Dear Editor,

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We would like to thank you for your reply. We agree that the manuscript can be improved by shortening some parts. We have made significant reductions but retained the essential information. The information of which data have partly been published in other papers is now included in table 3 and 4. We have addressed all specific comments made be the two referees.

We are looking forward to your reply, Best wishes,

10 Stephanie Jones

# **Referee 1**

## 15 General comment

We would like to thank the referee's comments. The novelty of our manuscript is related to the synthesis of a long dataset, which hasn't previously been undertaken in such detail over such a long timescale. We have addressed all specific comments.

### 20 Specific comments

Page 2, line 51: Insert 'from' here once again to make clear that it is all CH4 emission.

"from" has been inserted

Page 4, line134: Well, this seems a bit awkward. Can you quantify this 'something', also for the temperature?

We apologise for this mistake, the actual standard deviations have been added (947  $\pm$  234 mm, 9.0  $\pm$  0.4  $\odot$ )

Page 4, line136: perenne (do not start with capital P). It is unusual for permanent grassland to have such a clear dominance of one species. Are there any grassland renovation measures taken regularly?

*Lolium Perenne* has been changed to *Lolium perenne*. There have not been any grassland renovation measures taken place during the 9 years of presented measurements (2002-2010). The field has been ploughed and reseeded to improve the quality of the grassland, but only in March 2011.

35 Page 5, line 142: How large was the grassland area considered? What was the grazing management like? Rotational grazing? It seems rather erratic from Fig. 1, at least for heifers.

The size of the grassland field was 5.424 ha. The grazing management can be described as rotational grazing: The farmer moved animals from neighbouring fields to the Easter Bush field when the grass was high enough and then moved them back to neighbouring fields again when the grass was too short at the Easter Bush field to allow the recovery and growth of the pasture plants after grazing. This information has now been added and "grazed continuously" replaced with "grazed rotationally".

Page 5, line 148: Not at all in the later years?

Yes, the grass was not cut in all the later years

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Page 5, line 151: Rephrase (application was applied)

The sentence has been changed to: "In 2008 an additional fifth mineral N application was added, using urea instead of ammonium nitrate fertiliser."

50 Page 6, line 173: Does this include animal excretion?

> No, animal excretion is not included here as animal excretion is N that is recycled from plant and soil uptake.

Page 6 line 191: Similar for ewes with lamb.

We agree and included a sentence stating that ewes were not milked but their milk was only consumed 55 by their lambs.

Page 6, line 193: As it appears here as output, I feel the excretion should also be taken account of as input to the soil. Although it is recycled from plant and soil uptake, it is returned to the system and part of it is lost as CH4 (or leaching). Otherwise, the balance would suggest that animals fed on the site (which is considered in the balance), but were removed (e.g. to a stable) for excretion. This was not the case, however.

Carbon lost as CH<sub>4</sub> from animal excretion originates from C uptake from plants and is therefore included in the NEE, thus FC<sub>CO2</sub>. Therefore, if we counted C input from excretion as input, this C would be account for twice.

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Page 7, line 232: What was the estimate based on? Did he weigh his trucks before going into the silo? Was it estimated by plant height and density?

The farmer estimated the harvest amount based on information from plot experiments with similar management in the area, taking the plant height of the grass at Easter Bush at the time of cutting into account.

Page 7, line 239: Were they weighed again at the end/before giving birth?

Heifers were weight before they came onto the field and again at the end of the season, when they left the field.

Page 7, line 240: How realistic is the assumption of constant weight when they are lactating?

Most ewes will lose weight during lactation. However, as ewes were fed extra protein (standard cakeconcentrate) to reduce weight loss during lactation, we assumed a constant weight. The information that the ewes received extra feed during lactation is now added in the paper.

Page 8, line 242: Some of the weight of the lambs at least would be bones, though...

We agree with the referee. Using a bone content of 20 % for sheep (Lambe et al. 2007; Livestock Science 107, 37–52) and 14 % for Heifers (Navajas et al. 2010; Animal, 4:11, pp 1810–1817) and a C
and N content of bones of 20 and 7 %, respectively (Marchand, R.F. (1842): Ueber die chemische Zusammensetzung der Knochen. Journal für praktische Chemie 27, 83–97. DOI: 10.1002/prac.18420270117), we corrected the C and N offtake of meat by including the bone content and adjusted values in table 4 and 5 and Figure 3 and 4.

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Page 8, line 247: ...(Roche, 1995)`

Roche J., 1995 has been changed to Roche, 1995

Page 8, line 253: Well, but this is where the leachate would go to...

- 95 The reviewer is right that the hollow terrain position likely receives inputs of water and leached nitrogen compounds from upper terrain positions. Inputs of water and nitrogen into the hollow terrain position likely occur via lateral flow of groundwater or capillary rise from shallow groundwater. Since we used a water balance model based on water holding capacity, information of the quantity of (ground) water that is transported in the hollow laterally or via capillary rise was not available. Therefore we
- 100 restricted the quantification of leaching losses to the upslope position, where the downward movement of water and solutes prevailed.

We have changed the description of water balance model (page 8, line 263) into:

The latter was derived from a soil water model based on balancing daily precipitation and evaporation considering the water holding capacity of the soil (Kindler et al., 2011). This model did not allow the calculation of upward water fluxes with capillary rise from groundwater. We therefore only used the data for the upslope position for the calculation of leaching losses. The data of the hollow position were not used, because the soil was frequently water logged and likely influenced by capillaries from shallow groundwater and lateral flow of groundwater.

110 Page 8, line 256: How did you make sure there was no further mineralisation/denitrification in the samples before collection? Two to three weeks seem quite long time periods.

We agree that transformation of especially nitrogen species in sampling bottles before collection must be considered. Therefore, we placed the sampling bottles in an insulated aluminium box that was placed in a soil pit in order to keep the temperature of the sampling bottles as close as possible to the soil temperature and as a real as possible. We ministed the participation of "action in the soil temperature of the sampling bottles as close as possible to the soil

115 temperature and as cool as possible. We rejected the possibility of "poisoning" the leachate in the sampling bottles, because the addition of most poisoning agents like cyanide or strong acids (e.g. HCl) interferes with other analytical parameters, while the reliability of the reduction of microbial activity is questionable. Furthermore, we strived to minimize the use of harmful substances.

Microbial transformations in the sampling bottles are also reduced by the fact that the leachate passes a filter with very fine pores, the suction cup (pore width  $< 1.6 \mu$ m), before it enters the sampling bottle.

In the case of our study, a loss of nitrogen via denitrification also is unlikely, since the flasks were flushed with oxygen-rich atmospheric air every two weeks and the evacuated sampling bottles contained a rest of oxygen-rich air, because the vacuum that was applied equalled only 40kPa. Therefore, the oxidation of ammonium to nitrate in the sampling bottles is in principle possible. This

- 125 would mean that ammonium concentrations could be underestimated, while nitrate concentrations could be overestimated, without altering the total nitrogen concertation in the leachate and hence total nitrogen losses. The same applies to very biodegradable dissolved organic matter congaing organicallybound nitrogen (DON). However, we would like to note that these very biodegradable dissolved matter compounds as well as ammonium are commonly oxidized rapidly already in the soil.
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Page 9, line 289: Annual rainfall was measured directly at Easter Bush?

Yes, the annual rainfall was measured directly at Easter Bush

Page 9, line 299: Close second bracket.

135 The missing bracket has been added.

Page 9, line 300: Unite references. IDEM = Integrated Deposition Model The references have been united and the abbreviation of the model added 140 Page 9, line 306: When describing leaching measurement, you mention a slope, thus I suspect the area was not flat. How does this work with EC? How large was the area considered?

The maximum gradient in the field is 2.5 %, so although not completely flat, the topography is only gently sloping. The field has a mean slope is from NW to SE, with the steepest slope some 100 m to the NW of the flux tower. The mean flux footprint reflects the prevailing wind direction from the SW and secondarily from the NE, with the bulk of the contribution coming from within 50 m. The EC measurements thus sample the flatter areas of the field. Standard corrections are applied in processing to rotate co-ordinates relative to the mean wind flow in each half hour period. In this way, the fluxes are measured relative to the plane where mean vertical wind speed is zero, rather than assuming a horizontal ground surface.

150 For these reasons, we do not think slope is a serious issue at this site.

Page 10, line 313: And to deposit excreta?

Yes, chambers were also accessible for animals to deposit excreta, if they wished to. This information has been added.

Page 10, line 317: How?

Samples were transferred from tedlar bags to glass vials by a syringe, which was fitted with a three way tap. Glass vials were flushed with the samples using two needles (one fitted to the syringe and one stuck into the lid of the vial) in order not to over pressurise the vials.

Page 10. Line 325: calendar

Callendar has been changed to calendar

165 Page 10, line 326: Did you also simulate fluxes for the times you had data and compare the results with your measurements? How good was the fit?

LandscapeDNDC was tested in detail with available data on plant growth soil temperature, moisture, inorganic soil N concentration NO and  $N_2O$  which resulted in general good agreement of simulations and measurements. This sentence included in 2.8.3

170 Model simulations match the general pattern of high nitrate leaching (~50 kg N ha<sup>-1</sup>) and N<sub>2</sub>O emissions (~6 kg N ha<sup>-1</sup> yr<sup>-1</sup>) as well as moderate NO emissions (~2 kg N ha<sup>-1</sup> yr<sup>-1</sup>).

 $R^2$  of simulated and measured daily soil temp and moisture are 0.6- 0.9.  $r^2$  of soil inorganic N and N<sub>2</sub>O and NO are less good (<0.3) at daily time step but this is mostly related to the patchiness of emissions

by grazing and deposition of urine and faces. Nevertheless, aggregating to longer time steps (monthly to vearly) increases  $r^2$  to values up to 0.5-0.8. 175

Results except NO emissions are published in Molina et al., 2016. (Molina-Herrera S, Haas E, Klatt S, Kraus D., Augustin J., Magliulo V., Tallec T., Ceschia E., Ammann C., Lubet B., Skiba U., Jones S., Brümmer C., Butterbach-Bahl, Kiese R. 2016. A modeling study on mitigation of N<sub>2</sub>O emissions and NO3 leaching at different agricultural sites across Europe using Landscape DNDC. Science of the Total

180 Environment 553, 128-140.)

> Page 10, line 343: area Are has been changed to area

185 Page 10, line 344: Again, how did these compare to measured data? Please see comment to Page 10, line 326.

Page 11, line 353: This could have been measured easily. It will also change over the season. There are lots of assumptions in the dataset.

190 The N content in the grass has actually been measured monthly, although not in all years. Using those measured values to calculate CP we now re-calculated daily N excretions for ewes, lambs and heifers. For years, where no grass N content was measured we used an averaged value calculated over all years.

We agree with the referee that there are many assumptions in the dataset. Unfortunately we didn't have funding to measure all C and N concentrations and fluxes within the grassland over 10 years to

195 calculate a C and N budget, but the data were collected during three different projects (GREENGRASS, CarboEurope and NitroEurope), with each different objectives. Therefore gaps in measuring series were unavoidable.

Page 11, line 358: Add information that variation in excretion depends on variation in weight of the 200 animals.

The information that the variation in excretion depends on weight variation has been added.

Page 12, line 384: Similar information is missing for N2O-EC data above.

Details for the gap filling method of  $N_2O$ -EC data are described in Jones et al 2011. We have now 205 added this reference referring to gap filling method.

Page 13, line 433: Did you still take them into account in the balances?

No we, did not take them into account as they were assumed to be negligible.

210 Page 18, line 583: Below, it is said that this is the value for grazed years, while it was 2 g less over all years. Please check.

It is correct that the value for 2004-2010, which is the period for which soil stock measurements are available, corresponds to the "grazed only" years (when no cut was carried out). Over all years the N storage from flux calculations is indeed 2 g less than for the period of 2004-2010, mainly because N was lost in 2002 (see table 4).

215 was lost in 2002 (see table 4).

Page 18, line 583: Given the standard deviations, I guess there is no significant difference. So be careful not to overinterpret 'differences'.

We agree that there is no significant difference and therefore should not over interpret differences.

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Page 19, line 634: Repetition from above.

The first sentence of 3.5 has been deleted and the GWP of  $CH_4$  and  $N_2O$  included in the method section under 2.3

225 Page 19, line 644: Did you never measure uptake?

Yes, we did measure  $N_2O$  uptake on occasional days (see Jones et al. 2011). But on an annual basis, we measured a net loss of  $N_2O$  for each year and as budgets are presented annually we did not mention negative  $N_2O$  fluxes.

- 230 Page 20, line 663: For trying to estimate whether the system is in equilibrium, it would be good to have data about grassland renovation/ reseeding etc. For a more than 20 year-old permanent grassland, a cover of 99% Lolium perenne is rarely found. If it was renovated in between (and maybe not so long before or even during the study), this would make more sense. However, in that case, the soil would have been disturbed (including mineralisation etc.) and a new equilibrium would have to be established.
- 235 There have not been any grassland renovation measures taken place during the 9 years of presented measurements (2002-2010) nor in the last 20 years. This information has now been added.

The field has been ploughed and reseeded to improve the quality of the grassland, but only in March 2011 (after the end of the period reported in this manuscript).

240 Page 21, line 684: Delete 'is'.

'is' has been deleted

Page 21, line 704: Where/on which scale?

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The stated 85% are actually not crops but nitrogen in crops harvested or imported into the EU, whereas only 15% is used to feed humans directly (Sutton et al. 2011). We apologise for this mistake and corrected the numbers and added the relevant reference in the text.

Page 21, line 706: Or a larger fraction of meat produced from grassland only.

We agree with the referee's comment and included this information.

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Page 21, line 708: Delete ':'.

':' has been deleted

Page 23, line 759: Did you consider freeze-thaw cycles?

255 No, we did not specifically consider freeze-thaw cycles.

Page 23, line 769: IPCC No wonder, as there is a wealth of processes producing N2O in soils.

Yes, we appreciate that the processes producing  $N_2O$  are complex. These are discussed in our and other papers we referred to in this manuscript.

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Page 23, line 776: What are these two numbers? Minimum and maximum? Emission from mineral and organic fertilizer???

Those two numbers are minimum and maximum  $N_2O$  emission factors shown in Table 6, whereas the maximum is actually 7.2 not 7.4%. This has been corrected and clarified.

#### 265

Page 24, line 792: predominantly

'Predominately' has been change to 'predominantly'

Page 24, line 804: Please check sentence (text within/outside brackets).

# 270 The sentence has been corrected.

Page 24, line 806: cattle-grazed?

'cattle grazed' has been changed to 'cattle-grazed'

275 Page 24, line 817: ...for N losses. 'from' has been changed to 'for'

Page 25, line 838: Well, the grass is used for feeding animals also if it is cut. Just the emissions are produced somewhere else.

280 We agree with the referee's comment. However, as mentioned in section 2.3, we set the boundary for inputs and outputs for out budget by the field perimeters. Therefore emissions produced off the field are not included in the budget. This has been clarified.

Page 25, line 848: If water and nutrients are not limiting.

285 This has information has been added.

Page 26, line 873: ...where...? 'Whereas' has been changed to 'where'

290 Page 26, line 877: and CH4'and CH<sub>4</sub>' has been added in the sentence

Page 27, line 893: comma should not be superscript. Comma is now not superscript anymore

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Page 27, line 897: I do not see the contradiction. Or do you mean 'where' rather than 'whereas'? Yes, we actually mean 'where', not 'whereas'.

Page 27, line 899: Close bracket.

300 The bracket has been closed

Page 27, line 905: have been shown

'have shown' has been changed to 'have been shown'

305 Page 28, line 946: not be ... 'be' has been added after 'not'

Page 29, line 968: loose

'lose' has been changed to 'loose'

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Page 30, line 998: If the total number of animals increases. If the same number of animals is just fed in a different way, changes in emissions are rather small.

We agree with the referee's comment and clarified this in the text.

315 Page 30, line 1007: Insert reference to Smith 2014, as this is nothing you showed in this study. The reference of Smith 2014 has been inserted

Page 44, table 5: You give different numbers for all years. What do you mean with this? Or do you mean that the 2010 value is the average of 2002-2009? Then the asterisk after 'Leaching' is confusing.

320 The 2010 value is the average of 2002-2009, the asterisk after 'leaching' has been deleted.

# **Referee 2**

## **General comment**

325 We would like to thank the referee for the comments.

Firstly, we accept the observation of the referee that the many of the measurements reported covered only for a part of the experimental period, and that some modelled and literature values were included. The data were collected during three different projects (GREENGRASS, CarboEurope and NitroEurope), each with different objectives. Therefore some gaps in the series of observations were unavoidable. Although some of the data has already been published, preceding publications focus on shorter timescales and percover (often subannuel) objectives. The nevelty of this menuscript is indeed

- shorter timescales and narrower (often subannual) objectives. The novelty of this manuscript is indeed the aggregation of data to derive a total C and N budget and to present data on soil C and N stock over a long time period.
- 335 Secondly, we agree that by and large we cannot separate the effects of management and weather on C and N cycling processes. Our aim was firstly to characterise the overall C and N budgets for the site but then as a secondary objective provide commentary on clear relationships that were apparent between aspects of management and climate that were linked to the observed nutrient fluxes. Doing this over the long time period, with the typical variations in farm managements and climate, will provide a more
- 340 robust interpretation of the key components one may want to pay attention to in future with improved measurements and modelling (for C: NEE leaching of DOC and harvest and for N; N<sub>2</sub> loss and NO<sub>3</sub> and DON leaching, which are difficult to measure). So there is an important message to the researchers and modellers to characterise and improve our understanding of these poorly described nutrient fluxes.
- 345 Considering the third point addressed by the referee, we appreciate that there is a spatial variably. However, spatial heterogeneity was not a specific focus of this manuscript (for the GHGs this is covered in Skiba et al. 2013 (Biogeosciences, 10, 1231–1241, 2013). The hollow position (wet area) which was not used for NO<sub>3</sub> leaching measurements only covered a small part of the field and was therefore avoided for N<sub>2</sub>O, CH<sub>4</sub> and soil measurements, but included in the footprint of the EC measurements.
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## Specific comments

Page 2, line 42: Better "stocks". Pools in plural suggests that different pools per element were assessed.

We replaced "pool" with "stocks"

355 Page 3, line87: What is the source for this?

Increased N input through intensive N management will lead to a decrease in the C/N ratio of the soil resulting in increased nitrification and denitrification processes and thus  $N_2O$  losses. Nitrogen fertilisation is also a key factor inhibiting CH<sub>4</sub> oxidation in soils. The NH<sub>4</sub><sup>+</sup> from the added fertiliser has

an inhibitory effect on CH<sub>4</sub> oxidation, as it competes with CH<sub>4</sub> as a substrate of the enzyme methane monooxygenase (MMO). Mosier et al. (1991) reported an inhibition of CH<sub>4</sub> uptake on grassland by 41 % after the application of N fertilizer.

Page 3, line 99: It might be helpful to start with this part of the introduction, which contains the motivation to study grassland biogeochemistry. The paragraphs above should be shortened, e.g. very general scentences like the very first could be left out.

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We agree with the reviewer and have changed the structure of the introduction and shortened some parts.

Page 4, line 127: quotation mark a typo?

370 Yes, this is a typo; Greengrass, EC EVK"-CT2001-00105 has been changed to Greengrass, EC EVK2-CT2001-00105

Page 4 line 134: "something" - should not this be a number (for temperature es well)

We apologise for this mistake, the actual standard deviation have been added (947  $\pm$  234 mm, 9.0  $\pm$  375  $\,$  0.4 C)

Page 4, line 136: common names are "perennial ryegrass" and "white clover"; "perenne" in scientific name should be lower case.

This has been corrected

#### 380

Page 5, line 142: The size of the field should be given here already. What was the relief of the field (slope and hollow are mentioned later)?

The size of the grassland field (5.424 ha) has been added. The maximum gradient in the field is 2.5 %, so although not completely flat, the topography is only gently sloping. The field has a mean slope from NW to SE, with the steepest slope some 100 m to the NW of the EC flux tower. This information has been added.

Page 5, line 145: What was the grazing management? Continuous or rotational stocking? Animal numbers appear to change frequently (Figure 1a). Did animals receive any additional feed while on pasture? Was the field subdivided? It seems that in 2004 and 2005 spreading of organic manure occcurred during grazing or was immediately followed by grazing.

The grazing management can be described as rotational grazing; The farmer moved animals from neighbouring fields to the Easter Bush field when the grass was high enough and then moved them to neighbouring fields again when the grass was too short at the Easter Bush field to allow the recovery and growth of the pasture plants after grazing. This represents a common management practice by farmers in this region. This information has now been added and "grazed continuously" replaced with "grazed rotationally". The field was not subdivided. The ewes got additional feed in spring (standard cake concentrate), this information has now been added. The slurry spreading was indeed followed shortly by grazing; the slurry was spread specifically for a measurement campaign.

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Page 5, line 147: bracket missing

Instead of adding a bracket, the bracket before "Scottish Agricultural College" has been replaced by a semicolon.

405 Pate 7, line 215: I do not think the "equals" sign should be used here, since 1 kg N cannot be equal to 1 kg C (even if its climate forcing effect was equal).

We agree that this sign is confusing. We therefore replaced the equal sign with "corresponds to".

Page 8, line 265: bracket missing

410 Missing bracket has been added

Page 9, line 287: where

Were has been changed to where

415 Page 11, line 365: At how many quadrats of which size, and at which intervals?

50 quadrates of 0.25 m2 (0.5 m) were randomly thrown into the field to measure the species cover. The species composition has been measured once in 2002 and at monthly intervals in 2003. This information has been added.

420 Page 11, line 370: During which time period?

NEE was measured continuously from 1.January 2002 till 31.December 2010. This information has been added.

Page 12, line 384: The URL should probably be given in the reference list, or else in the text directly

425 The URL has been included in the text directly

Page 12, line 391: Is it legitimate to extrapolate from findings for forests to grasslands? Both vegetation types differ considerably in the proportion of photosynthetically active tissue to total tisse (and other aspects).

- 430 The Waring 1998 reference is indeed based on forest studies. The point is made more generally by Amthor (2000) and others, who show that there are strong theoretical constraints on this ratio, and that the estimate of 0.5 is reasonable. For example, Zhang et al (2009) estimate the variability across global biome types, and suggest a value of ~0.5 for grasslands. Note that the exact value assumed for this ratio is not critical to our results: the appearance of the pie chart would not change.
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Amthor, Jeffrey S. 2000. 'The McCree–de Wit–Penning de Vries–Thornley Respiration Paradigms: 30 Years Later'. Annals of Botany 86 (1): 1–20. doi:10.1006/anbo.2000.1175. Zhang, Yangjian, Ming Xu, Hua Chen, and Jonathan Adams. 2009. 'Global Pattern of NPP to GPP Ratio Derived from MODIS Data: Effects of Ecosystem Type, Geographical Location and Climate'. Global Ecology and Biogeography 18 (3): 280–90. doi:10.1111/j.1466-8238.2008.00442.x.

Page 13, line 438: A 10 m grid with 100 points would cover roughly one hectar. How was this positioned in the pasture (with respect to slope, to areas potentially preferred by livestock, etc.)? Was the same grid used for sampling in 2004 and 2011?

The objective for the soil sampling grid was to cover the main footprint area of the site, rather than covering the entire field. We apologies that this information was missing and have now added it to the manuscript. The grid was positioned independently from slope and potentially preferred areas to avoid biased sampling. For the resampling in 2011 the same grid was used, but the transect was chosen two meters further to the NW in order not to meet the same places we already sampled and disturbed before.

