



Temporal and spatial variations of soil CO₂, CH₄ and N₂O fluxes at three differently managed grasslands

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Received: 26 January 2013 – Published in Biogeosciences Discuss.: 14 February 2013

Revised: 26 July 2013 – Accepted: 5 August 2013 – Published: 10 September 2013

Abstract. A profound understanding of temporal and spatial variabilities of soil carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O) fluxes between terrestrial ecosystems and the atmosphere is needed to reliably quantify these fluxes and to develop future mitigation strategies. For managed grassland ecosystems, temporal and spatial variabilities of these three soil greenhouse gas (GHG) fluxes occur due to changes in environmental drivers as well as fertilizer applications, harvests and grazing. To assess how such changes affect soil GHG fluxes at Swiss grassland sites, we studied three sites along an altitudinal gradient that corresponds to a management gradient: from 400 m a.s.l. (intensively managed) to 1000 m a.s.l. (moderately intensive managed) to 2000 m a.s.l. (extensively managed). The alpine grassland was included to study both effects of extensive management on CH₄ and N₂O fluxes and the different climate regime occurring at this altitude. Temporal and spatial variabilities of soil GHG fluxes and environmental drivers on various timescales were determined along transects of 16 static soil chambers at each site. All three grasslands were N₂O sources, with mean annual soil fluxes ranging from 0.15 to 1.28 nmol m⁻² s⁻¹. Contrastingly, all sites were weak CH₄ sinks, with soil uptake rates ranging from -0.56 to -0.15 nmol m⁻² s⁻¹. Mean annual soil and plant respiration losses of CO₂, measured with opaque chambers, ranged from 5.2 to 6.5 μmol m⁻² s⁻¹. While the environmental drivers and their respective explanatory power for soil N₂O emissions differed considerably among the three grasslands (adjusted *r*² ranging from 0.19 to 0.42), CH₄ and CO₂ soil fluxes were much better constrained (adjusted *r*² ranging from 0.46 to 0.80) by soil water content and air temperature, respectively. Throughout the year, spatial heterogeneity was particularly high for soil N₂O and CH₄ fluxes. We found perma-

nent hot spots for soil N₂O emissions as well as locations of permanently lower soil CH₄ uptake rates at the extensively managed alpine site. Including hot spots was essential to obtain a representative mean soil flux for the respective ecosystem. At the intensively managed grassland, management effects clearly dominated over effects of environmental drivers on soil N₂O fluxes. For CO₂ and CH₄, the importance of management effects did depend on the status of the vegetation (LAI).

1 Introduction

About 10 % of the fossil fuel emissions that originate from EU-25 countries have been shown to be absorbed by terrestrial ecosystems (Janssens et al., 2003; Schulze et al., 2009). While most of the atmospheric CO₂ is sequestered by forests, grasslands are a small net sink for atmospheric CO₂. Croplands are reported to be CO₂ neutral, and managed peatlands act as net sources for atmospheric CO₂ (Ciais et al., 2010b). However, the net terrestrial sink for atmospheric CO₂ is almost counterbalanced by CH₄ and N₂O emissions from agriculture (Ciais et al., 2010a). Despite the relatively small atmospheric concentrations of CH₄ and N₂O, these two greenhouse gases (GHG) account for 26 % of the global warming effect due to their high global warming potential (GWP) on a per-mass basis over a 100 yr time horizon (IPCC, 2007). Thus, understanding temporal and spatial variabilities of soil CO₂, CH₄ and N₂O fluxes between terrestrial ecosystems and the atmosphere is of key importance to reliably estimate these GHG fluxes and to develop mitigation strategies. While Vleeshouwers and Verhagen (2002) and Janssens et al. (2003) reported that the GWP of European

grasslands is still highly uncertain, Soussana et al. (2007) and Schulze et al. (2009) estimated that European grasslands had negative GWPs (including vertical and lateral fluxes of the three GHGs: e.g., fertilizer input, harvested biomass, animal emissions). In particular, all these investigated European grasslands were net sinks for CO₂ on an annual timescale. Soil fluxes of CH₄ and N₂O, expressed in CO₂ equivalents, reduced the net CO₂ sink capacity, but were too small to change the overall effect into a positive GWP (Soussana et al., 2007; Schulze et al., 2009). These two studies, however, did not consider grasslands at higher elevations, with cooler and wetter climatic conditions that may subsequently lead to higher soil CH₄ and N₂O emissions. Mountain regions are characterized by more orographic precipitation under episodic influences of maritime air masses, with lower evapotranspiration rates due to lower temperatures, both contributing to wetter soils (Beniston, 2005) and potentially higher soil emissions of CH₄ and N₂O.

Many studies used manually operated closed soil chambers to measure soil fluxes of CO₂, CH₄ and N₂O (Flessa et al., 2002; Pumpanen et al., 2004; Wang et al., 2005; Jones et al., 2006; Jiang et al., 2010; Rochette, 2011), as they are applicable in a wide range of ecosystems with varying site conditions. Moreover, chamber methods, in contrast to the eddy covariance method, are capable of detecting small flux magnitudes that are characteristic for CH₄ and N₂O fluxes between climatic- or management-driven pulse events (Baldocchi et al., 2012). In addition, the use of chambers allows for detection of spatial patterns in GHG fluxes (Flessa et al., 2002; Merbold et al., 2011), an important aspect when studying managed or undulating grassland sites (Ambus and Christensen, 1994; Ball et al., 1997; Mathieu et al., 2006). Especially N₂O and CH₄ fluxes are known for their non-uniform spatial distribution of sources and sinks (e.g., Matthias et al., 1980; Folorunso and Rolston, 1984; van den Pol-van Dasselaar et al., 1998). The spatial patterns are often controlled by grazing, vegetation and soil properties such as soil type and texture, soil temperature and soil water content, as well as soil C and N contents and microtopography (e.g., Dalal and Allen, 2008). In managed grasslands, temporal variability of soil GHG fluxes needs to be taken into consideration additionally since flux magnitudes respond not only to environmental forcings but also to human-induced activities such as harvesting and fertilization (Buchmann, 2011).

