Biogeosciences Discussion BG_2019-333

"Carbon / nitrogen interactions in European forests and semi-natural vegetation. Part I: Fluxes and budgets of carbon, nitrogen and greenhouse gases from ecosystem monitoring and modelling" 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 and for their constructive comments, which have helped improve the manuscript. For clarity's sake, we have provided our point-by-point **responses to each comment in blue**, and provided **changes to the manuscript text in green**.

Anonymous Referee #1

General Comment This is a very well written paper in the suite of other papers on N deposition at the European scale lead by the principal author. In fact, this paper acts as a "prequel" of the companion paper, the Part II dedicated to untangling climatic, edaphic, management and N deposition effects on C sequestration. A previous paper to Part II is important since uncertainties and gaps of knowledge associated to the different components of the N and C cycles in terrestrial ecosystems need to be examined previously to the attempt of disentangling. Therefore, in this paper, to evaluate the uncertainties and gaps in the estimates in N and C budgets, the authors have made a remarkable effort of gathering N and C data from 31 forests and 9 seminatural ecosystems extended over Europe and covering a wide span of climates, from Mediterranean to boreal. To constrain the N budget they have taken advantage of local measurements of dry and wet deposition at specific sites from the NEU (NitroEurope) database, complemented with the use of deposition models (EMEP model) in some cases. Loss of N by nitrate leaching and by gaseous emissions have been estimated by measurements in some sites and modelling when no measurements were available. For the C budget, data were mostly obtained from eddy covariance sites within the CarboEurope Integrated Project (CEIP) combined with laboratory bioassays and literature mining. The results of this big effort of compilation constitute an important contribution to the evaluation of N deposition on C sequestration at a European scale, by critically evaluating the uncertainties in the quantification of some of the drivers. Also, it calls for attention to neglected fluxes that might have a considerable role in the budgets, e.g. N2 emissions by denitrification. The paper is well written, well documented, scientifically sound, and it fits the scope of Biogeosciences Discussions, so I recommend it for publication with only very minor changes.

We are grateful to Referee #1 for this positive assessment of our paper.

Specific Comments

Abstract The abstract summarizes the main findings, so it is very important to give accurate figures. In this sense, I suggest to review the sentence in line 110, since from Fig. 3F one can see a different range of values of % N losses to total N dep than those reported in the text (10-35% at Ndep below 1 gN m-1 yr-1 and 35-80% at Ndep above 3 gN m-1 yr-1 in the Figure). This sentence is followed with a consideration that 1/3 of the sites might be in a state of early – advanced N saturation. But, from Fig 3F, I deduce that one third of the sites result from considering a threshold value of 2 gN m-1 yr-1. I suggest rewriting this paragraph, also including a suggested of Ndep that might indicate early N saturation.

We have provided the mean value, as well as the range, for inorganic N losses at N_{dep} below 1 g N m^{-2} yr^{-1} and N_{dep} above 3 g N m^{-2} yr^{-1} (see below). However, as discussed in Section 4.3.2, there are several definitions of N saturation, which potentially lead to different thresholds, while the presence of different tree species, of different age classes and growing on very different soils, makes it difficult to pinpoint exactly at what N_{dep} level N saturation begins, and what level corresponds to advanced N saturation. This is why we were cautious and admittedly a little vague ('perhaps one third') in the second part of the sentence. We stand by the decision to remain cautious about N_{dep} thresholds for saturation in the abstract (while leaving the debate open in the discussion). The paragraph was rephrased thus:

'…Nitrogen losses in the form of NO, N_2O and especially NO_3^- were on average 27% (range 6-54%) of N_{dep} at sites with N_{dep} < 1 g (N) m^{-2} yr^{-1}, versus 65% (range 35-85%) for N_{dep} > 3 g (N) m^{-2} yr^{-1}. Such large levels of N_r loss likely indicate that different stages of N saturation occurred at a number of sites. The joint analysis of the C and N budgets provided further hints that N saturation could be detected in altered patterns of forest growth. Net ecosystem productivity increased with N_r deposition up to 2-2.5 g (N) m^{-2} yr^{-1}, …'

Introduction I like it very much. In line 175, wetlands might be also included for DOC leaching.

Yes, wetlands should in fact be mentioned first, before grasslands and croplands. A mention and reference was added: '…leaching can also be significant, especially for **wetlands (Dinsmore et al., 2010) and also** grassland and cropland ecosystems (Kindler et al., 2011; Gielen et al., 2011). This is relevant for…'

Methods For dry deposition, the inferential method is based in the same 4 models as in Flechard et al (2011). However, it is not clear to me whether the retained value is the averaged estimate from the 4 models, can you clarify?

Later on (line 533) it is mentioned that DD is calculated as ensemble average of 4 inferential models, but this should be stated and explained in Methods.

Yes, the ensemble average of the same 4 models was used for dry deposition. This is already stated in section 2.2.1, lines 264-265: '*…we tried here to minimise such uncertainties by using the ensemble average dry deposition predicted by four different models, as in Flechard et al. (2011).*'

For wet deposition, an estimated NO3 and NH4 deposition was attributed to every one of 40 sites through kriging interpolation of EMEP and ICP-Forest data. Furthermore, 13 sites were provided with BD samplers for 3 years so that BD Ndep was actually measured in these sites, and six more sites already were equipped with BD or WD collectors. Can you comment here on how well did compare the kriging estimates to the actual measures?

This issue is explored in Results section 3.1.1, lines 543-547. We have written that '… *By contrast, wet deposition was generally reasonably consistent between the different data sources for inorganic Nr (in situ bulk or wet-only measurement, kriging of monitoring network data, EMEP model output). For the 18 sites where all three sources of data were available, the mean CV of the three estimates was 21% (range 2%-56%, with 15 CV values out of 18 below 30%), and the mean (± 95% conf. int.) wet deposition estimates across the 18 sites were 0.63 ±0.14, 0.64 ±0.15 and 0.68 ±0.16 g (N) m-2 yr-1 for the three methods, respectively (Fig. S2), showing no systematic bias between methods…*' Figure S2 in the supplement shows the comparison between precipitation samplers, EMEP model, and spatial interpolation of network data.

When calculating losses by leaching, it is mentioned that lysimeters were used to obtain soil N (or DOC) concentrations that were combined with a hydrological drainage model to derive the export fluxes. Can you explain better this hydrological model? Was it possible, in any one of the sites of the survey, compare results from N and C exports calculated using the hydrological drainage model and from actual water runoff at gauged sites? I understand that this is not the main focus of the paper; however, just to know how the two approaches estimate losses can be of interest.

Several approaches were used to quantify dissolved C and N losses, and the sampling methods and hydrological models were site- or paper-specific. Unfortunately, there was no comparison anywhere of C/N exports estimated by actual water runoff and by lysimeter/suction cup setups. It would be difficult to describe each hydrological model in any detail; however, the text (lines 410-411) does provide references to the papers on DIC/DOC, in which the hydrological approaches are described: Kindler et al., 2011; Gielen et al., 2011; Verstraeten et al., 2014. We have added the following sentence and references to section 2.2.2 (ca line 325), where the method is first mentioned for the case of DIN leaching: "…One-dimensional (1-D) drainage models were based on the soil water balance equation using evapotranspiration, observed precipitation and changes in soil water content (Kindler et al., 2011; Gielen et al., 2011).

Results

In line 548, when considering organic N deposition, it is seen that WSON is a small fraction of Inorganic and organic N deposition. But, can you comment on the possible role of dry WSON (e.g. urea is important in some cases) deposition? Should this also need to be considered in the N budgets? And then, if the dry organic N flux is considered to be relevant, should it be included in Figure 1?

We had indeed omitted to show a conceptual arrow for N_{org} dry deposition in Fig.1; this was included in the revised version. We agree that the dry deposition of organic N_r (ON) is in principle a component of the total N_{dep} budget, but we believe this is a minor fraction of total N_{dep}. Gas-phase organic N_r (e.g. PAN and other organic nitrates, amines) have relatively small ambient concentrations and deposition velocities, as discussed in Flechard et al. (2011), and as shown by EMEP model calculations involving PAN and organic nitrates from isoprene chemistry. However, Kanakidou et al. (2016) suggest that particulate ON largely dominates the atmospheric ON load, and for particles the main atmospheric removal mechanism is through precipitation.

The original article version did mention that dry deposition of organic N_r is not quantified, p19, lines 808-809: '*…incoming organic nitrogen in precipitation (WSON) as well as dry deposition of organic Nr species, not quantified here (Fig. 1)…*' But we have added the following sentence to Section 2.2.1, line 265, to explain why we think this can be considered minor:

'…The dry deposition of atmospheric organic N_r (ON) species not accounted for by the EMEP model (e.g. amines, urea), and not included in DELTA measurements, can contribute a fraction of total N_r deposition. However, Kanakidou et al. (2016) suggest that particulate ON largely dominates the atmospheric ON load, and for particles the main atmospheric removal mechanism is through precipitation. Thus, dry deposition of ON is expected to be much smaller than wet deposition of water soluble organic compounds (see below).'

Additional reference:

Kanakidou, M., Myriokefalitakis, S., Daskalakis, N., Fanourgakis, G.S., Nenes, A., Baker, A.R.,Tsigaridis, K., Mihalopoulos, N.: Past, present, and future atmospheric nitrogen deposition, J. Atmos. Sci., 73, 2039–2047, https://doi.org/10.1175/JAS-D-15-0278.1, 2016.

When commenting on N losses, in line 556 reference is made to Fig 3D to indicate greater losses with N_{dep} above 2 gN m-1 yr-1. It should be specified that this statement is based on measured leaching and Dise's leaching model, but not on BASFOR estimates.

The manuscript does mention, a few lines further down (560-563), that '*… the DIN leaching estimate by BASFOR, shown for comparison on Fig. 3C, was not used in the calculation of total inorganic N losses in Fig. 3D; this is because BASFOR does not simulate N2 loss by denitrification, and thus part of the soil N surplus that would in reality denitrify is assumed to drain, resulting in an over-estimation of the leaching term…*', so we don't believe it is necessary to further stress the point.

Discussion

The N balance is presented in Fig 3D (N losses compared to N inputs) and it is shown that a non-linear fit best describes this relationship. Then the authors argue that above a Ndep of 4 gN m-2 yr-1, N losses might "even exceed " the estimated N deposition, but this should occur when extrapolating the line into a region devoided of data. On the other hand, sites of lower N dep (e.g. EN9) have a leaching loss as close to the Ndep value than other sites with higher deposition. In my opinion, the pattern towards N saturation is best shown in Fig. 3F, when plotting the % of N losses to Ndep. In this plot, to me it is clear that, at Ndep above 2 gN m-2 yr-1, all site mean leaching values are above 35%. The classification in three ranges of depositions (low, intermediate and high) is OK, but as commented in the Abstract, the % ranges of N losses to N_{dep} should be revised (e.g. for Ndep above 3 gN m-2 yr-1, mean loss% ranges from 35 to 80%.

What was meant by '…*N losses might even exceed the estimated N deposition*…' was actually not that N losses would exceed N_{dep} if one extrapolates N_{dep} to 5 or 6 g N m^{-2} yr^{-1} (a region in which we have no data indeed); what we meant was that the error bars on the total inorganic loss term are large when N_{dep} > 4 g N m^{-2} yr^{-1} and the confidence interval overlaps a range where N_{loss} can be larger than N_{dep}.

We have revised the sentence on the ranges of % Nlosses (lines 814-816) using mean values (and the range) for the three classes of N_{dep}, as suggested by the referee:

'…the large range of losses from 6% to 85%, with on average 27% loss (range 6-54%) for N_{dep} < 1 g (N) m^{-2} yr^{-1}, 45% loss (12-78%) for intermediate N_{dep} levels, and 65% loss (35-85%) for N_{dep} > 3 g (N) m^{-2} yr^{-1}…'

Minor corrections

Line 524: large Done

Line 569: include here Fig. 3A after Ndep 2 g m-2 yr-1 Done

Line 577: Fig 3A Done

Line 586: why include here the units in kg ha-1 yr-1, besides g m-2 y-1?

We have deleted the value in kg ha-1 yr-1, we agree this is not needed.

Line 631: This inter-annual peak in LAI, is it the average of various years? What does it mean "peak"?

This is the inter-annual mean value of the annual maximum leaf area index. We have rephrased thus: '…The inter-annual mean value of the annual maximum leaf area index (LAI_{max}) increased from…'

Line 574: better than? Maybe use: provide a good estimate. . .

We are not sure what the referee means here, we do not find any instance of '*better than*' in line 574.

If the Referee means **line 754** instead of 574 (?), we agree we should specify that we compare local depositon estimates to the outputs of a large-scale chemical transport model:

'…*Despite these uncertainties, measuring gas-phase and aerosol Nr concentrations locally* should provide a better estimate of total ecosystem N_r inputs than the outputs of a large-scale chemical transport model…'

Fig 6: for CSOM, r2 = 0.00, but this seems too low given the distribution of points. . .can you revise it?

CSOM is predictably a difficult variable to get right in forest ecosystem modelling, since this depends much less on carbon accumulated during the lifetime of the forest, than on the initial CSOM value at the start of the simulation, about which little is known in most cases. There is however stronger confidence in the change in CSOM over time than in the actual absolute CSOM value at any given time.

To answer the Referee's question, we are not sure how this can be revised. The R^2 is what it is. It is always possible to select data points to improve the correlation, but this is not necessarily helpful in this case.

Fig. 9: include regression, r2 and p in plot A and B. Done

Figures: Identification of sites are generally difficult to read, especially in Figs. 3 and 6

We have increased font size for site labels in these 2 figures, and also moved labels to avoid overlapping. We also now use black instead of grey for site labels in Fig.3, which improves readability.

Anonymous Referee #2

General comments: This article presents estimates of nitrogen, carbon and greenhouse fluxes and budgets at 40 European flux towers compiled from different sources (observations, models, literature). Overall, the article contains an integrated approach to estimate the total nitrogen flux at these sites as complete as possible, accounting for different N pathways. It includes a lot of useful measurements, such as local wet and dry Nr deposition from a collocated measurement network, as well as other in-situ observations (NO, N2O, soil samples etc.). These nitrogen flux estimates are complemented with carbon and greenhouse fluxes, as well as auxiliary information (such as climate variables, forest characteristics, etc.), and as such provide a useful database as a basis for assessment of carbon/nitrogen interactions in European ecosystems, which is also in part included in this article.

Even though the content of this article is useful and interesting, it is very lengthy and it can benefit largely from shortening and restructuring. It contains plenty of information which could be transferred as Supplementary information. The materials and method section is for instance very long (almost 8 pages) and puts a lot of emphasis on some of the different measurements and data sources. I would highly recommend adding a schematic overview of the different data sources at the beginning of this section to make clear what is used and from what source. Also, a couple of subsections may be added to the 'nitrogen fluxes' section to subdivide the text into different components. The section about the BASFOR forest ecosystem model is also unnecessarily lengthy, while the output of this model is only a small part of the discussed results. To make the article more focused, I recommend to greatly shorten this section or even better refer to existing publications.

We acknowledge that this is a long paper. This is because we have endeavoured, here (Part I) and also in Part II of this study (BG_2019-335), to bring together many of the results of two large European-scale projects (CarboEurope and NitroEurope), to investigate the linkages between the carbon cycle, the nitrogen cycle and greenhouse gas budgets, but also the interactions with the water cycle, climate and soil.

In this paper, we wish to demonstrate that significant patterns of interactions can be identified from the joint analysis of the various datasets involved. But, importantly, we also need to make it clear that, while significant advances have been made in measurements and observation networks, in our understanding of processes, and in biogeochemical and ecosystem models, there remain very significant uncertainties in measurements and models. Closing the C and N cycles of ecosystems is an ambitious objective that requires observations of many variables, but also many assumptions and inevitably a dose of modelling, and therefore, a critical assessment of methods and uncertainties is needed, as underlined by the other Referee (#1).

Therefore, this paper (part I of the study) was built on two axes: i) a clear methodological focus on the ways to assemble the main components – and evaluate the uncertainties – of the C and N budgets on the basis of observations, as far as possible; and ii) a scientific focus on the patterns of biogeochemical interactions across the monitoring networks, which need to be identified prior to the assessment of the quantitative links between C sequestration and N deposition (in Part II of the study). The second objective is tightly connected to the first: our understanding of the interactions between the C and N cycles is only as good as our measurements and models are at estimating the C and N cycle components.

We believe that the methodological part of the paper and the discussion of uncertainties in C and N budgets require a detailed description and assessment of the different methods used in compiling the budgets. Moving a large part of Materials and Methods to the supplement (as suggested by Referee #2) would weaken this methodological focus, since uncertainties are directly related – and must be discussed in relation to – the methods that are used. A good example of this is the description of methods for the interpretation of eddy covariance data (section 2.3.1), which Referee #2 suggests shortening. The discussion of uncertainties in the CO_2 budgets and carbon sequestration efficiency (4.2.1) relies on methods being described in some detail, because the assumptions made in EC data post-processing, gap-filling and partitioning are critical for the C budgets derived from the raw data.

Nevertheless, we do agree with Referee #2 that forest ecosystem (BASFOR) modelling plays a secondary role in this paper (Part I of the study). Its main quantitative contribution to the elemental budgets is the simulation of soil NO and N_2O fluxes, and indeed the size of the model description in 2.6 is not in proportion to the actual importance of BASFOR results in the overall paper. Meanwhile, in the companion paper (Part II, BG_2019-335), Referee #1 writes that it is not clear and transparent '*…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…*'.

We have therefore moved the detailed BASFOR model description from Part I to Part II. We now describe the BASFOR model in a few sentences in Part I, and we point to Part II for details of the modelling. This satisfies three objectives: i) the Part I paper is shortened by approximately 1000 words; ii) a more thorough description of BASFOR assumptions, workings and implementation will be found in the Part II paper, where it is more needed; iii) we have redressed the balance in size and contents of the two parts of the study.

In addition, we also agree with Referee #2 that a table summarizing the methods used in the compilation of fluxes would add clarity to the paper and guide the reader through the following sections. This new table (see 'Table 3' on the following page) is now placed in the manuscript at the start of section 2.2 and lists methods and references for each component flux of the C, N and GHG budgets. For each item a small bar diagram shows the percentage of forest and semi-natural sites where measured and modelled data were available.

The results section seems to focus on two major parts: one being the discussion of the resulting nitrogen, carbon and greenhouse fluxes and budgets themselves (and their inter-comparison and validation) and the other being the link between these N and C fluxes and the interpretation. I would recommend separating these two parts as much as possible.

For most of the flux tower sites, the inter-annual carbon fluxes have been presented and published in several previous papers (with key references provided in Table S1), sometimes on a site-by-site basis, sometimes as part of various meta-analyses. We felt it would be superfluous to dwell again on the C budgets by themselves, and that the added value of this paper was to describe the geographical variations in C fluxes in relation to a number of factors including N deposition (hence section 3.2.1). As for N deposition, N losses, and the GHG budgets, which have not all been published previously, they are described in detail by themselves in dedicated sections.

Moreover, the "results" and "discussion" sections seem to be partially intertwined. It would be better to exclusively include discussion points in the latter. Some sections, for instance, the one about "carbon sequestration efficiency", introduce new concepts and results and seem to fit better in the results section (see specific comments below). As for the "discussion" section, it would be more structured if there was a clear separation between the methodological uncertainties used in the estimation of the N, C and greenhouse gas fluxes and the uncertainties related to the interplay of N and C.

We agree that the carbon sequestration efficiency metric should be introduced in Methods (2.3.1), and that the description of CSE results should be moved to results sections 3.2.1 and 3.2.3. We have done this in the revised version.

The uncertainties related to the interplay of N and C are discussed in Section 4.3, while methodological uncertainties in N deposition, N losses, C balance and GHG budgets are discussed in Sections 4.1.1, 4.1.2, 4.2.1 and 4.2.2, respectively, so there is a clear separation of these two types of uncertainties in the discussion.

Table 3. Summary of the main methods used to quantify carbon, nitrogen and greenhouse gas fluxes and budgets for the 31 forests and 9 short semi-natural vegetation sites included in this study. Horizontal bars (green: forests; blue: short semi-natural vegetation) indicate the percentages of study sites with available data (filled bars), or without available data (open bars). See also Supplement Tables S6-S7 for details at individual sites.

Fluxes and budgets	**Components**	**Experimental data (this study)** Methods (selected references)	**Literature and other data mining** Methods (selected references)	**Modelling (this study)** Models (selected references)
Carbon	Net ecosystem exchange (NEE)	Eddy covariance (1)		
	Net ecosystem productivity (NEP)	Gap-filled from NEE (14)		BASFOR (18)
	Gross primary productivity (GPP)	Inferred from NEE (14)		BASFOR (18)
	Ecosystem respiration (R_{eco})	Inferred from NEE (14)		BASFOR (18)
	Soil respiration (R_{soil})	Static/dynamic chambers (12)	Static/dynamic chambers (19)	
	Heterotrophic respiration (R_{het}) Ratio R_{het} / R_{soil}		Root exclusion, trenching, girdling, isotopic methods (19)	BASFOR (18)
	Dissolved organic/inorganic carbon (DIC / DOC) losses	Suction cups (9); peatbog stream sampling (3)	Lysimeter / suction cups (6); weir (8) ; ground- and ditch-water sampling (7)	
	Soil-atmosphere CH_4 fluxes	Static chambers (12) Laboratory soil bioassay (15)	Eddy covariance (10) ; static chambers (7)	
Nitrogen	Atmospheric N_r concentrations	DELTA (17)		EMEP (16)
	Atmospheric dry deposition	Inferential method (5)		EMEP (16)
	Atmospheric wet deposition (Inorganic N_r)	Bulk samplers (2) Wet-only samplers	Regional networks / kriging	EMEP (16)
	Atmospheric wet deposition (wet-soluble organic N_r, WSON)	Bulk samplers (Dämmgen, 2006) Wet-only samplers		
	Throughfall N_r deposition	Throughfall precipitation collectors		
	Dissolved inorganic nitrogen (DIN) losses	Suction cups (9)	Lysimeter / suction cups (11)	IFEF model (4)
	Dissolved organic nitrogen (DON) losses	Suction cups (9)	Lysimeter / suction cups (11)	
	Soil-atmosphere NO fluxes	Dynamic open chambers (12) Laboratory soil bioassay (15)	Dynamic open chambers (13)	BASFOR (18)
	Soil-atmosphere N_2O fluxes	Static chambers (Luo et al., 2012) Laboratory soil bioassay (15)	Static chambers (13)	BASFOR (18)

1 Aubinet et al. (2000) ; 2 Dämmgen (2006) ; 3 Dinsmore et al. (2010) ; 4 Dise et al. (2009) ; 5 Flechard et al. (2011) ; 6 Gielen et al. (2011) ; 7 Hendriks et al. (2007) ; 8 Ilvesniemi et al. (2009) ; 9 Kindler et al. (2011) ; 10 Kowalska et al. (2013) ; 11 Legout et al. (2016) ; 12 Luo et al. (2012) ; 13 Pilegaard et al. (2006) ; 14 REddyProc (2019) ; 15 Schaufler et al. (2010) ; 16 Simpson et al. (2012) ; 17 Tang et al. (2009) ; 18 van Oijen et al. (2005) ; 19 See references in Table S7.

Some of the results need to be updated, e.g. in section 2.2.1 it is written that EMEP data were downloaded in 2013 and different model versions were used?

Some EMEP model results were indeed used for the calculation of total N deposition in this paper, but only for i) wet deposition of inorganic N and ii) the NO2 concentration used in dry deposition. The comment about different versions of the model leading to different results is really only relevant for dry deposition of inorganic N, but this output of the model was not used in our total N deposition estimate; it is only used as a comparison in Fig. S2 of the supplement. Regarding wet deposition and NO2 concentrations from EMEP, we have stated at the end of section 2.2.1 that '*…Evaluation of the model against measurements over this period has shown quite consistent results for the wet-deposited components and NO2 concentrations…*', and therefore the data obtained in 2013 are not significantly different from the most up-to-date version of the model.

In a lot of statements there is an unevidenced qualification, such as: 'there did not appear to be any systematic overestimation ..', '0,85 is plausible but also much variability ..', We assumed that all sites of the European network followed the same relative time course ..', 'reveals a potential cross correlation ..', 'reasonable overall model performance ..', etc. It would help if such statements are underpinned/quantified.

We address each of these statements below:

'*…there did not appear to be any systematic overestimation compared with wet deposition estimates from the monitoring networks or EMEP data…*' This statement appears in methods, and it would not be appropriate to provide numbers, i.e. results, here. The data substantiating this statement are provided on lines 543-547. We have added a pointer to Results in the statement. ('see Results')

'*…the applied ratio of 0.85 is plausible but also that much variability…*' We presume Referee means that the variability is not shown, but the whole sentence (lines 332-334) actually reads '*…A comparison with values of DINTF / Ndep ratios actually measured at the EN2, EN8, EN10, EN16 and DB2 sites (0.71, 0.80, 0.29, 0.85, 1.11, respectively; mean ± st. dev. 0.75 ± 0.30) shows that the applied ratio of 0.85 is plausible but also that much variability in canopy retention/leaching may be expected between sites…*", so we do in fact provide the numbers and the actual range, mean and standard deviation of observed data in the same sentence.

'*…We assumed that all sites of the European network followed the same relative time course…*' This sentence was removed from the revised version, since most of the detailed BASFOR model description has moved to the Part II paper, as described above.

'*…reveals a potential cross correlation…*' We assume Referee #2 means we should provide statistical measures (eg R^2) of the correlations we mention between N deposition and climate. We agree this is a not a straightforward message to convey, or correlation to demonstrate, for two reasons: i) the relationships of N_{dep} to MAT or MAP are circumstantial rather than strictly causal, as we explain in the first paragraph of Section 4.3.1 (and as also pointed out by Referee #2 of the Part II paper); and ii) the relationships are not linear but bell-shaped, as evident in the figures shown below. We do not mean to imply that a change in MAT or MAP automatically induces a change in N_{dep}, but that N_{dep} cannot be considered to be completely independent of, or unrelated to, climate.

We have added the following figure to the Supplement (Fig. S4), as suggested by the Editor, to show the relationship of N_{dep} to MAT and MAP:

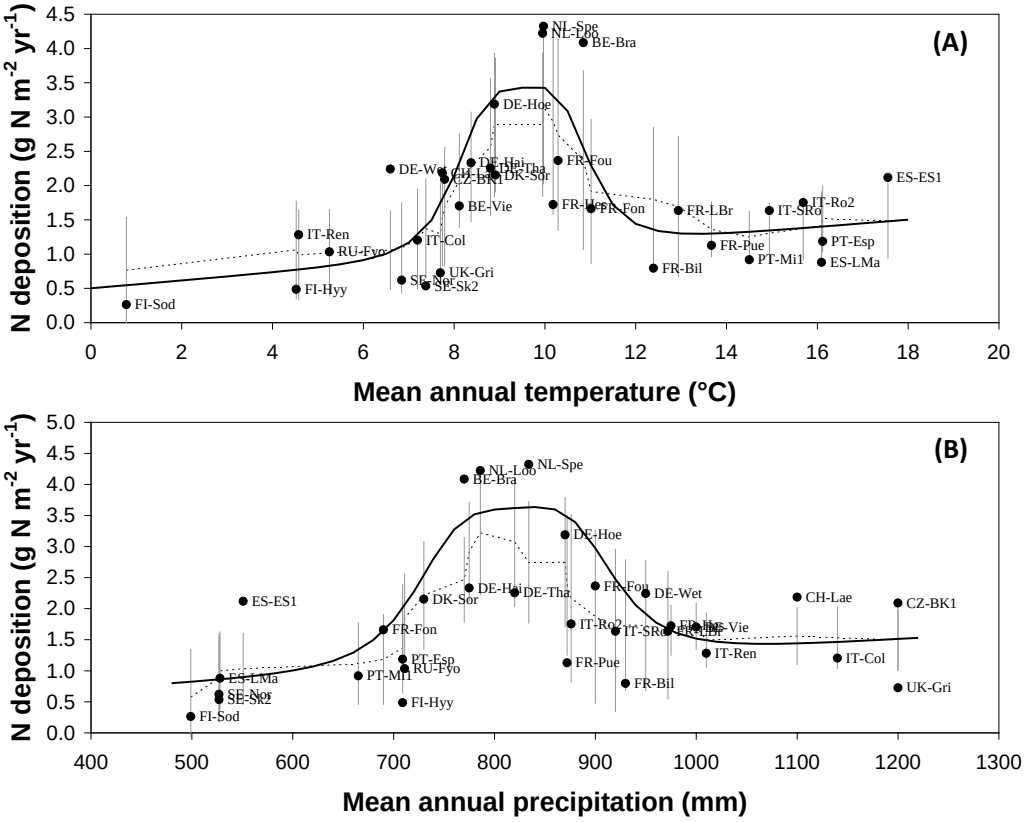

Figure S4. Spatial variations in measurement-based nitrogen deposition (N_{dep}), plotted as a function of (A) mean annual temperature (MAT) and (B) mean annual precipitation (MAP). Temperature and precipitation are not direct determinants of N_{dep}, but the geographical occurrence of peak N_{dep} levels in mid-range for both MAT and MAP means that the relationship of forest productivity to N_{dep} cannot be considered independently of climate at the European scale.

'*…reasonable overall model performance…*' We have provided in each sub-plot of Fig.6 (to which this sentence refers on line 675-676) the R^2, MAE and NRMSE statistics of modelled vs measured. It would be cumbersome and repetitive to add to the text all these numbers for each variable.

Finally, there seems to be some parts of the paper that are outdated, such as the start of the discussion about the Magnani paper. These can be omitted.

