

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

3  
4 Stephanie K. Jones<sup>1,2</sup>, Carole Helfter<sup>1</sup>, Margaret Anderson<sup>1</sup>, Mhairi Coyle<sup>1</sup>, Claire Campbell<sup>1</sup>,  
5 Daniela Famulari<sup>1</sup>, Chiara Di Marco<sup>1</sup>, Netty van Dijk<sup>1</sup>, Cairistiona F.E. Topp<sup>2</sup>, Ralf Kiese<sup>3</sup>,  
6 Reimo Kindler<sup>4</sup>, Jan Siemens<sup>5</sup>, Marion Schrumpp<sup>6</sup>, Klaus Kaiser<sup>7</sup>, Eiko Nemitz<sup>1</sup>, Peter Levy<sup>1</sup>,  
7 Robert M. Rees<sup>2</sup>, Mark A. Sutton<sup>1</sup>, Ute .M. Skiba<sup>1</sup>

8  
9 1) Centre for Ecology and Hydrology, Edinburgh, Bush Estate, Penicuik, Midlothian EH26 QB, UK

10 2) Scotland's Rural College, King's Buildings, West Mains Road, Edinburgh, EH9 3JG, UK

11 3) Karlsruhe Institute of Technology, Institute for Meteorology and Climate Research, Atmospheric  
12 Environmental Research (IMK-IFU), Kreuzeckbahnstr. 19, 82467 Garmisch-Partenkirchen, D

13 4) Chair of Waste Management and Environmental Research, Technische Universität Berlin,  
14 Franklinstr. 29, 10587 Berlin, D

15 5) Institute of Crop Science and Resource Conservation, Soil Science, Universität Bonn, Nussallee 13,  
16 53115 Bonn, D

17 6) Max Plank Institute for Biogeochemistry, Hans-Knöll-Str. 10, 07745 Jena, D

18 7) Martin Luther University Halle-Wittenberg, Von-Seckendorff-Platz 3, 06120 Halle (Saale), D

19

20 *Correspondence to:* Stephanie K. Jones (Stephanie.Jones@sruc.ac.uk)

21

22

23

24

25

26

27

28

29

30

31

32

33

34

35

36

37 **Abstract**

38 Intensively managed grazed grasslands in temperate climates are globally important  
39 environments for the exchange of the greenhouse gases (GHGs) carbon dioxide (CO<sub>2</sub>), nitrous  
40 oxide (N<sub>2</sub>O) and methane (CH<sub>4</sub>). We assessed the N and C budget of a mostly grazed,  
41 occasionally cut, and fertilized grassland in SE Scotland by measuring or modelling all relevant  
42 imports and exports to the field as well as changes in soil C and N pools over time. The N  
43 budget was dominated by import from inorganic and organic fertilisers (21.9 g N m<sup>2</sup> yr<sup>-1</sup>) and  
44 losses from leaching (5.3 g N m<sup>2</sup> yr<sup>-1</sup>), N<sub>2</sub> emissions and NO<sub>x</sub> and NH<sub>3</sub> volatilisation (6.4 g N  
45 m<sup>2</sup> yr<sup>-1</sup>). The efficiency of N use by animal products (meat and wool) averaged 11%. On  
46 average over nine years (2002-2010) the balance of N fluxes suggested that  $7.2 \pm 4.6$  g N m<sup>-2</sup> y<sup>-1</sup>  
47 <sup>1</sup> (mean  $\pm$  confidence interval at  $p > 0.95$ ) were stored in the soil. The largest component of the  
48 C budget was the net ecosystem exchange of CO<sub>2</sub> (NEE), at an average uptake rate of  $218 \pm$   
49  $155$  g C m<sup>-2</sup> y<sup>-1</sup> over the nine years. This sink strength was offset by carbon export from the  
50 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  
51 terms, CH<sub>4</sub> emissions from the soil, manure applications and enteric fermentation were  
52 negligible and only contributed to 0.02-4.2 % of the total C losses. Only a small fraction of C  
53 was incorporated into the body of the grazing animals. Inclusion of these C losses in the budget  
54 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  
55 taken in May 2004 and May 2011 indicated that the grassland sequestered N in the 0-60 cm soil  
56 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.  
57 Potential reasons for the discrepancy between these estimates are probably an underestimation  
58 of C and N losses, especially from leaching fluxes as well as from animal respiration. The  
59 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  
60 strongly affected by CH<sub>4</sub> and N<sub>2</sub>O emissions. The GHG sink strength of the NEE was reduced  
61 by 54% by CH<sub>4</sub> and N<sub>2</sub>O emissions. Enteric fermentation from the ruminating sheep proved to  
62 be an important CH<sub>4</sub> source, exceeding the contribution of N<sub>2</sub>O to the GHG budget in some  
63 years.

64

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

67

68 **Introduction**

69 Nitrogen (N) is an essential component of proteins and genetic material and therefore required  
70 by all living organisms. Before N can be used by most organisms, inert atmospheric molecular  
71 nitrogen (N<sub>2</sub>) has to be transformed to reactive nitrogen (Nr). In an agricultural system Nr is  
72 added from inorganic fertiliser and cultivation-induced biological N fixation and as organic  
73 compounds from organic manure applications and dung from grazing animals. Generally N  
74 inputs into agricultural systems exceed outputs in the form of crops or animal off-takes (meat,  
75 milk and wool). In a steady state system the exceeding Nr is converted back to N<sub>2</sub> through  
76 complete denitrification (Galloway et al., 2003). However, in agricultural systems the surplus  
77 of Nr is generally only partly converted to N<sub>2</sub>, while the rest is lost to the atmosphere or aquatic  
78 ecosystems as Nr, causing various environmental problems.

79 Carbon (C) and N cycles in grasslands are intricately linked and tightly coupled in  
80 extensively managed low N grasslands, with sinks and sources in equilibrium. Converting  
81 such systems to intensively managed N fertilised grasslands in the short term may increase the  
82 soil organic carbon (SOC) pool from decomposed plant litter and root material as well as  
83 through rhizodeposition (Rees et al., 2005) until a new equilibrium is reached (Soussana and  
84 Lemaire, 2014). In the case of the Broadbalk experiment, Rothamsted, this equilibrium was  
85 achieved after 50 years (Powelson et al., 2011). After the conversion to intensive N  
86 management, the tight coupling of the N and C cycles becomes disrupted, leading to  
87 emissions of N<sub>2</sub>O and CH<sub>4</sub> at rates which may outweigh the benefits of C sequestration.  
88 Several studies indicate that managed grasslands can sequester C (Kim et al., 1992; Jones et  
89 al., 2006; Soussana et al., 2004; Ammann et al., 2007) however, uncertainties are high  
90 (Janssens et al., 2003). On the contrary, Smith (2014) concluded from long-term experiments  
91 and chronosequence studies, that changes in agronomic management may lead to short-term C  
92 sequestration, but in the long-term, under constant management and environmental  
93 conditions, C stocks are relatively stable. In a grassland ecosystem the C balance is  
94 determined by the net biome exchange (the difference between total C input and losses). In  
95 managed grassland ecosystems exports through biomass harvesting, the addition of organic  
96 manures (from organic fertiliser additions and animal excretion) as well as CO<sub>2</sub> and CH<sub>4</sub>  
97 losses from animal respiration and enteric fermentation can make significant contributions to  
98 the C budget.

99 Worldwide an estimated 26 % of land consists of managed grassland (FAOstat, 2008).  
100 The impact of Nr losses, C sequestration and GHG emissions (CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O) from  
101 managed grasslands on the environment is therefore of global importance and will become  
102 even more relevant in the future as an increased standard of living in developed countries is

103 expected to result in a rapid growth of livestock farming (Caro et al, 2014). Nutrient budgets  
104 are a valuable tool to summarise and understand nutrient cycling in agroecosystems and to  
105 assess their impact on the environment. As imbalances are not sustainable in the long term, N  
106 and C budgets can be used as indicators and regulatory policy instruments for nutrient  
107 management in order to reduce losses and increase efficiency. So far, different Nr species  
108 have been looked at in separate studies according to their form and impact. Few studies have  
109 attempted to calculate N budgets from managed grasslands (e.g. Ammann et al., 2009; Chen  
110 et al., 2004; Nunez et al., 2010, Kramberger et al., 2015), whereas C budgets have been  
111 assessed more often and are available for various ecosystems (e.g. Aubinet et al., 2000;  
112 Soussana et al., 2007; Ammann et al., 2007, Rytter et al. 2015). To calculate the total C and N  
113 budget of an ecosystem all import and export processes have to be assessed by measuring or  
114 estimating all imports and exports to an ecosystem. The other method is to measure  
115 differences in N and C stocks in the soil over time. This approach has the advantage that it  
116 requires the measurement of only a single component of the system. However, a large number  
117 of samples are needed at an interval of more than 5 years before detectable changes may be  
118 statistically significant (Smith, 2004). Moreover this approach does not provide any  
119 information about the different processes leading to the final budget.

120 In this study we assessed the C and N budget from an intensively managed grassland  
121 in Southern Scotland using both approaches. Here we report one of the most detailed analyses  
122 of C and N fluxes from a grassland ecosystem over 9 years (2002-2010). This study allowed  
123 an analysis of the importance of common grassland management practices such as cutting for  
124 silage, grazing of cattle and sheep at different stocking densities, N input by inorganic and  
125 organic fertiliser applications, as well as different weather conditions on the N, C and GHG  
126 balance. The data were collected in the frame of the three European projects GREENGRASS  
127 (EC EVK"-CT2001-00105), CarboEurope (GOCE-CT2003-505572) and NitroEurope  
128 (contract 017841).

129

## 130 **2. Methods**

### 131 **2.1 Site description**

132 The experimental site, Easter Bush, is located in South East Scotland, 10 km South of  
133 Edinburgh (03°02'W, 55°52' N, 190 m a.s.l). Mean annual rainfall (2002-2010) was 947 ±  
134 something mm and the mean annual temperature was 9.0 ± something °C. The field has been  
135 under permanent grassland management for more than 20 years with a species composition of  
136 >99% rye grass (*Lolium Perenne*) and < 0.5% clover (*Trifolium repens*). The soil type is an

137 imperfectly drained Macmerry soil series, Rowanhill soil association (Eutric Cambisol) with a  
138 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  
139 0.85 m depth on average and the main rooting zone extends down to 0.31 m below soil surface.

140

## 141 **2.2 Grassland management**

142 The grassland was grazed continuously throughout the experimental period by heifers in calf,  
143 ewes and lambs at different stocking density (Table 1 and Figure 1a). Animals were counted  
144 several times per week and it was assumed that the animal number stayed constant between  
145 observations. Livestock units used for heifers, ewes and lambs were 0.75, 0.10 and 0.04,  
146 respectively (1 livestock unit has a standard live weight of 600 kg head<sup>-1</sup> (Scottish Agricultural  
147 College, 1995). Lambs were present on the field from April to September only. The grass was  
148 cut for silage on the 1<sup>st</sup> of June and 8<sup>th</sup> of August 2002 and on the 29<sup>th</sup> of May 2003.  
149 Ammonium nitrate fertiliser was applied to the field 3-4 times per year, usually between March  
150 and July (56 kg N ha<sup>-1</sup> application<sup>-1</sup> on average). In 2008 an additional fifth mineral N  
151 application was applied, using urea instead of ammonium nitrate fertiliser. Organic manure was  
152 applied on the 28<sup>th</sup> of September 2004 and 27<sup>th</sup> of March 2005 as cattle slurry, using a vacuum  
153 slurry spreader. Rates of N and C input from fertiliser and manure and export from harvest are  
154 shown in Table 4 and 5 and in Fig. 1 a) and b).

155

## 156 **2.3. Annual budget calculations**

157 We assessed the N and C budget by measuring or estimating the import and export of all  
158 relevant fluxes to and from the grassland field on an annual basis. Throughout the manuscript  
159 all fluxes are presented following the sign convention used in micrometeorology; fluxes from  
160 the ecosystem to the atmosphere are positive (exported from the field), while negative values  
161 indicate fluxes from the atmosphere to the ecosystem (imported to the field). We set the system  
162 boundary for inputs and exports of N and C by the field perimeters (covering an area of 5.4 ha).  
163 The balance of all imports and exports results in the observed changes of N and C on this field  
164 over time.

165

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

168

$$\begin{aligned} 169 \Delta N / \Delta t = & \text{FN}_{\text{org fert.}} + \text{FN}_{\text{synt fert.}} + \text{FN}_{\text{N}_2 \text{ (biol. fixation)}} + \text{FN}_{\text{dep.}} + \\ 170 & \text{FN}_{\text{harvest}} + \text{FN}_{\text{animal}} + \text{FN}_{\text{leaching}} + \text{FN}_{\text{NH}_3/\text{NO}_x \text{ (fert., manure, animal)}} + \\ 171 & \text{FN}_{\text{NO}_x \text{ (soil)}} + \text{FN}_{\text{N}_2\text{O}} + \text{FN}_{\text{N}_2 \text{ (denitr.)}} \end{aligned} \quad (1)$$

172  
173 N imports include the addition of N from organic and inorganic fertiliser ( $FN_{org\ fert.} + FN_{synt\ fert.}$ ),  
174 the fixation of  $N_2$  through biological fixation ( $FN_{N_2\ (biol.\ fixation)}$ ) and the deposition of  $NH_3$ ,  
175  $HNO_3$ ,  $NH_4^+$ ,  $NO_3^-$  from dry, and  $NH_4^+$  and  $NO_3^-$  from wet deposition (summarised as  $FN_{dep.}$ ).  
176 Exports include the N lost from plant biomass at cuts for silage ( $FN_{harvest}$ ), the off-take of N in  
177 meat and wool from animals ( $FN_{animal}$ ), the loss of organic and inorganic dissolved N through  
178 leaching ( $FN_{leaching}$ ), the  $NH_3$  and  $NO_x$  emissions from volatilisation of inorganic and organic  
179 fertiliser spreading as well as from animal excretion ( $FN_{NH_3/NO_x(fert.,\ manure,\ animal)}$ ), the emission  
180 of  $NO_x$  from the soil ( $FN_{NO_x(soil)}$ ), the emission of  $N_2O$  from the soil ( $FN_{N_2O}$ ) and the loss of  $N_2$   
181 from total denitrification ( $FN_{N_2(denitr.)}$ ).  
182

183 The change in the C balance ( $\Delta C$ ) over time equals the net biome production (NBP) and  
184 can be written for our site as:

185  
186 
$$\Delta C/\Delta t = NBP = FC_{CO_2} + FC_{org\ fert} + FC_{animal} + FC_{CH_4} + FC_{leaching} + FC_{harvest} \quad (2)$$
  
187

188  $FC_{CO_2}$  represents the net ecosystem exchange (NEE) of  $CO_2$  and  $FC_{org\ fert}$  is the C input through  
189 manure application. Carbon input from animal excretion was not included in the budget as it  
190 was assumed to be recycled C from plant and soil uptake.  $FC_{animal}$  includes the C off-take  
191 through animal weight increase and wool production. As grazing cows were heifers in calf,  
192 there was no C off-take through milk to be considered. Methane emissions from enteric  
193 fermentation by ruminants, animal excretion and manure application as well as  $CH_4$  fluxes  
194 from the soil are included in  $FC_{CH_4}$ .  $FC_{leaching}$  is the C lost through dissolved organic and  
195 inorganic C and dissolved  $CH_4$  leaching and  $FC_{harvest}$  represents the C lost from the system  
196 though plant biomass export from harvests (cut for silage). Carbon emissions from farm  
197 operations (i.e. tractor emissions) or off farm emissions (i.e. fertiliser manufacture) are not  
198 included in the C budget.  
199

200 Details of methods to quantify each N and C budget component, as listed in Eq. (1) and (2), are  
201 described under Sect. 2.4 to 2.11. Some budget components were measured throughout the 9  
202 years presented, while others were only measured in some years or not at all. Missing data were  
203 derived from the literature, models or averages from available data from other years.  
204

205 The annual net GHG exchange (NGHGE) was calculated from annual NEE ( $FC_{CO_2}$ ),  
206  $CH_4$  ( $FC_{CH_4}$ ) and  $N_2O$  ( $FN_{N_2O}$ ) fluxes using global warming potentials (GWPs) at the 100-year  
207 time horizon (IPCC, 2013):

$$208 \text{ NGHGE} = (FC_{CO_2}) + FC_{CH_4} * k_{CH_4} + FN_{N_2O} * k_{N_2O} \quad (3)$$

210 Where;

$$212 k_{CH_4} = 9.09, \text{ since } 1 \text{ kg } CH_4\text{-C} = 9.09 \text{ kg } CO_2\text{-C}$$

$$214 k_{N_2O} = 127, \text{ since } 1 \text{ kg } N_2O\text{-N} = 127 \text{ kg } CO_2\text{-C}$$

216  
217 In addition the net annual greenhouse gas balance (NGHGB) was calculated by including the  
218 loss of C through animal meat and wool production, harvest off take, C leaching and input by  
219 organic fertiliser application:

$$220 \text{ NGHGB} = \text{NGHGE} + FC_{\text{org fert}} + FC_{\text{animal}} + FC_{\text{leach}} + FC_{\text{harvest}} \quad (4)$$

## 223 **2.4 Nitrogen and carbon import by fertiliser and manure ( $FN_{\text{synt fert}} + FN_{\text{org fert}} + FC_{\text{org fert}}$ )**

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

230

## 231 **2.5 Nitrogen and carbon export by harvest ( $FN_{\text{harvest}} + FC_{\text{harvest}}$ )**

232 The farmer estimated a harvest of 15 t fresh weight (FW)  $ha^{-1} y^{-1}$  at the first cut and 10 t FW  $ha^{-1}$   
233  $y^{-1}$  at the second cut of a year. As there were two cuts in 2002 and one cut in 2003 the  
234 estimated harvest was 25 t FW  $ha^{-1} y^{-1}$  for 2002 and 10 t FW  $ha^{-1} y^{-1}$  for 2003. A subsample of  
235 harvested vegetation was collected and dried at 80°C for plant N and C analysis using a Carbo-  
236 Erba/400 automated N and C analyser.