To understand how annual soil GHG fluxes respond to environmental- and management-induced forcings, we quantified temporal and spatial variations of chamber-based soil CO₂, CH₄ and N₂O fluxes at three Swiss grasslands, which are part of a traditional three-stage farming system at different altitudes (Weiss, 1941; Boesch, 1951; Ehlers and Kreutzmann, 2000). Our specific objectives were as follows: (1) to investigate the source/sink behavior of soil CO₂, CH₄ and N₂O fluxes at three differently managed grasslands located along an altitudinal gradient under different climatic conditions; (2) to assess temporal variation of soil GHG fluxes at

seasonal and annual timescales; and (3) to identify the spatial variations within sites.

2 Materials and methods

2.1 Site description

The three-stage farming system that is typical for many parts of the Swiss Alps (Ehlers and Kreutzmann, 2000) was represented exemplarily by the three ETH research sites Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS). The lowland site is represented by Chamau, situated north of Lake Zug in the pre-alpine lowlands of Switzerland at 400 m a.s.l. (47°12'37" N, 8°24'38" E), with a mean annual temperature of 9.1 °C and 1151 mm of precipitation (Sieber et al., 2011; Finger et al., 2013). The dominating soil type at CHA is a Cambisol (pH = 5) with bulk density ranging between 0.9 and 1.2 10³ kg m⁻³ in the uppermost 20 cm. The grass mixture consists of two dominating species, i.e., Italian ryegrass (*Lolium multiflorum* Lam.) and white clover (*Trifolium repens* L.). In 2010 and 2011, the pastures were used for forage production (Sautier, 2007; Zeeman et al., 2010). The site is intensively managed, with 5 to 10 management events per calendar year, including harvests and subsequent slurry applications. The exact number of management events per calendar year naturally depends on seasonal weather conditions.

The so-called Maiensaess site (early season mountain rangeland) is represented by Frübül, situated at the Zugerberg mountain ridge, east of Lake Zug (47°6'57" N, 8°32'16" E) at 1000 m a.s.l., with a mean annual temperature of 6.1 °C and 1682 mm of precipitation (Sieber et al., 2011; Finger et al., 2013). At FRU, the dominating soil type is a Gleysol (pH = 4.5), with bulk density ranging between 1.3 and 1.4 10³ kg m⁻³ (uppermost 20 cm). The grass mixture is more diverse than at CHA, consisting of ryegrass (*Lolium multiflorum* Lam.), meadow foxtail (*Alopecurus pratensis* L.), cocksfoot grass (*Dactylis glomerata* L.), dandelion (*Taraxacum officinale*), buttercup (*Ranunculus* L.) and white clover (*Trifolium repens* L.) (Sautier, 2007; Zeeman et al., 2010). The pastures at FRU are used for cattle grazing in late spring (May) and early fall (September). The management includes two to four harvests and/or fertilization events (slurry/manure) per calendar year, depending on local weather conditions and resulting vegetation growth.

The alpine site is represented by Alp Weissenstein, situated near the Albula pass at 2000 m a.s.l. in the canton Grisons (46°34'59" N, 9°47'25" E), with a mean annual temperature of -1.4 °C and 877 mm of precipitation (Sieber et al., 2011; Finger et al., 2013; Michna et al., 2013). The soils at AWS are slightly humus to humus sandy loam (pH = 5.6), with comparable bulk density in the uppermost 20 cm as reported for FRU (1.3 to 1.4 10³ kg m⁻³). The grass composition is classified as *Deschampsio cespitosae-Poetum*

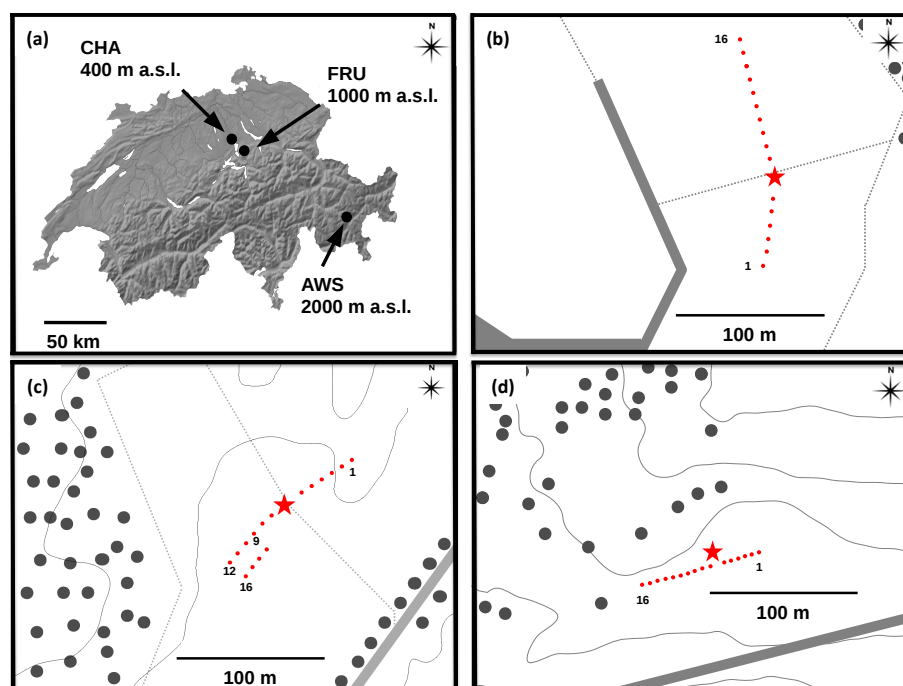


Fig. 1. Topographical map of Switzerland (a) showing the geographic position of the sites, (b) Chamau (CHA), (c) Frübül (FRU) and (d) Alp Weissenstein (AWS) as well as site setup maps (b–d). The red star indicates the position of the eddy covariance tower, and the red dots the position of the chambers along the prevailing wind direction (flux footprint; modified after Zeeman et al., 2010). The numbers refer to the chamber numbers along the transects. Dots denote individual trees, and gray bold lines paved roads.

alpinae, with red fescue (*Festuca rubra*), alpine cat's tail (*Phleum rhaeticum*), white clover (*Trifolium repens* L.) and dandelion (*Taraxacum officinale*) (Sautier, 2007; Zeeman et al., 2010). AWS represents the summer rangelands for cattle grazing without harvests in normal years. Manure is applied to the pastures at the end of the grazing season, typically in the second half of September. The geographic location of the three sites as well as the site setups are shown in Fig. 1, while dates for harvests and fertilization events are shown in Table 1.