Page 14, line 467: As I understand it, only confidence intervals for parameter means were calculated (while ANOVA should involve comparison of group means).

455 It is correct that CIs for individual parameters were calculated. The confidence intervals for group means (e.g. data presented in fig 5) were used to establish whether or not differences were significantly different from zero. This has now been clarified in the text.

Page 14, line 472: Correlations between annual means of parameters are presented in the results. What type of correlation were these? Were the averages used in these correlations calculated by calendar year?

Correlations presented were positive linear correlations. Averages used in these correlations were calculated by calendar year. This information has been added

465 Page 16, line 547: at various points in the results, coefficients of determination are given for subdatasets from which certain years were omitted (e.g. L545-547), but no reason is given for excluding these particular years. If the reason is only that these years decrease R<sup>2</sup>, this would not appear to be legitimate.

We agree with the referee's comment and decided not to include coefficients of determination for subdatasets from which certain years were omitted.

Page 17, line 578: Means and standard deviation of the soil C and N concentrations (or stocks) both in 2004 and 2011 should be given in addition to the difference.

Mean and 95% CI of soil C and N concentrations are now included in the result section.

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Page 19, line 640: Which average? Per calendar year?

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Annual NGHGE were highly correlated with annual NEE. This has been clarified in the manuscript

Page 20, line 655: Inhibition of biological N2 fixation by high soil mineral N, and competitive exclusion of legumes are two different (and only indirectly linked) issues. Additionally, higher proportions of T. repens than observed here can persist even under comparable management intensity, so that height of N inputs alone probably cannot explain the virtual absence of legumes in this grassland.

We agree that the inhibition of biological  $N_2$  fixation by high soil mineral N and the competitive exclusion of legumes are two different issues. This has been clarified in the text. The grass was sown as a grass mixture only and no clover was sown, which is the reason for the low clover content. This information has now been added.

Page 20, line 681: NUE of different studies using either forage or animal product as the basis for calculating cannot be compared directly at all. This is not a matter of grazing vs. cutting, but of the trophic level that is considered in the evaluation of NUE. This whole paragraph should be rewritten taking this into account.

We agree that the inclusion of grazing adds a new trophic level and the NUE in the grass itself is not lowered due to grazing, but the sum of NUE of harvest and a lower NUE for grazing leads to a total lower annual NUE. We have now clarified this in the text.

Page 21, line 689: See my comment above: It would appear that other factors apart from fertilization limit competitive strength of legumes at this site.

Yes, the main reason was that no clover was sown. Furthermore low pH and wet conditions are not favourable for clover, both factors were present at the Easter Bush field.

Page 21, line 692: Was it not stated that fertilization stayed within recommended maximum levels? (L670f)

Yes, the fertilisation stayed within the recommended maximum levels. WE have changed "avoidance to over fertilisation" to "reduction of fertiliser input".

Page 22, line 722: For better comparison, g N m-1 y-1 should be used as unit here as well.

The units have been changed from kg N ha<sup>-1</sup> y<sup>-1</sup> to g N m<sup>-1</sup> y<sup>-1</sup>

510 Page 23, line 776: Table 6 shows nine years (although 2010 is only the mean of the other eight), and it seems that only four of them exceed 3.0%.

This has been corrected in the manuscript.  $N_2O$  fluxes were measured in 2010, therefor the  $N_2O$  emission factor in 2010 is not the mean of the other eight.

515 Page 23, line 780: Can this be generalized over all Scottish soils?

Yes, generally Scottish soils are high in SOC, see "Buckingham,S., Rees,R.M. & Watson,C.A. 2013. Issues and pressures facing the future of soil carbon stocks with particular emphasis on Scottish soils. *The Journal of Agricultural Science*, **152**, 699-715." We added this reference in the text.

520 Page 24, line 813: missing space

The missing space has been added

Page 29, line 955: missing space

The missing space has been added

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Page 34, line 1185: Wrong alphabetical order

The wrong alphabetical order has been corrected

Page 35, line 1254: duplicate

530 The duplicate reference has been deleted

Page 37, line 1333: Initials before name.

The initials have been moved after the name.

535 Page 42, line 1539 (table 1): Correct term would be "stocking rates". Abbreviation LSU should be explained here.

"Livestock densities" has been changed to "stocking rates" and the abbreviation LSU has been explained (*with asterisk*)

540 Page 42, line 1545 (table 2): volumetric or gravimetric content?

Values are given for volumetric soil water content (% by volume). This information has been added.

Page 42, line 1546 (table 2): Based on which temperatures?

The plant growing season begins and ends with periods of consecutive days where daily temperatures average more than 5C without any five-day spells of temperatures below 5C. This information has been included in table 2. For some reason some of the values for the length of growing season were not correct; values have now been corrected.

Page 42, line 1552 (table 3): "how much for measurements?" - is something missing here?

550 We apologise for the missing value and have now included the systematic uncertainty value for measuring leaching of 10%.

Page 43, table 4: Why was this not modelled, as for the other times where no measurement data were available? (I did not find this stated in the methods section).

555 Wet N deposition fluxes 2002-2009 are measured values and dry N deposition fluxes for 2002-2009 were modelled using measured concentration values. Neither of these data were available for 2010 when we originally wrote the manuscript and therefore averages were used. But in the meantime those

data are available and are now included in table 4. We apologise for this mistake. Missing C and N leaching values for 2010 have also been modelled and are now included in table 4.

560

Page 45, table 5: Since this is only the mean of 2002-2009, it should not be included here.

 $N_2O$  fluxes were measured till November 2010 and therefore 2010 data are not a mean of 2002-2009. We corrected the information in the method section, where it was stated that  $N_2O$  fluxes were only measured till December 2009. Originally we were not planning to include 2010 data in the manuscript, we apologise for the confusion.

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Page 46, figure caption 1: "Stocking density" would be more accurate here; LSU should be explained; Letters for sub-figures a, b, c, are not quite correct; "input and export" should be replaced by fluxes, otherwise it is not intuitive that input is negative and output positive; "fluxes" should probably be added to the y axis as well.

We have now changed "Live stock density" to "Stocking density", explained the term LSU and corrected the letters fro sub-figures.

Page 46, figure caption 2: Volumetric or gravimetric soil water content? Given that only annual averages are presented for the measured values, this Figure might not be necessary.

Volumetric soil water content was measured, which is now stated in the figure capture. We agree that we don't discuss inter-annual variations of C and N fluxes/concentrations and that therefore this figure is not needed.

580 Page 46, figure caption 4: Figures 3 and 4 essentially duplicate information contained in Tables 4 and 5; regarding the division into grazing-only vs. grazing-and-cutting years, please see my general comments

We think that Figure 3 and 4 are valuable to investigate the influence of different managements on the C ad N budget and therefore prefer to keep these figures in the manuscript.

585 Page 46, figure caption 5: Abbreviation NEE should be explained here

We have now included the explanation of the abbreviation NEE in the figure capture.

The nitrogen, carbon and greenhouse gas budget of a grazed, cut and fertilised temperate grassland

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## Abstract

Intensively managed grazed grasslands in temperate climates are globally important environments for the exchange of the greenhouse gases (GHGs) carbon dioxide ( $CO_2$ ), nitrous oxide ( $N_2O$ ) and methane 620 (CH<sub>4</sub>). We assessed the N and C budget of a mostly grazed, occasionally cut, and fertilized grassland in SE Scotland by measuring or modelling all relevant imports and exports to the field as well as changes in soil C and N stockspools over time. The N budget was dominated by import from inorganic and organic fertilisers (21.9 g N m<sup>2</sup> vr<sup>-1</sup>) and losses from leaching (5.3 g N m<sup>2</sup> vr<sup>-1</sup>), N<sub>2</sub> emissions and NOx and NH<sub>3</sub> volatilisation (6.4 g N m<sup>2</sup> yr<sup>-1</sup>). The efficiency of N use by animal products (meat and wool) 625 averaged 104% of total N input. On average over nine years (2002-2010) the balance of N fluxes suggested that  $6.67.2 \pm 4.46$  g N m<sup>-2</sup> y<sup>-1</sup> (mean ± confidence interval at p > 0.95) were stored in the soil. The largest component of the C budget was the net ecosystem exchange of  $CO_2$  (NEE), at an average uptake rate of  $218 \pm 155$  g C m<sup>-2</sup>v<sup>-1</sup> over the nine years. This sink strength was offset by carbon export from the field mainly as harvest (48.9 g C m<sup>2</sup> yr<sup>-1</sup>) and leaching (16.4 g C m<sup>2</sup> yr<sup>-1</sup>). The other export 630 terms, CH<sub>4</sub> emissions from the soil, from manure applications and enteric fermentation were negligible and only contributed to 0.02-4.2 % of the total C losses. Only a small fraction of C was incorporated into the body of the grazing animals. Inclusion of these C losses in the budget resulted in a C sink strength of  $163 \pm 140$  g C m<sup>-2</sup>y<sup>-1</sup>. On the contrary, soil stock measurements taken in May 2004 and May 2011 indicated that the grassland sequestered N in the 0-60 cm soil layer at 4.51  $\pm$  2.64 g N m<sup>-2</sup> y<sup>-1</sup> and lost C at a rate of 29.08  $\pm$  38.19 g C m<sup>-2</sup> y<sup>-1</sup>, respectively. Potential reasons for the discrepancy between 635 these estimates are probably an underestimation of C and N-losses, especially from leaching fluxes as well as from animal respiration. The average greenhouse gas (GHG) balance of the grassland was -366  $\pm$  601 g CO<sub>2</sub> eq m<sup>-2</sup> y<sup>-1</sup> and strongly affected by CH<sub>4</sub> and N<sub>2</sub>O emissions. The GHG sink strength of the NEE was reduced by 54% by CH<sub>4</sub> and N<sub>2</sub>O emissions. Enteric fermentation from the ruminating sheep 640 proved to be an important  $CH_4$  source, exceeding the contribution of N<sub>2</sub>O to the GHG budget in some years.

Keywords: grassland, carbon stocks, carbon sequestration, nitrogen cycling, budget, greenhouse gases

### Introduction

Nitrogen (N) is an essential component of proteins and genetic material and therefore required by all living organisms. Before N can be used by most organisms, inert atmospheric molecular nitrogen (Na) has to be transformed to reactive nitrogen (Nr). In an agricultural system Nr is added from inorganic fertiliser and cultivation induced biological N fixation and as organic compounds from organic manure 650 applications and dung from grazing animals. Generally N inputs into agricultural systems exceed outputs in the form of crops or animal off-takes (meat, milk and wool). In a steady state system the exceeding Nr is converted back to N2 through complete denitrification (Galloway et al., 2003). However, in agricultural systems the surplus of Nr is generally only partly converted to N2, while the rest is lost to the atmosphere or aquatic ecosystems as Nr, causing various environmental problems. 655

Managed grasslands cover<del>Worldwide</del> an estimated 26 % of earth's land surfaceland consists of managed grassland (FAOstat, 2008). The impact of reactive nitrogen (Nr) losses, carbon (C) sequestration and greenhouse gas (GHG) emissions (CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O) from managed these grasslands on the environment-is therefore of global importance and will become even more relevant in the future as an-increased standards of living in developed countries areis expected to result in a 660 rapid growth of livestock farming (Caro et al, 2014). Carbon (C) and N cycles in grasslands are intricately linked and tightly coupled in extensively managed low N grasslands, with sinks and sources in equilibrium. Converting such systems to intensively managed N fertilised grasslands in the short term may increase the soil organic carbon (SOC) pool from decomposed plant litter and root material 665 as well as through rhizodeposition (Rees et al., 2005) until a new equilibrium is reached (Soussana and Lemaire, 2014). In the case of the long term Broadbalk experiment in the UK, Rothamsted, this equilibrium was achieved after 50 years (Powlson et al, 2011). After the conversion to intensive N management, the tight coupling of the N and C cycles becomes disrupted, leading to emissions of  $N_2O$ and  $CH_4$  at rates which may outweigh the benefits of C sequestration. Several studies indicate that 670 managed grasslands can sequester C (Kim et al., 1992; Jones et al., 2006; Soussana et al., 2004; Ammann et al., 2007) however, uncertainties are high (Janssens et al., 2003). On the contrary, Smith

(2014) concluded from long-term experiments and chronosequence studies, that changes in agronomic management may lead to short-term C sequestration, but in the long-term, under constant management and environmental conditions, C stocks are relatively stable. In a grassland ecosystem the C balance is determined by the net biome exchange (the difference between total C input and losses). In managed grassland ecosystems exports through biomass harvesting, the addition of organic manures (from organic fertiliser additions and animal excretion) as well as CO<sub>2</sub> and CH<sub>4</sub> losses from animal respiration and enteric fermentation can make significant contributions to the C budget.

Worldwide an estimated 26 % of land consists of managed grassland (FAOstat, 2008). The 680 impact of Nr losses, C sequestration and GHG emissions (CO2, CH4 and N2O) from managed grasslands on the environment is therefore of global importance and will become even more relevant in the future as an increased standard of living in developed countries is expected to result in a rapid growth of livestock farming (Caro et al, 2014). Nutrient budgets are a valuable tool to summarise and understand nutrient cycling in agroecoystems and to assess their impact on the environment. As 685 imbalances are not sustainable in the long term, N and C budgets can be used as indicators and regulatory policy instruments for nutrient management in order to reduce losses and increase efficiency. So far, different Nr species have been looked at in separate studies according to their form and impact. Few studies have attempted to calculate N budgets from managed grasslands (e.g. Chen et al., 2004; Ammann et al., 2009; Chen et al., 2004; Nunez et al., 2010, Kramberger et al., 2015), 690 whereas C budgets have been assessed more often and are available for various ecosystems (e.g. Aubinet et al., 2000; Soussana et al., 2007; Ammann et al., 2007, Rytter et al. 2015). To calculate the total C and N budget of an ecosystem all import and export processes have to be assessed by measuring or estimating all imports and exports to an ecosystem. The other method is to measure differences in N and C stocks in the soil over time. This approach has the advantage that it requires the 695 measurement of only a single component of the system. However, a large number of samples are needed at an interval of more than 5 years before detectable changes may be statistically significant (Smith, 2004). Moreover this approach does not provide any information about the different processes leading to the final budget.

In this study we assessed the C and N budget from an intensively managed grassland in 700 Southern Scotland using both approaches. Here we report one of the most detailed analyses of C and N fluxes from a grassland ecosystem over 9 years (2002-2010). This study allowed an analysis of the importance of common grassland management practices such as cutting for silage, grazing of cattle and sheep at different stocking densities, N input by inorganic and organic fertiliser applications, as well as different weather conditions on the N, C and GHG balance. The data provide a unique overview of research undertaken withinwere collected in the frame of the three European projects GREENGRASS (Soussana et al., 2007), CarboEurope (Schulze et al., 2009) and NitroEurope (Sutton et al., 2007; Skiba et al., 2009).(EC EVK"-2-CT2001-00105), CarboEurope (GOCE-CT2003-505572) and NitroEurope (contract 017841).

710 **2. Methods** 

## 2.1 Site description

The experimental site, Easter Bush, is located in South East Scotland, 10 km South of Edinburgh (03°02'W, 55°52' N, 190 m a.s.l) with a \_\_\_\_\_Mmean annual rainfall (2002 2010) of was 947 ± 234something mm and the mean annual temperature was of 9.0 ± 0.4something °C (2002-2010). The field (5.424 ha) has been under permanent grassland management for more than 20 years with a species composition of >99% perennial rye-grass (*Lolium Pperenne*) and < 0.5% with clover (*Trifolium repens*). The grass was sown as a grass mixture (no clover) and no grassland renovation measures have taken place in the last 20 years. The field has a mean slope from NW to SE, with the steepest slope some 100 m to the NW of the eddy covariance tower. The maximum gradient in the field is 2.5%, so although not completely flat, the topography is only gently sloping. The soil type is an imperfectly drained Macmerry soil series, Rowanhill soil association (Eutric Cambisol) with a pH of 5.1 (in H<sub>2</sub>O) and a clay fraction of 20-26%. The ground water table was assumed to be at 0.85 m depth on average and the main rooting zone extends down to 0.31 m below soil surface.

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# 2.2 Grassland management

The grassland was grazed rotationallycontinuously throughout the experimental period by heifers in calf, ewes and lambs at different stocking densitiesy (Table 1 and Figure 1a). Animals were moved to neighbouring fields when the grass was too short for grazing to allow the recovery and growth of 730 pasture plants and moved back to the field when the grass was high enough, which represent a common management practise by farmers in this. Animals were counted several times per week and it was assumed that the animal number stayed constant between observations. Livestock units used for heifers, ewes and lambs were 0.75, 0.10 and 0.04, respectively (1 livestock unit has a standard live weight of 600 kg head<sup>-1-</sup>; (SACeottish Agricultural College, 1995). Lambs were present on the field from April to September only. The grass was cut for silage only in the first two years, on the 1<sup>st</sup> of June and 8<sup>th</sup> of 735 August 2002 and on the 29<sup>th</sup> of May 2003. Ammonium nitrate fertiliser was applied to the field 3-4 times per year, usually between March and July (56 kg N ha<sup>-1</sup> application<sup>-1</sup> on average). In 2008 an additional fifth mineral N application was addedpplied, using urea instead of ammonium nitrate fertiliser. Organic manure was applied on the 28<sup>th</sup> of September 2004 and 27<sup>th</sup> of March 2005 as cattle slurry, using a vacuum slurry spreader, Rates of N and C input from fertiliser and manure and export 740 from harvest are shown in(Table 4 and 5 and in Fig. 1 a) and b).

## 745 2.3. Annual budget calculations

We assessed the N and C budget by measuring or estimating the import and export of all relevant fluxes to and from the grassland field on an annual basis. Throughout the manuscript all fluxes are presented following the sign convention used in micrometeorology; fluxes from the ecosystem to the atmosphere are positive (exported from the field), while negative values indicate fluxes from the atmosphere to the ecosystem (imported to the field). We set the system boundary for inputs and exports of N and C by the field parimeters (covariant on a cross of 5.4 hz). The balance of all imports and exports results in the

field perimeters (covering an area of 5.4 ha). The balance of all imports and exports results in the observed changes of N and C on this field over time.

The change in the N balance ( $\Delta$ N) over time ( $\Delta$ t) of our grassland ecosystem can be written as:

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- $\Delta N/\Delta t = FN_{org fert.} + FN_{synt fert.} + FN_{N2} (biol. fixation) + FN_{dep.} + FN_{harvest} + FN_{animal} + FN_{leaching} + FN_{NH3/NOX(fert.,manure, animal)} + (1)$   $FN_{NOX(soil)} + FN_{N2O} + FN_{N2(denitr.)}$
- N imports include the addition of N from organic and inorganic fertiliser (FN<sub>org fert.</sub> + FN<sub>synt fert.</sub>), the fixation of N<sub>2</sub> through biological fixation (FN<sub>N2 (biol. fixation)</sub>) and the deposition of NH<sub>3</sub>, HNO<sub>3</sub>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> from dry, and NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> from wet deposition (summarised as FN<sub>dep</sub>.). Exports include the N lost from plant biomass at cuts for silage (FN<sub>harvest</sub>), the off-take of N in meat\_(including bones)-and wool from animals (FN<sub>animal</sub>), the loss of organic and inorganic dissolved N through leaching (FN<sub>leaching</sub>), the NH<sub>3</sub> and NOx emissions from volatilisation of inorganic and organic fertiliser spreading as well as from animal excretion (FN<sub>NH3/NOX(fert., manure, animal</sub>)), the emission of NOx from the soil (FN<sub>NOX(soil</sub>)), the emission of N<sub>2</sub>O from the soil (FN<sub>N2(denir.)</sub>).
- 770 The change in the C balance ( $\Delta$ C) over time equals the net biome production (NBP) and can be written for our site as:

$$\Delta C/\Delta t = NBP = FC_{CO2} + FC_{org fert} + FC_{animal} + FC_{CH4} + FC_{leaching} + FC_{harvest}$$
(2)

FC<sub>CO2</sub> represents the net ecosystem exchange (NEE) of CO<sub>2</sub> and FC<sub>org fert</sub> is the C input through manure application. Carbon input from animal excretion was not included in the budget as it was assumed to be recycled C from plant and soil uptake. FC<sub>animal</sub> includes the C off-take through animal weight increase and wool production. As grazing cows were heifers in calf<u>and ewes milk was consumed by their lambs</u>, there was no C off-take through milk to be considered. Methane emissions from enteric fermentation by ruminants, animal excretion and manure application as well as CH<sub>4</sub> fluxes from the soil are included in FC<sub>CH4</sub>. FC<sub>leaching</sub> is the C lost through dissolved organic and inorganic C and dissolved CH<sub>4</sub> leaching and FC<sub>harvest</sub> represents the C lost from the system though plant biomass export from harvests (cut for silage). Carbon emissions from farm operations (i.e. tractor emissions) or off farm emissions (i.e. fertiliser manufacture) are not included in the C budget.

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Details of methods to quantify each N and C budget component, as listed in Eq. (1) and (2), are described under Sect. 2.4 to 2.11 and in the Appendix. Some budget components were measured throughout the 9 years presented, while others were only measured in some years or not at all. Missing data were derived from the literature, models or averages from available data from other years.

#### 790

The annual net GHG exchange (NGHGE) was calculated from annual NEE (FC<sub>CO2</sub>), CH<sub>4</sub> (FC<sub>CH4</sub>) and N<sub>2</sub>O (FN<sub>N2O</sub>) fluxes using global warming potentials (GWPs) at the 100-year time horizon (<u>1 for CO<sub>2</sub>, 298 for N<sub>2</sub>O and 25 for CH<sub>4</sub>, IPCC, 2013</u>):

795 NGHGE = 
$$(FC_{CO2}) + FC_{CH4} * k_{CH4} + FN_{N2O} * k_{N2O}$$

Where;

 $k_{CH4} = 9.09$ , since 1 kg CH<sub>4</sub>-C <u>corresponds to</u>= 9.09 kg CO<sub>2</sub>-C

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$$k_{N2O} = 127$$
, since 1 kg N<sub>2</sub>O-N corresponds to= 127 kg CO<sub>2</sub>-C

In addition the net annual greenhouse gas balance (NGHGB) was calculated by including the loss of C through animal meat and wool production, harvest off take, C leaching and input by organic fertiliser application:

$$NGHGB = NGHGE + FC_{org fert} + FC_{animal} + FC_{leach} + FC_{harvest}$$
(4)

# 2.4 Nitrogen and carbon import by fertiliser and manure (FN<sub>synt fert</sub> + FN<sub>org fert</sub>, + FC<sub>org fert</sub>)

- 810 Mineral fertiliser was applied by a spreader as either ammonium nitrate or urea. Data of application rates and N content were obtained from the farmer. Six month old cattle slurry was spread by a vacuum slurry tanker. Three samples from the slurry tank were taken at each application and analysed for ammoniacal nitrogen ( $NH_3$  and  $NH_4^+$ ), dry matter content, total N, total C, pH and nitrate. The total N and C import to the field by the slurry was calculated by the volume of the slurry applied and the N and
- 815 C analyses of the slurry.