237

## 238 **2.6 Nitrogen and carbon export by meat and wool ( $FN_{\text{animal}} + FC_{\text{animal}}$ )**

239 It was estimated by the farmer that heifers increased in weight by 0.8kg per day (starting  
240 weight of 450 kg). The ewe weight was assumed to be constant (60 kg), whereas lambs were  
241 brought to the field at a weight of 5 kg and removed when they reached a weight of 45 kg. The

242 total meat export was calculated from the daily weight increase of heifers and lambs multiplied  
243 by the animal number per day. To calculate the N and C export from meat we assumed an N  
244 content of 3.5 % and a C content of 21 % (Flindt, 2002). Ewes were sheared annually in June,  
245 yielding an estimated 2.5 kg of wool per sheep. Wool N and C export was calculated from wool  
246 production multiplied by the average sheep number in June, assuming a N and C content of  
247 wool of 16.5 and 50 %, respectively (Roche J., 1995)

248

## 249 **2.7 Nitrogen and carbon leaching ( $FN_{\text{leaching}} + FC_{\text{leaching}}$ )**

250 Two sets of ten glass suction cups (pore size  $<1 \mu\text{m}$ , ecoTech, Bonn, Germany) for soil water  
251 and four Teflon suction cups (ecoTech, Bonn, Germany) for soil gas collection were installed  
252 in August 2006. One set was located on a slope, another on a hollow. For the budget  
253 calculations we only used results from the slope location as the hollow location was frequently  
254 water logged. Suction cups were installed horizontally from a soil pit beneath the A horizon (30  
255 cm depth) and at 90cm depth and connected to 2-l glass bottles in an insulated aluminium box  
256 placed into the soil pit. Samples were collected every two to three weeks. For further details  
257 and description of dissolved organic and inorganic C (DIC, DOC) and dissolved  $\text{CH}_4$  analysis  
258 see Kindler et al. (2011). Dissolved inorganic and organic N (DIN, DON) and total N (TN)  
259 concentrations in leachate water were analysed by colorimetric analysis (San<sup>++</sup>, Automated Wet  
260 Chemistry Analyzer - Continuous Flow Analyzer (CFA), Skalar, The Netherlands). Leachate C  
261 and N concentrations were measured from October 1<sup>st</sup> 2006 - March 30<sup>th</sup> 2008. Dissolved C  
262 and N were calculated by multiplying concentrations of DIC, DOC and dissolved  $\text{CH}_4$  or DIN  
263 and DON respectively, with leachate volume. The latter was derived from a soil water model  
264 based on daily precipitation and evaporation data (Kindler et al., 2011). For the remaining years  
265 N was simulated using the LandscapeDNDC model (Haas et al., 2013, with the model tested  
266 and validated with comprehensive measured data. LandscapeDNDC is a process based  
267 biogeochemical model with unifying functionalities of the agricultural-DNDC (e.g. Li et al.,  
268 1992; Li 2000) and the ForestDNDC model (e.g. Kesik et al., 2005; Stange et al., 2000),  
269 particularly suitable for ecosystem N turnover and associated losses of N trace gases and nitrate  
270 leaching (Wolf et al., 2012; Chirinda et al., 2011; Kiese et al., 2011). For C leaching linear  
271 regression models describing the relationship between calculated C leaching fluxes and  
272 leachate volume for the measurement period (DOC;  $y = 0.0186x - 0.0695$ ,  $R^2 = 0.8663$ , DIC;  $y$   
273  $= 0.021x - 0.0008$ ,  $R^2 = 0.8056$  and dissolved  $\text{CH}_4$ :  $y = 0.0019x - 0.0135$ ,  $R^2 = 0.7623$ ) were  
274 used to extrapolate to the remaining years.

275




## 276 **2.8 Gaseous N fluxes**

277



### 278 **2.8.1 N deposition ( $FN_{dep}$ )**

#### 279 **Wet N deposition**

280 Wet N deposition was determined from daily samples collected by an automatic precipitation  
281 sampler (Eigenbrodt® precipitation collector 181/KS, Königsmoor, D) at Auchencorth Moss  
282 ( $3^{\circ}14'35W$ ,  $55^{\circ}47'34N$ ), 17 km south west of Easter Bush (Skiba et al., 2013, McKenzie et al.,  
283 2015). The precipitation collector was only open during rainfall and closed automatically when  
284 precipitation ceased. Precipitation samples were analysed for  $NO_3^-$  and  $NH_4^+$  by ion  
285 chromatography (Methrom AG, Switzerland). Typical detection limits were  $0.5 \mu M$  for  $NH_4^+$   
286 and  $0.4 \mu M$  for  $NO_3^-$ . Annual inorganic N deposition at this site was then adjusted to annual  
287 rainfall amounts at Easter Bush. For years where no data were available (2002, 2003), an  
288 average mineral N concentration per mm rainfall for 2004-2009 was taken and adjusted to the  
289 annual rainfall amount at Easter Bush in 2002 and 2003. 


290

#### 291 **Dry N deposition**

292 Cumulative monthly concentrations of gaseous and aerosol N species ( $NH_3$ ,  $HNO_3$ , particulate  
293  $NH_4^+$  and  $NO_3^-$ ) were collected from another field, about 300m distance from our study field,  
294 using a DELTA system (DENuder for Long Term Atmospheric) (Sutton et al., 2001). The  
295 DELTA system comprised of a denuder filter sampling train, an air pump (providing a  
296 sampling flow rate of  $0.2-0.4 L min^{-1}$ ) and a high sensitivity dry gas meter to record sampled  
297 volumes (Tang et al., 2009) set at 1.5 m height above ground. N dry deposition fluxes were  
298 calculated using the average flux from four different inferential models; the UK CBED scheme  
299 (Concentration Based Estimated Deposition technique (Smith et al., 2000),  the Dutch IDEM  
300 model (Bleeker, 2000),  (Erisman et al., 1994), the dry deposition module of the Environment  
301 Canada model CDRY (Zhang et al., 2001; Zhang et al., 2003) and the surface exchange scheme  
302 EMEP (Simpson et al., 2003; Tuovinen et al., 2009), as described in detail by Flechard et al.  
303 (2011).

304

### 305 **2.8.2 $N_2O$ fluxes ( $FN_{N_2O}$ )**

306 From June 2002 to July 2003  $N_2O$  fluxes were measured continuously by eddy covariance (EC)   
307 using an ultra-sonic anemometer coupled with a Tunable Diode Laser absorption spectrometer  
308 (TDL) at a frequency of 10 Hz. For details see Di Marco et al. (2004). The detection limit for  
309 the TDL was estimated to be 1 ppbV and the detection limit for a 30 min averaging period of  
310 the  $N_2O$  flux measurement was estimated at  $11 ng N_2O-N m^{-2} s^{-1}$ . From August 2006 to

311 December 2009 N<sub>2</sub>O fluxes were measured using manual closed static chambers (Clayton et  
312 al.,1994, Skiba et al., 2013). Four chambers (0.4 m diameter, 0.2 m height) were inserted into  
313 the soil to 0.03 – 0.07 m depth and were accessible for animals to graze. Chambers were closed  
314 usually between 10:00 and 12:00 for 60 minutes with an aluminium lid fitted with a draft  
315 excluder. Samples of 200 ml were collected by syringe and injected into Tedlar bags at the  
316 beginning and the end of the closure time through a three way tap fitted into the lid. In the  
317 laboratory samples were transferred to glass vials and analyzed for N<sub>2</sub>O using a Hewlett  
318 Packard 5890 series II gas chromatograph (Agilent Technologies, Stockport, UK), fitted with  
319 an electron capture detector (detection limit: N<sub>2</sub>O < 33 ppbV). Fluxes were calculated from the  
320 change of gas concentration with time of closure, multiplied by the volume of enclosed space and  
321 divided by its surface. Linearity tests were performed in between measurements showing a  
322 linearity of up to 120 minutes with an average R<sup>2</sup> = 0.96. The minimal detectable flux was 12  
323 ng N<sub>2</sub>O-N m<sup>-2</sup> s<sup>-1</sup>. Fluxes were measured weekly and more frequently during fertilisation.  
324 Cumulative fluxes were calculated by gapfilling data for missing days using linear interpolation  
325 and summing up all gapfilled data over each calendar year. For the periods where no N<sub>2</sub>O  
326 fluxes were measured (January -May 2002, July 2003-March 2004, May 2004-July 2006)  
327 fluxes were simulated by LandscapeDNDC (Haas et al., 2013).

328

### 329 **2.8.3 NO<sub>x</sub> fluxes (FN<sub>NO<sub>x</sub>(soil)</sub>)**

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

345

#### 346 **2.8.4 NH<sub>4</sub> + NO<sub>x</sub> volatilisation (FN<sub>NH3/NOx</sub> (fert.,manure, animal))**

347 The fraction of nitrogen that volatilises as NH<sub>4</sub> and NO<sub>x</sub> from applied synthetic fertiliser or  
348 cattle slurry application and animal excretion was estimated to be 10% and 20% of total N  
349 applied, respectively (IPCC, 2006b). The animal excretion amount was estimated in accordance  
350 with the IPCC Guidelines (IPCC, 2006a). The amount of N excretion (N<sub>ex</sub>) from animals  
351 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>  
352 (amount of N consumed by the animal) depends on the gross energy (GE) intake (see section  
353 2.10) and the crude protein content (CP%) of the feed, assumed to be 15.6% (MAFF, 1990).  
354 N<sub>retention</sub> represents the fraction of N intake retained by the animal as meat, milk or wool. For  
355 lactating ewes the milk production was estimated at 0.618 l animal<sup>-1</sup> d<sup>-1</sup> and the milk protein  
356 content (Milk PR%) at 5.3% (Atti et al., 2006). Daily N excretions were thus calculated as  
357 0.0263 kg N animal<sup>-1</sup> d<sup>-1</sup> for ewes and varied between 0.0019-0.0106 kg N animal<sup>-1</sup> d<sup>-1</sup> for  
358 lambs and 0.096-0.1013 kg N animal<sup>-1</sup> d<sup>-1</sup> for heifers.

359

#### 360 **2.8.5 N<sub>2</sub> emission by total denitrification (FN<sub>N2(denitr.)</sub>)**

361 Di-nitrogen (N<sub>2</sub>) emissions resulting from total denitrification in the soil was not measured in  
362 our experiment. We therefore used the N<sub>2</sub> emission rates from LandscapeDNDC simulations.

363


#### 364 **2.8.6 Biological N<sub>2</sub> fixation (FN<sub>N2</sub> (biol. fixation))**

365 The species composition was measured by the visual estimation method (Braun-Blanquet,  
366 1964). As the legume fraction (*Trifolium repens*) was smaller than 0.5% at each measuring  
367 point we assumed the nitrogen fixation through plants to be zero.

368

#### 369 **2.9 Exchange of CO<sub>2</sub> (FC<sub>CO2</sub>)**

370 NEE was measured by an eddy covariance system consisting of a fast response 3D ultrasonic  
371 anemometer (Metek USA-1, Metek GmbH, Elmshorn, Germany) and a fast closed path CO<sub>2</sub>-  
372 H<sub>2</sub>O analyser (LI-COR 7000 infra-red gas analyzer (IRGA), LI-COR, Lincoln, NE, USA).  
373 Wind velocity components were measured at 2.5m above ground and data were logged at 20  
374 Hz by a PC running a custom LabView data acquisition program. Air was sampled 0.2 m below  
375 the sensor head of the anemometer using 6.3 mm (1/4 in. OD) Dekabon tubing. The IRGA was  
376 located in a field laboratory ca. 10 m from the mast. Lag times between wind data and trace gas  
377 concentrations were synchronised and taken into account in the offline data-processing (Helfter  
378 et al., 2014). Quality control of the eddy covariance data followed the procedure proposed by

379 Foken and Wichura (1996). Data were filtered out if the friction velocity ( $u_*$ ) was smaller than  
380  $0.2 \text{ m s}^{-1}$  (insufficient turbulence),  $\text{CO}_2$  concentrations fell outside a plausible interval (330–  
381 450 ppm),  $\text{CO}_2$  fluxes fell outside the range  $-50$  to  $50 \text{ } \mu\text{mol m}^{-2} \text{ s}^{-1}$  and latent (LE) and sensible  
382 (H) heat fluxes fell outside the range  $-250$  to  $800 \text{ W m}^{-2}$ . Missing NEE data were gap-filled  
383 using the online tool developed at the Max Planck Institute for Biogeochemistry, Jena,   
384 Germany<sup>1</sup> (Reichstein et al., 2005) NEE is the arithmetic sum of the gross primary production  
385 (GPP) and total ecosystem respiration (TER). Flux partitioning of measured NEE into GPP and  
386 TER was calculated by the same online tool used for gapfilling. In this flux partitioning  
387 approach, daytime TER is obtained by extrapolation of a night time parameterisation of NEE  
388 on air temperature and GPP is the difference between ecosystem respiration and NEE.  
389 Contrarily to unmanaged ecosystem, TER at our site also includes the respiratory loss of  $\text{CO}_2$   
390 by grazing animals. Net primary production (NPP), which represents the annual plant growth  
391 (difference between GPP and autotrophic respiration) was calculated as 50% of GPP (Waring  
392 et al., 1998).

393

## 394 **2.10 Methane fluxes ( $\text{FC}_{\text{CH}_4}$ )**

395 Methane fluxes from the soil were measured with closed static chambers simultaneously with  
396 the  $\text{N}_2\text{O}$  measurements (see Sect. 2.8.2). The same GC was fitted with a flame injection  
397 detector (detection limit:  $\text{CH}_4 < 70 \text{ ppbV}$ ). The minimal detectable flux was  $17 \text{ ng CH}_4\text{-C m}^{-2} \text{ s}^{-1}$   
398 <sup>1</sup>. Fluxes were measured weekly and more frequently at fertiliser events. As measured soil  $\text{CH}_4$   
399 fluxes were close to zero and did not vary significantly between months, we calculated  $\text{CH}_4$  for  
400 months where no  $\text{CH}_4$  fluxes were measured (January-May 2002, July 2003-March 2004, May  
401 2004-July 2006), as an average monthly cumulative flux from other years.

402 Methane emissions from grazing animals, i.e. animal excretion and enteric  
403 fermentation, were estimated following the IPCC Tier 2 methodology (IPCC, 2006a: Stewart et  
404 al., 2009). For animal excretion only solid volatile production was considered, as urine has no  
405 effect on  $\text{CH}_4$  emissions (Jarvis et al., 1995). The calculation of  $\text{CH}_4$  emissions from excretion  
406 was based on the amount of volatile solids (VS) excreted, the maximum  $\text{CH}_4$  producing  
407 capacity ( $B_0$ ) of the manure and the  $\text{CH}_4$  conversion factor (MCF), which is specific to the  
408 storage type (pasture, in our study). The amount of VS excreted depended largely on the GE  
409 intake of the animal. The GE intake (based on digestible energy of feed intake, milk  
410 production, pregnancy, current weight, mature weight, rate of weight gain and IPCC constants)  
411 in our study was estimated at  $19.5 \text{ MJ animal}^{-1} \text{ d}^{-1}$  for ewes, while it ranged from 7.9 to 14.9

---


<sup>1</sup> <http://www.bgc-jena.mpg.de/~MDIwork/eddyproc/upload.php>

412 MJ animal<sup>-1</sup> d<sup>-1</sup> for lambs and from 160.9 to 169.7 MJ animal<sup>-1</sup> d<sup>-1</sup> for heifers. Emission factors  
413 for excretion were calculated as 0.198 kg CH<sub>4</sub> head<sup>-1</sup> y<sup>-1</sup> for ewes and varied between 1.64-1.73  
414 kg CH<sub>4</sub> head<sup>-1</sup> y<sup>-1</sup> for heifers and 0.081-0.152 kg CH<sub>4</sub> head<sup>-1</sup> y<sup>-1</sup> for lambs. Methane emission  
415 factors for enteric fermentation were calculated from GE intake and CH<sub>4</sub> conversion factors  
416 (Y<sub>m</sub>). Depending on animal type and live weight, emission factors were 7.6 kg CH<sub>4</sub> head<sup>-1</sup> y<sup>-1</sup>  
417 for ewes and varied between 60.1-63.8 kg CH<sub>4</sub> head<sup>-1</sup> y<sup>-1</sup> for heifers and 2.0-4.0 kg CH<sub>4</sub> head<sup>-1</sup>  
418 y<sup>-1</sup> for lambs. Annual emissions from excretion and enteric fermentation were calculated from  
419 daily CH<sub>4</sub> emissions per animal multiplied by the animal number.

420 Methane emissions from slurry applications were assumed to be small. As no chamber  
421 measurements were conducted at the time of slurry spreading, the emissions were estimated as  
422 0.07 % of the applied assuming that emissions were comparable to those in a related study  
423 (Jones et al., 2006), where CH<sub>4</sub> was measured from chambers after slurry application on a  
424 nearby field in 2002 and 2003.

425

## 426 **2.11 VOC**

427 Fluxes of non-methane volatile organic compounds (VOC) were not measured. We assumed  
428 similar VOC emissions to those reported by Davison et al. (2008) for an intensively managed  
429 grassland in Switzerland, where the daily average flux of methanol, acetaldehyde and acetone  
430 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  
431 values, annual VOC emissions from our field were estimated to be in the order of 0.03% of the  
432 annual C offtake in harvest and 0.08 % of annual C off-take by grazing animals. We therefore  
433 assumed VOC emissions to be negligible. 

434

## 435 **2.12 Soil N and C measurements**

436 Total N and C content of the soil were measured in May 2004 and May 2011. One hundred soil  
437 cores with an inner diameter of 8.7 (2004) and 8.3 cm (2009, both corers from Eijkelkamp  
438 Agrisearch Equipment BV, Giesbeek, The Netherlands) were collected along a regular grid  
439 with a distance of 10 m between sampling points on both occasions. Cores were separated into  
440 layers of 0-5, 5-10, 10-20, 20-30, 30-40, 40-50 and 50-60 cm. Coarse stones of a diameter > 4  
441 mm and roots of a diameter >1mm were removed from the samples prior to drying at 40 °C.  
442 Stone and root samples were air-dried separately. Then, soil samples were sieved to < 2 mm.  
443 Particles > 2 mm were combined with the coarse stones. Dry weights of roots and combined  
444 stone fractions were determined. Total N and C concentrations in < 2 mm soil separates were  
445 determined using dry combustion (VarioMax, Elementar Analysensysteme GmbH, Hanau,

446 Germany). As the site contains no inorganic C, total C equals organic C. As bulk density varies  
447 spatially and over time (e.g. through compaction by livestock), the soil N and C content per  
448 unit ground area to a fixed depth will also change, without any change in the mass fraction of N  
449 and C in dry soil. Therefore, total N and C stocks were calculated on an equivalent soil mass  
450 (ESM) basis, so that comparisons between years were valid (see Gifford and Roderick, 2003,  
451 Wendt and Hauser, 2013). A cubic polynomial was fitted to the data, to predict cumulative N  
452 and C with cumulative soil mass in the profile. A soil mass of  $800 \text{ kg m}^{-2}$  was used (Table 7),  
453 which corresponds to approximately 60-cm depth, which was the depth of the corer.  
454 Uncertainty in the estimates of stock change was based on the prediction intervals in the cubic  
455 polynomial at a soil mass of  $800 \text{ kg m}^{-2}$ .

456

### 457 **2.13 Ancillary measurements**

458 Soil temperature and volumetric soil moisture were continuously recorded at four depths (3.5,  
459 7.5, 15 and 30 cm) by temperature probes (temperature probe 107, Campbell Scientific,  
460 Loughborough, UK) and TDR probes (TDR 100, Campbell Scientific, Loughborough, UK),  
461 respectively, the latter installed in June 2002. Rain was measured by a tipping bucket rain  
462 gauge, while air temperature and relative humidity were measured by an integrated humidity  
463 and temperature transmitter (HUMITTER<sup>®</sup>, Vaisala Ltd, Suffolk, UK).

