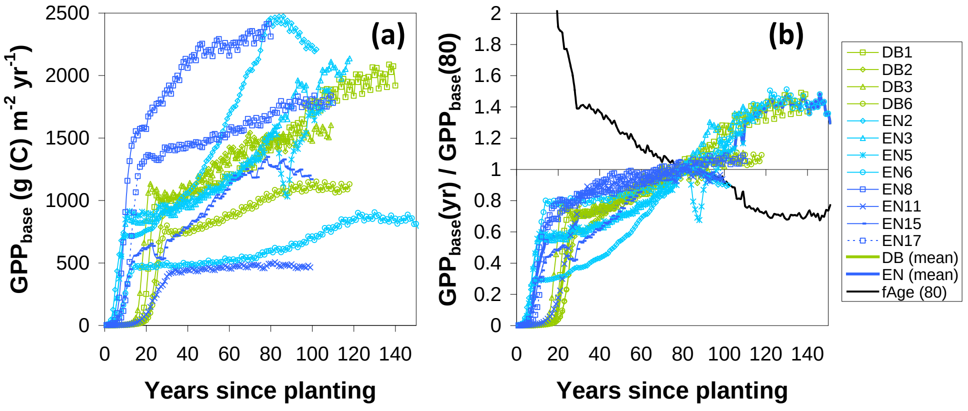


Figure 7. Steps in the calculation of a normalization factor for forest age (f_{AGE}, normalized to 80 yr) from modelled BASFOR growth curves for mature forests (12 sites older than 80 yr). (a) Modelled time course for baseline gross primary productivity (GPP_{base}); (b) Each site's GPP_{base} curve is normalized to the value at age 80 yr. A single f_{AGE} curve is then calculated as the mean of all sites after normalization to $GPP_{base}(80)$. The f_{AGE} curve is subsequently used as a scaling function to standardize all sites' measured GPP to a notional age of 80 (see Eq. (10), (16), (17)). DB: deciduous broadleaf; EN: coniferous evergreen needleleaf.

Commenté [CF36]: Provided additional explanations to help reading of figure

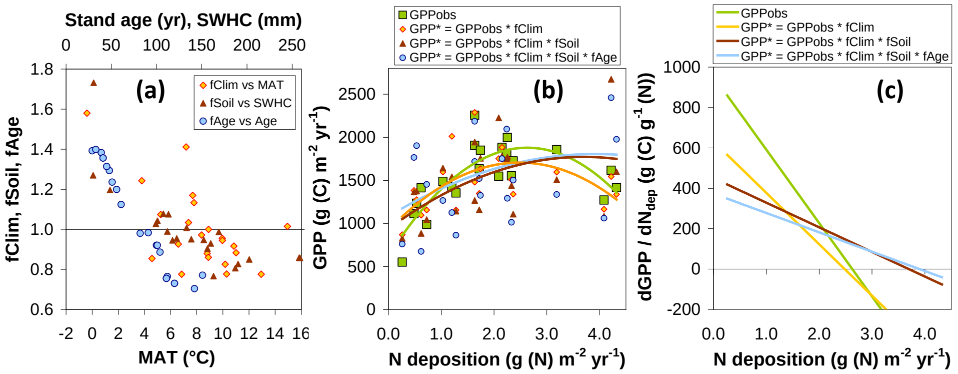


Figure 8. Model-based assessment of the sensitivity of gross primary productivity (GPP) to climate, soil, age and N_r deposition. (a) GPP standardization factors for climate (f_{CLIM}), soil (f_{SOIL}) and age (f_{AGE}) for observational (EC-based) data as a function of the dominant climatic and soil drivers (MAT: mean annual temperature; SWHC: soil water holding capacity; see text for details); (b) the resulting standardized GPP* compared with the original GPP_{obs} as a function of N_{dep} (one data point for each of 22 sites), with 2nd-order polynomial fits; (c) estimates of the GPP response to N_{dep}, calculated as the slope of the tangent line to the quadratic fits and plotted as a function of N_{dep}.

Commenté [CF37]: Acronyms made explicit.

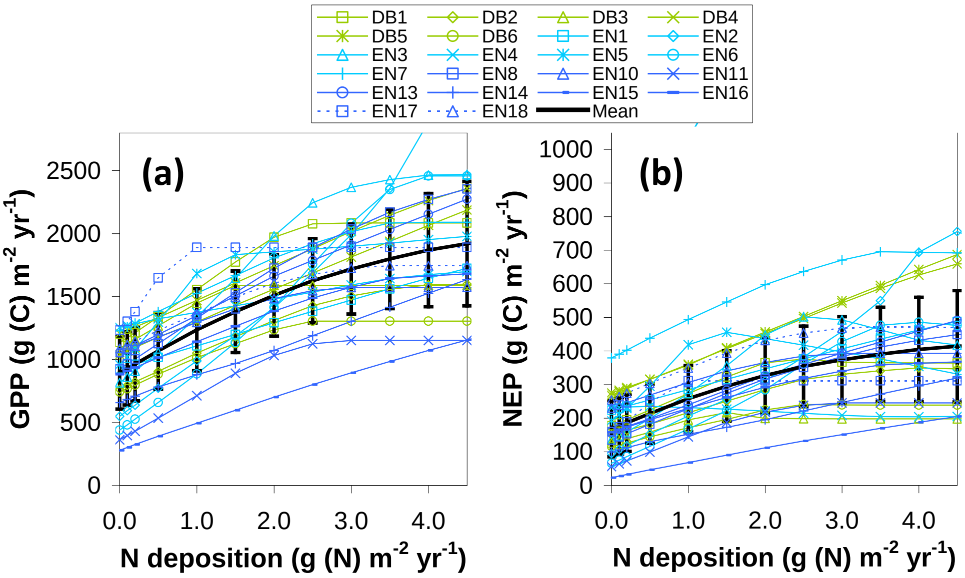


Figure 9. Simulated BASFOR model sensitivity to N deposition of (a) gross primary productivity (GPP) and (b) net ecosystem productivity (NEP) for 22 forest sites (with mean +/- standard deviation), derived from a purely modelled approach (not involving measured EC flux data). Each site was modelled using a range of N_{dep} values from 0 to 4.5 g (N) m^{-2} yr^{-1} (constant N_{dep} over the lifetime of the stands). DB: deciduous broadleaf; EN: coniferous evergreen needleleaf.

Commenté [CF38]: Acronyms made explicit.
Provided additional explanations to help reading of figure

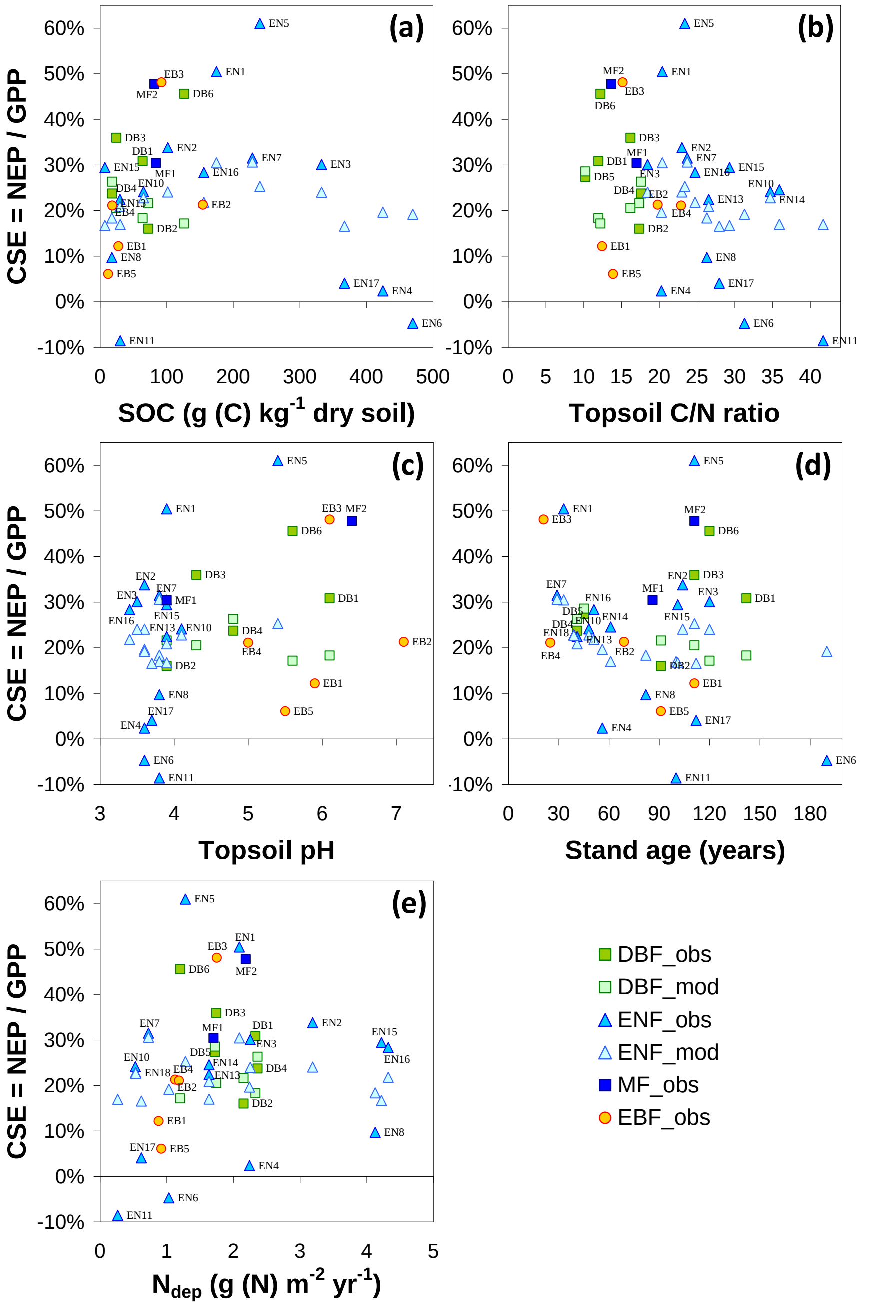


Figure 10. Variability of observation-based (obs) and modelled (mod) carbon sequestration efficiency (CSE) defined as the ratio of net ecosystem productivity (NEP) to gross primary productivity (GPP), calculated over a ~5-yr measurement period. The data are plotted versus (a) topsoil organic carbon content (SOC), (b) topsoil C/N ratio, (c) topsoil pH, (d) forest stand age, and (e) nitrogen deposition (N_{dep}). DBF: deciduous broadleaf forests; ENF: coniferous evergreen needleleaf forests; MF: mixed needleleaf/broadleaf forests; EBF: Mediterranean evergreen broadleaf forests.