(3)

# 2.5 Nitrogen and carbon export by harvest (FN<sub>harvest</sub>+ FC<sub>harvest</sub>)

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The farmer estimated a <u>forage</u> harvest of 15 t fresh weight (FW) ha<sup>-1</sup> y<sup>-1</sup> at the first cut and 10 t FW ha<sup>-1</sup> y<sup>-1</sup> at the second cut of a year, <u>based on the plant height at the field at the time of cutting and information from harvested plot experiments</u>. As there were two cuts in 2002 and one cut in 2003 the estimated harvest was 25 t FW ha<sup>-1</sup> y<sup>-1</sup> for 2002 and 10 t FW ha<sup>-1</sup> y<sup>-1</sup> for 2003. A subsample of harvested vegetation was collected and dried at 80°C for plant N and C analysis using a Carbo-Erba/400 automated N and C analyser.

# 825 2.6 Nitrogen and carbon export by meat and wool (FN<sub>animal</sub> + FC<sub>animal</sub>)

It was estimated by the farmer that heifers increased in weight by 0.8kg per day (starting weight of 450 kg). The ewe weight was assumed to be constant (60 kg). They were fed extra protein (standard cake concentrate) to reduce weight loss during lactation., whereas lambslambs were brought to the field at a weight of 5 kg and removed when they reached a weight of 45 kg. All animals were weight before they came onto the field at the beginning of the season and again at the end of the season. The total meat export, which includes bones, was calculated from the daily weight increase of heifers and lambs multiplied by the animal number per day. To calculate the N and C export from meat we assumed an N content of meat of 3.5 % and a C content of meat of 21 % (Flindt, 2002), a N content of bones of 7 % and a C content of bones of 20 % (Marchand, 1842), assuming a total bone content of 20 % for sheep (Lambe et al., 2002) and 14 % for heifers (Navajas et al., 2010) -(Flindt, 2002). Ewes were sheared annually in June, yielding an estimated 2.5 kg of wool per sheep. Wool N and C export was calculated from wool production multiplied by the average sheep number in June, assuming a N and C content of wool of 16.5 and 50 %, respectively (Roche-J., 1995).

## 840 2.7 Nitrogen and carbon leaching (FN<sub>leaching</sub> + FC<sub>leaching</sub>)

Two sets of ten glass suction cups (pore size  $<1 \mu$ m, ecoTech, Bonn, Germany) for soil water and four Teflon suction cups (ecoTech, Bonn, Germany) for soil gas collection were installed in August 2006. One set was located on a slope, another on a hollow. For the budget calculations we only used results

from the slope location as the hollow location was frequently water logged. Suction cups were installed 845 horizontally from a soil pit beneath the A horizon (30 cm depth) and at 90cm depth and connected to 2-1 glass bottles in an insulated aluminium box placed into the soil pit. Samples were collected every two to three weeks. To reduce microbial transformation in the sampling bottles, the leachate passed a filter with very fine pores, (the suction cup, pore width  $< 1.6 \mu m$ ), before it entered the sampling bottle and the bottles were placed in an insulated aluminium box that was placed in a soil pit in order to keep the bottles as cool as possible.\_For further details and description of dissolved organic and inorganic C 850 (DIC, DOC) and dissolved  $CH_4$  analysis see Kindler et al. (2011). Dissolved inorganic and organic N (DIN, DON) and total N (TN) concentrations in leachate water were analysed by colorimetric analysis (San<sup>++</sup>, Automated Wet Chemistry Analyzer - Continuous Flow Analyzer (CFA), Skalar, The Netherlands). Leachate C and N concentrations were measured from October 1<sup>st</sup> 2006 - March 30<sup>th</sup> 855 2008. Dissolved C and N were calculated by multiplying concentrations of DIC, DOC and dissolved  $CH_4$  or DIN and DON respectively, with leachate volume. The latter was derived from a soil water model based on balancing daily precipitation and evaporation considering the water holding capacity of the soil data (Kindler et al., 2011). This model did not allow the calculation of upward water fluxes with capillary rise from groundwater. We therefore only used the data for the upslope position for the calculation of leaching losses. The data of the hollow position were not used, because the soil was 860 frequently water logged and likely influenced by capillaries from shallow ground water and lateral flow of groundwater. For the remaining years N was simulated using the LandscapeDNDC model (Haas et al., 2013), with the model tested and validated with comprehensive measured data. LandscapeDNDC is a process based biogeochemical model with unifying functionalities of the agricultural-DNDC (e.g. Li et al., 1992; Li 2000) and the ForestDNDC model (e.g. Kesik et al., 2005; Stange et al., 2000), 865 particularly suitable for ecosystem N turnover and associated losses of N trace gases and nitrate leaching (Wolf et al., 2012; Chirinda et al., 2011; Kiese et al., 2011). For C leaching linear regression models describing the relationship between calculated C leaching fluxes and leachate volume for the measurement period (DOC; y = 0.0186x - 0.0695,  $R^2 = 0.8663$ , DIC; y = 0.021x - 0.0008,  $R^2 = 0.8056$ 870 and dissolved CH<sub>4</sub>: y = 0.0019x - 0.0135,  $R^2 = 0.7623$ ) were used to extrapolate to the remaining years.

#### 2.8 Gaseous N fluxes

2.8.1 N deposition (FN<sub>dep</sub>)

## 875 Wet N deposition

Wet N deposition was determined from daily samples collected by an automatic precipitation sampler (Eigenbrodt<sup>®</sup> precipitation collector 181/KS, Königsmoor, D) at Auchencorth Moss (3°14'35W, 55°47'34 N), 17 km south west of Easter Bush (Skiba et al., 2013, McKenzie et al., 2015). The precipitation collector was only open during rainfall and closed automatically when precipitation ceased. Precipitation samples were analysed for NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> by ion chromatography (Methrom AG, Switzerland). Typical detection limits were 0.5  $\mu$ M for NH<sub>4</sub><sup>+</sup> and 0.4  $\mu$ M for NO<sub>3</sub><sup>-</sup>. Annual inorganic N deposition at this site was then adjusted to annual rainfall amounts measured at Easter Bush. For years where no data were available (2002, 2003), an average mineral N concentration per mm rainfall for 2004 2009 was taken and adjusted to the annual rainfall amount at Easter Bush in 2002 and 2003.

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### Dry N deposition

Cumulative monthly concentrations of gaseous and aerosol N species (NH<sub>3</sub>, HNO<sub>3</sub>, particulate NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>) were collected from another field, about 300m distance from our study field, using a DELTA system (DEnuder for Long Term Atmospheric) (Sutton et al., 2001). The DELTA system comprised of a denuder filter sampling train, an air pump (providing a sampling flow rate of 0.2 0.4 L min<sup>-1</sup>) and a high sensitivity dry gas meter to record sampled volumes (Tang et al., 2009) set at 1.5 m height above ground. N dry deposition fluxes were calculated using the average flux from four different inferential models; the UK CBED scheme (Concentration Based Estimated Deposition technique) (Smith et al., 2000), the Dutch IDEM model (Integrated Deposition Model) (Bleeker, 2000);, (Erisman et al., 1994), the dry deposition module of the Environment Canada model CDRY (Zhang et al., 2001; Zhang et al., 2003) and the surface exchange scheme EMEP (Simpson et al., 2003; Tuovinen et al., 2009), as described in detail by Flechard et al. (2011).

2.8.2 N<sub>2</sub>O fluxes (FN<sub>N2O</sub>)

- 900 From June 2002 to July 2003  $N_2O$  fluxes were measured continuously by eddy covariance (EC) using an ultra-sonic anemometer coupled with a Tunable Diode Laser absorption spectrometer (TDL) at a frequency of 10 Hz. For details see Di Marco et al. (2004). The detection limit for the TDL was estimated to be 1 ppbV and the detection limit for a 30 min averaging period of the N<sub>2</sub>O flux measurement was estimated at 11 ng N<sub>2</sub>O N m<sup>2</sup> s<sup>4</sup>. Details for the gap filling method of the N<sub>2</sub>O-EC data are described in Jones et al., 2011. For details see . The mean flux footprint reflects the prevailing 905 wind direction from the SW and secondarily from the NE, with the bulk of the contribution coming from within 50 m. The EC measurements thus sample the flatter areas of the field. Standard corrections were applied in processing to rotate co-ordinates relative to the mean wind flow in each half hour period. In this way, the fluxes were measured relative to the plane where mean vertical wind speed is 910 zero, rather than assuming a horizontal ground surface From August 2006 to November 2010 December 2009 N<sub>2</sub>O fluxes were measured using manual closed static chambers (Clayton et al., 1994, Skiba et al., 2013). Four chambers (0.4 m diameter, 0.2 m height) were inserted into the soil to 0.03 0.07 m depth and were accessible for animals to graze and deposit exreta. Chambers were closed usually between 10:00 and 12:00 for 60 minutes with an aluminium lid fitted with a draft excluderSamples of 200 ml 915 were collected by syringe and injected into Tedlar bags at the beginning and the end of the closure time through a three way tap fitted into the lid. In the laboratory samples were transferred to glass vials Samples were analysed for N<sub>2</sub>O using a Hewlett Packard 5890 series II gas chromatograph (Agilent Technologies, Stockport, UK), fitted with an electron capture detector (detection limit:  $N_2O < 33$ ppbV), - Samples of 200 ml were collected by syringe and injected into Tedlar bags at the beginning and the end of the closure time through a three way tap fitted into the lid. In the laboratory samples were 920 transferred to glass yiels using a syringe fitted with a 3 way tap; yiels were flushed with the sample using two needles in order not to over pressurise the vials. Samples were then and analyzed for N2O using a Hewlett Packard 5890 series II gas chromatograph (Agilent Technologies, Stockport, UK), fitted with an electron capture detector (detection limit:  $N_2O < 33$  ppbV). Fluxes were calculated from 925 the change of gas concentration with time of closure, multiplied by the volume of enclosed space and divided by its surface. Linearity tests were performed in between measurements showing a linearity of up to 120 minutes with an average  $R^2 = 0.96$ . The minimal detectable flux was 12 ng N<sub>2</sub>O N m<sup>2</sup> s<sup>4</sup>.
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Fluxes were measured weekly and more frequently during fertilisation. Cumulative fluxes were calculated by gapfilling data for missing days using linear interpolation and summing up all gapfilled data over each <u>callendarcalendar</u> year. For the periods where no N<sub>2</sub>O fluxes were measured (January - May 2002, July 2003-March 2004, May 2004-July 2006) fluxes were simulated by LandscapeDNDC (Haas et al., 2013). <u>LandscapeDNDC was tested in detail with available data on plant growth, soil temperature, moisture, inorganic soil N concentration, NO and N<sub>2</sub>O, which resulted in general good agreement of simulations and measurements.</u>

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#### 2.8.3 NOx fluxes (FN<sub>NOx(soil)</sub>)

NOx fluxes from the soil were only measured for a short period (June 2009-August 2010). The NOx fluxes were measured using an autochamber system described in detail by Butterbach-Bahl et al. (1997). Measurements were made 4 times per day, every 6 hours for an 8 min period per chamber. Four Perspex chambers (0.5 m x 0.5 m x 0.15 m; total volume 0.0375 m<sup>3</sup>) were fastened onto shallow frames 940 and moved fortnightly to a second position to allow free grazing of the first chamber set. One control chamber was placed onto a Perspex surface to account for ozone/NOx reactions inside tubing and chamber. Measurements were made 4 times per day, every 6 hours for an 8 min period per chamber. An in house Labyiew program controlled chamber closure and activated a solenoid valve system to sample 945 from the 4 chambers in sequence, interlaced with sampling from the control chamber. PTFE tubing (25 m in length, ID x OD; 4.35 x 6.35 mm) connected chambers to the NOx (42i TL Trace Level NOx Analyzer, Thermo Scientific US) and ozone (Model 49i Ozone Analyzer, Thermo Scientific, US) analysers located inside the mains-powered field cabin. Fluxes were calculated from the difference between control (on Perspex) and sample chambers (on grass), the flowrate into the analysers (11 lpm) and the surface area of the frames  $(0.25 \text{ m}^2)$ . We used simulated data from Landscape DNDC for years 950 where no NOx fluxes were measured.

2.8.4 NH4-NH3 + NOx volatilisation (FNNH3/NOx (fert.,manure, animal))

The fraction of nitrogen that volatilises as  $NH_4$ - $NH_3$  and NOx from applied synthetic fertiliser or cattle slurry application and animal excretion was estimated to be 10% and 20% of total N applied,

respectively (IPCC, 2006b). The animal excretion amount was estimated in accordance with the IPCC Guidelines (IPCC, 2006a)<u>s</u>.-<u>for details, see Appendix.</u> The amount of N excretion (Nex) from animals depends on the total N intake (N<sub>intake</sub>) and total N retention (N<sub>retention</sub>) of the animal. N<sub>intake</sub> (amount of N consumed by the animal) depends on the gross energy (GE) intake (see section 2.10) and the crude protein content (*CP%*) of the feed. CP was calculated using the measured N content in the grass. Grass

Daily N exerctions were thus calculated as 0.0263 kg N animal<sup>4</sup> d<sup>4</sup> for ewes and varied between

0.0019 0.0106 kg N animal<sup>4</sup>-d<sup>-1</sup> for lambs and 0.096 0.1013 kg N animal<sup>-1</sup>-d<sup>-1</sup> for heifers, depending on

- 960 protein content (*CP%*) of the feed. <u>CP was calculated using the measured N content in the grass. Grass</u> <u>N content was measured monthly in most years, where data were missing we used an averaged value</u> <u>calculated over all years.</u>, assumed to be 15.6% (MAFF, 1990)... N<sub>retention</sub> represents the fraction of N intake retained by the animal as meat, milk or wool. For lactating ewes the milk production was estimated at 0.618 1 animal<sup>-1</sup>-d<sup>-1</sup> and the milk protein content (*Milk PR%*) at 5.3% (Atti et al., 2006).
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animal weight..

# 2.8.5 $N_2$ emission by total denitrification $(FN_{N2(denitr.)})$

970 Di-nitrogen  $(N_2)$  emissions resulting from total denitrification in the soil was not measured in our experiment. We therefore used the  $N_2$  emission rates from LandscapeDNDC simulations.

# 2.8.6 Biological N<sub>2</sub> fixation (FN N2 (biol. fixation))

The species composition was measured <u>once in 2002 and at monthly intervals in 2003</u> by the visual estimation method (Braun-Blanquet, 1964), where 50 quadrates of 0.25 m<sup>2</sup> were randomly thrown into the field. As the legume fraction (*Trifolium repens*) was smaller than 0.5% at each measuring point we assumed the nitrogen fixation through plants to be zero.

## 2.9 Exchange of CO<sub>2</sub> (FC<sub>CO2</sub>)

980 NEE was measured <u>continuously from 1.January 2002 till 31.Decmber 2010</u> by an eddy covariance system consisting of a fast response 3D ultrasonic anemometer (Metek USA-1, Metek GmbH, Elsmhorn, Germany) and a fast closed path CO<sub>2</sub>-H<sub>2</sub>O analyser (LI-COR 7000 infra-red gas analyzer (IRGA), LI-COR, Lincoln, NE, USA). Wind velocity components were measured at 2.5m above ground

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and data were logged at 20 Hz by a PC running a custom LabView data acquisition program. Air was sampled 0.2 m below the sensor head of the anemometer using 6.3 mm (1/4 in. OD) Dekabon tubing. The IRGA was located in a field laboratory ca. 10 m from the mast. Lag times between wind data and trace gas concentrations were synchronised and taken into account in the offline data processing (Helfter et al., 2014). Quality control of the eddy covariance data followed the procedure proposed by Foken and Wichura (1996). Data were filtered out if the friction velocity (u<sub>2</sub>) was smaller than 0.2 m s<sup>-1</sup> 990 (insufficient turbulence), CO<sub>2</sub> concentrations fell outside a plausible interval (330 450 ppm), CO<sub>2</sub> fluxes fell outside the range 50 to 50 umol m<sup>-2</sup> s<sup>-1</sup> and latent (LE) and sensible (H) heat fluxes fell outside the range -250 to 800 W m<sup>-2</sup>. Missing NEE data were gap-filled using the online tool developed Max Planck Institute for Biogeochemistry, Jena, Germany<sup>4</sup> (<sup>4</sup> http://www.bge--the jena.mpg.de/~MDIwork/eddyproc/upload.php, Reichstein et al., 2005) NEE is the arithmetic sum of the gross primary production (GPP) and total ecosystem respiration (TER). Flux partitioning of measured 995 NEE into GPP and TER was calculated by the same online tool used for gapfilling. In this flux partitioning approach, daytime TER is obtained by extrapolation of a night time parameterisation of NEE on air temperature and GPP is the difference between ecosystem respiration and NEE. Contrarily to unmanaged ecosystem, TER at our site also includes the respiratory loss of  $CO_2$  by grazing animals. Net primary production (NPP), which represents the annual plant growth (difference between GPP and autotrophic respiration) was calculated as 50% of GPP (Amthor 2000, Zhang et al., 2009 Waring et al.,

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<del>1998</del>).

### 2.10 Methane fluxes (FC<sub>CH4</sub>)

1005 Methane fluxes from the soil were measured with closed static chambers simultaneously with the  $N_2O$ measurements (see Sect. 2.8.2). The same GC was fitted with a flame injection detector (detection limit:  $CH_4 < 70$  ppbV). The minimal detectable flux was 17 ng  $CH_4$ -C m<sup>-2</sup> s<sup>-1</sup>. Fluxes were measured weekly and more frequently at fertiliser events. As measured soil CH<sub>4</sub> fluxes were close to zero and did not vary significantly between months, we calculated CH<sub>4</sub> for months where no CH<sub>4</sub> fluxes were measured

<sup>&</sup>lt;sup>4</sup> http://www.bgc.iena.mpg.de/~MDIwork/eddvproc/upload.php

1010 (January-May 2002, July 2003-March 2004, May 2004-July 2006), as an average monthly cumulative flux from other years.

Methane emissions from grazing animals, i.e. animal excretion and enteric fermentation, were estimated following the IPCC Tier 2 methodology (IPCC, 2006a: Stewart et al., 2009). For details, see Appendix. For animal excretion only solid volatile production was considered, as urine has no effect on 1015 CH₄-emissions (Jarvis et al., 1995). The calculation of CH₄ emissions from excretion was based on the amount of volatile solids (VS) excreted, the maximum CH4 producing capacity (B<sub>e</sub>) of the manure and the CH<sub>4</sub> conversion factor (MCF), which is specific to the storage type (pasture, in our study). The amount of VS exercted depended largely on the GE intake of the animal. The GE intake (based on digestible energy of feed intake, milk production, pregnancy, current weight, mature weight, rate of weight gain and IPCC constants) in our study was estimated at 19.5 MJ animal<sup>4</sup> d<sup>-1</sup> for ewes, while it 1020 ranged from 7.9 to 14.9 MJ animal<sup>+</sup> d<sup>+</sup> for lambs and from 160.9 to 169.7 MJ animal<sup>+</sup> d<sup>+</sup> for heifers. Emission factors for excretion were calculated as 0.198 kg  $CH_4$  head  $+ v^+$  for ewes and varied between 1.64 1.73 kg CH<sub>4</sub> head<sup>-1</sup> v<sup>-1</sup> for heifers and 0.081 0.152 kg CH<sub>4</sub> head<sup>-1</sup> v<sup>-1</sup> for lambs. Methane emission factors for enteric fermentation were calculated from GE intake and CH4 conversion factors (Ym). Depending on animal type and live weight, emission factors were 7.6 kg CH<sub>4</sub> head<sup>4</sup> v<sup>+</sup> for ewes and 1025 varied between 60.1 63.8 kg CH<sub>4</sub> head<sup>4</sup> v<sup>1-</sup>for heifers and 2.0 4.0 kg CH<sub>4</sub> head<sup>4</sup> v<sup>1-</sup>for lambs. Annual emissions from excretion and enteric fermentation were calculated from daily CH4 emissions per animal multiplied by the animal number.

Methane emissions from slurry applications were assumed to be small. As no chamber 1030 measurements were conducted at the time of slurry spreading, the emissions were estimated as 0.07 % of the applied assuming that emissions were comparable to those in a related study (Jones et al., 2006), where  $CH_4$  was measured from chambers after slurry application on a nearby field in 2002 and 2003.

### 2.11 VOC

1035 Fluxes of non-methane volatile organic compounds (VOC) were not measured. We assumed similar VOC emissions to those reported by Davison et al. (2008) for an intensively managed grassland in Switzerland, where the daily average flux of methanol, acetaldehyde and acetone over 3 days after

cutting were 21.1, 5.1. and 2.6 nmol m<sup>-2</sup> s<sup>-1</sup>, respectively. Based on those values, annual VOC emissions from our field were estimated to be in the order of 0.03% of the annual C offtake in harvest and 0.08 % of annual C off-take by grazing animals. We therefore assumed VOC emissions to be negligible and did not account for them in the C balance.

#### 2.12 Soil N and C measurements

Total N and C content of the soil were measured in May 2004 and May 2011. One hundred soil cores 1045 with an inner diameter of 8.7 (2004) and 8.3 cm (2009, both corers from Eijkelkamp Agrisearch Equipment BV, Giesbeek, The Netherlands) were collected along a regular grid with a distance of 10 m between sampling points on both occasions. The soil sampling grid covered the main footprint area of the site, not the entire field. The grid was positioned independently from slope and potentially preferred areas to avoid biased sampling. For the resampling in 2011 the same grid was used, but the transect was 1050 chosen two meters further to the NW in order not to meet the same place we already sampled and disturbed before. Cores were separated into layers of 0-5, 5-10, 10-20, 20-30, 30-40, 40-50 and 50-60 cm. Coarse stones of a diameter > 4 mm and roots of a diameter > 1mm were removed from the samples prior to drying at 40 °C. Stone and root samples were air-dried separately. Then, soil samples were sieved to < 2 mm. Particles > 2 mm were combined with the coarse stones. Dry weights of roots and 1055 combined stone fractions were determined. Total N and C concentrations in < 2 mm soil separates were determined using dry combustion (VarioMax, Elementar Analysensysteme GmbH, Hanau, Germany). As the site contains no carbonatesinorganic C, total C was assumed to equals organic C. As bulk density varies spatially and over time (e.g. through compaction by livestock), the soil N and C content per unit ground area to a fixed depth will also change, without any change in the mass fraction of N and C in dry 1060 soil. Therefore, total N and C stocks were calculated on an equivalent soil mass (ESM) basis, so that comparisons between years were valid (see Gifford and Roderick, 2003, Wendt and Hauser, 2013). A cubic polynomial was fitted to the data, to predict cumulative N and C with cumulative soil mass in the profile. A soil mass of 800 kg m<sup>-2</sup> was used (Table 7), which corresponds to approximately 60-cm

depth, which was the depth of the corer. Uncertainty in the estimates of stock change was based on the

1065 prediction intervals in the cubic polynomial at a soil mass of 800 kg m<sup>-2</sup>.
### 2.13 Ancillary measurements

Soil temperature and volumetric soil moisture were continuously recorded at four depths (3.5, 7.5, 15 and 30 cm) by temperature probes (temperature probe 107, Campbell Scientific, Loughborough, UK) and TDR probes (TDR 100, Campbell Scientific, Loughborough, UK), respectively, the latter installed in June 2002. Rain was measured by a tipping bucket rain gauge, while air temperature and relative humidity were measured by an integrated humidity and temperature transmitter (HUMITTER<sup>®</sup>, Vaisala Ltd, Suffolk, UK).