464

### 465 **2.14 Statistical and uncertainty analysis**

466 Random error was determined as  $2\sigma$ -standard error (95% confidence) of the overall mean  
467 according to Gaussian statistics. Analyses of variance (ANOVA) were used to test if values  
468 were significantly different from zero ( $p < 0.05$ ). For systematic errors the uncertainty range of  
469 measurements as well as of parameterisations and literature based estimates was estimated  
470 according to expert judgment. To calculate the combined effect of systematic uncertainties of  
471 each flux component on the C and N budget simple Gaussian error propagation rules were  
472 used. Confidence intervals are given at the 95% confidence level.

473

## 474 **3. Results**

### 475 **3.1 Climate and management**

476 The meteorological conditions exhibited substantial inter-annual variability in the study period  
477 2002-2010 (Table 2 and Fig. 2). Annual rainfall ranged from 575 mm to 1238 mm with highest  
478 monthly rainfalls at  $280 \text{ mm month}^{-1}$  in September 2002. Lowest annual reported rainfall was  
479 in 2010; this low value was caused by a gap in data from January-March, due to snowfall.

480 Average annual air temperature ranged from 8.3 to 9.6 °C with highest daily air temperatures of  
481 30.4 °C in August 2005 and lowest in December 2010 at -10.3 °C. Highest average monthly air  
482 temperatures were measured in July 2006 at 17°C and lowest monthly average air temperatures  
483 at 2°C in November 2009. In 2003 the highest average annual temperature (9.6° C) and lowest  
484 annual rainfall (680 mm) were measured with a correspondingly low annual soil water content  
485 of 31 %. The duration of the growing season was defined per calendar year as the period  
486 bounded by the first and last 5 consecutive days with mean daily air temperature  $\geq 5$  °C. The  
487 length of the growing season (LGS) varied between 151 days (2006) and 242 days (2009)  
488 (Table 2).

489 Livestock stocking density exhibited both intra- and inter-annual variability. The  
490 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  
491 LSU ha<sup>-1</sup> y<sup>-1</sup>, respectively (Table 1), which were the years where the grass was cut for silage  
492 and no lambs were present in the field. In 2007, 2008, 2009 and 2010 no heifers were present in  
493 the field. Highest annual average stocking density occurred in 2004 and 2007 at 0.99 LSU ha<sup>-1</sup>  
494 y<sup>-1</sup> and 0.91 LSU ha<sup>-1</sup> y<sup>-1</sup>, respectively. Maximum monthly stocking density occurred in  
495 September 2006 at 13.8 LSU ha<sup>-1</sup>, while interim periods with no grazing at all were observed in  
496 all years (Fig. 1a). Mineral N fertiliser was applied split into 3 to 5 applications per year,  
497 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  
498 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,  
499 which resulted in a C input of 55.4 and 171.8 g C m<sup>-2</sup> application<sup>-1</sup>, respectively (Fig. 1b and c).  
500 The grass was only cut in 2002 and 2003. Harvested biomass in 2002 and 2003 ranged from  
501 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>  
502 cut<sup>-1</sup> and a C removal from the field ranging from 113.1 to 169.5 g C m<sup>-2</sup> cut<sup>-1</sup> (Fig. 1c).

503

### 504 **3.2 Uncertainty analysis**

505 Systematic uncertainties for each component of the C and N budget are shown in Table 3.  
506 Uncertainty values were estimated according to expert judgment. The systematic uncertainty of  
507 the N input from mineral fertiliser was assumed to be minimal (1 %), while the systematic  
508 uncertainty of the N and C spread by the manure was assumed to be 17 % on average for the C  
509 and N analysis. Together with an uncertainty of 10 % of the volume spread, this resulted in a  
510 total uncertainty of 20 %. The uncertainty of the C and N analysis for harvest were 4 and 12 %,  
511 respectively. We assumed an error of 10% in the farmer's estimate of the harvest amount,  
512 which resulted in a total uncertainty of 16 % for N and 11 % for C off take. We attributed a  
513 systematic uncertainty of 30 % to the modelled data for C and N leaching. The systematic

514 uncertainty of the meat and wool consists of the estimated uncertainty in the animal weight,  
515 animal numbers and literature values for wool and meat C and N contents. We assign an  
516 uncertainty for animal weight of 10 %, for animal numbers of 5 % and for literature values of  
517 wool and meat C and N content of 3 %, resulting in a total uncertainty of 12 %. The uncertainty  
518 of wet N deposition was 30 % resulting from the error of sample analysis and a potential bias  
519 from dry deposition on the funnel. The uncertainty of dry N deposition consisted of an error of  
520 7 % for the analysis of DELTA samples and an 80% uncertainty of the variation of the output  
521 from the four models, which resulted in a total uncertainty of 80%. The systematic uncertainty  
522 attributed to the annual cumulative N<sub>2</sub>O fluxes was 30 %, due to the uncertainty of gapfilling.  
523 The uncertainty attributed to the modelled NO<sub>x</sub> fluxes is 30 %. The uncertainty attributed to the  
524 NH<sub>4</sub> and NO<sub>x</sub> volatilisation was 30 % from applied synthetic fertiliser and 50 % from cattle  
525 slurry application and animal excretion. The uncertainty attributed to the N<sub>2</sub> fluxes was 30 %.  
526 The total uncertainty for NEE values was estimated to be 80 g C m<sup>-2</sup> y<sup>-1</sup> (Levy et al.,  
527 submitted). The systematic uncertainty of annual cumulative soil CH<sub>4</sub> fluxes was very high at  
528 160 %, due to the uncertainty of gap filling and as values were close to zero. The uncertainty of  
529 CH<sub>4</sub> from enteric fermentation and animal excretion estimates were each assumed to be 20%,  
530 according to IPCC (2006a). The uncertainty of CH<sub>4</sub> fluxes from organic manure application  
531 was estimated at 120 %.

532

### 533 **3.3. N budget**

534 In our grassland system the N balance is the difference between the N input through fertiliser  
535 and atmospheric deposition and the N output through harvest, animal export, leaching and  
536 gaseous emissions. The total resulting balance over the nine years, derived from flux  
537 calculations and estimations, showed that N was stored at an average rate of  $-7.21 \pm 4.6$  g N m<sup>-2</sup>  
538 y<sup>-1</sup> ( $p < 0.05$ ). From 2003 to 2010, N was stored at a rate of -3.1 to -17.9 g N m<sup>-2</sup> y<sup>-1</sup>, whilst in  
539 2002 N was lost at a rate of 6.3 g N m<sup>-2</sup> y<sup>-1</sup> (Table 4). The major N input consisted of inorganic  
540 fertiliser, ranging from -11 to -25.9 g N m<sup>-2</sup> y<sup>-1</sup>, averaging at -19.2 g N m<sup>-2</sup> y<sup>-1</sup>, while N  
541 deposition represented only between 1.9 and 5.9% of the total N input. During the years where  
542 N was stored, a significant positive correlation between total N input from fertiliser and N  
543 storage was observed ( $R^2 = 0.55$ ). Largest losses resulted from leaching at an average rate of  
544  $5.34 \pm 3.4$  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  
545 2003 at 0.09 g N m<sup>2</sup> y<sup>-1</sup>. We found a strong correlation between N leaching and rainfall ( $R^2 =$   
546 0.82), if values from 2004 were excluded, a weak correlation between livestock density and N  
547 leaching if the years 2002 and 2004 were excluded ( $R^2 = 0.47$ ), while no correlation with total N



548 input could be found. The total N off take through meat and wool ranged from 0.15-3.12 g N  
549  $\text{m}^{-2} \text{y}^{-1}$ , while the total annual N offtake from harvest was 5.0 g N  $\text{m}^{-2} \text{y}^{-1}$  in 2002 and 4.68 g N  
550  $\text{m}^{-2} \text{y}^{-1}$  in 2003. Amongst gaseous exchanges, highest losses were estimated from  $\text{N}_2$  emissions,  
551 averaging at 2.76 g N  $\text{m}^{-2} \text{y}^{-1}$  with maximum losses of 4.12 g N  $\text{m}^{-2} \text{y}^{-1}$  in 2009, although in  
552 2004 and 2005 losses from  $\text{NO}_x/\text{NH}_3$  volatilisation from excretion and organic fertilisation  
553 exceeded losses from  $\text{N}_2$  emissions. Losses through  $\text{NO}_x$  from the soil were always less than  
554 1% of the total N exchange (0.2 g N  $\text{m}^{-2} \text{y}^{-1}$ ). Nitrous oxide emissions ranged from 0.11 to 1.27  
555 g N  $\text{m}^{-2} \text{y}^{-1}$ , representing 1.3-8.4 % of the total N export. Annual  $\text{N}_2\text{O}$  emissions showed no  
556 correlation with precipitation, livestock density or total N input. However, there was a positive  
557 correlation with rainfall if 2004 and 2007 data were excluded ( $R^2=0.78$ ); with livestock density  
558 if the years 2002 and 2004 were excluded ( $r^2=0.70$ ); and with total N input if the years 2002,  
559 2003 and 2010 were excluded ( $R^2=0.76$ ).  $\text{N}_2\text{O}$  emission factors (percentage of N lost from total  
560 N inputs by mineral and organic fertiliser), ranged between 0.6 and 7.5 % (Table 6).

561 To investigate the influence of different managements on the N and C budget, we  
562 separated experimental years into harvested and grazed (2002 and 2003) and grazed only years  
563 (2004-2010 Fig. 3 and 4). During the harvested years, the main loss of N from the system  
564 occurred through leaching (39.2% of total N inputs), followed by the export through harvest  
565 (25.2%), while the export from animals (meat and wool) accounted for less than 2 % of total  
566 losses (Fig. 3a). The main loss to the atmosphere resulted from total denitrification ( $\text{N}_2$ ;  
567 15.4%), followed by  $\text{NO}_x/\text{NH}_3$  volatilisation from inorganic N fertiliser applications (9.5%),  
568 while  $\text{N}_2\text{O}$  emissions accounted for 3.3%,  $\text{NO}_x/\text{NH}_3$  volatilisation from excretion for 2.7% and  
569  $\text{NO}_x$  from soil for less than 1%. The residual 2% represents the N storage in the soil and the  
570 uncertainty of the budget. When grazed-only years were considered (Fig. 3b), the residual part  
571 was the highest at 38.6%. Losses through leaching (19.9%) and  $\text{N}_2$  (11.4%) were lower in  
572 grazed years compared to harvested years, while the export through grazing animals were  
573 considerably higher at 15.8% (sum of N loss through meat, wool and  $\text{NO}_x/\text{NH}_3$  volatilisation  
574 from excretion). An additional loss occurred in grazed years through the volatilisation of  
575  $\text{NO}_x/\text{NH}_3$  from organic fertiliser applications in 2004 and 2005 (3%). Losses through  $\text{N}_2\text{O}$  and  
576  $\text{NO}_x/\text{NH}_3$  from inorganic fertiliser were comparable to harvested years at 2.5% and 8.3%,  
577 respectively.

578 Cumulative soil N stocks were derived from soil core measurements taken in May 2004 and  
579 May 2011. Nitrogen storage over the 7 years was calculated from the cumulative equivalent  
580 soil mass (ESM) for the soil mass increment of 800 kg  $\text{m}^{-2}$ , which corresponds to approximate  
581 60 cm depth. The estimated N storage over the 7 years was  $-4.51 \pm 2.64$  g N  $\text{m}^{-2} \text{y}^{-1}$  (Table 7)

582 and was a significant N accumulation to the soil ( $p < 0.01$ ). The estimated N storage derived  
583 from flux calculations between 2004 and 2010, however was  $-9.20 \pm 4.10 \text{ g N m}^{-2} \text{ y}^{-1}$ , which is  
584 2 times more than that calculated by sequential soil analysis.

585

### 586 **3.4. C budget**

587 Annual C inputs through photosynthesis (GPP) varied between  $-982.1$  and  $-2162.9 \text{ g C m}^{-2}$ , and  
588 losses through autotrophic and heterotrophic respiration (TER) varied between  $972.1$  and  
589  $2183.2 \text{ g C m}^{-2}$ , both considerably larger than any other C fluxes (Table 5). If only the NEE  
590 was considered (difference between GPP and TER), the grassland acted as a sink for  $\text{CO}_2$  at an  
591 average of  $218 \pm 155 \text{ g C g C m}^{-2} \text{ y}^{-1}$ , and the  $\text{CO}_2$  uptake was significantly different from zero  
592 ( $p < 0.05$ ). The sink strength ranged from  $-10 \text{ g C m}^{-2} \text{ y}^{-1}$  (2006) to  $-606 \text{ g C m}^{-2} \text{ y}^{-1}$  (2009),  
593 only in 2004, the grassland was a small source of  $\text{CO}_2$  ( $72 \text{ g C m}^{-2} \text{ y}^{-1}$ ). Taking into account all  
594 C inputs and outputs (NBP), C was sequestered on average at  $164 \pm 140 \text{ g C m}^{-2} \text{ y}^{-1}$  over the  
595 nine years, although the storage was not significantly different from zero ( $p < 0.05$ ). In 2004 and  
596 2006 C was lost from the ecosystem. The major C import resulted from NEE in all years apart  
597 from 2005, when the C input from manure application was larger. Highest C export occurred  
598 from harvest in 2002 and 2003 ( $270.6$  and  $169.5 \text{ g C m}^{-2} \text{ y}^{-1}$  respectively), while second largest  
599 export in 2002 and 2003 and largest exports in other years was leaching ( $6.8$  to  $25.1 \text{ g C m}^{-2} \text{ y}^{-1}$ ).  
600 The measured C leaching value for 2007 ( $15.4 \text{ g C m}^{-2} \text{ y}^{-1}$ , table 5) differs from the leaching  
601 value published for Easter Bush by Kindler et al. (2011), as we only used values of one of the  
602 two measured sites in this manuscript (slope, not hollow, as the hollow location was frequently  
603 water logged). The third largest C loss consisted of C export from meat in 2004-2010, ranging  
604 from  $6.4$ - $15.8 \text{ g C m}^{-2} \text{ y}^{-1}$ . In 2002 and 2003, when no lambs were present in the field, C export  
605 from meat was exceeded by  $\text{CH}_4$  losses from enteric fermentation. Carbon export from wool  
606 ranged from  $0.5$  to  $2.1 \text{ g C m}^{-2} \text{ y}^{-1}$ .  $\text{CH}_4$  emissions from organic fertilisation, soil processes and  
607 animal excretion were always less than 1 % of the total C losses.  $\text{CH}_4$  losses from enteric  
608 fermentation ranged from  $1.5$  to  $5.7 \text{ g C m}^{-2} \text{ y}^{-1}$ , corresponding to 0.5-22.5 % of all C losses  
609 from the ecosystem. The annual carbon balance (NBP) was dominated by the NEE. A high  
610 livestock density tended to reduce the net sink strength. A significant negative correlation of  
611 NEE as well as NBP with stocking density could be seen ( $R^2=0.77$  and  $R^2=0.83$ , respectively),  
612 if the years with cuts (2002 and 2003) were excluded. The NBP correlated positively with  
613 rainfall ( $R^2=0.48$ ) whereas the correlation improved if the dry year 2003 was excluded  
614 ( $R^2=0.78$ ). There was only a weak correlation between NEE and rainfall ( $R^2=0.38$  for all years,  
615  $R^2=0.47$  without the year 2003).

616 Net primary production (NPP) in years when grass was harvested and grazed (2002 and  
617 2003) and grazed only (2004– 2010) are presented in Figure 4. In both management types most  
618 C was lost through ecosystem respiration, (67% and 71% of NPP, respectively). Harvest export  
619 represented 21% of NPP. Leaching accounted for 1.5% of NPP during harvested years and  
620 2.2% in grazed only years. Animal export (meat and wool) consisted of 1.5% of NPP in grazed  
621 only years and was less than 0.2% of NPP in grazed and harvested years. The sum of all CH<sub>4</sub>  
622 emissions (from organic fertilisation, excretion, enteric fermentation and soil) was less than 1%  
623 of the NPP. The residual part, which includes the C storage in the soil as well as the uncertainty  
624 of the budget, was estimated at 10% and 24% of NPP in harvested and grazed or grazed years,  
625 respectively.

626 The C content for the cumulative soil mass increment 0-800 kg m<sup>-2</sup> (~ 0-60 cm) was lower in  
627 2011 compared to 2004, resulting in a C loss of 29.08 ± 38.19 g C m<sup>-2</sup> (Table 7). In  
628 comparison, based on flux calculations C was stored at 180 ±180 g C m<sup>-2</sup> y<sup>-1</sup> over the 7 years.  
629 However, neither C loss calculated by sequential soil analysis, nor C storage estimated from  
630 flux calculations were significantly different from zero.

631

### 632 3.5. Greenhouse gas budget


633 In order to calculate the global warming potential for the Easter Bush grassland fluxes of the  
634 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  
635 different global warming potentials for each gas at the 100 year time horizon (1 for CO<sub>2</sub>, 298  
636 for N<sub>2</sub>O and 25 for CH<sub>4</sub>, IPCC, 2013). Average greenhouse gas fluxes, net GHG exchange  
637 (NGHGE) and attributed net GHG balance (NGHGB) for 2002-2010 are shown in Figure 5.  
638 The CO<sub>2</sub> storage from the NEE provided the largest term in the annual GHG budget. Carbon  
639 dioxide (NEE) was sequestered over the 9 years at a rate of -799 ± 567 g CO<sub>2</sub> m<sup>-2</sup> y<sup>-1</sup>. This  
640 storage was significantly different from zero (p < 0.05). On average, the net GHG exchange  
641 (NGHGE) was highly correlated with NEE (R<sup>2</sup>=0.96). On average the grassland was a source  
642 of the GHGs CH<sub>4</sub> and N<sub>2</sub>O at a rate of 148 ± 30 and 285 ± 131 g CO<sub>2</sub> m<sup>-2</sup> y<sup>-1</sup>, respectively, both  
643 being significantly different from zero (p < 0.001 and p < 0.01, respectively). Nitrous oxide  
644 losses ranged from 52 g CO<sub>2</sub> eq. m<sup>-2</sup> y<sup>-1</sup> (2004) to 588 g CO<sub>2</sub> eq. m<sup>-2</sup> y<sup>-1</sup> (2007) (data for each  
645 year not shown). Methane from soil processes, manure input as well as animal excretion,  
646 accounted for less than 5% of total CH<sub>4</sub> emissions. Methane emissions from enteric  
647 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). The CH<sub>4</sub>  
648 emissions, which were predominately (> 97%) of ruminant origin weakened the sink strength  
649 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

650 substantial 54% and increased to a total of 67 %, if only grazed years were considered. On  
651 average the grassland represented a GHG sink of  $-366 \pm 601 \text{ g CO}_2 \text{ m}^{-2} \text{ y}^{-1}$ , if only NEE, CH<sub>4</sub>  
652 and N<sub>2</sub>O were included (NGHGE). If all C components (FC<sub>org.fert.</sub>, FC<sub>animal</sub>, FC<sub>leaching</sub>, FC<sub>harvest</sub>)  
653 are included, the sink strength of the grassland decreased to  $-182 \pm 560 \text{ g CO}_2 \text{ m}^{-2} \text{ y}^{-1}$   
654 (NGHGB). This represents a weakening of the sink strength of the NGHGE by 50 %, mainly  
655 due to the export of harvest. However, it has to be noted that in harvested years the return of the  
656 manure, resulting from the grass fed to livestock off -site, would reduce the GHG balance. If  
657 only grazed years were considered the sink strength increased slightly by 5.4 %, due to the C  
658 input from manure in 2004 and 2005. Both, NGHGE and NGHGB were not significantly  
659 different from zero.