Commenté [CF39]: Acronyms made explicit.
Site labels in plots moved to avoid text overlap

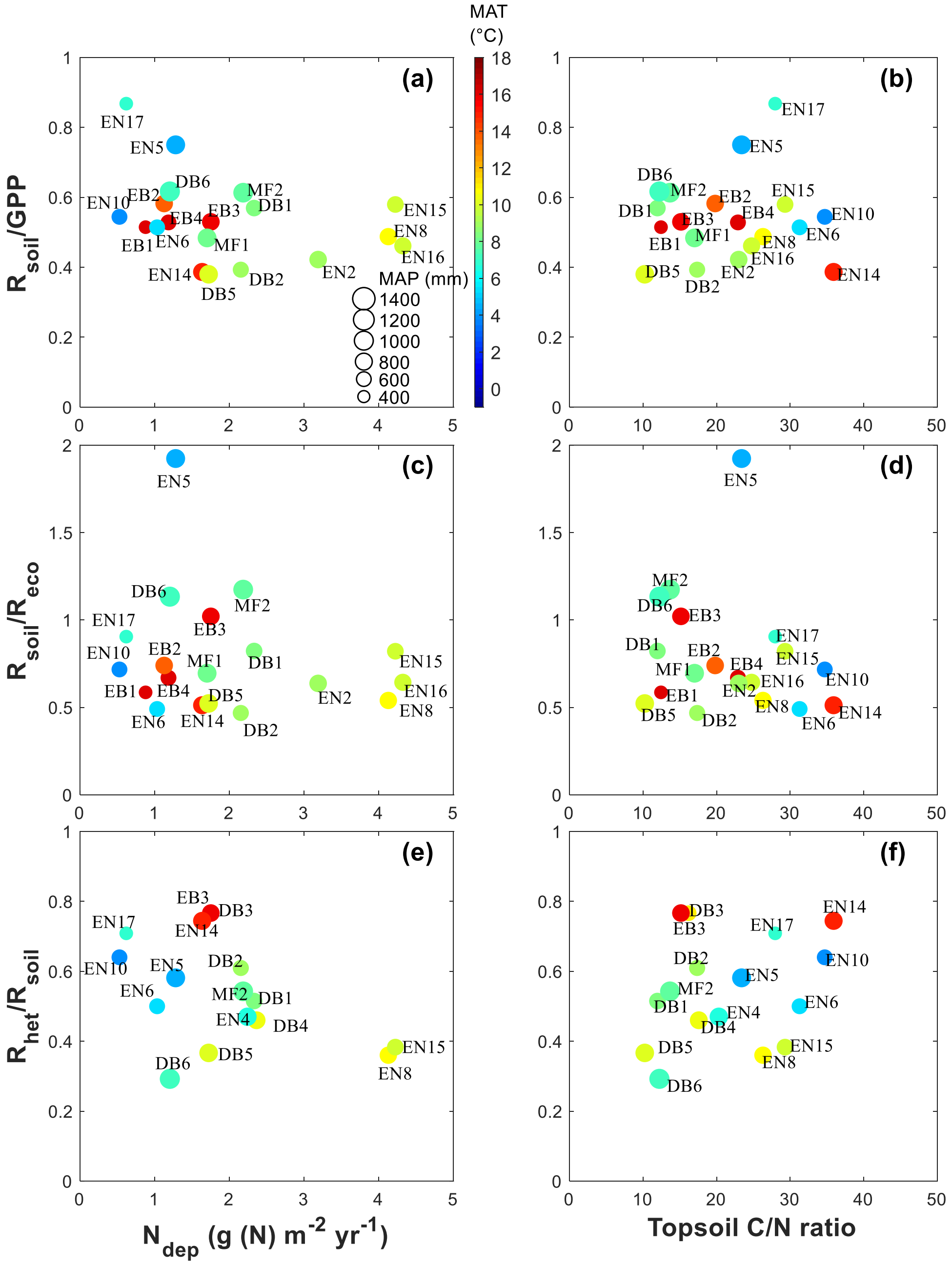


Figure 11. Variability of normalized soil respiration metrics as a function of nitrogen deposition (a, c, e) and soil organic carbon (b, d, f). In all plots, the color scale indicates mean annual temperature (MAT), and the symbol size is proportional to mean annual precipitation (MAP). R_{soil}: total soil respiration; R_{eco}: total ecosystem respiration; R_{het}: heterotrophic component of R_{soil}; GPP: gross primary productivity. DB: deciduous broadleaf; EN: coniferous evergreen needleleaf; EB: Mediterranean evergreen broadleaf; MF: mixed needleleaf/broadleaf forests.

Commenté [CF40]: Acronyms made explicit.
Site labels in plots moved to avoid text overlap

Table 1. BASFOR model state variables, inputs and outputs, and other acronyms used in the study.

BASFOR variables	Description
Tree state variables	
CL	Carbon pool in Leaves
CB	Carbon pool in Branches
CS	Carbon pool in Stems
CLBS	Carbon pool in Leaves, Branches and Stems
CR	Carbon pool in Roots
CRES	Carbon pool in Reserves
NL	Nitrogen pool in Leaves
SD	Forest stand density
Soil state variables	
CLITT	Carbon pool in Litter layers
CSOMF	Carbon pool in Soil Organic Matter (Fast turnover)
CSOMS	Carbon pool in Soil Organic Matter (Slow turnover)
NLITT	Nitrogen pool in Litter layers
NSOMF	Nitrogen pool in Soil Organic Matter (Fast turnover)
NSOMS	Nitrogen pool in Soil Organic Matter (Slow turnover)
NMIN	Soil Mineral (inorganic) Nitrogen pool
WA	Water pool in the root zone
Soil parameters	
Φ_{SAT}	Saturation soil water content
Φ_{FC}	Field capacity
Φ_{WP}	Wilting point
ROOTD	Root depth
Model inputs (daily time step)	
R_g	Daily global radiation
T_a	Daily average air temperature
P	Daily accumulated rain
WS	Daily average wind velocity
RH	Water vapour pressure
CO_2	Annual mean CO_2 mixing ratio
N_{dep}	Annual atmospheric nitrogen deposition
thinFR	Fraction of trees removed by thinning
Model outputs	
H	Tree height
DBH	Diameter at breast height
LAI	Leaf area index
LeafN	Leaf N content
GPP	Gross primary productivity
R_{eco}	Ecosystem respiration
R_{het}	Soil heterotrophic respiration
NPP	Net primary productivity
NEE	Net ecosystem exchange
ET	Evapotranspiration
N_{miner}	Nitrogen supply from SOM mineralization
N_{upt}	Root N uptake by trees
N_{leach}	Inorganic N leaching
NO	Nitric oxide
N_2O	Nitrous oxide
$N_{emission}$	Gaseous soil NO + N_2O emissions

Other variables	
GPP_{obs}, NEP_{obs}	Observation-based (eddy covariance) GPP or NEP
GPP_{base}	Baseline model run for GPP
GPP*, NEP*	Model-standardized observation-based GPP or NEP
f_{CLIM}, f_{SOIL}, f_{AGE}	Model-derived standardization factors to account for climate, soil, age
NECB	Modelled net ecosystem carbon balance, calculated as d(CLBS+CR+CSOM+CLITT)/dt
R_{aut}	Autotrophic respiration
R_{soil}	Soil (heterotrophic and rhizospheric) respiration
SCE	Soil CO_2 efflux measured by chamber methods
CSE_{obs}	Observation-based carbon sequestration efficiency (NEP_{obs}/GPP_{obs})
$CSE_{5\text{-yr, lifetime}}$	Modelled carbon sequestration efficiency; = NEP/GPP (5-yr), or NECB/GPP (lifetime)
NUPE	Modelled nitrogen uptake efficiency, calculated as N_{upt} / N_{supply}
N_{supply}	Total mineral N supply, calculated as (modelled) N_{miner} + (observation-based) N_{dep}
N_{loss}	Modelled percentage ecosystem N losses, calculated as (N_{leach} + $N_{emission}$) / N_{supply}
dC/dN, $dGPP/dN_{dep}$, $dNEP/dN_{dep}$	Response (slope) of ecosystem C productivity versus atmospheric N_r deposition
SWHC	Soil water holding capacity, = (Φ_{FC} - Φ_{WP}) x ROOTD
MAT, MAP	Mean annual temperature or precipitation
CEXP	Carbon exported by thinning or harvest in forests

Commenté [CF41]: Changed NUE to NUPE

Table 2. Estimates of ecosystem dC/dN response for gross and net productivity, calculated under different assumptions and expressed as g (C) photosynthesized or sequestered per g (N) deposited from the atmosphere. The stepwise method described in this paper (for forests only) first calculates dGPP/dN$_{dep}$, for both raw GPP$_{obs}$ and GPP* standardized by meta-modelling following Eq. (10)-(17); this is then multiplied by different estimates of CSE (from observations or from modelling) to provide an NEP (5-yr) or NECB (lifetime) equivalent. Quadratic regressions (Q) are used for productivity vs N$_{dep}$, whereby the mean tangent slope is calculated either over the whole N$_{dep}$ range (0-4.3 g (N) m^{-2} yr^{-1}) (italics), or discarding sites with N$_{dep}$ larger than 2.5 g (N) m^{-2} yr^{-1} (bold). Uncertainty ranges are calculated from combined standard errors in dGPP/dN$_{dep}$ and in CSE. For comparison purposes only, are also displayed i) simple linear regression (L) slopes of EC-based (not standardized) GPP$_{obs}$ and NEP$_{obs}$ versus N$_{dep}$ for both forests and semi-natural vegetation; and ii) results of the meta-modelling standardization method applied directly to NEP$_{obs}$ instead of GPP$_{obs}$.