# 1075 2.14 Statistical and uncertainty analysis

Random error was determined as  $2\sigma$ -standard error (95% confidence) of the overall mean according to Gaussian statistics. The confidence intervals for group means were used to establish whether or not differences were significantly different from zero. Analyses of variance (ANOVA) were used to test if values were significantly different from zero (p<0.05). Linear correlations between C and N inputs and outputs were calculated by calendar year. For systematic errors the uncertainty range of measurements as well as of parameterisations and literature based estimates was estimated according to expert judgment. To calculate the combined effect of systematic uncertainties of each flux component on the C and N budget simple Gaussian error propagation rules were used, details are, provided in Table S1 (Supplementary material). Confidence intervals are given at the 95% confidence level.

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A more detailed description of the methods are provided in the Appendix.

# 3. Results

# 3.1 Climate and management

The meteorological conditions exhibited substantial inter-annual variability in the study period 2002-2010 (Table 2 and Fig. 2). Annual rainfall ranged from 575 mm to 1238 mm with highest monthly rainfalls at of 280 mm month<sup>-1</sup> in September 2002. Lowest annual reported rainfall was in 2010; this low value was caused by a gap in data from January-March, due to snowfall. Average annual air

temperature ranged from 8.3 to 9.6 °C with highest daily air temperatures of 30.4 °C in August 2005 and
lowest in December 2010 at -10.3 °C. Highest average monthly air temperatures were measured in July
2006 at 17°C and lowest monthly average air temperatures at 2°C in November 2009. In 2003 the
highest average annual temperature (9.6° C) and lowest annual rainfall (680 mm) were measured with a
correspondingly low annual soil water content of 31 %. The duration of the growing season was defined
per calendar year as the period bounded by the first and last 5 consecutive days with mean daily air
temperature ≥ 5 °C. The length of the growing season (LGS) varied between 151 days (2006) and 242

Livestock stocking density exhibited both intra- and inter-annual variability. The average annual stocking density was lowest in 2002 and 2003 at 0.27 LSU ha<sup>-1</sup> y<sup>-1</sup> and 0.54 LSU ha<sup>-1</sup> y<sup>-1</sup>, respectively (Table 1), which were the years where the grass was cut for silage and no lambs were present in the field. In 2007, 2008, 2009 and 2010 no heifers were present in the field. Highest annual average stocking density occurred in 2004 and 2007 at 0.99 LSU ha<sup>-1</sup> y<sup>-1</sup> and 0.91 LSU ha<sup>-1</sup> y<sup>-1</sup>, respectively. Maximum monthly stocking density occurred in September 2006 at 13.8 LSU ha<sup>-1</sup>, while interim periods with no grazing at all were observed in all years (Fig. 1a). Mineral N fertiliser was applied split into 3 to 5 applications per year, ranging from 2.5 to 9.6 g N m<sup>-2</sup> application<sup>-1</sup> (Fig. 1b). Organic manure was applied in 2004 and 2005 as cattle slurry, spread at a rate of 6.9 and 15.8 g N m<sup>-2</sup> application<sup>-1</sup>, respectively (Fig. 1b and c). The grass was only cut in 2002 and 2003. Harvested biomass in 2002 and 2003 ranged from

C removal from the field ranging from 113.1 to 169.5 g C  $m^{-2}$  cut<sup>-1</sup> (Fig. 1c).

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#### **3.2 Uncertainty analysis**

days (2009) (Table 2).

Systematic uncertainties for each component of the C and N budget are shown in Table 3. Uncertainty values were estimated according to expert judgment. The systematic uncertainty of the N input from mineral fertiliser was assumed to be minimal (1 %), while the systematic uncertainty of the N and C spread by the manure was assumed to be 17 % on average for the C and N analysis. Together with an uncertainty of 10 % of the volume spread, this resulted in a total uncertainly of 20 %. The uncertainty of

2.60 to 3.75 t DW ha<sup>-1</sup> cut<sup>-1</sup> which resulted in an N off-take ranging from 1.7 to 4.7 g N m<sup>-2</sup> cut<sup>-1</sup> and a

the C and N analysis for harvest were 4 and 12 %, respectively. We assumed an error of 10% in the farmer's estimate of the harvest amount, which resulted in a total uncertainty of 16 % for N and 11 % for C off take. We attributed a systematic uncertainty of 30 % to the modelled data for C and N leaching. The systematic uncertainty of the meat and wool consists of the estimated uncertainty in the 1125 animal weight, animal numbers and literature values for wool and meat C and N contents. We assign an uncertainty for animal weight of 10 %, for animal numbers of 5 % and for literature values of wool and meat C and N content of 3 %, resulting in a total uncertainty of 12 %. The uncertainty of wet N deposition was 30 % resulting from the error of sample analysis and a potential bias from dry deposition on the funnel. The uncertainty of dry N deposition consisted of an error of 7 % for the analysis of 1130 DELTA samples and an 80% uncertainty of the variation of the output from the four models, which resulted in a total uncertainty of 80%. The systematic uncertainty attributed to the annual cumulative N<sub>2</sub>O fluxes was 30 %, due to the uncertainty of gapfilling. The uncertainty attributed to the modelled NOx fluxes is 30 %. The uncertainty attributed to the NH<sub>4</sub> and NOx volatilisation was 30 % from 1135 applied synthetic fertiliser and 50 % from cattle slurry application and animal excretion. The uncertainty attributed to the N<sub>2</sub> fluxes was 30 %. The total uncertainty for NEE values was estimated to be 80 g C m<sup>-2</sup>-v<sup>+</sup>-(Levv et al., submitted). The systematic uncertainty of annual cumulative soil CH<sub>4</sub>-fluxes was very high at 160 %, due to the uncertainty of gap filling and as values were close to zero. The uncertainty of CH4 from enteric fermentation and animal excretion estimates were each assumed to be 1140 20%, according to IPCC (2006a). The uncertainty of CH₄ fluxes from organic manure application was estimated at 120 %.

#### 3 <u>32</u>. N budget

In our grassland system the N balance is the difference between the N input through fertiliser and atmospheric deposition and the N output through harvest, animal export, leaching and gaseous emissions. The total resulting balance over the nine years, derived from flux calculations and estimations, showed that N was stored at an average rate of  $-7.216.6 \pm 4.46$  g N m<sup>-2</sup> y<sup>-1</sup> (p<0.05). From 2003 to 2010, N was stored at a rate of -1.93.1 to -17.29 g N m<sup>-2</sup> y<sup>-1</sup>, whilst in 2002 N was lost at a rate of 6.43 g N m<sup>-2</sup> y<sup>-1</sup> (Table 4). The major N input consisted of inorganic fertiliser, ranging from -11 to -

- 25.9 g N m<sup>-2</sup> y<sup>-1</sup>, averaging at -19.42 g N m<sup>-2</sup> y<sup>-1</sup>, while N deposition represented only between 1.9 and 1150 5.9% of the total N input. During the years where N was stored, a significant positive correlation between total N input from fertiliser and N storage was observed ( $R^2 = 0.55$ ). Largest losses resulted from leaching at an average rate of 5.34  $\pm$  3.4 g N m<sup>-2</sup> y<sup>-1</sup> and were estimated to be highest in 2002 at 14.9 g N m<sup>-2</sup> y<sup>-1</sup> and lowest in 2003 at 0.09 g N m<sup>2</sup> y<sup>-1</sup>. We found a strong correlation between N leaching and rainfall ( $\mathbb{R}^2 = 0.82$ ), if values from 2004 were excluded, a weak correlation between 1155 livestock density and N leaching if the years 2002 and 2004 were excluded (R<sup>2</sup>=0.47), while no correlation with total N input could be found. The total N off take through meat and wool ranged from 0.15-3.12 g N m<sup>-2</sup> y<sup>-1</sup>, while the total annual N offtake from harvest was 5.0 g N m<sup>-2</sup> y<sup>-1</sup> in 2002 and 4.68 g N  $m^{-2}y^{-1}$  in 2003. Amongst gaseous exchanges, highest losses were estimated from N<sub>2</sub> emissions, averaging at 2.76 g N m<sup>-2</sup> y<sup>-1</sup> with maximum losses of 4.12 g N m<sup>-2</sup>y<sup>-1</sup> in 2009, although in 2004 and 1160 2005 losses from NOx/NH<sub>3</sub> volatilisation from excretion and organic fertilisation exceeded losses from  $N_2$  emissions. Losses through NOx from the soil were always less than 1% of the total N exchange (0.2 g N m<sup>-2</sup> y<sup>-1</sup>). Nitrous oxide emissions ranged from 0.11 to 1.27 g N m<sup>-2</sup> y<sup>-1</sup>, representing 1.3-8.4 % of the total N export. Annual N<sub>2</sub>O emissions showed no correlation with precipitation, livestock density or total N input. However, there was a positive correlation with rainfall if 2004 and 2007 data were 1165 excluded ( $R^2$ =0.78); with livestock density if the years 2002 and 2004 were excluded ( $r^2$ =0.70); and with total N input if the years 2002, 2003 and 2010 were excluded ( $R^2 = 0.76$ ). N<sub>2</sub>O emission factors (percentage of N lost from total N inputs by mineral and organic fertiliser), ranged between 0.6 and 7.5 % (Table 6).
- To investigate the influence of different managements on the N and C budget, we separated experimental years into harvested and grazed (2002 and 2003) and grazed only years (2004-2010 Fig. 23 and 34). During the harvested years, the main loss of N from the system occurred through leaching (39.2% of total N inputs), followed by the export through harvest (25.2%), while the export from animals (meat and wool) accounted for less than 2 % of total losses (Fig. 23a). The main loss to the atmosphere resulted from total denitrification (N<sub>2</sub>; 15.4%), followed by NOx/NH<sub>3</sub> volatilisation from inorganic N fertiliser applications (9.5%), while N<sub>2</sub>O emissions accounted for 3.3%, NOx/NH<sub>3</sub> volatilisation from excretion for 2.7% and NOx from soil for less than 1%. The residual 2% represents

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the N storage in the soil and the uncertainty of the budget. When grazed-only years were considered (Fig. 23b), the residual part was the highest at 38.6%. Losses through leaching (19.9%) and  $N_2$  (11.4%) were lower in grazed years compared to harvested years, while the export through grazing animals were considerably higher at 15.8% -(sum of N loss through meat, wool and NOx/NH<sub>3</sub> volatilisation from excretion). An additional loss occurred in grazed years through the volatilisation of NOx/NH<sub>3</sub> from organic fertiliser applications in 2004 and 2005 (3%). Losses through N<sub>2</sub>O and NO<sub>x</sub>/NH<sub>3</sub> from inorganic fertiliser were comparable to harvested years at 2.5% and 8.3%, respectively.

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Cumulative soil N stocks were derived from soil core measurements taken in May 2004 and May 2011. In 2004 N stocks were 840.86 ( $\pm$ 11.89) g N m<sup>-2</sup> and in 2011 they were 870.02 ( $\pm$ 14.14) g N  $m^{-2}$ . Nitrogen storage over the 7 years was calculated from the cumulative equivalent soil mass (ESM) for the soil mass increment of 800 kg m<sup>-2</sup>, which corresponds to approximate 60 cm depth. The estimated N storage over the 7 years was  $-4.51 \pm 2.64$  g N m<sup>-2</sup> y<sup>-1</sup> (Table 7) and was a significant N accumulation to the soil (p < 0.01). The estimated N storage derived from flux calculations between 2004 and 2010, however was  $-\frac{8.449.20}{2} \pm 4.210$  g N m<sup>-2</sup> y<sup>-1</sup>, which is <u>almost 2</u> times more than that calculated by sequential soil analysis, however, values were not significantly different from each other.

# 3.43. C budget

Annual C inputs through photosynthesis (GPP) varied between -982.1 and -2162.9 g C m<sup>-2</sup>, and losses 1195 through autotrophic and heterotrophic respiration (TER) varied between 972.1 and 2183.2 g C m<sup>-2</sup>, both considerably larger than any other C fluxes (Table 5). If only the NEE was considered (difference between GPP and TER), the grassland acted as a sink for CO<sub>2</sub> at an average of  $218 \pm 155$  g C g C m<sup>-2</sup> v <sup>1</sup>, and the CO<sub>2</sub> uptake was significantly different from zero (p < 0.05). The sink strength ranged from -10 g C m<sup>-2</sup> y<sup>-1</sup> (2006) to -606 g C m<sup>-2</sup> y<sup>-1</sup> (2009), only in 2004, the grassland was a small source of CO<sub>2</sub> 1200 (72 g C m<sup>-2</sup> y<sup>-1</sup>). Taking into account all C inputs and outputs (NBP), C was sequestered on average at  $1634 \pm 140$  g C m<sup>-2</sup> y<sup>-1</sup> over the nine years, although the storage was not significantly different from zero (p<0.05). In 2004 and 2006 C was lost from the ecosystem. The major C import resulted from NEE in all years apart from 2005, when the C input from manure application was larger. Highest C export occurred from harvest in 2002 and 2003 (270.6 and 169.5 g C m<sup>-2</sup>y<sup>-1</sup> respectively), while second largest

export in 2002 and 2003 and largest exports in other years was leaching (6.8 to 25.1 g C m<sup>-2</sup>v<sup>-1</sup>). The measured C leaching value for 2007 (15.4 g C  $m^{-2}y^{-1}$ , table 5) differs from the leaching value published for Easter Bush by Kindler et al. (2011), as we only used values of one of the two measured sites in this manuscript (slope, not hollow, as the hollow location was frequently water logged). The third largest C loss consisted of C export from meat in 2004-2010, ranging from 6.4-15.8 g C m<sup>-2</sup> y<sup>-1</sup>. In 2002 and 1210 2003, when no lambs were present in the field, C export from meat was exceeded by CH<sub>4</sub> losses from enteric fermentation. Carbon export from wool ranged from 0.5 to 2.1 g C m<sup>-2</sup> y<sup>-1</sup>. CH<sub>4</sub> emissions from organic fertilisation, soil processes and animal excretion were always less than 1 % of the total C losses. CH<sub>4</sub> losses from enteric fermentation ranged from 1.5 to 5.7 g C m<sup>-2</sup> v<sup>-1</sup>, corresponding to 0.5-22.5 % of 1215 all C losses from the ecosystem. The annual carbon balance (NBP) was dominated by the NEE. A high livestock density tended to reduce the net sink strength. A significant negative correlation of NEE as well as NBP with stocking density could be seen  $(R^2=0.47)77$  and  $R^2=0.83$ , respectively), if the years with cuts (2002 and 2003) were excluded. The NBP correlated positively with rainfall ( $R^2$ =0.48) and whereas the correlation improved if the dry year 2003 was excluded ( $R^2 = 0.78$ ). T there was only a weak correlation between NEE and rainfall ( $R^2=0.38$ ) for all years,  $R^2=0.47$  without the year 2003). 1220

The nNet primary production (NPP) in years when grass was harvested and grazed (2002 and 2003) and grazed only (2004–2010) are presented in Figure 43. In both management types most C was lost through ecosystem respiration, (67% and 71% of NPP, respectively). Harvest export represented 21% of NPP. Leaching accounted for 1.5% of NPP during harvested years and 2.2% in grazed only years. Animal export (meat and wool) consisted of 1.5% of NPP in grazed only years and was less than 0.2% of NPP in grazed and harvested years. The sum of all CH<sub>4</sub> emissions (from organic fertilisation, excretion, enteric fermentation and soil) was less than 1% of the NPP. The residual part, which includes the C storage in the soil as well as the uncertainty of the budget, was estimated at 10% and 24% of NPP in harvested and grazed or grazed years, respectively.

1230 The C content for the cumulative soil mass increment 0-800 kg m<sup>-2</sup> (~ 0-60 cm) was lower in 2011 (12026.05  $\pm$  190.19 g C m<sup>-2</sup>) compared to 2004 (11824.87  $\pm$  187.84 g C m<sup>-2</sup>), resulting in a C loss of 29.08  $\pm$  38.19 g C m<sup>-2</sup> (Table 7). In comparison, based on flux calculations C was stored at 180  $\pm$ 180

g C  $m^{-2} y^{-1}$  over the 7 years. However, neither C loss calculated by sequential soil analysis, nor C storage estimated from flux calculations were significantly different from zero.

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# 3.54. Greenhouse gas budget

In order to calculate the global warming potential for the Easter Bush grassland fluxes of the greenhouse gases CO<sub>2</sub>, N<sub>2</sub>O and CH<sub>4</sub> were expressed in CO<sub>2</sub> equivalents considering the different global warming potentials for each gas at the 100 year time horizon (1 for CO<sub>2</sub>, 298 for N<sub>2</sub>O and 25 for CH4, IPCC, 2013). Average greenhouse gas fluxes, net GHG exchange (NGHGE) and attributed net 1240 GHG balance (NGHGB) for 2002-2010 are shown in Figure 45. The CO<sub>2</sub> storage from the NEE provided the largest term in the annual GHG budget. Carbon dioxide (NEE) was sequestered over the 9 years at a rate of  $-799 \pm 567$  g CO<sub>2</sub> m<sup>-2</sup> y<sup>-1</sup>. This storage was significantly different from zero (p < 0.05). On average, the annual net GHG exchange (NGHGE) was highly correlated with annual NEE ( $R^2$ =0.96). On average the grassland was a source of the GHGs CH<sub>4</sub> and N<sub>2</sub>O at a rate of 148 ± 30 and 1245  $285 \pm 131$  g CO<sub>2</sub> m<sup>-2</sup> y<sup>-1</sup>, respectively, both being significantly different from zero (p < 0.001 and p < 0.01, respectively). Nitrous oxide losses ranged from 52 g CO<sub>2</sub> eq.  $m^{-2}$  y<sup>-1</sup> (2004) to 588 g CO<sub>2</sub> eq.  $m^{-2}$ v<sup>-1</sup> (2007) (data for each year not shown). Methane from soil processes, manure input as well as animal excretion, accounted for less than 5% of total CH<sub>4</sub> emissions. Methane emissions from enteric fermentation ranged from 53 g CO<sub>2</sub> eq. m<sup>-2</sup> y<sup>-1</sup> (2002) to 199 g CO<sub>2</sub> eq. m<sup>-2</sup> y<sup>-1</sup> (2004). Annual total CH<sub>4</sub> 1250 emissions correlated positively with annual live stock density ( $R^2=0.99$ ). The CH<sub>4</sub> emissions, which were predominately (> 97%) of ruminant origin weakened the sink strength of NEE by 18 %. If both CH<sub>4</sub> and N<sub>2</sub>O were considered the total trade-off of NEE was a substantial 54% and increased to a total of 67 %, if only grazed years were considered. On average the grassland represented a GHG sink of -366  $\pm$  601 g CO<sub>2</sub> m<sup>-2</sup> y<sup>-1</sup>, if only NEE, CH<sub>4</sub> and N<sub>2</sub>O were included (NGHGE). If all C components 1255 (FCorg.fert, FCanimal, FCleaching, FCharvest) are included, the sink strength of the grassland decreased to -182  $\pm$  560 g CO<sub>2</sub> m<sup>-2</sup> y<sup>-1</sup> (NGHGB). This represents a weakening of the sink strength of the NGHGE by 50 %, mainly due to the export of harvest. However, it has to be noted that in harvested years the return of the manure, resulting from the grass fed to livestock off -site, would reduce the GHG balance. If only 1260 grazed years were considered the sink strength increased slightly by 5.4 %, due to the C input from manure in 2004 and 2005. Both, NGHGE and NGHGB were not significantly different from zero.

### 4. Discussion

# 4.1. Nitrogen balance

- 1265 The main N inputs in our study were from inorganic and organic fertilizer additions. The amount of N added through fertilizer was governed determined by national recommendations levels (SACRUC, 2013) - and lies within the range of N applied in other European studies with similar management (e.g. Laws et al., 2000; Allard et al., 2007; Ammann et al., 2009). Nitrogen added through the excretion from grazing animals was not considered an N input as this represents an internal 1270 redistribution of N within the system. Atmospheric N deposition (wet and dry) accounted only for a small fraction of the total N input toon our managed grassland. This is in contrast to semi natural systems, where atmospheric N deposition and biological fixation represents the main N input (Pheonix et al., 2006, Bleeker et al., 2011). As oour experimental field was sown as a grass mixture (without clover) has been under grazing/cutting management for more than 20 years with regular N inputs from 1275 mineral fertilizers, manure and animal excretion. As biological N<sub>2</sub> fixation by legumes is inhibited by soil mineral N (Streeter, 1988), the legume fraction was less than 1% and biological N<sub>2</sub> fixation therefore a negligible source of N-in our system. Atmospheric N-deposition (wet and dry) accounted only for a small fraction of the total N input on our managed grassland. This is in contrast to semi natural systems, where atmospheric N deposition represents the main N input (Pheonix et al., 2006, 1280 Blecker et al., 2011). The main N inputs in our study were from inorganic and organic fertilizer additions. The amount of N-added through fertilizer was governed by recommended maximum levels (SRUC 2013) and lies within the range of N applied in other European studies with similar management (e.g. Laws et al., 2000; Allard et al., 2007; Ammann et al., 2009). Nitrogen added through exerction from grazing animals was not considered an N input as this represents an internal redistribution of N within the system. 1285
- Field Code Changed Field Code Changed

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4.1.1 N use efficiency

The data obtained from our budget were used to calculate the Nitrogen Use Efficiency (NEU) expressed as the ratio between N in crop and animal products (in this case either the crop harvest or the 1290 sum of meat, wool and milk) to the total N inputs to the system (fertilizer, imported manure). The ratio between N input and percentage of N uptake into the crop or animal products (meat, wool and milk) is defined as the N use efficiency (NUE). In our study a substantial amount of N was removed by harvest, with an The NUE of herbage in cut years (2002 and 2003) of 25% (Figure 3a2a)). This seems low compared to reported N efficiencies of 55-80% in harvested herbage from managed temperate 1295 grasslands (Ball and Ryden 1984; Ammann et al., 2009). It has been shown that the NUE in crops is significantly higher compared to the NUE in animal production (Galloway and Cowling, 2002). The inclusion of grazing ruminants introduces an additional trophic level alterings the NUE of herbage as the nitrogen in the grazed grass is consumed is and converted to meat, milk, wool, or is excreted. The lower NUE in the grass production in our study is therefore partly due to grazing. Furthermore, it has 1300 been shown that the proportion partitioned to plant uptake decreases as the total amount of soil inorganic N increases (Scholefield et al., 1991), which is a further explanation for a low NUE in herbage in our high N input system. There are different mitigation options to reduce N losses and thus increase NUE. The introduction of clover into grassland has been shown to reduce the requirement of N input from fertilisation, thereby resulting in the same yield (Herrmann et al., 2001; Ledgard, 2001). 1305 Adherences with fertiliser recommendation systems and avoidance of over fertilisation is also likely to increase the efficiency of N use without compromising productivity (Rees et al., 2013). The use of nitrification inhibitors applied onto grassland has been shown to result in a reduction of N<sub>2</sub>O emissions (McTaggart et al., 1997). Furthermore, a novel approach to reduce N losses from sheep urine, by infusing N process inhibitors into the gastrointestinal tract of the animals, has been demonstrated by 1310 Ledgard et al. (2008), however, the evidence for this as a mitigation option is still limited, and could face legal and ethical challenges. The NUE in crops is significantly higher compared to the NUE in animal production. The NUE of animal products on our grassland system ranged from 65 to 2118% in grazed only years (2004-2010),

with an average of 9.910.6 %. This is in agreement with the NUE reported for sheep of 6.2 % by Van der Hoek (1998) and beef production systems, which reported N efficiencies range from 6 to 12%

(Whitehead et al., 1986; Tyson et al., 1992) and 5-20% (Ball and Ryden, 1984). Approximately 85% of total harvested N is used to feed livestock (Sutton 2011).erops produced are used for animal feed, which is significantly less efficient than if the crops were used to feed humans directly. A measure to reduce N pollution could therefore be the reduction of meat consumption or a larger fraction of meat produced from grassland only- (Smith et al. 2013).