660

## 661 **4. Discussion**


### 662 **4.1. N balance**



663 Our experimental field has been under grazing/cutting management for more than 20 years with   
664 regular N inputs from mineral fertilizers, manure and animal excretion. As biological N<sub>2</sub>  
665 fixation by legumes is inhibited by soil mineral N (Streeter, 1988), the legume fraction was less  
666 than 1% and therefore a negligible source of N in our system. Atmospheric N deposition (wet  
667 and dry) accounted only for a small fraction of the total N input on our managed grassland.  
668 This is in contrast to semi natural systems, where atmospheric N deposition represents the main  
669 N input (Pheonix et al., 2006, Bleeker et al., 2011). The main N inputs in our study were from  
670 inorganic and organic fertilizer additions. The amount of N added through fertilizer was  
671 governed by recommended maximum levels (SRUC, 2013) and lies within the range of N  
672 applied in other European studies with similar management (e.g. Laws et al., 2000; Allard et  
673 al., 2007; Ammann et al., 2009). Nitrogen added through the excretion from grazing animals  
674 was not considered an N input as this represents an internal redistribution of N within the  
675 system.

676

#### 677 **4.1.1 N use efficiency**

678 The ratio between N input and percentage of N uptake into the crop or animal products (meat,  
679 wool and milk) is defined as the N use efficiency (NUE). In our study a substantial amount of  
680 N was removed by harvest, with an NUE of herbage in cut years (2002 and 2003) of 25%  
681 (Figure 3a). This seems low compared to reported N efficiencies of 55-80% in harvested  
682 herbage from managed temperate grasslands (Ball and Ryden 1984; Ammann et al., 2009). The  
683 inclusion of grazing ruminants alters the NUE of herbage as the nitrogen in the grazed grass is

684 consumed  and converted to meat, milk, wool, or is excreted. The lower NUE in the grass  
685 production in our study is therefore partly due to grazing. Furthermore, it has been shown that  
686 the proportion partitioned to plant uptake decreases as the total amount of soil inorganic N  
687 increases (Scholefield et al., 1991), which is a further explanation for a low NUE in herbage in  
688 our high N input system. There are different mitigation options to reduce N losses and thus  
689 increase NUE. The introduction of clover into grassland has been shown to reduce the  
690 requirement of N input from fertilisation, thereby resulting in the same yield (Herrmann et al.,  
691 2001; Ledgard, 2001). Adherences with fertiliser recommendation systems and avoidance of  
692 over fertilisation is also likely to increase the efficiency of N use without compromising  
693 productivity (Rees et al., 2013). The use of nitrification inhibitors applied onto grassland has  
694 been shown to result in a reduction of N<sub>2</sub>O emissions (McTaggart et al., 1997). Furthermore, a  
695 novel approach to reduce N losses from sheep urine, by infusing N process inhibitors into the  
696 gastrointestinal tract of the animals, has been demonstrated by Ledgard et al. (2008), however,  
697 the evidence for this as a mitigation option is still limited, and could face legal and ethical  
698 challenges.

699 The NUE in crops is significantly higher compared to the NUE in animal production. The  
700 NUE of animal products on our grassland system ranged from 5 to 18% in grazed years (2004-  
701 2010), with an average of 10.6 %. This is in agreement with the NUE reported for sheep of 6.2  
702 % by Van der Hoek (1998) and studies for beef production systems, which reported N  
703 efficiencies range from 6 to 12% (Whitehead et al., 1986; Tyson et al., 1992) and 5-20% (Ball  
704 and Ryden, 1984). Approximately 85% of crops produced  are used for animal feed, which is  
705 significantly less efficient than if the crops were used to feed humans directly. A measure to  
706 reduce N pollution could therefore be the reduction of meat consumption  (Smith et al. 2013).

#### 708 **4.1.2 N loss to the environment:**

709 Nitrogen leaches from grassland soils in the form of nitrate (NO<sub>3</sub><sup>-</sup>), ammonium (NH<sub>4</sub><sup>+</sup>) and  
710 dissolved organic N (DON). Whereas NO<sub>3</sub><sup>-</sup> is highly mobile in water and can be easily leached  
711 into groundwater, NH<sub>4</sub><sup>+</sup> is less prone to leaching as it is mostly bound to soil particles (Brady  
712 and Weil, 2002). Leaching depends on the water-holding capacity of the soil, amount of  
713 rainfall, water use by plants and soil nutrient content, which are in turn influenced by  
714 management. Leaching occurs predominantly from late autumn to early spring when  
715 precipitation often exceeds evapotranspiration (Askegaard et al., 2005). In our field leaching  
716 losses varied widely over the years. This variation can mainly be explained by differences in  
717 precipitation. Overall, leaching from our field ( $5.3 \pm 3.4 \text{ g N m}^{-2} \text{ y}^{-1}$ ) was comparable to values

718 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  
719 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.,  
720 1998), max. leaching losses of 10.4 g N m<sup>-2</sup> y<sup>-1</sup>). However, leaching from our study was high  
721 compared to the Swiss NitroEurope site, where a maximum loss of 3.5 kg N ha<sup>-1</sup> y<sup>-1</sup> was  
722 estimated from an ungrazed grass/clover sward, despite annual rainfall and N inputs  
723 comparable to our site (Ammann et al., 2009). This difference can be explained by the different  
724 plant cover and management. It has been shown that clover introduction can reduce leaching  
725 (Owens et al., 1994), whereas grazing tends to increase leaching (Cuttle and Scholefield, 1995).  
726 Grazed grasslands tend to have higher N leaching rates than cut grasslands as the N added as  
727 fertiliser is not removed by harvest, but returned to the soil in urine and dung from consumed  
728 herbage, prone to leaching. The uneven distribution of excreted organic N further enhances  
729 leaching due to the formation of N hotspots, which has been observed at outdoor pig farms (e.g.  
730 (Eriksen, 2001). Ryden et al., (1984a) showed a 5.6 times higher leaching loss from grazed  
731 compared to cut grassland with 36% of total N inputs lost from grazed compared to 6% lost  
732 from cut grassland. On our site leaching equaled about 20 % of total inputs in grazed years,  
733 compared to 39% in the cut years. However, the higher value in cut years was due to the high  
734 rainfall in 2002.

735  
736 Due to high atmospheric N<sub>2</sub> background, N<sub>2</sub> fluxes cannot be measured directly in the  
737 field. However, there are different methods to measure N<sub>2</sub> fluxes indirectly, which have been  
738 summarized by Groffman et al. (2006). In our study, we estimated N<sub>2</sub> losses using the process  
739 based biogeochemical model LandscapeDNDC (Haas et al, 2013, Molina-Herrera et al. 2016).  
740 They represented the highest gaseous losses from our grassland in most years, with an average  
741 of 12.6 % of total N inputs and 14 % of inorganic fertilizer N inputs. This is comparable with  
742 the average N<sub>2</sub> loss of 12.5 % from inorganic N applications measured by the acetylene  
743 inhibition method from a fertilized and cut, but ungrazed grassland in Switzerland (Rudaz et  
744 al., 1999). Using the same method, van der Salm et al. (2007) reported a higher loss of 22% of  
745 total N input from a cattle grazed pasture on a heavy clay soil in the Netherlands. Apart from  
746 the impact of the heavy clay soil, which could have enhanced denitrification due to reduced  
747 oxygen concentrations, grazing is likely to have enhanced denitrification rates in van der  
748 Salm's study. Grazing not only enhances denitrification through soil compaction caused by  
749 trampling animals but also due to the formation of N hot spots resulting from unevenly  
750 distributed soil N from excretion. In our study N<sub>2</sub> losses simulated by LandscapeDNDC are  
751 based on average (per ha<sup>-1</sup>) changes of the soil N pool instead of the more uneven distribution

752 of soil N in hot spots like urine patches. Therefore it is likely that N<sub>2</sub>O losses in our study have  
753 been underestimated.

754

755 Nitrous oxide emissions are influenced by both management and environmental  
756 conditions (Flecharth et al., 2007, Bell et al., 2015; Cowan et al., 2015). In our study N<sub>2</sub>O fluxes  
757 showed typical temporal variations with high N<sub>2</sub>O peaks after N application decreasing to  
758 background levels after < 1 to 20 days, increased losses during wetter periods, and reduced  
759 losses during the colder winter months (Skiba et al., 2013). Spatial variability was high due to  
760 the uneven distribution of excreta and urine and uneven soil compaction from grazing animals  
761 (Jones et al., 2011). Values measured in our study (0.1 to 1.3 g N m<sup>-2</sup> y<sup>-1</sup>) are within the range  
762 of literature values from reported grazed as well as un-grazed European grasslands (Velthof  
763 and Oenema, 1997; Leahy et al., 2004; Flecharth et al., 2007). Generally N<sub>2</sub>O losses are higher  
764 from grazed grassland compared to cut, ungrazed pasture (Velthof and Oenema, 1995; Luo et  
765 al., 1999) due to a more anaerobic environment as a consequence of soil compaction caused by  
766 animal treading and the influence of N and C from the deposition of animal excreta to the soil  
767 (Oenema et al., 1997). We could only find correlations between annual N<sub>2</sub>O emissions and  
768 stocking density, rainfall or total N input if certain years were excluded. This shows that N<sub>2</sub>O  
769 emissions are not a uniform fraction of N applied, as suggested by the Tier 1 IPCC  
770 methodology, but are also influenced by the type of N applied, by stocking density, and by the  
771 rainfall at the time of fertilization (Jones et al., 2007; Flecharth et al., 2007). We found a  
772 relationship between the cumulative precipitation 1 week before plus 3 weeks after fertilization  
773 and N<sub>2</sub>O emissions (R<sup>2</sup>=0.53) (Skiba et al., 2013). This relationship, together with the influence  
774 of stocking density and type of N applied needs to be considered when developing Tier 2  
775 emission factors. Emission factors, calculated as a simple fraction of total N input (mineral and  
776 organic fertilizer) showed a variation of 0.6 and 7.4% on our field. In five out of eight years  
777 this value was above the uncertainty range (0.3 - 3 %) given by IPCC Tier 1 guidelines (IPCC,  
778 2006b). However, it has been shown that the N<sub>2</sub>O emission factor from managed grassland can  
779 be higher, especially under wet conditions and with a high soil C content as this is the case for  
780 Scottish soils (Jones et al., 2007; Dobbie et al., 1999; Bell et al., 2015).

781

782 In grazed pastures NH<sub>3</sub> volatilizes from urine patches, decomposing dung as well as  
783 from fertilizers containing urea and NH<sub>4</sub><sup>+</sup> (Twigg et al 2011). Increased rates of NH<sub>3</sub> losses  
784 have been associated with high stocking densities under a rotational grazing system by Ryden  
785 and Mc Neill (1984). In our study, N volatilized as NH<sub>3</sub> and NO<sub>x</sub> from inorganic and organic

786 fertiliser and animal excretion, before it was incorporated into the soil, and accounted for a  
787 considerable amount of total N, with losses of 13 % in cut and grazed years (2002, 2003) and  
788 17 % in grazed only years. Apart from 2004, where stocking rates were highest, NO<sub>x</sub> and NH<sub>3</sub>  
789 volatilizations from inorganic fertilizer applications exceeded those from animal excretion,  
790 while those from organic manure applications exceeded those from inorganic fertilizers (2004,  
791 2005). However there is a high uncertainty attributed to those estimates.

792 Soil NO<sub>x</sub> emissions result predominately from microbial nitrification of either added N  
793 fertilizers or following the mineralization of soil organic matter, animal excretions or added  
794 manure. Emissions tend to be linked with aerobic soil conditions (Davidson, 1991). In relation  
795 to the total N loss from our grassland system, soil NO<sub>x</sub> emissions were estimated to be  
796 negligible, accounting for less than 1% of the total budget.

797

#### 798 **4.1.3 N storage in the soil**

799 Results from soil analysis taken in May 2004 and May 2011 indicate that our field has stored  
800 N. The N budget assessed from the net N flux balance also showed that N was stored in the soil  
801 of our grassland, although at a higher rate (average N storage of  $-7.2 \pm 4.6 \text{ g N m}^{-2} \text{ y}^{-1}$  over all 9  
802 years and average N storage of  $-9.16 \pm 4.09 \text{ g N m}^{-2} \text{ y}^{-1}$  in grazed years, 2004-2011). The slight  
803 shifts in measurement periods (May 2004 – May 2011) for the soil stock calculations and the  
804 period (Jan 2004 – Dec 2010), is presumed to be insignificant in this comparison.

805 Results from both methods are within the range of literature values. Neeteson and  
806 Hassink (1997) found a N accumulation in SOM of  $0-25 \text{ g N m}^{-2} \text{ y}^{-1}$  from two cattle grazed  
807 farms in the Netherlands, while Watson et al. (2007) reported a N storage in grazed Irish  
808 grasslands ranging from  $10-15.2 \text{ g N m}^{-2} \text{ y}^{-1}$ , depending on N inputs. Soil N storage assessed  
809 from soil measurements from a cut grassland close to our field, where plots were treated with  
810 cattle slurry, stored N over 6 years at a rate of  $-2.17 \text{ g N m}^{-2} \text{ y}^{-1}$  in the top 10 cm, while, in the  
811 same experiment, a N loss was observed from mineral N and urea treatments ( $4.5$  and  $8.3 \text{ g N}$   
812  $\text{m}^{-1} \text{y}^{-1}$ , respectively) (Jones et al., 2007). In contrast, Schipper et al. (2007) reported an average  
813 loss of  $9.1 \text{ g N m}^{-2} \text{ y}^{-1}$  in the top 100cm from managed grasslands over 20 years in New  
814 Zealand.

815 The reason for the difference between methods (flux measurements vs sequential soil  
816 sampling) in our study might lie in a possible underestimation of losses from flux  
817 measurements. Uncertainties of our estimates are high, especially those from N losses. The  
818 largest absolute systematic uncertainty for the N balance was attributed to N leaching. Leaching  
819 was modelled for most years, whereas the model was validated using measured data from



820 October 1<sup>st</sup> 2006 - March 30<sup>th</sup> 2008. The spatial variability of leaching was not considered in  
821 the measured data, as only one location has been used. The uncertainty of the leaching estimate  
822 would therefore be reduced if the model could be validated with data measured from several  
823 locations. The second highest systematic uncertainty was attributed to losses through N<sub>2</sub>,  
824 NO<sub>x</sub>/NH<sub>3</sub> emission from excretion, NO<sub>x</sub>/NH<sub>3</sub> emission from inorganic fertilization and inputs  
825 from organic fertilization. Combined uncertainties from all components lead to a total  
826 systematic uncertainty in the N balance of 2.1 g N m<sup>-2</sup> y<sup>-1</sup> (2004-2010).

827

## 828 **4.2. Carbon balance**

### 829 **4.2.1. Net ecosystem exchange**

830 We observed large variations of NEE between years, caused by varying management and  
831 environmental conditions. The maximum uptake of CO<sub>2</sub> measured in our study is close to the  
832 upper range of NEE reported for temperate grasslands (100 to 600 g C m<sup>-2</sup> y<sup>-1</sup>, (IPCC, 1996).  
833 On an annual basis our grassland site was a sink for atmospheric CO<sub>2</sub> in most years. NEE was  
834 only positive in 2004, which was likely to be due to a combination of slurry spreading and a  
835 high livestock density. Generally, grazing causes a very gradual impact on the CO<sub>2</sub> uptake as a  
836 part of the field is defoliated each day. The reduced leaf area index (LAI) then leads to a  
837 reduced CO<sub>2</sub> uptake by plants. In addition to the reduced LAI, grazing presents a source of CO<sub>2</sub>  
838 from animal respiration, thereby reducing the CO<sub>2</sub> sink of the grassland (Levy et al.,  
839 submitted). Indeed, annual NEE of all years correlated negatively with livestock density if  
840 years with cuts were excluded. On average over the 9 years the magnitude of the NEE on our  
841 grassland (-218.0 ± 154.5 g C m<sup>-2</sup> y<sup>-1</sup>) was close to the average NEE measured in a comparison  
842 of nine European grasslands over two years (240 ± 70 g C m<sup>-2</sup> y<sup>-1</sup>) by Soussana et al. (2007)  
843 and comparable to the CO<sub>2</sub> sink capacity of managed Irish grasslands measured by Byrne et al.  
844 (2007) (290 ± 50 g C m<sup>-2</sup> y<sup>-1</sup>) or Leahy et al. (2004) (257 g C m<sup>-2</sup> y<sup>-1</sup>). Despite high variability  
845 over the 9 years, the average NEE value was significantly different from zero (p < 0.05). The  
846 NEE represents the difference between the gross primary production (GPP) and the total  
847 ecosystem respiration (TER), both influenced by temperature, precipitation and management,  
848 though GPP is mainly controlled by PAR above a certain temperature threshold. The range of  
849 the calculated annual GPP (-982 to -2163 g C m<sup>-2</sup> y<sup>-1</sup>) and TER (972 to 2183 g C m<sup>-2</sup> y<sup>-1</sup>) from  
850 our field were within reported values for other managed grasslands. Gilmanov et al. (2007)  
851 reported the GPP of 18 intensively managed European grasslands ranging from 467 to 1874 g  
852 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  
853 values of 2000 g C m<sup>-2</sup> y<sup>-1</sup> for GPP and TER from a intensively grazed dairy pasture in New

854 Zealand.

855

#### 856 **4.2.2. Net biome production**

857 The total C budget (=NBP), which includes all components of C import and export in addition  
858 to the CO<sub>2</sub> exchange, was negative on average, meaning that C was stored in the grassland over  
859 the 9 years. However, due to the high variability between years, NBP was not significantly  
860 different from zero ( $p = 0.05$ ), suggesting that our site is carbon neutral. The average C storage  
861 value on our site ( $164 \pm 140 \text{ g C m}^{-2} \text{ y}^{-1}$ ) is higher than most estimates reported in literature, but  
862 due to the high annual variation, still within the range of reported values; Soussana et al. (2007)  
863 reported C storage estimates from European grazed and cut grasslands of  $104 \pm 73 \text{ g C m}^{-2} \text{ y}^{-1}$ ,  
864 and Mudge et al. (2011) reported for a grazed and cut grassland in New Zealand fluxes of  $59 \pm$   
865  $56 \text{ g C m}^{-2} \text{ y}^{-1}$  and  $90 \pm 56 \text{ g C m}^{-2} \text{ y}^{-1}$  in two consecutive years. NBP estimates from a Swiss  
866 grassland cut for silage was shown to sequester C at a rate of  $147 \pm 130 \text{ g C m}^{-2} \text{ y}^{-1}$  (Ammann  
867 et al., 2007), while estimates from a cut grassland in Germany was shown to vary from being a  
868 sink ( $-28 \text{ g C m}^{-2} \text{ y}^{-1}$ ) to being a source of C ( $+25 \text{ g C m}^{-2} \text{ y}^{-1}$ ), depending on years (Prescher et  
869 al., 2010). The inclusion of all C imports and exports lead to a weakening of the C sink strength  
870 assessed from NEE measurements in 5 years and even changed the grassland from being a sink  
871 to being a source in 2006. Due to the C export from harvest, C sequestration tends to be lower  
872 in cut systems. This is represented in the lower residual value of NPP in cut years (Figure 4a)  
873 compared to the residual value from grazed only years (Figure 4 b), whereas the residual value  
874 represents the C storage in the soil as well as the uncertainty of the budget. The grassland off-  
875 take from harvest weakened the annual C sink capacity assessed from the NEE by 51 % (2002)  
876 and 43 % (2003). However, it has to be kept in mind that the herbage yielded from cuts will  
877 end up as animal feed; C will be digested and respired off-site, releasing CO<sub>2</sub> to the atmosphere  
878 as well as being returned to the grassland as manure. It is likely that much of the organic C in  
879 the manure is decomposed and evolved to the atmosphere as CO<sub>2</sub>, with very little being  
880 retained in soil because of the lack of contact between manure and soil: there is some evidence  
881 of this from two long-term grassland experiments in the UK (Hopkins et al., 2009). When the  
882 only management was grazing (2004-2010) the NEE showed to be a good proxy of the NBP. In  
883 those years the plant biomass was digested on-site by the grazing animals and thereby  
884 contributed to total ecosystem respiration

885 Only a small fraction of the digested C was incorporated into the body of the grazing  
886 animal as meat and wool, while the largest part was respired as CO<sub>2</sub> shortly after intake.

