

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

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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 (CO₂), nitrous
40 oxide (N₂O) and methane (CH₄). 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 pool over time. The N
43 budget was dominated by import from inorganic and organic fertilisers (21.9 g N m² yr⁻¹) and
44 losses from leaching (5.3 g N m² yr⁻¹), N₂ emissions and NO_x and NH₃ volatilisation (6.4 g N
45 m² yr⁻¹). 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 ± 4.6 g N m⁻² y⁻¹
47 ¹ (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 CO₂ (NEE), at an average uptake rate of $218 \pm$
49 155 g C m⁻² y⁻¹ over the nine years. This sink strength was offset by carbon export from the
50 field mainly as harvest (48.9 g C m² yr⁻¹) and leaching (16.4 g C m² yr⁻¹). The other export
51 terms, CH₄ 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 ± 140 g C m⁻² y⁻¹. 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 ± 2.64 g N m⁻² y⁻¹ and lost C at a rate of 29.08 ± 38.19 g C m⁻² y⁻¹, 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 ± 601 g CO₂ eq m⁻² y⁻¹ and
60 strongly affected by CH₄ and N₂O emissions. The GHG sink strength of the NEE was reduced
61 by 54% by CH₄ and N₂O emissions. Enteric fermentation from the ruminating sheep proved to
62 be an important CH₄ source, exceeding the contribution of N₂O 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 (N₂) 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 N₂ through
76 complete denitrification (Galloway et al., 2003). However, in agricultural systems the surplus
77 of Nr is generally only partly converted to N₂, 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 N₂O and CH₄ 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 CO₂ and CH₄
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 (CO₂, CH₄ and N₂O) 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 EVK²-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 H₂O) 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⁻¹ (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 1st of June and 8th of August 2002 and on the 29th 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⁻¹ application⁻¹ 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 28th of September 2004 and 27th 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 (ΔN) over time (Δt) 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 ($FN_{\text{org fert.}} + FN_{\text{synt fert.}}$),
174 the fixation of N_2 through biological fixation ($FN_{N_2 \text{ (biol. fixation)}}$) 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 $FN_{\text{dep.}}$).
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 ($FN_{NH_3/NO_x(\text{fert., manure, animal})}$), the emission
180 of NO_x from the soil ($FN_{NO_x(\text{soil})}$), the emission of N_2O from the soil (FN_{N_2O}) and the loss of N_2
181 from total denitrification ($FN_{N_2(\text{denitr.})}$).
182

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

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

187

188 FC_{CO_2} represents the net ecosystem exchange (NEE) of CO_2 and $FC_{\text{org fert}}$ 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;

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

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

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 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^{++} , Automated Wet
260 Chemistry Analyzer - Continuous Flow Analyzer (CFA), Skalar, The Netherlands). Leachate C
261 and N concentrations were measured from October 1st 2006 - March 30th 2008. Dissolved C
262 and N were calculated by multiplying concentrations of DIC, DOC and dissolved 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 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 ($3^{\circ}14'35W$, $55^{\circ}47'34 N$), 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 N₂O 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 N₂O using a Hewlett
318 Packard 5890 series II gas chromatograph (Agilent Technologies, Stockport, UK), fitted with
319 an electron capture detector (detection limit: N₂O < 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 $R^2 = 0.96$. The minimal detectable flux was 12
323 ng N₂O-N m⁻² s⁻¹. 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 callendar year. For the periods where no N₂O
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_x fluxes (FN_{NO_x(soil)})**

330 NO_x fluxes from the soil were only measured for a short period (June 2009-August 2010). The
331 NO_x 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 m³)
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_x 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_x (42i-TL Trace Level
340 NO_x 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 are of the frames (0.25 m²). We used simulated data
344 from Landscape DNDC for years where no NO_x fluxes were measured.

345

346 **2.8.4 NH₄ + NO_x volatilisation (FN_{NH3/NOx} (fert.,manure, animal))**

347 The fraction of nitrogen that volatilises as NH₄ and NO_x 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_{ex}) from animals
351 depends on the total N intake (N_{intake}) and total N retention (N_{retention}) of the animal. N_{intake}
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_{retention} 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⁻¹ d⁻¹ 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⁻¹ d⁻¹ for ewes and varied between 0.0019-0.0106 kg N animal⁻¹ d⁻¹ for
358 lambs and 0.096-0.1013 kg N animal⁻¹ d⁻¹ for heifers.