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# 4.1.2 N loss to the environment:

Nitrogen was lost from our grassland to the environment through different pathways. Nitrogen leaches from grassland soils in the form of nitrate  $(NO_3)$ , ammonium  $(NH_4^+)$  and dissolved organic N 1325 (DON). Whereas  $NO_3^{-}$  is highly mobile in water and can be easily leached into groundwater.  $NH_4^{+}$  is less prone to leaching as it is mostly bound to soil particles (Brady and Weil, 2002). Leaching depends on the water holding capacity of the soil, amount of rainfall, water use by plants and soil nutrient content, which are in turn influenced by management. Leaching occurs predominantly from late autumn to early spring when precipitation often exceeds evapotranspiration (Askegaard et al., 2005). In our field leaching losses varied widely over the years. This variation can mainly be explained by differences in 1330 precipitation. Overall, leaching from our field (5.3  $\pm$  3.4 g N m<sup>-2</sup> v<sup>-1</sup>) was comparable to values measured at intensively grazed pastures in Ireland (1.8-6.4 g N m<sup>-2</sup> y<sup>-1</sup>, Watson et al., 2007) and England (3.8-13.3 g N m<sup>-2</sup> y<sup>-1</sup>, Scholefield et al., 1993) or croplands (e.g. Bechmann et al., 1998), max. leaching losses of 10.4 g N m<sup>-2</sup> y<sup>-1</sup>). However, leaching from our study was high compared to the Swiss NitroEurope site, where a maximum loss of 0.33.5 kg N mha<sup>-24</sup> y<sup>-1</sup> was estimated from an ungrazed 1335 grass/clover sward, despite comparable annual rainfall and N inputs comparable to our site (Ammann et al., 2009). This difference can be explained by the different plant cover and management. It has been shown that clover introduction can reduce leaching (Owens et al., 1994), whereas grazing tends to increase leaching (Cuttle and Scholefield, 1995). Grazed grasslands tend to have higher N leaching rates 1340 than cut grasslands since highly concentrated N depositedas the N added as fertiliser is not removed by harvest, but returned to the soil in urine is inefficiently recovered-and dung from consumed by herbage and, prone to leaching. The uneven distribution of excreted organic N further enhances leaching due to the formation of N hotspots, which has been observed at outdoor pig farms (e.g. (Eriksen, 2001). Ryden

et al., (1984a) showed a 5.6 times higher leaching loss from grazed compared to cut grassland with 36% of total N inputs lost from grazed compared to 6% lost from cut grassland. On our site leaching represented equaled about 20 % of total inputs in grazed years, compared to 39% in the cut years. However, the higher value in cut years was due to the high rainfall in 2002.

Due to its high atmospheric N<sub>2</sub> background in the atmosphere, N<sub>2</sub> fluxes cannot be measured 1350 directly in the field. However, there There are different methods to measure  $N_2$  fluxes indirectly, which have been summarized by Groffman et al. (2006). In our study, we estimated N<sub>2</sub> losses using the process based biogeochemical model LandscapeDNDC (Haas et al., 2013, Molina-Herrera et al., 2016). These losses They represented the highest gaseous N losses from our grassland in most years, with an average of 12.6 % of total N inputs and 14 % of inorganic fertilizer N inputs. This is comparable with 1355 anthe average  $N_2$  loss of 12.5 % from inorganic N applications measured by the acetylene inhibition method from a fertilized and cut, but ungrazed grassland in Switzerland (Rudaz et al., 1999). Using the same method, V+an der Salm et al. (2007) reported a higher loss of 22% of total N input from a cattle grazed pasture on a heavy clay soil in the Netherlands. In addition to Apart from the impact of the heavy clay soil, which could have enhanced denitrification due to reduced oxygen concentrations, grazing is 1360 likely to have enhanced denitrification rates in van-Van der Salm's study. Grazing not only enhances denitrification through soil compaction caused by trampling animals but also due to the formation of N hot spots resulting from unevenly distributed soil N from excretion. In our study N<sub>2</sub> losses simulated by LandscapeDNDC were-are based on average (per ha<sup>-1</sup>) changes of the soil N pool instead of the more uneven distribution of soil N in hot spots like urine patches. Therefore is it is likely that N<sub>2</sub> losses in our 1365 study have been underestimated and better- estimation would contribute to a significant reduction in the uncertainties associated with the overall N budget.

Nitrous oxide emissions are influenced by both management and environmental conditions (Flechard et al., 2007, Bell et al., 2015; Cowan et al., 2015). In our study N<sub>2</sub>O fluxes showed typical temporal variations with high N<sub>2</sub>O peaks after N application decreasing to background levels after < 1 to 20 days, increased losses during wetter periods, and reduced losses during the colder winter months

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(Skiba et al., 2013). Spatial variability was high due to the uneven distribution of excreta and urine and uneven soil compaction from grazing animals (Jones et al., 2011). Annual N<sub>2</sub>O emissions Values measured in our study (0.1 to 1.3 g N m<sup>-2</sup> y<sup>-1</sup>) are within the range of literature values from reported grazed as well as un-grazed European grasslands (Velthof and Oenema, 1997; Leahy et al., 2004; 1375 Flechard et al., 2007). Generally N<sub>2</sub>O losses are higher from grazed grassland compared to cut, ungrazed pasture (Velthof and Oenema, 1995; Luo et al., 1999) due to a more anaerobic environment as a consequence of soil compaction caused by animal treading and the influence of N and C from the deposition of animal excreta to the soil-(Oenema et al., 1997). We did not observe could only find any 1380 correlations between annual N<sub>2</sub>O emissions and stocking density, rainfall or total N input-if certain vears were excluded. This demonstrates shows that N<sub>2</sub>O emissions are not simply a uniform fraction of N applied, as suggested by the Tier 1 IPPC methodology, but are also influenced by the type of N applied, by stocking density, and by the rainfall at the time of fertilization (Jones et al., 2007; Flechard et al., 2007). We found a relationship between the cumulative precipitation one-1 week before and plus three<sup>3</sup> weeks after fertilization withand N<sub>2</sub>O emissions ( $R^2=0.53$ ) (Skiba et al., 2013). This relationship, 1385 together with the influence of stocking density and type of N applied needs to be considered when developing Tier 2 N<sub>2</sub>O emission factors, Emission factors, calculated as a simple fraction of total N input (mineral and organic fertilizer) ranged betweenshowed a variation of 0.6 and 7.24% on our field (Table 6). In our study EFs were above the uncertainty range (0.3 - 3 %) given by IPCC Tier 1 1390 guidelines (IPCC, 2006b) iIn fourfive out of eight nine years, this value was above the uncertainty range (0.3 - 3 %) given by IPCC Tier 1 guidelines (IPCC, 2006b). However, it has been shown that the  $N_2O$  emission factor from managed grassland can be higher, especially under wet conditions and with a high soil C content as this is the case for Scottish soils (Jones et al., 2007; Dobbie et al., 1999; Buckingham et al., 2013Bell et al., 2015).

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In grazed pastures  $NH_3$  volatilizes from urine patches, decomposing dung as well as from fertilizers containing urea and  $NH_4^+$  (Twigg et al 2011). Increased rates of  $NH_3$  losses have been associated with high stocking densities under a rotational grazing system by Ryden and Mc Neill (1984). In our study, N volatilized as  $NH_3$  and NOx from inorganic and organic fertiliser and animal

excretion, before it was incorporated into the soil, and accounted for a considerable amount of total N, with losses of 13 % in cut and grazed years (2002, 2003) and 17 % in grazed only years. Apart from 2004, where stocking rates were highest, NOx and NH<sub>3</sub> volatilizations from inorganic fertilizer applications exceeded those from animal excretion, while those from organic manure applications exceeded those from inorganic fertilizers (2004, 2005). However there is a high uncertainty attributed to those estimates. In contrast, relation to the total N loss from our grassland system, soil NOx emissions from our grassland were estimated to be negligible, accounting for less than 1% of the total budget.

\_Soil NOx emissions result predominantely from microbial nitrification of either added N fertilizers or following the mineralization of soil organic matter, animal excretions or added manure. Emissions tend to be linked with aerobic soil conditions (Davidson, 1991). In relation to the total N loss from our grassland system, soil NOx emissions were estimated to be negligible, accounting for less than 1% of the total budget.

### 4.1.3 N storage in the soil

Results from soil analysis taken in May 2004 and May 2011 indicate that our field has stored N (-4.51  $\pm$  2.64 g N m<sup>-2</sup> y<sup>-1</sup>). The N budget assessed from the net N flux balance also showed that N was stored in the soil of our grassland over the same 7 years at a higher rate (-8.44  $\pm$  4.21 g N m<sup>-2</sup> y<sup>-1</sup>), although values were not significantly different from each other. at a higher rate (average N storage of 7.2  $\pm$  4.6 g N m<sup>-2</sup> y<sup>-1</sup> over all 9 years and average N storage of 9.16  $\pm$  4.09 g N m<sup>-2</sup> y<sup>-1</sup> in grazed years, 2004-2011). The slight shifts in measurement periods (May 2004 – May 2011) for the soil stock calculations and the period for flux budget calculations (Jan 2004 – Dec 2010), is presumed to be insignificant in this comparison.

Results from both methods are within the range of literature values. Neeteson and Hassink (1997) found a N accumulation in SOM of 0-25 g N m<sup>-2</sup> y<sup>-1</sup> from two <u>eattle-cattle-grazed</u> farms in the Netherlands, while Watson et al. (2007) reported a N storage in grazed Irish grasslands ranging from 10-15.2 g N m<sup>-2</sup> y<sup>-1</sup>, depending on N inputs. Soil N storage assessed from soil measurements from a cut grassland close to our field, where plots were treated with cattle slurry, stored N over 6 years at a rate of -2.17 g N m<sup>-2</sup> y<sup>-1</sup> in the top 10 cm, while, in the same experiment, a N loss was observed from mineral N

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and urea treatments (4.5 and 8.3 g N m<sup>-1</sup>y<sup>-1</sup>, respectively) (Jones et al., 2007). In contrast, Schipper et al. (2007) reported an average loss of 9.1 g N m<sup>-2</sup> y<sup>-1</sup> in the top 100 cm from managed grasslands over 20 years in New Zealand.

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The reason for the small difference between methods (flux measurements vs sequential soil sampling) in our study might lie in a possible underestimation of losses from flux measurements. Uncertainties of our estimates are high, especially those forrom N losses. The largest absolute systematic uncertainty for the N balance was attributed to N leaching as for most years values were modelled using data to valuate the model from only one location.- Leaching was modelled for most vears, whereas the model was validated using measured data from October 1<sup>st</sup> 2006 - March 30<sup>th</sup> 2008. The spatial variability of leaching was not considered in the measured data, as only one location has been used. The uncertainty of the leaching estimate would therefore be reduced if the model could be validated with data measured from several locations. The second highest systematic uncertainty was attributed to losses through  $N_2$ , followed by NOx/NH<sub>3</sub> emission from excretion, NOx/NH<sub>3</sub> emission from inorganic fertilization and inputs from organic fertilization- Combined uncertainties from all components lead to a total systematic uncertainty in the N balance of 2.1 g N m<sup>-2</sup> y<sup>-1</sup> (2004-2010).

#### 4.2. Carbon balance

#### 1445 4.2.1. Net ecosystem exchange

We observed large variations of NEE between years, caused by varying management and environmental conditions. The maximum uptake of CO2 measured in our study is close to the upper range of NEE reported for temperate grasslands (100 to 600 g C m<sup>-2</sup> y<sup>-1</sup>, (IPCC, 1996). On an annual basis our grassland site was a sink for atmospheric  $CO_2$  in most years. NEE was only positive in 2004, which was 1450 likely to be due to a combination of slurry spreading and a high livestock density. Generally, grazing causes a very gradual impact on the CO<sub>2</sub> uptake as a part of the field is defoliated each day. The reduced leaf area index (LAI) then leads to a reduced  $CO_2$  uptake by plants. In addition to the reduced LAI, grazing presents a source of  $CO_2$  from animal respiration, thereby reducing the  $CO_2$  sink of the grassland within the field (Levy et al., submitted). The maximum uptake of CO<sub>2</sub> measured in our study is close to the upper range of NEE reported for temperate grasslands (100 to 600 g C m<sup>-2</sup> y<sup>-1</sup>, (IPCC,

1996). Indeed, annual NEE of all years correlated negatively with livestock density if years with cuts were excluded. On average over the 9 years the magnitude of the NEE on our grassland (-218.0  $\pm$  154.5  $g C m^{-2} y^{-1}$ ) was close to the average NEE measured in a comparison of nine European grasslands over two years  $(240 \pm 70 \text{ g C m}^{-2} \text{ y}^{-1})$  by Soussana et al. (2007) and comparable to the CO<sub>2</sub> sink capacity of managed Irish grasslands measured by Byrne et al. (2007) (290  $\pm$  50 g C m<sup>-2</sup> y<sup>-1</sup>) or Leahy et al. (2004) 1460 (257 g C m<sup>-2</sup> y<sup>-1</sup>). Despite high variability over the 9 years, the average NEE value was significantly different from zero (p < 0.05). The NEE represents the difference between the gross primary production (GPP) and the total ecosystem respiration (TER), both influenced by temperature, precipitation and management, though GPP is mainly controlled by PAR above a certain temperature threshold if water 1465 and nutrients are not limiting. The range of the calculated annual gross primary production (GPP) (-982 to -2163 g C m<sup>-2</sup> y<sup>-1</sup>) and terrestrial ecosystem respiration (TER) (972 to 2183 g C m<sup>-2</sup> y<sup>-1</sup>) from our field were within reported values for other managed grasslands. Gilmanov et al. (2007) reported the GPP of 18 intensively managed European grasslands ranging from 467 to 1874 g C m<sup>-2</sup> y<sup>-1</sup> and TER ranging from 493 to 1541 g C m<sup>-2</sup> y<sup>-1</sup>, while Mudge et al. (2011) reported values of 2000 g C m<sup>-2</sup> y<sup>-1</sup> for 1470 GPP and TER from a intensively grazed dairy pasture in New Zealand.

# 4.2.2. Net biome production

The total C budget (=NBP), which includes \_When \_all components of C import and export \_were included in addition to the CO<sub>2</sub> exchange (NBP), was negative on average, meaning that \_C was stored in ourthe grassland over the 9 years. However, due to the high variability between years, NBP was not significantly different from zero (p = 0.05), suggesting that our site is carbon neutral. The average C storage value on our site (164 ± 140 g C m<sup>-2</sup> y<sup>-1</sup>) is higher than most estimates reported in literature, but due to the high annual variation, still within the range of reported values; Soussana et al. (2007) reported C storage estimates from European grazed and cut grasslands of 104 ± 73 g C m<sup>-2</sup> y<sup>-1</sup>, and Mudge et al. (2011) reported for a grazed and cut grassland in New Zealand fluxes of 59 ± 56 g C m<sup>-2</sup> y<sup>-1</sup> in two consecutive years. NBP estimates from a Swiss grassland cut for silage was shown to sequester C at a rate of 147 ± 130 g C m<sup>-2</sup> y<sup>-1</sup> (Ammann et al., 2007), while estimates from a cut grassland in Germany was shown to vary from being a sink (-28 g C m<sup>-2</sup> y<sup>-1</sup>) to being a

source of C (+25 g C m<sup>-2</sup> y<sup>-1</sup>), depending on years (Prescher et al., 2010). The inclusion of all C imports and exports lead to a weakening of the C sink strength assessed from NEE measurements in five5 years 1485 and even changed the grassland from being a sink to being a source in 2006. Due to the C export from harvest, C sequestration tends to be lower in cut systems. This is represented in our study in the lower residual value of NPP in cut years (Figure 4a) compared to the residual value from grazed only years (Figure 34-b), whereas the residual value represents the C storage in the soil as well as the uncertainty 1490 of the budget. The grassland off take from harvest weakened the annual C sink capacity assessed from the NEE by 51 % (2002) and 43 % (2003). However, it has to be kept in mind that the herbage yielded from cuts will end up as animal feed; C will be digested and respired off-site, releasing  $CO_2$  and  $CH_4$  to the atmosphere as well as being returned to the grassland as manure. It is likely that much of the organic C in the manure is decomposed and evolved to the atmosphere as  $CO_{27}$  with very little being retained in 1495 soil because of the lack of contact between manure and soil: there is some evidence of this from two long term grassland experiments in the UK (Hopkins et al., 2009). When the only management was grazing (2004-2010) the NEE showed to be a good proxy of the NBP. In those years the plant biomass was digested on site by the grazing animals and thereby contributed to total ecosystem respiration Only a small fraction of the digested C was incorporated into the body of the grazing animal as 1500 meat and wool, while the largest part was respired as CO<sub>2</sub> shortly after intake. Results from soil analysis indicate that our grassland has lost C from 2004-2010 (29±38 g C m<sup>-2</sup> v<sup>-1</sup> Table 7). We estimated that CH<sub>4</sub> emissions from grazing animals were only 0.7 % of NPP. Methane emissions were also measured by eddy covariance technique over several months in 2010 on the same field (Dengel et al., 2011). By dividing CH4 fluxes by the number of sheep in the field each day, Dengel et al. calculated CH<sub>4</sub> emissions per head of livestock as 7.4 kg CH<sub>4</sub> head + v<sup>+</sup> for sheep, which is close 1505 to the emission factor used in our budget of 7.6 kg CH<sub>4</sub> head  $^{+}$  v<sup>+</sup> for ewes, showing that our estimates were realistic. Methane emissions from slurry spreading were relatively high on specific days (up to 0.28 g C m<sup>-2</sup> d<sup>-1</sup>, measured with chamber method), however, they were negligible on an annual basis as peaks only lasted for 2-3 days.

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Carbon leaching from managed grasslands has not been reported in many studies. Kindler et al. (2011) reported C leaching from various European ecosystems, whereas the measured data (2007) from our experimental field was part of the study. Our data (30.0 g C m<sup>+</sup> v<sup>+</sup>, average of two locations as published in Kindler et al. (2011)) were close to the average value (29.4 g C m<sup>-1</sup> v<sup>-1</sup>) of the reported European grasslands, which showed a range of C losses of 6.5 42.5 g C m<sup>+</sup> v<sup>+</sup>. Higher losses have been 1515 observed by McTiernan et al. (2001), who measured DOC export from grassland lysimeter plots treated with N fertilizer and slurry over two months. Up scaled to one year, they measured DOC loss between 25.2 and 70.8 g C m<sup>-2</sup> v<sup>+</sup>, all above what we measured in our study. Important factors controlling the magnitude of C leaching have been shown to be drainage, the topsoil C/N ratio and the saturation of the 1520 subsoil's sorption capacity for organic C (Kindler et al., 2011; McTiernan et al., 2001). In waterlogged soils the soil organic matter (SOM) decomposition and groundwater recharge tend to be reduced and thus the amount of C prone to leaching compared to that under more aerobic conditions associated with drainage. Although our field was drained more than 50 years ago, the drainage system does not operate very well, resulting in large puddles of standing water during prolonged periods of rain. The measured 1525 data used for the budget were taken at one sampling point, which was not in a waterlogged area. Therefore our leaching estimates are highly uncertain and could be significantly lower and C exports overestimated. The spatial heterogeneity within the grassland field caused by uneven water management as well as faeces and urine patches requires to sample at more points in order to obtain a representative leaching value. Therefore our leaching estimates are highly uncertain and could be significantly lower 1530 and C exports overestimated.

The systematic uncertainty of the C balance is mainly determined by the error of the  $CO_2$  exchange, followed by the systematic uncertainty of the harvest export, organic fertilizer input and leaching losses. Combined uncertainties from all components lead to a total systematic uncertainty of the C balance of 18.3 c C m<sup>2</sup> · ·

4.2.3 C sequestration

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	Unlike forests, most of the C stored by grasslands is contained within soil organic matter.
	Carbon sequestration in grasslands can therefore be either determined directly from measuring soil
1540	organic carbon changes or indirectly by measuring the net C balance flux. If measuring soil C changes,
	the internationally recommended practice in carbon accounting is to express soil C stocks to a depth of
	30 cm (IPCC, 1997). However, as the bulk density often changes over time with land use, the soil C
	content per unit ground area to a fixed depth will also change even without any change in the mass
	fraction of C in dry soil. By using the ESM method this problem is avoided, by considering the whole
1545	soil mass present in the 0 60 cm soil layer. A comparison of the C storage calculated from the net C flux
	balance from 2004-2010 with C stock changes measured from soil sample analysis (Table 7) show that,
	although the flux balance estimated a C sequestration, while based on soil measurements C was lost,
	neither value was significantly different from zero. In the literature, losses as well as storage of C at
	various rates have been reported from managed grasslands assessed from soil stock measurements. Soil
1550	stock change measurements from our field are comparable with values found in the literature.
	Depending on the study, managed grasslands in Belgium were shown to either loose (90 g C m <sup>-2</sup> y <sup>-1</sup> ,
	Lettens et al., 2005a) or sequester carbon (4.4 g C m <sup>-2</sup> y <sup>-1</sup> in 0-30 cm, Goidts and Van Wesemael, 2007;
	22.5 g C m <sup>-2</sup> y <sup>-1</sup> in 0-30 cm, Lettens et al. 2005b). Schipper et al. (2007) reported losses of C from
	pastures in New Zealand over 20 years at an average rate of 106 g C m <sup>-2</sup> y <sup>-1</sup> (top 100 cm), whereas these
1555	losses were a result of an earlier land use change from forestry. Schuman, et al., (2002) measured a C
	sequestration of 10-30 g C m <sup>-2</sup> y <sup>-1</sup> from US rangelands (0-60 cm)), while Watson et al. (2007) measured
	a C storage at 112-145 g C m <sup>-2</sup> y <sup>-1</sup> in the top 15 cm soil layer from a grazed Irish grassland. Bellamy et
	al. (2005) showed no evidence of increased C in the topsoil of grasslands in England and Wales and
	Hopkins et al. (2009) found no significant change of SOC over time in two UK long term experiments.
1560	The above mentioned results are contrasting and inconclusive, because observed C sinks in grasslands
	are the effect of land management or land use change prior to the beginning of the C stock change
	measurement. Soussana et al (2014) concluded in a theoretical study that grassland SOC sequestration
	has a strong potential to partly mitigate the GHG balance of ruminant production systems at low grazing
	intensities, but not with intensive systems. Smith (2014) examined evidence from repeated soil surveys,
1565	long term grassland experiments and simple mass balance calculations and concluded that, although

grasslands can act as C sinks, they cannot act as a perpetual C sink and thus could not be used as an offset for GHG emissions.