		FORESTS				**SEMI-NATURAL**
		GPP$_{obs}$	GPP*			GPP$_{obs}$
			GPP$_{obs}$ x f$_{CLIM}$	GPP$_{obs}$ x f$_{CLIM}$ x f$_{SOIL}$	GPP$_{obs}$ x f$_{CLIM}$ x f$_{SOIL}$ x f$_{AGE}$	
Gross primary productivity per unit N$_{dep}$ dGPP /dN$_{dep}$ (g (C) g^{-1} (N))		*260 [38, 483]*$^{(M,Q)}$ **425 [203, 648]** $^{(M,Q)}$	*146 [-121, 412]* $^{(M,Q)}$ **261 [-5, 528]** $^{(M,Q)}$	*218 [-174, 609]* $^{(M,Q)}$ **273 [-119, 664]** $^{(M,Q)}$	*190 [-375, 755]* $^{(M,Q)}$ **234 [-331, 799]** $^{(M,Q)}$	
		146 [89, 203] $^{(A,L)}$ **432 [355, 509]** $^{(A,L)}$				*374 [275, 474]* $^{(A,L)}$ **504 [331, 677]** $^{(A,L)}$
Net ecosystem productivity per unit N$_{dep}$ (g (C) g^{-1} (N))	CSE$_{obs}$ * (dGPP /dN$_{dep}$)	*64 [8, 136]* $^{(M,Q)}$ **105 [43, 182]** $^{(M,Q)}$	*36 [-25, 116]* $^{(M,Q)}$ **64 [-1, 149]** $^{(M,Q)}$	*53 [-36, 172]* $^{(M,Q)}$ **67 [-25, 187]** $^{(M,Q)}$	*47 [-79, 213]* $^{(M,Q)}$ **57 [-69, 225]** $^{(M,Q)}$	
	CSE$_{5\text{-}yr}$ * (dGPP /dN$_{dep}$)	*57 [8, 110]* $^{(M,Q)}$ **93 [42, 147]** $^{(M,Q)}$	*32 [-25, 94]* $^{(M,Q)}$ **57 [-1, 120]** $^{(M,Q)}$	*47 [-36, 138]* $^{(M,Q)}$ **59 [-25, 151]** $^{(M,Q)}$	*41 [-78, 172]* $^{(M,Q)}$ **51 [-69, 181]** $^{(M,Q)}$	
	CSE$_{lifetime}$ * (dGPP /dN$_{dep}$)	*47 [7, 91]* $^{(M,Q)}$ **77 [35, 122]** $^{(M,Q)}$	*26 [-21, 78]* $^{(M,Q)}$ **47 [-1, 100]** $^{(M,Q)}$	*39 [-30, 115]* $^{(M,Q)}$ **49 [-20, 125]** $^{(M,Q)}$	*34 [-65, 143]* $^{(M,Q)}$ **42 [-57, 151]** $^{(M,Q)}$	
		NEP$_{obs}$	NEP*			NEP$_{obs}$
			NEP$_{obs}$ x f$_{CLIM}$	NEP$_{obs}$ x f$_{CLIM}$ x f$_{SOIL}$	NEP$_{obs}$ x f$_{CLIM}$ x f$_{SOIL}$ x f$_{AGE}$	
	dNEP /dN$_{dep}$	*108 [-118, 333]* $^{(M,Q)}$ **178 [-47, 403]** $^{(M,Q)}$	*93 [-166, 352]* $^{(M,Q)}$ **161 [-98, 420]** $^{(M,Q)}$	*120 [-162, 403]* $^{(M,Q)}$ **165 [-117, 447]** $^{(M,Q)}$	*112 [-146, 370]* $^{(M,Q)}$ **146 [-112, 404]** $^{(M,Q)}$	
		71 [29, 114] $^{(A,L)}$ **224 [157, 292]** $^{(A,L)}$				*89 [46, 132]* $^{(A,L)}$ **34 [-41, 109]** $^{(A,L)}$

GPP: gross primary productivity; NEP: net ecosystem productivity; NECB: net ecosystem carbon balance

GPP$_{obs}$, NEP$_{obs}$: observation-based (eddy covariance) GPP or NEP

GPP*, NEP*: GPP or NEP standardized through meta-modelling for the effects of climate (f$_{CLIM}$), soil (f$_{SOIL}$), age (f$_{AGE}$)

CSE: carbon sequestration efficiency

CSE$_{obs}$ = NEP$_{obs}$ / GPP$_{obs}$ (eddy covariance-based, mean value across all sites)

CSE$_{5\text{-}yr}$ = NEP$_{5\text{-}yr}$ / GPP$_{5\text{-}yr}$ (BASFOR model-based over 5-yr period, mean value across all sites)

CSE$_{lifetime}$ = NECB$_{lieftime}$/ GPP$_{lifetime}$ (BASFOR model-based over lifetime, mean value across all sites)

^{Q}: calculated by quadratic regression

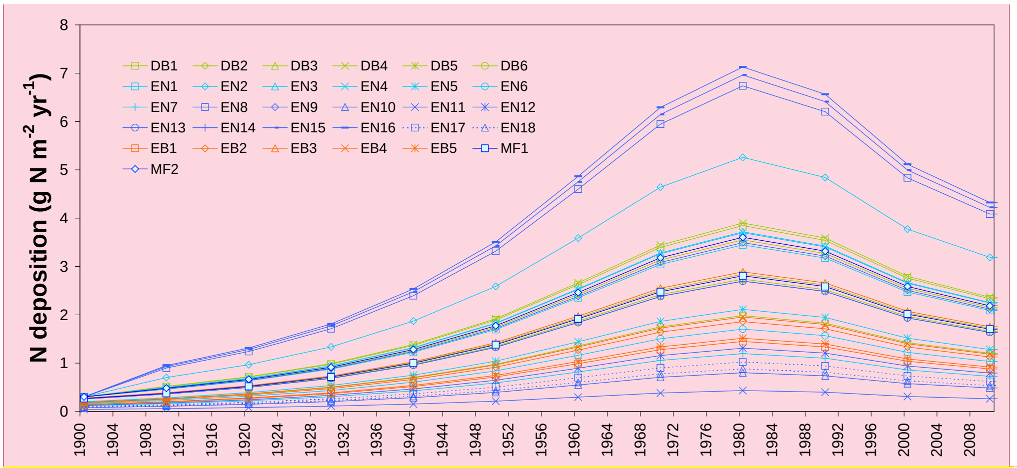
^{L}: calculated by simple linear regression

^{A}: calculated on the basis of all sites in the monitoring network (31 forests, 9 semi-natural sites)

^{M}: calculated on the basis of the subset of 22 forest sites included in BASFOR meta-modelling

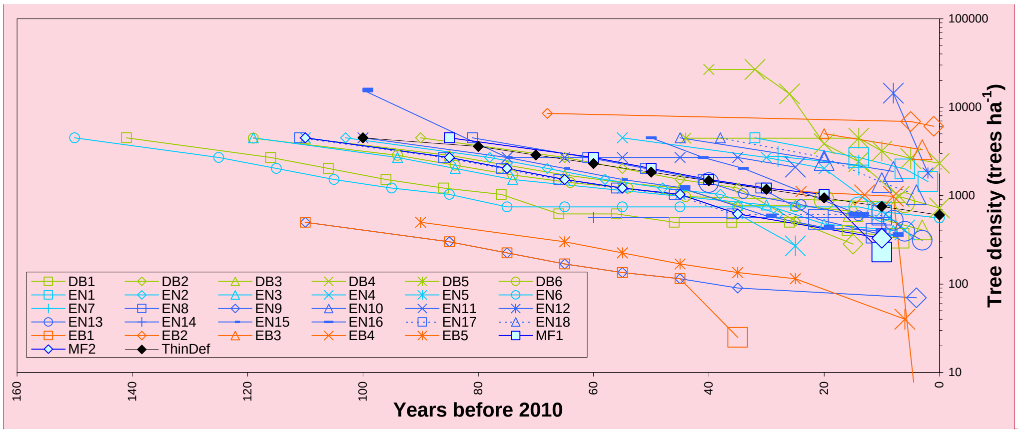
Supplement to 'Carbon/nitrogen interactions in European forests and semi-natural vegetation. Part II: Untangling climatic, edaphic, management and nitrogen deposition effects on carbon sequestration potentials'

by C.R. Flechard et al., Biogeosciences, 2020



Commenté [CF1]: Fig. added to Supplement, moved from the Supplement of the Part I paper

Figure S1. Historical N deposition over the 20th century used as inputs to the BASFOR model. The relative temporal evolution of N_{dep} was assumed to be identical for all sites and was derived from Fig.3, p72, in van Oijen et al. (2008).



Commenté [CF2]: Fig. added to Supplement, moved from the Supplement of the Part I paper

Figure S2. Reconstructed time courses of stand density, driven by thinning events over the lifetimes of CEIP/NEU forest monitoring sites, with DBF, ENF(1-7: spruce), ENF(8-18: pine), EBF and MF shown as different colours. Actual (observed and reported) tree density is shown by larger symbols on each time series, while small symbols indicate values reconstructed on the basis of the default density/thinning scheme ("ThinDef", black symbols, illustration for a 100-yr old forest in 2010) adopted from Cameron et al. (2013).