359

360 **2.8.5 N₂ emission by total denitrification (FN_{N2(denitr.)})**

361 Di-nitrogen (N₂) emissions resulting from total denitrification in the soil was not measured in
362 our experiment. We therefore used the N₂ emission rates from LandscapeDNDC simulations.

363

364 **2.8.6 Biological N₂ fixation (FN_{N2} (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 CO₂ (FC_{CO2})**

370 NEFF was measured by an eddy covariance system consisting of a fast response 3D ultrasonic
371 anemometer (Metek USA-1, Metek GmbH, Elsmhorn, Germany) and a fast closed path CO₂-
372 H₂O 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 m s^{-1} (insufficient turbulence), CO_2 concentrations fell outside a plausible interval (330–
381 450 ppm), 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 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 (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 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 (FC_{CH_4})**

395 Methane fluxes from the soil were measured with closed static chambers simultaneously with
396 the N_2O 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 Fluxes were measured weekly and more frequently at fertiliser events. As measured soil CH_4
399 fluxes were close to zero and did not vary significantly between months, we calculated CH_4 for
400 months where no 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 CH_4 emissions (Jarvis et al., 1995). The calculation of CH_4 emissions from excretion
406 was based on the amount of volatile solids (VS) excreted, the maximum CH_4 producing
407 capacity (B_0) of the manure and the 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

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

412 MJ animal⁻¹ d⁻¹ for lambs and from 160.9 to 169.7 MJ animal⁻¹ d⁻¹ for heifers. Emission factors
413 for excretion were calculated as 0.198 kg CH₄ head⁻¹ y⁻¹ for ewes and varied between 1.64-1.73
414 kg CH₄ head⁻¹ y⁻¹ for heifers and 0.081-0.152 kg CH₄ head⁻¹ y⁻¹ for lambs. Methane emission
415 factors for enteric fermentation were calculated from GE intake and CH₄ conversion factors
416 (Y_m). Depending on animal type and live weight, emission factors were 7.6 kg CH₄ head⁻¹ y⁻¹
417 for ewes and varied between 60.1-63.8 kg CH₄ head⁻¹ y⁻¹ for heifers and 2.0-4.0 kg CH₄ head⁻¹
418 y⁻¹ for lambs. Annual emissions from excretion and enteric fermentation were calculated from
419 daily CH₄ 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 CH₄ 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⁻² s⁻¹, 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 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 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[®], Vaisala Ltd, Suffolk, UK).