The comparison of the C storage calculated from the net C flux balance with soil C stock changes show that, the flux balance estimated a C sequestration, while based on C stock changes, C was 1570 lost, although neither value was significantly different from zero (Table 7). A literature search by Soussana et al. (2010) showed that generally C sequestration calculations on grassland were lower if derived from SOC stock changes (average  $-5\pm30$  g C m<sup>-2</sup> y<sup>-1</sup>) compared to C flux balances (average - $22\pm56$  g C m<sup>-2</sup> y<sup>-1</sup>), although these estimates were not significantly different from each other. However, in none of those reviewed studies were C flux and C stock change measured in the same field experiment. A reason for the discrepancy between <del>calculation</del> estimation methods in our study might lie 1575 in a possible underestimation of C exports in the flux balance calculation, leading to an overestimation of C storage in the soil. One underestimated flux could be the export of DIC and DOC. Carbon leaching from managed grasslands has not been reported in many studies. Kindler et al. (2011) reported C leaching from various European ecosystems, where the measured data (2007) from our experimental field was part of the study. Our data (30.0 g C m<sup>-1</sup> y<sup>-1</sup>, average of two locations as published in Kindler 1580 et al. (2011)) were close to the average value (29.4 g C m<sup>-1</sup> y<sup>-1</sup>) of the reported European grasslands, which showed a range of C losses of 6.5-42.5 g C  $m^{-1}$  v<sup>-1</sup>. Higher losses have been observed by McTiernan et al. (2001), who measured DOC export from grassland lysimeter plots treated with N fertilizer and slurry over two months. Up-scaled to one year, they measured DOC loss between 25.2 and 70.8 g C m<sup>-2</sup> y<sup>-1</sup>, all above what we measured in our study. Important factors controlling the magnitude 1585 of C leaching have been shown to be drainage, the topsoil C/N ratio and the saturation of the subsoil's sorption capacity for organic C (Kindler et al., 2011; McTiernan et al., 2001). In waterlogged soils the soil organic matter (SOM) decomposition and groundwater recharge tend to be reduced and thus the amount of C prone to leaching compared to that under more aerobic conditions associated with 1590 drainage. Although our field was drained more than 50 years ago, the drainage system does not operate very well, resulting in large puddles of standing water during prolonged periods of rain. The measured data used for the budget were taken at one sampling point, which was not in a waterlogged area. The spatial heterogeneity within the grassland field caused by uneven water management as well as faeces

and urine patches requires to sample at more points in order to obtain a representative leaching value. 1595 Therefore our leaching estimates are highly uncertain and could be significantly lower and C exports overestimated. Furthermore, IL-eaching was only measured in one year (2008), while values for remaining years were estimated using a simple regression model with an attributed high uncertainty of 30-32 % (5.34.9 g C m<sup>-2</sup> y<sup>-1</sup> of average fluxes). Further uncertainty could be due to the use of only one sampling location, which might not be representative of the whole field due to high spatial heterogeneity (see Sect. 4.1.2.). Indeed, Siemens (2003) hypothesized that the underestimation of C 1600 leaching from soils can explain a large part of the difference between atmosphere- and land-based estimates of the C uptake of European terrestrial ecosystems. Another underestimated flux could be the loss of CO<sub>2</sub> in the NEE measurements. Gapfilling can introduce uncertainties in the NEE data especially for years with low data capture. Furthermore, CO<sub>2</sub> losses from animal respiration could be underestimated at times due to the animals moving out of the footprint of the EC mast. Using animal 1605 respiration values from chamber experiments of 12.1 g CO<sub>2</sub> kg<sup>-1</sup> live weight d<sup>-1</sup> for cows and 11.7 g CO<sub>2</sub> kg<sup>-1</sup> live weight d<sup>-1</sup> for sheep and lambs (Shane Troy, SRUC, personal communication), we estimated a maximum CO<sub>2</sub> loss from animal respiration of 53 g C m<sup>-2</sup> y<sup>-1</sup> (2002-2010) or 59 g C m<sup>-2</sup> y<sup>-1</sup> (2004-2010).\_\_So if we assume that all animal respiration has been missed by eddy covariance 1610 measurements then the C sink estimated from NEE measurements would be reduced by 24 % (2002-2010) or 33 % (2004-2010). This theoretical maximum 33% reduction would reduce the net carbon balance to ~ 122 g C m<sup>-2</sup> y<sup>-1</sup> (2004-2010).

The systematic uncertainty of the C balance is mainly determined by the error of the CO2 exchange, followed by the systematic uncertainty of the harvest export, organic fertilizer input and leaching losses. Combined uncertainties from all components lead to a total systematic uncertainty of the C balance of 18.3 g C m<sup>2</sup> v<sup>4</sup>. In the literature, losses as well as storage of C at various rates have been reported from managed grasslands assessed from soil stock measurements. Soil stock measurements from our field are comparable with Depending on the study, managed grasslands in Belgium were shown to either loose (90 g C m<sup>-2</sup> v<sup>-1</sup>, Lettens et al., 2005a) or sequester carbon (4.4 g C  $m^2 y^+$ in 0 30 cm, Goidts and Van Wesemael, 2007; 22.5 g C  $m^2 y^+$ in 0 30 cm, Lettens et al. 2005b). 1620 Schipper et al. (2007) reported losses of C from pastures in New Zealand over 20 years at an average

rate of 106 g C m<sup>-2</sup> v<sup>-1</sup> (top 100 cm), whereas these losses were a result of an earlier land use change from forestry, the C sequestration of 10 30 g C m<sup>2</sup> y<sup>4</sup>, measured from US rangelands (0 60 cm, Schuman, et al., 2002), while Watson et al. (2007) measured a C storage at 112 145 g C m<sup>-2</sup> v<sup>+</sup> in the top 15 cm soil layer from a grazed Irish grassland. Bellamy et al. (2005) showed no evidence of 1625 increased C in the topsoil of grasslands in England and Wales and Hopkins et al. (2009) found no significant change of SOC over time in two UK long term experiments. Depending on the study, managed grasslands in Belgium were shown to either loose (90 g C m<sup>2</sup> y<sup>4</sup>, Lettens et al., 2005a) or sequester earbon (4.4 g C m<sup>2</sup> y<sup>4</sup> in 0-30 cm, Goidts and Van Wesemael, 2007; 22.5 g C m<sup>2</sup> y<sup>4</sup> in 0-30 em, Lettens et al. 2005b). Schipper et al. (2007) reported losses of C from pastures in New Zealand over 1630 20 years at an average rate of 106 g C m<sup>2</sup> y<sup>++</sup>(top 100 cm), whereas these losses were a result of an earlier land use change from forestry. The above mentioned results are contrasting and inconclusive. because observed C sinks in grasslands are the effect of land management or land use change prior to the beginning of the C stock change measurement. Soussana et al (2014) concluded in a theoretical study that grassland SOC sequestration has a strong potential to partly mitigate the GHG balance of 1635 ruminant production systems at low grazing intensities, but not with intensive systems. Smith (2014) examined evidence from repeated soil surveys, long term grassland experiments and simple mass balance calculations and concluded that, although grasslands can act as C sinks, they cannot act as a perpetual C sink and thus could not be used as an offset for GHG emissions. 1640 In addition to uncertainties in the flux budget calculations, uncertainties are also attributed to

In addition to uncertainties in the flux budget calculations, uncertainties are also attributed to soil C and N stock measurements. Soil inventory data in our study indicated a loss of C and a storage of N over 7 years, which seems contradictory, although C storage was not significantly different from zero. The uncertainty of soil C and N stock measurements arise from the variability of soil C and N concentrations due to errors from laboratory and to their high spatial variability as well as from the variability in the rock fragment content (Goidts et al., 2009).

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# 4.3 Greenhouse gas budget

In the overall N and C budget N<sub>2</sub>O and CH<sub>4</sub> emissions were negligible in terms of N and C losses from 1650 the system (1 - 8%) of total N losses and 0.6 - 4.5% of total C losses, respectively). However, in terms of CO<sub>2</sub> equivalents, N<sub>2</sub>O emissions as well as CH<sub>4</sub> emissions strongly affected the GHG budget. Since the radiative forcing effect of  $N_2O$  is 298 times greater than that of  $CO_2$  a relatively small emission of N<sub>2</sub>O can exert a strong influence on the total radiative forcing budget of an ecosystem. Indeed, the sink strength of the NEE was weakened by N<sub>2</sub>O emissions by 29 % over all years. Methane emissions from 1655 soil processes, manure input and animal excretion were negligible in terms of the C budget as well as in the GHG budget. In contrast, enteric fermentation proved to be an important GHG source-Methane emissions were also measured by eddy covariance technique over several months in 2010 on the same field (Dengel et al., 2011). By dividing CH<sub>4</sub> fluxes by the number of sheep in the field each day, Dengel et al. calculated CH<sub>4</sub> emissions per head of livestock as 7.4 kg CH<sub>4</sub> head<sup>-1</sup> v<sup>-1</sup> for sheep, which is close to the emission factor used in our budget of 7.6 kg CH<sub>4</sub> head<sup>-1</sup> y<sup>-1</sup> for ewes, showing that our estimates 1660 were realistic. The positive correlation of  $CH_4$  emissions with the stock density indicates that any changes in animal production will have a major impact on the global  $CH_4$  budget. The weakening of the GHG sink strength of the NEE by N<sub>2</sub>O and CH<sub>4</sub> emissions, show the importance of those two gases in terms of global warming. Thus, adapting the management of grasslands by adding fertilizer or manure to increase plant growth and thus improve C sequestration in soils may increase N<sub>2</sub>O emissions, while 1665 changing land use from cropland to pasture in the attempt to reduce C losses from soils might lead to increased CH<sub>4</sub> losses from grazing animals if the total number of animals increases rather than animals are fed in a different way.

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### 5. Conclusion

In our study only a small proportion of the N inputs from inorganic fertilizer and organic manure were converted to animal outputs or stored in the soil, while the main part was lost through leaching and gaseous emissions. An improvement of the NUE would mean both an economic profit for the farmer as well as an environmental benefit. Estimates from flux budget calculations indicated that our grassland

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was sequestering C. However, although grasslands can act as C sinks, they can not cannot act as a

perpetual C sink and thus could not be used as an offset for GHG emissions (Smith et al., 2014). Instead, as it is easier and faster for soils to lose than to gain carbon, care must be taken to preserve C loss by management options, rather than trying to increase carbon stocks in grasslands. There was a 1680 discrepancy between soil stock measurements and flux budget calculations for the C as well as the N budget. The reason for the discrepancy between C budgetstorage estimates might lie in a possible underestimation of C exports such as leaching and animal respiration as well as the uncertainty due to gapfilling in the NEE data. The N accumulation budget storage might have been overestimated by the flux calculations through a possible overestimation of N losses, mainly through leaching as well as 1685 through N<sub>2</sub> and NOxNH<sub>4</sub> emissions. Furthermore, uncertainties are also attributed to soil C and N stock measurements. Our data have shown that the information about the potential of managed grasslands to act as sinks or sources for GHG is important for mitigation and adaption purposes. High plant productivity, stimulated by fertilisation, resulted in high plant CO<sub>2</sub> fixation. However, increased N losses through N<sub>2</sub>O emissions counteracted the benefits of C sequestration in terms of GHG emissions. 1690 Furthermore, CH<sub>4</sub> emissions from enteric fermentation largely reduced the positive effect of CO<sub>2</sub> uptake, especially in years where NEE rates were small. We therefore conclude that CO<sub>2</sub> exchange alone is not sufficient for the estimation of the GWP of a managed grassland ecosystem. Only a comprehensive approach, combining C and N cycling will help us to better understand functionalities of ecosystems and to improve modelling by integrating this knowledge.

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**Appendix A. Supplementary information** 

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# **Reference List**

1710	Allard, V., Soussana, J. F., Falcimagne, R., Berbigier, P., Bonnefond, J. M., Ceschia, E., D'hour, P., Henault, C., Laville, P., Martin, C., and Pinares-Patino, C.: The role of grazing management for the net biome productivity and greenhouse gas budget (CO <sub>2</sub> , N <sub>2</sub> O and CH <sub>4</sub> ) of semi-natural grassland, Agr. Ecosyst. Environ., 121.1-2, 47-58, 2007.
1715	Ammann, C., Flechard, C. R., Leifeld, J., Neftel, A., and Fuhrer, J.: The carbon budget of newly established temperate grassland depends on management intensity, Agr. Ecosyst. Environ., 121.1-2, 5-20, 2007.
	Ammann, C., Neftel, A., Spirig, C., Leifeld, and J., Fuhrer, J.: Nitrogen balance of hay meadows with and without fertilization, Agrarforschung, 16.9, 348-53, 2009.
1720	Amthor, J.S.: The McCree-de Wit-Penning de Vries-Thornley Respiration Paradigms: 30 Years Later, Annals of Botany 86 (1): 1-20, 2000.
1725	Askegaard, M., Olesen, J. E., and Kristensen, K.: Nitrate leaching from organic arable crop rotations: effects of location, manure and catch crop. Soil Use Manage., 21.2, 181–88, 2005.
	Atti, N., Rouissi H., and Othmane, M. H.: Milk production, milk fatty acid composition and conjugated linoleic acid (CLA) content in dairy ewes raised on feedlot or grazing pasture, Livest. Sci., 104.1-2, 121-27, 2006.
1730	Aubinet, M., Grelle, A., Ibrom, A., Rannik, U., Moncrieff, J., Foken, T., Kowalski, A. S., Martin, P. H., Berbigier, P., Bernhofer, C., Clement, R., Elbers, J., Granier, A., Grunwald, T., Morgenstern, K., Pilegaard, K., Rebmann, C., Snijders, W., Valentini, R., and Vesala, T.: Estimates of the annual net carbon and water exchange of forests: The EUROFLUX methodology, Adv. Ecol. Res., 30, 113, 75, 2000
1755	Ball, P. R. and Ryden. J. C.: Nitrogen Relationships in Intensively Managed Temperate Grasslands, Plant Soil, 76.1-3, 23-33, 1984.
1740	Bell. M. J., Rees, R.M., Cloy, J. M., Topp, C. F. E., Bagnall A., Chadwick, D. R.: Nitrous oxide emissions from cattle excreta applied to a Scottish grassland: Effects of soil and climatic conditions and a nitrification inhibitor, Sci Total Environ, 508, 343–353, 2015.
ļ	Bechmann, M., Eggestad, H. O., and Vagstad, N.: Nitrogen balances and leaching in four

agricultural catchments in southeastern Norway, Environ. Pollut., 102, 493-99, 1998.

Bellamy, P. H., Loveland, P. J., Bradley, R. I., Lark R. M., and Kirk, G. J. D.: Carbon	
osses from all soils across England and Wales 1978-2003, Nature, Vol 437, 245-48, 2005	<i>.</i>

1	750	
L	/ 10	
	/ . / / /	

Bleeker, A., Reinds G. J., Vermeulen A. T., de Vries W., and Erisman J. W.: Critical loads and resent deposition thresholds of nitrogen and acidity and their exceedances at the level II and level I monitoring plots in Europe, ECN report ECN-C-04-117, Petten, The Netherlands, December 2004. 2004.

1755

1

Bleeker, A., Hicks, W. K., Dentener, F., Galloway, J., Erisman, J. W.: N deposition as a threat to the World's protected areas under the Convention on Biological Diversity. Environ. Pollut. 159, 2280e2288, 2011.

760	Brady N.C. Wail P. P. The Nature and Properties of Soils 13th Edition Prentice Hall
/00	Brady, N. C., Wen, R. R. The Nature and Hoperites of Bons, 15th Edition, Frentice Han,
	Upper Saddle River NI 960 pp ISRN 0 13 016763 0 2002
	- Opper Suddre River, 13, 900 pp., 15bit 0 15 010705 0, 2002.

Braun-Blanquet J.: Pflanzensoziologie, Grundzüge der Vegetationskunde, 3. Aufl. Springer, 1964, Verlag, Wien and New York.

1765

Byrne, K. A., Kiely, G., and Leahy, P: Carbon sequestration determined using farm scale carbon balance and eddy covariance, Agr. Ecosyst. Environ., 121.4, 357-64, 2007.

Buckingham, S., Rees, R.M. & Watson, C.A. 2013. Issues and pressures facing the future of soil carbon
 stocks with particular emphasis on Scottish soils. The Journal of Agricultural Science, 152, 699-715.

Butterbach-Bahl, K., Gasche, R., Breuer, L., and Papen, H.: Fluxes of NO and N<sub>2</sub>O from temperate forest soils: impact of forest type, N deposition and of liming on the NO and N<sub>2</sub>O emissions, Nutr. Cycl. Agroecosys., 48, 79-90, 1997.

1775

Caro, D., Davis S. J., Bastianoni, S., Caldeira K.: Global and regional trends in greenhouse gas emissions from livestock. Climatic Change, 126:203–216, 2014

 Chen, W., McCaughey, W. P., and Grant, C. A:. Pasture type and fertilization effects on
 N-2 fixation, N budgets and external energy inputs in western Canada, Soil. Biol. Biochem., 36.8, 1205-12, 2004.

 Chirinda N., Kracher D., Laegdsmand M., Porter J. R., Olesen J.E., Petersen B. M., Doltra J., Kiese R., Butterbach Bahl K.:Simulating soil N2O emissions and heterotrophic CO2 respiration in arable systems using FASSET and MoBiLE DNDC. Plant Soil, 343, 139-169, 2011. Clayton, H., Arah, J. R. M., and Smith, K. A.: Measurement of Nitrous-Oxide Emissions from Fertilized Grassland Using Closed Chambers. J. Geophys. Res.-atmos., 99. D8, 16599-607, 1994.

1790 19

Cowan, N. J., Norman, P., Famulari, D., Levy, P. E., Reay, D. S., Skiba, U. M.(2015). Spatial variability and hotspots of soil N<sub>2</sub>O fluxes from intensively grazed grassland. *Biogeosciences* 12 1585 – 1596, 2015.

#### 1795

1815

- Cuttle, S. P. and Scholefield, D.: Management options to limit nitrate leaching from grassland. J. Cont. Hydrol., 20, 299-312, 1995.
- Davidson, E. A., 1991. Fluxes of nitrous oxide and nitric oxide from terrestrial ecosystems. In: Rogers,
  J. E., Whitman, W. B. (Eds.), Microbial Production and Consumption of Greenhouse Gases: Methane,
  Nitrogen Oxides and Halomethanes. American Society of Microbiology,
  Washington, DC, pp. 219–236.
- Davison, B., Brunner, A., Ammann, C., Spirig, C., Jocher, M., and Neftel, A.: Cutinduced VOC emissions from agricultural grasslands, Plant Biol., 10.1, 76-85, 2008.

Dengel, S., Levy, P. E., Grace, J., Jones, S. K., and Skiba, U. M.: Methane emissions from sheep pasture, measured with an open-path eddy covariance system, Glob. Change Biol., 17.12, 3524-33, 2011.

1810 Di Marco, C., Skiba, U., Weston ,K., Hargreaves, K., and Fowler, D.: Field scale N<sub>2</sub>O flux measurements from grassland using eddy covariance, Water Air Soil Poll.: Focus, 4.6, 143-49, 2004.

Dobbie, K. E., McTaggart, I. P., and Smith, K. A.: Nitrous oxide emissions from intensive agricultural systems: Variations between crops and seasons, key driving variables, and mean emission factors, J. Geophys. Res.-atmos., 104.D21, 26891-99, 1999.

Eriksen, J.: Nitrate leaching and growth of cereal crops following cultivation of contrasting temporary grasslands, J. Agr. Sci., 136, 271-81, 2001.

1820 Erisman, J. W., Vanpul A., and Wyers P.: Parametrization of Surface Resistance for the Quantification of Atmospheric Deposition of Acidifying Pollutants and Ozone. Atmos. Environ., 28.16, 2595-607, 1994.

Flechard, C. R., Ambus, P., Skiba, U., Rees, R.M., Hensen, A., van Amstel, A., Pol-van
Dasselaar, A.V., Soussana, J. F., Jones, M., Clifton-Brown, J., Raschi, A., Horvath, L., Neftel, A.;
Jocher, M., Ammann, C., Leifeld, J., Fuhrer, J., Calanca, P., Thalman, E., Pilegaard, K., Di Marco, C.,
Campbell, C., Nemitz, E., Hargreaves, K. J., Levy, P. E., Ball, B. C., Jones, S. K., van de Bulk, W. C.
M., Groot, T., Blom, M., Domingues, R., Kasper, G., Allard, V., Ceschia, E., Cellier, P., Laville, P.,

1830	Henault, C., Bizouard, F., Abdalla, M., Williams, M., Baronti, S., Berretti, F., and Grosz, B.: Effects of climate and management intensity on nitrous oxide emissions in grassland systems across Europe, Agr. Ecosyst. Environ., 121.1-2, 135-52, 2007.
1835	Flechard, C. R., Nemitz, E., Smith, R. I., Fowler, D., Vermeulen, A. T., Bleeker, A., Erisman, J. W., Simpson, D., Zhang, L., Tang, Y. S., and Sutton, M. A.: Dry deposition of reactive nitrogen to European ecosystems: a comparison of inferential models across the NitroEurope network, Atmos. Chem. Phys., 11.6, 2703-28, 2011.
	Flindt, R.: Biologie in Zahlen: Eine Datensammlung inTabellen mit ueber 10000 Einzelwerten. Spektrum akademischerVerlag. Gustav Fischer, 249, 2002.
1840	Foken, T., and Wichura, B.: Tools for quality assessment of surface based flux measurements, Agr. Forest Meteorol., 78.1-2, 83-105, 1996.
1845	Galloway, J. N., Aber, J. D., Erisman, J. W., Seitzinger, S. P., Howarth, R.W., Cowling, E.B., Cosby, B.J.: The nitrogen cascade, Bioscience, 53.4, 341–56, 2003. Galloway, J.N. and Cowling, E.B.: Reactive Nitrogen and the World: 200 Years of Change. 2002. Ambio, Vol. 31, No. 2, Optimizing Nitrogen Management in Food and EnergyProductions, and Environmental Change (Mar., 2002), pp. 64-71.
1850	Gilmanov, T. G., Aires, L., Barcza, Z., Baron, V. S., Belelli, L., Beringer, J., Billesbach, D., Bonal, D., Bradford, J., Ceschia, E., Cook, D., Corradi, C., Frank, A., Gianelle, D., Gimeno, C., Gruenwald, T., Guo, H.Q., Hanan, N., Haszpra, L., Heilman, J., Jacobs, A., Jones, M. B., Johnson, D.
1855	A., Kiely, G., Li,S. G., Magliulo, V., Moors, E., Nagy, Z., Nasyrov, M., Owensby, C., Pinter, K., Pio, C., Reichstein, M., Sanz, M. J., Scott, R., Soussana, J. F., Stoy, P. C., Svejcar, T., Tuba, Z., and Zhou, G. S.: Partitioning European grassland net ecosystem CO <sub>2</sub> exchange into gross primary productivity and ecosystem respiration using light response function analysis, Agr. Ecosyst. Environ., 121.1-2, 93-120, 2007.
1860	Goidts, E. and Van Wesemael, B.: Regional assessment of soil organic carbon changes under agriculture in Southern Belgium (1955-2005), Geoderma, 141.3-4, 341-54, 2007.
	Goidts, E., Van Wesemael, B. and Crucifix, M.: Magnitude and sources of uncertainties in soil organic carbon (SOC) stock assessments at various scales. European Journal of Soil Science, October 2009, 60,

1865

723-739, 2009.

Groffman, P. M., Altabet, M. A., Bohlke, J. K., Butterbach-Bahl, K., David, M. B., Firestone, M. K., Giblin, A. E., Kana, T. M., Nielsen, L. P., and Voytek, M. A.: Methods for measuring
denitrification: Diverse approaches to a difficult problem, Ecol. Appl., 16.6, 2091-122, 2006.