887 We estimated that CH<sub>4</sub> emissions from grazing animals were only 0.7 % of NPP. Methane  
888 emissions were also measured by eddy covariance technique over several months in 2010 on  
889 the same field (Dengel et al., 2011). By dividing CH<sub>4</sub> fluxes by the number of sheep in the field  
890 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> y<sup>-1</sup>  
891 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  
892 ewes, showing that our estimates were realistic. Methane emissions from slurry spreading were  
893 relatively high on specific days (up to 0.28 g C m<sup>-2</sup> d<sup>-1</sup>, measured with chamber method),  
894 however, they were negligible on an annual basis as peaks only lasted for 2–3 days.

895  
896 Carbon leaching from managed grasslands has not been reported in many studies.  
897 Kindler et al. (2011) reported C leaching from various European ecosystems, whereas the  
898 measured data (2007) from our experimental field was part of the study. Our data (30.0 g C m<sup>-1</sup>  
899 y<sup>-1</sup>, average of two locations as published in Kindler et al. (2011) were close to the average  
900 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  
901 of 6.5–42.5 g C m<sup>-1</sup> y<sup>-1</sup>. Higher losses have been observed by McTiernan et al. (2001), who  
902 measured DOC export from grassland lysimeter plots treated with N fertilizer and slurry over  
903 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>  
904 <sup>1</sup>, all above what we measured in our study. Important factors controlling the magnitude of C  
905 leaching have been shown to be drainage, the topsoil C/N ratio and the saturation of the subsoil's  
906 sorption capacity for organic C (Kindler et al., 2011; McTiernan et al., 2001). In waterlogged  
907 soils the soil organic matter (SOM) decomposition and groundwater recharge tend to be  
908 reduced and thus the amount of C prone to leaching compared to that under more aerobic  
909 conditions associated with drainage. Although our field was drained more than 50 years ago,  
910 the drainage system does not operate very well, resulting in large puddles of standing water  
911 during prolonged periods of rain. The measured data used for the budget were taken at one  
912 sampling point, which was not in a waterlogged area. Therefore our leaching estimates are  
913 highly uncertain and could be significantly lower and C exports overestimated. The spatial  
914 heterogeneity within the grassland field caused by uneven water management as well as faeces  
915 and urine patches requires to sample at more points in order to obtain a representative leaching  
916 value.

917  
918 The systematic uncertainty of the C balance is mainly determined by the error of the  
919 CO<sub>2</sub> exchange, followed by the systematic uncertainty of the harvest export, organic fertilizer  
920 input and leaching losses. Combined uncertainties from all components lead to a total

921 systematic uncertainty of the C balance of  $18.3 \text{ g C m}^{-2} \text{ y}^{-1}$ .

922

### 923 **4.2.3 C sequestration**

924 Unlike forests, most of the C stored by grasslands is contained within soil organic matter.  
925 Carbon sequestration in grasslands can therefore be either determined directly from measuring  
926 soil organic carbon changes or indirectly by measuring the net C balance flux. If measuring soil  
927 C changes, the internationally recommended practice in carbon accounting is to express soil C  
928 stocks to a depth of 30 cm (IPCC, 1997). However, as the bulk density often changes over time  
929 with land use, the soil C content per unit ground area to a fixed depth will also change even  
930 without any change in the mass fraction of C in dry soil. By using the ESM method this  
931 problem is avoided, by considering the whole soil mass present in the 0-60 cm soil layer. A  
932 comparison of the C storage calculated from the net C flux balance from 2004-2010 with C  
933 stock changes measured from soil sample analysis (Table 7) show that, although the flux  
934 balance estimated a C sequestration, while based on soil measurements C was lost, neither  
935 value was significantly different from zero. A literature search by Soussana et al. (2010)  
936 showed that generally C sequestration calculations on grassland were lower if derived from  
937 SOC stock changes (average  $-5 \pm 30 \text{ g C m}^{-2} \text{ y}^{-1}$ ) compared to C flux balances (average  $-22 \pm 56 \text{ g C m}^{-2} \text{ y}^{-1}$ ), although these estimates were not significantly different from each other. However,  
938 in none of those reviewed studies were C flux and C stock change measured in the same field  
939 experiment. A reason for the discrepancy between calculation methods in our study might lie in  
940 a possible underestimation of C exports in the flux balance calculation, leading to an  
941 overestimation of C storage in the soil. One underestimated flux could be the export of DIC and  
942 DOC. Leaching was only measured in one year (2008), while values for remaining years were  
943 estimated using a simple regression model with an attributed high uncertainty of 30 % ( $4.9 \text{ g C m}^{-2} \text{ y}^{-1}$  of average fluxes). Further uncertainty could be due to the use of only one sampling  
944 location, which might not be representative of the whole field due to high spatial heterogeneity  
945 (see Sect. 4.1.2.). Indeed, Siemens (2003) hypothesized that the underestimation of C leaching  
946 from soils can explain a large part of the difference between atmosphere- and land-based  
947 estimates of the C uptake of European terrestrial ecosystems. Gapfilling can introduce  
948 uncertainties in the NEE data especially for years with low data capture. Furthermore,  $\text{CO}_2$   
949 losses from animal respiration could be underestimated at times due to the animals moving out  
950 of the footprint of the EC mast. Using animal respiration values from chamber experiments of  
951  $12.1 \text{ g CO}_2 \text{ kg}^{-1} \text{ live weight d}^{-1}$  for cows and  $11.7 \text{ g CO}_2 \text{ kg}^{-1} \text{ live weight d}^{-1}$  for sheep and  
952 lambs (Shane Troy, SRUC, personal communication), we estimated a maximum  $\text{CO}_2$  loss from  
953  
954


955 animal respiration of  $53 \text{ g C m}^{-2} \text{ y}^{-1}$  (2002-2010) or  $59 \text{ g C m}^{-2} \text{ y}^{-1}$  (2004-2010). So if we assume  
956 that all animal respiration has been missed by eddy covariance measurements then the C sink  
957 estimated from NEE measurements would be reduced by 24 % (2002-2010) or 33 % (2004-  
958 2010). This theoretical maximum 33% reduction would reduce the net carbon balance to  $\sim 122$   
959  $\text{g C m}^{-2} \text{ y}^{-1}$  (2004-2010).

960 In the literature, losses as well as storage of C at various rates have been reported from  
961 managed grasslands assessed from soil stock measurements. Soil stock measurements from our  
962 field are comparable with the C sequestration of  $10\text{-}30 \text{ g C m}^{-2} \text{ y}^{-1}$ , measured from US  
963 rangelands (0-60 cm, Schuman, et al., 2002), while Watson et al. (2007) measured a C storage  
964 at  $112\text{-}145 \text{ g C m}^{-2} \text{ y}^{-1}$  in the top 15 cm soil layer from a grazed Irish grassland. Bellamy et al.  
965 (2005) showed no evidence of increased C in the topsoil of grasslands in England and Wales  
966 and Hopkins et al. (2009) found no significant change of SOC over time in two UK long term  
967 experiments. Depending on the study, managed grasslands in Belgium were shown to either  
968 lose  $10 \text{ g C m}^{-2} \text{ y}^{-1}$ , Lettens et al., 2005a) or sequester carbon ( $4.4 \text{ g C m}^{-2} \text{ y}^{-1}$  in 0-30 cm,  
969 Goidts and Van Wesemael, 2007;  $22.5 \text{ g C m}^{-2} \text{ y}^{-1}$  in 0-30 cm, Lettens et al. 2005b). Schipper et  
970 al. (2007) reported losses of C from pastures in New Zealand over 20 years at an average rate  
971 of  $106 \text{ g C m}^{-2} \text{ y}^{-1}$  (top 100 cm), whereas these losses were a result of an earlier land use change  
972 from forestry. The above mentioned results are contrasting and inconclusive, because observed  
973 C sinks in grasslands are the effect of land management or land use change prior to the  
974 beginning of the C stock change measurement. Soussana et al (2014) concluded in a theoretical  
975 study that grassland SOC sequestration has a strong potential to partly mitigate the GHG  
976 balance of ruminant production systems at low grazing intensities, but not with intensive  
977 systems. Smith (2014) examined evidence from repeated soil surveys, long term grassland  
978 experiments and simple mass balance calculations and concluded that, although grasslands can  
979 act as C sinks, they cannot act as a perpetual C sink and thus could not be used as an offset for  
980 GHG emissions.

981


### 982 **4.3 Greenhouse gas budget**

983 In the overall N and C budget  $\text{N}_2\text{O}$  and  $\text{CH}_4$  emissions were negligible in terms of N and C  
984 losses from the system (1 – 8 % of total N losses and 0.6 - 4.5 % of total C losses,  
985 respectively). However, in terms of  $\text{CO}_2$  equivalents,  $\text{N}_2\text{O}$  emissions as well as  $\text{CH}_4$  emissions  
986 strongly affected the GHG budget. Since the radiative forcing effect of  $\text{N}_2\text{O}$  is 298 times  
987 greater than that of  $\text{CO}_2$  a relatively small emission of  $\text{N}_2\text{O}$  can exert a strong influence on the  
988 total radiative forcing budget of an ecosystem. Indeed, the sink strength of the NEE was

989 weakened by N<sub>2</sub>O emissions by 29 % over all years. Methane emissions from soil processes,  
990 manure input and animal excretion were negligible in terms of the C budget as well as in the  
991 GHG budget. In contrast, enteric fermentation proved to be an important GHG source. The  
992 positive correlation of CH<sub>4</sub> emissions with the stock density indicates that any changes in  
993 animal production will have a major impact on the global CH<sub>4</sub> budget. The weakening of the  
994 GHG sink strength of the NEE by N<sub>2</sub>O and CH<sub>4</sub> emissions, show the importance of those two  
995 gases in terms of global warming. Thus, adapting the management of grasslands by adding  
996 fertilizer or manure to increase plant growth and thus improve C sequestration in soils may  
997 increase N<sub>2</sub>O emissions, while changing land use from cropland to pasture in the attempt to  
998 reduce C losses from soils might lead to increased CH<sub>4</sub> losses from grazing animals. 

999

## 1000 **5. Conclusion**

1001 In our study only a small proportion of the N inputs from inorganic fertilizer and organic  
1002 manure were converted to animal outputs or stored in the soil, while the main part was lost  
1003 through leaching and gaseous emissions. An improvement of the NUE would mean both an  
1004 economic profit for the farmer as well as an environmental benefit. Estimates from flux budget  
1005 calculations indicated that our grassland was sequestering C. However, although grasslands can  
1006 act as C sinks,  they can not act as a perpetual C sink and thus could not be used as an offset for  
1007 GHG emissions. Instead, as it is easier and faster for soils to lose than to gain carbon, care must  
1008 be taken to preserve C loss by management options, rather than trying to increase carbon stocks  
1009 in grasslands. There was a discrepancy between soil stock measurements and flux budget  
1010 calculations for the C as well as the N budget. The reason for the discrepancy between C  
1011 storage estimates might lie in a possible underestimation of C exports such as leaching and  
1012 animal respiration as well as the uncertainty due to gapfilling in the NEE data. The N budget  
1013 storage might have been overestimated by the flux calculations through a possible  
1014 overestimation of N losses, mainly through leaching as well as through N<sub>2</sub> and NO<sub>x</sub>NH<sub>4</sub>  
1015 emissions. Our data have shown that the information about the potential of managed  
1016 grasslands to act as sinks or sources for GHG is important for mitigation and adaption  
1017 purposes. High plant productivity, stimulated by fertilisation, resulted in high plant CO<sub>2</sub>  
1018 fixation. However, increased N losses through N<sub>2</sub>O emissions counteracted the benefits of C  
1019 sequestration in terms of GHG emissions. Furthermore, CH<sub>4</sub> emissions from enteric  
1020 fermentation largely reduced the positive effect of CO<sub>2</sub> uptake, especially in years where NEE  
1021 rates were small. We therefore conclude that CO<sub>2</sub> exchange alone is not sufficient for the  
1022 estimation of the GWP of a managed grassland ecosystem.

1023

1024 **Acknowledgments**

1025 We would like to thank Chris Flechard for help with N deposition modelling, Patricia Ricci for  
1026 help with IPCC methodology used for CH<sub>4</sub> animal emissions and NO<sub>x</sub>/NH<sub>3</sub> volatilisation  
1027 calculations and Sue Owen for help with VOC emission calculations. We are grateful to Ivan  
1028 Simmons, Julia Drewer and Ian Leith for their help with field infrastructure. The authors  
1029 gratefully acknowledge funding from the EU project GREENGRASS (EC EVK<sup>2</sup>-CT2001-  
1030 00105) and the NitroEurope Integrated Project (contract 017841).

1031

1032 **Reference List**

1033 Allard, V., Soussana, J. F., Falcimagne, R., Berbigier, P., Bonnefond, J. M., Ceschia, E.,  
1034 D'hour, P., Henault, C., Laville, P., Martin, C., and Pinares-Patino, C.: The role of grazing  
1035 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  
1036 semi-natural grassland, *Agr. Ecosyst. Environ.*, 121.1-2, 47-58, 2007.

1037

1038 Ammann, C., Flechard, C. R., Leifeld, J., Neftel, A., and Fuhrer, J.: The carbon budget of  
1039 newly established temperate grassland depends on management intensity, *Agr. Ecosyst.*  
1040 *Environ.*, 121.1-2, 5-20, 2007.

1041

1042 Ammann, C., Neftel, A., Spirig, C., Leifeld, and J., Fuhrer, J.: Nitrogen balance of hay  
1043 meadows with and without fertilization, *Agrarforschung*, 16.9, 348-53, 2009.

1044

1045 Askegaard, M., Olesen, J. E., and Kristensen, K.: Nitrate leaching from organic arable  
1046 crop rotations: effects of location, manure and catch crop. *Soil Use Manage.*, 21.2, 181-88,  
1047 2005.

1048

1049 Atti, N., Rouissi H., and Othmane, M. H.: Milk production, milk fatty acid composition  
1050 and conjugated linoleic acid (CLA) content in dairy ewes raised on feedlot or grazing pasture,  
1051 *Livest. Sci.*, 104.1-2, 121-27, 2006.

1052

1053 Aubinet, M., Grelle, A., Ibrom, A., Rannik, U., Moncrieff, J., Foken, T., Kowalski, A. S.,  
1054 Martin, P. H., Berbigier, P., Bernhofer, C., Clement, R., Elbers, J., Granier, A., Grunwald, T.,  
1055 Morgenstern, K., Pilegaard, K., Rebmann, C., Snijders, W., Valentini, R., and Vesala, T.:  
1056 Estimates of the annual net carbon and water exchange of forests: The EUROFLUX  
1057 methodology, *Adv. Ecol. Res.*, 30, 113-75, 2000.

1058

1059 Ball, P. R. and Ryden, J. C.: Nitrogen Relationships in Intensively Managed Temperate  
1060 Grasslands, *Plant Soil*, 76.1-3, 23-33, 1984.

1061

1062 Bell, M. J., Rees, R.M., Cloy, J. M., Topp, C. F. E., Bagnall A., Chadwick, D. R.: Nitrous oxide  
1063 emissions from cattle excreta applied to a Scottish grassland: Effects of soil and climatic  
1064 conditions and a nitrification inhibitor, *Sci Total Environ*, 508, 343–353, 2015.

1065

1066 Bechmann, M., Eggestad, H. O., and Vagstad, N.: Nitrogen balances and leaching in four  
1067 agricultural catchments in southeastern Norway, *Environ. Pollut.*, 102, 493-99, 1998.