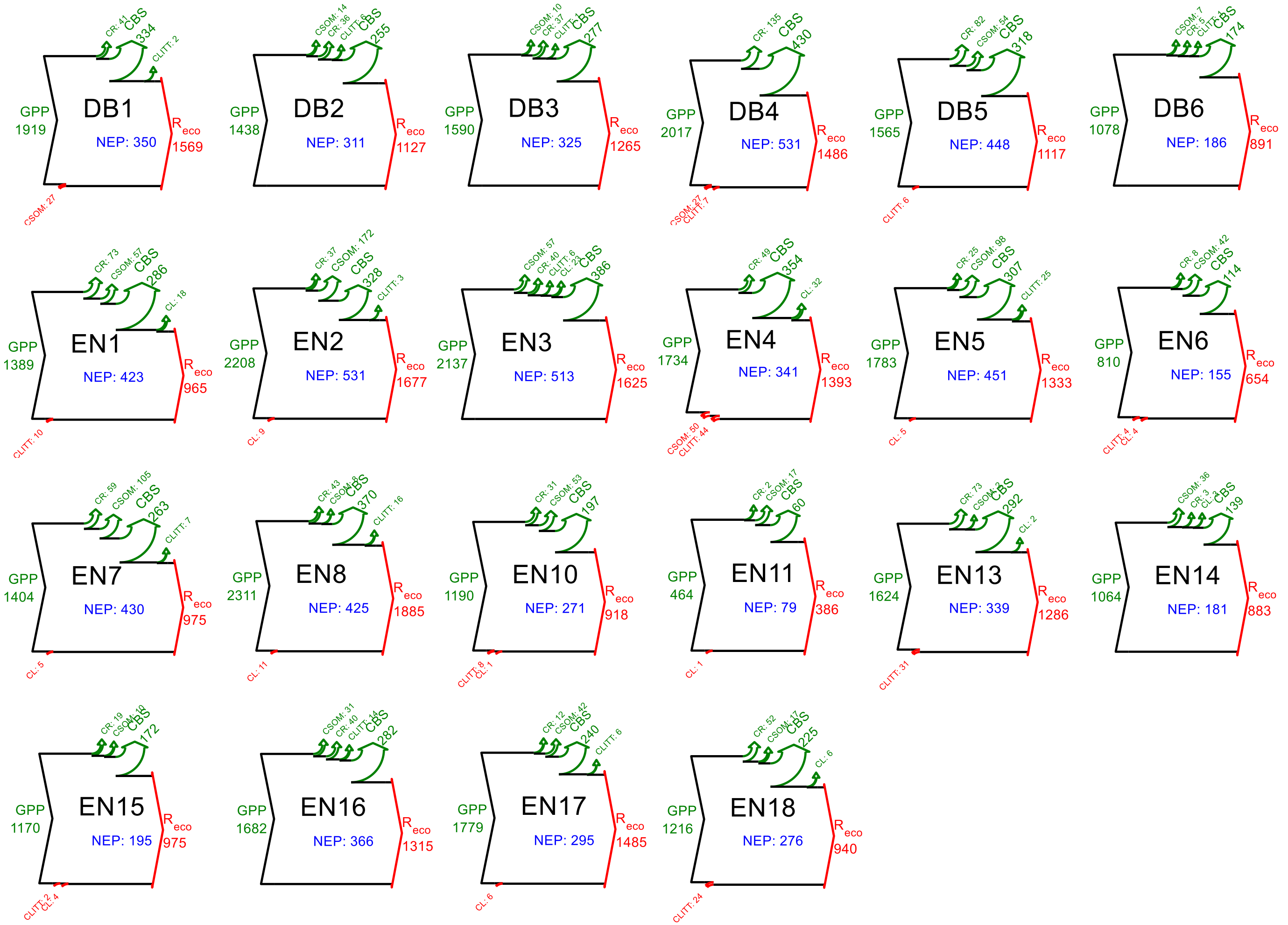


Figure S3. Modelled (BASFOR) partitioning of C fluxes at deciduous broadleaf (DB) and coniferous evergreen needleleaf (EN) forests, and associated changes in C pools in soil organic matter (CSOM), roots (CR), litter layers (CLITT), branches and stems (CBS) and leaves (CL) (units g (C) m^{-2} yr^{-1}). The simulations were run over the most recent 5-year period which did not include any thinning event ('5-yr' in the text). In this case (no disturbance, no export), NEP = NECB = d(CSOM+CR+CLITT+CBS+CL) / dt. Green indicates ecosystem C gain (photosynthesis and C pool increase), red denotes ecosystem C loss (respiration and C pool decrease). The sizes of the Sankey plots are not proportional to the C fluxes of the different study sites.

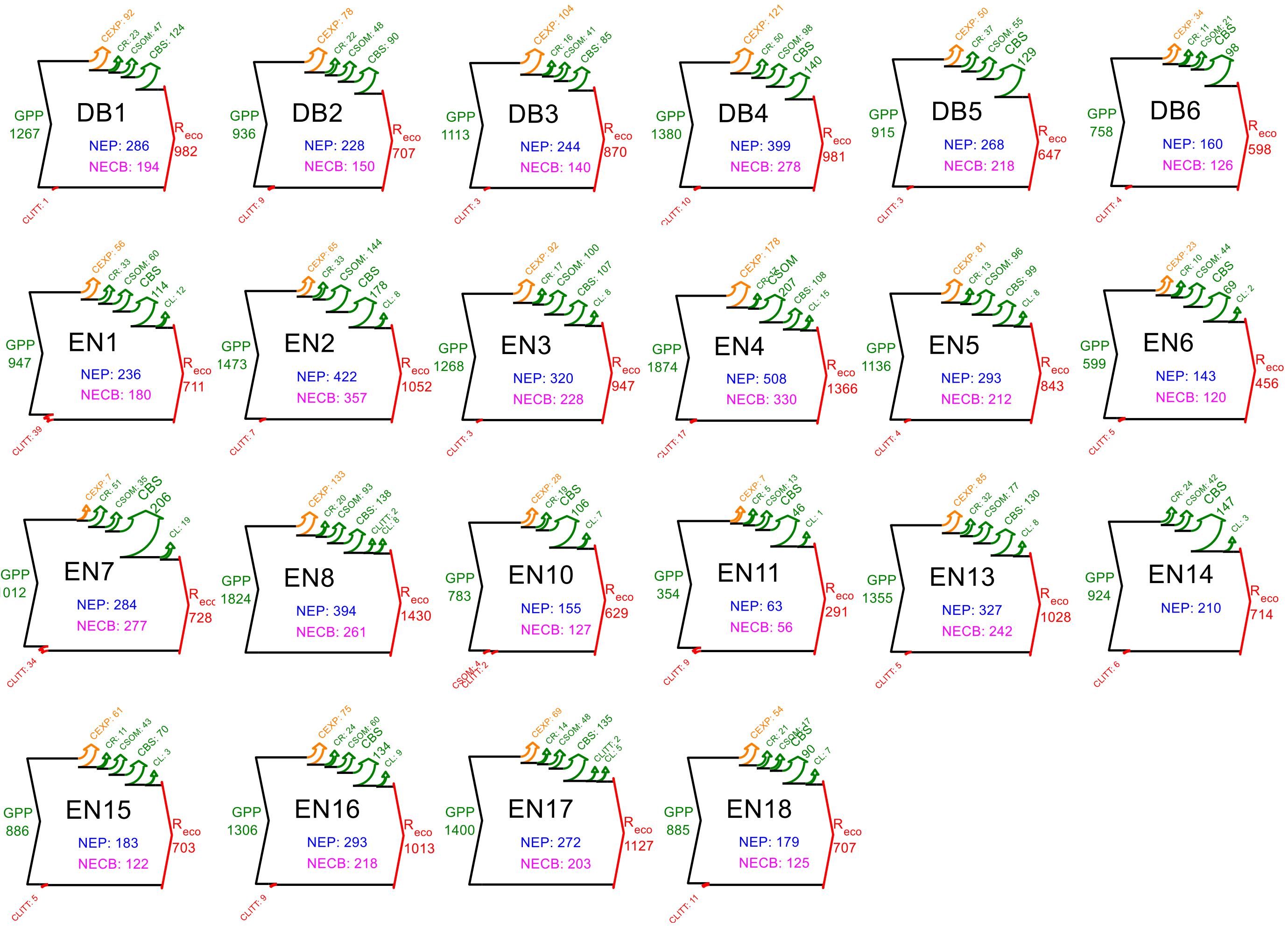


Figure S4. Same as Fig. S3, but simulated over the whole time period since the forest was established (‘lifetime’ in the text), i.e. including C exports (CEXP) through all thinning/management events (units g (C) m^{-2} yr^{-1}). In this case, NEP - CEXP = NECB = d(CSOM+CR+CLITT+CBS+CL) / dt. The sizes of the Sankey plots are not proportional to the C fluxes of the different study sites.

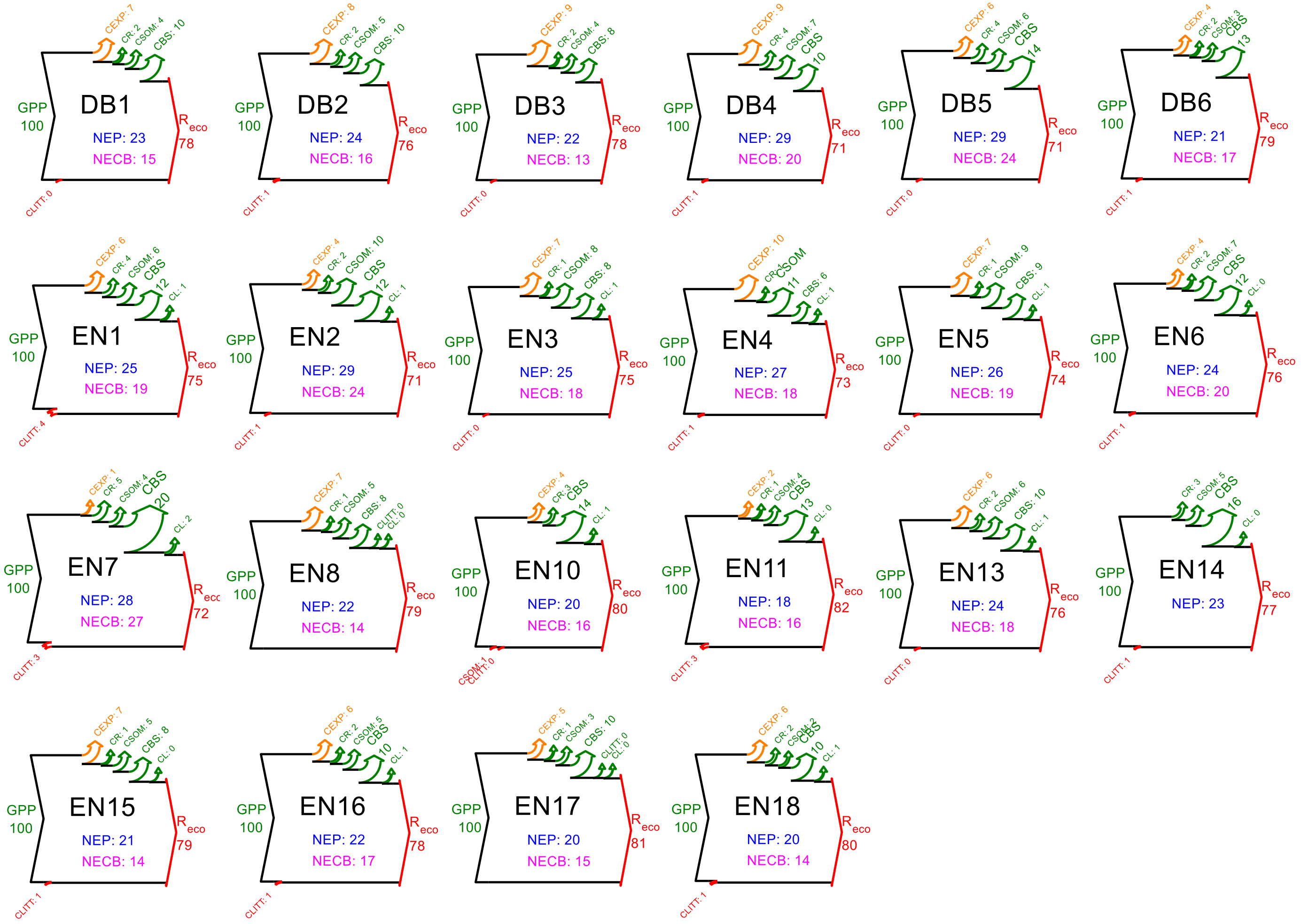


Figure S5. Same as Fig. S4, but normalized to the mean lifetime GPP of each site (units % GPP). The NECB percentage value corresponds to the lifetime carbon sequestration efficiency (CSE). The sizes of the Sankey plots are not proportional to the C fluxes of the different study sites.