The publication of the Magnani et al. (2007) paper actually served the purpose of triggering much critical discussion, constructive debate and further studies and reviews on the complicated issue of the impact of nitrogen pollution on the carbon cycle. It is correct that there is now a consensus that the dC/dNdep values calculated by Magnani et al. were over-estimated, but the present paper has a different focus. We believe it is still necessary to stress the importance of two aspects, which were overlooked in the Magnani et al. (2007) paper, but not only there:

1- The size (and often dominance) of dry deposition relative to wet deposition, as well as the uncertainty in dry deposition. Other studies have also downplayed the role of dry deposition, probably because it is more difficult to quantify, and less visible, than wet deposition.
2- The linear relationship of NEP to N_{dep} derived from the Magnani et al. (2007) dataset did not make it apparent that N saturation in polluted areas may be counter-productive to forest growth. Later studies have indicated non-linear relationships, while accounting for other drivers, and we now refer more specifically to them in the text.

We believe that contrasting findings from this study with those from Magnani et al. (2007), and others before and after them, benefits the reader in providing a more balanced view. In this, we are supported by Referee #1 of the Part II paper, who writes: '*…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.*'

To highlight the contrast between the different studies, we have added the following two sentences:

Section 4, start of Discussion, line 721: *'…Sutton et al., 2008).* Other attempts have subsequently been made to assess impacts of N deposition on forest growth and carbon sequestration, while accounting for other drivers, at more than 350 long-term monitoring plots in Europe (Solberg et al., 2009; Laubhann et al., 2009; De Vries et al., 2008). *A special feature of the present study is that it aims to assemble N deposition rates …*'

Section 4.1, line 739: '*… especially not with wet N deposition only.* Based on a review of experimental N addition studies (e.g. Högberg et al., 2006; Pregitzer et al., 2008) and monitoring based field studies along N deposition gradients (e.g., Solberg et al., 2009; Laubhann et al., 2009; Thomas et al., 2010), De Vries et al. (2014) suggested that the C response reaches a plateau near 1.5-2.0 g N m^{-2} yr^{-1} and then starts to decrease. *The linear relationship between C sequestration and wet Nr deposition …*'

Additional references:

Högberg P., Fan, H., Quist, M., Binkley, D. and Tamm, C.O.: Tree growth and soil acidification in response to 30 years of experimental nitrogen loading on boreal forest, Glob. Change Biol., 12, 489–499, https://doi.org/10.1111/j.1365-2486.2006.01102.x, 2006.

Pregitzer, K.S., Burton, A.J., Zak, D.R. and Talhelm, A.F.: Simulated chronic nitrogen deposition increases carbon storage in Northern Temperate forests, Glob. Change Biol., 14, 142–153, https://doi.org/10.1111/j.1365-2486.2007.01465.x, 2008.

Thomas, R.Q., Canham, C.D., Weathers, K.C. and Goodale, C.L.: Increased tree carbon storage in response to nitrogen deposition in the US, Nat. Geosci., 3(1), 13–17, https://doi.org/10.1038/ngeo721, 2010.

We have in addition delete an unnecessary reference to Magnani et al. (2007) (line 604-605).

It raises the question to me about what the purpose of the paper is and how it relates to the second part. If it is a description of the database, including reference to the data sources, models and including an uncertainty estimate it can be a much shorter well focused paper with detailed information as supplementary information. The second part can then focus on the use and interpretation of the data. It would be my recommendation to split it in such a way. I therefore recommend major revision of the paper.

We hope we have convinced Referee #2 that this paper is about much more than the description of the database (i.e. a data paper) underpinning Part II of the study. In our response to the Referee's opening general comments (see above), we have argued that constructing realistic C and N budgets at many observation sites, and closing and connecting the C and N cycles (our main scientific objective), on the basis of measurement-based data and complemented where required by models, requires a thorough critical examination of the methods employed. We do not wish to show that our data are flawless (which they are not), but that they are as good as can be under the constraints we are facing, and that it is important to recognize, identify and rank important gaps in data, knowledge and models, whatever they are (e.g. N2 loss by denitrification; DOC leaching; organic N deposition; etc). We believe that the space taken up by method descriptions and uncertainty assessments is justified by these objectives.

To address the Referee's comment that it is not clear '*…what the purpose of the paper is and how it relates to the second part…*', we have made the following changes:

- The current abstract mentions uncertainty in the beginning but then does not mention it anymore. We have summarized our main findings on uncertainty in one sentence added towards the end of the abstract : 'Uncertainties in elemental budgets were much larger for nitrogen than carbon, especially at sites with elevated N_{dep} where N_r leaching losses were also very large, and compounded by the lack of reliable data on organic nitrogen and N_2 losses by denitrification.'
- The formulation of the goals of the paper, as defined in the last paragraph of the Introduction, may have been slightly misleading. We wrote in a single sentence: "*The main aim of this paper is to build tentative C, N and GHG budgets … prior to an assessment in the companion paper …*", which could be misinterpreted as meaning that Part 1 is just the Materials and Methods for Part 2. We have rephrased the opening sentence in the following way: 'A main objective of this paper is to build tentative C, N and GHG budgets, and analyse C/N interactions empirically, for a wide range of European monitoring sites, by using measurements or observation-based data as far as possible, complemented by modelling. Important methodological goals are to critically examine uncertainties in measurement methods and elemental budgets, to identify knowledge and data gaps, and to assess the current state of process understanding as encoded in models. To this end, we compiled …'. We have postponed the mentioning of the Part 2 paper until the very last sentence of the Introduction.

Specific comments:

Line 127-130– The information in the introduction about tropical forest could be shortened, as the paper focusses on temperate/boreal forest.

The three lines about tropical forests are provided in the introduction as background and contrast to temperate/boreal forests, which are the focus of this paper. It is relevant to briefly contrast the different regions of the globe at the start of the introduction, which discusses the likely magnitudes of global and hemispheric CO2 sinks.

Line 159 – 1 by 1 degree does not correspond to 10 by 10 km, it covers a larger area.

We do not mean that 1 x 1 degree corresponds to 10 x 10 km. We mean that the resolutions of models are typically 10 x10 km for regional CTMs, or 1° x 1° for global CTMs, as indicated by the adverb '*respectively*' at the end of the sentence.

Line 195 – How about vegetation changes at all sited, e.g. due to growth and/or composition change?

We have assumed (because we have no data to suggest otherwise) that vegetation species composition has not evolved since the beginning of measurements, or (in the case of BASFOR modelling) since forests were planted. But we have collected as much data as possible on tree heights, diameters, density, etc at various dates in the past, from publications and databases, as shown for example in Table S2 and Figure S5 of the Supplement. All these data were used in the Bayesian calibration of the BASFOR model (Cameron et al., 2018).

Line 220 – Not all FLUXNET sites seem to be included in this analysis, what are the criteria used to in- or exclude individual sites?

We did not conduct a meta-analysis of all FLUXNET sites, and there was no selection process. We stated in Introduction (lines 205-210) and in Materials and Methods (lines 237-240) that the sites were those included in the CarboEurope IP and NitroEurope IP projects, at which we installed N_r deposition monitoring equipment (and measured other variables). We could not use all FLUXNET sites for this study, since our aim was to quantify as many components of the C and N cycles on the basis of measurements as far as possible, and such data are not available for most FLUXNET sites.

Line 262 – In this section it is sometimes unclear to what "models" are referred. Specify what type of models, e.g. dry deposition models. What are the key differences between these deposition models? It would also be good to specify the EMEP model domain and resolution in this section.

The dry deposition models were described in some detail in Flechard et al. (2011), where four models were compared and their uncertainties analyzed. The main principle of such models is described in the present paper on lines 258-261; i.e., the dry deposition flux is obtained by the product of measured ambient concentration and a deposition velocity (V_d), that depends on meteorology, surface roughness, leaf area index, chemical species, etc. We recognize that it is difficult for a non-specialist to see clearly how this works on the basis of such a short description, but we believe that a full description of the method is outside the scope of this paper, especially if we do not wish to make the paper longer than it already is. The most effective way is to refer the reader to specialized papers (see line 261), as we do for all other methods.

The EMEP model resolution is given in this section (line 288), and the modelling domain is shown in Fig. 5.

Line 284 – leaving out the organic N data leads to a systematic bias.

We acknowledge that failing to account for organic N_r deposition in our total N_r deposition estimate leads to a small bias. We were unfortunately unable to estimate the missing organic terms (both wet and dry) by modelling. But as we state on line 287, the underestimation is a small one (<5% of total N_r deposition), and much smaller than the overall uncertainty in N_{dep} (as discussed in Section 4.1.1).

Line 290 – There are probably new data available?

We copy below the reply we made to Referee #1 on the same topic:

Some EMEP model results were indeed used for the calculation of total N deposition in this paper, but only for i) wet deposition of inorganic N and ii) the NO2 concentration used in dry deposition. The comment about different versions of the model leading to different results is really only relevant for dry deposition of inorganic N, but this output of the model was not used in our total N deposition estimate; it is only used as a comparison in Fig. S2 of the supplement. Regarding wet deposition and NO2 concentrations from EMEP, we have stated at the end of section 2.2.1 that '*…Evaluation of the model against measurements over this period has shown quite consistent results for the wet-deposited components and NO2 concentrations…*', and therefore the data obtained in 2013 are not significantly different from the most up-to-date version of the model.

Line 308 – Could you elaborate on why you decided to scale up the N2O and NO fluxes using linear interpolation?

This a common but admittedly not ideal procedure in such studies, where flux measurements are not continuous and gaps need to be filled somehow to yield annual flux estimates. We acknowledge that linear interpolation can lead to large uncertainties in annual flux estimates, e.g. by missing emission peaks that can occur (unnoticed) between two consecutive flux measurement dates, or by over-representing over time large fluxes measured on certain days. The issue is related to whether the statistical NO or N_2O flux distribution is normal (Gaussian) or quasi log-normal. The latter is frequent in managed, fertilized agro-systems, where large emission peaks occur during just a few days or weeks per year, in response to fertilization, and emissions are otherwise very small. In forests and non-fertilized semi-natural systems, fluxes tend to be more normally distributed, and we believe that the uncertainty associated with linear interpolation is much smaller.

Ideally a more mechanistic gap-filling procedure for NO or N_2O fluxes, based on soil moisture, temperature, and other indicators of soil microbial activity and SOM turnover, should be used. However, in practise this is often very hard to achieve using field measurements, either due to the low temporal resolution of measurements, or because no significant patterns can be derived between fluxes and environmental macro-drivers. Linear interpolation then represents the default alternative. We have added the following text in this section, to briefly summarize the issue:

'*…scaled up to yearly values by linear interpolation or using the arithmetic mean of all flux measurements*. There may be considerable uncertainty in the annual flux if gap-filling is based on linear interpolation between discrete values, when flux measurements are made manually and therefore discontinuous and infrequent (Parkin, 2008). This is due to the episodic nature and lognormal distribution of NO and N_2O emissions, observed particularly in fertilized croplands and grasslands. However, this 'episodicity' is less pronounced in semi-natural ecosystems, or at least the magnitude of the episodic fluxes is generally much smaller than in fertilized agro-systems (Barton et al., 2015). The uncertainty in annual emissions estimated in our study from manual chamber measurements is related to the observation frequency (bi-weekly or monthly), and larger than in the case of automatic (continuous) chamber measurements.'

Added References:

Barton, L., Wolf, B., Rowlings, D., Scheer, C., Kiese, R., Grace, P., Stefanova, K. and Butterbach-Bahl, K.: Sampling frequency affects estimates of annual nitrous oxide fluxes, Sci. Rep.-UK, 5:15912, https://doi.org/10.1038/srep15912, 2015.

Parkin, T.B.: Effect of sampling frequency on estimates of cumulative nitrous oxide emissions, J. Environ. Qual., 37, 1390–1395, https://doi.org/10.2134/jeq2007.0333, 2008.

Line 326 – DINTF is not included in the abbreviations table.

DIN_{TF} was added to Table 2.

Line 334 – there is an enormous range, why not used in some way?

We did not find a pattern to the ratio of throughfall to total N_{dep} for these five sites. More observations would be needed to link this ratio to forest and climatic characteristics.

Line 336 – This section, about the EC processing, is lengthy. Consider shortening it.

We have touched upon this specific issue as part of our response to Referee #2's general comments (see above). The discussion of uncertainties in the CO_2 budgets is dependent on methods being described in some detail, because the assumptions made in EC data post-processing, gap-filling and partitioning are critical for the C budgets derived from the raw data. Ecosystem carbon budgets and sequestration are the main motivations for this work, and collective experience from 25 years of flux monitoring networks around the world shows that there are many ways and options to measure and analyze eddy covariance data, with the end results (inter-annual mean CO_2 budgets) being heavily influenced by methodological choices. One illustration is the discussion on night-time fluxes and advection issues (lines 843-909).

Line 389 – Missing comma between "some sites" and "such as"

Done

Line 427 – Add a reference to the table in the supplement that includes the major publications per site.

Done

Line 480 – It would make more sense to move the primary purpose of the BASFOR model to the beginning of the section.

Most of the BASFOR description (Section 2.6) was moved to the companion paper, as suggested above. But we have kept a very brief summary (a few lines) describing the purpose and basic principles of BASFOR.

Line 538 – 550 – The discussion on the model uncertainties would fit better in the discussion section.

We agree that lines 538-542 consitute discussion material for dry deposition; they were moved to Section 4.1.1.

Lines 543-550 describe wet deposition results by different methods without further discussion.

Line 580 – 594 – This section discusses the computation of the denitrification losses and the uncertainty associated with it. Consider moving a part of this section to the methods/discussion.

We have very little data on denitrification N_2 losses. This paragraph provides a couple of very tentative numbers as part of results, as a background to the more informed and detailed inorganic N fluxes; but it is really too small to split into Methods and Discussion as suggested by the Referee.

Line 603– 612 – It seems a bit sudden to address the ultimate objective of the whole project here. It would be better to discuss this earlier on in the paper.

The objectives of the paper were laid out in the final paragraph of Introduction, and were strengthened in the revised version by the addition of a couple of sentences, as described above in response to the Referee's request for a clearer description of the aims of the paper.

However, the sentence on line 603-604 did not actually refer solely to this paper's objectives, but to the '*ultimate objective of the project*' as a whole, including the companion paper (Part II of the study).

Line 640 – You state that there is a "broad negative correlation between MAP and MAT", could you support this with a R2 value? Moreover, the MAP alone does not seem appropriate to address site-specific water-availability, without considering other parameters such as soil water holding capacity etc.

We have added the R^2 value to the text:

'…for sites with MAT > 7 °C there was a broad negative correlation between MAT and MAP (R^2 = 0.24, p = 0.01) i.e. the warmest sites in southern Europe…'

We agree entirely with the Referee's comment that MAP is not '*appropriate to address site-specific water availability, without considering other parameters such as soil water holding capacity etc*'. As we state in introduction to 3.2.1, the first stage of the study is a '*descriptive approach*', which however '*…is done with the strong reservation that a simple empirical relationship does not necessarily prove causality, as other confounding and co-varying factors, e.g., climate, soil, age, etc, may exist.*' (lines 606-608). We further emphasized, later on (lines 969-976), that '*…In addition, a range of other potential explanatory variables such as soil type, especially the water holding capacity (Φ_{FC} - Φ_{WP}), soil fertility (Vicca et al., 2012; Legout et al., 2014), tree species, stand age (Besnard et al., 2018), are potentially needed to explain the observed variability (Flechard et al., 2019).*'

Thus our intention in this paper was first to show the apparent inter-relationships between carbon sequestration, N deposition and climate, in a similar way to previous studies, but at the same time pointing to the difficulties and risks of single factor interpretations. Multiple factors are clearly involved (climate, soil, fertility, age, species, etc), which, owing to the limited size of our database and the non-independence of controlling factors, could not be untangled using statistical methods (see lines 959-965 and 972-976). Having established this methodological challenge, we therefore proposed to investigate the issue in the companion paper (Part II) using mechanistic modelling.

Line 647 – Missing "were"

The verb for the sentence was on the next line (648)

Line 659 – 674 – In this section you are discussing results related to Figure 4 again. Consider moving this section up.

Fig.4 contains the data described in both Sections 3.2.1 and 3.2.2. We think it is more logical to start describing the results with '*Spatial variability of the carbon sink in relation to climate and nitrogen deposition*' (3.2.1) and then move on to '*Differences between plant functional types*' (3.2.2).

Line 737 – Add a reference for this number.

We have added the reference to Figure 3:

'…when Ndep exceeds a critical load of approximately 2-2.5 g (N) m-2 yr-1 (Fig. 3),…'

Line 754 – how can you demonstrate that it 'provides a better estimate' and that it is more realistic than an transport model?

We agree that the wording of this sentence is perhaps too strong. Indeed, we can't prove from our data and with 100% certainty that '*…measuring gas-phase and aerosol Nr concentrations locally did provide a better estimate of total ecosystem Nr inputs…*'. The only way to prove this would be to measure all dry deposition fluxes for all gaseous and particulate N_r species by micrometeorological methods, for several years, which is practically impossible.

We nonetheless believe that having a record of actual measured gas-phase and aerosol N_r concentrations at all sites, for several years, at least reduces the uncertainty in N_r concentrations, compared with a large-scale chemical transport model.

We have changed the verb of the sentence from '*did provide*' to '*should provide*'.

Line 804 – It would be better to either add the definitions for N saturation here or refer to this later.

This section discusses the uncertainties in N losses and budgets, rather than the concepts of saturation, whose definitions come later (4.3.2); however it makes sense to at least mention N saturation at this stage.

Line 930 – Why is the uncertainty in the non-CO2 fluxes possibly >100% larger?

'*…the uncertainty in non-CO2 GHG fluxes is much larger (possibly > 100%) than for multi-annual EC-based CO2 datasets…*' for several reasons, which we have explained in the text:

- because, unlike CO_2, in our datasets the non-CO_2 GHG fluxes were not measured in situ continuously (manual soil chamber measurements made periodically, except at a few sites equipped with auto-chambers);
- because non-CO_2 GHG flux measurements were mostly made by chambers and thus not at the ecosystem scale, unlike eddy covariance, which integrates a large footprint;
- because for some sites, there were no in-situ flux measurements, and fluxes were estimated from the bioassay experiments, or from BASFOR modelling.

Line 972–975 – You state here that your attempts with more advanced forms of regression analysis were not successful. This is a bit on the vague side. You should either elaborate on the attempts that were made and/or for instance add some references, or exclude this statement from this section.

We have rephrased this sentence in the following way:

'*…are potentially needed to explain the observed variability*. In order to account for, and untangle, the multiple inter-relationships, we chose a mechanistic model (BASFOR) based approach, described in Flechard et al. (2019), whereby most of the known interactions of plant, soil, climate, age, species, are encoded and parameterised to the best of our current knowledge. Given the limited size and very large diversity of the dataset, such an approach appears to be preferable to regression-based statistical analyses, since a simple pattern to explain the coupling of carbon and nitrogen budgets with the available data and knowledge is unlikely.'

Line 1016 – It is unclear why considering the uncertainties would amount to a confident threshold of 2-2.5 g N, rather than just widening the range of the given estimate, could you elaborate on this?

We agree that the wording was confusing. What we meant is that there may be less uncertainty in the threshold for the advanced stage of saturation (2-2.5 g N m^{-2} yr^{-1}) than in the thresholds for early or moderate saturation. We changed the sentence starting line 1016:

'*…Following definition ii) of N saturation given above, the sum of inorganic Nr losses, heavily dominated by DIN leaching at the upper end of the Ndep range in our datasets (Fig. 3), may indicate various stages of N saturation in all forests with Ndep > 1-1.5 g (N) m-2 yr-1.* A threshold for a more advanced saturation stage could be placed at 2-2.5 g (N) m-2 yr-1, where inorganic N_r losses are consistently larger than 50% of N_{dep}. *Such numbers are entirely consistent…*'

Line 1052 –1055 – This sentence is a bit confusing. It refers to the large uncertainty in dry deposition modelling, but both deposition estimates from CTM and denuders make use of dry deposition models in the end. So I would say that it is not so much the deposition models that improved substantially, but more the estimates of the concentrations of these compounds.

We did not state that '*…the deposition models … improved substantially…*', but that '*…the low-cost network to monitor atmospheric gas-phase and aerosol N_r contributed to substantially reducing the large uncertainty in total N_{dep} rates at individual sites*'. We agree however that the sentence was a little long and confusing, and therefore we split it into two:

'Nevertheless, the low-cost network to monitor atmospheric gas-phase and aerosol N_r contributed to substantially reducing the large uncertainty in total N_{dep} rates at individual sites (compared with gridded outputs of a regional chemical transport model). This was because dry deposition almost systematically heavily dominates over wet deposition in forests, except at very remote sites (away from sources of atmospheric pollution), and directly measured N_r concentrations reduced the uncertainty in dry deposition fluxes'

Figure 4 – What type of LAI data is used to determine LAImax?

Leaf are index is defined both in the text (line 629) and in Table 1 (footnote) as '*1-sided for broad-leaf, or half of total for needle-leaf*'.

Figure 8 – Some of the labels seem to be missing.

For each site in each plot, there are two values (observed and modelled). We provided only one label per site (attached to the 'observed' series) in order not to clutter the plots. The site name for the 'modelled' series can be easily deduced by following a vertical path, since both 'observed' and 'modelled' data points have the same x-value.

Figure 9 – "to each tohet" – to each other OK

Consider splitting up / reformulating these sentences for clarity/readability:

For each sentence we provide the revised text.

Line 119-121 – "The global terrestrial . . . Pg (C) yr-1"

'The global terrestrial net sink for atmospheric carbon dioxide (CO2) is approximately 1.7 Pg (C) yr-1, i.e. roughly one fifth of global CO2-C emissions by fossil fuel combustion and industry (9.4 ± 0.5 Pg (C) yr-1). This corresponds to the land based carbon (C) uptake of 3.2 ± 0.8 Pg (C) yr-1 minus emissions from deforestation and other land-use changes of 1.5 ± 0.7 Pg (C) yr-1.'

Line 127-130 – "Tropical forest areas . . . generally believed."

'Tropical forest areas are believed to be closer to carbon neutral (Pan et al., 2011), or even a net C source globally (Baccini et al., 2017), due to emissions from deforestation, forest degradation and land use change offsetting their

sink potential. However, others (Stephens et al., 2007) have argued that the tropical land CO2 sink may be stronger – and the Northern hemispheric land CO2 sink weaker – than was generally believed.'

Line 297-299 – "Nitrogen losses to . . . empirical methods."

'Nitrogen losses to the atmosphere (gaseous emissions) and to groundwater (N leaching) are especially hard to quantify and thus typically cause large uncertainties in ecosystem N budgets. These N_r losses were estimated by direct flux measurements or by indirect empirical methods.'

Line 308 -309 – "To address . . . responses of soils."

'Direct in situ Nr and non-CO2 GHG gas flux measurements were unavailable at many sites. These soil N2O, NO (and also CH4) fluxes were therefore also estimated, as part of NEU, from the temperature and moisture responses of soils.'

Line 525-528 – "Total Nr deposition . . . Schwede et al., 2018)."

'Total Nr deposition was around 25% smaller on average at short semi-natural vegetation sites compared with forests (Fig. S2), even though the mean total atmospheric Nr concentrations (reduced and oxidized, N-containing gas and aerosol compounds) were quite similar between the two data sets (Flechard et al., 2011). The difference was driven by higher dry deposition rates over forests due to higher aerodynamic roughness and deposition velocities (Fig. S3; see also Schwede et al., 2018).'

Line 548-550 – "Wet deposition . . . most forest sites."

This sentence is fairly straightforward.

Line 754-756 – "Despite these . . . cycling processes."

'Despite these uncertainties, measuring gas-phase and aerosol Nr concentrations locally should provide a better estimate of total ecosystem Nr inputs than the outputs of a large-scale chemical transport model. In addition, the partitioning of wet *vs* dry deposition, reduced *vs* oxidized N, and canopy absorption *vs* soil deposition, should also be improved, all of which are useful in interpreting ecosystem N cycling processes.'

Line 790-793 – "In addition . . . as beech or oak."

'In addition, it is noteworthy that the two sites with the largest N_{dep} and DIN leaching rates (EN15, EN16) were dominated by pine or Douglas fir (Table S2). These species have been shown in a common garden experiment (Legout et al., 2016) to cause larger nitrification, NO3- leaching and acidification rates (as well as larger losses of calcium, magnesium and aluminium), compared with other tree species such as beech or oak.'

Line 824-828 – "By analogy . . . observation periods."

'Previous studies have normalised data through the carbon use efficiency (CUE, commonly defined from a plant's perspective as the NPP/GPP ratio), or the biomass production efficiency (BPE = BP/GPP; Vicca et al., 2012), which is a CUE proxy. By analogy, we define here an ecosystem-scale, medium-term indicator of carbon sequestration efficiency (CSE) as the NEP/GPP ratio, calculated from measurable fluxes over the CEIP/NEU project observation periods.'

Line 959-963 – "Through the continent-wide. . . continental scale."

'Nitrogen deposition patterns at the European scale result from the continent-wide geographical distribution of population, human, industrial and agricultural activities, and of precursor emissions, combined with mesoscale patterns of meteorology-driven atmospheric circulation and chemistry. Through the interplay of these factors, the elevated Ndep levels in this study happened to co-occur geographically with temperate climatic zones of Central-Western Europe (Fig. 5 C-D) that are the most conducive to vegetation growth at the continental scale.'

Line 972-975 – "Our attempts . . . the dataset."

This has been addressed and rephrased as part of a previous comment (see above).

Line 1052-1055 – “Nevertheless, . . . is much larger.”

This has been addressed and rephrased as part of a previous comment (see above).

Carbon/nitrogen interactions in European forests and semi-natural vegetation. Part I: Fluxes and budgets of carbon, nitrogen and greenhouse gases from ecosystem monitoring and modelling

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Abstract. The impact of atmospheric reactive nitrogen (N_r) deposition on carbon (C) sequestration in soils and biomass of unfertilised, natural, semi-natural and forest ecosystems has been much debated. Many previous results of this dC/dN response were based on changes in carbon stocks from periodical soil and ecosystem inventories, associated with estimates of N_r deposition obtained from large-scale chemical transport models. This study and a companion paper (Flechard et al., 2020) strive to reduce uncertainties of N effects on C sequestration by linking multi-annual gross and net ecosystem productivity estimates from 40 eddy covariance flux towers across Europe to local measurement-based estimates of dry and wet N_r deposition from a dedicated collocated monitoring network. To identify possible ecological drivers and processes affecting the interplay between C and N_r inputs and losses, these data were also combined with in situ flux measurements of NO, N_2O and CH_4 fluxes, soil NO_3^- leaching sampling, as well as results of soil incubation experiments for N and greenhouse gas (GHG) emissions, surveys of available data from online databases and from the literature, together with forest ecosystem (BASFOR) modelling.

Multi-year averages of net ecosystem productivity (NEP) in forests ranged from -70 to 826 g (C) m^{-2} yr^{-1} at total wet + dry inorganic N_r deposition rates (N_{dep}) of 0.3 to 4.3 g (N) m^{-2} yr^{-1}; and from -4 to 361 g (C) m^{-2} yr^{-1} at N_{dep} rates of 0.1 to 3.1 g (N) m^{-2} yr^{-1} in short semi-natural vegetation (moorlands, wetlands and unfertilised extensively managed grasslands). The GHG budgets of the forests were strongly dominated by CO_2 exchange, while CH_4 and N_2O exchange comprised a larger proportion of the GHG balance in short semi-natural vegetation. Uncertainties in elemental budgets were much larger for

nitrogen than carbon, especially at sites with elevated N_{dep} where N_r leaching losses were also very large, and compounded by the lack of reliable data on organic nitrogen and N_2 losses by denitrification. Nitrogen losses in the form of NO, N_2O and especially NO_3^- were on average 27% (range 6-54%) of N_{dep} at sites with N_{dep} < 1 g (N) m^{-2} yr^{-1}, versus 65% (range 35-85%) for N_{dep} > 3 g (N) m^{-2} yr^{-1}. Such large levels of N_r loss likely indicate that different stages of N saturation occurred at a number of sites. The joint analysis of the C and N budgets provided further hints that N saturation could be detected in altered patterns of forest growth. Net ecosystem productivity increased with N_r deposition up to 2-2.5 g (N) m^{-2} yr^{-1}, with large scatter associated with a wide range in carbon sequestration efficiency (CSE, defined as the NEP/GPP ratio). At elevated N_{dep} levels (> 2.5 g (N) m^{-2} yr^{-1}), where inorganic N_r losses were also increasingly large, NEP levelled off and then decreased. The apparent increase in NEP at low to intermediate N_{dep} levels was partly the result of geographical cross-correlations between N_{dep} and climate, indicating that the actual mean dC/dN response at individual sites was significantly lower than would be suggested by a simple, straightforward regression of NEP vs. N_{dep}.

Commentaire [C1]: Provided exact (average) numbers for N losses

1 Introduction

The global terrestrial net sink for atmospheric carbon dioxide (CO_2) is approximately 1.7 Pg (C) yr^{-1}, i.e. roughly one fifth of global CO_2-C emissions by fossil fuel combustion and industry (9.4 ± 0.5 Pg (C) yr^{-1}). This corresponds to the land based carbon (C) uptake of 3.2 ± 0.8 Pg (C) yr^{-1} minus emissions from deforestation and other land-use changes of 1.5 ± 0.7 Pg (C) yr^{-1}. The ocean sink is of the same order (2.4 ± 0.5 Pg (C) yr^{-1}), while twice as much CO_2-C (4.7 ± 0.02 Pg (C) yr^{-1}) is added yearly to the atmosphere (Le Quéré et al., 2018). Data from atmospheric CO_2 inversion methods (e.g. Bousquet et al., 1999, Ciais et al., 2010), from national to global forest C inventory approaches (Goodale et al., 2002; Pan et al., 2011), and from eddy covariance (EC) flux networks (Luyssaert et al., 2007), have suggested that a dominant part of this terrestrial CO_2 sink is currently occurring in forests, and especially in boreal and temperate forests of the Northern hemisphere (Ciais et al., 2010; Pan et al., 2011). Tropical forest areas are believed to be closer to carbon neutral (Pan et al., 2011), or even a net C source globally (Baccini et al., 2017), due to emissions from deforestation, forest degradation and land use change offsetting their sink potential. However, others (Stephens et al., 2007) have argued that the tropical land CO_2 sink may be stronger – and the Northern hemispheric land CO_2 sink weaker – than was generally believed. At the European scale, Schulze et al. (2010) calculated that the net biome productivity (NBP, the mean long-term carbon sink at a large spatial scale) of temperate and boreal forests was 81% of the total continental-scale land sink.

