1 # Memory effects on greenhouse gas emissions (CO_2, N_2O and CH_4) following

2 # grassland restoration?

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28 ## Abstract

29 A five-year greenhouse gas (GHG) exchange study of the three major gas species (CO_2, CH_4

30 and N_2O) from an intensively managed permanent grassland in Switzerland is presented.

31 Measurements comprise two years (2010/2011) of manual static chamber measurements of

32 CH_4 and N_2O, five years of continuous eddy covariance (EC) measurements (CO_2/H_2O – 2010-

33 2014) and three years (2012-2014) of EC measurement of CH_4 and N_2O. Intensive grassland

management included both regular and sporadic management activities. Regular management practices encompassed mowing (3-5 cuts per year) with subsequent organic fertilizer amendments and occasional grazing whereas sporadic management activities comprised grazing or similar activities. The primary objective of our measurements was to compare pre-ploughing to post-ploughing GHG exchange and to identify potential memory effects of such a substantial disturbance on GHG exchange and carbon (C) and nitrogen (N) gains/losses. In order to include measurements carried out with different observation techniques, we tested two different measurement techniques jointly in 2013, namely the manual static chamber approach and the eddy covariance technique for N_2O, to quantify the GHG exchange from the observed grassland site.

Our results showed that there were no memory effects on N_2O and CH_4 emissions after ploughing, whereas the CO_2 uptake of the site considerably increased when compared to post-restoration years. In detail, we observed large losses of CO_2 and N_2O during the year of restoration. In contrast, the grassland acted as a carbon sink under usual management, i.e. the time periods (2010-2011 and 2013-2014). Enhanced emissions/emission peaks of N_2O (defined as exceeding background emissions 0.21 ± 0.55 nmol m^{-2} s^{-1} (SE = 0.02) for at least two sequential days and the seven-day moving average exceeding background emissions) were observed for almost seven continuous months after restoration as well as following organic fertilizer applications during all years. Net ecosystem exchange of CO_2 (NEE_{CO2}) showed a common pattern of increased uptake of CO_2 in spring and reduced uptake in late fall. NEE_{CO2} dropped to zero and became positive after each harvest event. Methane (CH_4) exchange fluctuated around zero during all years. Overall, CH_4 exchange was of negligible importance for both, the GHG budget as well as for the carbon budget of the site.

Our results stress the inclusion of grassland restoration events when providing cumulative sums of C sequestration potentials and/or global warming potentials (GWPs). Consequently, this study further highlights the need for continuous long-term GHG exchange observations as well as the implementation of our findings into biogeochemical process models to track potential GHG mitigation objectives as well as to predict future GHG emission scenarios reliably.

1 Introduction

Grassland ecosystems are commonly known for their provisioning of forage, either directly via grazing of animals on site, or indirectly by regular biomass harvest and preparation of silage or hay. Simultaneously, grasslands have further been acknowledged for their greenhouse gas (GHG) mitigation and soil carbon sequestration potential (Lal, 2004; Smith et al., 2008). However, greenhouse gas emissions from grasslands, particularly N_2O and CH_4 have been shown to offset net carbon dioxide equivalent (CO_2-eq.) gains (Ammann et al., 2020; Dengel et al., 2011; Hörtnagl et al., 2018; Hörtnagl and Wohlfahrt, 2014; Merbold et al., 2014; Schulze et al., 2009). Still, datasets containing continuous measurements of all three major GHGs (CO_2, CH_4 and N_2O) in grassland ecosystems remain limited (Hörtnagl et al., 2018), include a single GHG only, or focus on specific management activities (Fuchs et al., 2018; Krol et al., 2016). At the same time such datasets are extremely valuable by providing key training datasets for biogeochemical process models (Fuchs et al., 2020a).

Here we investigate the GHG exchange of the three major trace gases (CO_2, CH_4 and N_2O) over five consecutive years in a typical managed grassland on the Swiss plateau. Our study includes the application of traditional GHG chamber measurements and state-of-the-art GHG concentration measurements with a quantum cascade laser absorption spectrometer and a sonic anemometer in an eddy covariance setup (Eugster and Merbold, 2015). Prior to our measurements we hypothesized short-term losses of CO_2 and more continuous losses of primarily N_2O following dramatic managements events such as ploughing occurring at irregular time intervals. We further hypothesized an increased carbon uptake strength compared to the pre-ploughing years. Methane emissions were hypothesized to be of minor importance due to the limited time of grazing animals on site (Merbold et al., 2014).

Up to date the majority of greenhouse gas exchange research has focused on CO_2, with less focus on the other two important GHGs N_2O and CH_4, even though an increased interest in these other gas species has become visible in recent years (Ammann et al., 2020; Ball et al., 1999; Cowan et al., 2016; Krol et al., 2016; Kroon et al., 2007, 2010; Necpálová et al., 2013; Rutledge et al., 2017). The existing exceptions are often referred to as "high-flux" ecosystems, namely wetlands and livestock production system in terms of CH_4 (Baldocchi et al., 2012; Felber et al., 2015; Laubach et al., 2016; Teh et al., 2011) and agricultural ecosystems such as bioenergy system with considerable N_2O emissions (Cowan et al., 2016; Fuchs et al., 2018; Krol et al., 2016; Skiba et al., 1996, 2013; Wecking et al., 2020; Zenone et al., 2016; Zona et

al., 2013). Agricultural ecosystems and specifically grazed systems are characterized by GHG emissions caused through anthropogenic activities. These activities lead to changes in GHG emission patterns and include harvests, amendments of fertilizer and/or pesticides and less frequently occurring ploughing, harrowing and re-sowing events. While ploughing has been shown to lead to considerable short-term emissions of CO_2 and N_2O (Buchen et al., 2017; Cowan et al., 2016; Hörtnagl et al., 2018; MacKenzie et al., 1997; Merbold et al., 2014; Rutledge et al., 2017; Vellinga et al., 2004), regular harvests have been shown to lead to increased CO_2 uptake (Zeeman et al., 2010) and grazing leads to large CH_4 emissions (Dengel et al., 2011; Felber et al., 2015). Other studies showed contrary results with reduced N_2O emissions following ploughing of a drained grassland when compared to a fallow in Canada (MacDonald et al., 2011).