	Haas E., Klatt, S., Fröhlich, A., Kraft, P., Werner, C., Kiese, R., Grote, R., Breuer, L., and Butterbach Babl. K.: Landscape DNDC: a process model for simulation of biosphere	
1875	atmosphere hydrosphere exchange processes at site and regional scale, Landscape Ecol., 28, 615–636, 2013.	
1880	Molina-Herrera S., Haas, E., Klatt, S., Kraus, D., Augustin, J., Magliulo, V., Tallee, T., Ceschia, E., Ammann C., Loubet, B., Skiba, U., Jones, S., Brümmer, C., Butterbach-Bahl, K., Kiese, R.:A modelling study on mitigation of N <sub>2</sub> O emissions and NO <sub>3</sub> leaching at different agricultural sites across Europe using LandscapeDNDC, Science of the Total Environment, 553, 128–140, 2016.	
1885	Haas, E., Klatt, S., Frohlich, A., Kraft, P., Werner, C., Kiese, R., Grote, R., Breuer, L., Butterbach-Bahl, K.: LandscapeDNDC: a process model for simulation of biosphere–atmosphere–hydrosphere exchange processes at site and regional scale. Landscape Ecol, 28, 615–636, 2013. DOI 10.1007/s10980-012-9772-	
1890	Helfter, C., Campbell, C., Dinsmore, K. J., Drewer, J., Coyle, M., Anderson, M., Skiba, U., Nemitz, E., Billett, M. F., and Sutton, M. A.: Drivers of long term variability in CO <sub>2</sub> net ecosystem exchange in a temperate peatland, Biogeosciences, 12, 1799–1811, 2015	
	Herrmann, B., Jones, S. K., Fuhrer, J., Feller, U., and Neftel, A.: N budget and NH <sub>3</sub> exchange of a grass/clover crop at two levels of N application, Plant Soil, 235.2, 243–52, 2001.	
1895	Hopkins, D. W., Waite, I. S., McNicol, J. W., Poulton, P. R., Macdonald, A. J. and O'Donnell, A. G.: Soil organic carbon contents in long-term experimental grassland plots in the UK (Palace Leas and Park Grass) have not changed consistently in recent decades, Glob. Change Biol., 15, 1739-1754, 2009.	
1900	IPCC 1996. Climate change 1995. Impacts, adaptation and mitigation of climate change: scientic, technical analysis, contribution of working group II to the 2nd assessment reports of the IPCC. 1996. Intergovernmental Panel on Climate Change and Cambridge University Press Cambridge UK.	
1905	IPCC 2006a . Guidelines for National Greenhouse Gas Inventories, Agriculture, Forestry and Other Land Use. Chapter 10: Emissions from Livestock and Manure Management. Intergovernmental Panel on Climate Change (IPCC), Volume 4. 2006. Institute for Global Environmental Strategies, Tokyo, Japan.	
1910	IPCC 2006b. Guidelines for National Greenhouse Gas Inventories, Agriculture, Forestry and Other Land Use. Chapter 11: N2O Emissions from Managed Soils, and CO <sub>2</sub> Emissions from Lime and Urea Application. Intergovernmental Panel on Climate Change (IPCC) Volume 4.	

2013.	Institute for	Global H	Environmental	Strategies,	Tokyo, Japan.

 1915 IPCC 2013. Myhre, G., Shindell, D., Bréon, F. M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J. F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T. and Zhang, H.: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Stocker, T. F., D. Qin, G. K. Plattner, M. Tignor, S.K.
 1920 Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P. M. Midgley (eds.), Cambridge University Press.

Cambridge, United Kingdom and New York, NY, USA. Anthropogenie and Natural Radiative Forcing

Janssens, I. A., Freibauer, A., Ciais, P., Smith, P., Nabuurs, G. J., Folberth, G.,

Schlamadinger, B., Hutjes, R. W. A., Ceulemans, R., Schulze, E. D., Valentini, R., and Dolman, A.J.:
Europe's terrestrial biosphere absorbs 7 to 12% of European anthropogenic CO<sub>2</sub> emissions, Science, 300.5625, 1538-42, 2003.

Jarvis, S. C., Lovell, R. D., and Panayides, R.: Patterns of methane emission from excreta of grazing animals, Soil. Biochem., 27.12, 1581-88, 1995.

1930

Jones, S. K., Famulari, D., Di Marco, C. F., Nemitz, E., Skiba, U. M., Rees, R. M., and Sutton, M.A.: Nitrous oxide emissions from managed grassland: a comparison of eddy covariance and static chamber measurements, *Atmospheric Measurement Techniques*, 4,10, 2179-94, 2011.

1935 Jones, S. K., Rees, R. M., Kosmas, D., Ball, B. C., and Skiba, U. M.: Carbon sequestration in a temperate grassland; management and climatic controls, Soil Use Manage., 22.2, 132-42, 2006.

Jones, S. K., Rees, R. M., Skiba, U. M., and Ball, B. C.: Influence of organic and mineral N fertiliser on N<sub>2</sub>O fluxes from a temperate grassland, Agr. Ecosyst. Environ., 121.1-2, 74-83, 2007.

1940

Jones, S. K., Famulari, D., Di Marco, C. F., Nemitz, E., Skiba, U. M., Rees, R. M., and Sutton, M.A.: Nitrous oxide emissions from managed grassland: a comparison of eddy covariance and static chamber measurements, Atmospheric Measurement Techniques, 4.10, 2179-94, 2011.

1945 McKenzie, R. M., Özel, M. Z., Cape, J. N., Drewer, J., Dinsmore, K. J., Nemitz, E., Hamilton, J. F., Sutton, M. A., Gallagher, M. W. and Skiba, U.: The import and export of organic nitrogen species at a Scottish ombrotrophic peatland, Biogeosciences Discussions, 12, 515-554, 2015.

 Kesik M., Ambus P., Baritz R., Brüggemann N., Butterbach Bahl K., Damm M., Duyzer
 J., Horváth L., Kiese R., Kitzler B., Leip A., Li C., Pihlatie M., Pilegaard K., Seufert G., Simpson D., Skiba U., Smiatek G., Vesala T., and Zechmeister Boltenstern S.: Inventories of N<sub>2</sub>O and NO emissions from European forest soils, Biogeosciences, 2, 353–375, 2005.

Kiese R., Heinzeller C., Werner C., Wochele S., Grote R., and Butterbach Bahl K.:

1955 Quantification of nitrate leaching from German forest ecosystems by use of a process oriented biogeochemical model. Environ. Pollut., 159, 3204–3014, 2011.

Kim, J., Verma, S. B., and Clement, R. J.: Carbon-Dioxide Budget in A Temperate Grassland Ecosystem. J. Geophys. Res-Atmos., 97.D5, 6057-63, 1992.

1965

1995

Kindler, R., Siemens, J., Kaiser, K., Walmsley, D. C., Bernhofer, C., Buchmann,
N., Cellier, P., Eugster, W., Gleixner, G., Grunwald, T., Heim, A., Ibrom, A., Jones, S. K., Jones, M.,
Klumpp, K., Kutsch, W., Larsen, K. S., Lehuger, S., Loubet, B., McKenzie, R., Moors, E., Osborne, B.,
Pilegaard, K., Rebmann, C., Saunders, M., Schmidt, M. W. I., Schrumpf, M., Seyfferth, J., Skiba, U.,
Soussana, J. F., Sutton, M.A., Tefs, C., Vowinckel, B., Zeeman, M. J., and Kaupenjohann, M.:

- Dissolved carbon leaching from soil is a crucial component of the net ecosystem carbon balance, Glob. Change Biol., 17.2, 1167-85, 2011.
- Kramberger, M., Podvršnik, A., Gselman, V., Šuštar, J., Kristl, M., Muršec, M., Lešnik, D., Škorjanc:
  The effects of cutting frequencies at equal fertiliser rates on bio-diverse permanent grassland: Soil
  organic C and apparent N budget. Agriculture, Ecosystems & Environment, Volume 212, , Pages 13-20B, 2015.
- Lal, R.: Carbon emission from farm operations. Environ. Int., 30, 981–990, 2004. Lambe, N.R., Navajas,
   E.A, McLean, K.A., Simm, G., Bünger, L.: Changes in carcass traits during growth in lambs of two contrasting breeds, measured using computer tomography, Livestock Science, 107, 37–52, 2007.

Laws, J. A., Falge, E., Gu, L., Baldocchi, D. D., Bakwin, P., Berbigier, P., Davis, K.,

Dolman, A. J., Falk, M., Fuentes, J. D., Goldstein, A., Granier, A., Grelle, A., Hollinger, D., Janssens, I. A., Jarvis, P., Jensen, N. O., Katul, G., Mahli, Y., Matteucci, G., Meyers, T., Monson, R., Munger, W., Oechel, W., Olson, R., Pilegaard, K., Paw, K. T., Thorgeirsson, H., Valentini, R., Verma, S., Vesala, T., Wilson, K., and Wofsy, S.: Comparison of grassland management systems for beef cattle using self-contained farmlets: effects of contrasting nitrogen inputs and management strategies on nitrogen budgets, and herbage and animal production, Agr. Ecosyst. Environ., 80.3, 243-54, 2000.

Leahy, P., Kiely, G., and Scanlon, T. M.: Managed grasslands: A greenhouse gas sink or source? Geophys. Res. Lett., 31.20, 2004.

1990 Ledgard, S. F.: Nitrogen cycling in low input legume based agriculture, with emphasis on legume/grass pastures, Plant Soil, 228,1, 43–59, 2001.

Ledgard, S. F., Menneer, J.C., Dexter, M. M., Kear, M. J., Lindsey, S., Peters, J. S., and
 Pacheco, D.: A novel concept to reduce nitrogen losses from grazed pastures by administering soil
 nitrogen process inhibitors to ruminant animals: A study with sheep. Agr. Ecosyst. Environ., 125.1 4, 148-58, 2008.

<sup>1960</sup> 

2000	Lettens, S., Van Orshoven, J, Van Wesemael, B., De Vos, B., and Muys, B.: Stocks and fluxes of soil organic carbon for landscape units in Belgium derived from heterogeneous data sets for 1990 and 2000, Geoderma, 127.1-2, 11-23, 2005 a.
2005	Lettens, S., Van Orshoven, J., Van Wesemael, B., Muys, B., and Perrin, D.: Soil organic carbon changes in landscape units of Belgium between 1960 and 2000 with reference to 1990, Glob. Change Biol., 11.12, 2128-40, 2005b.
2003	Li, C. S., Frolking, S., and Frolking, T. A.: A Model of Nitrous Oxide Evolution from Soil Driven by Rainfall Events. 1. Model Structure and Sensitivity, J. Geophys. Res. atmos., 97.D9, 9759- 76, 1992.
2010	Li C.: Modeling trace gas emissions from agricultural ecosystems, Nutr. Cycl. Agroecosys. 58, 259, 276, 2000
	Luo, J., Tillman, R. W., and Ball, P. R.: Grazing effects on denitrification in a soil under pasture during two contrasting seasons, Soil. Biol. Biochem, 31.6, 903-12, 1999.
2015	MAFF, 1990. Ministry of agriculture fisheries and food. UK tables of nutritive value and chemical composition of feedingstuffs, 1990. 1st ed. Rowett Research. <u>MAFF. Guidelines for farmers</u> in NVZs. 32 pp. 1998. Ministry of Agriculture, Fisheries and
2020	<u>Food, London.</u> Marchand, R.F.: Ueber die chemische Zusammensetzung der Knochen. Journal für praktische Chemie 27, 83–97, 1842. DOI: 10.1002/prac.18420270117
2025	McTaggart, I. P., Clayton, H., Parker, J., Swan, L., and Smith, K. A.: Nitrous oxide emissions from grassland and spring barley, following N fertiliser application with and without nitrification inhibitors, Biol. Fert. Soils, 25.3, 261–68, 1997.
	MAFF. Guidelines for farmers in NVZs. 32 pp. 1998. Ministry of Agriculture, Fisheries and Food, London.
2030	R. M. McKenzie, J. N. Cape, M. Z. Ozel, J. Drewer, K. J. Dinsmore, J. F. Hamilton, E. Nemitz, M. A. Sutton, M. W. Gallagher, and U. Skiba.: The import and export of organic nitrogen species at a Scottish ombrotrophic peatland, Biogeosciences Discussions, 12, 515–554, 2015.
2035	McKenzie, R. M., Özel, M. Z., Cape, J. N., Drewer, J., Dinsmore, K. J., Nemitz, E., Hamilton, J. F., Sutton, M. A., Gallagher, M. W. and Skiba, U.: The import and export of organic nitrogen species at a Scottish ombrotrophic peatland, Biogeosciences Discussions, 12, 515-554, 2015.
	McTiernan, K. B., Jarvis, S.C., Scholefield, D., and Hayes, M. H. B.: Dissolved organic carbon losses from grazed grasslands under different management regimes, Water Res, 35.10, 2565-69, 2001.

2040	Molina-Herrera S., Haas, E., Klatt, S., Kraus, D., Augustin, J., Magliulo, V., Tallec, T., Ceschia, E.,
	Ammann C., Loubet, B., Skiba, U., Jones, S., Brümmer, C., Butterbach-Bahl, K., Kiese, R.: A modelling
	study on mitigation of N <sub>2</sub> O emissions and NO <sub>3</sub> leaching at different agricultural sites across Europe
	using LandscapeDNDC, Science of the Total Environment,
	553, 128–140, 2016.

2045

- Mudge, P. L., Wallace, D. F., Rutledge, S., Campbell, D. I., Schipper, L. A., Hosking, C. L.: Carbon balance of an intensively grazed temperate pasture in two climatically contrasting years, Agr. Ecosyst. Environ., 144.1, 271-80, 2011.
- 2050 <u>Navajas, E. A., Richardson, R. I., Fisher, A. V., Hyslop, J. J., Ross, D. W., Prieto, N.,</u> <u>Simm, G. and Roehe, R.: Predicting beef carcass composition using tissue weights</u> <u>of a primal cut assessed by computed tomography, Animal, 4:11, pp 1810–1817, 2010.</u> <u>doi:10.1017/S1751731110001096</u>
- 2055 Neeteson, J. J. and Hassink, J.: Nitrogen Budgets of Three Experimental and Two Commercial Dairy Farms in the Netherlands. In: M.K. van Ittersum & S.C. van de Geijn (Eds.), Perspectives for Agronomy. Adopting ecological prinicples and managing resource use. Elsevier Science BV, Amsterdam. Developments in Crop Science 25, pp. 171-178. 1997.
- 2060 Nunez, R. P., Demanet, R., Alfaro, M., and Mora, M. L.: Nitrogen Soil Budgets in Contrasting Dairy Grazing Systems of Southern Chile, A Short Term Study, Rev. Cienc. Suelo Nutr., 10.2, 170-83, 2010.

2065 Oenema, O., Velthof, G.L., Yamulki, and S., Jarvis, S.C.: Nitrous oxide emissions from grazed grassland, Soil Use Manage., 13,4, 288 95, 1997.

Owens, L. B., Edwards, W. M., and Vankeuren, R. W.: Groundwater Nitrate Levels Under Fertilized Grass and Grass-Legume Pastures. J. Environ. Qual., 23.4, 752-58, 1994.

2070 Phoenix, G. K., Hicks, W.K., Cinderby, S., Kuylenstierna, J.C.I., Stock, W. D., Dentener, F. J., Giller, K. E., Austin, A. T., Lefroy, R. D. B., Gimeno, B. S., Ashmore, M. R., Ineson, P.: Atmospheric nitrogen deposition in world biodiversity hotspots: the need for a greater global perspective in assessing N deposition impacts. Global Change Biology, 12, pp. 470–476, 2006.

2075

Powlson, D.S., Whitmore, A.P. and Goulding, K.W.T.: Soil carbon sequestration to mitigate climate change: A critical re-examination to identify the true and the false. European Journal of Soil Science, 62, 42-55, 2011.

2080 Prescher, A. K., Grunwald, T. and Bernhofer, C.: Land use regulates carbon

budgets in eastern Germany: From NEE to NBP, Agr. Forest Meteorol., 150.7-8, 1016-25, 2010.

Rees, R. M., Bingham, I. J., Baddeley, J. A., and Watson, C.A.: The role of plants and land management in sequestering soil carbon in temperate arable and grassland ecosystems. Geoderma, 128.1-2, 130-54, 2005.

Rees, R. M., Bingham, I. J.; Baddeley, J. A.; and Watson, C.A.: Nitrous oxide mitigation in UK agriculture. Soil Sci. Plant Nutr., 59.1, 3-15, 2013.

- 2090 Reichstein, M., Falge, E., Baldocchi, D., Papale, D., Aubinet, M., Berbigier, P., Bernhofer, C., Buchmann, N., Gilmanov, T., Granier, A., Grunwald, T., Havrankova, K., Ilvesniemi, H., Janous, D., Knohl, A., Laurila, T., Lohila, A., Loustau, D., Matteucci, G., Meyers, T., Miglietta, F., Ourcival, J. M., Pumpanen, J., Rambal, S., Rotenberg, E., Sanz, M., Tenhunen, J., Seufert, G., Vaccari, F., Vesala, T., Yakir, D., and Valentini, R.: On the separation of net ecosystem exchange into assimilation and ecosystem respiration: review and improved 2095
  - algorithm, Global Change Biology, 11.9, 1424-39, 2005.

Rose Marie Rytter, R. M., Rytter, L., Hogbom, L.: Carbon sequestration in willow (Salix spp.) plantations on former arable land estimated by repeated field sampling and C budget calculation. Biomass and Bioenergy, 83, 483-492, 2015.

Roche J.: The International Wool Trade. Woodhead Publishing, 1995.

Rudaz, A. O., Walti, E., Kyburz, G., Lehmann, P., and Fuhrer, J.: Temporal variation in N<sub>2</sub>O and N-2 2105 fluxes from a permanent pasture in Switzerland in relation to management, soil water content and soil temperature, Agr. Ecosyst. Environ., 73.1, 83-91, 1999.

Ryden, J. C., Ball, P. R., and Garwood, E. A.: Nitrate Leaching from Grassland, Nature 311.5981, 50-53, 1984a.

2110

2100

2085

Ryden, J. C., and McNeill, J. E.: Application of the micrometeorological mass balance method to the determination of ammonia loss from a grazed sward, J. Sci. Food Agric., 35, 1297-310. <del>1984.</del>

Rytter, R, M., Rytter, L., Hogbom, L.: Carbon sequestration in willow (Salix spp.) plantations on former arable land estimated by repeated field sampling and C budget calculation. Biomass and Bioenergy, 83, 2115 483-492, 2015.

Schipper, L. A., Baisden, W. T., Parfitt, R. L., Ross, C., Claydon, J. J., and Arnold, G.: Large losses of soil C and N from soil profiles under pasture in New Zealand during the past 20 years, Glob. Change Biol, 13.6, 1138-44, 2007.

2120

Scholefield, D., Lockyer, D. R., Whitehead, D. C., and Tyson, K. C. A.: A Model to Predict

Transformations and Losses of Nitrogen in Uk Pastures Grazed by Beef-Cattle, Plant Soil, 132.2, 165-77, 1991.

2125

Scholefield, D., Tyson, K. C., Garwood, E.A., Armstrong, A.C., Hawkins, J., and Stone, A. C.: Nitrate Leaching from Grazed Grassland Lysimeters - Effects of Fertilizer Input, Field Drainage, Age of Sward and Patterns of Weather, J. Soil Sci., 44.4, 601-13, 1993.

2130 Schuman, G. E., Janzen, H. H., and Herrick, J. E.: Soil carbon dynamics and potential carbon sequestration by rangelands, Environ. Pollut., 116.3, 391-96, 2002.

Schrumpf, M., Schulze, E. D., Kaiser, K., and Schumacher, J.: How accurately can soil organic carbon stocks and stock changes be quantified by soil inventories? Biogeosciences, 8, 1193-1212, 2011.

2135 1

Schulze, E.D., Luyssaert, S., Ciais, P., Freibauer, A., Jannsens, I.A., Soussana, J.F., Grace, J., Levin, I., Thiruchittampalam, B., Heimann, M., Dolman, A.J., Valentini, R., Bousquet, P., Peylin, P., Peters, W., Rödenbeck, C., Etiope, G., Vuichard, N., Wattenbach, M., Nabuurs, G.J., Poussi, Z., Nieschulze, J., Gach, J.H.: Importance of methane and nitrous oxide for Europe's terrestrial greenhouse-gas balance. Nature Geoscience 2: 842-850, 2009.

Scottish Agricultural College, Farm management handbook SAC; edt. Linda Chadwick, Edinburgh, 1995.

2145

2140

Scottish Agricultural College, Technical Note TN652: Fertiliser recommendations for grasslands. A., Sinclair, P.A., Shipway, B., Crooks, 2013.

Siemens, J.: The European carbon budget: A gap, Science, 302.5651, 1681, 2003.

2150 Simpson, D., Fagerli, H., Jonson, J. E., Tsyro, S., Wind, P., and Tuovinen, J.-P.: Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe. Part I: Unified EMEP Model Description. EMEP Status Report 2003, ISSN 0806-4520, Det . 2003. Meteorologisk Institutt, Oslo, 2003.

2155 Skiba, U., Drewer, J., Tang, Y. S., van Dijk, N., Helfter, C., Nemitz, E., Famulari, D., Cape, J. N., Jones, S. K., Twigg, M., Pihlatie, M., Vesala, T., Larsen, K. S., Carter, M. S., Ambus, P., Ibrom, A.,Beier, C., Hensen, A., Frumau, A., Erisman, J. W., Br<sup>\*</sup>uggemann, N.,Gasche, R., Butterbach-Bahl, K., Neftel, A., Spirig, C., Horvath, L., Freibauer, A., Cellier, P., Laville, P., Loubet, B., Magliulo, E., Bertolini, T., Seufert, G., Andersson, M., Manca, G., Laurila, T., Aurela, M., Lohila A., Zechmeister-Boltenstern, S., Kitzler, B., Schaufler, G., Siemens, J., Kindler, R., Flechard, C., and Sutton, M. A.: Biosphere atmosphere exchange of reactive nitrogen and greenhouse gases at the NitroEurope core flux measurement sites: Measurement strategy and first annual data set, Agr. Ecosys. Environ., 133, 139– 149, 2009

2165	<u>Skiba U, Drewer J, Tang Y, van Dijk N, Helfter C, Nemitz E, Famulari D, Cape J, Jones S,</u>
	Twigg M.:. Biosphere-atmosphere exchange of reactive nitrogen and greenhouse gases at the
	NitroEurope core flux measurement sites: Measurement strategy and first data sets. Agriculture,
	Ecosystems & Environment 133 (3-4): 139-149, 2009. doi: 10.1016/j.agee.2009.05.018

2170 Skiba, U., Jones, S. K., Drewer, J., Helfter, C., Anderson, M., Dinsmore, K., McKenzie, R., Nemitz, E., and Sutton, M. A.: Comparison of soil greenhouse gas fluxes from extensive and intensive grazing in a temperate maritime climate, Biogeosciences 10, 1231-1241, 2013.

 Smith, R. I., Fowler, D., Sutton, M. A., Flechard, C., and Coyle, M.: Regional estimation of pollutant
 gas dry deposition in the UK: model description, sensitivity analyses and outputs. Atmos. Environ., 34,22, 3757-77, 2000.

Smith, P. : How long before a change in soil organic carbon can be detected? Glob. Change Biol., 10.11, 1878-83, 2004

2180

Smith, P., Haberl, H., Popp, A., Erb, K.-H., Lauk, C., Harper, R., et al.: How much land based greenhouse gas mitigation can be achieved without compromising food security and environmental goals? Global Change Biology, 19, 2285–2302, 2013.