1068

1069 Bellamy, P. H., Loveland, P. J., Bradley, R. I., Lark R. M., and Kirk, G. J. D.: Carbon  
1070 losses from all soils across England and Wales 1978–2003, *Nature*, Vol 437, 245-48., 2005.  
1071

1072 Bleeker, A., Reinds G. J., Vermeulen A. T., de Vries W., and Erisman J. W.: Critical loads and  
1073 resent deposition thresholds of nitrogen and acidity and their exceedances at the level II and  
1074 level I monitoring plots in Europe, ECN report ECN-C-04-117, Petten, The Netherlands,  
1075 December 2004, 2004.  
1076

1077 Bleeker, A., Hicks, W. K., Dentener, F., Galloway, J., Erisman, J. W.: N deposition as a threat  
1078 to the World's protected areas under the Convention on Biological Diversity. *Environ. Pollut.*  
1079 159, 2280e2288, 2011.  
1080

1081 Brady, N. C., Weil, R. R.: *The Nature and Properties of Soils*, 13th Edition, Prentice Hall,  
1082 Upper Saddle River, NJ, 960 pp., ISBN 0-13-016763-0, 2002.  
1083

1084 Braun-Blanquet J.: *Pflanzensoziologie, Grundzüge der Vegetationskunde*, 3. Aufl. Springer,  
1085 1964, Verlag, Wien and New York.  
1086

1087 Byrne, K. A., Kiely, G., and Leahy, P: Carbon sequestration determined using farm scale  
1088 carbon balance and eddy covariance, *Agr. Ecosyst. Environ.*, 121.4, 357-64, 2007.  
1089

1090 Butterbach-Bahl, K., Gasche, R., Breuer, L., and Papen, H.: Fluxes of NO and N<sub>2</sub>O from  
1091 temperate forest soils: impact of forest type, N deposition and of liming on the NO and N<sub>2</sub>O  
1092 emissions, *Nutr. Cycl. Agroecosys.*, 48, 79-90, 1997.  
1093

1094 Caro, D., Davis S. J., Bastianoni, S., Caldeira K.: Global and regional trends in greenhouse gas  
1095 emissions from livestock. *Climatic Change*, 126:203–216, 2014  
1096

1097 Chen, W., McCaughey, W. P., and Grant, C. A.: Pasture type and fertilization effects on  
1098 N-2 fixation, N budgets and external energy inputs in western Canada, *Soil. Biol. Biochem.*, 36.8,  
1099 1205-12, 2004.  
1100

1101 Chirinda N., Kracher D., Laegdsmand M., Porter J. R., Olesen J.E., Petersen B. M., Doltra J.,  
1102 Kiese R., Butterbach-Bahl K. : Simulating soil N<sub>2</sub>O emissions and heterotrophic CO<sub>2</sub>  
1103 respiration in arable systems using FASSET and MoBiLE-DNDC. *Plant Soil*, 343, 139-169,  
1104 2011.  
1105

1106 Clayton, H., Arah, J. R. M., and Smith, K. A.: Measurement of Nitrous-Oxide Emissions from  
1107 Fertilized Grassland Using Closed Chambers. *J. Geophys. Res.-atmos.*, 99. D8, 16599-607,  
1108 1994.  
1109

1110 Cowan, N. J., Norman, P., Famulari, D., Levy, P. E., Reay, D. S., Skiba, U. M.(2015). Spatial  
1111 variability and hotspots of soil N<sub>2</sub>O fluxes from intensively grazed grassland. *Biogeosciences*  
1112 12 1585 - 1596, 2015.  
1113

1114 Cuttle, S. P. and Scholefield, D.: Management options to limit nitrate leaching from grassland.  
1115 *J. Cont. Hydrol.*, 20, 299-312, 1995.  
1116

1117 Davidson, E. A., 1991. Fluxes of nitrous oxide and nitric oxide from terrestrial ecosystems. In:  
1118 Rogers, J. E., Whitman, W. B. (Eds.), *Microbial Production and Consumption of Greenhouse*  
1119 *Gases: Methane, Nitrogen Oxides and Halomethanes*. American Society of Microbiology,



1120 Washington, DC, pp. 219–236.  
1121  
1122 Davison, B., Brunner, A., Ammann, C., Spirig, C., Jocher, M., and Neftel, A.: Cut-  
1123 induced VOC emissions from agricultural grasslands, *Plant Biol.*, 10.1, 76-85, 2008.  
1124  
1125 Dengel, S., Levy, P. E., Grace, J., Jones, S. K., and Skiba, U. M.: Methane emissions from  
1126 sheep pasture, measured with an open-path eddy covariance system, *Glob. Change Biol.*, 17.12,  
1127 3524-33, 2011.

1128 Di Marco, C., Skiba, U., Weston, K., Hargreaves, K., and Fowler, D.: Field scale N<sub>2</sub>O flux  
1129 measurements from grassland using eddy covariance, *Water Air Soil Poll.: Focus*, 4.6, 143-49,  
1130 2004.  
1131  
1132 Dobbie, K. E., McTaggart, I. P., and Smith, K. A.: Nitrous oxide emissions from intensive  
1133 agricultural systems: Variations between crops and seasons, key driving variables, and mean  
1134 emission factors, *J. Geophys. Res.-atmos.*, 104.D21, 26891-99, 1999.  
1135  
1136 Eriksen, J.: Nitrate leaching and growth of cereal crops following cultivation of contrasting  
1137 temporary grasslands, *J. Agr. Sci.*, 136, 271-81, 2001.  
1138  
1139 Erisman, J. W., Vanpul A., and Wyers P.: Parametrization of Surface-Resistance for the  
1140 Quantification of Atmospheric Deposition of Acidifying Pollutants and Ozone. *Atmos.*  
1141 *Environ.*, 28.16, 2595-607, 1994.  
1142  
1143 Flechard, C. R., Ambus, P., Skiba, U., Rees, R.M., Hensen, A., van Amstel, A., Pol-van  
1144 Dasselaar, A.V., Soussana, J. F., Jones, M., Clifton-Brown, J., Raschi, A., Horvath, L., Neftel,  
1145 A.; Jocher, M., Ammann, C., Leifeld, J., Fuhrer, J., Calanca, P., Thalman, E., Pilegaard, K., Di  
1146 Marco, C., Campbell, C., Nemitz, E., Hargreaves, K. J., Levy, P. E., Ball, B. C., Jones, S. K.,  
1147 van de Bulk, W. C. M., Groot, T., Blom, M., Domingues, R., Kasper, G., Allard, V., Ceschia,  
1148 E., Cellier, P., Laville, P., Henault, C., Bizouard, F., Abdalla, M., Williams, M., Baronti, S.,  
1149 Berretti, F., and Grosz, B.: Effects of climate and management intensity on nitrous oxide  
1150 emissions in grassland systems across Europe, *Agr. Ecosyst. Environ.*, 121.1-2, 135-52, 2007.  
1151  
1152 Flechard, C. R., Nemitz, E., Smith, R. I., Fowler, D., Vermeulen, A. T., Bleeker, A.,  
1153 Erisman, J. W., Simpson, D., Zhang, L., Tang, Y. S., and Sutton, M. A.: Dry deposition of  
1154 reactive nitrogen to European ecosystems: a comparison of inferential models across the  
1155 NitroEurope network, *Atmos. Chem. Phys.*, 11.6, 2703-28, 2011.  
1156  
1157 Flindt, R.: *Biologie in Zahlen: Eine Datensammlung in Tabellen mit ueber 10000 Einzelwerten.*  
1158 *Spektrum akademischer Verlag. Gustav Fischer*, 249, 2002.  
1159  
1160 Foken, T., and Wichura, B.: Tools for quality assessment of surface-based flux measurements,  
1161 *Agr. Forest Meteorol.*, 78.1-2, 83-105, 1996.  
1162  
1163 Galloway, J. N., Aber, J. D., Erisman, J. W., Seitzinger, S. P., Howarth, R.W., Cowling, E.B.,  
1164 Cosby, B.J.: The nitrogen cascade, *Bioscience*, 53.4, 341-56, 2003.  
1165  
1166 Gilmanov, T. G., Aires, L., Barcza, Z., Baron, V. S., Belelli, L., Beringer, J., Billesbach, D.,  
1167 Bonal, D., Bradford, J., Ceschia, E., Cook, D., Corradi, C., Frank, A., Gianelle, D., Gimeno, C.,  
1168 Gruenwald, T., Guo, H.Q., Hanan, N., Haszpra, L., Heilman, J., Jacobs, A., Jones, M. B.,  
1169 Johnson, D. A., Kiely, G., Li, S. G., Magliulo, V., Moors, E., Nagy, Z., Nasyrov, M., Owensby,

1170 C., Pinter, K., Pio, C., Reichstein, M., Sanz, M. J., Scott, R., Soussana, J. F., Stoy, P. C.,  
1171 Svejcar, T., Tuba, Z., and Zhou, G. S.: Partitioning European grassland net ecosystem CO<sub>2</sub>  
1172 exchange into gross primary productivity and ecosystem respiration using light response  
1173 function analysis, *Agr. Ecosyst. Environ.*, 121.1-2, 93-120, 2007.

1174  
1175 Goidts, E. and Van Wesemael, B.: Regional assessment of soil organic carbon changes under  
1176 agriculture in Southern Belgium (1955-2005), *Geoderma*, 141.3-4, 341-54, 2007.

1177  
1178 Groffman, P. M., Altabet, M. A., Bohlke, J. K., Butterbach-Bahl, K., David, M. B., Firestone,  
1179 M. K., Giblin, A. E., Kana, T. M., Nielsen, L. P., and Voytek, M. A.: Methods for measuring  
1180 denitrification: Diverse approaches to a difficult problem, *Ecol. Appl.*, 16.6, 2091-122, 2006.

1181 Haas E., Klatt, S., Fröhlich, A., Kraft, P., Werner, C., Kiese, R., Grote, R., Breuer, L., and  
1182 Butterbach-Bahl, K.: LandscapeDNDC: a process model for simulation of biosphere  
1183 atmosphere–hydrosphere exchange processes at site and regional scale, *Landscape Ecol.*, 28,  
1184 615-636, 2013.

1185  
1186 Molina-Herrera S., Haas, E., Klatt, S., Kraus, D., Augustin, J., Magliulo, V., Tallec, T.,  
1187 Ceschia, E., Ammann C., Loubet, B., Skiba, U., Jones, S., Brümmer, C., Butterbach-Bahl, K.,  
1188 Kiese, R.: A modelling study on mitigation of N<sub>2</sub>O emissions and NO<sub>3</sub> leaching at different  
1189 agricultural sites across Europe using LandscapeDNDC, *Science of the Total Environment*,  
1190 553, 128–140, 2016.

1191  
1192 Haas, E., Klatt, S., Frohlich, A., Kraft, P., Werner, C., Kiese, R., Grote, R., Breuer, L.,  
1193 Butterbach-Bahl, K.: LandscapeDNDC: a process model for simulation of biosphere–  
1194 atmosphere–hydrosphere exchange processes at site and regional scale. *Landscape Ecol*, 28,  
1195 615–636, 2013. DOI 10.1007/s10980-012-9772-

1196  
1197 Helfter, C., Campbell, C., Dinsmore, K. J., Drewer, J., Coyle, M., Anderson, M.,  
1198 Skiba, U., Nemitz, E., Billett, M. F., and Sutton, M. A.: Drivers of long-term variability in CO<sub>2</sub>  
1199 net ecosystem exchange in a temperate peatland, *Biogeosciences*, 12, 1799-1811, 2015

1200  
1201 Herrmann, B., Jones, S. K., Fuhrer, J., Feller, U., and Neftel, A.: N budget and NH<sub>3</sub>  
1202 exchange of a grass/clover crop at two levels of N application, *Plant Soil*, 235.2, 243-52, 2001.

1203  
1204 Hopkins, D. W., Waite, I. S., McNicol, J. W., Poulton, P. R., Macdonald, A. J. and  
1205 O'Donnell, A. G.: Soil organic carbon contents in long-term experimental grassland plots in the  
1206 UK (Palace Leas and Park Grass) have not changed consistently in recent decades, *Glob.*  
1207 *Change Biol.*, 15, 1739-1754, 2009.

1208  
1209 IPCC 1996. Climate change 1995. Impacts, adaptation and mitigation of climate change:  
1210 scientific, technical analysis, contribution of working group II to the 2nd assessment reports of  
1211 the IPCC. 1996. Intergovernmental Panel on Climate Change and Cambridge University Press  
1212 Cambridge UK.

1213  
1214 IPCC 2006a . Guidelines for National Greenhouse Gas Inventories, Agriculture, Forestry and  
1215 Other Land Use. Chapter 10: Emissions from Livestock and Manure Management.  
1216 Intergovernmental Panel on Climate Change (IPCC), Volume 4. 2006. Institute for Global  
1217 Environmental Strategies, Tokyo, Japan.

1218  
1219 IPCC 2006b. Guidelines for National Greenhouse Gas Inventories, Agriculture, Forestry and  
1220 Other Land Use. Chapter 11: N<sub>2</sub>O Emissions from Managed Soils, and CO<sub>2</sub> Emissions from

1221 Lime and Urea Application. Intergovernmental Panel on Climate Change (IPCC) Volume 4.  
1222 2013. Institute for Global Environmental Strategies, Tokyo, Japan.  
1223  
1224 IPCC 2013. Myhre, G., Shindell, D., Bréon, F. M., Collins, W., Fuglestedt, J., Huang, J.,  
1225 Koch, D., Lamarque, J.-F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G.,  
1226 Takemura, T. and Zhang, H.: Anthropogenic and Natural Radiative Forcing. In: Climate  
1227 Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth  
1228 Assessment Report of the Intergovernmental Panel on Climate Change. Stocker, T. F., D. Qin,  
1229 G. K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P. M.  
1230 Midgley (eds.). Cambridge University Press, Cambridge, United Kingdom and New York, NY,  
1231 USA. Anthropogenic and Natural Radiative Forcing  
1232  
1233 Janssens, I. A., Freibauer, A., Ciais, P., Smith, P., Nabuurs, G. J., Folberth, G.,  
1234 Schlamadinger, B., Hutjes, R. W. A., Ceulemans, R., Schulze, E. D., Valentini, R., and  
1235 Dolman, A.J.: Europe's terrestrial biosphere absorbs 7 to 12% of European anthropogenic CO<sub>2</sub>  
1236 emissions, *Science*, 300.5625, 1538-42, 2003.  
1237  
1238 Jarvis, S. C., Lovell, R. D., and Panayides, R.: Patterns of methane emission from excreta  
1239 of grazing animals, *Soil. Biol. Biochem.*, 27.12, 1581-88, 1995.  
1240  
1241 Jones, S. K., Famulari, D., Di Marco, C. F., Nemitz, E., Skiba, U. M., Rees, R. M., and  
1242 Sutton, M.A.: Nitrous oxide emissions from managed grassland: a comparison of eddy  
1243 covariance and static chamber measurements, *Atmospheric Measurement Techniques*, 4.10,  
1244 2179-94, 2011.  
1245  
1246 Jones, S. K., Rees, R. M., Kosmas, D., Ball, B. C., and Skiba, U. M.: Carbon sequestration  
1247 in a temperate grassland; management and climatic controls, *Soil Use Manage.*, 22.2, 132-42,  
1248 2006.  
1249  
1250 Jones, S. K., Rees, R. M., Skiba, U. M., and Ball, B. C.: Influence of organic and mineral N  
1251 fertiliser on N<sub>2</sub>O fluxes from a temperate grassland, *Agr. Ecosyst. Environ.*, 121.1-2, 74-83,  
1252 2007.  
1253  
1254 McKenzie, R. M., Özel, M. Z., Cape, J. N., Drewer, J., Dinsmore, K. J., Nemitz, E.,  
1255 Hamilton, J. F., Sutton, M. A., Gallagher, M. W. and Skiba, U.: The import and export of  
1256 organic nitrogen species at a Scottish ombrotrophic peatland, *Biogeosciences Discussions*, 12,  
1257 515-554, 2015.  
1258  
1259 Kesik M., Ambus P., Baritz R., Brüggemann N., Butterbach-Bahl K., Damm M., Duyzer  
1260 J., Horváth L., Kiese R., Kitzler B., Leip A., Li C., Pihlatie M., Pilegaard K., Seufert G.,  
1261 Simpson D., Skiba U., Smiatek G., Vesala T., and Zechmeister-Boltenstern S.: Inventories of  
1262 N<sub>2</sub>O and NO emissions from European forest soils, *Biogeosciences*, 2, 353-375, 2005.  
1263  
1264 Kiese R., Heinzeller C., Werner C., Wochele S., Grote R., and Butterbach-Bahl K.:  
1265 Quantification of nitrate leaching from German forest ecosystems by use of a process oriented  
1266 biogeochemical model. *Environ. Pollut.*, 159, 3204-3014, 2011.  
1267  
1268 Kim, J., Verma, S. B., and Clement, R. J.: Carbon-Dioxide Budget in A Temperate  
1269 Grassland Ecosystem. *J. Geophys. Res-Atmos.*, 97.D5, 6057-63, 1992.  
1270  
1271 Kindler, R., Siemens, J., Kaiser, K., Walmsley, D. C., Bernhofer, C., Buchmann,

1272 N., Cellier, P., Eugster, W., Gleixner, G., Grunwald, T., Heim, A., Ibrom, A., Jones, S. K.,  
1273 Jones, M., Klumpp, K., Kutsch, W., Larsen, K. S., Lehuger, S., Loubet, B., McKenzie, R.,  
1274 Moors, E., Osborne, B., Pilegaard, K., Reibmann, C., Saunders, M., Schmidt, M. W. I.,  
1275 Schrumpf, M., Seyfferth, J., Skiba, U., Soussana, J. F., Sutton, M.A., Tefs, C., Vowinckel, B.,  
1276 Zeeman, M. J., and Kaupenjohann, M.: Dissolved carbon leaching from soil is a crucial  
1277 component of the net ecosystem carbon balance, *Glob. Change Biol.*, 17.2, 1167-85, 2011.  
1278  
1279 Kramberger, M., Podvršnik, A., Gselman, V., Šuštar, J., Kristl, M., Muršec, M., Lešnik, D.,  
1280 Škorjanc: The effects of cutting frequencies at equal fertiliser rates on bio-diverse permanent  
1281 grassland: Soil organic C and apparent N budget. *Agriculture, Ecosystems & Environment*,  
1282 Volume 212, , *Pages 13-20B*, 2015.  
1283  
1284 Lal, R.: Carbon emission from farm operations. *Environ. Int.*, 30, 981–990, 2004.  
1285  
1286 Laws, J. A., Falge, E., Gu, L., Baldocchi, D. D., Bakwin, P., Berbigier, P., Davis, K.,  
1287 Dolman, A. J., Falk, M., Fuentes, J. D., Goldstein, A., Granier, A., Grelle, A., Hollinger, D.,  
1288 Janssens, I. A., Jarvis, P., Jensen, N. O., Katul, G., Mahli, Y., Matteucci, G., Meyers, T.,  
1289 Monson, R., Munger, W., Oechel, W., Olson, R., Pilegaard, K., Paw, K. T., Thorgeirsson, H.,  
1290 Valentini, R., Verma, S., Vesala, T., Wilson, K., and Wofsy, S.: Comparison of grassland  
1291 management systems for beef cattle using self-contained farmlets: effects of contrasting  
1292 nitrogen inputs and management strategies on nitrogen budgets, and herbage and animal  
1293 production, *Agr. Ecosyst. Environ.*, 80.3, 243-54, 2000.  
1294  
1295 Leahy, P., Kiely, G., and Scanlon, T. M.: Managed grasslands: A greenhouse gas sink or  
1296 source? *Geophys. Res. Lett.*, 31.20, 2004.  
1297  
1298 Ledgard, S. F.: Nitrogen cycling in low input legume-based agriculture, with emphasis on  
1299 legume/grass pastures, *Plant Soil*, 228.1, 43-59, 2001.  
1300  
1301 Ledgard, S. F., Menneer, J.C., Dexter, M. M., Kear, M. J., Lindsey, S., Peters, J. S., and  
1302 Pacheco, D.: A novel concept to reduce nitrogen losses from grazed pastures by administering  
1303 soil nitrogen process inhibitors to ruminant animals: A study with sheep. *Agr. Ecosyst.*  
1304 *Environ.*, 125.1-4, 148-58, 2008.  
1305  
1306 Lettens, S., Van Orshoven, J, Van Wesemael, B., De Vos, B., and Muys, B.: Stocks and  
1307 fluxes of soil organic carbon for landscape units in Belgium derived from heterogeneous data  
1308 sets for 1990 and 2000, *Geoderma*, 127.1-2, 11-23, 2005 a.  
1309  
1310 Lettens, S., Van Orshoven, J., Van Wesemael, B., Muys, B., and Perrin, D.: Soil organic  
1311 carbon changes in landscape units of Belgium between 1960 and 2000 with reference to 1990,  
1312 *Glob. Change Biol.*, 11.12, 2128-40, 2005b.  
1313  
1314 Li, C. S., Frolking, S., and Frolking, T. A.: A Model of Nitrous-Oxide Evolution from Soil  
1315 Driven by Rainfall Events. 1. Model Structure and Sensitivity, *J. Geophys. Res.-atmos.*, 97.D9,  
1316 9759-76, 1992.  
1317  
1318 Li C.: Modeling trace gas emissions from agricultural ecosystems, *Nutr. Cycl. Agroecosys.* 58,  
1319 259-276, 2000  
1320 Luo, J., Tillman, R. W., and Ball, P. R.: Grazing effects on denitrification in a soil under  
1321 pasture during two contrasting seasons, *Soil. Biol. Biochem.*, 31.6, 903-12, 1999.  
1322