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The large European and North American CO_2 sinks have been attributed to a combination of factors including afforestation of abandoned land and formerly cut forests, reduced forest harvest, CO_2 fertilisation, changes in management and age structure legacy effects in Europe (Vilén et al., 2016), and atmospheric reactive nitrogen (N_r) deposition (Reay et al., 2008; Ciais et al., 2013, and references therein; De Vries et al., 2017). However, some studies (Nadelhoffer et al., 1999; Gundale et al., 2014; Fernández-Martínez et al., 2017) have questioned the widespread theory that elevated N_r deposition boosts forest C sequestration, and the magnitude of the N «fertilisation» effect on forest C sequestration has been a matter of much debate (Magnani et al.,2007; Högberg, 2007; De Schrijver et al., 2008; de Vries et al., 2008; Magnani et al., 2008; Sutton et al., 2008; Dezi et al., 2010; Binkley and Högberg, 2016). A better understanding of the impact of nitrogen deposition on natural and semi-natural ecosystems, in particular over forests, and the impact on the carbon and nitrogen cycles as an indirect effect resulting from anthropogenic activities (Canadell et al., 2007), remains key to improve the forecast of regional (de Vries et al., 2017) and global (Du and de Vries, 2018) models.

The relevance of N_r deposition for the global C sequestration potential, or more explicitly the dC/dN response (change in C storage with change in N_r deposition), has been estimated typically through meta-analyses of N_r addition experiments (e.g. Schulte-Uebbing and de Vries, 2018), or by combining forest growth inventories, together with estimates of Nr deposition obtained from large-scale forest monitoring plots (Solberg et al., 2009; Laubhann et al., 2009; De Vries et al., 2008). Both

methods have many sources of uncertainty. One key difficulty in the latter approach lies in estimating total (wet+dry) N_r deposition (N_{dep}), especially dry deposition, which is highly variable spatially, very challenging to measure, and consequently hard to parameterize in regional-scale chemical transport models (CTM) (Flechard et al., 2011; Simpson et al., 2014; Schwede et al., 2018). The annual or long-term dry deposition component of N_{dep} to forests, in all the diversity of N-containing forms (gaseous vs. aerosol, reduced vs. oxidized, inorganic vs. organic, e.g. Zhang et al., 2009), has been actually measured (by micrometeorological methods) in very few forests worldwide (Neirynck et al., 2007; Erisman et al., 1996). Due to the large diversity of atmospheric compounds that contribute to total N_r and the complexity of the measurement techniques required for each compound (Flechard et al., 2011), it is even debatable that complete measurements of all N_r deposition terms have ever been achieved anywhere. Thus virtually all studies of the forest dC/dN response so far have relied on modelled atmospheric N_r deposition estimates, at least for the dry and occult deposition fractions, and further, that the N_r deposition data being used were systematically provided by the outputs of large-scale regional (e.g. Sutton et al., 2008; Fernández-Martínez et al., 2017) or even global (Fleischer et al., 2013) models, with resolutions of typically 10 km x 10 km or 1° x 1°, respectively. Grid averaging in such large-scale models introduces a large uncertainty in local (ecosystem-scale) N_r dry deposition rates (Schwede et al., 2018), particularly when the forest sites are located near agricultural or industrial N_r sources (Loubet et al., 2009; Fowler et al., 1998).

Additionally, nitrogen losses may significantly offset atmospheric N_r inputs at eutrophicated and acidified sites, with the consequence that dC/dN may correlate better with net, rather than gross, atmospheric N_r inputs. Depending especially on the extent of ecosystem N saturation (De Schrijver et al., 2008), substantial N losses may occur in the form of nitrate (NO_3^-) leaching (Dise et al., 2009), nitric oxide (NO) and nitrous oxide (N_2O) emissions (Pilegaard et al., 2006), ammonia (NH_3) bi-directional exchange (Hansen et al., 2013), as well as emissions of di-nitrogen (N_2) from total denitrification (Butterbach-Bahl et al., 2002) (Fig. 1). The implication is that the carbon response to N_{dep} would be non-linear, with larger dC/dN at low N_{dep} rates, and a lowering of dC/dN as N_{dep} increases, as suggested in the review by Butterbach-Bahl and Gundersen (2011) and further elaborated in De Vries et al. (2014). The latter authors show in their review that above a certain N deposition level, the dC/dN response declines due to adverse effects of excess N_r deposition and high soil ammonium (NH_4^+) concentration and nitrification (e.g. acidification, nutrient base cation losses, aluminium mobility), which are known to reduce soil fertility and affect ecosystem health and functioning (Aber, 1992).

Carbon losses through dissolved organic carbon (DOC) and biogenic dissolved inorganic carbon (DIC) leaching can also be significant, especially for wetlands (Dinsmore et al., 2010) and also grassland and cropland ecosystems (Kindler et al., 2011; Gielen et al., 2011). This is relevant for the net ecosystem carbon balance (NECB) or the net biome productivity (NBP) estimates obtained on the basis of EC flux systems, and needs to be accounted for as a part of the net ecosystem productivity (NEP) that is not actually stored in the system (Chapin et al., 2006; Schulze et al., 2010) (Fig. 1). Dissolved and/or emitted methane (CH_4) may further represent a significant loss from organic soils (Hendriks et al., 2007), while CH_4 oxidation, which is often observed in well-aerated soils and can be suppressed by N_r addition, especially NH_4^+ (Steudler et al., 1989), may affect the net greenhouse gas (GHG) budget. Nitrogen deposition-induced N_2O emissions from the forest floor (Pilegaard et al., 2006; Liu and Greaver, 2009), or from denitrification triggered by deposited NO_3^- in peatland (Francez et al., 2011), can also offset the gain in the ecosystem GHG balance resulting from a hypothetical nitrogen fertilisation effect.

Nitrogen deposition or addition is known to affect soil microbial C cycling in many different ways, for example high level N enrichment generally leading to reduced microbial biomass and suppressed soil CO_2 respiration (Treseder, 2008); a reduction of basal respiration without significant decline in total microbial biomass, following N addition to incubated peat cores (Francez et al., 2011); and added NO_3^- altering directly the oxidative enzyme production by microbial communities and hence controlling extracellular enzyme activity (Waldrop and Zak, 2006). Nitrate addition can lead to a reduction in CH_4 emissions from wetlands and peatlands (Francez et al., 2011), since in anaerobic conditions and in the presence of NO_3^- as electron acceptor, denitrifying bacteria can oxidize organic C-substrates (*e.g.* acetate) and thus out-compete methanogenic

communities (Boone, 1991). However, if chronic N enrichment of peatland ecosystems leads to floristic changes, especially an increase in vascular plants at the expense of bryophytes, the net effect may be an increase in CH_4 emissions (Nykänen et al., 2002), as the aerenchyma of tracheophytes provides a direct diffusion path to the atmosphere for soil-produced CH_4, bypassing oxidation in the peat by methanotrophs. Excess nitrogen-induced vegetation composition changes in Sphagnum moss peatland are believed to reduce C sequestration potentials, and the effect is likely to be exacerbated by climate change (Limpens et al., 2011).

This complex web of interactions between the C and N cycles and losses shows the need for integrated approaches for studying the impacts of N_r deposition on C sequestration and net GHG budgets. Ideally, all C and N gain and loss pathways (including infrequently or rarely measured fluxes such as N_r dry deposition, organic C and N leaching fluxes, GHG fluxes, etc; see Fig. 1) should be quantified at long-term experimental sites to improve and calibrate process-based models. Closing the C and N budgets experimentally at each site of large (e.g. FLUXNET) monitoring networks is unlikely to occur in the near future, but realistic and cost-effective measurement approaches can be used to progressively reduce the uncertainties for the large terms of the budgets. Such approaches were tested and implemented in this study, as part of a large-scale effort, within the NitroEurope Integrated Project (NEU, 2013; Sutton and Reis, 2011), to quantify N_r deposition and N losses from ecosystems, in parallel and coordinated with the CarboEurope Integrated Project (CEIP, 2011) to estimate the net C and GHG balance, for forest and semi-natural ecosystems in Europe.

A main objective of this paper is to build tentative C, N and GHG budgets, and analyse C/N interactions empirically, for a wide range of European monitoring sites, by using measurements or observation-based data as far as possible, complemented by modelling. Important methodological goals are to critically examine uncertainties in measurement methods and elemental budgets, to identify knowledge and data gaps, and to assess the current state of process understanding as encoded in models. To this end, we compiled the C, N and GHG flux data from NEU, CEIP and other complementary datasets, using a combination of in situ measurements, empirical relationships, ecosystem modelling, literature and database surveys, at the scale of the CEIP and NEU flux monitoring networks. This study presents the methodologies and discusses the different terms of the budgets, including atmospheric deposition from gas, aerosol and precipitation N_r concentration monitoring, soil NO_3^- leaching measurements and modelling, GHG and N_r emission estimates from chamber measurements and laboratory-based soil bioassays, EC tower-based C budgets, as well as historical published data. Forest ecosystem modelling (BASFOR) is used to simulate C, N and GHG fluxes, with the double objective to compare with actual measurements and to fill some gaps in the datasets. Wherever possible, alternative measurements, datasets or modelled data are shown alongside the primary data in order to provide an estimate of the uncertainty in the different terms. In the companion paper (Flechard et al., 2020), the response of C sequestration to N_{dep} is quantified using the same datasets.

{Insert Figure 1 here}

Commentaire [c4]: Clarified the main goals of the paper

Commentaire [c5]: Reference to the companion paper for the analysis of the dC/dN response

2 Materials and methods

2.1 Monitoring sites

The study comprised 40 terrestrial ecosystem-scale, carbon and nitrogen flux monitoring sites, including 31 forests (F) and 9 natural or semi-natural (SN) short vegetation ecosystems, primarily moorlands, wetlands and extensively managed, unfertilised grasslands (Table 1). The sites spanned a European geographical and climatic gradient from the Mediterranean to the Arctic and from the Atlantic to western Russia (Fig. S1), an elevation range of -2 m to 1765 m a.m.s.l., a mean annual temperature (MAT) range of -1.0°C to 17.6°C, and a mean annual precipitation (MAP) range of 500 mm to 1365 mm. Selected references are provided for each site in Table S1. A list of the main acronyms and abbreviations used in the paper is provided in Table 2.

{Insert Table 1 here}

{Insert Table 2 here}

The forest sites of the study ranged from very young (< 10 years old) to mature (> 150 years old), and can be broadly classified into four plant functional types (PFT) or five dominant tree categories (Table 1): deciduous broadleaf (DB), evergreen needle-leaf (EN, comprising mostly spruce and pine species), mixed deciduous/coniferous (MF), and Mediterranean evergreen broadleaf (EB). Forest species composition, stand characteristics, C and N contents of different ecosystem compartments (leaves, wood, soil), soil physical properties and micro-climatological characteristics are described in Tables S2-S5. Semi-natural short vegetation ecosystems included unimproved (mountainous and semi-arid) grasslands, wetlands and peatlands; they are included in the study as unfertilised, C-rich soil systems, providing a contrast with forests where storage also occurs above ground (thus with different C/N ratios). Among the 40 EC-CO_2 flux measurement stations, most sites (36) were part of the CEIP CO_2 flux network. A further three CO_2 flux sites were operated as part of the NEU network (EN2, EN16, and SN3), and one site (DB4) was included from the French F-ORE-T observation network (F-ORE-T, 2012). Table S6 provides an overview of the available C, N and GHG flux measurements, detailed hereafter.

2.2 Nitrogen fluxes

Input and output fluxes of the ecosystem nitrogen and carbon budgets are represented schematically in Fig. 1. The following sections describe the methods used to quantify the different terms, summarized in Table 3.

{Insert Table 3 here}

Commentaire [c6]: Added new Table 3 to provide overview of methods used in compiling C and N budget terms

2.2.1 Atmospheric deposition

To obtain realistic estimates of total (dry + wet) N_r deposition at the 40 sites of the network, it was necessary to measure ambient air concentrations of the main N-containing chemical species at each location, due to the large spatial heterogeneity in gas phase concentrations, especially for NH_3. The requirement for local measurements of wet deposition was relaxed because this is much less spatially variable. For both dry and wet components, measurements had to be complemented by models, either to calculate fluxes based on local concentration data at each site, or to obtain local estimates from a large-scale CTM when data were missing.

Atmospheric inorganic N_r concentrations, available from the NEU (2013) database, were measured monthly for 2-4 years in the gas phase (NH_3, HNO_3, HONO) and in the aerosol phase (NH_4^+, NO_3^-), using DEnuder for Long-Term Atmospheric sampling (DELTA) systems (Sutton et al., 2001; Tang et al., 2009). Concentrations of nitrogen dioxide (NO_2), not covered by DELTA sampling, were measured by chemiluminescence at a few sites only, and were otherwise taken from gridded concentration outputs of the European-scale EMEP CTM (details given below). The N_r data initially reported in Flechard et al. (2011) covered the first 2 years of the NEU project (2007-2008); here, the data from the entire 4-yr NEU monitoring period (2007-2010) were used and averaged to provide a more robust long-term 4-year estimate of N_r dry deposition. The inferential modelling method was used to calculate dry deposition for N-containing gas and aerosol species, whereby measured ambient N_r concentrations were multiplied by a vegetation-, meteorology- and chemical species-dependent deposition velocity (V_d) (Flechard et al., 2011, 2013; Bertolini et al., 2016; Thimonier et al., 2018). In the case of NH_3, a canopy compensation point scheme was applied in some models, allowing bi-directional exchange between the surface and the atmosphere. Considering notoriously large uncertainties in deposition velocities and large discrepancies between the surface exchange schemes currently used in different CTMs, we tried here to minimise such uncertainties by using the ensemble average dry deposition predicted by four different models, as in Flechard et al. (2011).

The dry deposition of atmospheric organic N_r (ON) species not accounted for by the EMEP model (e.g. amines, urea), and not included in DELTA measurements, can contribute a fraction of total N_r deposition. However, Kanakidou et al. (2016) suggest that particulate ON largely dominates the atmospheric ON load, and for particles the main atmospheric removal

Commentaire [C7]: Stated explicitly that ON dry deposition is not considered

mechanism is through precipitation. Thus, dry deposition of ON is expected to be much smaller than wet deposition of water soluble organic compounds (see below).

For wet deposition, several sources of data were used, and the final wet deposition estimate was derived from the arithmetic mean of the different sources, where available. First, within the NEU project, a survey was made of the available national and/or trans-national (e.g. EMEP, 2013; ICP Forests Level-II, ICP, 2019) wet deposition monitoring network concentration data for inorganic N (NH_4^+, NO_3^-) in the different European countries hosting one or several CEIP/NEU flux sites. These data were checked for consistency and outliers, harmonized, and then spatially interpolated by kriging to provide measurement-based estimates of solute concentrations in rainfall for each of the 40 sites of this study. Wet deposition was then calculated as the product of interpolated concentration times measured precipitation at each site.

Next, thirteen sites (DB1, DB3, DB4, EN4, EN9, EN13, EN14, EB2, EB3, MF1, MF2, SN3, SN8) were identified as lacking local or nearby wet deposition measurements. These sites were equipped for three years (2008-2010) with bulk (open funnel) precipitation samplers (Model B, Rotenkamp, Germany; Dämmgen, 2006), mounted above the canopy or inside a clearing for some of the forest sites, with monthly sample change and analysis. The precipitation samples were stabilized by addition of thymol at the beginning of each exposure period, and were analyzed subsequently for inorganic N_r (NH_4^+ and NO_3^-) as well as SO_4^{2-}, Cl^-, PO_4^{3-}, base cations (Mg^{2+}, Ca^{2+}, K^+, Na^+) and pH. A few other sites (EN2, EN8, EN10, EN16, DB2, SN9) were already equipped with wet-only or bulk precipitation collectors. No correction was applied to the bulk deposition estimates to account for a possible contribution by dry deposition within the sampler glass funnel (e.g. Dämmgen et al., 2005), since there did not appear to be any systematic overestimation compared with wet deposition estimates from the monitoring networks or EMEP data (see Results and Fig. S2), even if a more significant bias may be expected in dry (Mediterranean) regions.

In addition to inorganic nitrogen, the wet deposition of water-soluble organic N_r (WSON) compounds was also investigated in precipitation samples at 16 sites (Cape et al., 2012). However, since WSON data were not available for all sites and the measurements were subject to considerable uncertainties (Cape et al., 2012), and also because the contribution of WSON to total N_r deposition was on average less than 5%, WSON was not included in the final estimates of total N_r deposition.

The last data source was the ca. 50 km x 50 km gridded modelled wet inorganic N_r deposition (also NO_2 concentrations, discussed above), simulated by the European-scale EMEP CTM (Simpson et al., 2006a, 2006b, 2012, 2014) for the years 2007-2010, available from EMEP (2013). The data were downloaded in 2013, and it should be noted that in this data series different model versions were used for the different years. This leads to some uncertainty, especially in the dry deposition estimates, but it is hard to say which model version is the most realistic. Evaluation of the model against measurements over this period has shown quite consistent results for the wet-deposited components and NO_2 concentrations, but the dry deposition rates cannot be evaluated versus actual measurements at the European scale. We chose therefore to make use of all versions and years, giving a small ensemble of simulations.

2.2.2 Soil gaseous and leaching losses

Nitrogen losses to the atmosphere (gaseous emissions) and to groundwater (N leaching) are especially hard to quantify and thus typically cause large uncertainties in ecosystem N budgets. These N_r losses were estimated by direct flux measurements or by indirect empirical methods. Soil NO and N_2O emissions were measured in the field using closed static and dynamic chamber methods, as part of NEU (e.g. EN2, EN10, EN16, DB2, SN3, SN8, SN9) and/or collected from the literature (e.g. EN2, EN10, EN14, EN16, DB2, Pilegaard et al., 2006; long term data at EN2 in Luo et al., 2012). Such data were available for N_2O at seven forest sites and four semi-natural sites, and at five forest sites for NO (Table S6). Manual static chamber N_2O measurements were made manually at a typically bi-weekly (growing season) or monthly (winter half-year) frequency at many sites. Automatic chamber systems, allowing continuous N_2O measurements at a frequency of four times per day, were deployed at EN2, EN10, DB2 and SN3. Fluxes of NO were only measured by automatic dynamic (open) chambers.

Commentaire [c8]: Split sentence to improve readability

Measured fluxes were scaled up to yearly values by linear interpolation or using the arithmetic mean of all flux measurements. There may be considerable uncertainty in the annual flux if gap-filling is based on linear interpolation between discrete values, when flux measurements are made manually and therefore discontinuous and infrequent (Parkin, 2008). This is due to the episodic nature and lognormal distribution of NO and N_2O emissions, observed particularly in fertilized croplands and grasslands. However, this 'episodicity' is less pronounced in semi-natural ecosystems, or at least the magnitude of the episodic fluxes is generally much smaller than in fertilized agro-systems (Barton et al., 2015). The uncertainty in annual emissions estimated in our study from manual chamber measurements is related to the observation frequency (bi-weekly or monthly), and likely larger than in the case of automatic (continuous) chamber measurements.

Direct in situ N_r and non-CO_2 GHG gas flux measurements were unavailable at many sites. These soil N_2O, NO (and also CH_4) fluxes were therefore also estimated, as part of NEU, from empirical temperature and moisture responses of soils. These responses were established in a series of factorial soil incubation experiments in controlled conditions with four levels of temperature (5-20°C) and water-filled pore space (20-80 WFPS%), following the protocol described in Schaufler et al. (2010). Twenty-four undisturbed soil cores (top 5 cm of the mineral soil, Ah horizon) were taken from each of 27 forests and 8 semi-natural sites in spring after soils had warmed up above 8°C for one week in order to guarantee phenological comparability of the different climatic zones. Sampling was conducted in 2008, 2009 and 2010 and cores were sent to a common laboratory at the Federal Research and Training Centre for Forests (BFW, Vienna, Austria) for the controlled environment bioassays, which were carried out straight away. The 5 cm top soil layer was selected as it represents the highest microbial activity and correspondingly high GHG production/consumption rates, although processes in deeper soil layers should not be neglected (Schaufler et al., 2010). Site-specific, empirical bi-variate (T, WFPS) relationships describing soil fluxes for CO_2, N_2O, NO and CH_4 were derived from the incubation results and then applied to multi-annual time series of soil temperature and moisture measured at the sites, mimicking field conditions and providing scaled up estimates of potential annual trace gas emissions.

Leaching of dissolved inorganic nitrogen (DIN = NH_4^+ + NO_3^-) was measured using lysimeter setups, or estimated from a combination of suction cup measurements (typically ~1m soil depth) and a hydrological drainage model, at a few sites during the NEU monitoring period (EN2, EN4, EN10, EN15, EN16, DB1, DB2) and as part of parallel projects (EN8, DB4). One-dimensional (1-D) drainage models were based on the soil water balance equation using evapotranspiration, observed precipitation and changes in soil water content (Kindler et al., 2011; Gielen et al., 2011). For the forest sites where no leaching measurements were available, the empirical algorithm by Dise et al. (2009) was applied to predict DIN leaching based on key variables (throughfall inorganic N_r deposition DIN_{TF}, organic horizon C/N ratios, MAT). The algorithm, developed from the extensive Indicators of Forest Ecosystem Functioning (IFEF) database (>300 European forest sites), simulates the non-linearity of DIN leaching with respect to DIN_{TF} and soil C/N ratio, with critical thresholds for the onset of leaching of DIN_{TF} = 0.8 g (N) m^{-2} yr^{-1} and C/N = 23, respectively. Since the algorithm requires DIN_{TF} as input, as opposed to total (above canopy) N_{dep}, in the present study we applied a reduction factor of 0.85 from N_{dep} to DIN_{TF} (i.e. a canopy retention of 15% of atmospheric N), which was calculated as the average of all available individual DIN_{TF} / N_{dep} ratios in the IFEF database. A comparison with values of DIN_{TF} / N_{dep} ratios actually measured at the EN2, EN8, EN10, EN16 and DB2 sites (0.71, 0.80, 0.29, 0.85, 1.11, respectively; mean ± st. dev. 0.75 ± 0.30) shows that the applied ratio of 0.85 is plausible but also that much variability in canopy retention/leaching may be expected between sites.

2.3 Carbon fluxes

2.3.1 Ecosystem-atmosphere CO_2 exchange

Half-hourly rates of net ecosystem-atmosphere CO_2 exchange (NEE) were measured over several years (on average 5 years; see Table S6) by the eddy covariance (EC) technique at all sites. The long term net ecosystem productivity (NEP) is defined following Chapin et al. (2006) as the difference between gross primary production (GPP) and ecosystem respiration (R_{eco}),

Commentaire [c9]: Added text to address issue of uncertainty in gaseous emissions, related to sampling frequency and gap-filling by linear interpolation.

Commentaire [c10]: Split sentence to improve readability

Commentaire [C11]: Short sentence to clarify what type of drainage models are used

and thus calculated as the straightforward annual sum of NEE fluxes (with opposite sign). The net ecosystem carbon balance (NECB) may differ from the NEP if C fluxes other than assimilation and respiration, such as DIC/DOC leaching, CH_4 and other volatile organic compound (VOC) emissions, as well as lateral fluxes (harvest, thinning) and other disturbances (fire), are significant over the long term (Chapin et al., 2006). 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. Previous studies have normalised C flux data through the carbon use efficiency (CUE), commonly defined from a plant's perspective as the ratio of net to gross primary productivity (NPP/GPP), or the biomass production efficiency (BPE = BP/GPP; Vicca et al., 2012), which is a CUE proxy. By analogy, we define here an ecosystem-scale, medium-term indicator of carbon sequestration efficiency (CSE) as the NEP/GPP ratio, calculated from measurable fluxes over the CEIP/NEU project observation periods.

Commentaire [c12]: Introduce definition of CSE in Methods, by analogy to CUE

The EC technique is based on fast-response (sampling rates typically 10-20Hz) open-path or closed-path infra-red gas analyzer (IRGA) measurements of turbulent fluctuations in CO_2 concentration (c) in the surface layer above the ecosystem, coupled with ultra-sonic anemometer measurements of the three components of wind (u, v, w) and temperature. The NEE flux is calculated as the average product of c and w fluctuations, i.e. the covariance (Swinbank, 1951; Lee et al., 2004).

The EC-CO_2 flux measurements reported here followed the protocols established during the CEIP project, largely based on the EUROFLUX methodology (Aubinet et al., 2000). Briefly, post-processing of the raw high frequency EC data included typically: de-spiking to remove outliers; 2-D rotation of the coordinate system; time lag optimization by maximization of the covariance between CO_2 concentration and vertical component of wind speed (w); block-averaging over the flux averaging interval of 30 minutes. Corrections were applied for various methodological artefacts, including notably i) flux losses at the different frequencies of flux-carrying eddies, caused e.g. by attenuation/damping in the inlet/tubing system (Ibrom et al. 2007; Fratini et al. 2012), path averaging, sensor separation, analyzer response time, high- and low-pass filtering; ii) effects of temperature fluctuations and dilution by water vapor on measured fluctuations in concentrations of CO_2 (Webb-Pearman-Leuning corrections; Webb et al., 1980); iii) CO_2 storage below sensor height. Quality assurance and quality control procedures were further developed and agreed upon within CEIP, including statistical tests, non-stationarity, integral turbulence characteristics (Foken et al., 2004), and footprint evaluation (Göckede et al., 2008). Friction velocity (u_*) threshold filtering was implemented using the moving point test according to Papale et al. (2006) and as described in REddyProc (2019), in order to discard flux data from periods of low turbulence.

Different EC post-processing softwares were used at the different sites within the project, such that the data were not evaluated in exactly the same way across the CEIP network, but a reasonably good overall agreement was found among the different softwares, within 5-10% difference for 30-minute CO_2 flux values (Mauder et al., 2008; Mammarella et al., 2016).

Similarly, for the gap-filling of the 30-minute flux time series, during periods of instrument malfunction or unsuitable measurement conditions (low turbulence, insufficient fetch, etc.), and for the partitioning of NEP into GPP and R_{eco}, a number of alternative algorithms have been developed in the past, based on different sets of principles (Falge et al., 2001; Barr et al., 2004; Reichstein et al., 2005; Lasslop et al., 2010). The gap-filling and partitioning algorithm used by default in this study was the generic online REddyProc (2019) software, implemented also in the European Fluxes Database Cluster. REddyProc was based on i) Reichstein et al. (2005) for the filling of gaps in the NEE flux data on the basis of information from environmental conditions; ii) Reichstein et al. (2005) for the nighttime data based R_{eco} parameterization (using an Arrhenius-type function of temperature); and iii) on Lasslop et al. (2010) for the daytime data based GPP evaluation (using a rectangular hyperbolic light–response curve for NEE and including a temperature sensitivity of respiration and limitation of GPP by vapour pressure deficit).

In this study, for all CEIP flux sites, we have retrieved the fully analysed and validated half-hourly (level-3) and daily to annual (level-4) CO_2 flux (NEP, GPP, R_{eco}) data as available, initially from the CEIP database, later from the European Fluxes Database Cluster (2012) or from the GHG-Europe portal (GHG-Europe, 2012). For these data, although the

evaluation methods were not necessarily harmonized between sites, we hold that the data available in the database were obtained using the best possible, state-of-the-art evaluation methods at the time of retrieval. For the four non-CEIP flux sites, flux evaluation closely followed CEIP protocols; in the case of DB4 the EddyPro (v6.2) software was used, which was based on a synthesis of calculation and correction methods from CEIP and other FLUXNET flux networks around the globe.

The EC-CO_2 flux measurements used in this study mostly spanned the 5-year period of CEIP (2004-2008), except for a dozen sites where measurements continued until 2010, i.e. the end of NEU and of atmospheric N_r sampling. Older EC data (since the mid-late 1990's) were also available at DB5, EN6 and EN13. Data collection started and ended later at DB4, at which both EC-CO_2 flux and DELTA-N_r measurements spanned the 7-year period 2009-2015. Data analyses presented in the paper, based on inter-annual mean CO_2 budgets and mean N_r deposition, assume that five or more years of monitoring yield reasonably robust estimates of long-term fluxes for the different sites, and that the small time shift between CEIP and NEU project periods (2-3 year overlap) does not affect the results significantly. At some sites, such as DB2, long-term NEE measurements showed multi-decadal variations (Pilegaard et al. 2011; Wu et al. 2013), thus it was essential to use the years overlapping with NEU.

2.3.2 Soil CO_2 and CH_4 fluxes

In situ soil CO_2 efflux (SCE) measurements by opaque (static or dynamic) manual chambers were carried out at 24 of the forest sites, with typically weekly to monthly sampling frequency, with fluxes being measured continuously (hourly) by automated chambers at a few sites (e.g. EN2). The SCE is usually considered a proxy for CO_2 production by soil respiration (R_{soil}), though the two may not be equal as part of the CO_2 production is dissolved into pore water and may reach the atmosphere only later, either on-site, or even off-site if dissolved CO_2 (DIC) leaches to groundwater. Annual R_{soil} data, scaled-up from SCE measurements, are available for 18 forest sites and were collected from the CEIP or GHG Europe databases and/or from various peer-reviewed publications for the different sites (see Table S7). The ratio of heterotrophic respiration (R_{het}) to R_{soil} was determined on an annual scale at 15 sites by different techniques (root-exclusion meshes, trenching experiments, radiocarbon or stable isotope tracing, tree girdling; e.g. Subke et al., 2006) (Table S7).