Still, the full range of management activities occurring in intensively managed grasslands and their respective impact on GHG exchange has not been investigated in detail. In a recent synthesis including grasslands located along an altitudinal gradient in Central Europe, Hörtnagl et al. (2018) highlighted the most important abiotic drivers of CO_2 (light, water availability and temperature), CH_4 (soil water content, temperature and grazing) and N_2O exchange (water filled pore space and soil temperature). The study by Hörtnagl et al. (2018) further elaborated the variation in management intensity and related variations in GHG exchange across sites, stressing the need for more case studies based on continuous GHG observations to improve existing knowledge and close remaining knowledge gaps. To complete the picture on factors driving ecosystem GHG exchange, irregular occurring events such as dry spells or extraordinary wet periods can further lead to enhanced or reduced GHG emissions (Chen et al., 2016; Hartmann and Niklaus, 2012; Hopkins and Del Prado, 2007; Mudge et al., 2011; Wolf et al., 2013).

While drought has been shown to reduce CO_2 uptake in forests (Ciais et al., 2005) whereas dry spells did not affect CO_2 uptake in grasslands (Wolf et al., 2013), flooding leads primarily to enhanced CH_4 emissions (Knox et al., 2015) and large precipitation events can lead to plumes of N_2O (Fuchs et al., 2018; Zona et al., 2013) similar to freeze-thaw events (Butterbach-Bahl et al., 2011; Matzner and Borken, 2008) to name only some examples. Consequently, understanding both, anthropogenic impacts such as management besides environmental impacts on ecosystem GHG exchange, are crucially important to suggest appropriate climate change mitigation as well as adaptation strategies for future land management with ongoing climate change.

Different measurement techniques to quantify the net GHG exchange in ecosystems are known and the most common approaches are either GHG chamber measurements or the eddy covariance (EC) technique. Static manual chamber measurements have been used for more than a century to quantify CO_2 emissions (Lundegardh, 1927) and their application has further been expanded during the last decades to quantify losses of the three major GHGs, CO_2, N_2O and CH_4 from soils (Imer et al., 2013; Pavelka et al., 2018a; Pumpanen et al., 2004; Rochette et al., 1997). Even though more complex in technology and assumptions made before carrying out measurements, the eddy covariance (EC) technique has become a valuable tool to derive ecosystem integrated CO_2 and H_2O_{vapour} exchange across the globe (Baldocchi, 2014; Eugster and Merbold, 2015). The technique has been further extended to continuous measurements of CH_4 and N_2O with the development of easy field-deployable fast-response analyzers during the last decade (Brümmer et al., 2017; Felber et al., 2015; Kroon et al., 2007; Nemitz et al., 2018a; Wecking et al., 2020). Each of the two approaches has its strengths and weaknesses and it is beyond the scope of this study to discuss each of them in detail. However, we refer to a set of reference papers highlighting the advantages and disadvantages of each technique separately (chambers: (Ambus et al., 1993; Brümmer et al., 2017; Pavelka et al., 2018a); eddy covariance: (Baldocchi, 2014; Denmead, 2008; Eugster and Merbold, 2015; Nemitz et al., 2018).

The overall objective of this study was to investigate the net GHG exchange (CO_2, CH_4 and N_2O) before and after grassland restoration and thus fill existing knowledge gaps caused by limited amounts of available GHG exchange data from intensively managed grasslands. The specific goals were: (i) to assess pre- and post-ploughing GHG exchange in a permanent grassland in central Switzerland accounting for changes in GHG exchange following frequent management activities; (ii) to compare two different measurement techniques, namely eddy covariance and static greenhouse gas flux chambers to quantify the GHG exchange in a business-as-usual year; and (iii) to provide a five year GHG budget of the site and quantify losses/gains of C and N. Based on our results we provide suggestions for future research approaches to further understand ecosystem GHG exchange, to mitigate GHG emissions and to ensure nutrient retention at the site for sustainable production from permanent grasslands in the future.

2 Material and Methods

2.1 Study site

The Chamau grassland site (Fluxnet identifier - CH-Cha) is located in the pre-alpine lowlands of Switzerland at an altitude of 400 m a.s.l. (47°12′ 37″N, 8°24′38″E) and characterized by intensive management (Zeeman et al., 2010). The site is divided into two parcels (Parcel A and B) with occasionally slightly different management regimes [see also *Fuchs et al., 2018*]. Mean annual temperature (MAT) is 9.1 °C, and mean annual precipitation (MAP) is 1151 mm. The soil type is a Cambisol with a pH ranging between 5 and 6, a bulk density between 0.9 and 1.3 kg m^{-3} and a carbon stock of 55.5–69.4 t C ha^{-1} in the upper 20 cm of the soil. The common species composition consists of Italian ryegrass (*Lolium multiflorum*) and white clover (*Trifolium repens L.*). For more details of the site we refer to Zeeman et al., (2010).

CH-Cha is intensively managed, with activities being either recurrent – referred to as usual/regular - or sporadic. Usual management refers to regular mowing and subsequent organic fertilizer application in form of liquid slurry (up to 7 times per year). In addition, the site is occasionally grazed by sheep and cattle for few days in early spring and/or fall (H.-R. Wettstein personal communication, Table S1). Sporadic activities aim at maintaining the typical fodder species composition and comprise reseeding, herbicide and pesticide application or irregular ploughing and harrowing on an approximately decadal timescale (Merbold et al., 2014). By such activity, mice are eradicated and a high-quality sward for fodder production is re-established following weed contamination. Specific information on management activity (timing, type of management, amount of biomass harvested) were reported by the farmers on site (Table S1). Additionally, representative samples of organic fertilizer were collected shortly before fertilizer application events and sent to a central laboratory for nutrient content analysis (Labor fuer Boden- und Umweltanalytik, Eric Schweizer AG, Thun, Switzerland). Harvest estimates were compared to estimates based on destructive sampling of randomly chosen plots (n = 10) in the years 2010, 2011, 2013 and 2014. The amount of harvested biomass in the year 2012 was based on a calibration of the values presented by the farmer in comparison to the on-site destructive harvests in previous and following years (Table S1).