1323 MAFF, 1990. Ministry of agriculture fisheries and food. UK tables of nutritive value and  
1324 chemical composition of feedingstuffs, 1990. 1st ed. Rowett Research.  
1325

1326 McTaggart, I. P., Clayton, H., Parker, J., Swan, L., and Smith, K. A.: Nitrous oxide emissions  
1327 from grassland and spring barley, following N fertiliser application with and without  
1328 nitrification inhibitors, *Biol. Fert. Soils*, 25.3, 261-68, 1997.  
1329

1330 MAFF. Guidelines for farmers in NVZs. 32 pp. 1998. Ministry of Agriculture, Fisheries and  
1331 Food, London.  
1332

1333 R. M. McKenzie, J. N. Cape, M. Z. Ozel, J. Drewer, K. J. Dinsmore, J. F. Hamilton, E. Nemitz,  
1334 M. A. Sutton, M. W. Gallagher, and U. Skiba.: The import and export of organic nitrogen  
1335 species at a Scottish ombrotrophic peatland, *Biogeosciences Discussions*, 12, 515-554, 2015.  
1336

1337 McTiernan, K. B., Jarvis, S.C., Scholefield, D., and Hayes, M. H. B.: Dissolved organic carbon  
1338 losses from grazed grasslands under different management regimes, *Water Res*, 35.10, 2565-  
1339 69, 2001.  
1340

1341 Mudge, P. L., Wallace, D. F., Rutledge, S., Campbell, D. I., Schipper, L. A., Hosking, C. L.:  
1342 Carbon balance of an intensively grazed temperate pasture in two climatically contrasting  
1343 years, *Agr. Ecosyst. Environ.*, 144.1, 271-80, 2011.  
1344

1345 Neeteson, J. J. and Hassink, J.: Nitrogen Budgets of Three Experimental and Two Commercial  
1346 Dairy Farms in the Netherlands. In: M.K. van Ittersum & S.C. van de Geijn (Eds.), *Perspectives  
1347 for Agronomy. Adopting ecological principles and managing resource use*. Elsevier Science  
1348 BV, Amsterdam. *Developments in Crop Science* 25, pp. 171-178. 1997.  
1349

1350 Nunez, R. P., Demanet, R., Alfaro, M., and Mora, M. L.: Nitrogen Soil Budgets in  
1351 Contrasting Dairy Grazing Systems of Southern Chile, A Short-Term Study, *Rev. Cienc. Suelo  
1352 Nutr.*, 10.2, 170-83, 2010.  
1353

1354 Oenema, O., Velthof, G.L., Yamulki, and S., Jarvis, S.C.: Nitrous oxide emissions from grazed  
1355 grassland, *Soil Use Manage.*, 13.4, 288-95, 1997.  
1356

1357 Owens, L. B., Edwards, W. M., and Vankeuren, R. W.: Groundwater Nitrate Levels Under  
1358 Fertilized Grass and Grass-Legume Pastures. *J. Environ. Qual.*, 23.4, 752-58, 1994.

1359 Phoenix, G. K., Hicks, W.K., Cinderby, S., Kuylenstierna, J.C.I., Stock, W. D., Dentener, F. J., Giller,  
1360 K. E., Austin, A. T., Lefroy, R. D. B., Gimeno, B. S., Ashmore, M. R., Ineson, P.: Atmospheric  
1361 nitrogen deposition in world biodiversity hotspots: the need for a greater global perspective in assessing  
1362 N deposition impacts. *Global Change Biology*, 12, pp. 470-476, 2006.  
1363

1364 Prescher, A. K., Grunwald, T. and Bernhofer, C.: Land use regulates carbon  
1365 budgets in eastern Germany: From NEE to NBP, *Agr. Forest Meteorol.*, 150.7-8, 1016-25,  
1366 2010.  
1367

1368 Rees, R. M., Bingham, I. J., Baddeley, J. A., and Watson, C.A.: The role of plants and land  
1369 management in sequestering soil carbon in temperate arable and grassland ecosystems.  
1370 *Geoderma*, 128.1-2, 130-54, 2005.  
1371

1372 Rees, R. M., Bingham, I. J.; Baddeley, J. A.; and Watson, C.A.: Nitrous oxide mitigation in UK  
1373 agriculture. *Soil Sci. Plant Nutr.*, 59.1, 3-15, 2013.

1374

1375 Reichstein, M., Falge, E., Baldocchi, D., Papale, D., Aubinet, M., Berbigier, P., Bernhofer, C.,  
1376 Buchmann, N., Gilmanov, T., Granier, A., Grunwald, T., Havrankova, K., Ilvesniemi, H.,  
1377 Janous, D., Knohl, A., Laurila, T., Lohila, A., Loustau, D., Matteucci, G., Meyers, T.,  
1378 Miglietta, F., Ourcival, J. M., Pumpanen, J., Rambal, S., Rotenberg, E., Sanz, M., Tenhunen, J.,  
1379 Seufert, G., Vaccari, F., Vesala, T., Yakir, D., and Valentini, R.: On the separation of  
1380 net ecosystem exchange into assimilation and ecosystem respiration: review and improved  
1381 algorithm, *Global Change Biology*, 11.9, 1424-39, 2005.

1382

1383 Rose-Marie Rytter, R. M., Rytter, L., Hogbom, L.: Carbon sequestration in willow (*Salix* spp.)  
1384 plantations on former arable land estimated by repeated field sampling and C budget  
1385 calculation. *Biomass and Bioenergy*, 83, 483-492, 2015.

1386

1387 Roche J.: *The International Wool Trade*. Woodhead Publishing, 1995.

1388

1389 Rudaz, A. O., Walti, E., Kyburz, G., Lehmann, P., and Fuhrer, J.: Temporal variation in N<sub>2</sub>O  
1390 and N<sub>2</sub> fluxes from a permanent pasture in Switzerland in relation to management, soil water  
1391 content and soil temperature, *Agr. Ecosyst. Environ.*, 73.1, 83-91, 1999.

1392

1393 Ryden, J. C., Ball, P. R., and Garwood, E. A.: Nitrate Leaching from Grassland, *Nature*  
1394 311.5981, 50-53, 1984a.

1395

1396 Ryden, J. C., and McNeill, J. E.: Application of the micrometeorological mass balance method  
1397 to the determination of ammonia loss from a grazed sward, *J. Sci. Food Agric.*, 35, 1297-310.  
1398 1984.

1399

1400 Schipper, L. A., Baisden, W. T., Parfitt, R. L., Ross, C., Claydon, J. J., and Arnold, G.: Large  
1401 losses of soil C and N from soil profiles under pasture in New Zealand during the past 20 years,  
1402 *Glob. Change Biol*, 13.6, 1138-44, 2007.

1403

1404 Scholefield, D., Lockyer, D. R., Whitehead, D. C., and Tyson, K. C. A.: A Model to Predict  
1405 Transformations and Losses of Nitrogen in Uk Pastures Grazed by Beef-Cattle, *Plant Soil*,  
1406 132.2, 165-77, 1991.

1407

1408 Scholefield, D., Tyson, K. C., Garwood, E.A., Armstrong, A.C., Hawkins, J., and Stone, A. C.:  
1409 Nitrate Leaching from Grazed Grassland Lysimeters - Effects of Fertilizer Input, Field  
1410 Drainage, Age of Sward and Patterns of Weather, *J. Soil Sci.*, 44.4, 601-13, 1993.

1411

1412 Schuman, G. E., Janzen, H. H., and Herrick, J. E.: Soil carbon dynamics and potential carbon  
1413 sequestration by rangelands, *Environ. Pollut.*, 116.3, 391-96, 2002.

1414

1415 Schrumpf, M., Schulze, E. D., Kaiser, K., and Schumacher, J.: How accurately can soil organic  
1416 carbon stocks and stock changes be quantified by soil inventories? *Biogeosciences*, 8, 1193-  
1417 1212, 2011.

1418

1419 Scottish Agricultural College, *Farm management handbook SAC*; edt. Linda Chadwick,  
1420 Edinburgh, 1995.

1421

1422 Scottish Agricultural College, *Technical Note TN652: Fertiliser recommendations for*

1423 grasslands. A., Sinclair, P.A., Shipway, B., Crooks, 2013.  
1424  
1425 Siemens, J.: The European carbon budget: A gap, *Science*, 302.5651, 1681, 2003.

1426 Simpson, D., Fagerli, H., Jonson, J. E., Tsyro, S., Wind, P., and Tuovinen, J.-P.: Transboundary  
1427 Acidification, Eutrophication and Ground Level Ozone in Europe. Part I: Unified EMEP Model  
1428 Description. EMEP Status Report 2003, ISSN 0806-4520, Det . 2003. Meteorologisk Institutt,  
1429 Oslo, 2003.  
1430  
1431 Skiba, U., Jones, S. K., Drewer, J., Helfter, C., Anderson, M., Dinsmore, K., McKenzie, R.,  
1432 Nemitz, E., and Sutton, M. A.: Comparison of soil greenhouse gas fluxes from extensive and  
1433 intensive grazing in a temperate maritime climate, *Biogeosciences* 10, 1231-1241, 2013.  
1434  
1435 Smith, R. I., Fowler, D., Sutton, M. A., Flechard, C., and Coyle, M.: Regional estimation of  
1436 pollutant gas dry deposition in the UK: model description, sensitivity analyses and outputs.  
1437 *Atmos. Environ.*, 34.22, 3757-77, 2000.  
1438  
1439 Smith, P. : How long before a change in soil organic carbon can be detected? *Glob. Change*  
1440 *Biol.*, 10.11, 1878-83, 2004  
1441  
1442 Smith, P., Haberl, H., Popp, A., Erb, K.-H., Lauk, C., Harper, R., et al.: How much land based  
1443 greenhouse gas mitigation can be achieved without compromising food security and  
1444 environmental goals? *Global Change Biology*, 19, 2285–2302, 2013.  
1445  
1446 Smith, P.: Do grasslands act as a perpetual sink for carbon? *Glob. Change Biol.*, 20, 2708–  
1447 2711, 2014.  
1448  
1449 Soussana, J. F., Loiseau, P., Vuichard, N., Ceschia, E., Balesdent, J., Chevallier, T., and  
1450 Arrouays, D.: Carbon cycling and sequestration opportunities in temperate grasslands, *Soil Use*  
1451 *Manage.*, 20, 219-30, 2004.  
1452  
1453 Soussana, J. F., Allard, V, Pilegaard, K., Ambus, P., Amman, C., Campbell, C., Ceschia, E.,  
1454 Clifton-Brown, J., Czobel, S., Domingues, R., Flechard, C., Fuhrer, J., Hensen, A., Horvath, L.,  
1455 Jones, M., Kasper, G., Martin, C., Nagy, Z., Neftel, A., Raschi, A., Baronti, S., Rees, R.M.,  
1456 Skiba, U., Stefani, P., Manca, G., Sutton, M., Tubaf, Z., and Valentini, R.: Full accounting of  
1457 the greenhouse gas (CO<sub>2</sub>, N<sub>2</sub>O, CH<sub>4</sub>) budget of nine European grassland sites, *Agr. Ecosyst.*  
1458 *Environ.*, 121.1-2, 121-34, 2007.  
1459  
1460 Soussana, J. F., Tallec, T., and Blanfort, V.: Mitigating the greenhouse gas balance of ruminant  
1461 production systems through carbon sequestration in grasslands, *Animal*, 4.3, 334-50, 2010.  
1462  
1463 Soussana, J. F. and Lemaire, G.: Coupling carbon and nitrogen cycles for environmentally  
1464 sustainable intensification of grasslands and crop-livestock systems, *Agr. Ecosyst. Environ.*,  
1465 190 9–17, 2014.  
1466  
1467 Stange F., Butterbach-Bahl K., Papen H., Zechmeister-Boltenstern S., Li C., and Aber J.: A  
1468 process oriented model of N<sub>2</sub>O and NO emissions from forest soils: 2: sensitivity analysis and  
1469 validation, *J. Geophys. Res.*, 105, 4385-4398, 2000.  
1470

- 1471 Stewart, A. A., Little, S. M., Ominski, K.H., Wittenberg, K. M., and Janzen, H. H.: Evaluating  
 1472 greenhouse gas mitigation practices in livestock systems: an illustration of a whole-farm  
 1473 approach, *J. Agr. Jci.*, 147, 367-82, 2009.
- 1474  
 1475 Streeter, J.: Inhibition of Legume Nodule Formation and N-2 Fixation by Nitrate, *Crc. Cr. Rev.*  
 1476 *Plant Sci.*, 7.1, 1-23, 1988.
- 1477  
 1478 Sutton, M. A., Tang, Y. S., Miners, B., and Fowler, D.: A New Diffusion Denuder System for  
 1479 Long-Term, Regional Monitoring of Atmospheric Ammonia and Ammonium, *Water Air Soil*  
 1480 *Poll.: Focus*, 1.5-6, 145-56, 2001.
- 1481  
 1482 Tang, Y. S., Simmons, I.; van Dijk, N., Di Marco, C., Nemitz, E., Dammgén, U., Gilke, K.,  
 1483 Djuricic, V., Vidic, S., Gliha, Z., Borovecki, D., Mitosinkova, M., Hanssen, J. E., Uggerud, T.  
 1484 H., Sanz, M.J., Sanz, P., Chorda, J.V., Flechard, C. R., Fauvel, Y., Ferm, M., Perrino, C., and  
 1485 Sutton, M.A.: European scale application of atmospheric reactive nitrogen measurements in a  
 1486 low-cost approach to infer dry deposition fluxes, *Agr. Ecosyst. Environ.*, 133.3-4, 183-95,  
 1487 2009.
- 1488  
 1489 Tuovinen, J. P., Emberson, L., and Simpson, D.: Modelling ozone fluxes to forests for risk  
 1490 assessment: status and prospects, *Ann. For Sci.*, 66.4, 2009.
- 1491  
 1492 Tyson, K. C., Garwood, E. A., Armstrong, A.C., and Scholefield, D.: Effects of Field Drainage  
 1493 on the Growth of Herbage and the Liveweight Gain of Grazing Beef-Cattle, *Grass Forage Sci.*,  
 1494 47.3, 290-301, 1992.
- 1495  
 1496 Twigg, M. M., House, E., Thomas, R., Whitehead, J., Phillips, G.J., Famulari, D., Fowler, D.,  
 1497 Gallagher, M.W., Cape, J.N., Sutton, M.A., Nemitz, E.: Surface/atmosphere exchange and  
 1498 chemical interactions of reactive nitrogen compounds above a manured grassland *Agricultural*  
 1499 *and Forest Meteorology*, Volume 151, Issue 12, , Pages 1488-1503, 2011.
- 1500  
 1501 Van der Hoek, K. W.: Nitrogen efficiency in global animal production, *Environ. Pollut.*, 102,  
 1502 127-32, 1998.
- 1503  
 1504 Van der Salm, C., Dolfing, J., Heinen, M., and Velthof, G. L.: Estimation of nitrogen losses via  
 1505 denitrification from a heavy clay soil under grass, *Agr. Ecosyst. Environ.*, 119, 311-319, 2007.
- 1506  
 1507 Velthof, G. L. and Oenema, O.: Nitrous oxide fluxes from grassland in the Netherlands .2.  
 1508 Effects of soil type, nitrogen fertilizer application and grazing, *Eur. J. Soil Sci.*, 46.4, 541-49,  
 1509 1995.
- 1510  
 1511 Velthof, G. L. and Oenema, O.: Effects of nitrogen fertilization and grazing on the emission of  
 1512 nitrous oxide from grassland. 1997. RIVM-Dutch National Research Programme on Global  
 1513 Air Pollution and Climate Change, Velthof, G.L. & Oenema, O. (eds.), Bilthoven (1997)  
 1514 Report no. 410 100 055.
- 1515  
 1516 Waring, R. H., Landsberg J. J., and Williams, M.: Net primary production of forests: a constant  
 1517 fraction of gross primary production? *Tree Physiol.*, 18.2, 129-34, 1998.
- 1518  
 1519 Watson, C. J., Jordan, C., Kilpatrick, D., McCarney, B., and Stewart, R.: Impact of grazed  
 1520 grassland management on total N accumulation in soil receiving different levels of N inputs.  
 1521 *Soil Use Manage.*, 23.2, 121-128, 2007.



1522  
1523 Whitehead, D. C., Pain, B. F., and Ryden J. C.: Nitrogen in UK grassland agriculture, J. R.  
1524 Agric. Soc., 147, 190-201. 1986.  
1525  
1526 Wolf, B., Kiese R., Chen W.W., Grote R., Zheng X. H., and Butterbach-Bahl K.: Modeling  
1527 N<sub>2</sub>O emissions from steppe in Inner Mongolia, China, with consideration of spring thaw and  
1528 grazing intensity, Plant Soil, 350, 297-310, 2012.  
1529  
1530 Zhang, L. M., Gong, S.L., Padro, J., and Barrie, L.: A size-segregated particle dry deposition  
1531 scheme for an atmospheric aerosol module, Atmos. Environ., 35.3, 549-60, 2001.  
1532  
1533 Zhang, L., Brook, J. R. and Vet R.: A revised parameterization for gaseous dry deposition in  
1534 air-quality models. Atmos. Chem. Phys., 3, 2067-82, 2003.  
1535  
  
1536  
1537

1538 **Tables**

1539

1540 Table 1. Average annual livestock densities [LSU ha<sup>-1</sup> y<sup>-1</sup>].

1541

	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

1542

1543

1544 Table 2. Weather characteristics of each measurement year.

1545

	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 [%]	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	180	196	156	177	151	186	193	242	226

1546

1547

1548 Table 3. Systematic uncertainties attributed to each budget component. Combined uncertainties were  
1549 calculated according to simple Gaussian error propagation rules.