2.2 Experimental setup

From June 2010 to June 2011, we measured soil fluxes of CO₂, CH₄ and N₂O using opaque static soil chambers at all three sites. The diameter of the polyvinyl chloride chambers was 0.3 m, the average headspace height 0.136 m (± 0.015 m) and average insertion depth of the collars was 0.08 m (± 0.05 m). On sampling campaigns with vegetation inside the chamber > 0.20 m, collar extensions (0.45 m) were used. Chamber lids were equipped with reflective aluminum foil to minimize heating inside the chamber during the period of actual measurement. At each site, transects of 16 soil chamber collars were installed in May 2010 (Fig. 1). Spacing between the chambers was 7 m at CHA and FRU, and 5 m at AWS. At FRU, two transects were established, one consisting of 12 chambers and a second consisting of four

chambers (Fig. 1c). This was done to adopt the sampling approach to the usual management regime, which is coerced by two differently managed field parcels. Spacing was chosen so that the transects would fit into the previously calculated footprint of the eddy covariance towers at the respective sites (Zeeman et al., 2010), as well as to cover the topographic differences at each site. At FRU and AWS, chamber collars were fenced to avoid trampling and/or removal by the cattle. Sampling of the chambers was performed on a weekly basis during the growing season, and at least once a month during the winter season (except for AWS, which is inaccessible in winter). The vegetation inside the chamber collars was manually harvested at the times of regular management activities, i.e., harvests and grazing.

Additionally, we measured diel patterns of soil CH₄ and N₂O fluxes in an intensive observation campaign in September 2010. During this campaign, soil CH₄ and N₂O fluxes were measured over 48 consecutive hours at all three sites simultaneously, at intervals of two hours. During this 48 h intensive observation campaign (21–23 September 2010), 75 mean chamber fluxes of CH₄ and N₂O were obtained at the three sites ($N_{\text{CHA}} = 25$; $N_{\text{FRU}} = 25$; $N_{\text{AWS}} = 25$).

Table 1. Management activities (harvests and fertilization: SL = slurry; MA = manure) and nitrogen inputs (kg N ha^{-1}) during flux measurement periods at the three sites Chamau (CHA), Frübül (FR) and Alp Weissenstein (AWS). Numbers indicate nitrogen inputs from slurry/manure in kg ha^{-1} . At AWS, no harvest was performed (-).

	CHA		FRU		AWS	
Cuts	20 Aug 2010		2 Jul 2010		-	
	2 Oct 2010		2 Aug 2010			
	21 Apr 2011					
	15 Jun 2011					
Fertilization	6 Jul 2010	SL, 18	19 Jul 2010	MA, 33	30 Aug 2010	MA, n.a.
	25 Aug 2010	SL, 30	11 Sep 2010	SL, 10		
	28 Oct 2010	SL, 44	23 Mar 2011	SL, 140		
	10 Mar 2011	SL, 60				
	28 Apr 2011	SL, 35				
	22 Jun 2011	SL, 52				

2.3 Data acquisition and processing

2.3.1 Flux sampling and calculations

GHG fluxes were calculated based on the rate of changing gas concentrations inside the chamber headspace. After closing the chambers, four samples were taken, one immediately after closure and then at 10 to 13 min increments so that the chamber was no longer closed than 40 min. This closing time was sufficiently short to avoid saturation effects inside the chamber headspace. We inserted 60 mL syringes into the chambers lid septums to take the gas samples, and then injected the gas into pre-evacuated 12 mL vials (Labco Limited, Buckinghamshire, UK). Prior to the second, third and fourth sampling of each chamber, the chamber headspace was flushed with the syringe volume of air from the chamber headspace to minimize effects of built-up concentration gradients inside the chamber. Gas samples were analyzed for their CO_2 , CH_4 and N_2O concentrations using gas chromatography (Agilent 6890 gas chromatograph equipped with a flame ionization detector, a methanizer and an electron capture detector, Agilent Technologies Inc., Santa Clara, USA) as described by Hartmann et al. (2011).

Data processing, which included flux calculation and quality checks, was carried out with the R statistical software (R Development Core Team, 2010). The rate of change was calculated by the slope of the linear regression between gas concentration and time. Fluxes were always small enough (r^2 and visual inspection of concentration changes with time) that no saturation in measured concentration data could be detected that would be indicative of saturation effects inside the chamber. We used the following equation to derive the flux estimate F_{GHG} :

$$F_{\text{GHG}} = \frac{\delta c}{\delta t} \cdot \frac{p \cdot V}{R \cdot T \cdot A}, \quad (1)$$

where c is the respective GHG concentration ($\mu\text{mol mol}^{-1}$ for CO_2 ; nmol mol^{-1} for CH_4 and N_2O). Time (t) is given in seconds, atmospheric pressure (p) in Pa, the headspace volume (V) in m^3 , the universal gas constant (R) is $8.3145 \text{ m}^3 \text{ Pa K}^{-1} \text{ mol}^{-1}$, T is ambient air temperature (K) and A is the surface area enclosed by the chamber (m^2). Individual chamber fluxes were only computed if the linear regression for each individual GHG yielded a $r^2 \geq 0.8$. If the slope between the first and second concentration obviously deviated from the one of the remaining three concentration measurements, then we omitted it and calculated the flux from the remaining three. Chambers for which the rate of change of CO_2 was negative were also discarded, as photosynthesis is assumed to be zero inside the opaque chamber. The mean chamber flux was then calculated as the arithmetic mean of all available individual chamber fluxes for each date and site.

A total of 81 sampling campaigns were performed between June 2010 and June 2011 ($N_{\text{CHA}} = 35$; $N_{\text{FRU}} = 32$; $N_{\text{AWS}} = 17$), resulting in an equivalent number of mean chamber fluxes of N_2O , CH_4 and CO_2 . We follow the micrometeorological convention, where positive fluxes are directed to the atmosphere, and negative fluxes to the ecosystem.