2.2 Eddy covariance flux measurements

2.2.1 Eddy covariance setup

The specific site characteristics with two prevailing wind directions (North-northwest and South-south east) allows continuous observations of both management parcels. It is

noteworthy, that the separation of the two parcels is done exactly at the location of the tower. See Zeeman et al. (2010) and Fuchs et al. (2018) for further details. The eddy covariance setup consisted of a three-dimensional sonic anemometer (2.4 m height, Solent R3, Gill Instruments, Lymington, UK), an open-path infrared gas analyzer (IRGA, LI-7500A, LiCor Biosciences, Lincoln, NE, USA) to measure the concentrations of CO_2 and H_2O_{vapour} and a recently developed continuous-wave quantum cascade laser absorption spectrometer (mini-QCLAS - CH_4, N_2O, H_2O configuration, Aerodyne Research Inc., Billerica, MA, USA) to measure the concentrations of CH_4, N_2O, and H_2O_{vapour}. 3D wind components (u, v, w), CO_2 and H_2O_{vapour} concentration data from the IRGA were collected at a 20 Hz time interval, whereas concentrations of CH_4 and N_2O were collected at a 10 Hz rate from the QCLAS. The QCLAS provided the dry mole fraction for both trace gases (CH_4 and N_2O), and data were transferred to the data acquisition system (MOXA embedded Linux computer, Moxa, Brea, CA, USA) via an RS-232 serial data link and merged with the sonic anemometer and IRGA data streams in near-real time (Eugster and Plüss, 2010). Important to note is that the QCLAS was stored in a temperature-controlled box (temperature variation during the course of a single day was reduced to < 2 K) and located approximately 4 meters away from the EC tower to avoid long tubing. Total tube length from the inlet near the sonic anemometer to the measurement cell was 6.5 m. The inlet consisted of a coarse sinter filter (common fuel filter used in model cars) and a fine vortex filter (mesh size 0.3μm and a water trap) installed directly before the QCLAS. Filters were changed monthly or if the cell pressure in the laser dropped by more than 2 torr. Flow rate of approximately 15 l min^{-1} was achieved with a large vacuum pump (BOC Edwards XDS-35i, USA and TriScoll 600, Varian Inc., USA – the latter was used during maintenance of the Edwards pump). The pumps were maintained annually and replaced twice due to malfunction during the observation period. The infrared gas analyzer was calibrated to known concentrations of CO_2 and H_2O each year. The QCLAS did not need calibration due to its operating principles, and an internal reference cell (mini-QCL manual, Aerodyne Research Inc., Billerica, MA, USA) eased finding the absorption spectra after each restart of the analyzer.

2.2.2 Eddy covariance flux processing, post-processing and quality control

Raw fluxes of CO_2, CH_4, N_2O (F_{GHG}, μmol m^{-2} s^{-1}) were calculated as the covariance between turbulent fluctuations of the vertical wind speed and the trace gas species mixing ratio, respectively (Baldocchi, 2003; Eugster and Merbold, 2015). Open-path infrared gas analyzer (IRGA) CO_2 measurements were corrected for water vapor transfer effects (Webb et al., 1980). A 2-dimensional coordinate rotation was performed to align the coordinate system with the

235 mean wind streamlines so that the vertical wind vector $\acute{w} = 0$. Turbulent departures were
236 calculated by Reynolds (block) averaging of 30 min data blocks. Frequency response
237 corrections were applied to raw fluxes, accounting for high-pass and low-pass filtering for the
238 CO_2 signal based on the open-path IRGA as well as for the closed-path CH_4 and N_2O data
239 (Fratini et al., 2014). All fluxes were calculated using the software *EddyPro* (version 6.0, LiCor
240 Biosciences, Lincoln, NE, USA) (Fratini and Mauder, 2014).

241 The quality of half-hourly raw time series was assessed during flux calculations following
242 (Vickers and Mahrt, 1997). Raw data were rejected if (a) spikes accounted for more than 1 %
243 of the time series, (b) more than 10 % of available data points were significantly different from
244 the overall trend in the 30 min time period, (c) raw data values were outside a plausible range
245 ($\pm$ 50 $\mu mol\ m^{-2}\ s^{-1}$ for CO_2, $\pm$ 300 $nmol\ m^{-2}\ s^{-1}$ for N_2O and $\pm$ 1 $\mu mol\ m^{-2}\ s^{-1}$ for CH_4) and (d)
246 window dirtiness of the IRGA sensor exceeded 80 %. Only raw data that passed all quality
247 tests were used for flux calculations.

248 Half-hourly flux data were rejected if (e) fluxes were outside a physically plausible range (ie.
249 +/- 50 $\mu mol\ m^{-2}\ s^{-1}$ for CO_2) (f) the steady state test exceeded 30 % and (g) the developed
250 turbulent conditions test exceeded 30 % (Foken et al., 2006). Between 1st January 2010 and
251 31st December 2014 64572 (88% of all possible data) 30-min flux values were calculated for
252 CO_2, of which 42865 (57.8%) passed all quality tests and were used for analyses in the present
253 study (Table 1). The amount of available flux values for N_2O and CH_4 were less, since we were
254 only capable to continuously measure both gases from 2012 onwards (Table 1). Flux values in
255 this manuscript are given as number of moles of matter/mass per ground surface area and unit
256 time. Negative fluxes represent a flux of a specific gas species from the atmosphere into the
257 ecosystem, whereas positive fluxes represent a net loss from the system.

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259 **2.3 Static greenhouse gas flux chambers**

260 *2.3.1 Manual static GHG chamber setup*

261 Static manual opaque GHG chambers were installed within the footprint of the site to measure
262 soil fluxes in 2010 and 2011 (n =16) as well as during summer 2013 (n = 10). The chambers
263 were made of polyvinyl chloride tubes with a diameter of 0.3 m (Imer et al., 2013). The average
264 headspace height was 0.136 m $\pm$ 0.015 m and average insertion depth of the collars into the
265 soil was 0.08 m $\pm$ 0.05 m. During sampling days with vegetation larger than 0.3 m inside the
266 chamber, collar extensions (0.45 m) were used (2013 only). Chamber lids were equipped with
267 reflective aluminium foil to minimize heating inside the chamber during the period of actual
268 measurement. Spacing between the chambers was approximately seven m and an equal number

269   of chambers were installed in each parcel. For further details we refer to Imer et al. (2013).