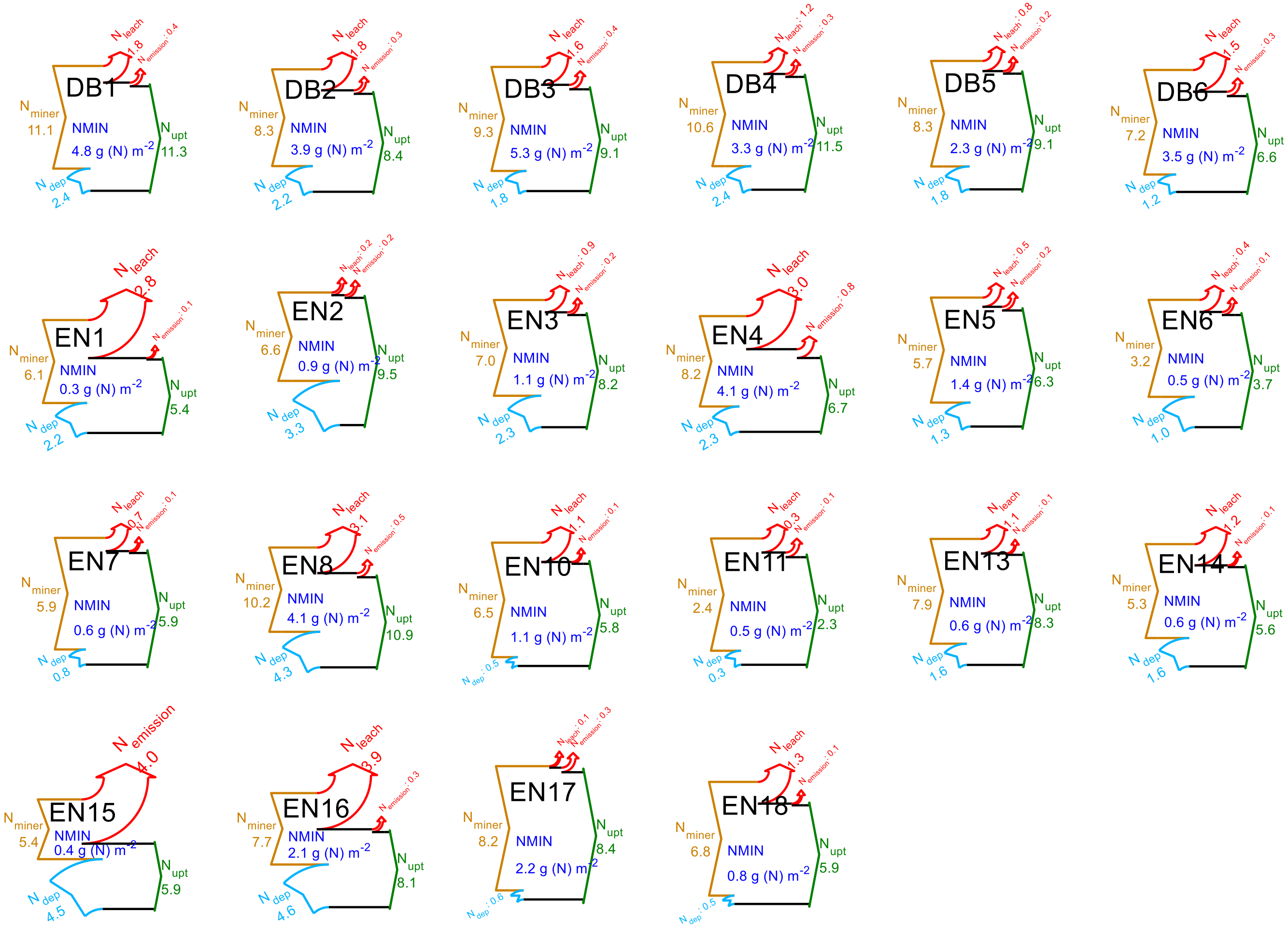


Figure S6. Modelled (BASFOR) nitrogen budgets at deciduous broadleaf (DB) and coniferous evergreen needleleaf (EN) forests. The simulations were run over the most recent 5-year period which did not include any thinning event ('5-yr' in the text). The data show ecosystem SOM mineralisation (N_{miner}) and atmospheric Nr deposition (N_{dep}) (together making up N_{supply}), balanced by vegetation uptake (N_{upt}) and the sum of losses as dissolved N (N_{leach}) and gaseous NO + N_2O ($N_{emission}$) (units: g (N) m^{-2} yr^{-1}). NMIN indicates the mean size of the soil inorganic N pool (g (N) m^{-2}) over the modelling period. The sizes of the Sankey plots are not proportional to the N fluxes of the different study sites.

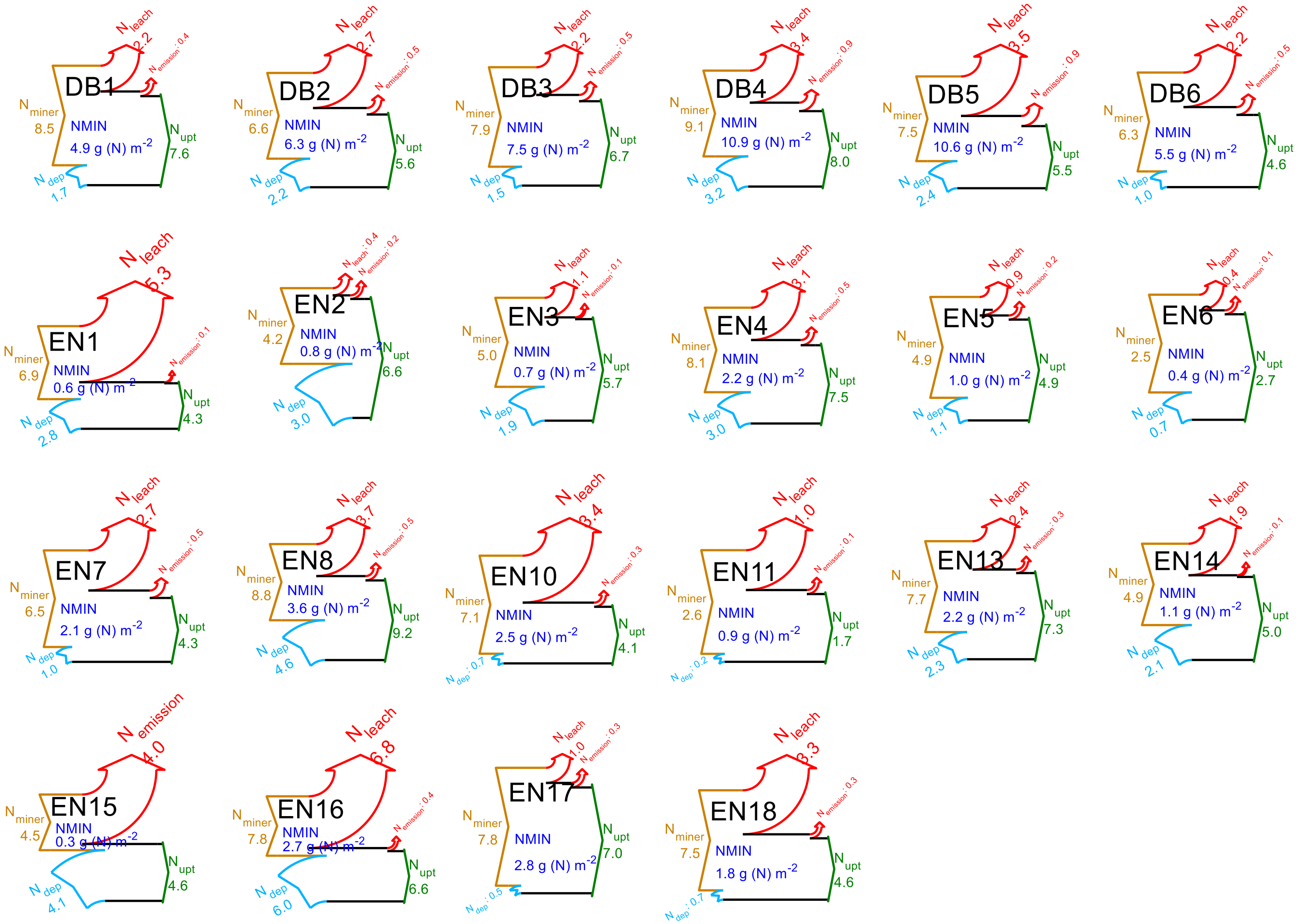


Figure S7. Same as Fig. S6, but simulated over the whole time period since the forest was established (‘lifetime’ in the text) (units: g (N) m^{-2} yr^{-1}, except for NMIN). The sizes of the Sankey plots are not proportional to the N fluxes of the different study sites.