1550

Nitrogen budget component	N [%]	Carbon budget component	C [%]
Mineral fertiliser	1		
Organic manure <sup>a</sup>	20	Organic manure <sup>a</sup>	20
Harvest <sup>b</sup>	16	Harvest <sup>b</sup>	11
Leaching <sup>c</sup>	30	Leaching <sup>c</sup>	30
Animal (wool and meat) <sup>d</sup>	12	Animal (wool and meat) <sup>a</sup>	12
Wet deposition	30	CH <sub>4</sub> soil	160
Dry deposition <sup>e</sup>	80	CH <sub>4</sub> enteric	20
N <sub>2</sub> O	30	CH <sub>4</sub> excretion	20
NO <sub>x</sub> soil	30	CH <sub>4</sub> organic	120
NH <sub>4</sub> volatilisation	30		
NO <sub>x</sub> volatilisation	50		
N <sub>2</sub>	30		

1551 combined uncertainties of C and N analysis (17%) and volume spread (10%)

1552 combined uncertainty of total C (4%) and N (12%) analysis and farmer's estimate in harvest amount (10%)

1553 modelled 30, how much for measurements?

1554 combined uncertainties from animal numbers (5%), animal weight (10%) and literature values for C and N content for  
1555 meat and wool (3%)

1556 combined uncertainty of DELTA sample analysis (7%) and variation of outputs from the four models (80%)

1557

1558 |

1559

Table 4. 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 [ $\text{g N m}^{-2} \text{y}^{-1}$ ]. Negative numbers represent uptake while positive numbers represent loss of N from this grassland ecosystem.

	2002	2003	2004	2005	2006	2007	2008	2009	2010	2002-2010		
										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	-0.5*	-0.5	0.1	0.2
Dry deposition	-0.5	-0.4	-0.3	-0.3	-0.2	-0.3	-0.2	-0.2	-0.3*	-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	0.2	0.5	1.9	2.6	2.2	2.4	1.5	1.1	1.2	1.5	0.5	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
Leaching	15.0	0.1	0.1	4.6	10.6	4.2	5.6	2.6	5.3*	5.3	3.4	1.6
N <sub>2</sub>	3.7	2.2	1.3	1.7	2.8	3.0	3.3	4.1	2.8*	2.8	0.6	0.8
N <sub>2</sub> O	1.1	0.1	0.1	0.4	0.9	1.3	0.8	0.4	0.4	0.6	0.3	0.2
NO <sub>x</sub> (soil)	0.3	0.1	0	0.1	0.2	0.2	0.3	0.1	0.1	0.2	0.1	0.1
NO <sub>x</sub> ,NH <sub>3</sub> (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)	0.4	0.7	1.7	1.3	1.3	1.6	1.5	1.3	1.2	1.2	0.3	0.6
N balance	6.3	-7.0	-10.6	-17.9	-2.5	-3.1	-10.3	-13.2	-6.6	-7.2	4.6	2.1

\*average value of 2002-2009

Table 5. 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 [ $\text{g C m}^{-2} \text{y}^{-1}$ ]. Negative numbers represent uptake, while positive numbers represent loss of C from the grassland ecosystem.

	2002	2003	2004	2005	2006	2007	2008	2009	2010	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
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	0.9	3.0	11.5	15.8	13.1	14.5	9.1	6.4	7.3	9.1	3.4	1.1
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
Leaching*	25.1	7.0	22.1	18.7	19.4	15.4	17.0	6.8	16.4*	16.4	4.3	4.9
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
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)	-137.8	-73.3	57.7	-154.7	29.3	-78.6	-84.0	-587.6	-440.3	-163.2	139.5	15.9

\*average value of 2002-2009

**Table 6.** 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.

	N <sub>2</sub> O flux [g N m <sup>-2</sup> y <sup>-1</sup> ]	Total N input [g N m <sup>-2</sup> y <sup>-1</sup> ]	EF [%]
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 7.** N and C budget (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	-4.51 (2.64)	29.08 (38.19)
flux budget	-9.20 (4.10)	-180.7 (180)

## Figure captions

Figure 1. Livestock density (c), nitrogen (c) and carbon (b) input and export from inorganic and organic fertiliser and harvest from 2002-2010.

Figure 2. Maximum, minimum and average monthly air temperature, derived from daily averages (a) and monthly cumulative rainfall and soil water content (b) from 2002-2010.

Figure 3. 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 4. 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 5. Average greenhouse gas fluxes, net GHG exchange (NGHGE) and attributed net GHG balance (NGHGB, includes  $FC_{\text{org fert}}$ ,  $FC_{\text{animal}}$ ,  $FC_{\text{leaching}}$ ,  $FC_{\text{harvest}}$ ) for 2002-2010. Positive values correspond to losses and negative values to storage of greenhouse gases to and from the grassland system, respectively. The  $CH_4$  component comprises  $CH_4$  fluxes from enteric fermentation, animal excretion, slurry application and soil exchange, while the  $N_2O$  component is the  $N_2O$  flux from the soil. Global warming potentials of 298 and 25 were used for  $N_2O$  and  $CH_4$  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.

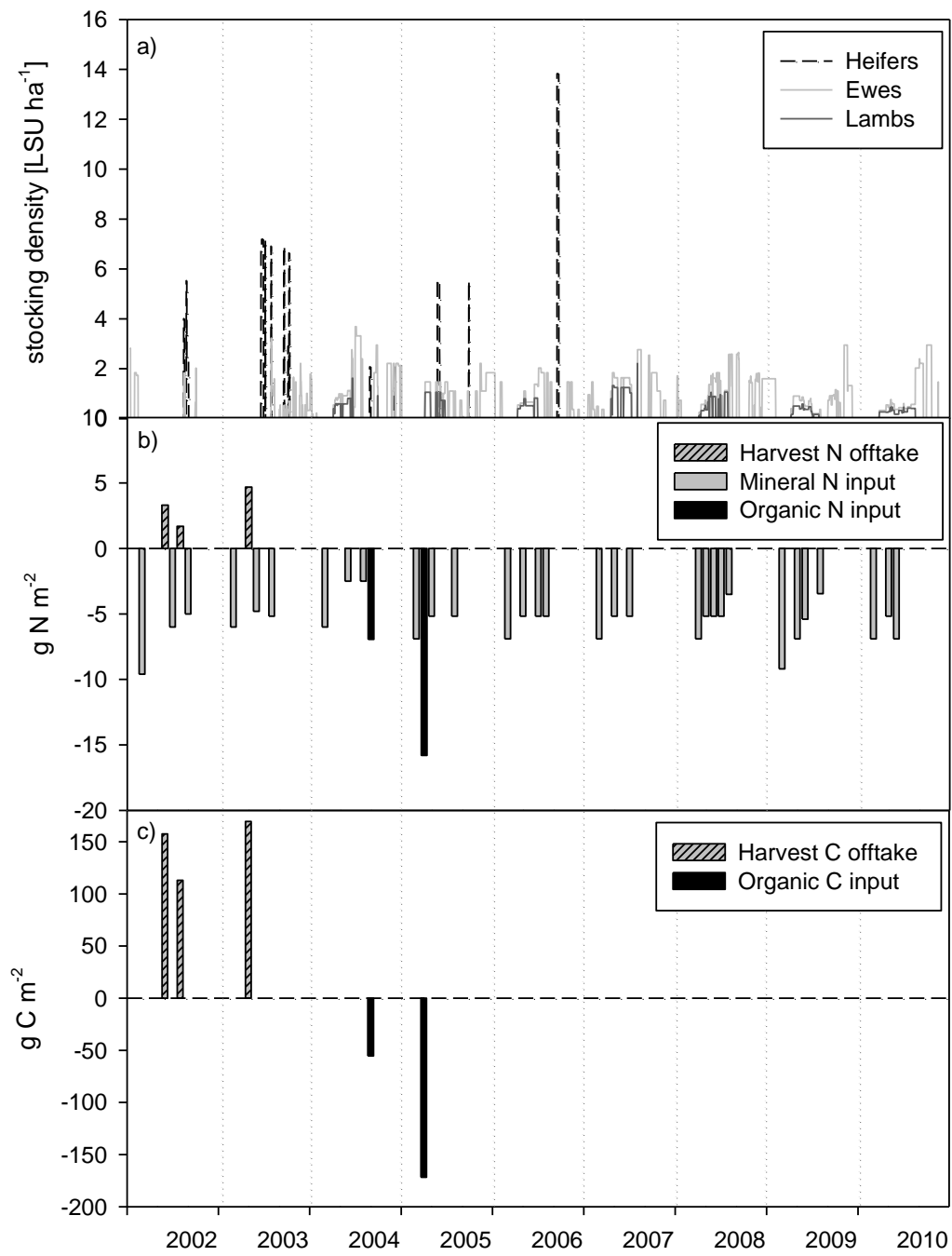


fig01

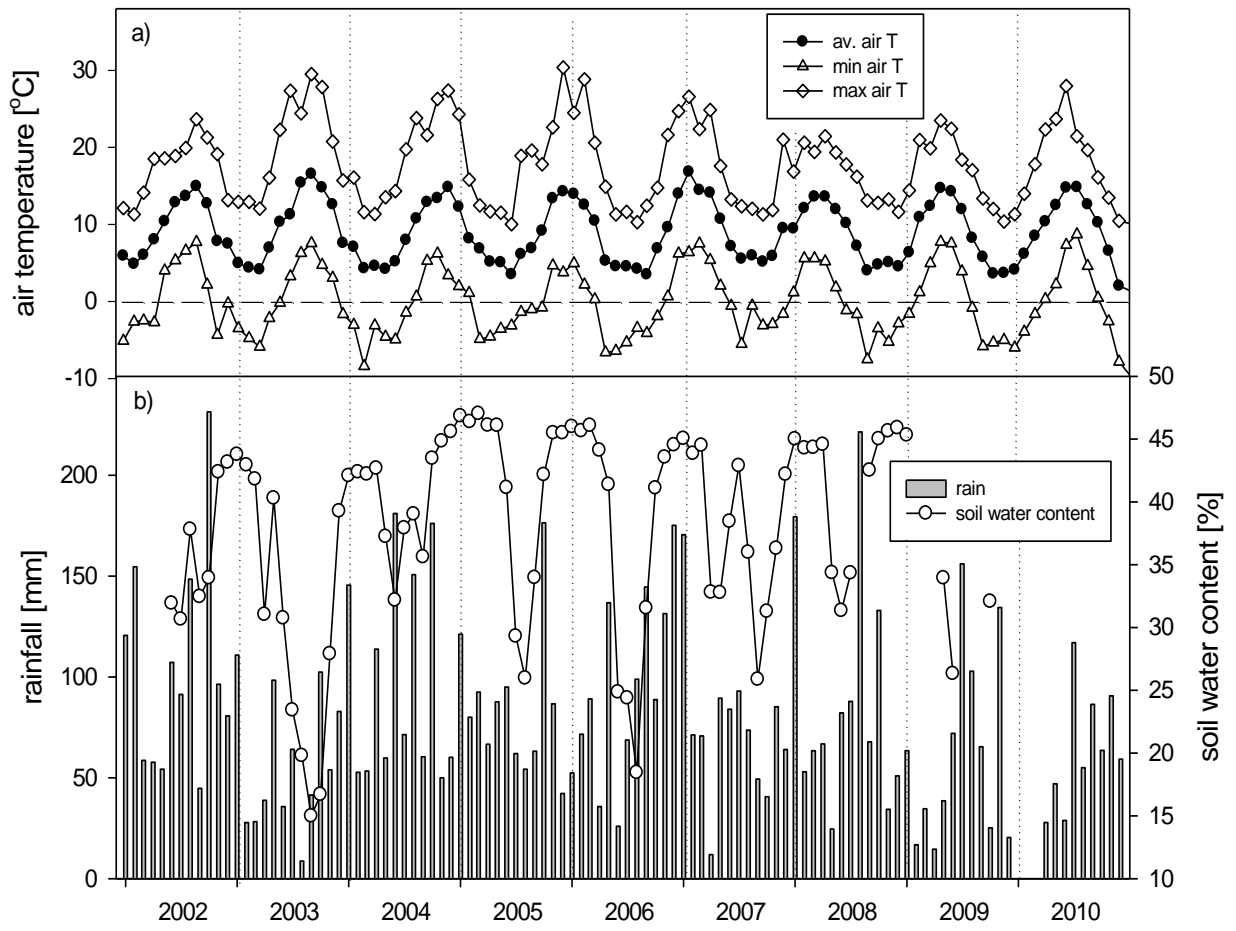


fig02



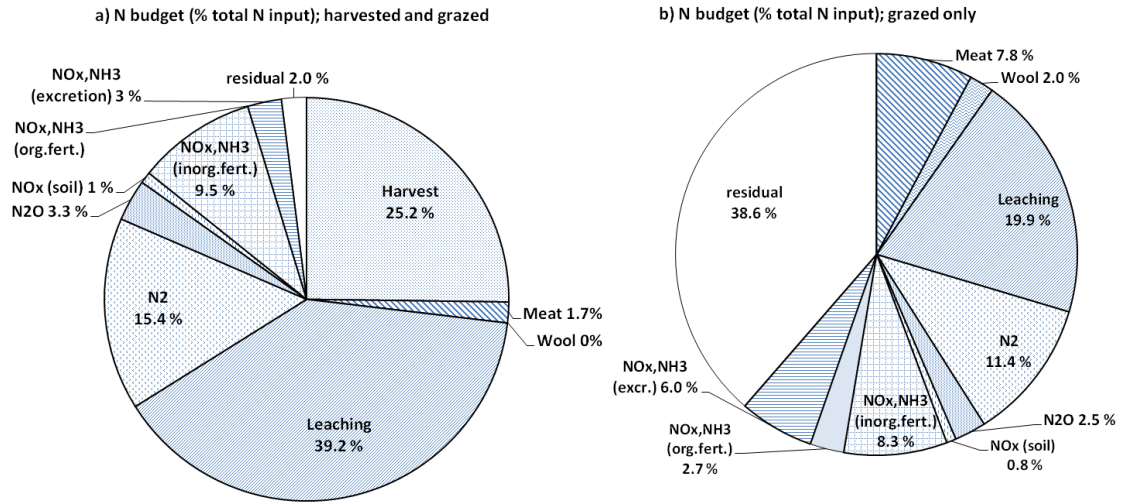


fig03

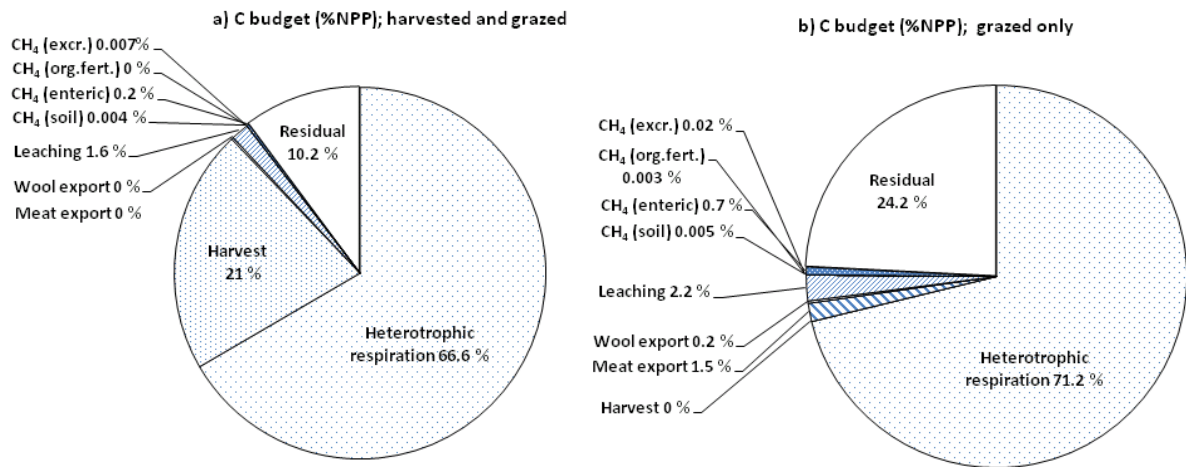


fig04

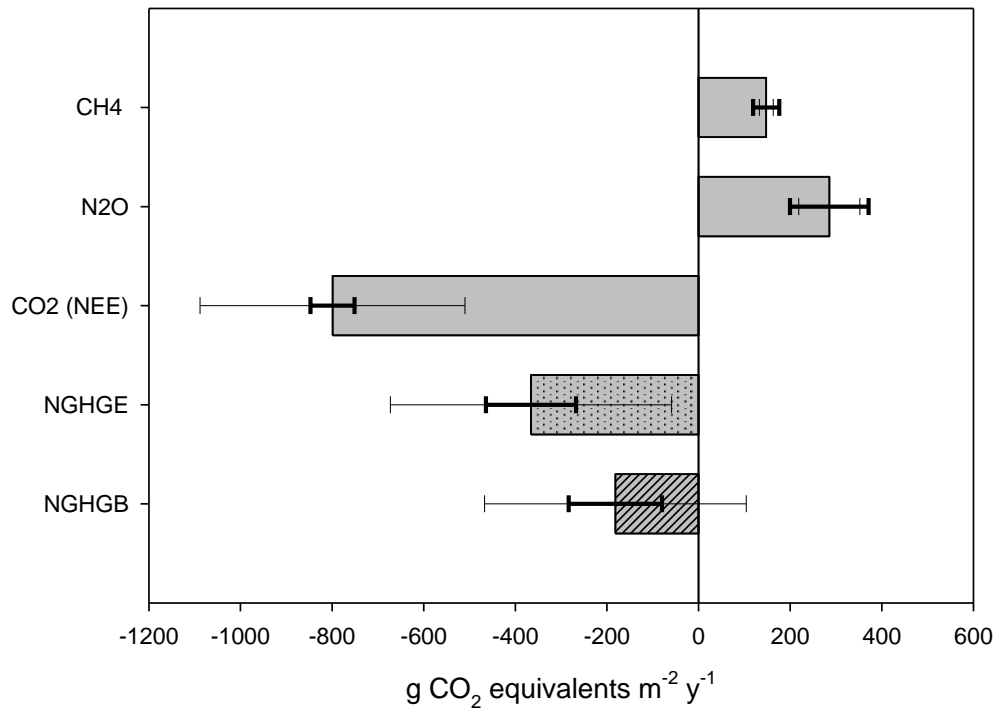


fig05