464

465 **2.14 Statistical and uncertainty analysis**

466 Random error was determined as 2σ -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 ≥ 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⁻¹ y⁻¹ and 0.54
491 LSU ha⁻¹ y⁻¹, 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⁻¹
494 y⁻¹ and 0.91 LSU ha⁻¹ y⁻¹, respectively. Maximum monthly stocking density occurred in
495 September 2006 at 13.8 LSU ha⁻¹, 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⁻² application⁻¹ (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⁻² application⁻¹, respectively,
499 which resulted in a C input of 55.4 and 171.8 g C m⁻² application⁻¹, 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⁻¹ cut⁻¹ which resulted in an N off-take ranging from 1.7 to 4.7 g N m⁻²
502 cut⁻¹ and a C removal from the field ranging from 113.1 to 169.5 g C m⁻² cut⁻¹ (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 N₂O fluxes was 30 %, due to the uncertainty of gapfilling.
523 The uncertainty attributed to the modelled NO_x fluxes is 30 %. The uncertainty attributed to the
524 NH₄ and NO_x volatilisation was 30 % from applied synthetic fertiliser and 50 % from cattle
525 slurry application and animal excretion. The uncertainty attributed to the N₂ fluxes was 30 %.
526 The total uncertainty for NEE values was estimated to be 80 g C m⁻² y⁻¹ (Levy et al.,
527 submitted). The systematic uncertainty of annual cumulative soil CH₄ 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 CH₄ from enteric fermentation and animal excretion estimates were each assumed to be 20%,
530 according to IPCC (2006a). The uncertainty of CH₄ 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 ± 4.6 g N m⁻²
538 y⁻¹ (p<0.05). From 2003 to 2010, N was stored at a rate of -3.1 to -17.9 g N m⁻² y⁻¹, whilst in
539 2002 N was lost at a rate of 6.3 g N m⁻² y⁻¹ (Table 4). The major N input consisted of inorganic
540 fertiliser, ranging from -11 to -25.9 g N m⁻² y⁻¹, averaging at -19.2 g N m⁻² y⁻¹, 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 (R² = 0.55). Largest losses resulted from leaching at an average rate of
544 5.34 ± 3.4 g N m⁻² y⁻¹ and were estimated to be highest in 2002 at 14.9 g N m⁻² y⁻¹ and lowest in
545 2003 at 0.09 g N m² y⁻¹. We found a strong correlation between N leaching and rainfall (R² =
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 (R²=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 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 NO_x/NH_3 volatilisation from excretion and organic fertilisation
553 exceeded losses from N_2 emissions. Losses through 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 N_2O 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 ($R^2=0.76$). N_2O 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 (N_2 ;
567 15.4%), followed by NO_x/NH_3 volatilisation from inorganic N fertiliser applications (9.5%),
568 while N_2O emissions accounted for 3.3%, NO_x/NH_3 volatilisation from excretion for 2.7% and
569 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 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 NO_x/NH_3 volatilisation
574 from excretion). An additional loss occurred in grazed years through the volatilisation of
575 NO_x/NH_3 from organic fertiliser applications in 2004 and 2005 (3%). Losses through N_2O and
576 NO_x/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 m^{-2} , which corresponds to approximate
581 60 cm depth. The estimated N storage over the 7 years was -4.51 ± 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 CO_2 at an
591 average of $218 \pm 155 \text{ g C g C m}^{-2} \text{ y}^{-1}$, and the 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 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 CH_4 losses from enteric fermentation. Carbon export from wool
606 ranged from 0.5 to $2.1 \text{ g C m}^{-2} \text{ y}^{-1}$. CH_4 emissions from organic fertilisation, soil processes and
607 animal excretion were always less than 1 % of the total C losses. 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 CH₄
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⁻² (~ 0-60 cm) was lower in
627 2011 compared to 2004, resulting in a C loss of 29.08 ± 38.19 g C m⁻² (Table 7). In
628 comparison, based on flux calculations C was stored at 180 ±180 g C m⁻² y⁻¹ 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 CO₂, N₂O and CH₄ were expressed in CO₂ equivalents considering the
635 different global warming potentials for each gas at the 100 year time horizon (1 for CO₂, 298
636 for N₂O and 25 for CH₄, 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 CO₂ 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 CO₂ m⁻² y⁻¹. This
640 storage was significantly different from zero (p < 0.05). On average, the net GHG exchange
641 (NGHGE) was highly correlated with NEE (R²=0.96). On average the grassland was a source
642 of the GHGs CH₄ and N₂O at a rate of 148 ± 30 and 285 ± 131 g CO₂ m⁻² y⁻¹, respectively, both
643 being significantly different from zero (p < 0.001 and p < 0.01, respectively). Nitrous oxide
644 losses ranged from 52 g CO₂ eq. m⁻² y⁻¹ (2004) to 588 g CO₂ eq. m⁻² y⁻¹ (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 CH₄ emissions. Methane emissions from enteric
647 fermentation ranged from 53 g CO₂ eq. m⁻² y⁻¹ (2002) to 199 g CO₂ eq. m⁻² y⁻¹ (2004). The CH₄
648 emissions, which were predominately (> 97%) of ruminant origin weakened the sink strength
649 of NEE by 18 %. If both CH₄ and N₂O 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, CH₄
652 and N₂O were included (NGHGE). If all C components ($FC_{\text{org.fert}}$, FC_{animal} , FC_{leaching} , FC_{harvest})
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 N₂
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 is 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 N₂O 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).

707

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

709 Nitrogen leaches from grassland soils in the form of nitrate (NO₃⁻), ammonium (NH₄⁺) and
710 dissolved organic N (DON). Whereas NO₃⁻ is highly mobile in water and can be easily leached
711 into groundwater, NH₄⁺ 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⁻² y⁻¹, Watson et al., 2007) and
719 England (3.8-13.3 g N m⁻² y⁻¹, Scholefield et al., 1993) or croplands (e.g. Bechmann et al.,
720 1998), max. leaching losses of 10.4 g N m⁻² y⁻¹). However, leaching from our study was high
721 compared to the Swiss NitroEurope site, where a maximum loss of 3.5 kg N ha⁻¹ y⁻¹ 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 N₂ background, N₂ fluxes cannot be measured directly in the
737 field. However, there are different methods to measure N₂ fluxes indirectly, which have been
738 summarized by Groffman et al. (2006). In our study, we estimated N₂ 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 N₂ 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 N₂ losses simulated by LandscapeDNDC are
751 based on average (per ha⁻¹) 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 N₂ losses in our study have
753 been underestimated.