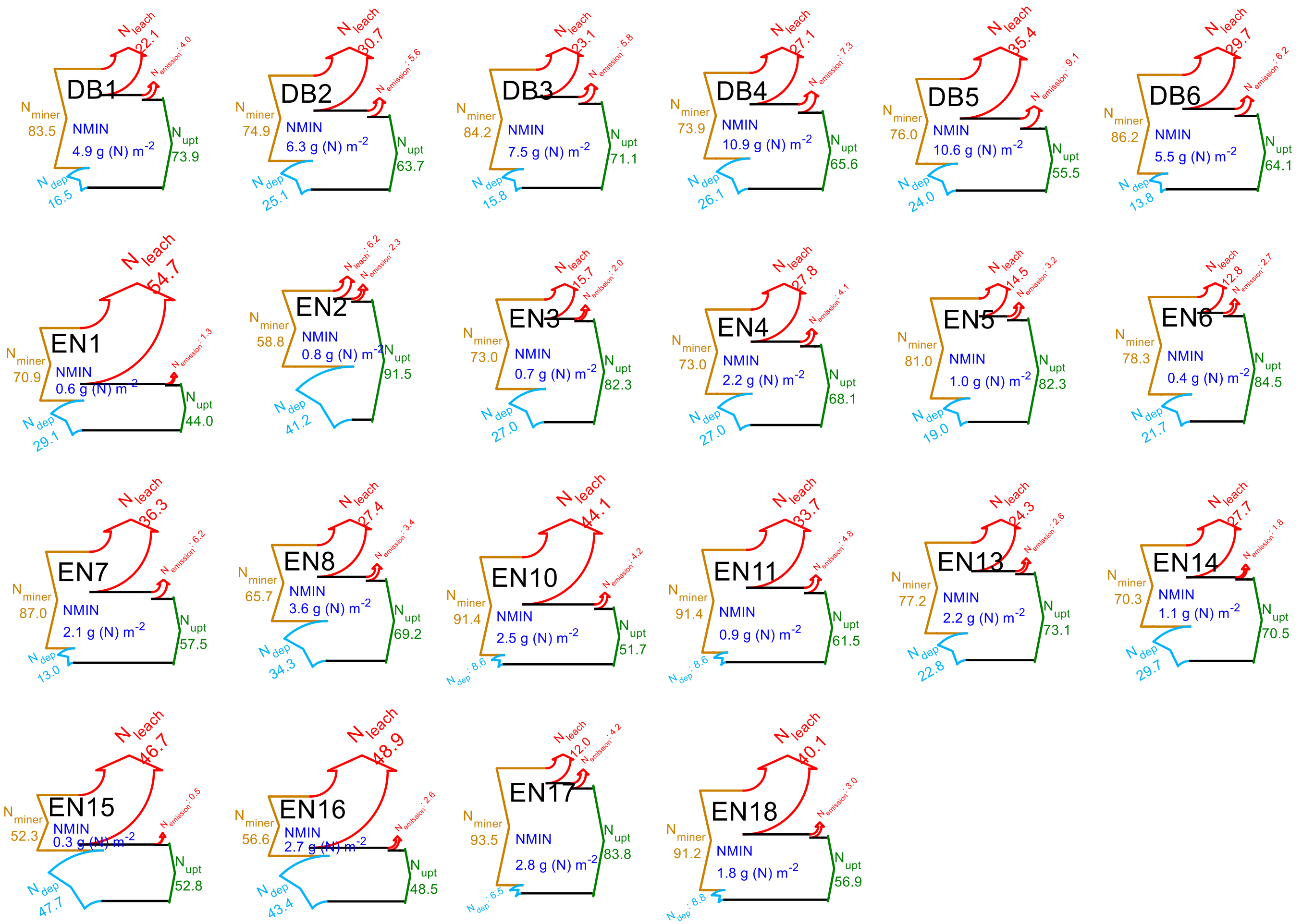


Figure S8. Same as Fig. S7, but fluxes are normalized and expressed as % of total N_{supply} (= N_{miner} + N_{dep} = 100). The N uptake percentage value corresponds to the lifetime nitrogen uptake efficiency (NUPE). The sizes of the Sankey plots are not proportional to the N fluxes of the different study sites.

Commenté [CF3]: Changed NUE to NUPE

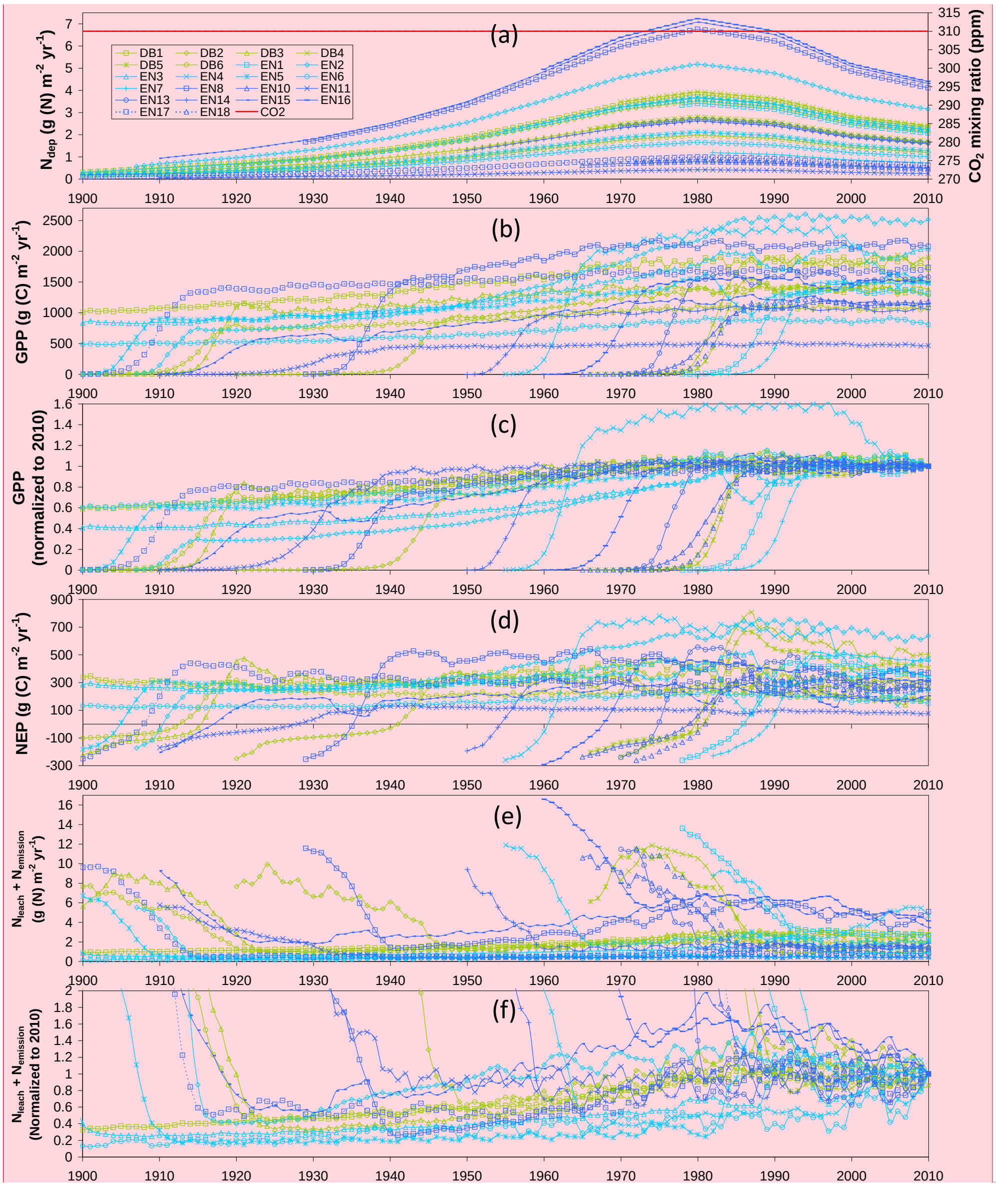


Figure S9. Alternative BASFOR model scenario using a constant CO$_2$ mixing ratio of 310 ppm through the entire modelling period (a), showing simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching and gaseous emissions, and (f) total N losses normalized to 2010.

Commenté [CF4]: Figures S9-S11 added to Supplement to show sensitivity of model results to CO$_2$ and N_{dep}

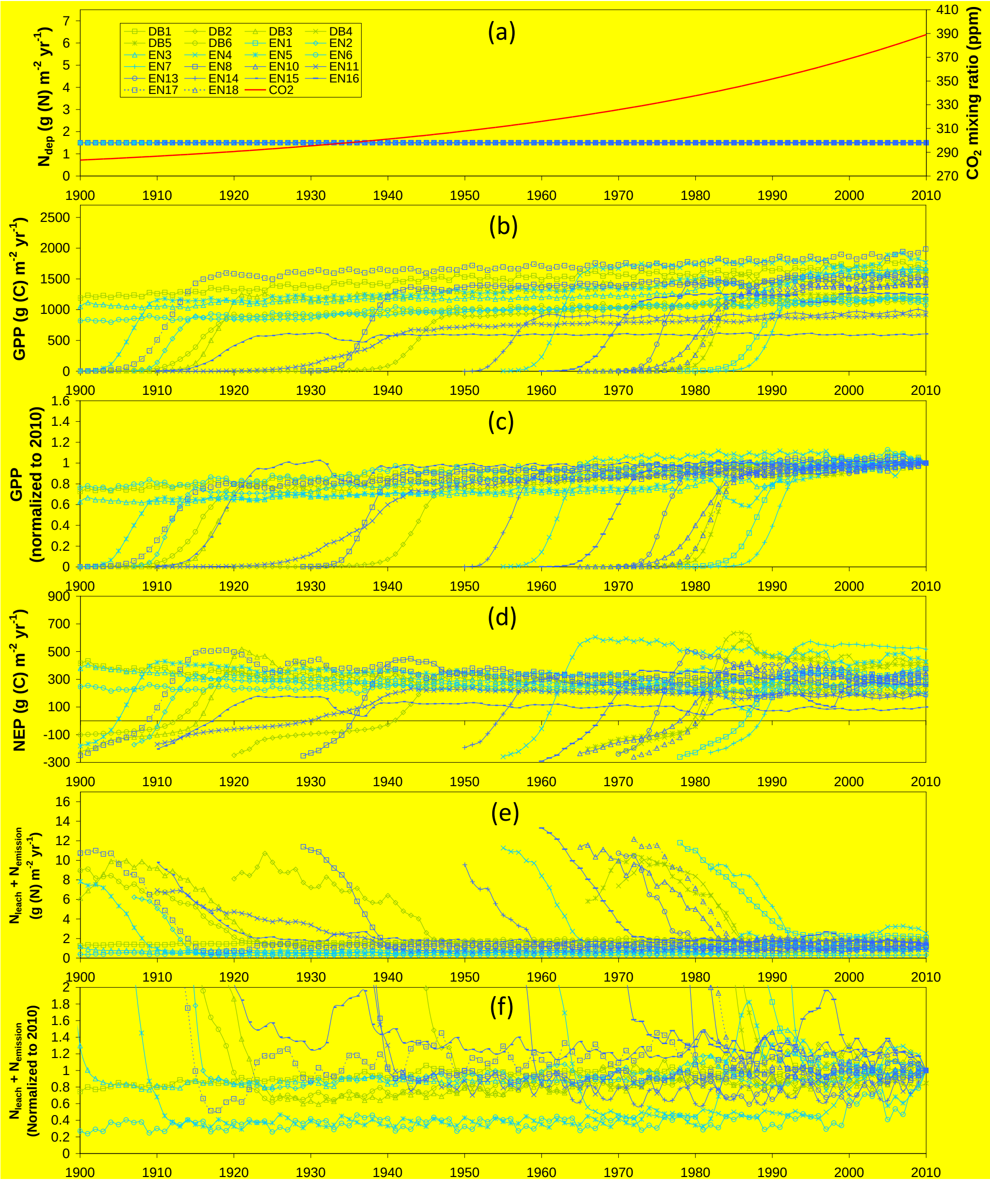


Figure S10. Alternative model scenario using a constant N_{dep} level of 1.5 g (N) m^{-2} yr^{-1} at all sites through the entire modelling period (a), showing simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching and gaseous emissions, and (f) total N losses normalized to 2010.