Methane fluxes were measured by chamber methods or eddy covariance at six forest sites and five semi-natural (peatland, wetland) sites (Hendriks et al., 2007; Skiba et al., 2009; Drewer et al., 2010; Shvaleva et al., 2011; Luo et al., 2012; Kowalska et al., 2013; Juszczak and Augustin 2013) (Table S6). These data were complemented by bioassay measurements of CH_4 emission or uptake (net oxidation) by the laboratory soil cores, as described previously for NO and N_2O estimates (Schauffler et al., 2010).

2.3.3 Dissolved carbon losses

Dissolved inorganic (excluding CO_2 from weathering of carbonate rocks) and organic carbon (DIC/DOC) fluxes were measured at six forest sites (DB1, DB2, EN4, EN8, EN10, EN15), using suction cups for sampling soil water and combined with soil drainage data, or by monitoring water runoff through weirs, as part of CEIP, NEU and other projects (Ilvesniemi et al., 2009; Kindler et al., 2011; Gielen et al., 2011; Verstraeten et al., 2014). Data were also available for peatland at SN7, with DIC, DOC and also dissolved CH_4 concentrations in pore water of the clayey peat, in groundwater from the sand aquifer and in ditch water, as described in Hendriks et al. (2007). For the peatland within SN9, Dinsmore et al. (2010) measured stream concentrations and export of DIC, DOC as well as particulate organic carbon (POC), and also estimated stream evasion of CO_2, CH_4 and N_2O in addition to the land-based flux (EC, chamber) measurements in the tower footprint.

2.4 Ecosystem greenhouse gas balance

Net GHG budgets were constructed from inter-annual mean EC-based NEP combined with measured and scaled up N_2O and CH_4 fluxes wherever available (nine and six sites, respectively), or with bioassay-derived fluxes (most sites) or modelled

data (BASFOR, forests/N_2O only), using 100-yr global warming potentials (GWP) of 265 and 28 for N_2O and CH_4, respectively (Fifth Assessment Report, IPCC, 2013). The sign convention for non-CO_2 GHG fluxes and for the net ecosystem GHG balance in this paper adopts an atmospheric warming perspective, i.e. positive fluxes for emissions toward the atmosphere (warming), negative for uptake by the surface (cooling).

2.5 Ancillary soil, plant and ecosystem measurements

Ancillary data were collected mainly for the purpose of assembling input parameters and calibration datasets for forest ecosystem (BASFOR) modelling (see below). Texture (% clay, % sand, % silt), pH, soil organic carbon concentration (SOC) and C/N ratios were measured in soils of 35 sites as part of the bioassay experiments described previously, but were otherwise also documented in the CEIP database and in papers previously published for the majority of sites (Table S1). For the forest sites, ecosystem data for soil water content (SWC), porosity, saturation water content (Φ_{SAT}), field capacity (Φ_{FC}) and wilting point (Φ_{WP}), and for canopy height (H), leaf area index (LAI), diameter at breast height (DBH), basal area (BA), number of trees per unit area or stand density (SD) and thinning events, were obtained from CEIP and other project (e.g. FLUXNET) databases and complemented by various publications (Tables S2-S5). Such was also the case for ecosystem carbon stocks in soil organic matter (CSOM) and in roots (CR), stems (CS), branches (CB), leaves (CL) and litter layers (CLITT), for which the global database assembled by Luyssaert et al. (2007) provided additional data. At sites for which published values of Φ_{FC} and Φ_{WP} were not available, default estimates were inferred from soil texture by means of van Genuchten (1980) pedo-transfer functions, using tabulated values from the German soil description handbook (Eckelmann et al., 2005)

Foliar C and N contents (LeafC, LeafN) were measured as part of NEU for EN1, EN2, EN5, EN8, EN10, EN15, EN16, DB2 (Wang et al., 2013), DB4, SN3, SN4, SN8 and SN9, or were otherwise taken from CEIP, GHG Europe and FLUXNET databases as well as various publications; in total, leaf C/N measurements were available for 31 sites. By contrast, data were much rarer for C/N ratios for other compartments of the forest ecosystem, with data available at only 15 sites for litter, and only five sites for roots, stems and branches.

2.6 Modelling of C and N fluxes and pools by the BASFOR ecosystem model

The BASic FORest model (BASFOR) is a deterministic forest ecosystem model that simulates the growth (from planting or natural regeneration) and the biogeochemistry of temperate deciduous and coniferous even-aged stands (van Oijen et al., 2005). A description of the model and the fortran code are available in BASFOR (2016). The model was calibrated through a multiple site Bayesian calibration (BC) procedure, applied to three groups of plant functional types (DBF, ENF-spruce, ENF-pine), based on C/N/H_2O flux and pool data from the CEIP/NEU databases (Cameron et al., 2018). Details on model implementation as part of this study are provided in Flechard et al. (2020).

Briefly, the C, N and water cycles are simulated at a daily time step in interaction with the soil and climate environments and constrained by management (pruning and thinning). Carbon and nitrogen pools are simulated in the different ecosystem compartments (tree stems, branches, leaves and roots, litter layers and SOM with fast and slow turnover), which are inter-connected by internal flows and transformations (e.g. SOM mineralization, nitrogen retranslocation). Carbon, nitrogen and water enter the ecosystem from the atmosphere (photosynthesis, N_r deposition, rainfall). Inorganic nitrogen is taken up from the soil by tree roots; C and N return to the litter and soil pools upon senescence of leaves, branches and roots, and also when trees are pruned or thinned. Losses of C occur through autotrophic (root and shoot) respiration and microbial decomposition into CO_2 of litter and SOM (heterotrophic respiration); losses of N occur through nitrate leaching below the root zone and soil emissions to the atmosphere of NO and N_2O. The water balance is constrained by incoming rainfall, soil water holding capacity, and evapotranspiration (ET) simulated by the Penman equation.

Commentaire [C13]: Much shorter description of BASFOR model than in original version.

(Detailed description has moved to the companion paper II, together with Figs S4-S5 of Supplement)

3 Results

3.1 Nitrogen inputs and outputs

3.1.1 Nitrogen deposition

Total inorganic N_r deposition ranged from 0.1 to 4.3 g (N) m^{-2} yr^{-1} across the CEIP/NEU networks (Table 1), with the largest values observed in The Netherlands, northern Belgium and southern Germany, and the lowest levels observed at latitudes > 60°N (Fennoscandia). Nitrogen deposition was dominated by the dry fraction in forests (Fig. 2), with an average contribution to total deposition of 63% versus 39% for short semi-natural vegetation. This contribution was even larger (> 2/3) for high deposition sites (N_{dep} > 2 g (N) m^{-2} yr^{-1}). Total N_{dep} was more strongly correlated to dry deposition across all sites (R^2 = 0.94) than to wet deposition (R^2 = 0.56). Important differences in the ratio of dry to wet deposition are evident across climatic regions, with the share of dry deposition being especially large at Mediterranean sites (e.g. Sanz et al., 2002), where annual rainfall is smaller. However, the share of dry deposition was also large for sites that are located near (large) anthropogenic (industrial, vehicular, agricultural) N_r emission sources. Total N_r deposition was around 25% smaller on average at short semi-natural vegetation sites compared with forests (Fig. S2), even though the mean total atmospheric N_r concentrations (reduced and oxidized, N-containing gas and aerosol compounds) were quite similar between the two data sets (Flechard et al., 2011). The difference was driven by higher dry deposition rates over forests due to higher aerodynamic roughness and deposition velocities (Fig. S3; see also Schwede et al., 2018). Reduced N_r (NH_3 gas and NH_4^+ in aerosol and rain, collectively NH_x) contributed on average 56% of total deposition; oxidised N_r (HNO_3 + NO_2 gas and NO_3^- in aerosol and rain, collectively NO_y) was dominant at only six forest sites of the network (EN7, EN10, EN18, EB2, SN3, SN5; Fig. 2).

For comparison, dry deposition, calculated here as the ensemble average of four inferential model estimates based on in situ N_r concentration measurements, was on average more than a factor of two larger than the ca. 50 km x 50 km grid square-averaged EMEP model estimate (taken from EMEP, 2013) (see Fig. S2). However, since each EMEP grid square contains variable proportions of different land uses with different deposition velocities, it is more meaningful to compare DELTA-based inferential estimates for each study site with ecosystem-specific EMEP dry deposition rates in the relevant grid squares. In this case, the EMEP dry deposition rates are 32% smaller than the inferential estimates.

By contrast, wet deposition was generally reasonably consistent between the different data sources for inorganic N_r (in situ bulk or wet-only measurement, kriging of monitoring network data, EMEP model output). For the 18 sites where all three sources of data were available, the mean CV of the three estimates was 21% (range 2%-56%, with 15 CV values out of 18 below 30%), and the mean (± 95% conf. int.) wet deposition estimates across the 18 sites were 0.63 ±0.14, 0.64 ±0.15 and 0.68 ±0.16 g (N) m^{-2} yr^{-1} for the three methods, respectively (Fig. S2), showing no systematic bias between mehtods. Wet deposition of organic nitrogen (WSON), measured at 16 sites, represented on average 11% (range 2-36%) of total inorganic + organic wet deposition (Fig. S2), but only 4% (range 1-30%) of total dry + wet N_r deposition, since total N_{dep} was dominated by dry deposition at most forest sites.

{Insert Figure 2 here}

3.1.2 Nitrogen losses

Total ecosystem losses of inorganic N_r were computed for the forest sites as the sum of DIN leaching and NO and N_2O emissions (Fig. 3 A-D). We assumed that NH_3 emissions by soil and vegetation were negligible due to generally acidic forest soils, as well as low values of stomatal compensation point (the leaf NH_3 emission potential), respectively (Flechard et al., 2013). Inorganic N_r losses (Fig. 3D) increased sharply with N_r deposition and were largely dominated by DIN leaching at N_{dep} levels above 2 g (N) m^{-2} yr^{-1} (Fig. 3C). For these large N_{dep} levels, the fraction of deposited N_r lost as DIN, NO or N_2O was generally larger than 50% (Fig. 3F). The inorganic N_r balance (N_r deposition minus NO, N_2O and DIN losses) was probably still positive for most sites (Fig. 3E), although the confidence intervals of the budget term (accounting for

Commentaire [c14]: Split sentence for improved readability

uncertainties in all terms including deposition) were very large for the elevated N_r deposition sites. Note that the DIN leaching estimate by BASFOR, shown for comparison on Fig. 3C, was not used in the calculation of total inorganic N losses in Fig. 3D; this is because BASFOR does not simulate N_2 loss by denitrification, and thus part of the soil N surplus that would in reality denitrify is assumed to drain, resulting in an over-estimation of the leaching term, though not necessarily of the total N losses.

Emissions of NO estimated from bioassay measurements (Schaufler et al., 2010) and by BASFOR modelling were generally of the same order in forests (average values across all forest sites of 0.22 and 0.21 g (N) m^{-2} yr^{-1}, respectively), but validation by in situ chamber flux data was difficult owing to the limited number of available measurements (only five forest sites, mean value 0.27 g (N) m^{-2} yr^{-1}). Nonetheless, the largest NO emissions by the three methods were all found at N_{dep} levels above 2 g (N) m^{-2} yr^{-1} (Fig. 3A). By contrast, N_2O emissions did not show any marked dependence on N_{dep} and were on average smaller than NO emissions by a factor of two to five, with mean values across all sites of 0.12, 0.08 and 0.04 g (N) m^{-2} yr^{-1} for bioassay, BASFOR and chamber fluxes, respectively. The mean N_2O fluxes (averaged over the different methods) were larger than mean NO fluxes at only one third of the forests sites; by contrast, at SN sites N_2O emissions were larger than NO emissions at all but one location. The dominance of NO over N_2O in forests could in principle reflect the generally well aerated conditions of (especially coniferous) forest litter layers on well-drained top soils, more conducive to NO formation by nitrification than N_2O by denitrification (Davidson et al., 2000; Pilegaard et al., 2006). This would be perhaps especially true for the four highest (>3 g (N) m^{-2} yr^{-1}) N_r deposition sites (EN2, EN8, EN15, EN16, all coniferous forests) with the highest NO emissions (Fig. 3), which all had sand-dominated (64-96%) soil textures (Table S4). On the other hand, given the acidity of many forest top soils (Table S4), nitrification could be inhibited, but chemodenitrification could produce significant amounts of NO (Pilegaard, 2013).

For a complete ecosystem net N budget, additional measurements of dissolved organic nitrogen (DON) leaching, as well as dinitrogen (N_2) fluxes (biological fixation and total denitrification) would be required (Fig. 1), but they were not quantified in most cases. A tentative ballpark estimate of the potential magnitude of denitrification N_2 emissions for the DB2 forest site may be calculated by considering the mean N_2/N_2O ratio of 74 (± 0.85 st. err.), which was measured in He-O_2 mixture soil incubation experiments performed on DB2 soil cores (unpublished data). This mean ratio, multiplied by the mean field measured N_2O emission flux of 0.074 g (N_2O-N) m^{-2} yr^{-1} (Pilegaard et al., 2006), yields an estimate of the order of 5.5 g (N_2-N) m^{-2} yr^{-1}. There is considerable uncertainty in this number, since the mean N_2/N_2O ratio was calculated from short-term investigations in the laboratory, which may or may not be representative of the prevailing soil and weather conditions in the field. This uncertainty is reinforced by the low sensitivity of the N_2 detector, which was a factor of 20-80 lower than that of the N_2O detector used in the experiment (Buchen et al., 2019). Another estimate of forest soil denitrification loss obtained through a soil core incubation method was given by Butterbach-Bahl et al. (2002) for the EN2 spruce site, with an annual N_2 emission flux of 0.72 g (N_2-N) m^{-2} yr^{-1} and a mean N_2/N_2O ratio of 7. The N_2 emissions thus estimated suggest that total denitrification may be a very significant term in the total N budget of forests, possibly of the same order as atmospheric N_r deposition.

Measurements of DON leaching were available at very few sites, but proved to be significant. At the pine forest site of EN8, DON leaching was of the order of 0.3 g (N) m^{-2} yr^{-1}, i.e. a factor of three lower than DIN losses (Verstraeten et al., 2014). At the beech forest site of DB2, DIN and DON leaching were of the same order (0.07-0.08 g (N) m^{-2} yr^{-1}), but both very small in comparison to N_{dep} (2.15 g (N) m^{-2} yr^{-1}); while at the pine forest site of EN10 the leaching/runoff N_r loss was actually dominated by DON (0.012 g (N) m^{-2} yr^{-1}), which was around an order of magnitude larger than DIN leaching (Korhonen et al., 2013) and a factor of four smaller than N_{dep}.

{Insert Figure 3 here}

3.2 Net carbon and greenhouse gas balance

3.2.1 Spatial variability of the carbon sink in relation to climate and nitrogen deposition

The ultimate objective of the project was to quantify the response of C sequestration to atmospheric N_r deposition (addressed in Flechard et al., 2020), but this is not straightforward. We follow first in this paper a descriptive approach, whereby variations of C fluxes and other productivity indicators (*e.g.* leaf area index and N content) are examined graphically as a function of N_{dep} (Fig. 4). However, this is done with the strong reservation that a simple empirical relationship does not necessarily prove causality, as other confounding and co-varying factors, *e.g.*, climate, soil, age, etc, may exist. Figures 4-5 show for example that the large inter-site differences in MAT and MAP at the European scale also need to be considered, beside the variability in N_{dep}. Note that in assessing the variability of ecosystem carbon sink strength within the network, we use EC-derived NEP (the long term NEE sum) as a proxy for the net ecosystem carbon balance (NECB), because estimates of DIC/DOC leaching, CH_4 emissions and other C loss processes were not systematically measured at all sites.

Inter-annual mean NEP ranged from a small net source of -70 g (C) m^{-2} yr^{-1} (EN6, a waterlogged peat-based spruce stand in the southern Russian taiga) to a large net sink of +826 g (C) m^{-2} yr^{-1} (EN5, upland spruce forest in N. Italy) (Table 1, Fig. 4C); GPP ranged from 377 g (C) m^{-2} yr^{-1} (SN3, a boreal peatland site with the lowest MAT = -0.6°C) to 2256 g (C) m^{-2} yr^{-1} (EN14, a pine stand in Italy, one of the warmest sites with MAT of 14.9 °C and non-limiting rainfall with MAP = 920 mm) (Fig. 4A). Ecosystem respiration peaked at 1767 g (C) m^{-2} yr^{-1} at EN4 (upland spruce forest in E. Germany) and was lowest at 345 g (C) m^{-2} yr^{-1} at SN3 (boreal peatland), the coldest site (Fig. 4B); R_{eco} was strongly and positively related to GPP (Fig. 4F) (R^2 = 0.62, slope = 0.64). The resulting carbon sequestration efficiency values based on the ratio of observed NEP/GPP (CSE_{obs}) varied widely among observation sites, ranging from -9 to 61%, with an average of 25%.

The data show a positive correlation between GPP and N_{dep} in the range 0-2.5 g (N) m^{-2} yr^{-1} (R^2 = 0.55, $p < 0.01$). By contrast the five sites with N_{dep} > 2.5 g (N) m^{-2} yr^{-1} tend to show visually an inverse relationship (Fig. 4A), despite the fact that they lie in comparatively favourable climates. Similar patterns are observed for R_{eco} and NEP (Fig. 4B-C), but with much larger scatter and lower R^2 (0.24, $p < 0.01$, and 0.30, $p < 0.01$, respectively, for the N_{dep} range 0-2.5 g (N) m^{-2} yr^{-1}), with the same apparent decline for higher deposition sites. However, a closer inspection of Fig. 4A-C reveals a potential cross-correlation with climate (see also Fig. S4): i) the lower end of the N_{dep} range, coinciding with the lowest GPP, R_{eco} and NEP, also coincides with the lowest MAT and MAP (e.g. Finnish sites); and ii) the sites in the intermediate N_{dep} range (1.5-2.5 g (N) m^{-2} yr^{-1}), coinciding mostly with the largest observed GPP values (>1500 g (C) m^{-2} yr^{-1}), were on average 1.8°C warmer (10.2 vs. 8.4 °C) and 89 mm yr^{-1} wetter (887 vs. 798 mm) than the sites in the lower N_{dep} range (0-1.5 g (N) m^{-2} yr^{-1}). Other proxies of the ecosystem C and N cycles and productivity, such as the LAI (defined as 1-sided for broad-leaf, or half of total for needle-leaf; Table 1 and Fig. 4D) and the foliar N content (LeafN, Fig. 4E), also showed positive relationships to N_{dep} (see below for differences between vegetation types). The inter-annual mean value of the annual maximum leaf area index (LAI_{max}) increased from around 1 to 7 m^2 m^{-2} for N_{dep} increasing from 0.1 to 4.5 g (N) m^{-2} yr^{-1}, with the lower half of the LAI_{max} distribution (< 4.5 m^2 m^{-2}) mostly occurring at boreal, Mediterranean and upland sites and thus under temperature and/or water limitations.

{Insert Figure 4 here}

Clearly, therefore, the continental-scale variability in ecosystem/atmosphere CO_2 fluxes was to a large extent controlled by climate, namely by limitations in temperature and water availability. Gross ecosystem productivity was limited, as expected, by low temperatures at high latitudes (or high elevations) and by low rainfall and/or high evaporative demand at Mediterranean, boreal and continental sites. The distribution of the forest monitoring sites in the European climate space, with MAP and MAT on the x and y axes, respectively (Fig. 5A, 5B), shows that for sites with MAT > 7 °C there was a broad negative correlation between MAT and MAP (R2 = 0.24, p = 0.01) i.e. the warmest sites in southern Europe tend to be the

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driest and therefore potentially water-limited. Maximum GPP (and also R_{eco}, not shown) occurred in the mid-climate range, around 9-15 °C MAT and around 700-1000 mm MAP. Similarly, the larger N_{dep} values (> 2 g (N) m^{-2} yr^{-1}) occurred almost exclusively at sites with MAT in the narrow range of 6-11°C, and although these large N_{dep} values were found in a broad MAP range (550-1200 mm), they peaked sharply around 800-900 mm MAP (Fig. 5A; Fig. S4). Modelled N_{dep} values from the EMEP CTM (Fig. 5C, 5D) show that this is a generic pattern at the European scale.

{Insert Figure 5 here}

Ecosystem DIC + DOC losses estimated by Kindler et al. (2011) for 4 forest sites of this study (DB1, DB2, EN4, EN15) were on average 13 ± 7 g (C) m^{-2} yr^{-1} (range 3-35 g (C) m^{-2} yr^{-1}), with contributions by DIC to total (DIC + DOC) losses varying between 18% and 83%. By contrast, Gielen et al. (2011) estimated DOC leaching losses of 10 ± 2 g (C) m^{-2} yr^{-1} for the EN8 pine stand on an acidic sandy soil, in which DIC concentrations in soil water were negligibly small. Ilvesniemi et al. (2009) found DOC losses in runoff at EN10 of 0.8 g (C) m^{-2} yr^{-1}, which was negligible compared with NEP. These leaching or runoff losses of DOC and DIC were on average over all forest sites equivalent to a very small mean fraction of 0.6% of GPP (range 0.1-1.9%), but a more significant fraction of NEP (mean 6%, range 0.3-13%). At the SN7 peatland site, fluxes of total dissolved carbon (including CH_4) through seepage, infiltration and drainage were relatively small by comparison to NEP and to other peat bogs (17 g (C) m^{-2} yr^{-1}, only 5% of NEP) (Hendriks et al., 2007); by contrast, at the SN9 peatland site, net stream C export (including DIC, DOC and POC) was on average 29.1 g (C) m^{-2} yr^{-1} (81% of which being DOC), equivalent to a mean leached fraction of 37% of NEP (Dinsmore et al., 2010).

3.2.2 Differences between plant functional types

Forests (F) and short semi-natural (SN) vegetation showed similar relationships with GPP as a function of N_r deposition, increasing with a broadly similar slope at low N_{dep} values, then levelling off beyond 2g (N) m^{-2} yr^{-1}, except for the fact that GPP was lower by typically 200-500 g (C) m^{-2} yr^{-1} in SN compared with F sites, for a given N_{dep} level (Fig. 4). The behaviour was different for NEP, where the slope against N_{dep} in the range 0-2 g (N) m^{-2} yr^{-1} was much steeper for F than for SN, which occurred because R_{eco} values are of the same order for F and SN at a given N_{dep} level. No systematic difference was observed between the forest PFT, based on the available data, in the apparent relationships of the C fluxes *vs.* N_{dep}. However, this may be a result of the small number - and large diversity - of deciduous broadleaf (DB) and evergreen broadleaf (EB) forest sites in the dataset, compared with evergreen needleleaf (EN) sites (Table 1).

The relationship of LeafN to N_{dep} (Fig. 4E) showed three distinct groups, with the smallest values (0.8-1.8 % N in dry weight, DW) for evergreen needleleaf and broadleaf (EN, EB) forests being positively correlated to N_{dep} in the range 0.5-4.3 g (N) m^{-2} yr^{-1} (R^2 = 0.71, $p < 0.01$). Values for short semi-natural (SN) vegetation were found in an intermediate range (1-2.7 % N DW), with a steep and significant relationship to N_{dep} (R^2 = 0.51, $p < 0.05$). The largest values occurred for deciduous broadleaf (DB) forests (mostly >2 % N DW), but with little relationship to N_{dep} (R^2 = 0.18, not significant). Seasonal variations in forest LeafN could reach a factor of 2, as did differences between tree species within the same forest, which may account for some of the scatter observed in Fig. 4E.

3.2.3 Carbon fluxes and pools derived from forest ecosystem modelling

In the BASFOR base run (Fig. 6), reasonable overall model performance was achieved for GPP, ecosystem C pools, H, DBH, LAI and LeafN, while more scatter was present for R_{eco}, NEP and ET. In particular, in apparent contrast to GPP, R_{eco} stands out as a more challenging variable to model. Predictably, because BASFOR was calibrated using a subset of 22 sites from this dataset (Cameron et al., 2018), the range and mean values of modelled R_{eco} were close to mean observations by EC across the study sites, but differences between sites were poorly reproduced with much scatter around the 1/1 line and a low R^2. The modelled carbon sequestration efficiency (CSE_{mod}), simulated over the same time period as the flux measurements, was much less variable (range 17-31%, mean 22%) than observation-based values (CSE_{obs}) (comparison made for the 22

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sites used in model calibration). One possible reason was that BASFOR assumed that autotrophic respiration (R_{aut}) is a constant fraction of GPP, which may be an over-simplification (Collalti and Prentice, 2019). Also, heterotrophic respiration (R_{het}) appeared to be a much more variable fraction of R_{eco} in reality (Table S7) than was predicted by the model, leading to sizeable divergence in the overall modelled R_{eco}. As the direct measurement, NEP was the least uncertain term in EC-derived data, compared with GPP and daytime R_{eco}, which were inferred from measured (half-hourly) EC-NEE by empirical partitioning models. By contrast, in BASFOR, NEP was calculated as the residual between two large numbers (GPP and R_{eco}) and thus compounds the uncertainties of both component terms. The modelled result for NEP appeared to be an over-estimation of net C uptake at low productivity sites and an under-estimation at high productivity ones (slope <1). A broadly similar pattern emerged for ET.

{Insert Figure 6 here}

3.2.4 Net ecosystem greenhouse gas budgets

Carbon dioxide largely dominated the net GHG budget at all forest sites, with only three sites where either N_2O or CH_4 GWP-equivalent fluxes were larger than 10% of NEP in absolute terms (Fig. 7). Most of the forest soils (22 out of 27 sites) investigated in the bioassay experiment behaved as small net sinks for CH_4, with a mean (± st. err.) net oxidation flux of -0.14 ± 0.03 g (C) m^{-2} yr^{-1} (range -0.61 to +0.16 g (C) m^{-2} yr^{-1}). The mean CH_4 flux measured by soil chambers at the 6 forest sites where such measurements were available (EN2, EN6, EN10, EN16, DB2, EB5) was also a net oxidation flux of -0.32 ± 0.15 g (C) m^{-2} yr^{-1} (range -1.0 to -0.0 g (C) m^{-2} yr^{-1}). For these 6 sites, there was a significant correlation (R^2 = 0.74, $p < 0.05$) between annual soil CH_4 flux estimates derived from the bioassay experiment and from in situ flux measurements (Figure S5 in Supplement), with the largest net annual soil CH_4 uptake flux being observed by both methods at the EN10 pine forest site (Skiba et al., 2009). By contrast, at the elevated N_{dep} sites EN2 and EN16, the net soil CH_4 flux was close to zero, consistent with previous research (e.g. Steudler et al., 1989; Smith et al., 2000) showing that the CH_4 oxidation capacity of forest soils in negatively affected by N_r addition or deposition. In terms of C uptake, soil CH_4 oxidation was negligible compared to CO_2 fluxes, representing on average only 0.1% of NEP (range 0.0-0.4%). In terms of GWP the CH_4 flux was larger, being equivalent to 0.8% of NEP (range 0-4.5%), but on average still a factor of three smaller than the warming by N_2O emissions equal to 3.9% of NEP (range 0-18.5%).

By contrast to forests, at semi-natural, short vegetation sites N_2O or CH_4 emissions had a larger impact on the net GHG balance, where most (seven out of nine) sites showed non-CO_2 GHG contributions larger than 10% of NEP. Three of these seven sites were unfertilised, extensively grazed upland (SN2, SN5, SN6) grasslands (small N_2O sources), while three sites (SN3, SN7, SN8) were CH_4-emitting peatlands or wetlands (EC-CH_4 and chamber flux data from Drewer et al., 2010; Hendriks et al., 2007; Juszczak and Augustin, 2013 and Kowalska et al., 2013). At SN3 and SN8, the small to moderate NEP sinks were turned by large CH_4 emissions into net GHG sources (net warming budgets of +127 and +242 g CO_2-C Eq m^{-2} yr^{-1}, respectively), though not into actual net C sources (Fig. 7). At SN8, CH_4 emissions generally ranged from 25-45 g CH_4-C m^{-2} yr^{-1} but reached 86 g CH_4-C m^{-2} yr^{-1} during a particularly wet year, when the whole area was flooded. At the SN9 peatland site, Dinsmore et al. (2010) calculated that stream GHG evasion – at the scale of the 335-ha peatbog encompassing the flux tower footprint – together with downstream export represented 50-60 g CO_2-Eq m^{-2} yr^{-1} (13-16 g CO_2-C Eq m^{-2} yr^{-1}), 96% of which being de-gassed CO_2, i.e. in the range 11-23% of the GHG budget from the tower footprint.

{Insert Figure 7 here}

4 Discussion

Previous observations of simple empirical relationships found between N deposition and forest productivity have been criticized for, amongst other things, their low number of replications, unreasonably high sensitivities of productivity to N

additions, and limitations of the data and simplistic univariate statistical approaches used (Magnani et al., 2007; Högberg, 2007; de Vries et al., 2008; Sutton et al., 2008). Other attempts have subsequently been made to assess impacts of N deposition on forest growth and carbon sequestration, while accounting for other drivers, at more than 350 long-term monitoring plots in Europe (Solberg et al., 2009; Laubhann et al., 2009; De Vries et al., 2008). A special feature of the present study is that it aims to assemble N deposition rates and budgets, together with variables of the carbon cycle, for a wide range of sites across the European continent in more depth and completeness than hitherto attempted, in order to seek more robust empirical evidence for the response of the terrestrial carbon cycle to different regimes of atmospheric N inputs. The quality of the individual data sets is, however, not uniformly high. Some of the data were measured in situ with known uncertainty, while others were simulated, derived from laboratory experiments and adapted to the field situation using measured time series of soil T and soil moisture, or taken from existing databases and literature. Also, data may not be fully comparable between sites (different methods used), nor even fully representative of each site (spatial heterogeneity). In the following sections, we discuss limitations of the measured, empirical and simulated data, both in terms of the component C and N fluxes, their budgets and interactions, as well as the challenges faced when attempting to establish empirical/statistical evidence for possible N effects on carbon sequestration in natural and semi-natural terrestrial ecosystems in Europe.