754
755 Nitrous oxide emissions are influenced by both management and environmental
756 conditions (Flecharad et al., 2007, Bell et al., 2015; Cowan et al., 2015). In our study N₂O fluxes
757 showed typical temporal variations with high N₂O 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⁻² y⁻¹) 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; Flecharad et al., 2007). Generally N₂O 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 N₂O emissions and
768 stocking density, rainfall or total N input if certain years were excluded. This shows that N₂O
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; Flecharad et al., 2007). We found a
772 relationship between the cumulative precipitation 1 week before plus 3 weeks after fertilization
773 and N₂O emissions (R²=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 N₂O 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 NH₃ volatilizes from urine patches, decomposing dung as well as
783 from fertilizers containing urea and NH₄⁺ (Twigg et al 2011). Increased rates of NH₃ 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 NH₃ and NO_x 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_x and NH₃
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_x 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_x 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 $0\text{-}25 \text{ g N m}^{-2} \text{ y}^{-1}$ 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 $10\text{-}15.2 \text{ g N m}^{-2} \text{ y}^{-1}$, 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 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 1st 2006 - March 30th 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 N₂,
824 NO_x/NH₃ emission from excretion, NO_x/NH₃ 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⁻² y⁻¹ (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 CO₂ measured in our study is close to the
832 upper range of NEE reported for temperate grasslands (100 to 600 g C m⁻² y⁻¹, (IPCC, 1996).
833 On an annual basis our grassland site was a sink for atmospheric CO₂ 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 CO₂ uptake as a
836 part of the field is defoliated each day. The reduced leaf area index (LAI) then leads to a
837 reduced CO₂ uptake by plants. In addition to the reduced LAI, grazing presents a source of CO₂
838 from animal respiration, thereby reducing the CO₂ 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⁻² y⁻¹) was close to the average NEE measured in a comparison
842 of nine European grasslands over two years (240 ± 70 g C m⁻² y⁻¹) by Soussana et al. (2007)
843 and comparable to the CO₂ sink capacity of managed Irish grasslands measured by Byrne et al.
844 (2007) (290 ± 50 g C m⁻² y⁻¹) or Leahy et al. (2004) (257 g C m⁻² y⁻¹). 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⁻² y⁻¹) and TER (972 to 2183 g C m⁻² y⁻¹) 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⁻² y⁻¹ and TER ranging from 493 to 1541 g C m⁻² y⁻¹, while Mudge et al. (2011) reported
853 values of 2000 g C m⁻² y⁻¹ 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 CO₂ 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 CO₂ 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 CO₂, 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 CO₂ shortly after intake.

887 We estimated that CH₄ 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 CH₄ fluxes by the number of sheep in the field
890 each day, Dengel et al. calculated CH₄ emissions per head of livestock as 7.4 kg CH₄ head⁻¹ y⁻¹
891 for sheep, which is close to the emission factor used in our budget of 7.6 kg CH₄ head⁻¹ y⁻¹ 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⁻² d⁻¹, 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⁻¹
899 y⁻¹, average of two locations as published in Kindler et al. (2011) were close to the average
900 value (29.4 g C m⁻¹ y⁻¹) of the reported European grasslands, which showed a range of C losses
901 of 6.5–42.5 g C m⁻¹ y⁻¹. 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⁻² y⁻¹
904 ¹, all above what we measured in our study. Important factors controlling the magnitude of C
905 leaching have 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 CO₂ 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 $-22 \pm 56 \text{ g}$
938 $\text{C m}^{-2} \text{ y}^{-1}$), although these estimates were not significantly different from each other. However,
939 in none of those reviewed studies were C flux and C stock change measured in the same field
940 experiment. A reason for the discrepancy between calculation methods in our study might lie in
941 a possible underestimation of C exports in the flux balance calculation, leading to an
942 overestimation of C storage in the soil. One underestimated flux could be the export of DIC and
943 DOC. Leaching was only measured in one year (2008), while values for remaining years were
944 estimated using a simple regression model with an attributed high uncertainty of 30 % (4.9 g C
945 $\text{m}^{-2} \text{ y}^{-1}$ of average fluxes). Further uncertainty could be due to the use of only one sampling
946 location, which might not representative of the whole field due to high spatial heterogeneity
947 (see Sect. 4.1.2.). Indeed, Siemens (2003) hypothesized that the underestimation of C leaching
948 from soils can explain a large part of the difference between atmosphere- and land-based
949 estimates of the C uptake of European terrestrial ecosystems. Gapfilling can introduce
950 uncertainties in the NEE data especially for years with low data capture. Furthermore, CO_2
951 losses from animal respiration could be underestimated at times due to the animals moving out
952 of the footprint of the EC mast. Using animal respiration values from chamber experiments of
953 $12.1 \text{ g CO}_2 \text{ kg}^{-1} \text{ live weight d}^{-1}$ for cows and $11.7 \text{ g CO}_2 \text{ kg}^{-1} \text{ live weight d}^{-1}$ for sheep and
954 lambs (Shane Troy, SRUC, personal communication), we estimated a maximum CO_2 loss from