270   Chamber measurements were carried out on a weekly basis during the growing season in all

271   three years (2010, 2011 and 2013), and at least once a month during the winter season in 2010

272   and 2011. More frequent measurements of N_2O emissions (every day) were performed

273   following fertilization events in 2013 for seven consecutive days after each event. Besides this,

274   an intensive measurement campaign lasting 48 hours (two-hour measurement interval) was

275   carried out in September 2010.

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277   *2.3.2 GHG concentrations measurements*

278   During each chamber closure four gas samples were taken, one immediately after closure and

279   then in approximately ten-minute time increments. With this approach, we guaranteed that the

280   chambers were closed no longer than 40 minutes to avoid potential saturation effects. Syringes

281   (60 ml volume) were inserted into the chambers lid septa to take the gas samples. The collected

282   air sample was injected into pre-evacuated 12 ml vials (Labco Limited, Buckinghamshire, UK)

283   in the next step. Prior to the second, third and fourth sampling of each chamber, the air in

284   chamber headspace was circulated with the syringe volume of air from the chamber headspace

285   to minimize effects of built-up concentration gradients inside the chamber.

286   Gas samples were analyzed for their respective CO_2, CH_4 and N_2O concentrations in the lab as

287   soon as possible after sample collection and not stored for more than a few days. Gas sample

288   analysis was performed with a gas chromatograph (Agilent 6890 equipped with a flame

289   ionization detector, a methanizer - Agilent Technologies Inc., Santa Clara, USA - and an

290   electron capture detector – SRI Instruments Europe GmbH, 53604 Bad Honnef, Germany) as

291   described by Hartmann and Niklaus (2012).

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293   *2.3.3 GHG chamber flux calculations and quality control*

294   GHG fluxes were calculated based on the rate of gas concentration change inside the chamber

295   headspace. Data processing, which included flux calculation and quality checks, was carried

296   out with the statistical software R (R Development Core Team, 2010). Thereby the rate of

297   change was calculated by the slope of the linear regression of gas concentration over time. Flux

298   calculation was based on the common equation containing GHG concentration (c in nmol mol^{-1}

299   ^{1} for CH_4 and N_2O), time (t in seconds), atmospheric pressure (p in Pa), the headspace volume

300   (V in m^{-3}), the universal gas constant (R = 8.3145 m^{-3} Pa K^{-1} mol^{-1}), ambient air temperature

301   (Ta in K) and the surface area enclosed by the chamber (A in m^{-2}) (equation 1 in Imer et al.

302   (2013)).

303 Flux quality criteria were based on the fit of the linear regression. If the correlation coefficient

304 of the linear regression (r^2) was < 0.8 the actual flux value was rejected from the subsequent

305 data analysis (see Imer et al. 2013 for further details on data quality control, and Table 1).

306 Furthermore, if the slope between the 1st and 2nd GHG concentration measurement deviated

307 considerably from the following concentrations we omitted the first value and calculated the

308 flux based on three instead of four samples. Mean chamber GHG fluxes were then calculated

309 as the arithmetic mean of all available individual chamber fluxes for each date. A total of 35

310 GHG flux calculations (CH_4 and N_2O) were available for the years 2010 and 2011. Another 52

311 N_2O flux values were available for the five-month peak-growing season in 2013.

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313 *2.4 Gapfilling and annual sums of CO_2, CH_4, and N_2O*

314 To date a common strategy to fill gaps in EC data of CH_4 and N_2O has not been agreed on. The

315 commonly used methods are simple linear approaches (Mishurov and Kiely, 2011) or the

316 application of more sophisticated tools such as artificial neural networks (Dengel et al., 2011).

317 The difficulty of finding an adequate gap-filling strategy results from the fact that emission

318 pulses of either N_2O or CH_4 remain challenging to predict. Similarly, different measurement

319 approaches – i.e. low temporal resolution manual GHG chambers compared to high temporal

320 resolution eddy covariance measurements - need different gap-filling approaches (Mishurov

321 and Kiely, 2011; Nemitz et al., 2018). In order to keep the gap-filling methods as simple and

322 reliable as possible, we used a running median (30 and 60 days for eddy covariance based and

323 chamber N_2O fluxes, respectively). A similar approach was recently chosen by Hörtnagl et al.

324 (2018) due to its reduced sensitivity to peaks in the N_2O exchange data. The approach was

325 particularly chosen as it minimizes the bias occurring from linear gap filling or simply using

326 an overall average value. While the gapfilling approach may be of less importance for EC flux

327 measurements with its high temporal data availability, it is the more important for less

328 frequently available GHG fluxes derived via manual chambers. Given the occurrence of

329 sporadic N_2O peaks which occur mostly in relation to management activities and last for few

330 hours/days only as well as the labour needed to carry out GHG chambers measurements,

331 researchers commonly aim at having weekly or biweekly flux data (i.e. Imer et al. 2013). The

332 respective sampling design is commonly designed to capture potential N_2O flux peaks as well

333 as some background values (Mishurov and Kiely, 2011). If one then uses either a linear

334 interpolation or an overall average value, one can derive a budget which is than a likely

335 overestimation of the annual flux budget caused by the few flux peaks observed in such

336 managed systems. The same bias is likely to occur if just flux averages are used since few very

337 high emission peaks will affect such an average. Thus, and in order to simulate N_2O emission
338 peaks more reliably, we have chosen the approach as taken by Hörtnagl et al. (2018).

339 In contrast to CH_4 and N_2O various well-established approaches to fill CO_2 flux data exist
340 (Moffat et al., 2007). Here, we filled gaps in CO_2 exchange data following the marginal
341 distribution sampling method (Reichstein et al., 2005) which was implemented in the R
342 package REddyProc (https://r-forge.r-project.org/projects/reddyproc/).

343 Calculation of the global warming potential (GWP) given in CO_2-equivalents followed the
344 recommendations given in the 5th Assessment Report of the Intergovernmental Panel on
345 Climate Change (IPCC), with CH_4 having a 28 and N_2O a 265 times greater GWP than CO_2
346 on a per mass basis over a time horizon of 100 years (Stocker et al., 2013).