2.3.2 Ancillary measurements

At each field site, the following environmental variables were recorded in the center of the transects (eddy covariance towers, Fig. 1) as 10 min averages: air temperature (T_a) at 2 m (HydroClip S3, Rotronic AG, Basserdorf, CH), soil temperature (T_s) at -0.02 m (TL107 sensors, Markasub AG, Olten, CH), volumetric soil water content (SWC) at -0.05 m (ML2x, Delta-T Devices Ltd, Cambridge, UK), and photosynthetic active radiation (PAR) at 2 m (PARlite, Kipp and Zonen B.V., Delft, The Netherlands). Leaf area index (LAI) of the vegetation outside the chambers was measured at each flux sampling campaign (LAI-2000, Licor, Lincoln,

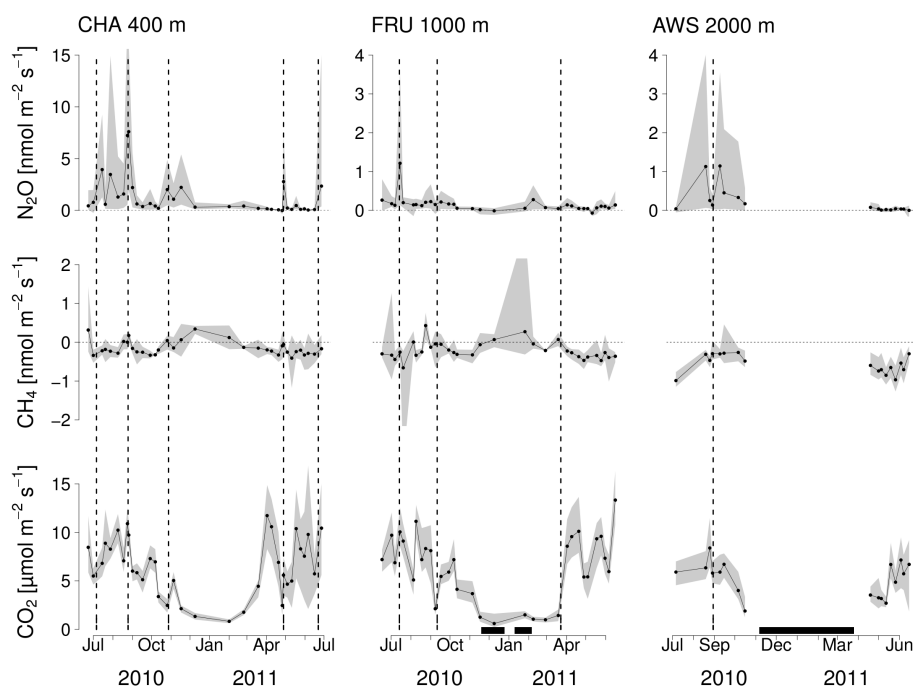


Fig. 2. Annual courses of N_2O , CH_4 and CO_2 fluxes at Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS). The black lines indicate the mean soil flux of the respective greenhouse gas (GHG), and the gray-shaded areas indicate the 95 % confidence intervals. Dashed, vertical lines indicate fertilizer applications. Black boxes for FRU and CHA denote periods of permanent snow cover. Note the different scaling for N_2O fluxes at the three sites. Fluxes of N_2O and CH_4 are given in $\text{nmol m}^{-2} \text{s}^{-1}$ and CO_2 in $\mu\text{mol m}^{-2} \text{s}^{-1}$.

USA), i.e., every week during the growing period and at least monthly during winter (when there was no snow cover). LAI measurements represent averages of 12 measurements along the chamber transects.

2.3.3 Statistics

For each site, mean chamber GHG fluxes were used to establish functional relationships with environmental drivers on annual timescales, using stepwise multiple linear regression models for annual timescales. A standardized principal component analysis (PCA) was performed prior to the multiple linear regressions to minimize potential artifacts from colinearities between environmental drivers. Since CH_4 fluxes at CHA and FRU showed exponential relationships with soil water content, data were log-transformed for the respective multiple linear models. The relative importance of each driver within the regression model was estimated using hierarchical partitioning (Chevan and Sutherland, 1991). The uncertainty of the calculated relative importance was assessed using parametric bootstrapping methods (Efron, 1979).

To investigate spatial variability of GHG fluxes, a one-way analysis of variance (ANOVA) was performed. The chamber number along the transect (Fig. 1) was used as a predictor, and all valid annual flux values per chamber were included. To assess the statistical significance of pairwise comparisons

of ANOVA results, the Tukey honestly significant differences (HSD) test was chosen.

3 Results

3.1 Temporal variation of GHG fluxes

3.1.1 Seasonal flux patterns and management

Soil fluxes of N_2O and CO_2 showed a commonly known seasonal pattern, with highest emission rates during the summer months (Fig. 2). Soil emissions of CH_4 were mostly observed during winters, whereas uptake rates were prevailing in summers.

At all three sites, soil N_2O fluxes were mostly positive, indicating a source for N_2O . Occasional uptake was observed for mean and individual chamber fluxes, as indicated by the 95 % confidence interval in Fig. 2. Highest N_2O efflux rates were observed at CHA (intensively managed) shortly after slurry applications. These peak fluxes exceeded maximum N_2O fluxes at FRU (moderately managed) and AWS (extensively managed) by a factor of 2 and higher. Emission factors at the intensively at CHA were on average 3.1 ± 3.6 %. At FRU, emission factors were 0.8 ± 0.8 %. At AWS, peak emissions occurred around the manure application and biomass removal, which was done manually during the period of grazing. Mean N_2O soil efflux over the 12-month

study period was highest at CHA with $1.28 \text{ nmol m}^{-2} \text{ s}^{-1}$, and lowest at FRU with $0.15 \text{ nmol m}^{-2} \text{ s}^{-1}$. At AWS, an average efflux of $0.23 \text{ nmol m}^{-2} \text{ s}^{-1}$ was observed. The low mean N_2O efflux at FRU is likely due to low emission rates during winter, which were missing from the AWS data. Hence mean emissions were larger at AWS than at FRU.

At CHA and FRU, both positive and negative methane fluxes were observed, yet uptake dominated at both sites. During eight sampling campaigns, which represented 20% of all campaigns, CHA was a source of CH_4 . FRU was a source during five sampling dates, representing 12.5% of all campaigns, while AWS acted as a sink for atmospheric CH_4 throughout the measurement period. However, no measurements are available for the winter period at this site, due to the inaccessibility of the site. The response of CH_4 fluxes to fertilization, i.e., reduced uptake rates, was not as distinct as for N_2O fluxes. Over the 12-month study period, mean uptake rates for CH_4 were -0.15 , -0.22 and $-0.56 \text{ nmol m}^{-2} \text{ s}^{-1}$ at CHA, FRU and AWS, respectively.