4.1 Constraining the ecosystem nitrogen balance through combined measurements and modelling

The compilation of N_r flux data (Fig. 3), based on several independent sources for each component term, provides a realistic picture of inorganic N_r inputs and losses; their balance suggests that for forests subjected to large deposition loads (> 2 g (N) m^{-2} yr^{-1}), typically more than half of the incoming N_r is lost to neighbouring environmental compartments such as groundwater and the atmosphere, and thus not available to promote C storage in the forest ecosystem. Since N losses increase - and N retention decreases - exponentially when N_{dep} exceeds a critical load of approximately 2-2.5 g (N) m^{-2} yr^{-1} (Fig. 3), it seems unlikely that the C sink strength of semi-natural ecosystems, including forests, increases linearly with N_r deposition, especially not with wet N deposition only. Based on a review of experimental N addition studies (e.g. Högberg et al., 2006; Pregitzer et al., 2008) and monitoring based field studies along N deposition gradients (e.g., Solberg et al., 2009; Laubhann et al., 2009; Thomas et al., 2010), De Vries et al. (2014) suggested that the C response reaches a plateau near 1.5-2.0 g (N) m^{-2} yr^{-1} and then starts to decrease. The linear relationship between C sequestration and wet N_r deposition as proposed e.g., by Magnani et al. (2007) is also challenged by the large contribution of dry N_r deposition and therefore by the poor correlation between total N_{dep} and wet deposition. We argue that our multiple-constraint approach for the nitrogen balance (measurement-model combination, model ensemble averaging, alternative data sources) provides overall a more robust basis for studying the impact of N_{dep} on the C cycle, even though uncertainties in individual terms remain significant.

4.1.1 Reducing uncertainty in nitrogen deposition

The uncertainty in dry deposition based on measured N_r concentrations and inferential modelling is likely not smaller than 30%, due to limitations in process understanding. The difference between ecosystem-specific EMEP values and the mean inferential estimates (Fig. S2) reflects discrepancies and uncertainties in the four dry deposition schemes used (Flechard et al., 2011); the mean coefficient of variation (CV = σ/μ) between the four inferential model estimates was 36%, i.e. larger than the difference between ecosystem-specific EMEP values and the mean inferential estimates. Other sources of discrepancy between the two methods include the use of measured *vs.* modelled meteorology to drive the deposition models, and site-specific *vs.* generic values of canopy height and leaf area index, as discussed in Flechard et al. (2011).

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The uncertainty in total N_r deposition is probably of the same order since even wet deposition can be deceptively difficult to measure (Dämmgen et al., 2005), and organic N, especially wet soluble organic N (WSON), may be significant but challenging to quantify (Cape et al., 2012) and generally ignored in the literature. WSON appears to be a generally small fraction of total (wet + dry) N_{dep} at most sites except at remote locations in Fennoscandia (EN10, SN3), where WSON

deposition could represent up to 20-30% of total N_{dep}. Also, potential double-counting due to dry deposition to the bulk deposition collectors (e.g. Thimonier et al., 2018) was not considered in this study, although on the basis of the comparison to other data sources (Fig. S2), bulk samplers did not appear to significantly over-estimate wet deposition.

Despite these uncertainties, measuring gas-phase and aerosol N_r concentrations locally should provide a better estimate of total ecosystem N_r inputs than the outputs of a large-scale chemical transport model. In addition, the partitioning of wet *vs.* dry deposition, reduced vs oxidized N, and canopy absorption *vs.* soil deposition, should also be improved, all of which are useful in interpreting ecosystem N cycling processes. In particular, for ammonia, with its high spatial variability on a local scale, the inferential modelling approach based on local measurements is likely to provide more realistic deposition estimates than a coarse-resolution chemical transport model (Flechard et al., 2013; Thimonier et al., 2018). In addition to low-cost methods for N_r concentrations, more actual micrometeorological N_r flux measurements are needed to further process understanding and better constrain surface exchange models over many ecosystems (Fowler et al., 2009). For example, ammonia flux measurements at DB2 have revealed unexpected features such as net NH_3 emissions from the forest in summer and autumn, in particular in response to leaf fall (Hansen et al., 2013, 2017). DB2 is likely not a net NH_3 source at the annual scale, but short-term emission pulses, which are not represented in most dry deposition models (Flechard et al., 2011), could significantly offset total N_r deposition.

An improved knowledge of N_r exchange patterns over CO_2 flux monitoring sites, either through inferential modelling or direct flux measurements, is also essential to quantify the fraction of deposited N_r that is absorbed by the canopy, reaching more or less directly the seat of photosynthesis in leaves, thus favouring a higher nitrogen use efficiency (NUE) (Nair et al., 2016; Wortman et al., 2012; Gaige et al., 2007). Canopy nitrogen retention occurs via several processes, including gaseous uptake by stomatal diffusion, a well-documented process (Monteith and Unsworth, 1990), but also through cuticular diffusion and stomatal penetration by aqueous solutions, with surface-deposited and dissolved gases and particles acting as direct leaf nutrients (Burkhardt, 2010; Burkhardt et al., 2012). By contrast, the N_r fraction initially deposited to soil (as simulated by the majority of fertilisation tracer experiments, e.g. Nadelhoffer et al., 1999) is subject to various losses via nitrification, denitrification and microbial uptake, before being eventually taken up by roots and moving upwards in xylem flow. The more advanced, emerging multi-layer canopy exchange models for atmospheric pollutants (N_r species, but also O_3, SO_2, etc.) can now partition dry deposition into stomatal, non-stomatal and soil pathways with increasing detail (Zhou et al., 2017; Simpson and Tuovinen, 2014; Flechard et al., 2013), thanks to improved understanding and parameterizations of surface and air column interactions and of photosynthesis-driven stomatal conductance (Büker et al., 2007; Grote et al., 2014). However, particular attention must be paid to measurement quality for an improved deposition accuracy, because such models are still very much dependent on local atmospheric concentration data for all main N_r forms (gas and aerosol, reduced and oxidized, mineral and organic).

4.1.2 Uncertainty in ecosystem nitrogen losses and net balance

The comparison of DIN leaching values by different methods shows that the Dise et al. (2009) algorithm performs reasonably well for low to moderate N_r deposition levels, but underestimates DIN losses for some of the highest (>4 g (N) m^{-2} yr^{-1}) deposition sites. This observation was also made by Dise et al. (2009) themselves, who argued that their simple relationships involving external forcings (N_{dep}) and internal factors (soil N status) are adequate "for early to intermediate stages of nitrogen saturation", but may fail at sites where historical, chronically enhanced N_r deposition has so strongly impacted forest ecosystems that N leaching has become dependent also on stochastic factors such as e.g. insect defoliation or a drought period followed by re-wetting of the soil. As was the case for field measured NO emissions (Fig. 3A), the four highest DIN leaching fluxes (0.9-3.2 g (N) m^{-2} yr^{-1}) occurred in the four highest N_{dep} forests growing on well-drained acidic sandy soils. In addition, it is noteworthy that the two sites with the largest N_{dep} and DIN leaching rates (EN15, EN16) were dominated by pine or Douglas fir (Table S2). These species have been shown in a common garden experiment (Legout et al.,

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Changed 'did provide' to 'should provide'

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2016) to cause larger nitrification, NO_3^- leaching and acidification rates (as well as larger losses of calcium, magnesium and aluminium), compared with other tree species such as beech or oak. This is consistent with deciduous trees being known to take up and store more nitrogen per unit biomass in stems and branches than coniferous trees (Jacobsen et al., 2003). Typical stem N content values, proposed for N uptake calculations in the Convention on Long-range Transboundary Air Pollution (CLRTAP) manual for critical loads mapping, are 1 and 1.5 g N kg^{-1} dry matter for conifers and deciduous trees, respectively, for steady state conditions (CLRTAP, 2017). Tree species traits may therefore, in our study, have exacerbated an existing DIN leaching predisposition resulting from edaphic factors and pollution climate. At the lower end of the N_{dep} range, the dataset is consistent with previous studies, which have shown that DIN leaching is unlikely to occur in forests where N_{dep} < 1 g (N) m^{-2} yr^{-1} (de Vries et al., 2009), although under these conditions there may still be significant N losses as NO and N_2O (Fig. 3).

The best empirical fit for the relationship of the sum DIN + NO + N_2O to N_{dep} was slightly non-linear (Fig. 3D) and may indicate that at the upper end of the N_{dep} range, above 4 g (N) m^{-2} yr^{-1}, the sum of inorganic N_r losses might approach or even exceed the estimated atmospheric deposition, which corresponds to one of the several existing definitions of ecosystem N saturation (see below). Whether these ecosystems turn into net N sources depends on the relative magnitudes of the missing terms: N_2 fixation (likely small in temperate compared with tropical forests; Vitousek et al., 2002), N_2 losses from denitrification (possibly the largest of the unknown terms at forest sites that are frequently waterlogged), N_2O losses from the litter layers of the forest floor, DON leaching; and also incoming organic nitrogen in precipitation (WSON) as well as dry deposition of organic N_r species, not quantified here (Fig. 1). The presumably small, and unaccounted for, N inputs via N_2 fixation and organic N_r deposition are at least partly compensated by denitrification N_2 losses and DON leaching losses. Moreover DON leaching typically responds much less strongly than DIN leaching to N inputs (Siemens and Kaupenjohann, 2002). Under these assumptions, the inorganic N_r budget calculated from Fig. 3 may provide a reasonable proxy for the overall ecosystem N balance. In this case, N outputs by gaseous and dissolved losses represent on average across all forest sites 43% of N inputs. More important than the average N loss for judging N_r deposition effects on C sequestration, is the large range of losses from 6% to 85%, with on average 27% loss (range 6-54%) for N_{dep} < 1 g (N) m^{-2} yr^{-1}, 45% loss (12-78%) for intermediate N_{dep} levels, and 65% loss (35-85%) for N_{dep} > 3 g (N) m^{-2} yr^{-1}. However, if the very few available data or estimates for DON leaching and especially denitrification N_2 fluxes are correct and may be extrapolated to other sites, they may often outweigh the inputs through organic N_r deposition and biological N_2 fixation, and thus the inorganic N_r budget (Fig. 3) may under-estimate the overall N losses.

4.2 Drivers and uncertainties of the carbon and GHG balance

4.2.1 Variability of carbon sequestration efficiency

The CSE ratio (=NEP/GPP) calculated over the CEIP/NEU project observation periods provides an indicator of the fraction of accumulated carbon in the ecosystem relative to gross CO_2 uptake by photosynthesis. This is a useful metric to compare carbon cycling in different terrestrial ecosystems and it is directly related to climate effects and other drivers such as site fertility (Vicca et al., 2012) and management (Campioli et al., 2015). By contrast, quantifying the accumulated carbon in terrestrial ecosystems requires much longer observations (one or several decades), to ensure statistical significance of a small change over a large C stock, particularly when soils are considered. This is often impractical, but also of limited use, because N deposition rates are unlikely to be constant over such long periods.

Over the time frame of the CEIP/NEU projects, observation-based CSE_{obs} values were much more variable than their modelled counterparts. Negative CSE_{obs} values (EN6, EN11) imply a net carbon source and may be explained by a number of factors, including soil carbon loss, lateral DOC/DIC water flow from adjacent ecosystems, tree mortality, low fertility, poor ecosystem health, a recently planted forest or other disturbances with long-lasting consequences on the C budget. At

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EN6 the main reasons may be a large SOC concentration, leading to large R_{eco} values, and a relatively old age of the forest, responsible for a small GPP.

However, the large discrepancy between observation-based and modelled CSE estimates may not be entirely caused by the model's inability to reproduce all fine patterns of GPP and especially R_{eco} across all ecosystems (Fig. 6). Some of the largest CSE_{obs} values may be less ecologically plausible and might result from methodological biases and/or incorrect interpretation of the EC measurements, in terms of their representativeness for the ecosystem considered. Multi-annual values of GPP and R_{eco} derived from EC flux data are not measurements *sensu stricto*; they compound problems in EC measurements, post-processing of high frequency data, gap-filling and partitioning. Some partitioning algorithms (Barr et al., 2004; Reichstein et al., 2005) evaluate GPP as the difference between measured daytime NEE and an estimate of daytime R_{eco} that is based on an empirical model of night-time R_{eco} measurements. In this case, any problem with nighttime and thus with estimated daytime R_{eco} would directly impact GPP in the same way (Vickers et al., 2009): GPP and R_{eco} would both be under-estimated, or both over-estimated, in absolute terms and by the same absolute magnitude, thereby impacting the annual or long term NEP/GPP (CSE_{obs}) ratio.

In this study, however, the use of the daytime data based partitioning method by Lasslop et al. (2010), within the REddyProc algorithm embedded in the European Fluxes Database Cluster, was intended to ensure the independence of GPP and R_{eco} estimates, since R_{eco} was estimated from the intercept of the Michaelis-Menten light–response curve fitted to daytime measured NEE. This partitioning procedure should avoid the propagation into the GPP estimate of potential errors in nighttime R_{eco} data, although it still assumes similar dependencies of day- and nighttime respiration to environmental factors, which is debatable from a biological standpoint (e.g., Kok, 1949; Wehr et al., 2016; Wohlfahrt and Galvagno, 2017). From a micrometeorological perspective, the nighttime flux can be underestimated due to low turbulence conditions and the transport of CO_2 by horizontal and/or vertical advection, and the decoupling of soil-level and understorey fluxes from the turbulent fluxes measured above the canopy (Feigenwinter et al., 2008; Etzold et al., 2010; Montagnani et al., 2010; Paul-Limoges et al., 2017). Further, in principle, the u_* threshold filtering (Gu et al., 2005; Papale et al., 2006), carried out to discard low turbulence flux data at the start of the gap-filling and partitioning algorithm (REddyProc, 2019), should alleviate the issue of nighttime R_{eco} underestimation, which affects annual R_{eco} and CSE_{obs} even if the error does not propagate into GPP in the Lasslop et al. (2010) method. However, the choice of the value for the u_* threshold can be critical if advection-affected flux values are to be discarded, especially for sites and data sets where the independence of the gap-filled annual NEP value from the u_* threshold value cannot be demonstrated. Advective flux contributions remain a largely unresolved issue, as Aubinet et al. (2010) conclude that «direct advection measurements do not help to solve the night-time CO_2 closure problem». Others (e.g. Kutsch and Kolari, 2015) have commented on the need to assign appropriate uncertainties when dealing with CSE and C balances derived from EC flux towers, which only measure turbulent fluxes and CO_2 storage change in the air column underneath the sensor but not the other terms of the conservation equation of a scalar in the atmospheric boundary layer (see Eq. (1) in Aubinet et al., 2000).

Despite all these precautions, at sloping or complex terrain sites where advection can be important, it cannot be excluded that the Lasslop et al. (2010) daytime data based approach may still underestimate R_{eco} (and overestimate CSE_{obs}) if advection is not accounted for explicitly. This is because the R_{eco} estimate based on the the intercept of the light response curve for the measured NEE (at PAR = 0) is strongly influenced by measurements made around sunrise and sunset, when a clear impact of advection on the light response curve ordinate has been observed, as shown at the EN5 subalpine site by Montagnani et al. (2009) (see their Fig. 13).

It is important to note that advection may also be a problem at flat lowland sites if there is strong spatial land surface heterogeneity, e.g. differences in albedo or in Bowen ratio, a gradient in tree species, a nearby lake, a gradient in water availability. Conversely, there may also be sites where EC underestimates CSE_{obs} for similar reasons, albeit in the opposite direction, for example additional CO_2 being advected into the ecosystem, then released by turbulent diffusion to the

atmosphere within the tower footprint. Another possibility is that basal R_{eco}, measured at dawn or dusk over a different (larger) footprint, is lower than during the day. Flux partitioning may again in this case underestimate R_{eco} during the warmer daytime hours, and therefore also underestimate GPP, resulting in overestimated NEP/GPP (CSE_{obs}) ratios.

Given this uncertainty, the fact that most of the forest stands with CSE_{obs} values larger than 40% (EN1, EN5, DB6, MF2) were located at elevations above 700 m a.m.s.l. (Table 1 and Fig. 8A), i.e. in hilly or mountainous areas with topographically more complex terrain than typically encountered at lowland sites, may be coincidental, or partly a consequence of advection or decoupling issues (Paul-Limoges et al., 2017). In such conditions, consistency crosschecks involving additional flux, advection, soil and biometric measurements, even ecosystem modelling, provide useful reference points to assess the plausibility of EC-derived C budgets and to better constrain the problem. At the EN5 site, the annual total tree biomass C increment based on biometric measurements was on average 218 g (C) m^{-2} yr^{-1} over the period 2010-2017 (L. Montagnani, unpublished data), i.e. 26% of the reported mean EC-derived NEP value of 826 g (C) m^{-2} yr^{-1} for the CEIP-NEU period, and it seems unlikely that the increase in soil carbon and fine roots stocks could account for the large difference. By contrast, the DB6 site was a fertile and managed beech forest, with a significantly higher efficiency conversion of photosynthates into biomass compared to less fertile and unmanaged sites (Vicca et al., 2012; Campioli et al., 2015). The long term annual total NPP at the site was 780 g (C) m^{-2} yr^{-1} over the period 1992-2007, with a significant part allocated belowground (Alberti et al., 2015), while heterotrophic respiration estimated at the site using either bomb-carbon (Harrison et al., 2000) or mineralization rates (Persson et al., 2000) was around 200 g (C) m^{-2} yr^{-1}, resulting in similar NEP estimates by EC flux measurements versus biometric data combined with process studies.

At the MF2 site, Etzold et al. (2011) calculated inter-annual mean EC-derived NEP, GPP and R_{eco} values (for the same 2005-2009 period used in this study) of 415, 1830 and 1383 g (C) m^{-2} yr^{-1}, respectively, using customized gap-filling and partitioning algorithms and thus providing alternative estimates to those from the REddyProc algorithm within the European Fluxes Database Cluster (Table 1). Values of R_{eco} and NEP were 82% larger and 40% lower, respectively, in Etzold et al. (2011) compared with the default database values that do not explicitly correct for advection. However, the Etzold et al. (2011) mean EC-derived NEP was much closer to NEP values calculated from the net annual increment in the woody and non-woody biomass and soil C storage using four different biometric and modelling methods (range 307-514 g (C) m^{-2} yr^{-1}, mean 421 g (C) m^{-2} yr^{-1}). The CSE_{obs} value derived from Etzold et al. (2011) was 23%, and comparable to the value of 25% that can be calculated from the decoupling-corrected EC budget computed by Paul-Limoges et al. (2017) for the same site for the years 2014-2015, in which the decoupling correction to account for undetected below-canopy fluxes doubled R_{eco} and reduced NEP from 758 to 327 g (C) m^{-2} yr^{-1}. These alternative CSE_{obs} estimates were thus much lower than the default CSE_{obs} value of 48% (Fig. 8) but fully consistent with model predictions (Fig. 8A).

The four upland sites EN1, EN5, DB6, MF2, were also among the wettest, with MAP > 1000 mm (Fig. 8B) in principle promoting larger leaching and runoff. The overall distribution of CSE_{obs} as a function of MAP (Fig. 8B) shows an apparent increase of CSE_{obs} with precipitation, though with large scatter, which would be consistent with a reduction in EC tower-based R_{eco} through an increase in the dissolved leached fraction. At sites where significant leaching occurs, R_{eco} determined from the atmospheric flux is no longer a reliable indicator of total C losses by respiration since the dissolved, then leached fraction of R_{soil} is not captured by the flux tower (Gielen et al., 2011), which implies that CSE_{obs} is over-estimated. As observed in the case of GPP, such apparent correlations of CSE_{obs} to single factors like elevation or MAP may not be (entirely) causal, potentially concealing underlying cross-correlations (such as large but unmeasured advection components occurring at the same sites where MAP is largest). The data by Kindler et al. (2011) and Gielen et al (2011) do suggest that the overestimation of C sequestration (as estimated by EC-derived NEP), caused by not accounting for dissolved C leaching, was likely smaller than 10% for forests (7% of NEP on average), but all five sites they investigated had MAP < 1000 mm and only one (EN4) was an upland site (785 m).

To summarize a set of unresolved issues, the largest CSE_{obs} values (> 45%) are likely to result from a combination of ecological factors and methodological biases, but they occurred at sites in mid range for N_{dep} (1.2–2.2 g (N) m^{-2} yr^{-1}) and thus did not introduce confounding trends in the overall C/N relationships we seek to establish across the whole N_{dep} spectrum in this study.

{Insert Figure 8 here}

4.2.2 Forest net greenhouse gas balance dominated by carbon

Based on the available data, the net GHG balance of the 31 forests investigated was generally not significantly affected by N_2O or CH_4 (Fig. 7), with the caveat that these fluxes were not actually measured in situ everywhere, nor with the same intensity and duration as CO_2. Thus, the uncertainty in non-CO_2 GHG fluxes is much larger (possibly > 100%) than for multi-annual EC-based CO_2 datasets, where a typical uncertainty is of the order of 10-30% (Loescher et al., 2006). Nonetheless, the N_2O and CH_4 emissions observed by different methods in forest soils were typically two orders of magnitude smaller than the CO_2 sink (in GWP equivalents), which means that the quality of CO_2 estimates dominates the overall uncertainty in our forest GHG budgets. Note that such results cannot be extended to waterlogged, organic soils of temperate and boreal zones, where CH_4 emissions can be large (Morison et al., 2012), nor to the tropics especially in degraded forests (Pearson et al., 2017). Also, N_2O fluxes can be highly episodic, with emission events linked to, e.g., freeze-thaw cycles (Risk et al., 2013; Medinets et al., 2017) and such episodes would have been missed by the bioassay approach, and in some cases by discontinuous (manual) chamber measurements.

By contrast, for the short semi-natural vegetation sites of our study, NEP was on average a factor of 2.7 smaller than in forests, but only a factor of 1.5 smaller for GPP, which implies that total C losses were much larger in proportion to gross assimilation, especially non-respiratory, non-CO_2 losses (i.e., a much lower CSE). Large wetland CH_4 emissions and dissolved DIC/DOC fluxes were much more likely to offset or even determine the C and GHG balance (Fig. 7; Kindler et al., 2011). In these systems, studying the impact of N_r deposition on C sequestration requires much more robust estimates of the gaseous and dissolved budgets for all components and over the long term, since the estimation of NECB requires in addition to EC-CO_2 the knowledge of non-atmospheric, non-CO_2 fluxes (Fig. 1). Technological developments in the field of (routine) EC measurements for N_2O and CH_4 (e.g. Nemitz et al., 2018) are likely to reduce uncertainties in net GHG budgets in the foreseeable future, but DIC/DOC losses in wetlands probably represent a bigger challenge.

It should however be remembered that such short-term GHG budgets, based on a few years flux data and GWP multipliers for a 100-yr time horizon, do not actually reflect the long term climate impact of northern mires, which may be thousands of years old, and despite their CH_4 emissions, typically have an overall climate-cooling effect. As shown by Frolking et al. (2006), pristine mires typically start cooling the climate some hundreds of years after their formation, the exact timing of course depending on the magnitude of the CH_4 and CO_2 fluxes; thus the history of the site should be accounted for when dealing with ecosystem radiative forcing assessments. For the SN3 site, Drewer et al. (2010) actually used a 500-yr time horizon GWP (instead of the usual 100-yr) for CH_4, reducing the GHG source strength of the site by a factor of 4 to 10, depending on the year considered.

4.3 Challenges in understanding the coupling of carbon and nitrogen budgets

4.3.1 Tangled effects of nitrogen deposition and climate on ecosystem productivity

The analysis of N_{dep} variability and spatial patterns at the scale of the monitoring network, as well as the European scale (Fig. 5), showed that the impact of N_r deposition on ecosystem C sequestration cannot be considered independently of climate in the regional context of this study. Nitrogen deposition patterns at the European scale result from the continent-wide geographical distribution of population, human, industrial and agricultural activities, and of precursor emissions, combined with mesoscale patterns of meteorology-driven atmospheric circulation and chemistry. Through the interplay of

Commentaire [c23]: Rephrased and split sentence for improved readability

these factors, the elevated N_{dep} levels in this study happened to co-occur geographically with temperate climatic zones of Central-Western Europe (Fig. 5 C-D) that are the most conducive to vegetation growth at the continental scale. This means adequate water supply as precipitation, reasonably low summertime evaporative demand, mild winters and temperate summers, long growing seasons. In other words, there are many gaps in the multi-dimensional variable space, which is incompletely explored by the available dataset. Thus, any regression analysis that would correlate NEP and other C fluxes with N_{dep}, without simultaneously accounting for climate, would be flawed, as Sutton et al. (2008) concluded from their re-analysis of the data used by Magnani et al. (2007). A dC/dN slope calculated directly from a (linear or non-linear) mono-factorial regression analysis of GPP or NEP vs. N_{dep} would misleadingly attribute the whole C flux variability to N_{dep} while ignoring climate effects (Fleischer et al., 2013). In addition, a range of other potential explanatory variables such as soil type, especially the water holding capacity (Φ_{FC} - Φ_{WP}), soil fertility (Vicca et al., 2012; Legout et al., 2014), tree species, stand age (Besnard et al., 2018), are potentially needed to explain the observed variability. In order to account for, and untangle, the multiple inter-relationships, we chose a mechanistic model (BASFOR) based approach, described in Flechard et al. (2020), whereby most of the known interactions of plant, soil, climate, age, species, are encoded and parameterised to the best of our current knowledge. Given the limited size and very large diversity of the dataset, such an approach appears to be preferable to regression-based statistical analyses, since a simple pattern to explain the coupling of carbon and nitrogen budgets with the available data and knowledge is unlikely.

Commentaire [c24]: Text rephrased in order to explain why model-based (mechanistic) approach to untangle the relationships is preferable to multivariate statistical approach

4.3.2 Evidence of nitrogen saturation from various indicators

Various definitions of nitrogen saturation have been proposed (Aber, 1992; De Schrijver et al., 2008; Binkley and Högberg, 2016), including i) the absence of a growth response in the case of further N addition (dC/dN = 0); ii) the onset of NO_3^- leaching and/or gaseous emissions; and iii) the equivalence of N inputs and N losses. The underlying concept of a dC/dN response is that the C and N cycles are closely coupled through stoichiometric ratios in the different parts of the ecosystem, with very different C/N ratios in soil organic matter, roots, leaves, tree branches and stems (de Vries et al., 2009; Zechmeister-Boltenstern et al., 2015). A difference in dC/dN response could, for example, be expected between forests, where carbon is stored in both woody and root biomass (C/N ratio 300-500) and below ground in SOM (C/N ratio 30-40), versus short semi-natural vegetation, where most of the stock is in SOM, and thus with a much lower overall ecosystem C/N ratio. This would be consistent with the observations in Fig. 4, where the apparent increase of NEP with increasing N_{dep} is smaller in short semi-natural vegetation than in forests. But the theoretical stoichiometric approach becomes more uncertain in the event of N saturation, as the C and N cycles have become much less tightly coupled than in pristine, N-limited environments, and thus defining a dose-response relationship requires a precise quantification of all C and N inputs and losses, not just productivity and N_r deposition.

Another possible indicator of N saturation in the present dataset may be provided by the comparison of the relationships of C/N ratios of foliage and top soil (5 cm) to atmospheric N_r deposition (Fig. 9A-B). Since leaf N content was not only dependent on N_r deposition but also on the ecosystem type (Fig. 4E), C/N ratios are shown separately for the different vegetation classes in Fig. 9. There was a clear negative correlation of leaf C/N ratio to N_{dep} for coniferous forests (ENF, spruce and pine pooled: exponential fit $R^2 = 0.86$, $p < 0.01$) and a similar but not significant trend for SN (linear $R^2 = 0.29$) (Fig. 9A); for the other ecosystems (DBF, MF, EBF) there were not enough data to derive trends. In top soils (Fig. 9B), there was also a broad downward trend of C/N ratios with increasing N_{dep} within the ENF and SN classes, but only for N_{dep} up to 2.5 g (N) m^{-2} yr^{-1}. Again, as for GPP and NEP, the relationship is highly non-linear as the four ENF sites above this N_{dep} threshold break the trend observed in the lower N_{dep} sites, and the overall best fit is quadratic ($R^2 = 0.49$, $p < 0.01$) with an inflexion point around this threshold. While the relationship of foliar C/N ratio to N_{dep} was almost linear for ENF (a consequence of the linear trend in ENF leaf N content, Fig. 4E), the non-linear behaviour of the topsoil C/N ratio and its stabilization or increase for N_{dep} > 2.5 g (N) m^{-2} yr^{-1} indicate a possible threshold for saturation. Atmospheric nitrogen was

therefore apparently efficiently taken up by vegetation when reaching the leaves; but after leaf fall, and following litter decomposition and incorporation into the topsoil, there appeared to be a limit to the amount of nitrogen that can be stabilized into soil organic matter of the ENF sites. However, forest soil organic N stocks are very large (in the range 200-700 g (N) m^{-2} at the sites we investigated), and therefore changes in C/N ratios in response to atmospheric N_r deposition must be very slow. The soil C/N ratio at a given time reflects centuries of land use as well as a more recent history of multi-decadal changes in N_r deposition (Flechard et al., 2020). This complicates the interpretation of the downward trends observed from instantaneous snapshots of soil and foliar C/N ratios versus N_{dep} since the ecosystems cannot be considered to be in steady state, neither for N_{dep} nor for growth or productivity. There was a positive correlation across all vegetation types between topsoil and foliar C/N ratios (Fig. 9C; R^2 = 0.19, $p < 0.05$), but this was mostly driven by differences between plant functional types (no significant correlation within each PFT).