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348 *2.5 Meteorological and phenological data*

349 Flux measurements were accompanied by standard meteorological measurements. These
350 included observations of soil temperature (depths of 0.01, 0.02, 0.05, 0.10, and 0.15 m, TL107
351 sensors, Markasub AG, Olten, Switzerland), soil moisture (depths of 0.02 and 0.15 m, ML2x
352 sensors, Delta-T Devices Ltd., Cambridge, UK) and air temperature (2 m height, Hydroclip S3
353 sensor, Rotronic AG, Switzerland). Furthermore, we measured the radiation balance including
354 short-wave incoming and outgoing radiation, long-wave incoming and outgoing radiation
355 (CNR1 sensor with ventilated Markasub housing, Kipp and Zonen, Delft, the Netherlands) as
356 well as photosynthetically active radiation at 2 m height (PARlite sensor, Kipp and Zonen,
357 Delft, the Netherlands). All data were stored as 30 min averages on a datalogger in a climate-
358 controlled box on site (CR10X, Campbell Scienctific, Logan, UT, USA).

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3 Results

3.1 General site conditions

The Chamau study site (CH-Cha) experienced meteorological conditions typical for the site during the five-year observation period. Summer precipitation commonly exceeded winter precipitation (Figure 1a). A spring drought was recorded from March till May 2011 (Wolf et al., 2013), leading to considerably lower soil water content than in previous and following years (Figure 1a). Average daily air temperatures rose up to 26.7 °C (27th July 2013) during summer and average daily temperature in winter dropped as low as -12.7 °C (6th February 2012, Figure 1b) with soil temperature following in a dampened pattern (Figure 1b). Average daily photosynthetic photon flux density did not differ considerably over the five-year observation period (Figure 1c). The site rarely experienced snow cover during winter (Figure 1b).

The complexity in management activities becomes apparent when comparing business as usual years (e.g. 2011) with the restoration year (2012, Figure 2a and b), highlighting the importance of grassland restoration to maintain productivity yields. Prior to 2012 an obvious decline in productivity with larger C and N inputs was found compared to the outputs in the years after restoration (2013 and 2014, Figure 2a and b).

3.2 EC N_2O fluxes vs. chamber derived N_2O fluxes

In 2013, we had the chance of comparing N_2O fluxes measured with two considerably different GHG measurement techniques, namely eddy covariance and static chambers. The chambers (n=10) were installed within the EC footprint. Our results reveal a similar temporal pattern, with increased N_2O losses being captured by both methodologies following fertilizer application. However, we could not identify a consistent bias of either technique (Figure 3a). Direct comparison of both measurements revealed a reasonable correlation (slope m = 0.61, r^2 = 0.4) and larger variation between both techniques with increasing flux values (Figure 3b).

3.3 Temporal variation of GHG exchange

Fluxes of CO_2 and N_2O showed considerable variation between and within years. This variation primarily occurs due to management activities and seasonal changes in meteorological variables (Figures 1 and 4). In contrast, methane fluxes did not show a distinct seasonal pattern.

CO₂ exchange

In pre-ploughing years (2010 and 2011), the Chamau site showed 60 % lower CO_2 uptake compared to the post-ploughing years (2013 and 2014, Table 2). All four non-ploughing years revealed largest CO_2 uptake rates in late spring (daily averaged peak uptake rates were >10 $\mu mol\ CO_2\ m^{-2}\ s^{-1}$, March and April, Figure 4a). Besides the seasonal effects a clear impact of harvest events could be identified, with abrupt changes from net uptake of CO_2 to either reduced uptake or net loss of CO_2 (light blue arrows indicate harvest event, Figure 4a). A similar but less pronounced effect was found following grazing periods (light and dark brown arrow, Figure 4a). A complete switch from net uptake to net CO_2 release was observed during the first three months of 2012, after ploughing and during re-cultivation of the grassland. In this specific year, the site only experienced snow cover for few days (Figure 1c) and temperatures below 5 ºC occurred more regularly than in all other years (Figure 1 b). Seasonal CO_2 exchange was characterized by net release of CO_2 in winter (DJF), highest CO_2 uptake rates were observed in spring (MAM), constant uptake rates during summer (JJA) which however were lower than those measured in spring, and very low net release of CO_2 in fall (Table 3). Average winter CO_2 exchange for the five-year observation period (gap-filled 30 min data) was $0.28 \pm 5.68\ \mu mol\ CO_2\ m^{-2}\ s^{-1}$ (SE = 0.04, Table 3). The restoration year 2012 showed a slightly different pattern with relatively large CO_2 release in winter and spring and considerably lower uptake rates in summer. The years before the restoration (2010 and 2011) were characterized by smaller net uptake rates during spring and summer when compared to the post-ploughing years (2013 and 2014). Additionally, winter fluxes in 2010 and 2011 were positive (net release of CO_2), while winter fluxes in the years 2013 and 2014 were showing a small but consistent net uptake of CO_2 (Figure 4a, Table 3).

CH₄ exchange

The individual static chamber measurements (2011&2011) were often below the detection limit and fluctuated around zero similar to the eddy covariance measurements (Figure 4b). Any methane peaks expected due to freezing and thawing in late winter and early spring were not observed. Also, commonly reported net emissions of methane during grazing of animals were not seen (Figure 4b). Seasonal differences of methane exchange did not show a clear pattern (Table 3). A comparison of methane fluxes obtained by both, static GHG chambers and EC measurements as done for N_2O (see next paragraph) could not be performed due to a malfunction of the respective detector in the gas chromatograph.

N₂O exchange

N₂O exchange was low during the majority of the days over the five-year observation period, fluctuating around zero (Figure 4c). However, clear peaks in N₂O emissions were observed following fertilization events or periods with high rainfall after a dry period in summer (i.e. summer 2013 and 2014, Figures 3a and 4c). While event driven N₂O emissions were commonly on the order of 4 to 8 nmol N₂O m^{-2} s^{-1} (Figure 4c), N₂O emissions following ploughing and subsequent re-sowing of the grassland in 2012 lead to up to three times as high N₂O emissions (Figure 4c, year 2012, see also Merbold et al. (2014)). Similar to methane, enhanced N₂O emissions in late winter or early spring as reported by other studies could not be identified (Figure 4c).