Temporal flux patterns of CO_2 were comparable at all three sites. The annual range of flux magnitudes was slightly smaller at AWS than at CHA and FRU. Respiration increased after fertilization at all three sites, yet the magnitude of this response was variable (Fig. 2). Over the 12-month study period, mean respiration rates were 6.5 and $6.3 \text{ } \mu\text{mol m}^{-2} \text{ s}^{-1}$ at CHA and FRU, respectively. At AWS, respiration rates were slightly smaller, with an overall mean flux of $5.2 \text{ } \mu\text{mol m}^{-2} \text{ s}^{-1}$ (growing season average).

3.1.2 Seasonal response of fluxes to environmental drivers

With the PCA, we were able to identify similarities among potential driver variables (T_a , T_s , SWC, PAR and LAI) and to reduce the set of driver variables for exploring functional relationships to those drivers that (a) are as independent from each other as possible, and (b) that are of similar relevance at all three elevations, such that functional relationships built with the selected drivers can be compared among sites. Nitrogen inputs in the form of slurry/manure applications were not considered for the PCA, as only six and three data points would have been available at CHA and FRU, respectively, and LAI can already be seen as a proxy for management activity. At CHA and FRU, T_a and T_s had similar loadings (Fig. 3) because both followed a similar annual cycle. Since T_a generally had the higher explanatory power than T_s , we selected T_a . The first and second principal components further indicated that PAR and SWC may be treated as one variable at CHA and FRU (Fig. 3), where variations in PAR were of opposite sign compared to variations in SWC, simply indicating that episodic increases in SWC after rain events at the lower two sites coincided with periods of high cloudiness that reduced PAR over several days. In contrast, under fair weather conditions, the typical diurnal cycle of PAR was likely linked to a similar cycle of SWC in the opposite

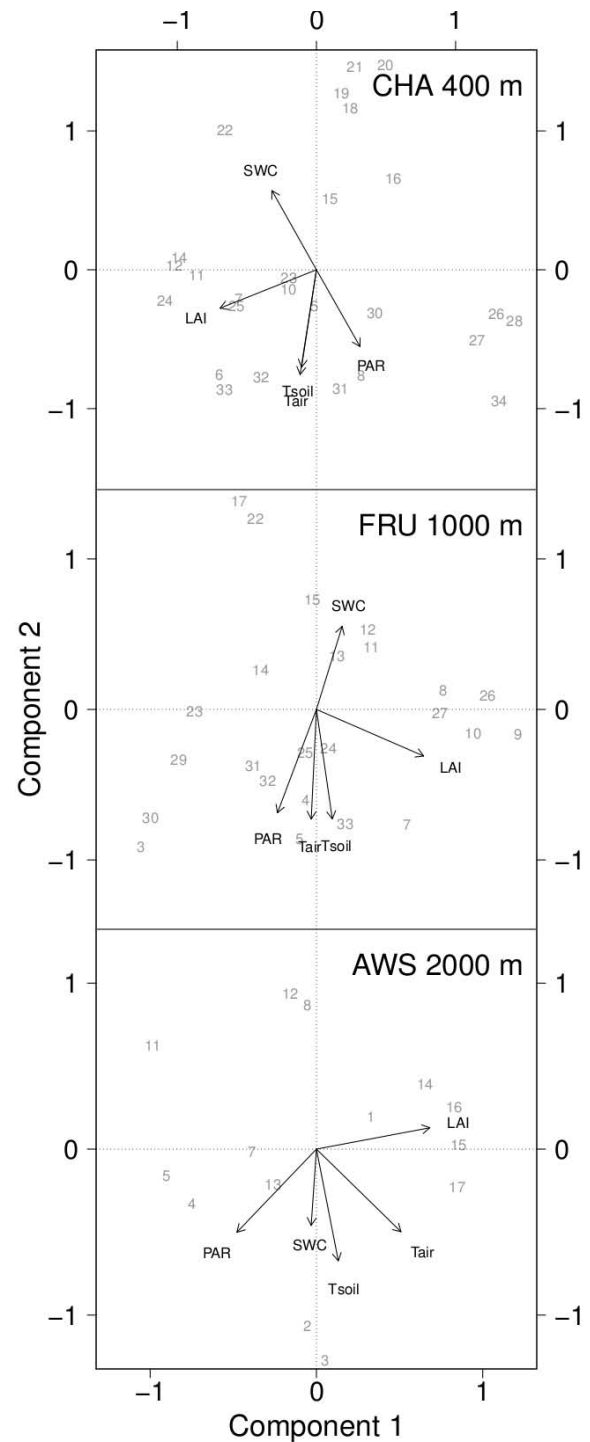


Fig. 3. Biplots of the first two components of a standardized principal component analysis of environmental variables for the three sites Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS). Environmental variables measured during sampling campaigns within the 12-month study period were considered (numbers 1 to maximum 34 at CHA). T_a = air temperature; T_s = soil temperature; SWC = soil water content; LAI = leaf area index; and PAR = photosynthetic active radiation.

direction. Since our selection of T_a already represented atmospheric conditions at a site, we selected SWC instead of PAR as the second variable. The third variable selected via PCA was LAI, which was almost independent of SWC at all three sites (Fig. 3), and hence was expected to add explanatory value to a functional model where the three compartments atmosphere (T_a), soil (SWC) and vegetation (LAI) were represented (Fig. 4).

The explanatory power of the multiple linear model with regard to the temporal variation of N_2O fluxes varied considerably at the three sites, with adjusted r^2 values ranging from 0.19 to 0.42. The relative importance (RI) of each selected driver, i.e., the contribution to the overall variance of flux variability explained by the model, was not consistent among sites (Fig. 4). At CHA, the model was able to explain 42% of the total variance inherent in annual soil N_2O flux data, with LAI and T_a being the most important explanatory variables ($RI_{LAI} = 45\%$; $RI_{T_a} = 38.9\%$). SWC had much less influence on the N_2O efflux, with a RI of 16.1%. At FRU, T_a was clearly the most important driver, with a RI of 84.7%, followed by SWC with a RI of 14%. LAI was of minor importance, with a RI of 1.3%. In total, only 19% of the variance in soil N_2O fluxes was explained by the model at FRU. At AWS, 34% of the variance was explained. Here, SWC was the most powerful explanatory variable, with a RI of 54.7%, followed by LAI ($RI = 43.7\%$). T_a had almost no impact on the variability of the N_2O flux at AWS ($RI = 1.7\%$) (Fig. 4).