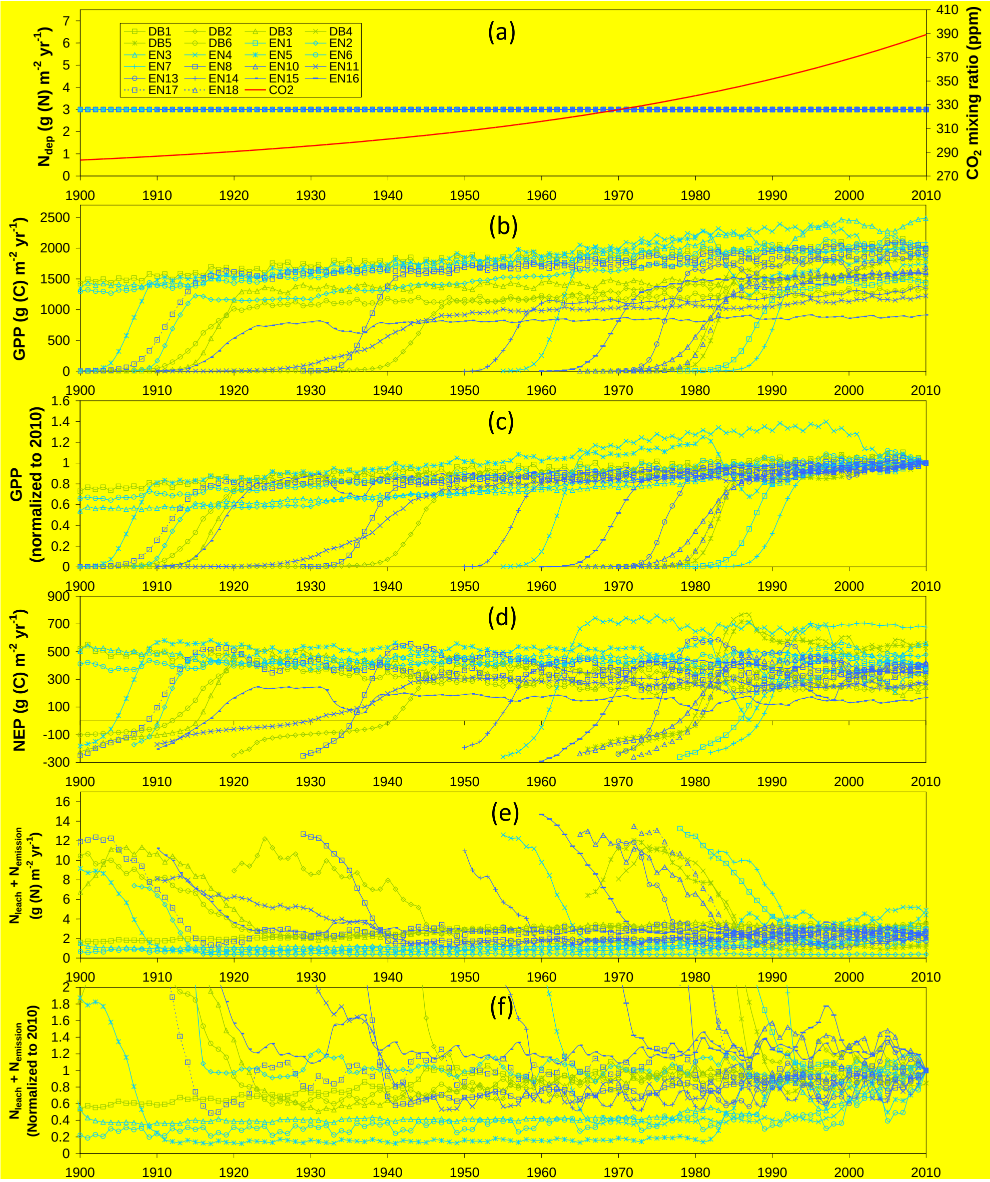


Figure S11. Alternative model scenario using a constant N_{dep} level of 3.0 g (N) m^{-2} yr^{-1} at all sites through the entire modelling period (a), showing simulations of (b) gross primary productivity (GPP), (c) GPP normalized to the 2010 value, (d) net ecosystem productivity (NEP), (e) total N losses by leaching and gaseous emissions, and (f) total N losses normalized to 2010.

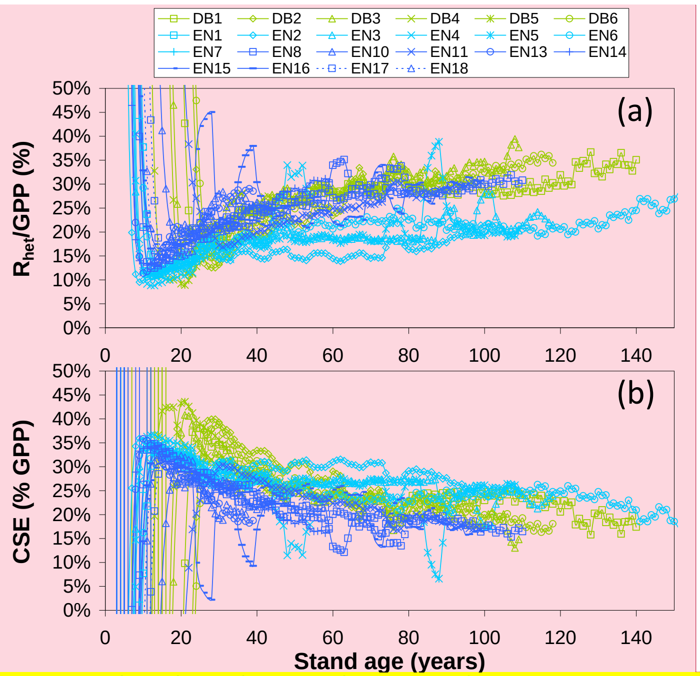


Figure S12. Modelled time courses for all forests of the study of (a) the ratio of heterotrophic respiration (R_{het}) to gross primary productivity (GPP) and (b) the carbon sequestration efficiency (CSE = NEP/GPP). Short term excursions are related to thinning events.

Commenté [CF5]: Figure added to Supplement to illustrate relative changes in heterotrophic respiration and GPP over time

Table S1. Overview of ecosystem and climatic characteristics and inter-annual mean ecosystem/atmosphere exchange fluxes for forest and semi-natural short vegetation sites in the CEIP-NEU network. Only the 22 sites highligthed in blue were included in the meta-modelling study (see text for details).