Background N₂O fluxes were estimated by analysing all high temporal resolution flux data but excluding the restoration year 2012 and all values one week after a management event. Daily average background fluxes were 0.21 ± 0.55 nmol m^{-2} s^{-1} (SE = 0.02). Differences in N₂O exchange over the course of individual years became obvious when splitting the dataset into the four seasons (winter – DJF, spring – MAM, summer – JJA and fall – SON). In contrast to CO₂ exchange that showed large net uptake rates in spring, N₂O emissions were largest during summer (JJA) and lowest in winter (DJF). As highlighted for the other gases, the year of grassland restoration showed a completely different picture (Table 3).

3.4 Annual sums and Global Warming Potential (GWP) of CO₂, CH₄ and N₂O

Annual sums showed a net uptake of CO₂ during the two pre-ploughing years (-695 g CO₂ m^{-2} yr^{-1} and -978 g CO₂ m^{-2} yr^{-1} in 2010 and 2011 respectively). Up to three times of this net uptake was reached in 2013 and 2014, the two post-ploughing years (-2046 g CO₂ m^{-2} yr^{-1} and -2751 g CO₂ m^{-2} yr^{-1}, Table 2). In contrast, the ploughing year 2011 was characterized by a net release of CO₂ (1447 g CO₂ m^{-2} yr^{-1}).

Methane budgets for the years 2010 and 2011 were not be calculated as many of the available measurements were below the limit of detection. For the years 2012 – 2014, the annual methane budget showed a minor release of 26.8 – 55.2 g CH₄ m^{-2} yr^{-1}.

The Chamau site was characterized by a net release of nitrous oxide over the five-year study period. While annual average N₂O emissions ranging between 0.34 and 1.17 g N₂O m^{-2} yr^{-1} in the non-ploughing years, the site emitted 4.36 g N₂O m^{-2} yr^{-1} in 2012. As an important note, due to the limited data availability for the years 2010 and 2011, the budgets of those years are likely incomplete.

The global warming potential (GWP), expressed as the yearly cumulative sum of all gases after their conversion to CO_2-equivalents, was negative during all years (between -387 and -2577 CO_2-eq. m^{-2}) except for the ploughing year 2012 (+2629 CO_2-eq. m^{-2}).

Overall, CO_2 exchange contributed more than 90% to the total GHG balance in 2011, 2013 and 2014. Clearly, CH_4 exchange was of minimal importance for the GHG budget (Table 2). In 2010, the contribution of CO_2 to the site's GHG budget was almost 70%, and N_2O contributed about 30%. Only in 2012, the year of restoration, CO_2 and N_2O exchange contributed almost equally to the site's overall GHG budget (55.1% and 43.9%, respectively).

3.5. Carbon gains/losses of the Chamau site between 2010 and 2014

The Chamau site assimilated on average -441 ± 260 g CO_2-C m^{-2} yr^{-1} (4410 kg C ha^{-1} yr^{-1}) during the "business as usual" years (2010 and 2011 as well as 2013 and 2014). During the restoration year the site lost 395 g CO_2-C m^{-2} (3950 kg C ha^{-1}) (Table 2). Carbon losses (and/or gains) from methane were < 1 g CH_4-C m^{-2} during all five years.

Carbon was gained in both parcels during the pre-ploughing years (Table 4). Considerable net losses of carbon were calculated for the ploughing year. In contrast, the post-ploughing years were again recognized as years with large net gains in carbon. Over the observation period of 5 years, the Chamau grassland gained approximately 4 t C ha^{-1}, excluding losses via leaching and deposition of C in form of dust.

4 Discussion

The five-year measurement period is representative for other similarly managed grassland ecosystems in Switzerland. Climate conditions were similar to the long-term average as described in Wolf et al. (2013). Management activities, such as harvests and subsequent fertilizer applications, were driven by overall weather conditions, (i.e. 2013 late spring, Figure 2a and b).

4.1 Technical and methodological aspects of the study

Different techniques are currently applied to measure GHG fluxes from a variety of ecosystems (Denmead, 2008), each having its advantages and disadvantages or being chosen for a specific purpose or reason. A common approach to study individual processes or time periods contributing to specific greenhouse gas emissions is to measure with GHG chambers on the plot scale (Pavelka et al., 2018). Chamber methods have been widely used to derive annual GHG and nutrient budgets (Barton et al., 2015; Butterbach-Bahl et al., 2013). Critical assessments of the suitability and associated uncertainty in chamber derived GHG budgets in relation to sampling frequency have been published by Barton et al. (2013). Existing studies have not only compared the two measurement techniques employed in this study (manual chambers and eddy covariance) in grasslands before, but also estimated annual emissions based on differing methodologies (Flechard et al., 2007; Jones et al., 2017). Additional confidence in our approach was obtained from the N_2O emissions during the summer period 2013, where both measurement techniques ran in parallel (Figure 3a and b). Annual budgets derived by applying similar gap-filling approaches to the individual datasets led to comparable results (Table 2).

We calculated detection limits for the individual GHGs from our manual chambers following (Parkin et al., 2012). Detection limits were 0.34 ± 0.26 nmol m^{-2} s^{-1}, 0.05 ± 0.02 nmol m^{-2} s^{-1}, and 0.06 ± 0.06 μmol m^{-2} s^{-1} for CH_4, N_2O and CO_2, respectively. Following this, methane flux measurements frequently were below this limit of detection, hence we did not calculate methane budgets for 2010 and 2011. The flux values measured with the EC technique between 2012 and 2014 compare well to similar measurements made by Felber et al. (2016) in an intensively managed grassland in Western Switzerland. The observed values have been identified to represent the soil methane exchange in EC measured fluxes (Felber et al. 2016). N_2O fluxes in contrast were much better constrained by both methods due to clear N_2O sources (i.e. fertilizer amendments) and better sensitivity of the instruments used by both techniques

538 for N_2O as compared to CH_4. Background N_2O emissions as observed in this study (0.21 ±

539 0.55 nmol m^{-2} s^{-1} (SE = 0.02)) compare well to estimates suggested by Rafique et al., (2011)

540 whom suggest an annual background N_2O losses of 1.8 kg N_2O-N for a grazed pasture (i.e.

541 0.20 nmol m^{-2} s^{-1}).