{Insert Figure 9 here}

Following definition ii) of N saturation given above, the sum of inorganic N_r losses, heavily dominated by DIN leaching at the upper end of the N_{dep} range in our datasets (Fig. 3), may indicate various stages of N saturation in all forests with N_{dep} > 1-1.5 g (N) m^{-2} yr^{-1}. A threshold for a more advanced saturation stage could be placed at 2-2.5 g (N) m^{-2} yr^{-1}, where inorganic N_r losses are consistently larger than 50% of N_{dep}. Such numbers are entirely consistent with the leaching risk classification of European forests proposed by Dise and Wright (1995), with low leaching risk at N_{dep} <1 g (N) m^{-2} yr^{-1}, intermediate risk at N_{dep} in the range 1-2.5 g (N) m^{-2} yr^{-1}, and high risk at N_{dep} > 2.5 g (N) m^{-2} yr^{-1}. The results are also in line with the review by De Vries et al. (2014); based on literature results of dC/dN responses derived from stoichiometric scaling, meta-analysis of N addition experiments and field observations of both growth changes and N_r deposition, accounting for other drivers, the data showed beneficial N_r deposition effects up to 2-3 g (N) m^{-2} yr^{-1} and adverse effects at higher levels. A lower N_{dep} threshold of 1 g (N) m^{-2} yr^{-1} had also been suggested by de Vries et al. (2007), but this was using throughfall deposition, which generally under-estimates total deposition through canopy retention processes (Thimonier et al., 2018). It must be stressed, however, that the definition of an all-purpose, generic N_{dep} threshold for N saturation may be misleading, or at least qualified with an uncertainty, since some tree species (Douglas fir, pine, spruce), grown on the same soil and under the same climate and N_{dep} regime, may result in significantly higher NO_3^- leaching rates than others (Legout et al., 2016). This also means that the NO_3^- leaching flux is not necessarily a good proxy of the severity of N saturation, though this depends on which of the several definitions of N saturation is considered.

The upper threshold of 2-2.5 g (N) m^{-2} yr^{-1} happens to coincide with the levelling off of GPP, R_{eco} and NEP, and the further reduction in C fluxes at higher N_{dep} levels (Fig. 4A-C). Whether this should be interpreted as a negative impact of advanced N saturation on soil processes and plant functioning and, hence, C sequestration potential, is not straightforward (Binkley and Högberg, 2016). If the parallel effects of climate, soil fertility, other nutrient limitations, tree species traits, age and planting density are overlooked in a simplistic, first-order interpretation, the dataset hints at an "optimum" N_{dep} level around 2 g (N) m^{-2} yr^{-1}, beyond which no further benefits (in carbon terms) could be gained from further atmospheric N_r additions, which would be consistent with the 2-2.5 g (N) m^{-2} yr^{-1} N_{dep} threshold derived by Etzold et al. (2014) for Swiss forests. The high soil N_r losses observed in these ecosystems growing under relatively favourable climates would then suggest that whatever fertilisation effect N_r deposition may have at low to moderate deposition rates (<2 g (N) m^{-2} yr^{-1}) is unlikely to be sustained at high deposition levels, especially on acidic sandy soils. However, the very limited number of affected sites with N_{dep} > 3 g (N) m^{-2} yr^{-1} leaves too few degrees of freedom to make the argument statistically compelling. More importantly, a knowledge of all other limitations to growth (climate, soil, fertility, nutrients, age structure) would be required to confirm the hypothesis. Additional measurement- and model-based investigations to untangle the N_{dep} effect on C sequestration (the dC/dN term) are presented in Flechard et al. (2020), drawing from the results, fluxes and budgets presented here.

5 Conclusion

We provided estimates of carbon, nitrogen and greenhouse gas budgets for 40 flux tower sites over European forests and semi-natural vegetation, compiled from a large variability of state of the art methods that can be applied in such a network approach. The CO_2 budgets from well-established EC methods were the least uncertain, followed by GHG budgets of forests, then the CH_4 and DIC/DOC fluxes of wetlands; uncertainty levels were likely highest in the net N budgets, especially at the elevated N_r deposition sites where NO_3^- leaching was almost of the same order as N_{dep}. The uncertainty was still compounded by the lack of some data on biological N_2 fixation, N_2 loss by denitrification, and organic N_r in rainwater, in dry deposition and in soil leaching, but some of these unknown terms would compensate mutually to some extent. Nevertheless, the low-cost network to monitor atmospheric gas-phase and aerosol N_r contributed to substantially reducing the large uncertainty in total N_{dep} rates at individual sites (compared with gridded outputs of a regional chemical transport model). This was because dry deposition almost systematically heavily dominates over wet deposition in forests, except at very remote sites (away from sources of atmospheric pollution), and directly measured N_r concentrations reduced the uncertainty in dry deposition fluxes.

Commentaire [c25]: Re-phrased to make the original statement less confusing

The greenhouse gas balances of the 31 forest sites included in this study were almost entirely determined by the CO_2 budgets, with small to negligible contributions by N_2O and CH_4. The GHG balance of nine extensively managed and upland grasslands, moorlands and wetlands was much more dependent on CH_4 and N_2O fluxes. Ecosystem productivity (GPP, NEP) data across Europe showed an apparent increase with atmospheric N_{dep}, though only up to 2.5 g (N) m^{-2} yr^{-1}, while the larger N_{dep} rates also happen to coincide geographically with regions of Europe where climate is optimal for tree growth (neither too cold nor too dry). The data thus underpinned a strong covariation of N_r deposition with variables like elevation and climate, and indicated that the ecosystem response of carbon sequestration to nitrogen deposition cannot be calculated simply and directly from the observed apparent $dNEP/dN_{dep}$ using bivariate statistics. Other co-varying influences such as climate, soil, fertility, nutrient availability, forest age, ecophysiological processes, etc., should be analyzed alongside, so the nitrogen deposition effect can be isolated.

The site-specific analysis of C and N fluxes and budgets across a large geographical and climatic gradient supports the concept of a non-linear response of C sequestration to N deposition. Large nitrogen losses (especially nitrate) from forests suggest that up to one third of the sites investigated can be classified as in early to advanced stages of N saturation. At the sites with the largest N_r deposition rates (> 2.5 g (N) m^{-2} yr^{-1}), a stagnation or reduction in forest productivity, compared to mid-range deposition sites, was observed. Beyond the conclusion that the apparent C response to increased N_r deposition was non-linear, we do not have enough data to test the hypothesis that the reduction in productivity and C sequestration is linked to N saturation-induced ecological impacts on soil and ecosystem functioning, rather than just the confounding effects of variability in meteorological and other drivers. Further efforts are required to disentangle N_{dep} effects and climatic as well as pedological effects on C sequestration at the continental scale.

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, MAS, AI, WdV, MvO, UMS conceived the paper; MAS, EN, UMS, KBB, WdV conceived or designed the NEU study; 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; YST, NvD, HU, UD, SV, VD, MMit, FS, YF performed DELTA and bulk deposition chemical analyses; AF collected wet deposition databases; DS provided modelled EMEP N_r deposition data; BKit, SZB conceived and performed the soil bioassay experiment; JKS provided foliar nitrogen analyses; AI, UMS, JFJK, ALeg, MJS, MAub, DL, LM, JN, IAJ, MP, RK, JA, AV, JO, RJ, MAur, BHC, JD, WE, AF, AG, PG, YH, CH, AH, LH, BKru, WLK, RLdV, ALoh, BL, MVM, GM, VM, JMO, KP, GP, MTS, MU, TV, CV, TW provided eddy covariance and/or other field data, or contributed to data collection from external databases and literature; AI, WdV, MAS, UMS, MvO, EN, KBB, SZB, DRC, NBD, JFJK, NB, ALeg, DS, MJS, MAub, DL, LM, JN, IAJ, MP, RK, JS, AJF, JA, AV, JO, RJ, MAur, WE, BKit, BKru, RLdV, ALoh, GM, AN, MU 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

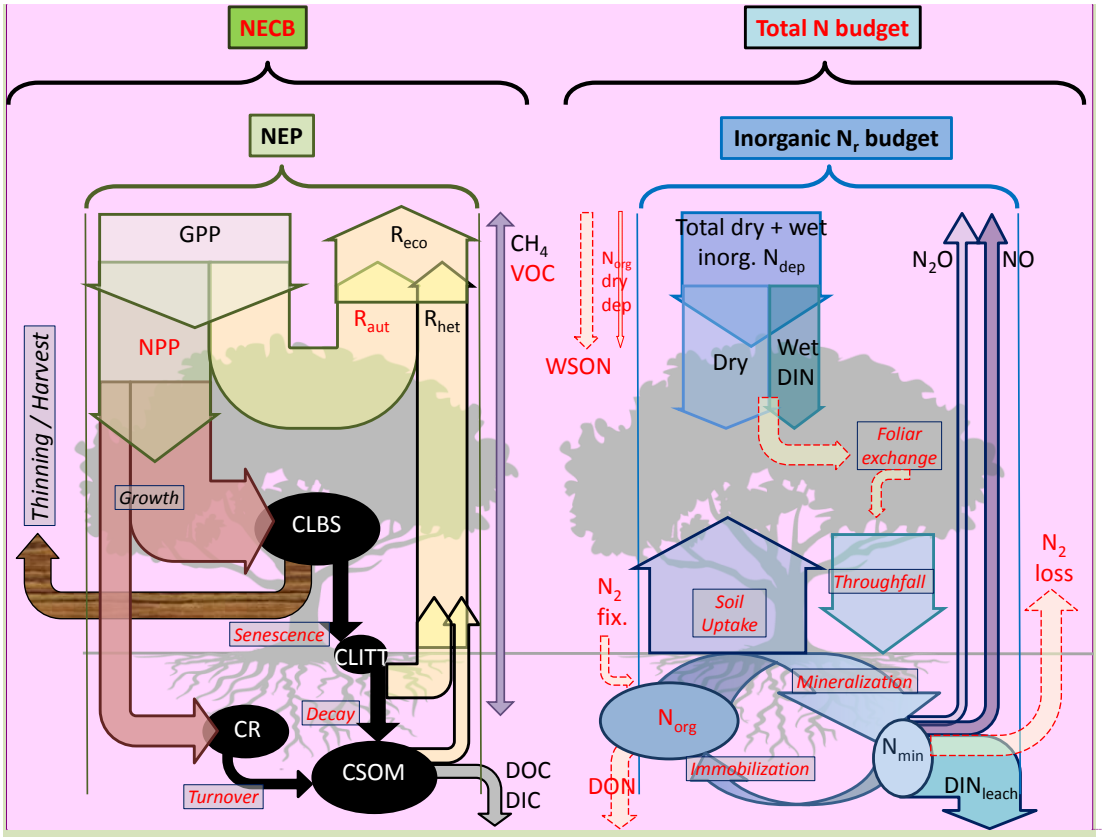

Figure 1. Flux terms and boundaries of the carbon (left) and nitrogen (right) budgets discussed in this paper. Net ecosystem productivity NEP = GPP – R_{eco} (≈ NPP – R_{het}) based on multi-annual eddy covariance CO_2 flux data. The net ecosystem carbon balance (NECB) includes in addition other C loss fluxes such as DIC/DOC, CH_4 and VOC, as well as harvest, thinning or other disturbances (e.g. fire). Inorganic reactive nitrogen (N_r) budget = N_{dep} – DIN_{leach} – NO – N_2O. The total N budget includes in addition organic nitrogen deposition (WSON) and leaching (DON), as well as N_2 inputs and losses from biological fixation and denitrification, respectively. CLBS, CSOM, CR, CLITT: carbon stocks in leaves, branches and stems, in soil organic matter, in roots, and in litter layers, respectively. Terms highlighted in red indicate that direct or measurement-based estimates were not available for some or all sites in our datasets (see also Table 2 for a list of acronyms, Table 3 for a summary of methods, and Table S6 for data availability).

Commentaire [c26]: Added arrow for dry deposition of Nord
Added 'Senescence' and 'Turnover' boxes

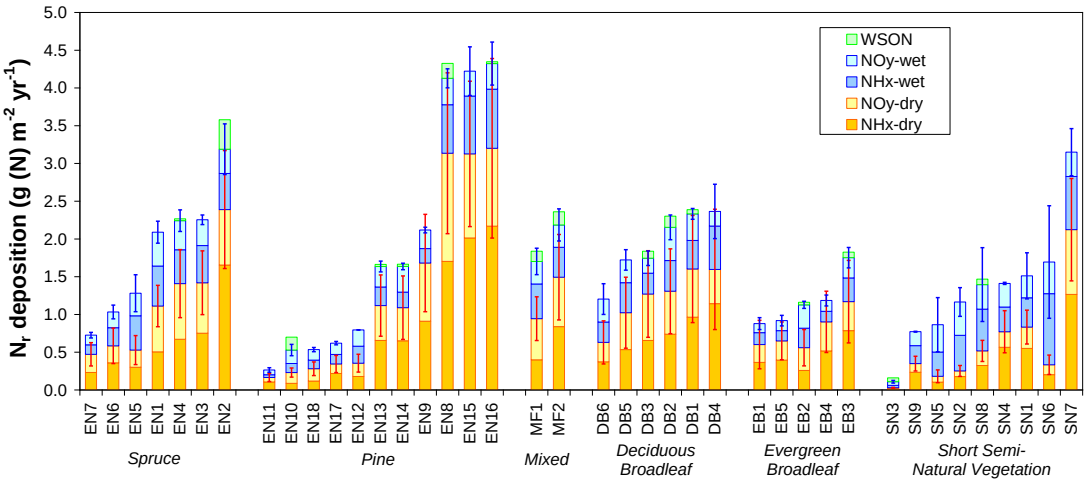

Figure 2. Total reactive nitrogen deposition (N_{dep}) and breakdown into inorganic wet and dry, oxidized (NO_y) and reduced (NH_x) deposition estimates at the 31 forest sites (evergreen needleleaf EN1-7 (spruce), EN8-18 (pine), mixed MF, deciduous broadleaf DB, evergreen broadleaf EB), and at 9 short semi-natural (SN) vegetation sites of the NitroEurope monitoring network. Data are arithmetic means over the years 2007-2010 of i) inferential dry deposition estimates by four different models based on in situ atmospheric N_r measurements, and ii) of different wet deposition estimates from precipitation monitoring datasets and from European-scale atmospheric chemistry and transport modelling (EMEP). Error bars indicate standard deviations of the four dry deposition models (red bars) and standard deviations of the different data sources for inorganic N_r wet deposition (blue bars). Wet deposition of water-soluble organic nitrogen (WSON) was measured at a few selected sites and is shown here for comparison with total inorganic N_r deposition.

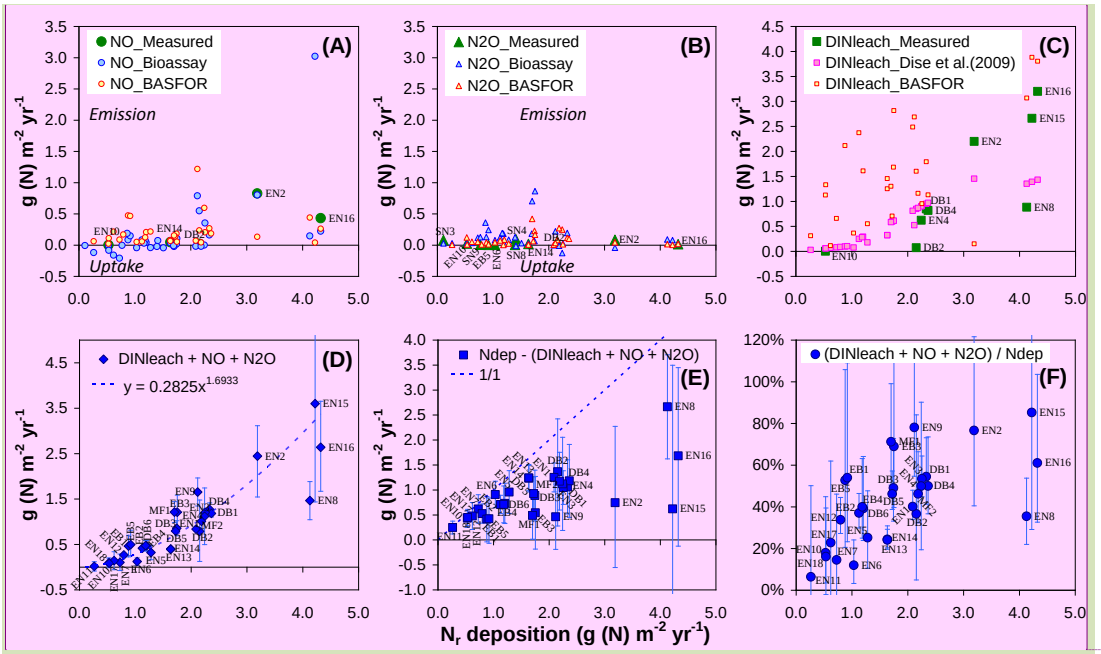

Figure 3. Comparison of measured and estimated ecosystem inorganic N_r losses and their relationships to total atmospheric N_r deposition (x-axis) at the forest sites. NO fluxes (A) and N_2O fluxes (B) were either i) measured in situ using static or dynamic flux chambers, ii) scaled up from laboratory bioassay-derived T/WFPS relationships, or iii) simulated using the BASFOR ecosystem model (see text for details). DIN leaching (C) was either measured (lysimeter or suction cups), or predicted from the Dise et al. (2009) empirical algorithm. The sum of inorganic N_r losses (DINleach + NO+ N_2O) was computed as the mean of measured values and modelled estimates. In panels A-C, site names are indicated for sites where in situ measurements were available.

Commentaire [c27]:
Improved site label readability:
-Increased font size for site labels
-Moved labels to avoid overlapping

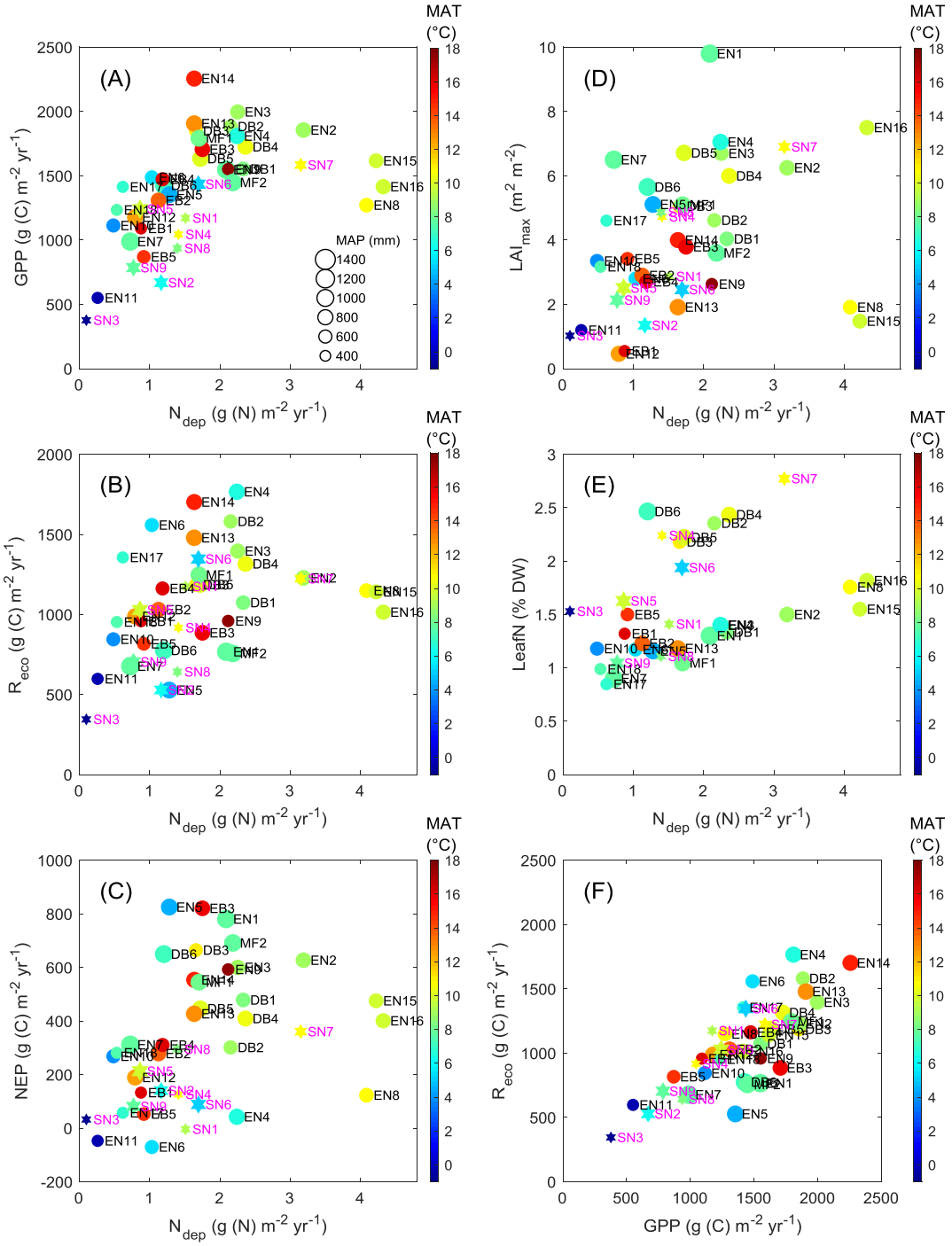

Figure 4. Overview of inter-annual mean EC-derived C flux estimates (GPP, R_{eco} and NEP), ecosystem LAI and leaf N content, in relation to total (dry + wet) atmospheric N_r deposition (A-E), and relationship of R_{eco} to GPP (F), for forests (filled circles, black labels) and short semi-natural vegetation (filled stars, magenta labels). In all plots, the colour scale indicates mean annual temperature (MAT), while the symbol size is proportional to mean annual precipitation (MAP, scale provided in panel A).

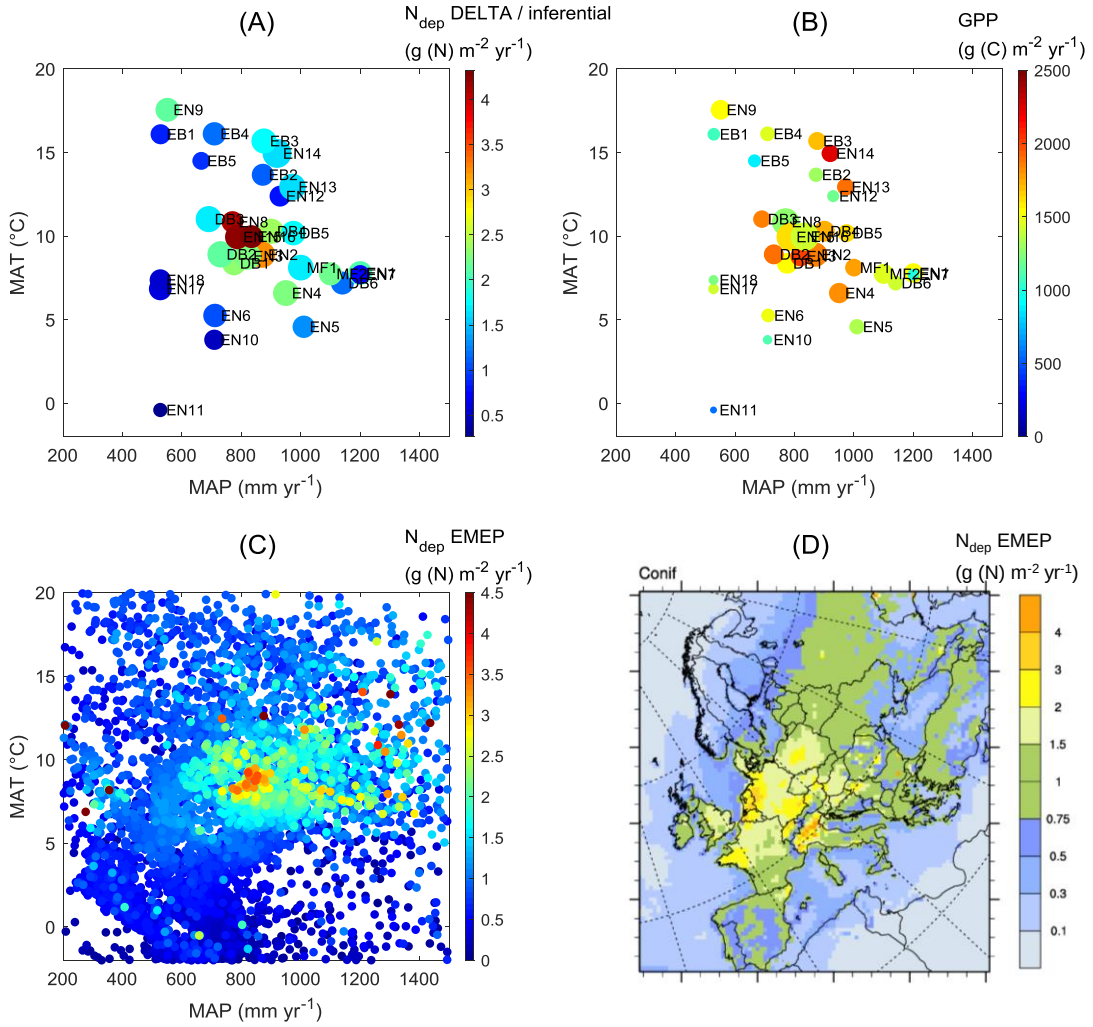

Figure 5. Distribution of observation-based nitrogen deposition (N_{dep}) (A) and gross primary productivity (GPP) (B) for the forest sites of this study, within the European climate space represented by mean annual temperature (MAT) and precipitation (MAP). In plot A the symbol color indicates N_{dep} while the symbol size is proportional to GPP; in plot B the symbol color indicates GPP, while the symbol size is proportional to N_{dep}. Plots C shows modelled N_{dep} from the EMEP model over coniferous forests (year 2010), represented in climate space (1 data point for each grid square of the EMEP domain containing coniferous forests), also shown as a map (D). The MAT axis can be seen as a proxy for latitude and/or elevation, while the MAP axis expresses to some extent longitude (distance to the ocean) and/or orographic precipitation enhancement.

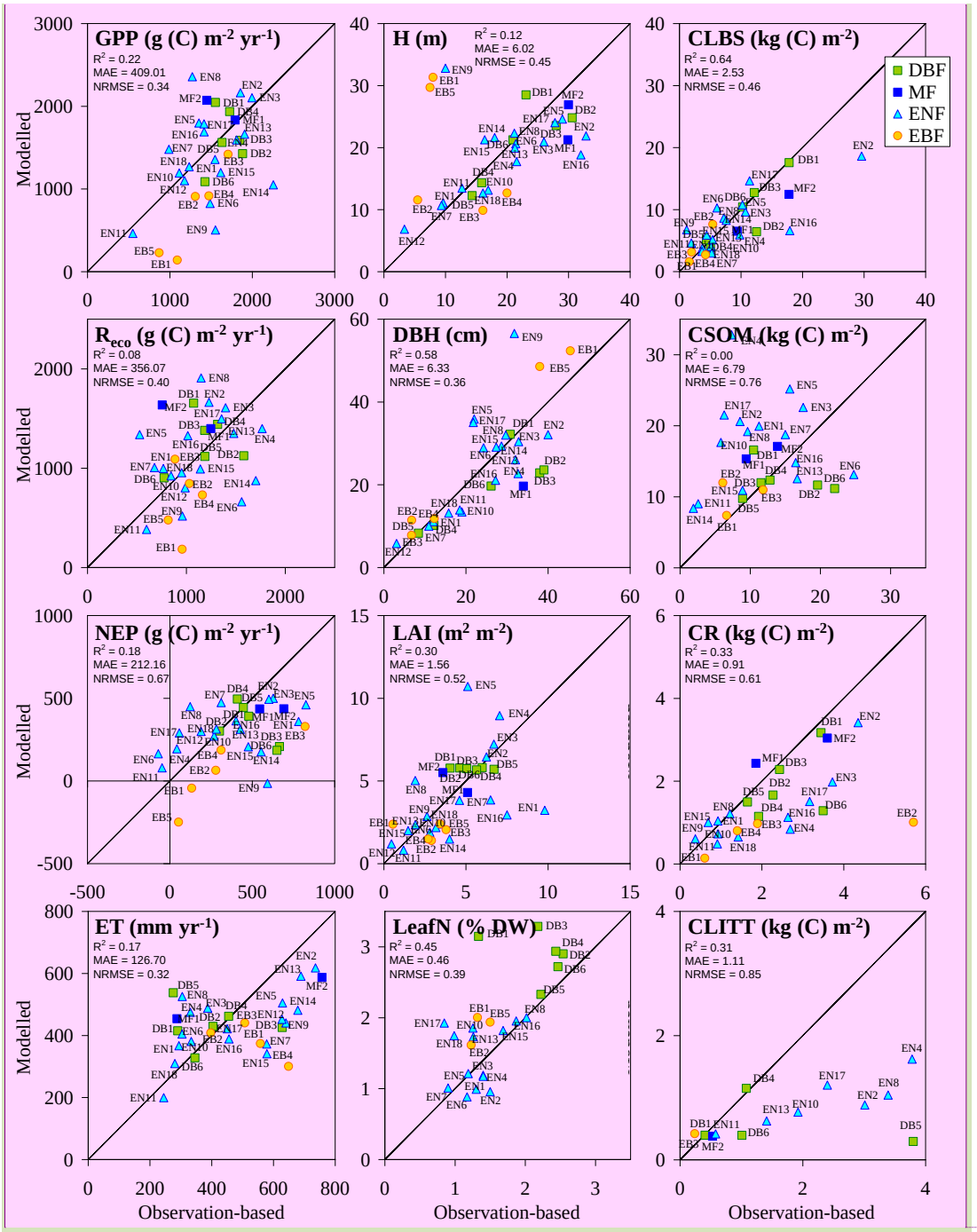

Figure 6. BASFOR baseline simulations for all forest sites; model outputs and observation-based values were averaged over the years between the first and last available observations. Note that model simulations include MF and EBF sites, for which the model was not calibrated in Cameron et al. (2018); the two MF runs were made using the parameter table for DBF, while the five EBF runs were made using the parameter table for ENF to allow continued growth throughout the year. H: mean tree height; DBH: mean diameter at breast height; CLBS, CSOM, CR, CLITT: carbon stocks in leaves, branches and stems, in soil organic matter, in roots, and in litter layers, respectively; MAE: mean absolute error; NRMSE: root mean square error normalised to the mean.