2185 Smith, P.: Do grasslands act as a perpetual sink for carbon? Glob. Change Biol., 20, 2708–2711, 2014.

Soussana, J. F., Loiseau, P., Vuichard, N., Ceschia, E., Balesdent, J., Chevallier, T., and Arrouays, D.: Carbon cycling and sequestration opportunities in temperate grasslands, Soil Use Manage., 20, 219-30, 2004.

2190

Soussana, J. F., Allard, V, Pilegaard, K., Ambus, P., Amman, C., Campbell, C., Ceschia, E., Clifton-Brown, J., Czobel, S., Domingues, R., Flechard, C., Fuhrer, J., Hensen, A., Horvath, L., Jones, M., Kasper, G., Martin, C., Nagy, Z., Neftel, A., Raschi, A., Baronti, S., Rees, R.M., Skiba, U., Stefani, P., Manca, G., Sutton, M., Tubaf, Z., and Valentini, R.: Full accounting of the greenhouse gas (CO<sub>2</sub>, N<sub>2</sub>O, CH<sub>4</sub>) budget of nine European grassland sites, Agr. Ecosyst. Environ., 121.1-2, 121-34, 2007.

Soussana, J. F., Tallec, T., and Blanfort, V.: Mitigating the greenhouse gas balance of ruminant production systems through carbon sequestration in grasslands, Animal, 4.3, 334-50, 2010.

2200 Soussana, J. F. and Lemaire, G.: Coupling carbon and nitrogen cycles for environmentally sustainable intensification of grasslands and crop-livestock systems, Agr. Ecosyst. Environ., 190 9–17, 2014.

Stange F., Butterbach Bahl K., Papen H., Zechmeister Boltenstern S., Li C., and Aber J.: A
 process oriented model of N<sub>2</sub>O and NO emissions from forest soils: 2: sensitivity analysis and

validation, J. Geophys. Res., 105, 4385-4398, 2000.

Stewart, A. A., Little, S. M., Ominski, K.H., Wittenberg, K. M., and Janzen, H. H.: Evaluating greenhouse gas mitigation practices in livestock systems: an illustration of a whole-farm approach, J. Agr. Jci., 147, 367-82, 2009.

Streeter, J.: Inhibition of Legume Nodule Formation and N-2 Fixation by Nitrate, Cre. Cr. Rev. Plant Sci., 7.1, 1-23, 1988.

Sutton, M. A., Tang, Y. S., Miners, B., and Fowler, D.: A New Diffusion Denuder System for Long-Term, Regional Monitoring of Atmospheric Ammonia and Ammonium, Water Air Soil Poll.: Focus, 1.5-6, 145-56, 2001.

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

Sutton M.A., Oenema, O., Erisman, J.W., Leip, A., van Grinsven, H. and Winiwarten, W.: Too much of a good thing. Nature 472, 159-161, 2011. doi:10.1038/472159a

2230

2210

Tang, Y. S., Simmons, I.; van Dijk, N., Di Marco, C., Nemitz, E., Dammgen, U., Gilke, K., Djuricic, V., Vidic, S., Gliha, Z., Borovecki, D., Mitosinkova, M., Hanssen, J. E., Uggerud, T. H., Sanz, M.J., Sanz, P., Chorda, J.V., Flechard, C. R., Fauvel, Y., Ferm, M., Perrino, C., and Sutton, M.A.: European scale application of atmospheric reactive nitrogen measurements in a low cost approach to infer dry deposition fluxes, Agr. Ecosyst. Environ., 133.3 4, 183 95, 2009.

2240 Tuovinen, J. P., Emberson, L., and Simpson, D.: Modelling ozone fluxes to forests for risk assessment: status and prospects, Ann. For Sci., 66.4, 2009.

Tyson, K. C., Garwood, E. A., Armstrong, A.C., and Scholefield, D.: Effects of Field Drainage on the Growth of Herbage and the Liveweight Gain of Grazing Beef-Cattle, Grass Forage Sci., 47.3, 290-301, 1992.

2245

Twigg, M. M., House, E., Thomas, R., Whitehead, J., Phillips, G.J., Famulari, D., Fowler, D., Gallagher, M.W., Cape, J.N., Sutton, M.A., Nemitz, E.: Surface/atmosphere exchange and chemical
2250	Meteorology, Volume 151, Issue 12, , Pages 1488-1503, 2011.	
2250	Van der Hoek, K. W.: Nitrogen efficiency in global animal production, Environ. Pollut., 102, 127-32, 1998.	
2255	Van der Salm, C., Dolfing, J., Heinen, M., and Velthof, G. L.: Estimation of nitrogen losses via denitrification from a heavy clay soil under grass, Agr. Ecosyst. Environ., 119, 311-319, 2007.	
2260	Velthof, G. L. and Oenema, O.: Nitrous oxide fluxes from grassland in the Netherlands .2. Effects of soil type, nitrogen fertilizer application and grazing, Eur. J. Soil Sci., 46.4, 541-49, 1995.	
2200	Velthof, G. L. and Oenema, O.: Effects of nitrogen fertilization and grazing on the emission of nitrous oxide from grassland. 1997. RIVM Dutch National Research Programme on Global Air Pollution and Climate Change, Velthof, G.L. & Oenema, O. (eds.), Bilthoven (1997) Report no. 410 100 055.	
2265	Waring, R. H., Landsberg J. J., and Williams, M.: Net primary production of forests: a constant fraction of gross primary production? Tree Physiol., 18.2, 129–34, 1998.	
2270	Watson, C. J., -Jordan, C., Kilpatrick, D., McCarney, B., and Stewart, R.: Impact of grazed grassland management on total N accumulation in soil receiving different levels of N inputs. Soil Use Manage., 23.2, 121-128, 2007.	
2275	Whitehead, D. C., Pain, B. F., and Ryden J. C.: Nitrogen in UK grassland agriculture, J. R. Agric. Soc., 147, 190-201. 1986.	
2213	Wolf, B., Kiese R., Chen W.W., Grote R., Zheng X. H., and Butterbach Bahl K.: Modeling N <sub>2</sub> O emissions from steppe in Inner Mongolia, China, with consideration of spring thaw and grazing intensity, Plant Soil, 350, 297–310, 2012.	
2280	Zhang, L. M., Gong, S.L., Padro, J., and Barrie, L.: A size segregated particle dry deposition scheme for an atmospheric aerosol module, Atmos. Environ., 35.3, 549-60, 2001.	
2285	Zhang, L., Brook, J. R. and Vet R.: A revised parameterization for gaseous dry deposition in air quality models. Atmos. Chem. Phys., 3, 2067–82, 2003.	Formatted: Normal Indent: Left:
2205	Zhang, Y., Xu, M., Chen, H. and Adams, J.: Global Pattern of NPP to GPP Ratio Derived from MODIS Data: Effects of Ecosystem Type, Geographical Location and Climate. Global Ecology and Biogeography 18 (3): 280–90, 2009, doi:10.1111/j.1466-8238.2008.00442 x	cm, Hanging: 1.27 cm, Tab stops: ( cm, Left

2290	Table 1. Average annual	stocking <del>livestock</del>	rates <del>densities</del>	$[LSU^{+}ha^{-1}]$	$y^{-1}$ ].
				L	

			2004	2007	2005	2007		• • • • •	
	2002	2003	2004	2005	2006	2007	2008	2009	2010
Heifers	0.12	0.38	0.05	0.15	0.27	0	0	0	0
Ewes	0.14	0.16	0.82	0.56	0.51	0.68	0.68	0.61	0.53
Lambs	0	0	0.12	0.12	0.12	0.23	0.14	0.11	0.12
all animals	0.27	0.54	0.99	0.83	0.90	0.91	0.83	0.72	0.65
*									

<sup>\*</sup>LSU stands for Live stock units

 Table 2. Weather characteristics of each measurement year.

$\gamma$	$\gamma$	n	4
4	4	7	•

	2002	2003	2004	2005	2006	2007	2008	2009	2010
Annual mean temperature [°C]	9.2	9.6	8.9	8.8	9.3	9.1	8.6	8.9	8.3
Maximum temperature [°C]	23.6	29.5	27.4	30.4	26.6	21.4	23.5	28.0	24.0
Minimum temperature [°C]	-5.1	-8.4	-4.9	-6.6	-5.5	-7.5	-5.8	-7.8	-10.3
Annual rainfall [mm]	1238	680	1169	1028	1120	904	1065	744	575
Soil water content [% by volume]	36.9	31.0	40.3	45.2	36.6	37.7	41.5	39.4	-
Water filled pore space [%]	68.0	57.2	74.3	83.3	67.5	69.5	76.5	72.6	-
Length of growing season <sup>*</sup> [days]	180	196	156	177	151	186	193	242	226

The plant growing season begins and ends with periods of consecutive days, where daily temperatures average more than 5 °C without any five-day spells of temperatures below 5 °C.

Tab	lo 3 Systematic	uncortaintios	attributed to a	ach hudget e	omponent	Combined 1	incortaintiac	Woro
100	ie o. bystematic	uncertainties	annouled to c	ach buuget e	omponent.	<del>comonica a</del>	meentainties	were
230010	ulated according	<del>r to simple Ga</del>	ussian error n	ronagation ru	les			
			assian enter p	opuguiton tu				

Nitrogen budget component	<del>N [%]</del>	Carbon budget component	<del>C [%]</del>
Mineral fertiliser	1		
Organic manure <sup>*</sup>	<del>20</del>	Organic manure <sup>*</sup>	<del>20</del>
Harvest <sup>b</sup>	<del>16</del>	Harvest <sup>b</sup>	++
Leaching	<del>30</del>	Leaching	<del>30</del>
Animal (wool and meat) <sup>e</sup>	<del>12</del>	Animal (wool and meat) <sup>#</sup>	<del>12</del>
Wet deposition	<del>30</del>	<del>CH<sub>4</sub> soil</del>	<del>160</del>
Dry deposition <sup>d</sup>	<del>80</del>	CH <sub>4</sub> -enteric	<del>20</del>
N₂⊖	<del>30</del>	CH <sub>4</sub> -excretion	<del>20</del>
NOx soil	<del>30</del>	<del>CH<sub>4</sub> organic</del>	<del>120</del>
NH <sub>4</sub> volatilisation	<del>30</del>	-	
NOx volatilisation	<del>50</del>		
N	30		

<sup>#</sup>combined uncertainties of C and N analysis (17%) and volume spread (10%)

<sup>b</sup>eombined uncertainty of total C (4%) and N (12%) analysis and farmer's estimate in harvest amount (10%)

<sup>e</sup>combined uncertainties from animal numbers (5%), animal weight (10%) and literature values for C and N content for meat and 2305 wool (3%)

<sup>d</sup>combined uncertainty of DELTA sample analysis (7%) and variation of outputs from the four models (80%)

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**Table 43.** Nitrogen budget and balance for each measurement year and average values, confidence intervals at p > 0.95 (CI) and systematic uncertainties (uncert.) for 2002-2010 [g N m<sup>-2</sup> y<sup>-1</sup>]. Negative numbers represent uptake while positive numbers represent loss of N from this grassland ecosystem. Letters indicate data published in previous publications.

	2002	2003	2004	2005	2006	2007	2008	2009	2010	20	002-201	0
										average	CI	uncert.
Organic fertilisation	0	0	-6.9	-15.8	0	0	0	0	0	-2.5	3.6	0.2
Inorganic fertilisation	-20.6	-16.0	-11.0	-17.3	-22.4	-17.3	-25.9	-25.0	-19.0	-19.4	3.1	0.2
Wet deposition	-0.4	-0.6	-0.6	-0.7	-0.6	-0.6	-0.5	-0.4	-0. <u>4</u> 5*	-0.5	0.1	0.2
<sup>a</sup> Dry deposition	-0.5	-0.4	-0.3	-0.3	-0.2	-0.3	-0.2	-0.2	-0. <del>3</del> 2*	-0.3	0.1	0.2
Harvest	5.0	4.7	0	0	0	0	0	0	0	1.1	1.4	0.2
Meat (incl. bones)											<u>0.7</u> 0	
	<u>0.2</u> 0.2	<u>0.6</u> 0.5	<u>2.3</u> 1.9	<u>3.1<del>2.6</del></u>	<u>2.6<del>2.2</del></u>	<u>2.9</u> 2.4	<u>1.8</u> 1.5	<u>1.3</u> 1.1	<u>1.5</u> 1.2	<u>1.80</u> 1.5	<del>.5</del>	0.2
Wool	0	0	0.6	0.5	0.4	0.7	0.7	0.2	0.2	0.4	0.2	0.0
<sup>b</sup> Leaching	1 <u>4.9</u> 5.								<u>5.0</u> 5.3			
<b>N</b>	0	0.1	0.1	4.6	10.6	4.2	5.6	2.6	*	5.3	3.4 <u>1</u>	<del>1.6<u>1.71</u></del>
$N_2$	27	2.2	1.2	17	20	2.0	2.2	4.1	<u>3.6</u> 2.8	280	0.6	0.8
<sup>c</sup> N <sub>2</sub> O	5.7	2.2	1.5	1./	2.0	5.0	5.5	4.1		2. <del>6 <u>9</u></del>	0.0	0.8
NO (soil)	1.1	0.1	0.1	0.4	0.9	1.5	0.8	0.4	0.4	0.0	0.3	0.2
	0.3	0.1	0	0.1	0.2	0.2	0.3	0.1	0.1	0.2	0.1	0.1
$NO_x, NH_3$ (inorg.fert.)	2.1	1.6	1.1	1.7	2.2	1.7	2.6	2.5	1.9	1.9	0.3	0.6
NO <sub>x</sub> ,NH <sub>3</sub> (org.fert.)	0	0	1.4	3.2	0	0	0	0	0	0.5	0.7	0.3
NO <sub>x</sub> NH <sub>3</sub> (excretion)	<u>0.5</u> 0.4	<u>0.8</u> 0.7	<u>1.5</u> 1.7	<u>1.6<del>1.3</del></u>	<u>1.5</u> 1.3	<u>2.4</u> 1.6	<u>2.1</u> 1.5	<u>1.6</u> 1.3	<u>1.5<del>1.2</del></u>	1. <u>5</u> 2	0. <u>4</u> 3	0. <del>6</del> 7
N balance	-	<u>-6.9</u> -	<u>-10.4</u> -	<u>-17.2</u> -	<u>-1.9</u> -	<u>-1.9</u> -	<u>-9.5</u> -	<u>-12.7</u> -	<u>-5.6</u> -			
	<u>6.4<del>6.3</del></u>	<del>7.0</del>	<del>10.6</del>	<del>17.9</del>	<del>2.5</del>	<del>3.1</del>	<del>10.3</del>	<del>13.2</del>	<del>6.6</del>	<u>-6.6</u> 7.2	4. <u>4</u> 6	2. <u>2</u> 1

\*average value of 2002-2009

<sup>a</sup>Flechard et al. 2011: Dry deposition, modelled average value of the two years 2007/2008

<sup>b</sup>Mo ina-Herrera et al. 2016: N leaching modelled 2005-2010

<sup>c</sup>Molina-Herrera et al. 2016: N<sub>2</sub>O fluxes modelled 2005-2010.

<sup>c</sup>Di Marco et al. 2004: N<sub>2</sub>O fluxes measured by eddy covariance (half hourly) June 2002 to June 2003

<sup>c</sup>Jones et al. 2011: N<sub>2</sub>O fluxes measured by eddy covariance (half hourly) and chambers (hourly) during measurement campaigns in June 2003, March/May/July 2007 and May/July 2008.

Flechard et al. 2007: annual N<sub>2</sub>O fluxes measured by eddy covariance in 2002/2003 and by chambers in 2004

<sup>c</sup>Skiba et al. 2013: annual N<sub>2</sub>O fluxes measured by chambers from Jan. 2007 – Sept. 2010

**Table 54.** Carbon budget and balance for each measurement year and average values, confidence intervals at p > 0.95 (CI) and systematic uncertainties (uncert.) for 2002-2010 [g C m<sup>-2</sup> y<sup>-1</sup>]. Negative numbers represent uptake, while positive numbers represent loss of C from the grassland ecosystem. Letters indicate data published in previous publications.

	2002	2003	2004	2005	2006	2007	2008	2009	2010	2	2002-2010	)
										average	CI	uncert.
GPP	-2162.9	-1982.0	-2111.4	-1662.4	-982.1	-1722.7	-1441.2	-1722.4	-2015.4	-1755.8	244.4	105.3
TER	1726.9	1725.9	2183.2	1638.5	972.1	1606.7	1324.0	1116.7	1547.0	1537.9	236.2	92.3
NPP	-1081.5	-991.0	-1055.7	-831.2	-491.1	-861.3	-720.6	-861.2	-1007.7	-877.9	122.2	-52.8
<sup>a</sup> CO <sub>2</sub> (NEE)	-436.0	-256.1	71.8	-24.0	-10.0	-115.9	-117.1	-605.7	-468.4	-217.9	154.5	80.0
Organic fert.	0.0	0.0	-55.4	-171.8	0.0	0.0	0.0	0.0	0.0	-25.2	37.8	5.0
Harvest	270.6	169.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	48.9	65.5	5.4
Meat (incl. bones)			<u>11.4</u> 11.	<u>15.6</u> 15.	<u>12.9</u> 4	<u>14.3</u> 14.						
	<u>0.9</u> 0.9	<u>2.9</u> 3.0	5	8	<del>3.1</del>	5	<u>9.0</u> 9.1	<u>6.3</u> 6.4	<u>7.3</u> 7.3	9. <mark>1-0</mark>	3. <mark>4</mark> 3	1 <del>.1</del> 0
Wool	0.0	0.0	1.7	1.5	1.3	2.1	2.0	0.7	0.5	1.1	0.5	0.1
<sup>b</sup> Leaching <u>*</u>									<del>16.4<u>14.</u></del>			4 <del>.9<u>5.2</u></del>
	25.1	7.0	22.1	18.7	19.4	15.4	17.0	6.8	<u>3*</u>	16.4	4.3	<u>6</u>
CH <sub>4</sub> (organic fert.)	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
<sup>c</sup> CH <sub>4</sub> (soil)	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.1
CH <sub>4</sub> (excretion)	0.0	0.1	0.2	0.1	0.2	0.2	0.1	0.1	0.1	0.1	0.0	0.0
CH <sub>4</sub> (enteric ferm.)	1.5	3.2	5.7	4.8	5.2	5.2	4.8	4.1	3.8	4.3	0.8	0.9
C balance (NBP)	<u>-137.8</u> -	<u>-73.3</u> -	<u>57.6<del>57.</del></u>	<u>-154.9</u> -	<u>29.1</u> 2	<u>-78.7</u> -	<u>-84.1</u> -	<u>-587.7</u> -	<u>-440.3</u> -	<u>-163.3</u> -	<u>139.5</u> 1	<u>16.0</u> 15
	<del>137.8</del>	73.3	7	154.7	<del>9.3</del>	<del>78.6</del>	<del>84.0</del>	<del>587.6</del>	<del>440.3</del>	<del>163.2</del>	<del>39.5</del>	<del>.</del> .

\*average value of 2002 2009

<sup>a</sup>Soussana et al. 2007: NEE July 2002- Dec 2004

<sup>a</sup>Skiba et al. 2013: NEE 2007-2010

<sup>a</sup>Kindler et al. 2011: NEE average multiyear value 2004-2007

<sup>b</sup>Kindler et al. 2011: C leaching losses October 2006- Sept 2008 (Slope value corresponds to data used in this publication).

<sup>c</sup>Skipa et al. 2013: CH<sub>4</sub> (soil) 2007-2010

	N <sub>2</sub> O flux	Total N input	EF
	$[g N m^{-2} y^{-1}]$	$[g N m^{-2} y^{-1}]$	[%]
2002	1.14	20.60	5.5
2003	0.14	15.98	0.9
2004	0.11	11.00	0.6
2005	0.36	17.25	1.1
2006	0.88	22.43	3.9
2007	1.25	17.25	7.2
2008	0.84	25.93	3.2
2009	0.41	24.95	1.6
2010	0.35	18.98	1.9

**Table 65**. Annual N<sub>2</sub>O exchange, total N input by fertiliser (mineral and organic) and N<sub>2</sub>O emission factors, expressed as percentage of total N inputs in 2002-2010.

**Table 76.** <u>N and C soil stocks (g N or C m<sup>-2</sup>) in May 2004 and May 2011 and N and C budgets</u> (g N or C m<sup>-2</sup> y<sup>-1</sup>) over 7 years based on repeated soil N and C stock inventories (May 2004 and May 2011) and flux budget calculations (January 2004 - December 2010). Soil stock changes are based on a soil mass of 800 kg m<sup>-2</sup>, which corresponds to approximately 60 cm depth. The flux budgets are averages for the years 2004 – 2010 (Table 4 & 5). Numbers in brackets represent confidence intervals. Negative numbers are sinks.

	N-balance	-C-balance
soil stock change	<del>4.51 (2.64)</del>	<del>-29.08 (38.19)</del>
flux budget	<del>-9.20 (4.10)</del>	<del>-180.7 (180)</del>

	<u>Nitrogen</u>	<u>Carbon</u>
soil stocks in 2004	<u>840.68 (11.89)</u>	<u>12026.05 (190.19)</u>
soil stocks in 2011	870.02 (14.14)	<u>11824.87 (187.84)</u>
soil stock change	<u>-4.51 (2.64)</u>	29.08 (38.19)
flux budget	<u>-8.44 (4.21)</u>	<u>-179.7 (180)</u>



**Figure 1**. <u>StockingLivestock</u> density (ea), nitrogen (eb) and carbon (bc) input and export from inorganic and organic fertiliser and harvest from 2002-2010. <u>LSU stands for livestock unit</u>, where 1 livestock unit has a standard live weight of 600 kg head<sup>-1</sup>.







**Figure 32.** Mean annual nitrogen budget for Easter Bush, showing the fate of total N input (fertiliser and deposition) in (a) years when harvested for silage (2002 and 2003) and (b) in years when only grazing took place (2004–2010). The residual term includes all the error in the budget calculation, as well as any net accumulation of soil organic nitrogen.



**Figure 43.** Mean annual carbon budget for Easter Bush, showing the fate of net primary productivity (NPP) in (a) years when harvested for silage (2002 and 2003) and (b) in years when only grazing took place (2004–2010). Heterotrophic respiration includes the respiration of soil microbes, cows and sheep.

The residual term includes all the error in the budget calculation, as well as any net accumulation of soil organic carbon.



**Figure 54.** Average greenhouse gas fluxes, net GHG exchange (NGHGE) and attributed net GHG balance (NGHGB, includes  $FC_{org fert}$ ,  $FC_{animal}$ ,  $FC_{leaching}$ ,  $FC_{harvest}$ ) for 2002-2010. Positive values correspond to losses and negative values to storage of greenhouse gases to and from the grassland system, respectively. The CH<sub>4</sub> component comprises CH<sub>4</sub> fluxes from enteric fermentation, animal excretion, slurry application and soil exchange, while the N<sub>2</sub>O component is the N<sub>2</sub>O flux from the soil. <u>CO<sub>2</sub> represent the Net Ecosystem Exchange (NEE)</u>. Global warming potentials of 298 and 25 were used for N<sub>2</sub>O and CH<sub>4</sub> respectively, using a time horizon of 100 yrs (IPCC, 2013). Thin error bars represent variations (confidence intervals at p > 0.95) between years, while thick error bars represent the systematic uncertainty of each value.