The variation of explanatory power among the driver variables within the multiple linear model for the prediction of CH_4 fluxes was more pronounced than that for N_2O fluxes. However, soil CH_4 fluxes were better constrained by the set of drivers, with adjusted r^2 values ranging from 0.46 to 0.83. Again, the RI of the drivers was not consistent among sites (Fig. 4). At CHA, the model explained 46% of the total variance inherent in all annual soil CH_4 flux data, with LAI and SWC being the most influential variables ($RI_{LAI} = 45.4\%$; $RI_{SWC} = 35.4\%$), followed by T_a with a RI of 19.2%. At FRU, 72% of the total variance was explained, with SWC being the most important variable, exhibiting a RI of 89%. At AWS, 83% of variance was explained by the model, of which 82.7% were due to changes in SWC. At both sites, FRU and AWS, the variables LAI and T_a had minor influences on soil CH_4 fluxes.

The explanatory power of the multiple linear model for soil CO_2 fluxes was almost the same at CHA and FRU, with 80% total explained variance. At AWS, the model was still able to explain 47%. At CHA and FRU, T_a was the most influential variable, with RI values of 71 and 81%, respectively. Yet, at CHA, LAI had a considerable influence on the temporal variability of CO_2 fluxes, with a RI of 21.8%. At FRU, the contribution of seasonal changes in LAI was less important ($RI = 9.1\%$). At AWS, T_a was the most important variable in the model similar to CHA and FRU (RI of 55.7%), followed by SWC with a RI of 30.9% (Fig. 4).

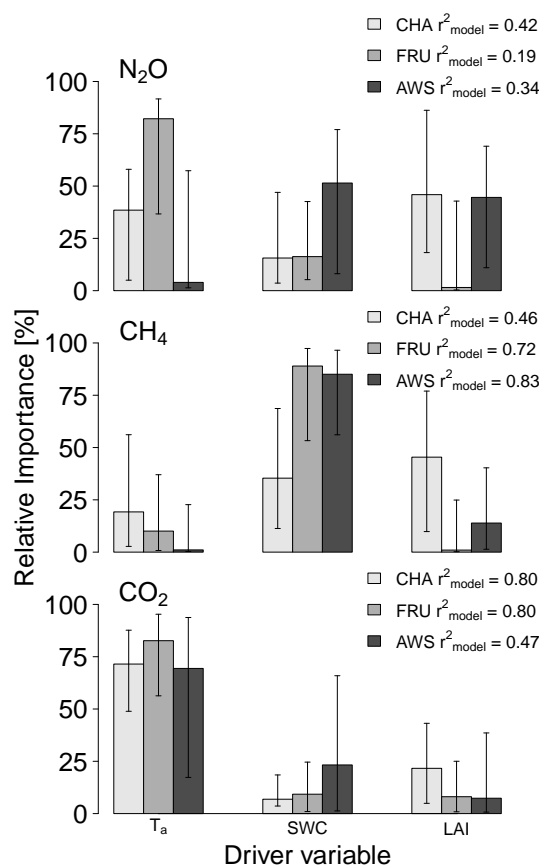


Fig. 4. Explanatory power of driver variables for GHG fluxes on the annual timescale at Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS), with T_a as air temperature, SWC as soil water content and LAI as leaf area index. Contribution (relative importance) to the overall variance explained is given in %. Error bars indicate 95% confidence intervals as determined from bootstrapping ($N_{runs} = 1000$). r^2 values represent overall model performance. Significance levels of each driver can be found in Table 2. The upper panel shows the model performance at all three sites for N_2O fluxes, the middle panel for CH_4 fluxes and the lower panel for CO_2 fluxes.

3.1.3 Diel variation of N_2O and CH_4 fluxes

Mean chamber efflux rates of N_2O and mean chamber uptake rates of CH_4 were observed during the intensive observation campaign at all sites in September 2010 (Fig. 5). The diel N_2O flux magnitudes along the elevational transect yielded a different ranking among sites than that observed at the annual scale. During the intensive observation campaign, highest emissions of N_2O were measured at AWS, with an average flux of $0.54 \text{ nmol m}^{-2} \text{ s}^{-1}$, followed by CHA with 0.21 and FRU with $0.15 \text{ nmol m}^{-2} \text{ s}^{-1}$. Diel variations of soil N_2O fluxes were clearly found only at FRU, where high emission rates were observed during the day, and smaller emissions during nights (Fig. 5). T_a was a good predictor for N_2O efflux rates at the diel timescale at CHA and FRU, explaining

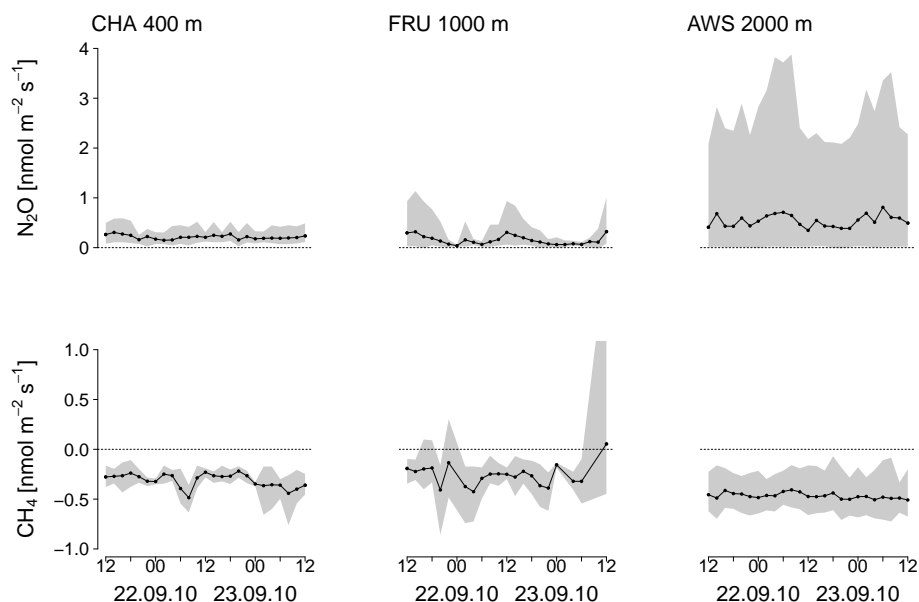


Fig. 5. Diel courses of N_2O (top) and CH_4 (bottom) fluxes at Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS). The gray-shaded areas indicate the 95 % confidence intervals of mean chamber fluxes (black line). Measurements started at noon (12:00) on September 21 and ended at noon (12:00) on 23 September.