Commentaire [c28]:
Improved site label readability:
-Increased font size for site labels
-Moved labels to avoid overlapping

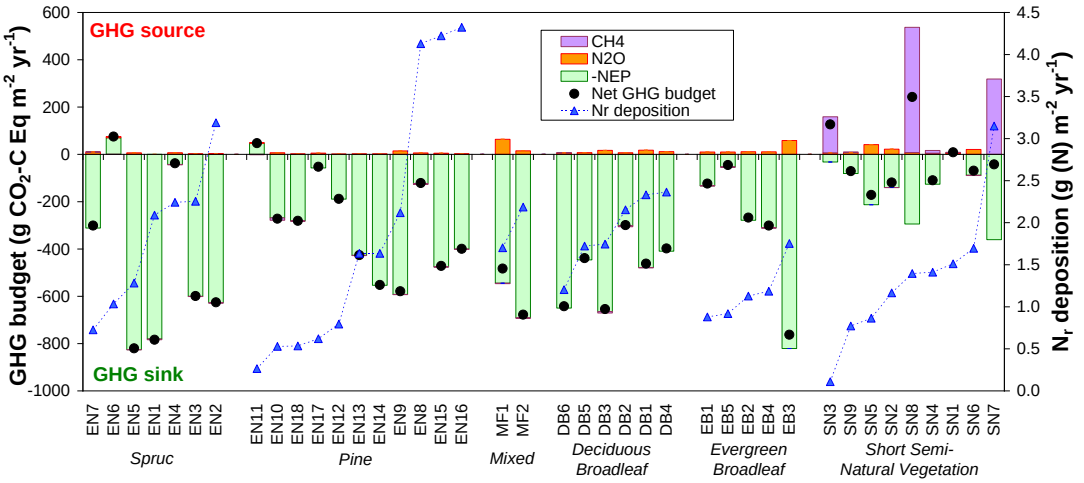

Figure 7. Net greenhouse gas (GHG) budgets calculated from a combination of inter-annual mean (around 2005-2010) net ecosystem productivity (NEP) from eddy covariance, and N_2O and CH_4 flux data measured in situ or estimated by extrapolated bioassay data and forest ecosystem BASFOR modelling. Global warming potential values (100-yr time horizon) of 265 and 28 were used for N_2O and CH_4, respectively; the sign convention is with respect to the atmosphere, negative for a sink, positive for a source. The data were grouped by ecosystem type (evergreen needleleaf EN-spruce and EN-pine, MF-mixed forests, DB-deciduous broadleaf, EB-evergreen broadleaf, SN-short semi-natural vegetation); within each group the data were sorted by increasing N_r deposition.

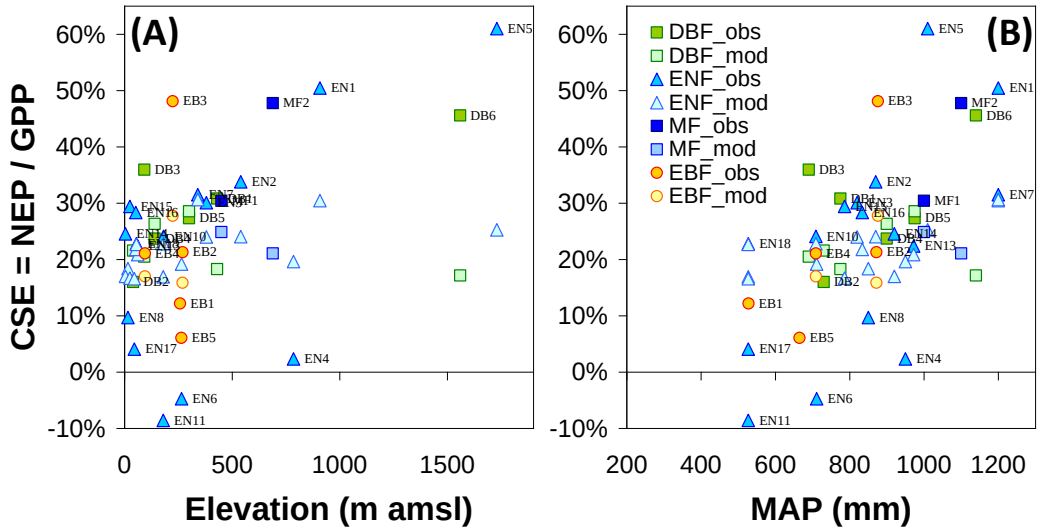

Figure 8. Variability of observation-based and modelled carbon sequestration efficiency (CSE, defined as the NEP/GPP ratio), as a function of (A) site elevation above mean sea level (m), and (B) MAP: mean annual precipitation (mm).

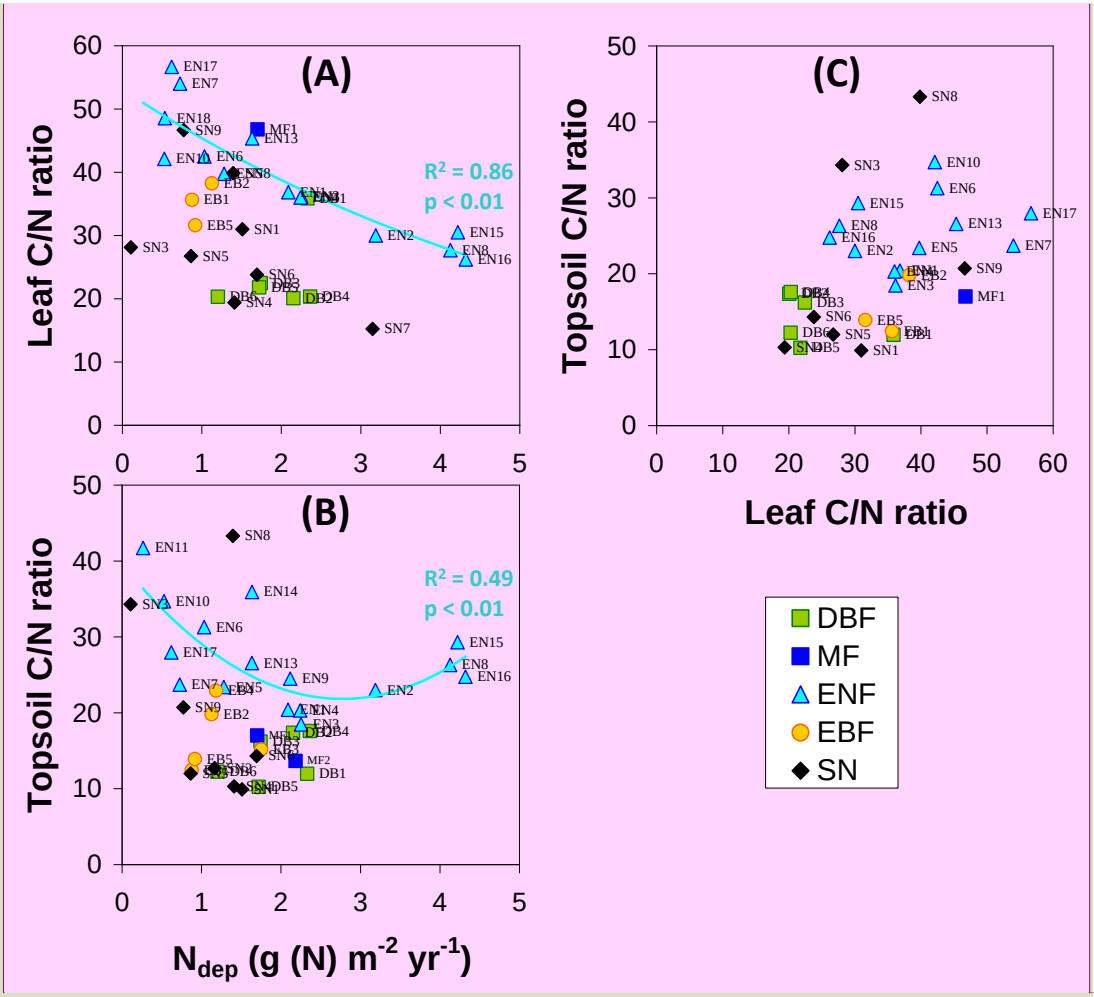

Figure 9. Relationships of leaf (A) and top soil (B) C/N ratios with atmospheric nitrogen deposition (N_{dep}), and to each other (C), in different ecosystem types (DBF: deciduous broadleaf forests, MF: mixed forests, ENF: evergreen needleleaf forests, EBF: evergreen broadleaf forests, SN: short semi-natural vegetation).

Commentaire [c29]:
Added R^2 and p statistics to trend lines for ENF

Table 1. Overview of ecosystem and climatic characteristics and inter-annual mean ecosystem/atmosphere exchange fluxes for forest and semi-natural short vegetation sites.

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 2. Main acronyms and abbreviations used in the study

Carbon fluxes and stocks	
NEE	Net ecosystem exchange
GPP	Gross primary productivity
NPP	Net primary productivity
NEP	Net ecosystem productivity
NECB	Net ecosystem carbon balance
NBP	Net biome productivity
R_{eco}	Ecosystem respiration
R_{aut}	Autotrophic respiration
R_{het}	Heterotrophic respiration
R_{soil}	Soil (heterotrophic and rhizospheric) respiration
SCE	Soil CO_2 efflux measured by chamber methods
CSE_{obs}, CSE_{mod}	Carbon sequestration efficiency, calculated from EC observations or by modelling
SOM	Soil organic matter
CSOM	Carbon stock in soil organic matter
CR	Carbon stock in roots
CLITT	Carbon stock in litter layers of the forest floor
CLBS	Carbon stock in leaves, branches and stems
LeafC	Leaf carbon content
DIC, DOC	Dissolved inorganic or organic carbon
dC/dN, $dNEP/dN_{dep}$	Response (slope) of ecosystem C productivity versus atmospheric N_r deposition
Nitrogen fluxes and stocks	
N_{dep}	Total (wet+dry) atmospheric reactive nitrogen deposition
N_r	Reactive nitrogen
N_{min}, N_{org}	Mineral or organic reactive nitrogen forms
LeafN	Leaf nitrogen content
DIN, DON	Dissolved inorganic or organic nitrogen
DIN_{TF}	Throughfall inorganic N_r deposition
WSON	Wet deposition of water-soluble organic nitrogen
Water budget terms	
SWC	Soil water content
WFPS	Water-filled pore space
ET	Evapotranspiration
Ecosystem characteristics	
PFT	Plant functional type
ENF	Evergreen needleleaf forest
DBF	Deciduous broadleaf forest
MF	Mixed (needleleaf/broadleaf) forest
EBF	Evergreen broadleaf forest
SN	Short semi-natural vegetation
H	Canopy height
DBH	Tree diameter at breast height (forests)
LAI	Leaf area index
SD	Stand density (forests): number of trees per unit area
MAT	Mean annual temperature
MAP	Mean annual precipitation
Methods and general terminology	
EC	Eddy covariance
DELTA	DEnuder for Long-Term Atmospheric sampling
BASFOR	BASic FORest ecosystem model
CTM	Chemical transport model
EMEP	European Monitoring and Evaluation Programme (www.emep.int)
GHG	Greenhouse gas
GWP	Global warming potential
CEIP	CarboEurope Integrated Project
NEU	NitroEurope Integrated Project
FLUXNET	Worldwide carbon flux monitoring network

Table 3. Summary of the main methods used to quantify carbon, nitrogen and greenhouse gas fluxes and budgets for the 31 forests and 9 short semi-natural vegetation sites included in this study. Horizontal bars (green: forests; blue: short semi-natural vegetation) indicate the percentages of study sites with available data (filled bars), or without available data (open bars). See also Supplement Tables S6-S7 for details at individual sites.

Fluxes and budgets	Components	Experimental data (this study) Methods (selected references)	Literature and other data mining Methods (selected references)	Modelling (this study) Models (selected references)
Carbon	Net ecosystem exchange (NEE)	Eddy covariance (1)		
	Net ecosystem productivity (NEP)	Gap-filled from NEE (14)		BASFOR (18)
	Gross primary productivity (GPP)	Inferred from NEE (14)		BASFOR (18)
	Ecosystem respiration (R_{eco})	Inferred from NEE (14)		BASFOR (18)
	Soil respiration (R_{soil})	Static/dynamic chambers (12)	Static/dynamic chambers (19)	
	Heterotrophic respiration (R_{het}) Ratio R_{het} / R_{soil}		Root exclusion, trenching, girdling, isotopic methods (19)	BASFOR (18)
	Dissolved organic/inorganic carbon (DIC / DOC) losses	Suction cups (9); peatbog stream sampling (3)	Lysimeter / suction cups (6); weir (8) ; ground- and ditch-water sampling (7)	
	Soil-atmosphere CH_4 fluxes	Static chambers (12) Laboratory soil bioassay (15)	Eddy covariance (10) ; static chambers (7)	
Nitrogen	Atmospheric N_r concentrations	DELTA (17)		EMEP (16)
	Atmospheric dry deposition	Inferential method (5)		EMEP (16)
	Atmospheric wet deposition (Inorganic N_r)	Bulk samplers (2) Wet-only samplers	Regional networks / kriging	EMEP (16)
	Atmospheric wet deposition (wet-soluble organic N_r, WSON)	Bulk samplers (Dämmgen, 2006) Wet-only samplers		
	Throughfall N_r deposition	Throughfall precipitation collectors		
	Dissolved inorganic nitrogen (DIN) losses	Suction cups (9)	Lysimeter / suction cups (11)	IFEF model (4)
	Dissolved organic nitrogen (DON) losses	Suction cups (9)	Lysimeter / suction cups (11)	
	Soil-atmosphere NO fluxes	Dynamic open chambers (12) Laboratory soil bioassay (15)	Dynamic open chambers (13)	BASFOR (18)
	Soil-atmosphere N_2O fluxes	Static chambers (Luo et al., 2012) Laboratory soil bioassay (15)	Static chambers (13)	BASFOR (18)

[1] Aubinet et al. (2000) ; [2] Dämmgen (2006) ; [3] Dinsmore et al. (2010) ; [4] Dise et al. (2009) ; [5] Flechard et al. (2011) ; [6] Gielen et al. (2011) ; [7] Hendriks et al. (2007) ; [8] Ilvesniemi et al. (2009) ; [9] Kindler et al. (2011) ; [10] Kowalska et al. (2013) ; [11] Legout et al. (2016) ; [12] Luo et al. (2012) ; [13] Pilegaard et al. (2006) ; [14] REddyProc (2019) ; [15] Schaufler et al. (2010) ; [16] Simpson et al. (2012) ; [17] Tang et al. (2009) ; [18] van Oijen et al. (2005) ; [19] See references in Table S7.

Commentaire [c30]: Table 3 added to summarize methods

<u>Supplement to</u> 'Carbon/nitrogen interactions in European forests and semi-natural vegetation. Part I: Fluxes and budgets of carbon, nitrogen and greenhouse gases from ecosystem monitoring and modelling'

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

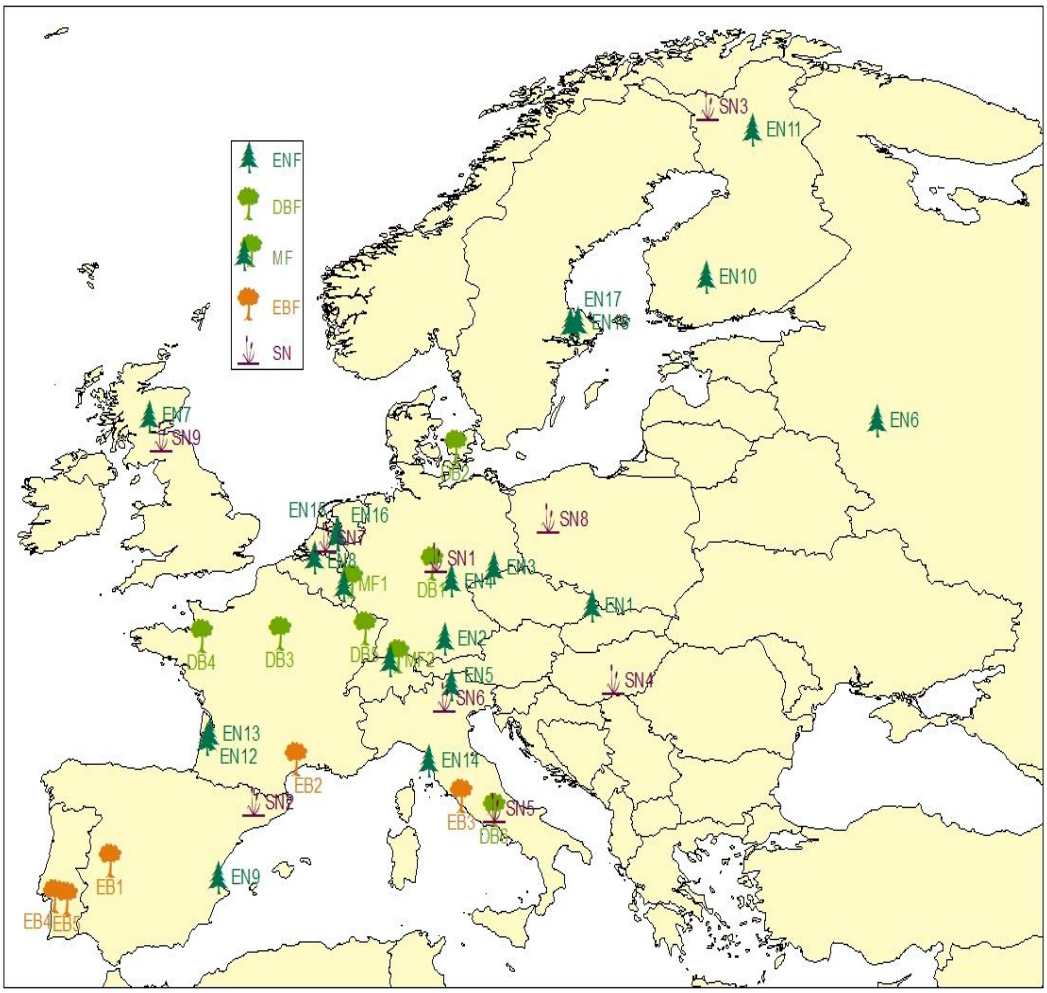

Figure S1. Geographical distribution of NitroEurope nitrogen deposition monitoring sites. Key: ENF: evergreen needle leaf forest; DBF: deciduous broadleaf forest; MF: mixed deciduous/needle leaf forest; EBF: Mediterranean evergreen broadleaf forest; SN: short semi-natural vegetation.

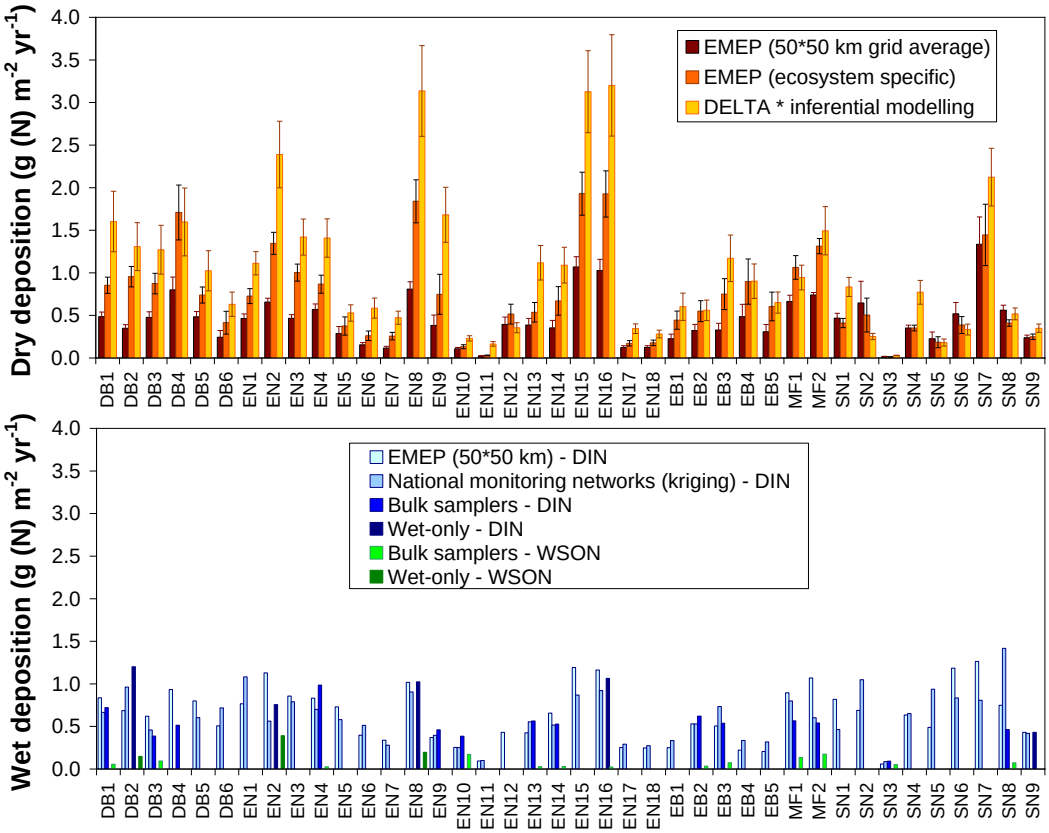

Figure S2. Comparison of annual dry and wet atmospheric inorganic nitrogen deposition estimates at the 31 forest (DB, EN, EB, MF) and 9 short semi-natural (SN) monitoring sites of this study, obtained using different methods and data sources. Top panel: dry deposition estimates from i) grid-average modelled outputs of the European-scale EMEP chemistry and transport model (Simpson et al., 2012) taken from the 50*50 km grid cells corresponding to each site (average and standard deviation for the years 2007-2010); ii) ecosystem-specific EMEP modelled dry deposition within each grid; and iii) inferential ensemble modelling estimates (in-situ atmospheric DELTA-Nr measurements coupled to several dry deposition models applied at the ecosystem scale; Flechard et al., 2011). Lower panel: dissolved inorganic (DIN) or water- soluble organic (WSON) N_r wet deposition estimates from i) modelled EMEP 50*50 km gridded outputs (average for the years 2007-2010); ii) spatial interpolation by kriging of rainfall concentration data from national and continental networks of precipitation monitors, scaled for local rainfall; iii) bulk precipitation samplers installed at 13 sites of the NEU network; iv) wet-only precipitation samplers at 6 sites.

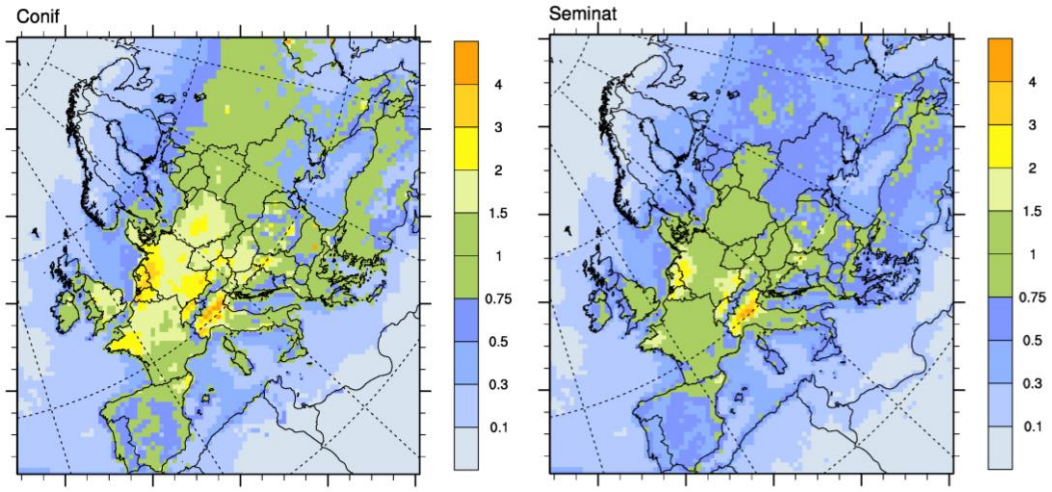

Figure S3. Comparison of total atmospheric nitrogen deposition (g (N) m^{-2} yr^{-1}) to coniferous forests (left) and to semi-natural vegetation (right), modelled by the EMEP chemical transport model (year 2010). The difference is mainly due to dry deposition, which is larger over forests due to larger surface roughness, which generates faster vertical turbulent transfer and larger deposition velocities.

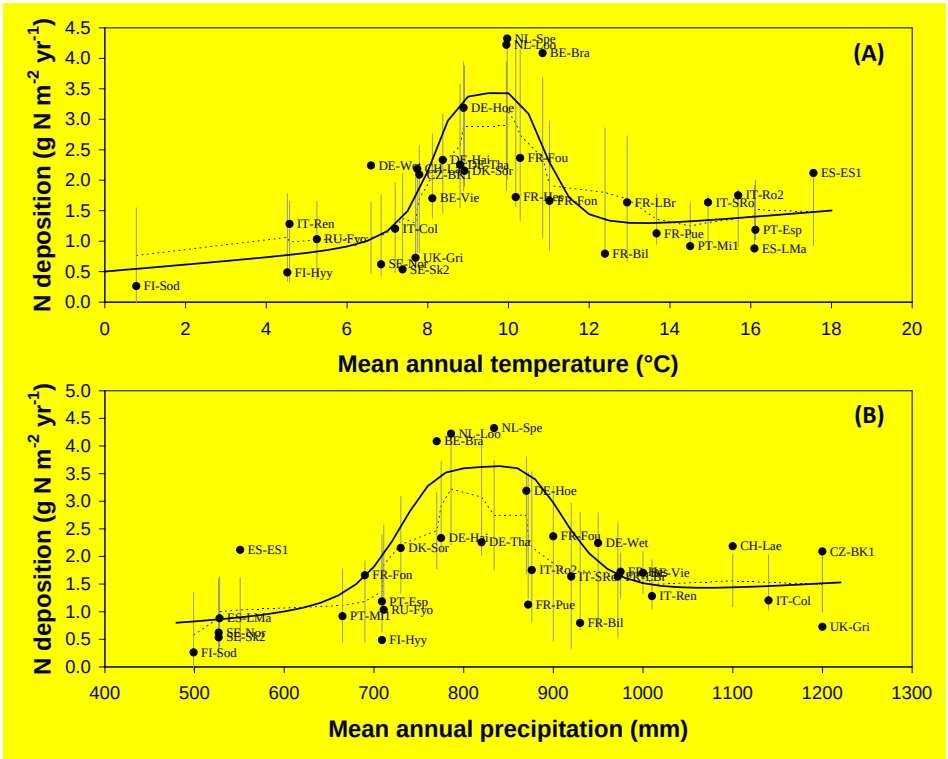

Figure S4. Spatial variations in measurement-based nitrogen deposition, plotted as a function of (A) mean annual temperature (MAT) and (B) mean annual precipitation (MAP). Temperature and precipitation are not direct determinants of N_{dep}, but the geographical occurrence of peak N_{dep} levels in mid-range for both MAT and MAP means that the relationship of forest productivity to N_{dep} cannot be considered independently of climate at the European scale.

Commentaire [c1]: New Figure S4 added to Supplement

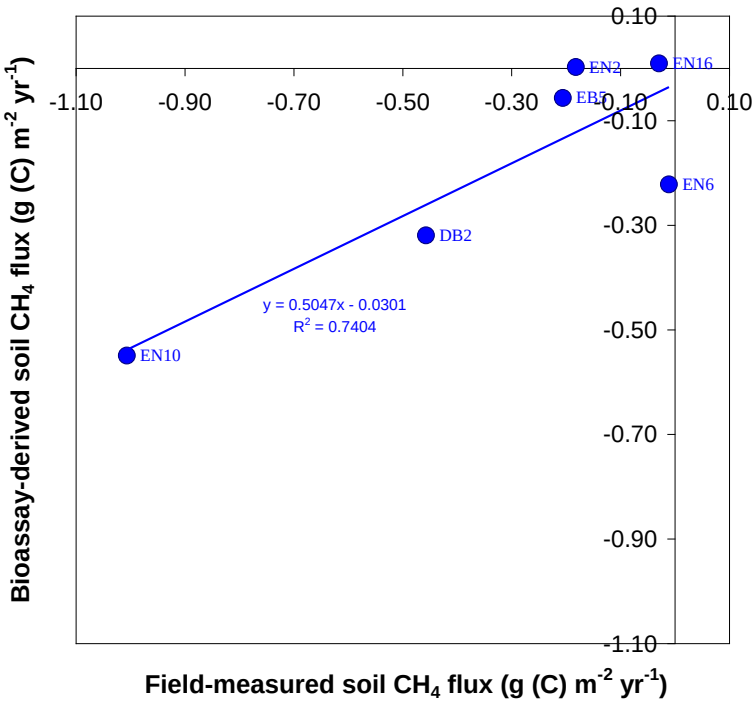

Figure S5. Comparison of net annual forest soil CH_4 fluxes calculated from field measurements using closed soil chambers and CH_4 flux values derived from laboratory-based bioassay experiments (see Methods for details).