542

543 *4.2 Annual GHG and C and N gains/losses*

544 Net carbon losses and gains estimated for the CH-Cha site between 2010 and 2015 were in

545 general within the range of values estimated by Zeeman et al., (2010) for the years 2006 and

546 2007. The slightly higher losses observed prior to ploughing may result from reduced

547 productivity of the sward. This becomes particularly visible when compared to the net

548 ecosystem exchange (NEE) of CO_2 values for the years after restoration. Losses via leaching

549 have previously been estimated to be of minor importance at this site (Zeeman et al., 2010) and

550 were therefore not considered in this study. Considerably higher C gains during post-ploughing

551 years were caused be enhanced plant growth in spring and summer. Restoration is primarily

552 done to eradicate weeds and rodents, favouring biomass productivity of the fodder grass

553 composition. Other grasslands in Central Europe, i.e. sites in Austria, France and Germany,

554 showed similar values for net ecosystem exchange (Hörtnagl et al., 2018). Still, total C budgets

555 as presented here are subject to considerable uncertainty which is strongly depending on

556 assumptions made for gap-filling etc. (Foken et al., 2004). Nevertheless, the values reported

557 here show the overall trend on C uptake/release of the site and clearly exceed the uncertainty

558 of ± 50 g C per year for eddy covariance studies as suggested by Baldocchi (2003).

559 Methane was of negligible importance for the C budget of this site. We did not observe distinct

560 peaks in CH_4 emissions in relation to grazing which is primarily due to the low grazing pressure

561 at CH-Cha. Studies carried out on pastures in Scotland, Mongolia, France and Western

562 Switzerland have shown that grazing can largely contribute to ecosystem-scale methane fluxes,

563 in particular if ruminants such as cattle are populating the EC footprint (Dengel et al., 2011;

564 Felber et al., 2015; Schönbach et al., 2012). If we included an approximation of methane

565 emissions of cattle which we may have missed in the EC flux measurements, we would have

566 to add 3.67 g CH_4-C m^{-2} y^{-1} to the current value of 1.48 g CH_4-C m^{-2} in 2014 (Table 2). This

567 value is based on the average methane emissions of 404 g CH_4 $head^{-1}$ d^{-1} stated in Felber et al.

568 (2016) and linking this to the average stocking density (4.04 head ha-1) on the Chamua site

569 and the stocking duration (30 days in 2014). Still, the GHG budget as well as the C budget of

570 the site would not be altered.

571   The nitrous oxide budget reported for the years without ploughing in this study coincides with
572   values reported for other grasslands in Europe, ranging from moist to dry climates and lower
573   to higher elevations in Austria and Switzerland (Cowan et al., 2016; Hörtnagl et al., 2018; Imer
574   et al., 2013; Skiba et al., 2013).
575   Nitrogen inputs and losses via N_2O varied largely between the years before and after ploughing.
576   While the site was characterized by large N amendments prior to ploughing and with reduced
577   harvest, the picture was completely the opposite during the years after ploughing, with
578   considerably less N inputs compared to the nitrogen removed from the field via harvests.
579   Farmers aim every year at having a balanced N budget (fertilizer inputs = nutrients removed
580   from the field). Pasture degradation is the main motivation for enhanced fertilizer inputs in
581   order to stabilize forage productivity. Similarly, regular restoration of permanent pastures is
582   absolutely necessary (Cowan et al., 2016). So far, we identified only one study that investigated
583   the net effects on the overall GHG exchange following grassland restoration (Drewer et al.,
584   2017).
585

586   **5 Conclusion**
587   This study in combination with an overview of available datasets on grassland restoration and
588   their consequences on GHG budgets highlights the overall need of additional observational
589   data. While restoration changed the previous C sink to a C source at the Chamau site, the wider
590   implication in terms of the GWP of the site when including other GHGs have long-term
591   consequences (i.e. in mitigation assessments). Furthermore, this study showed the large
592   variations in N inputs and N outputs from this grassland and the difficulty farmers face when
593   aiming for balanced N budgets in the field. Still, the current study focused on GHGs only and
594   can thus not constrain the N budget but assess the losses of N via N_2O. Losses in form of NH_3,
595   N_2 and NO_x will have to be quantified to fully assess N budgets besides the overall fact that
596   GHG data following grassland restoration remain largely limited to investigate long-term
597   consequences. Fortunately, these are likely to become available in the near future by the
598   establishment of environmental research infrastructures (i.e. ICOS in Europe, NEON in the
599   USA or TERN in Australia) that aim at standardized, high quality and high temporal resolution
600   trace gas observation of major ecosystems, including permanent grasslands. With these
601   additional data, another major constraint of producing defensible GHG and nutrient budgets,
602   namely gap-filling procedures, will likely be overcome. New and existing data can be used to
603   derive reliable functional relations and artificial neural networks (ANNs) at field to ecosystem

604   scale that are capable of reproducing in-situ measured data. Once this step is achieved, both

605   the available data as well the functional relations can be used to improve, to train and to validate

606   existing biogeochemical process models (Fuchs et al., 2020). Subsequently, reliable

607   projections on both nutrient and GHG budgets at the ecosystem scale that are driven by

608   anthropogenic management as well as climatic variability become reality.

609   The study stresses the necessity of including management activities occurring at low frequency

610   such as ploughing in GHG and nutrient budget estimates. Only then, the effect of potential

611   best-bet climate change mitigation options can be thoroughly quantified. The next steps in

612   GHG observations from grassland must not only focus on observing business as usual

613   activities, but also aim at testing the just mentioned best-bet mitigation options jointly in the

614   field while simultaneously in combination with existing biogeochemical process models.

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619 **6 Tables and Figures**

620

621 **Table 1**: Table 1: Data availability of GHG fluxes measured over the five-year observation
622 period. Values are given as all data possible, raw processed values and high quality (HQ) data
623 or for the chamber flux data if above the detection limit, which were then used in the analysis.
624 Grey shaded areas represent time period where both methods (EC and static chambers) were
625 used simultaneously to estimate F_{N2O}. Static chamber flux data are further marked in *italic* font.

626

627 **Table 2:** Annual average CO_2, CH_4 and N_2O fluxes and annual sums for the three GHGs as
628 well as carbon and nitrogen gain/losses per gas species. GWP were calculated for a 100-year
629 time horizon and based on the most recent numbers provided by IPCC (Stocker et al., 2013).
630 Annual budgets were derived from either gap-filled manual chamber (MC) or eddy covariance
631 (EC) measurements. n.c. stands for not calculated. Numbers in italic for N2O in the years
632 2010/2011 are likely incomplete due to limited data availability. Sign convention: positive
633 values denote export/release, negative values import/uptake.