Site Name	Location, Country	PFT (1) Short name	Dominant vegetation	Forest age (2010)	H_{max}(2) m	LAI_{max}(3) m^2 m^{-2}	Lat. °N	Long. °E	Elevation m amsl(4)	MAT(5) °C	MAP(6) mm	N_{dep}(7) g N m^{-2} yr^{-1}	GPP(8) g C m^{-2} yr^{-1}	R_{eco}(9) g C m^{-2} yr^{-1}	NEP(10) g C m^{-2} yr^{-1}
DE-Hai	Hainich, Germany	DB1	*Fagus sylvatica*	142	23	4.0	51.079	10.452	430	8.4	775	2.3	1553	1074	479
DK-Sor	Sorø, Denmark	DB2	*Fagus sylvatica*	91	31	4.6	55.487	11.646	40	8.9	730	2.2	1883	1581	301
FR-Fon	Fontainebleau-Barbeau, France	DB3	*Quercus petraea*	111	28	5.1	48.476	2.780	92	11.0	690	1.7	1850	1185	665
FR-Fgs	Fougères, France	DB4	*Fagus sylvatica*	41	20	6.0	48.383	-1.185	140	10.3	900	2.4	1725	1316	409
FR-Hes	Hesse, France	DB5	*Fagus sylvatica*	45	16	6.7	48.674	7.066	300	10.2	975	1.7	1634	1187	446
IT-Col	Collelongo, Italy	DB6	*Fagus sylvatica*	120	22	5.7	41.849	13.588	1560	7.2	1140	1.2	1425	776	650
CZ-BK1	Bily Kriz, Czech Rep.	EN1	*Picea abies*	33	13	9.8	49.503	18.538	908	7.8	1200	2.1	1548	767	781
DE-Hoe	Höglwald, Germany	EN2	*Picea abies*	104	35	6.3	48.300	11.100	540	8.9	870	3.2	1856	1229	627
DE-Tha	Tharandt, Germany	EN3	*Picea abies*	120	27	6.7	50.964	13.567	380	8.8	820	2.3	1997	1396	601
DE-Wet	Wetzstein, Germany	EN4	*Picea abies*	56	22	7.1	50.453	11.458	785	6.6	950	2.2	1809	1767	43
IT-Ren	Renon, Italy	EN5	*Picea abies*	111	29	5.1	46.588	11.435	1730	4.6	1010	1.3	1353	528	826
RU-Fyo	Fyodorovskoye, Russia	EN6	*Picea abies*	190	21	2.8	56.462	32.922	265	5.3	711	1.0	1488	1559	-70
UK-Gri	Griffin, UK	EN7	*Picea sitchensis*	29	12	6.5	56.617	-3.800	340	7.7	1200	0.7	989	677	311
BE-Bra	Brasschaat, Belgium	EN8	*Pinus sylvestris*	82	21	1.9	51.309	4.521	16	10.8	850	4.1	1272	1149	123
ES-ES1	El Saler, Spain	EN9	*Pinus halepensis*	111	10	2.6	39.346	-0.319	5	17.6	551	2.1	1552	960	593
FI-Hyy	Hyytiälä, Finland	EN10	*Pinus sylvestris*	48	18	3.4	61.848	24.295	181	3.8	709	0.5	1114	845	268
FI-Sod	Sodankylä, Finland	EN11	*Pinus sylvestris*	100	13	1.2	67.362	26.638	180	-0.4	527	0.3	551	598	-47
FR-Bil	Bilos, France	EN12	*Pinus pinaster*	9	4	0.5	44.522	-0.896	50	12.4	930	0.8	1178	989	189
FR-LBr	Le Bray, France	EN13	*Pinus pinaster*	41	22	1.9	44.717	-0.769	61	12.9	972	1.6	1906	1479	427
IT-SRo	San Rossore, Italy	EN14	*Pinus pinaster*	61	18	4.0	43.728	10.284	4	14.9	920	1.6	2256	1702	554
NL-Loo	Loobos, Netherlands	EN15	*Pinus sylvestris*	101	18	1.5	52.168	5.744	25	10.0	786	4.2	1617	1141	476
NL-Spe	Speulderbos, Netherlands	EN16	*Pseudotsuga menziesii*	51	32	7.5	52.252	5.691	52	10.0	834	4.3	1416	1015	401
SE-Nor	Norunda, Sweden	EN17	*Pinus sylvestris*	112	28	4.6	60.083	17.467	45	6.8	527	0.6	1414	1356	58
SE-Sk2	Skyttorp, Sweden	EN18	*Pinus sylvestris*	39	16	3.2	60.129	17.840	55	7.4	527	0.5	1235	953	282
ES-LMa	Las Majadas, Spain	EB1	*Quercus ilex*	111	8	0.6	39.941	-5.773	258	16.1	528	0.9	1091	958	133
FR-Pue	Puechabon, France	EB2	*Quercus ilex*	69	6	2.9	43.741	3.596	270	13.7	872	1.1	1309	1030	279
IT-Ro2	Roccarespampani, Italy	EB3	*Quercus cerris*	21	16	3.8	42.390	11.921	224	15.7	876	1.8	1707	886	821
PT-Esp	Espirra, Portugal	EB4	*Eucalyptus globulus*	25	20	2.7	38.639	-8.602	95	16.1	709	1.2	1473	1163	311
PT-Mi1	Mitra, Portugal	EB5	*Quercus ilex, Quercus suber*	91	8	3.4	38.541	-8.000	264	14.5	665	0.9	870	817	53
BE-Vie	Vielsalm, Belgium	MF1	*Fagus sylvatica, Pseudotsuga menziesii*	86	30	5.1	50.305	5.997	450	8.1	1000	1.7	1792	1247	545
CH-Lae	Lägeren, Switzerland	MF2	*Fagus sylvatica, Picea abies*	111	30	3.6	47.478	8.365	689	7.7	1100	2.2	1448	757	692
DE-Meh	Mehrstedt, Germany	SN1	Afforestated grassland	n.a.	0.5	2.9	51.276	10.657	293	9.1	547	1.5	1171	1175	-4
ES-VDA	Vall d'Alinya, Spain	SN2	Upland grassland	n.a.	0.1	1.4	42.152	1.448	1765	6.4	1064	1.2	669	528	140
FI-Lom	Lompolojänkkä, Finland	SN3	Peatland	n.a.	0.4	1.0	67.998	24.209	269	-1.0	521	0.1	377	345	32
HU-Bug	Bugac, Hungary	SN4	Semi-arid grassland	n.a.	0.5	4.7	46.692	19.602	111	10.7	500	1.4	1044	918	126
IT-Amp	Amplero, Italy	SN5	Upland grassland	n.a.	0.4	2.5	41.904	13.605	884	9.8	1365	0.9	1241	1028	213
IT-MBo	Monte Bondone, Italy	SN6	Upland grassland	n.a.	0.3	2.5	46.029	11.083	1550	5.1	1189	1.7	1435	1347	89
NL-Hor	Horstemeer, Netherlands	SN7	Peatland	n.a.	2.5	6.9	52.029	5.068	-2	10.8	800	3.1	1584	1224	361
PL-wet	POLWET/Rzecin, Poland	SN8	Wetland (reeds, sedges, mosses)	n.a.	2.1	4.9	52.762	16.309	54	8.5	550	1.4	937	642	295
UK-AMo	Auchencorth Moss, UK	SN9	Peatland	n.a.	0.6	2.1	55.792	-3.239	270	7.6	1165	0.8	786	705	81

(1) PFT (plant functional types): DB: deciduous broadleaf forest; EN: evergreen needleleaf coniferous forest; EB: evergreen broadleaf Mediterranean forest; MF: mixed deciduous/coniferous forest; SN: short semi-natural, including moorland, peatland, shrubland and unimproved/upland grassland; (2) maximum canopy height; (3) maximum leaf area index, defined as 1-sided or half of total; (4) above mean sea level; (5) mean annual temperature; (6) mean annual precipitation; (7) nitrogen deposition; (8) gross primary productivity; (9) ecosystem respiration; (10) net ecosystem productivity; n.a.: not available/ not applicable.

Table S2. Procedure for the calculation of climate / soil standardization factors (f_{CLIM} or f_{SOIL}) through BASFOR meta-modelling for the n=22 forest sites. The indices i and j stand for the site being modelled (i = 1..n), and for the scenarios being applied for climate data or for soil parameters (j=1..n), respectively. See main text and Eqs (10-15) for details.

GPP(i,j)	Site modelled i=1	i=2	i=3	i=…	i=n-1	i=n
Scenario j=1	GPP(1,1) = GPP$_{base}$(1)	GPP(2,1)	GPP(3,1)	…	GPP(n-1,1)	GPP(n,1)
j=2	GPP(1,2)	GPP(2,2) = GPP$_{base}$(2)	GPP(3,2)	…	GPP(n-1,2)	GPP(n,2)
j=3	GPP(1,3)	GPP(2,3)	GPP(3,3) = GPP$_{base}$(3)	…	GPP(n-1,3)	GPP(n,3)
j=…	…	…	…	GPP(i=j) = GPP$_{base}$(i)	…	…
j=n-1	GPP(1,n-1)	GPP(2,n-1)	GPP(3,n-1)	…	GPP(n-1,n-1) = GPP$_{base}$(n-1)	GPP(n,n-1)
j=n	GPP(1,n)	GPP(2,n)	GPP(3,n)	…	GPP(n-1,n)	GPP(n,n) = GPP$_{base}$(n)

X(i,j) = GPP(i,j) / GPP$_{base}$(i)	Site modelled i=1	i=2	i=3	i=…	i=n-1	i=n	**Mean $\overline{\mathbf{X(j)}}$**
Scenario j=1	1	GPP(2,1) / GPP$_{base}$(2)	GPP(3,1) / GPP$_{base}$(3)	…	GPP(n-1,1) / GPP$_{base}$(n-1)	GPP(n,1) / GPP$_{base}$(n)	$\overline{\mathbf{X(1)}}$
j=2	GPP(1,2) / GPP$_{base}$(1)	1	GPP(3,2) / GPP$_{base}$(3)	…	GPP(n-1,2) / GPP$_{base}$(n-1)	GPP(n,2) / GPP$_{base}$(n)	$\overline{\mathbf{X(2)}}$
j=3	GPP(1,3) / GPP$_{base}$(1)	GPP(2,3) / GPP$_{base}$(2)	1	…	GPP(n-1,3) / GPP$_{base}$(n-1)	GPP(n,3) / GPP$_{base}$(n)	$\overline{\mathbf{X(3)}}$
j=…	…	…	…	1	…	…	**…**
j=n-1	GPP(1,n-1) / GPP$_{base}$(1)	GPP(2,n-1) / GPP$_{base}$(2)	GPP(3,n-1) / GPP$_{base}$(3)	…	1	GPP(n,n-1) / GPP$_{base}$(n)	$\overline{\mathbf{X(n-1)}}$
j=n	GPP(1,n) / GPP$_{base}$(1)	GPP(2,n) / GPP$_{base}$(2)	GPP(3,n) / GPP$_{base}$(3)	…	GPP(n-1,n) / GPP$_{base}$(n-1)	1	$\overline{\mathbf{X(n)}}$

X$_{norm}$(i,j) = X(i,j) / $\overline{\mathbf{X(j)}}$	Site modelled i=1	i=2	i=3	i=…	i=n-1	i=n
Scenario j=1	X(1,1) / X(1)	X(2,1) / X(1)	X(3,1) / X(1)	...	X(n-1,1) / X(1)	X(n,1) / X(1)
j=2	X(1,2) / X(2)	X(2,2) / X(2)	X(3,2) / X(2)	...	X(n-1,2) / X(2)	X(n,2) / X(2)
j=3	X(1,3) / X(3)	X(2,3) / X(3)	X(3,3) / X(3)	...	X(n-1,3) / X(3)	X(n,3) / X(3)
j=…	...	...	...	X(i,j) / X(j)	...	...
j=n-1	X(1,n-1) / X(n-1)	X(2,n-1) / X(n-1)	X(3,n-1) / X(n-1)	...	X(n-1,n-1) / X(n-1)	X(n,n-1) / X(n-1)
j=n	X(1,n) / X(n)	X(2,n) / X(n)	X(3,n) / X(n)	...	X(n-1,n) / X(n)	X(n,n) / X(n)
f(i) = mean $\overline{\boldsymbol{X_{norm}}\mathbf{(i)}}$	**f(1) = $\overline{\boldsymbol{X_{norm}}\mathbf{(1)}}$**	**f(2) = $\overline{\boldsymbol{X_{norm}}\mathbf{(2)}}$**	**f(3) = $\overline{\boldsymbol{X_{norm}}\mathbf{(3)}}$**	**…**	**f(n-1) = $\overline{\boldsymbol{X_{norm}}\mathbf{(n-1)}}$**	**f(n) = $\overline{\boldsymbol{X_{norm}}\mathbf{(n)}}$**

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