Table S1. Selected references for the CarboEurope IP and NitroEurope IP monitoring sites included in this sudy.

Site Name	Location, Country	PFT short name	Reference
DE-Hai	Hainich, Germany	DB1	Knohl et al. (2003)
DK-Sor	Sorø, Denmark	DB2	Pilegaard et al. (2003)
FR-Fon	Fontainebleau-Barbeau, France	DB3	Delpierre et al. (2016)
FR-Fgs	Fougères, France	DB4	Huet et al. (2004)
FR-Hes	Hesse, France	DB5	Granier et al. (2008)
IT-Col	Collelongo, Italy	DB6	Scartazza et al. (2004)
CZ-BK1	Bily Kriz, Czech Rep.	EN1	Sedlak et al. (2010)
DE-Hoe	Höglwald, Germany	EN2	Kreutzer et al. (2009)
DE-Tha	Tharandt, Germany	EN3	Grünwald and Bernhofer (2007)
DE-Wet	Wetzstein, Germany	EN4	Anthoni et al. (2004)
IT-Ren	Renon, Italy	EN5	Marcolla et al. (2005)
RU-Fyo	Fyodorovskoye, Russia	EN6	Milyukova et al. (2002)
UK-Gri	Griffin, UK	EN7	Clement et al. (2003)
BE-Bra	Brasschaat, Belgium	EN8	Neirynck et al. (2005)
ES-ES1	El Saler, Spain	EN9	Sanz et al. (2002)
FI-Hyy	Hyytiälä, Finland	EN10	Vesala et al. (2005)
FI-Sod	Sodankylä, Finland	EN11	Thum et al. (2008)
FR-Bil	Bilos, France	EN12	Moreaux et al. (2011)
FR-LBr	Le Bray, France	EN13	Rivalland et al. (2005)
IT-SRo	San Rossore, Italy	EN14	Chiesi et al. (2005)
NL-Loo	Loobos, Netherlands	EN15	Dolman et al. (2002)
NL-Spe	Speulderbos, Netherlands	EN16	Erisman et al. (1999)
SE-Nor	Norunda, Sweden	EN17	Grelle et al. (1999)
SE-Sk2	Skyttorp, Sweden	EN18	Lindroth et al. (2008)
ES-LMa	Las Majadas, Spain	EB1	Casals et al. (2009)
FR-Pue	Puechabon, France	EB2	Allard et al. (2008)
IT-Ro2	Roccarespampani, Italy	EB3	Tedeschi et al. (2006)
PT-Esp	Espirra, Portugal	EB4	Pereira et al. (2007)
PT-Mi1	Mitra, Portugal	EB5	Pereira et al. (2007)
BE-Vie	Vielsalm, Belgium	MF1	Aubinet et al. (2018)
CH-Lae	Lägeren, Switzerland	MF2	Ruehr et al. (2010)
DE-Meh	Mehrstedt, Germany	SN1	Don et al. (2009)
ES-VDA	Vall d'Alinya, Spain	SN2	Sebastià (2007)
FI-Lom	Lompolojänkkä, Finland	SN3	Aurela et al. (2009)
HU-Bug	Bugac, Hungary	SN4	Nagy et al. (2007)
IT-Amp	Amplero, Italy	SN5	Gavrichkova et al. (2010)
IT-MBo	Monte Bondone, Italy	SN6	Vescovo and Gianelle (2006)
NL-Hor	Horstemeer, Netherlands	SN7	Hendriks et al. (2007)
PL-wet	POLWET/Rzecin, Poland	SN8	Kowalska et al. (2013)
UK-AMo	Auchencorth Moss, UK	SN9	Flechard et al. (1998)

Table S2. Forest species composition and stand characteristics

Site Name	PFT [(1)] Short name	Dominant species	%	Secondary species	%	Density (year) trees ha^{-1}	Thinning (year) Fraction removed	DBH (year) cm	Basal area (year) m^2 ha^{-1}
DE-Hai	DB1	Fagus sylvatica	64	Fraxinus excelsior	27	330 (2003)		35 (2004)	34.2 (2000)
DK-Sor	DB2	Fagus sylvatica	80	Picea abies, Larix decidua	20	283 (2003)		38 (2002)	29.1 (2006)
FR-Fon	DB3	Quercus petraea	75	Carpinus betulus	20	410 (2007)		39 (2006)	27.5 (2006)
FR-Fgs	DB4	Fagus sylvatica	100			725 (2010)	26% (2010)	17.24 (2010)	15.3 (1997)
FR-Hes	DB5	Fagus sylvatica	90			2328 (2010)	25% (1999-2000)	9.554 (2008)	19.8 (2005)
IT-Col	DB6	Fagus sylvatica	100			825 (2007)		25 (2007)	40.6 (2007)
CZ-BK1	EN1	Picea abies	99			1440 (2008)		15.6 (2008)	27.4 (2008)
DE-Hoe	EN2	Picea abies	100			621 (2009)		41.3 (2009)	70.9 (2009)
DE-Tha	EN3	Picea abies	72	Pinus sylvestris	15	477 (1999)	15% (2002)	33 (1999)	35.8 (1999)
DE-Wet	EN4	Picea abies	100			410 (2004)		32.7 (2004)	35.7 (2004)
IT-Ren	EN5	Picea abies	85	Pinus cembra	12	270 (2000)		22 (2000)	29.0 (2000)
RU-Fyo	EN6	Picea abies	85	Betula pendula	15	558 (2011)		26.3 (2011)	30.5 (2011)
UK-Gri	EN7	Picea sitchensis	97	Pseudotsuga menziesii	2	2215 (1996)	20% (1996)	12.4 (2001)	31.2 (2001)
BE-Bra	EN8	Pinus sylvestris	55	Quercus robur	23	362 (2003)	30% (1999)	29.7 (2003)	28 (2001)
ES-ES1	EN9	Pinus halepensis	90	Pinus pinea	10	70 (2006)		31.8 (2006)	6.96 (2006)
FI-Hyy	EN10	Pinus sylvestris	75-100	Picea abies, Betula pubescens	5-15	2500 (2002)	25% (2002)	19.5 (2008)	24.0 (2008)
FI-Sod	EN11	Pinus sylvestris	95			2100 (2005)		18.5 (2000)	19.4 (2000)
FR-Bil	EN12	Pinus pinaster	100			1800 (2008)	88% (2008)	3.99 (2010)	
FR-LBr	EN13	Pinus pinaster	100			313 (2007)		33.6 (2007)	28.2 (2007)
IT-SRo	EN14	Pinus pinaster	84	Pinus pinea	12	565 (2005)		29 (2005)	39 (2003)
NL-Loo	EN15	Pinus sylvestris	89	Betula pendula	3	362 (2002)		28.9 (2008)	23.26 (2008)
NL-Spe	EN16	Pseudotsuga menziesii	100			612 (2010)	42%(1996)	31.46 (2010)	47.21(2010)
SE-Nor	EN17	Pinus sylvestris	63	Picea abies	33	600 (2000)		21.8 (2004)	41.7 (2004)
SE-Sk2	EN18	Pinus sylvestris	76	Picea abies	24	1023 (2006)		15.8 (2004)	22.3 (2005)
ES-LMa	EB1	Quercus ilex	100			25 (2009)		45 (2010)	4.1 (2006)
FR-Pue	EB2	Quercus ilex	95			6074 (2009)	15% (2005)	6.8 (2009)	33 (2009)
IT-Ro2	EB3	Quercus cerris	90	Quercus pubescens, Q. suber and Q. ilex	10	3300 (2007)		8.6 (2007)	22.8 (2007)
PT-Esp	EB4	Eucalyptus globulus	100			983 (2002)		9.9 (2002)	19.2 (2002)
PT-Mi1	EB5	Quercus ilex, Quercus suber	90			35-45 (2004)		34-42 (2005)	
BE-Vie	MF1	Fagus sylvatica	43	Pseudotsuga menziesii	37	230 (2007)		34 (2002)	33.72 (2002)
CH-Lae	MF2	Fagus sylvatica	25	Picea abies	21			36 (2010)	

[(1)] PFT (plant functional types): DB: deciduous broadleaf forest; EN: evergreen needleleaf forest; EB: evergreen broadleaf forest; MF: mixed forest.

Table S3. Ecosystem carbon and nitrogen contents and C/N ratios at the sites included in this study.

Site Name	PFT Short name	Foliar N % DW	Foliar C % DW	Foliar C/N	Branches C/N	Stems C/N	Roots C/N	Topsoil C/N	Topsoil SOC gC/kg dry soil
DE-Hai	DB1	1.33	47.9	36	n.a.	n.a.	n.a.	12.0	64
DK-Sor	DB2	2.36	47.3	20	215 *	215 *	173	17.4	72
FR-Fon	DB3	2.18	49.0	22	n.a.	n.a.	n.a.	16.2	24
FR-Fgs	DB4	2.44	49.5	20	n.a.	n.a.	n.a.	17.6	18
FR-Hes	DB5	2.22	48.4	22	n.a.	n.a.	n.a.	10.2	n.a.
IT-Col	DB6	2.47	50.0	20	214	646	300[§] / 40[¶]	12.2	126
CZ-BK1	EN1	1.30	47.9	37	n.a.	n.a.	n.a.	20.4	175
DE-Hoe	EN2	1.50	45.0	30	259 *	259 *	261	23.0	102
DE-Tha	EN3	1.40	50.6	36	316	681	n.a.	18.5	333
DE-Wet	EN4	1.40	50.5	36	n.a.	n.a.	n.a.	20.3	425
IT-Ren	EN5	1.16	46.1	40	n.a.	n.a.	n.a.	23.4	240
RU-Fyo	EN6	1.17	49.8	42	n.a.	n.a.	n.a.	31.3	470
UK-Gri	EN7	0.90	48.6	54	n.a.	n.a.	n.a.	23.7	229
BE-Bra	EN8	1.76	48.6	28	n.a.	184***	n.a.	26.3	18
ES-ES1	EN9	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	24.5	110
FI-Hyy	EN10	1.18	49.7	42	274 *	274 *	240	34.7	65
FI-Sod	EN11	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	41.7	30
FR-Bil	EN12	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
FR-LBr	EN13	1.18	53.6	45	n.a.	n.a.	n.a.	26.6	30
IT-SRo	EN14	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	35.9	n.a.
NL-Loo	EN15	1.55	47.3	30	n.a.	n.a.	n.a.	29.3	7
NL-Spe	EN16	1.82	47.6	26	169 *	169 *	121	24.8	156
SE-Nor	EN17	0.85	48.0	57	n.a.	n.a.	n.a.	28.0	367
SE-Sk2	EN18	0.99	48.0	49	n.a.	n.a.	n.a.	n.a.	n.a.
ES-LMa	EB1	1.32	47.0	36	n.a.	n.a.	n.a.	12.4	28
FR-Pue	EB2	1.23	47.0	38	n.a.	n.a.	n.a.	19.8	155
IT-Ro2	EB3	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	15.2	92
PT-Esp	EB4	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	22.9	18
PT-Mi1	EB5	1.5**	47.4**	32**	n.a.	n.a.	n.a.	13.9	12
BE-Vie	MF1	1.05	48.9	47	n.a.	n.a.	n.a.	17.0	84
CH-Lae	MF2	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	13.7	82
DE-Meh	SN1	1.41	43.7	31	n.a.	n.a.	n.a.	9.9	24
ES-VDA	SN2	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	12.7	67
FI-Lom	SN3	1.53	43.0	28	n.a.	n.a.	n.a.	34.3	476
HU-Bug	SN4	2.24	43.5	19	n.a.	n.a.	n.a.	10.3	86
IT-Amp	SN5	1.63	43.5	27	n.a.	n.a.	n.a.	12.0	67
IT-MBo	SN6	1.94	46.2	24	n.a.	n.a.	n.a.	14.3	107
NL-Hor	SN7	2.77	42.2	15	n.a.	n.a.	n.a.	n.a.	n.a.
PL-wet	SN8	1.11	44.1	40	n.a.	n.a.	n.a.	43.3	456
UK-AMo	SN9	1.04	48.7	47	n.a.	n.a.	n.a.	20.7	533

* Stems and branches pooled; ** Measured on an adjacent younger stand at PT-Mi3; ***Measured in 2015 at EN8; [§] Coarse roots; [¶] Fine roots.

Table S4. Soil physical properties at the sites included in this study.

Site Name	PFT Short name	Depth m	Terrain slope °	Soil texture	Soil FAO class	Clay %	Sand %	Silt %	Φ_{WP} $m^3 m^{-3}$	Φ_{FC} $m^3 m^{-3}$	Φ_{SAT} $m^3 m^{-3}$	Topsoil pH
DE-Hai	DB1	0.48-0.71	2.5	Loamy clay	Dystric Cambisol	40	4	56	0.16	0.24	0.59	5.6
DK-Sor	DB2	0.70 - 0.85	0.0	Loamy sand	Stagnic Luvisol/Mollisol	15	51	34	0.09	0.20	0.46	3.4
FR-Fon	DB3	0.8	0.6	Loamy sand	Gleyic Luvisol	n.a.	n.a.	n.a.	0.09	0.20	0.30	3.8
FR-Fgs	DB4	0.4-1.5	1.1	Silty loam	Alocrisol/Neoluvisol	15	10	75	0.15	0.41	0.56	4.3
FR-Hes	DB5	1.45	1.4	Silty clay	Stagnic Luvisol	29	7	64	0.10	0.40	0.60	n.a.
IT-Col	DB6	0.3-1.5	n.a.	Silty clay	Humic Alisol	59	8	32	0.19	0.29	0.60	5.1
CZ-BK1	EN1	0.55	15.0	Loamy sand	Ferric Podzol	21	22	58	0.09	0.20	0.37	3.4
DE-Hoe	EN2	1	2.0	Sandy loam	Dystric Cambisol	5	64	31	0.18	0.27	0.37	3.1
DE-Tha	EN3	1.15	2.0	Silty loam	Dystric Cambisol	38	21	41	0.07	0.16	0.44	3
DE-Wet	EN4	0.44	n.a.	Sandy loam	Podzol	33	23	44	0.18	0.27	0.47	3.1
IT-Ren	EN5	0.5	11.0	Loamy sand	Haplic Podzol	11	55	34	0.14	0.34	0.61	4.9
RU-Fyo	EN6	0.3	1.0	Peat	Umbric Albeluvisol	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	3.1
UK-Gri	EN7	1	5.0	Loamy clay	Haplic Podzol	n.a.	n.a.	n.a.	0.34	0.44	0.62	3.3
BE-Bra	EN8	2	0.2	Loamy sand	Albic Hypoluvic Arenosol	4	89	7	0.05	0.13	0.37	3.3
ES-ES1	EN9	2	0.0	Loamy sand	Arenosol	0	72	28	0.09	0.20	0.37	6.3
FI-Hyy	EN10	0.61	2.4	Loamy sand	Haplic podzol	9	61	18	0.08	0.55	0.57	3.6
FI-Sod	EN11	1.5	0.0	Loamy sand	Haplic podzol	0	94	4	0.11	0.26	0.47	3.3
FR-Bil	EN12	1	0.0	Pure sand	Umbric Podzol	2	98	0	0.04	0.17	0.44	n.a.
FR-LBr	EN13	1.2	3.6	Pure sand	Haplic Podzol	2	98	0	0.03	0.17	0.44	3.4
IT-SRo	EN14	0.5-3	0.0	Loamy sand	Regosol	4	93	3	0.05	0.09	0.38	n.a.
NL-Loo	EN15	1	0.0	Pure sand	Haplic Arenosol/Podzol	2	96	2	0.03	0.12	0.25	3.4
NL-Spe	EN16	1	2.0	Loamy sand	Haplic Podzol	1	70	29	0.09	0.20	0.37	2.9
SE-Nor	EN17	0.7	0.0	Silty sand	Dystric Regosol	5	65	30	0.05	0.17	0.40	3.2
SE-Sk2	EN18	1	0.0	Silty sand	Eutric Cambisol	n.a.	n.a.	n.a.	0.06	0.18	0.37	n.a.
ES-LMa	EB1	>0.8	2.9	Loamy sand	Stagnic Alisol	9	80	11	0.11	0.26	0.48	5.4
FR-Pue	EB2	0.5	0.0	Silty loam	Rhodo-chromic luvisol	39	26	35	0.22	0.35	0.42	6.6
IT-Ro2	EB3	0.5	0.0	Clayey sand	Chromic Luvisol/Cambisol	33	54	14	0.37	0.52	0.55	5.6
PT-Esp	EB4	0.65	0.0	Clayey loam	Dystric Cambisol	30	n.a.	n.a.	0.05	0.14	0.41	4.5
PT-Mi1	EB5	1	0.0	Loamy sand	Dystric Cambisol	6	89	5	0.05	0.14	0.37	5
BE-Vie	MF1	1.5	1.7	Sandy silt	Dystric Cambisol	18	27	55	0.19	0.34	0.44	3.4
CH-Lae	MF2	1	24.0	Clayey loam	Rendzic Leptosol	n.a.	n.a.	n.a.	0.26	0.36	0.41	5.9
DE-Meh	SN1	n.a.	n.a.	Clayey loam	Haplic Luvisol	31	2	67	0.31	0.43	0.50	7.2
ES-VDA	SN2	n.a.	n.a.	Loamy clay	Lithic Cryrendoll	n.a.	n.a.	n.a.	0.34	0.44	0.48	5.9
FI-Lom	SN3	n.a.	n.a.	Peat	Dystric Histosol	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	4.4
HU-Bug	SN4	n.a.	n.a.	Loamy sand	Chernozem	20	60	20	0.09	0.20	0.25	6.9
IT-Amp	SN5	n.a.	n.a.	Silty clay	Haplic Phaeozem	55	13	32	0.37	0.53	0.59	5.2
IT-MBo	SN6	n.a.	n.a.	Sandy loam	Humic Umbrisol	18	40	42	0.37	0.59	0.79	4.8
NL-Hor	SN7	n.a.	n.a.	Peat	Eutric Histosol	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
PL-wet	SN8	n.a.	n.a.	Peat	Haplic Podzol	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	4.9
UK-AMo	SN9	n.a.	n.a.	Peat	Dystric Cambisol	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	3.4

Table S5. Site micro-climatological characteristics

Site Name	PFT Short name	MAT °C	GDD5[(1)] °C days	R_g[(2)] GJ m^{-2} yr^{-1}	MAP mm yr^{-1}	AET(EC)[(3)] mm yr^{-1}	AET=(Rn-H-G)/λ[(4)] mm yr^{-1}	PET[(5)] (Penman 1948) mm yr^{-1}	PET[(6)] (Penman-Monteith FAO) mm yr^{-1}	IWA1[(7)] AET/PET	IWA2[(8)] λE / (λE+H)	O3 AOT40[(9)] ppb hours
DE-Hai	DB1	8.4	1915	3.83	775	290	592	763	1029	0.33	0.59	16062
DK-Sor	DB2	8.9	1896	3.83	730	404	632	766	774	0.53	0.74	9318
FR-Fon	DB3	11.0	2535	4.25	690	628	689	876	1021	0.66	0.74	15470
FR-Fgs	DB4	10.3	2310	4.30	900	418	550	770	957	0.48	0.67	10049
FR-Hes	DB5	10.2	2347	4.28	975	276	836	945	961	0.29	0.68	18109
IT-Col	DB6	7.2	1596	5.65	1140	346	698	1034	1087	0.32	0.45	26492
CZ-BK1	EN1	7.8	2150	3.54	1200	294	513	790	1357	0.27	0.44	17814
DE-Hoe	EN2	8.9	2141	4.44	870	736	796	878	1245	0.69	0.68	20923
DE-Tha	EN3	8.8	2104	4.00	820	387	570	962	1708	0.29	0.50	17030
DE-Wet	EN4	6.6	1571	3.69	950	331	658	787	1218	0.36	0.61	16534
IT-Ren	EN5	4.6	1219	4.86	1010	629	197	867	1095	0.65	0.47	22229
RU-Fyo	EN6	5.3	1699	3.38	711	304	544	668	707	0.44	0.55	5814
UK-Gri	EN7	7.7	1444	2.78	1200	578	453	564	721	0.84	0.81	4591
BE-Bra	EN8	10.8	2402	3.79	850	305	433	915	815	0.35	0.48	10593
ES-ES1	EN9	17.6	4677	5.88	551	640	565	1252	1231	0.48	0.30	24709
FI-Hyy	EN10	3.8	1358	2.89	709	334	453	639	971	0.40	0.52	3329
FI-Sod	EN11	-0.4	905	2.76	527	245	-87	412	264	0.72	0.43	1083
FR-Bil	EN12	12.4	3053	4.61	930	628	582	1015	508	0.82	0.55	9647
FR-LBr	EN13	12.9	3142	4.48	972	689	677	1057	1148	0.62	0.60	12530
IT-SRo	EN14	14.9	3866	5.01	920	679	770	1192	1407	0.46	0.48	36194
NL-Loo	EN15	10.0	2232	3.70	786	579	847	925	534	0.79	0.61	7399
NL-Spe	EN16	10.0	2207	3.52	834	456	561	811	1634	0.37	0.60	7399
SE-Nor	EN17	6.8	1534	3.43	527	450	446	678	996	0.58	0.64	4258
SE-Sk2	EN18	7.4	1676	3.38	527	281	433	661	800	0.38	0.47	3741
ES-LMa	EB1	16.1	4309	6.01	528	558	880	1551	692	0.50	0.49	16116
FR-Pue	EB2	13.7	3385	5.23	872	397	622	1239	1123	0.34	0.43	20756
IT-Ro2	EB3	15.7	3876	5.81	876	507	835	1242	1424	0.37	0.60	29879
PT-Esp	EB4	16.1	4297	6.22	709	649	486	1771	1705	0.39	0.52	19437
PT-Mi1	EB5	14.5	3808	6.20	665	n.a.	n.a.	1365	1457	n.a.	n.a.	17076
BE-Vie	MF1	8.1	1752	3.63	1000	288	463	754	789	0.37	0.48	14750
CH-Lae	MF2	7.7	1836	4.53	1100	757	427	823	974	0.84	0.62	20927
DE-Meh	SN1	9.1	2081	3.56	547	235	482	740	609	0.35	0.56	15630
ES-VDA	SN2	6.4	1319	5.30	1064	440	615	884	537	0.62	0.62	18254
FI-Lom	SN3	-1.0	748	2.76	521	254	142	353	237	0.86	0.63	784
HU-Bug	SN4	10.7	2744	4.47	500	441	533	953	945	0.47	0.58	21047
IT-Amp	SN5	9.8	2722	5.27	1365	693	817	1105	899	0.69	0.68	26492
IT-MBo	SN6	5.1	1134	4.96	1189	480	616	639	503	0.85	0.76	29924
NL-Hor	SN7	10.8	2430	3.83	800	602	657	685	890	0.78	0.81	5568
PL-wet	SN8	8.5	2086	3.72	550	521	649	871	716	0.67	0.73	11809
UK-AMo	SN9	7.6	1430	2.83	1165	130	230	466	275	0.27	0.34	4174

[(1)] sum of growing degree days >5°C; [(2)] annual global radiation sum; [(3)] and [(4)] actual evapotranspiration from eddy covariance or from energy balance; [(5)] and [(6)] potential evapotranspiration from Penman 1948 or Penman-Monteith/FAO equations; [(7)] and [(8)] indices of water availability calculated from two different ratios; [(9)] accumulated O_3 exposure over 40-ppb threshold (EMEP model).

Table S6. Overview of available measured C, N and GHG flux data from the NEU and CEIP projects and from online databases and the literature. See Materials and Methods in the main body of the article for details.

Site	**PFT**	**CO_2 / NEE**	**Soil CO_2 efflux**	**R_{het}/R_{soil} ratio**	**DIC/DOC**	**Soil CH_4 flux**	**Soil core CH_4 flux**	**Soil N_2O flux**	**Soil core N_2O flux**	**Soil NO flux**	**Soil core NO flux**	**DIN leaching**	**Dry dep.**	**Wet dep.**	**Wet dep.**
Name	*Short name*	*EC flux N years*	*Field chamb.*	*Field exp.*	*Field exp.*	*EC/ field chamb.*	*Lab. bioassay*	*EC/ field chamb.*	*Lab. bioassay*	*Field chamb.*	*Lab. bioassay*	*Field exp.*	*DELTA * dep. model*	*Bulk/wet-only sampler*	*National network*
DE-Hai	DB1	4	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	n	**Y**	**Y**	**Y**	**B**	**Y**
DK-Sor	DB2	6	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**W-O**	**Y**
FR-Fon	DB3	3	n	**Y**	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	**B**	**Y**
FR-Fou	DB4	7	**Y**	**Y**	n	n	n	n	n	n	n	**Y**	**Y**	**B**	n
FR-Hes	DB5	6	**Y**	**Y**	n	n	n	n	n	n	n	n	**Y**	n	**Y**
IT-Col	DB6	5	**Y**	**Y**	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
CZ-BK1	EN1	6	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
DE-Hoe	EN2	3	**Y**	n	n	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**W-O**	**Y**
DE-Tha	EN3	7	**Y**	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
DE-Wet	EN4	5	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	n	**Y**	**Y**	**Y**	**B**	**Y**
IT-Ren	EN5	7	**Y**	**Y**	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
RU-Fyo	EN6	7	n	n	n	**Y**	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	n	**Y**
UK-Gri	EN7	1	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
BE-Bra	EN8	7	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	n	**Y**	**Y**	**Y**	**W-O**	**Y**
ES-ES1	EN9	3	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	**B**	**Y**
FI-Hyy	EN10	6	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**B**	**Y**
FI-Sod	EN11	5	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
FR-Bil	EN12	4	n	n	n	n	n	n	n	n	n	n	**Y**	n	n
FR-LBr	EN13	5	**Y**	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	**B**	**Y**
IT-SRo	EN14	7	**Y**	**Y**	n	n	n	**Y**	n	**Y**	n	n	**Y**	**B**	**Y**
NL-Loo	EN15	7	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	n	**Y**	**Y**	**Y**	n	**Y**
NL-Spe	EN16	2	**Y**	n	n	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**Y**	**W-O**	**Y**
SE-Nor	EN17	3	**Y**	**Y**	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
SE-Sk2	EN18	1	**Y**	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
ES-LMa	EB1	6	**Y**	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
FR-Pue	EB2	6	**Y**	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	**B**	**Y**
IT-Ro2	EB3	7	**Y**	**Y**	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	**B**	**Y**
PT-Esp	EB4	7	**Y**	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
PT-Mi1	EB5	3	**Y**	n	n	**Y**	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	n	**Y**
BE-Vie	MF1	7	**Y**	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	**B**	**Y**
CH-Lae	MF2	5	**Y**	**Y**	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	**B**	**Y**
DE-Meh	SN1	3	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
ES-VDA	SN2	5	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
FI-Lom	SN3	3	n	n	n	**Y**	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	**B**	**Y**
HU-Bug	SN4	5	n	n	n	**Y**	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	n	**Y**
IT-Amp	SN5	3	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
IT-MBo	SN6	5	n	n	n	n	**Y**	n	**Y**	n	**Y**	n	**Y**	n	**Y**
NL-Hor	SN7	4	n	n	**Y**	**Y**	n	n	n	n	n	n	**Y**	n	**Y**
PL-wet	SN8	4	n	n	n	**Y**	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	**B**	**Y**
UK-AMo	SN9	6	n	n	**Y**	**Y**	**Y**	**Y**	**Y**	n	**Y**	n	**Y**	**W-O**	**Y**

Table S7. Published estimates of annual soil respiration fluxes and ratios of heterotrophic to total soil respiration (R_{het}/R_{soil}) for the forest sites of this study

Site	R_{soil} (g (C) m^{-2} yr^{-1})	R_{het}/R_{soil}	References
DB1	884	0.52	Kutsch et al. (2010), Knohl et al. (2008), Moyano et al. (2008)
DB2	740	0.61	Wu et al. (2013), Janssens and Pilegaard (2003)
DB3	na	0.77	Chemidlin Prevost-Boure et al. (2009)
DB4	na	0.46	Huet, S. (2004)
DB5	620	0.37	Epron et al. (1999a,b), Ngao et al. (2007, 2012)
DB6	879	0.29	Matteucci et al. (2000), Luyssaert et al. (2007)
EN1	na	na	
EN2	783	na	Luo et al. (2012)
EN3	na	na	
EN4	na	0.47	Moyano et al. (2008)
EN5	1015	0.58	Rodeghiero et al. (2005), Luyssaert et al. (2007)
EN6	765	0.38-0.62	Šantrůčková et al. (2010), Kurbatova et al. (2013)
EN7	na	na	
EN8	620	0.36	Curiel Yuste et al. (2005), Chiti et al. (2011)
EN9	na	na	
EN10	606	0.64	Kolari et al. (2009), Korhonen et al. (2009)
EN11	na	na	
EN12	na	na	
EN13	na	na	
EN14	872	0.74	Matteucci et al. (2015)
EN15	937	0.38	Kruit et al. (2003), Luyssaert et al. (2007)
EN16	653	na	Frumau et al. (2011)
EN17	1227	0.71	Moren et al. (2000), Widen and Majdi (2001)
EN18	na	na	
MF1	867	na	Longdoz et al. (2000)
MF2	888	0.54	Ruehr et al. (2010), Ruehr and Buchmann (2010)
EB1	561	na	Casals et al. (2009), Gimeno et al. (unpublished)
EB2	762	na	Misson et al. (unpublished)
EB3	904	0.77	Rey et al. (2002)
EB4	778	na	Luyssaert et al. (2007)
EB5	na	na	

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