634

635 **Table 3:** Average GHG flux rates per season: winter (DJF), spring (MAM), summer (JJA) and
636 fall (SON). Values are based on gap-filled data to avoid bias from missing nighttime data
637 (predominantly relevant for CO_2). Data are only presented when continuous measurements
638 (eddy covariance data) were available. Sign convention: positive values denote export/release,
639 negative values import/uptake.

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641 **Table 4:** Table 4: Carbon and nitrogen gains/losses through fertilization, harvest and GHGs
642 for the Chamau (CH-Cha) site in 2010- 2014. Values are given in kg ha^{-1}. Gains are indicated
643 with "-" and losses/exports are indicated with "+". While management information was

644   available for both parcels (A and B), flux measurements are an integrate of both parcels. n.c. =
645   not calculated

646

647   **Table 5:** Existing studies investigating the GHG exchange over pastures following ploughing.
648   Results presented show the flux magnitude following ploughing and are rounded values of the
649   individual presented in the papers. Values were converted to similar units (mg CO_2-C m^{-2} h^{-1},
650   μg CH_4-C m^{-2} h^{-1} and μg N_2O-N m^{-2} h^{-1}). Based on Web of Knowledge search July 15th 2017
651   with the search terms "grassland", "pasture", "greenhouse gas", "ploughing" and/or "tillage".
652   Only two studies representing conversion from pasture to cropland or other systems were
653   included in this table.

654

655   **Table S1:** Detailed management information for the two parcels under investigation at the
656   Chamau research station. Data are based on fieldbooks provided by the farm personnel as well
657   as in-situ measurements. Organic fertilizer samples were sent to a central laboratory for nutrient
658   content analysis (Labor fuer Boden- und Umweltanalytik, Eric Schweizer AG, Thun,
659   Switzerland). Destructive harvests (n = 10) of biomass were carried out in the years 2010, 2011,
660   2013 and 2014. Harvest estimates are based on values derived from the in-situ measurements
661   and data provided by the farm personnel. Detailed information on the grazing regime was
662   furthermore provided by the farm personnel in hand-written form (not shown).

663

664   **Figure 1:** Weather conditions during the years 2010 – 2014. Weather data were measured with
665   our meteorological sensors installed on site. (a) Daily sum of precipitation (mm) and soil water
666   content (SWC, blue line, m^3 m^{-3}) measured at 5 cm soil depth; (b) daily averaged air
667   temperature (°C), daily averaged soil temperature (grey line, °C) and days with snow cover
668   (horizontal bars); (c) daily averaged photosynthetic photon flux density (PPFD, μmol m^{-2} s^{-1}).
669   Days with snow cover were identified with albedo calculations. Days with albedo > 0.45 were
670   identified as days with either snow or hoarfrost cover.

671

672   **Figure 2:** Management activities for both parcels (A and B in panels (a) and (b), respectively)
673   on the CH-Cha site. Overall management varied particularly in 2010 between both parcels,
674   whereas similar management took place between 2011 and 2014. Arrow direction indicates
675   whether carbon (C in kg ha^{-1}) and/or nitrogen (N in kg ha^{-1}) were amended to, or exported from
676   the site ("F_o" and "F_{o*}"- organic fertilizers, slurry/manure (red); "F_m" - mineral fertilizer (light
677   orange); "H" - harvest (light blue); "G_s" and "G_c" - grazing with sheep/cows (light/dark
678   brown). Other colored arrows visualize any other management activities such as pesticide
679   application ("P_h"- herbicide (light pink); "P_m"- molluscicide (dark pink); "T"- tillage (black),
680   "R"- rolling (light grey) and "S"- sowing (dark grey) which occurred predominantly in 2010
681   (parcel B) and 2012 (parcels A and B). Carbon imports and exports are indicated by black and
682   grey bars. Thereby black indicated the start of the specific management activities and grey the
683   duration (e.g. during grazing, "G_s"). Green colors indicate nitrogen amendments or losses, with
684   dark green visualizing the start of the activity and light green colors indicating the duration.
685   Sign convention: positive values denote export/release, negative values import/uptake.

686

687   **Figure 3:** (a) Temporal dynamics of N_2O fluxes measured with the eddy covariance (white
688   circles) and manual greenhouse gas chambers (black circles measured in 2013) – grey lines
689   indicate standard deviation. Arrows indicate management events ("H" = harvest, "F_o" =
690   organic fertilizer application (slurry), "Ph" = pesticide (herbicide) application). (b) 1:1
691   comparison between chamber based and eddy covariance based N_2O fluxes in 2013. The
692   dashed line represents the 1:1 line. (y = mx + c, r^2 = 0.4, m = 0.61, c = 0.17, p < 0.0001). Sign
693   convention: positive values denote export/release, negative values import/uptake.

694
695 **Figure 4:** Temporal dynamics of gap-filled (except methane in 2010/2011) daily averaged
696 greenhouse gas (GHG) fluxes (white circles): a) (CO_2 exchange in μmol m^{-2} s^{-1}; (b) CH_4
697 exchange in nmol m^{-2} s^{-1} and (c) N_2O exchange in nmol m^{-2} s^{-1}. Coloured circles indicate
698 manual chamber measurements. While both GHGs, CH_4 and N_2O were measured in 2010 and
699 2011 (blue cirlces), N_2O only was measured in 2013 (light blue circles). The grey dashed lines
700 indicate the beginning of a new year. Same color coding as used in Figure 3 a was used to
701 highlight management activities. Sign convention: positive values denote export/release,
702 negative values import/uptake. Grey lines behind the circles indicate standard deviation.
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8 Author contributions

LM and LH designed the study and wrote the first manuscript version. LM, CD, WE KF and BW collected the data in the field. LH further provided the code for flux processing. All authors revised and commented on the manuscript.

9 Author contributions

All flux and meta data are openly available via Fluxnet. The flux processing code is available via the Grassland Sciences Group at ETH Zurich. Greenhouse gas chamber data is available via Imer et al. 2013.

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