54 and 59 % of the variance, respectively. In contrast, N_2O emissions did not significantly correlate with T_a at the diel scale (nor at the annual scale) at AWS (Fig. 4).

Highest mean uptake rates of CH_4 were measured at AWS with $-0.47 \text{ nmol m}^{-2} \text{ s}^{-1}$, followed by CHA and FRU with -0.31 and $-0.16 \text{ nmol m}^{-2} \text{ s}^{-1}$ (Fig. 5). Although considerable variation in soil CH_4 flux rates was visible at CHA and FRU, no obvious diel trend was identified. At AWS, CH_4 uptake rates were almost constant, with only very little temporal variation. Hence, regression analysis to determine flux drivers was not successful. At CHA, 13 % of the variance in CH_4 uptake rates could be explained by T_a , while no significant relationship could be established at FRU and AWS. SWC variations, important at the annual scale, were affected only slightly during the 48 h intensive observation campaign, and were therefore omitted for developing any explanatory power at the diel scale.

3.2 Spatially invariant hot spots of GHG fluxes on the seasonal scale

Spatial variability of annually averaged soil N_2O fluxes, i.e., the flux variation among chambers along the transect, was highest at CHA (Fig. 6), in contrast to the spatial variation seen over 48 h. The one-way ANOVA with chamber as a factor yielded a p value of 0.57 for soil N_2O fluxes, indicating that all chambers showed high variation during the 12 months of measurements. At AWS, chambers one to three showed a wider range of annual average N_2O efflux rates relative to the other chambers along the transect. Yet, due to some very high flux estimates, the ANOVA yielded a p value of

0.52, indicating no significant differences among individual chambers. This suggested that the spatial variability of soil N_2O fluxes was not larger than the temporal variability of the fluxes measured at AWS. Spatial variations of annual average N_2O fluxes at FRU were negligible (Fig. 6), and in a similar magnitude as at the diel scale (not shown).

Spatially invariant annual averages of soil CH_4 fluxes were found at CHA and FRU (Fig. 6). At AWS, we observed a spot of significantly ($p = 0.02$) lower CH_4 uptakes rates around chambers two and three (Fig. 6).

4 Discussion

It was not a priori expected that the three grasslands at different elevations, and thus management intensities, were all weak net sinks for CH_4 due to abundant precipitation in mountainous areas, whereas the tight coupling between soil N_2O efflux rates and fertilization confirmed earlier studies. The alpine grassland (AWS), characterized by sufficient rain-water supply and hypothesized to be a net source of CH_4 , acted as a net sink of methane, given the fact that the long winter period could not be included in our measurement regime. In addition, soil N_2O as well as CH_4 fluxes were highly variable in time and space, supporting previous studies (e.g., Folorunso and Rolston, 1984; Mosier et al., 1991; Velthof et al., 2000).

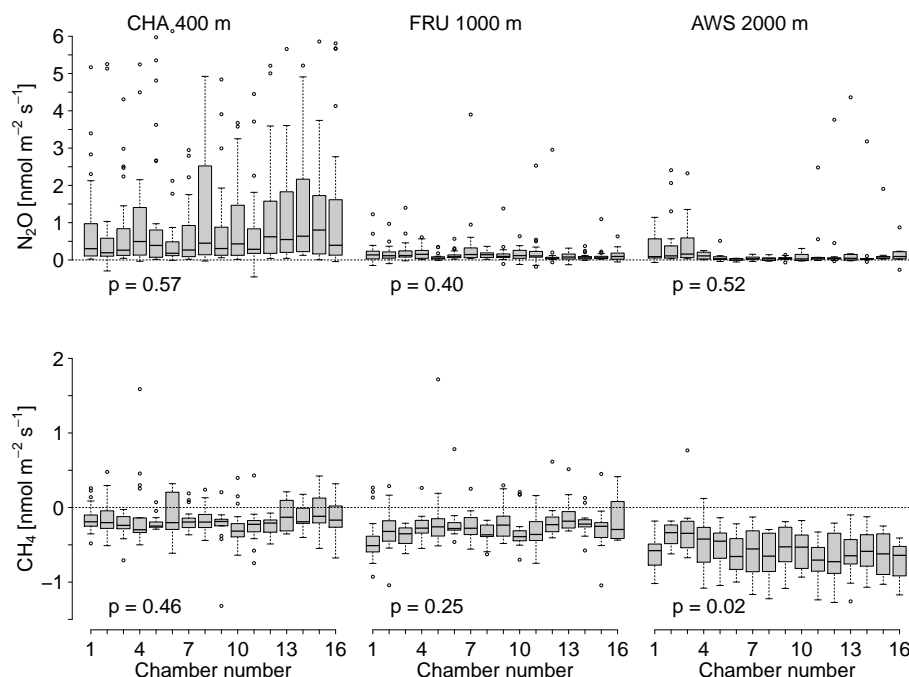


Fig. 6. Box plots depicting spatial gradients of mean annual N_2O and CH_4 chamber fluxes (1–16 on x axis) at Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS). The p values refer to the ANOVA results with chamber as a factor. Values below 0.05 indicate significant differences among mean chamber fluxes over the 12-month study period.

4.1 Seasonal GHG fluxes and drivers of their temporal variability

4.1.1 N_2O fluxes

Our measurements give strong evidence that managed grasslands are a constant source of N_2O , as hardly any uptake was observed (Ryden, 1981; Wagner-Riddle et al., 1997; Glatzel and Stahr, 2001; Neftel et al., 2007). The small number of negative soil N_2O fluxes (uptake) observed was evenly distributed throughout the 12-month study period. As expected, the intensively managed site, CHA, was the strongest source of N_2O . While total N addition (per application) at CHA was comparable to FRU, mean annual emissions were more than 8 times higher than those at FRU, leading to much higher emission factors at CHA. FRU was characterized by the lowest mean annual N_2O emissions. Emission factors at the intensively managed grassland (CHA) were on average $3.1 \pm 3.6\%$, and therefore considerably higher than the IPCC (2007) default value of 1.25% (without grazing). At FRU (moderately managed), emission factors were only $0.8 \pm 0.8\%$.