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 ~ 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 ($90 \text{ g C m}^{-2} \text{ y}^{-1}$, 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 N_2O and 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 CO_2 equivalents, N_2O emissions as well as CH_4 emissions
986 strongly affected the GHG budget. Since the radiative forcing effect of N_2O is 298 times
987 greater than that of CO_2 a relatively small emission of N_2O 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 N₂O 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 CH₄ emissions with the stock density indicates that any changes in
993 animal production will have a major impact on the global CH₄ budget. The weakening of the
994 GHG sink strength of the NEE by N₂O and CH₄ 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 N₂O emissions, while changing land use from cropland to pasture in the attempt to
998 reduce C losses from soils might lead to increased CH₄ 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 N₂ and NO_xNH₄
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 CO₂
1018 fixation. However, increased N losses through N₂O emissions counteracted the benefits of C
1019 sequestration in terms of GHG emissions. Furthermore, CH₄ emissions from enteric
1020 fermentation largely reduced the positive effect of CO₂ uptake, especially in years where NEE
1021 rates were small. We therefore conclude that CO₂ exchange alone is not sufficient for the
1022 estimation of the GWP of a managed grassland ecosystem.

1023

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1538 **Tables**

1539 
 1540 Table 1. Average annual livestock densities [LSU ha⁻¹ y⁻¹].
 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 ^a | 20 | Organic manure ^a | 20 |
| Harvest ^b | 16 | Harvest ^b | 11 |
| Leaching ^c | 30 | Leaching ^c | 30 |
| Animal (wool and meat) ^d | 12 | Animal (wool and meat) ^a | 12 |
| Wet deposition | 30 | CH ₄ soil | 160 |
| Dry deposition ^e | 80 | CH ₄ enteric | 20 |
| N ₂ O | 30 | CH ₄ excretion | 20 |
| NOx soil | 30 | CH ₄ organic | 120 |
| NH ₄ volatilisation | 30 | | |
| NOx volatilisation | 50 | | |
| N ₂ | 30 | | |

1551 combined uncertainties of C and N analysis (17%) and volume spread (10%)
 1552 combined uncertainty of total C (4%) and  2%) 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 ₂ | 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 ₂ 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 _x (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 _x ,NH ₃ (inorg.fert.) | 2.1 | 1.6 | 1.1 | 1.7 | 2.2 | 1.7 | 2.6 | 2.5 | 1.9 | 1.9 | 0.3 | 0.6 |
| NO _x ,NH ₃ (org.fert.) | 0 | 0 | 1.4 | 3.2 | 0 | 0 | 0 | 0 | 0 | 0.5 | 0.7 | 0.3 |
| NO _x ,NH ₃ (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 ₂ (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 ₄ (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 ₄ (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 ₄ (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 ₄ (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 N₂O exchange, total N input by fertiliser (mineral and organic) and N₂O emission factors, expressed as percentage of total N inputs in 2002-2010.

| | N ₂ O flux [g N m ⁻² y ⁻¹] | Total N input [g N m ⁻² y ⁻¹] | 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⁻² y⁻¹) 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⁻², 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 $FC_{org\ fert}$, FC_{animal} , $FC_{leaching}$, $FC_{harvest}$) for 2002-2010. Positive values correspond to losses and negative values to storage of greenhouse gases to and from the grassland system, respectively. The CH_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.

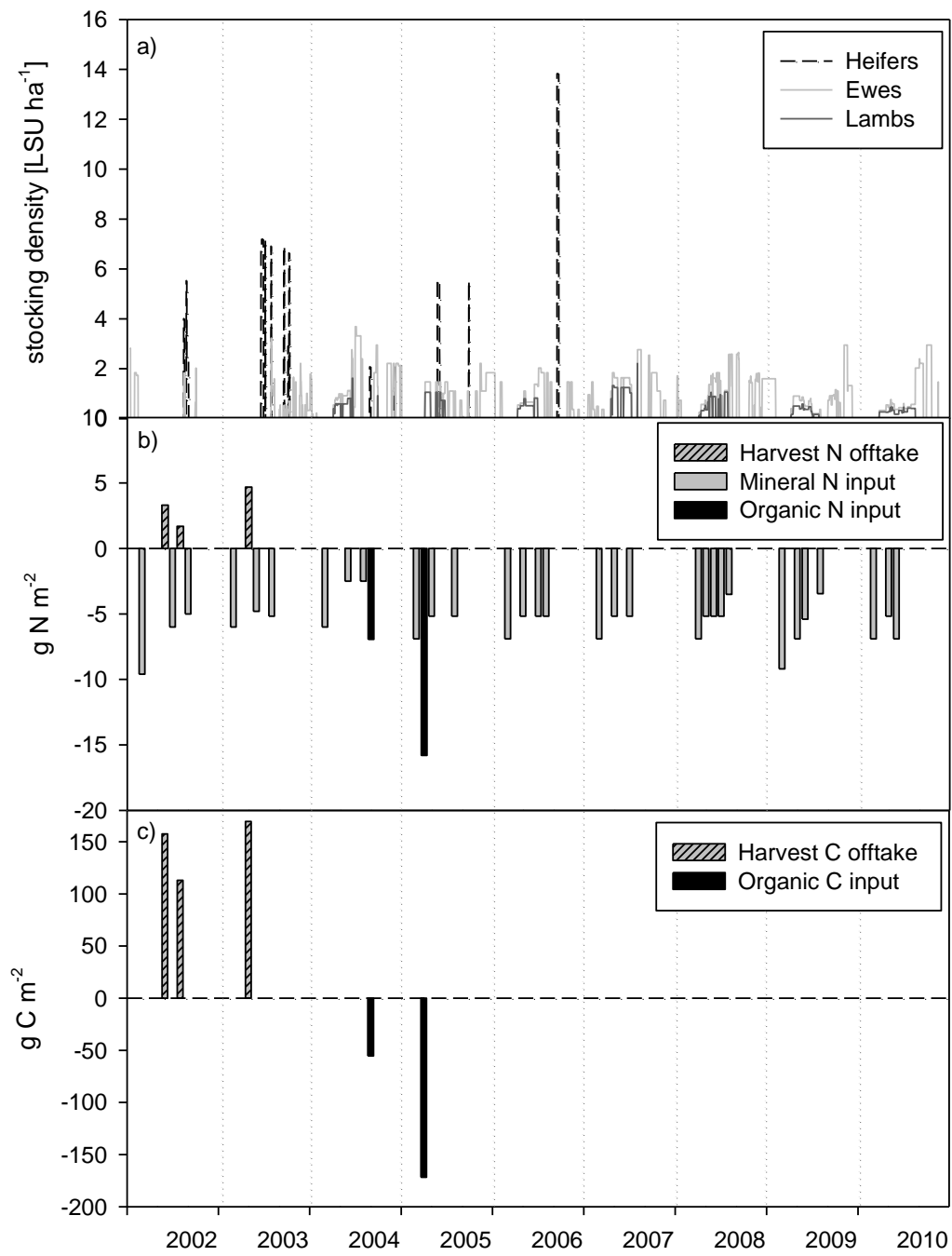


fig01

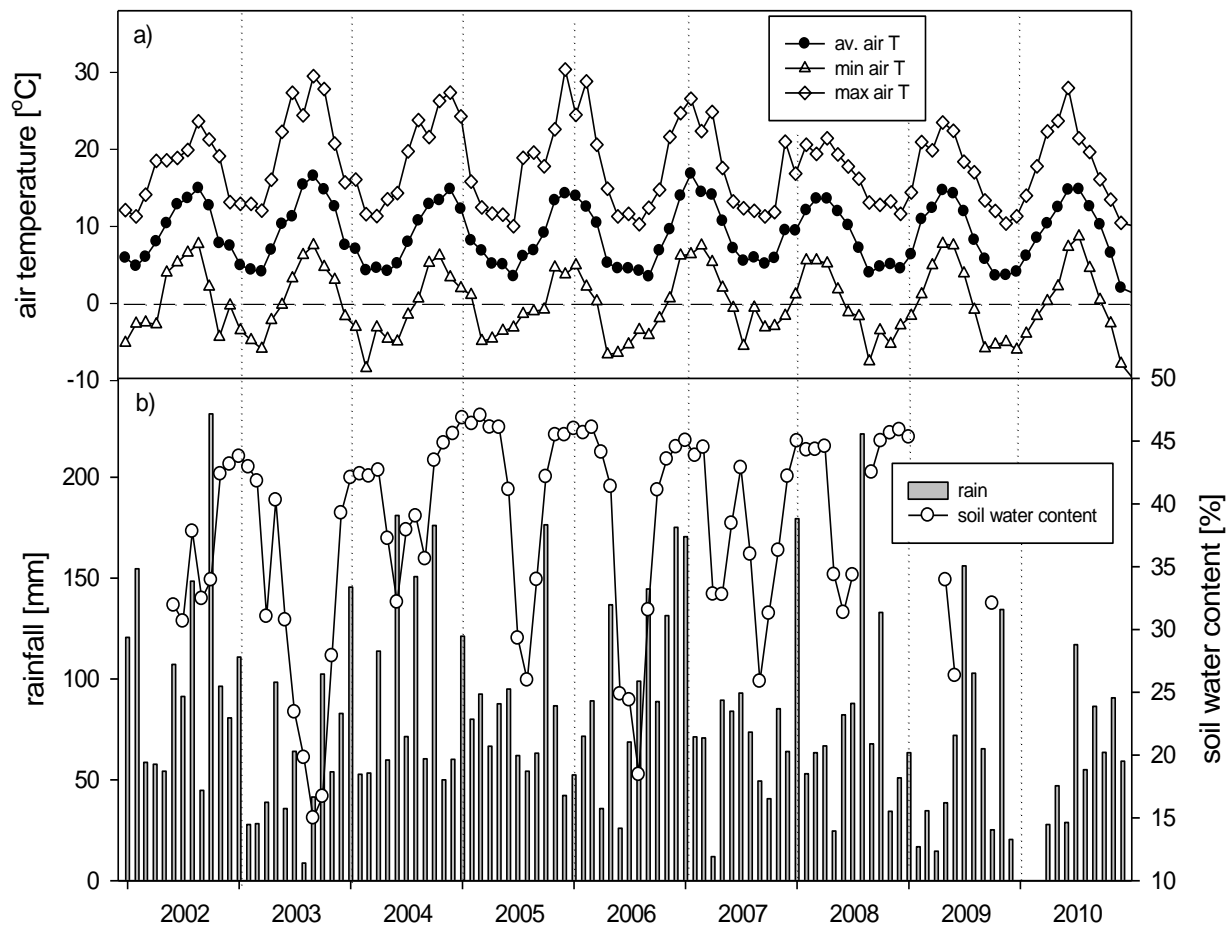


fig02

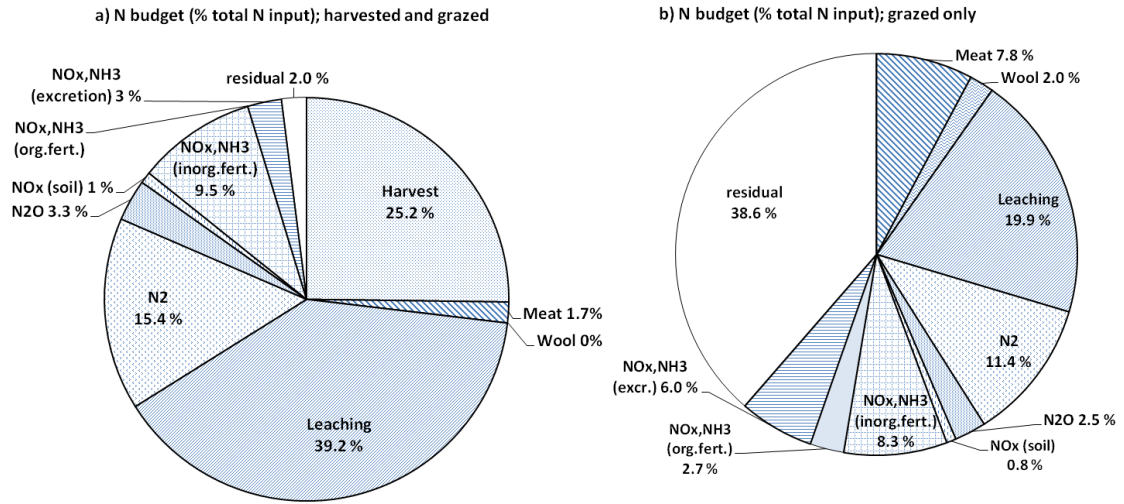


fig03

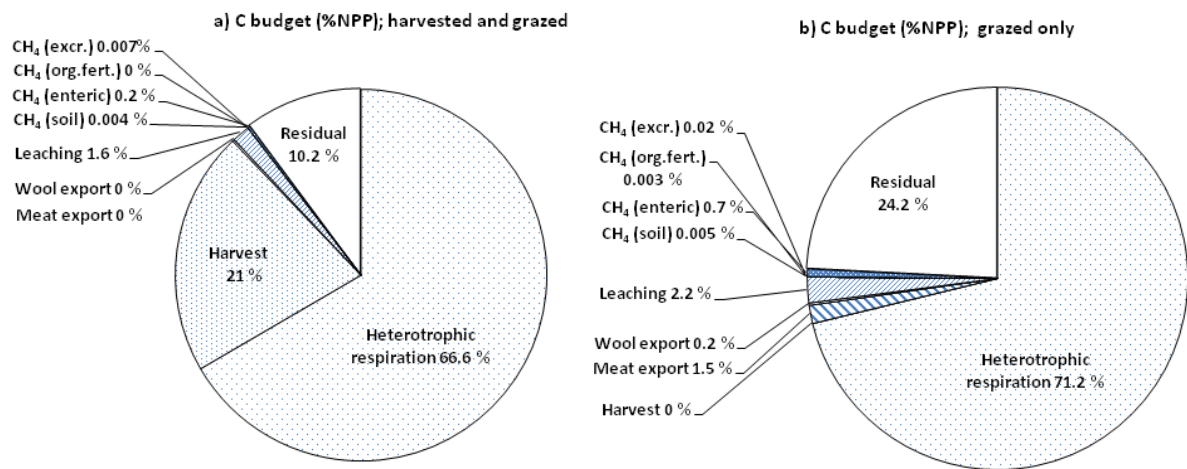


fig04

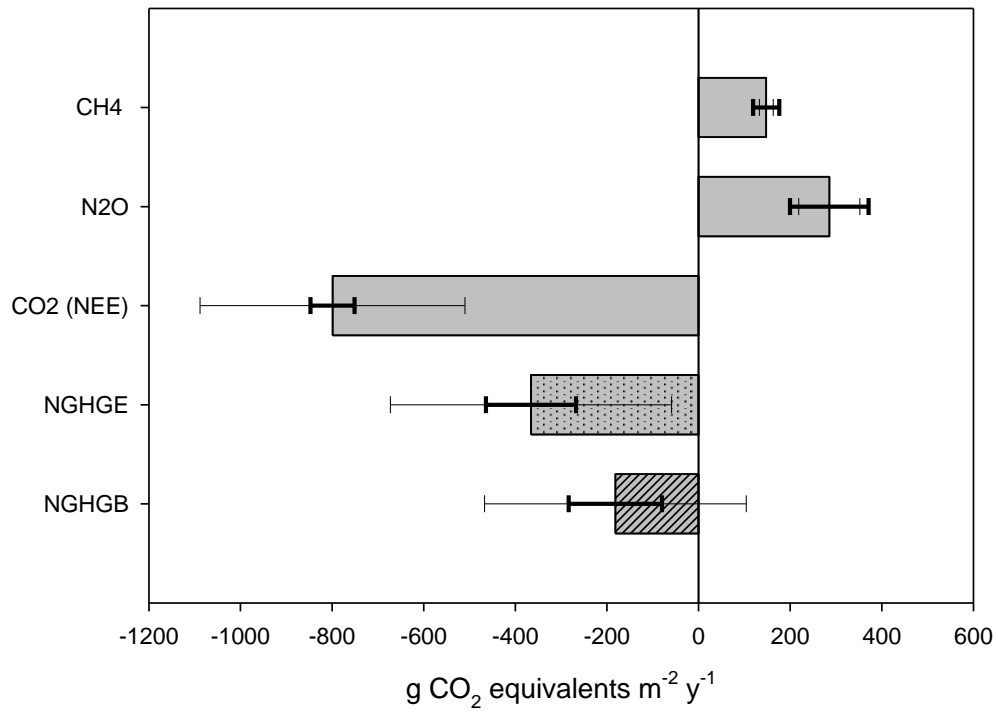


fig05