Our findings also underline the challenge of predicting N_2O fluxes from grasslands. N_2O fluxes were weakly constrained by a set of three environmental variables. Their influence on the flux varied temporally and from site to site, indicating the importance of additional factors (e.g., land management, fertilization). Yet, our results are in agreement with

previous studies, which identified air temperature and soil water content as influential variables (e.g., Wang et al., 2005; Liebig et al., 2010; Schauffer et al., 2010). The high relative importance of LAI at CHA (intensively managed) can be explained by the fact that step changes in LAI due to regular harvests during the growing season reflect subsequent slurry applications at CHA (N addition usually within five days after the harvest). This finding underlines the importance of a realistic management consideration when predicting N_2O fluxes. This was corroborated in Fig. 7, which shows the response of the three GHG fluxes to their primary environmental driver (at two LAI classes) on the annual scale (Table 2). For N_2O we found that T_a had no significant influence on flux magnitudes at constant LAI ranges. Thus, management (with LAI as proxy) had a larger effect on N_2O fluxes than the environment (with T_a as proxy).

4.1.2 CH_4 fluxes

Our results showed consistently small CH_4 sinks at all three sites, which is in agreement with other studies on temperate grasslands at low altitudes (Mosier et al., 1997; Liebig et al., 2010). Positive soil CH_4 fluxes mostly occurred during periods with high SWC and/or after fertilization (Lessard et al., 1997), and hence were observed most frequently at the intensively and moderately managed site, CHA and FRU. For FRU, our results are in contrast to the results presented by Hartmann et al. (2011), who exclusively measured uptake of

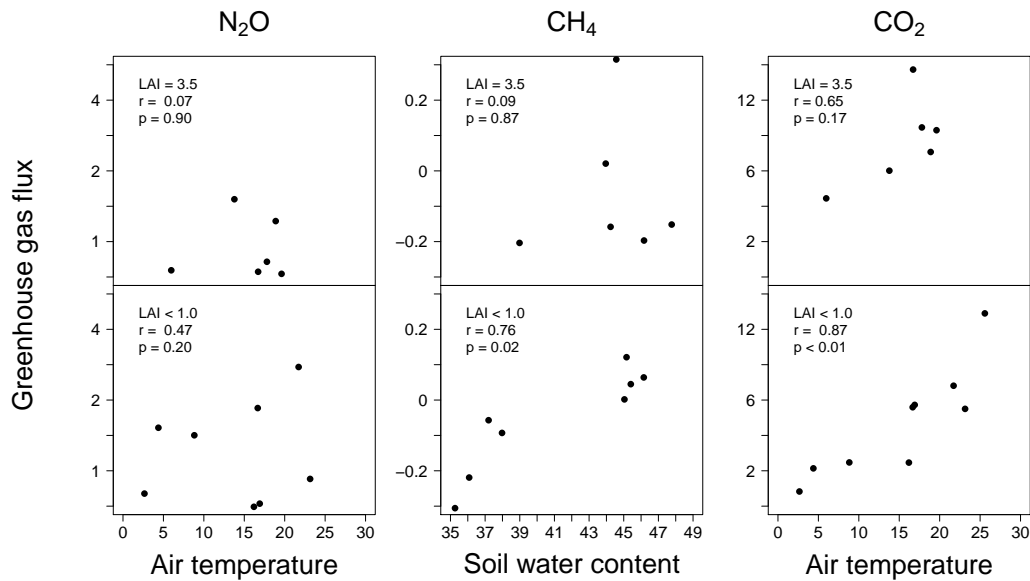


Fig. 7. Relationships of mean chamber GHG fluxes and their primary environmental drivers at two different LAI classes for the intensively managed site Chamau (CHA). For CH₄ and N₂O, fluxes are given in nmol m⁻² s⁻¹; for CO₂ in μmol m⁻² s⁻¹. Soil water content is given in vol. %, and air temperature in °C. Correlation coefficients and *p* values are given.

Table 2. Multiple linear model equations for annual GHG fluxes at the three sites Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS); *p* values are given for the individual drivers (*T_a* = air temperature; SWC = soil water content; LAI = leaf area index).

CHA	
N ₂ O	$= 6e^{-6} \pm 2e^{-6} \cdot T_a + 8e^{-4} \pm 3e^{-4} \cdot SWC - 2e^{-5} \pm 6e^{-6} \cdot LAI$ $\alpha = 0.002; \beta = 0.014; \gamma = 0.002$
log(CH ₄)	$= -0.01 \pm 0.02 \cdot T_a + 7.14 \pm 2.77 \cdot SWC - 0.17 \pm 0.05 \cdot LAI$ $\alpha = 0.751; \beta = 0.016; \gamma = 0.003$
CO ₂	$= 0.38 \pm 0.05 \cdot T_a + 24.24 \pm 8.43 \cdot SWC + 0.45 \pm 0.16 \cdot LAI$ $\alpha < 0.001; \beta = 0.024; \gamma = 0.009$
FRU	
N ₂ O	$= 7e^{-7} \pm 9e^{-5} \cdot T_a + 3e^{-5} \pm 2e^{-5} \cdot SWC - 1e^{-7} \pm 1e^{-6} \cdot LAI$ $\alpha = 0.038; \beta = 0.222; \gamma = 0.900$
log(CH ₄)	$= 8e^{-5} \pm 0.01 \cdot T_a + 4.36 \pm 0.68 \cdot SWC + 0.01 \pm 0.03 \cdot LAI$ $\alpha = 0.990; \beta < 0.001; \gamma = 0.860$
CO ₂	$= 0.39 \pm 0.05 \cdot T_a + 0.47 \pm 4.11 \cdot SWC + 0.20 \pm 0.17 \cdot LAI$ $\alpha < 0.001; \beta = 0.910; \gamma = 0.250$
AWS	
N ₂ O	$= 0.01 \pm 0.02 \cdot T_a + 1.79 \pm 1.17 \cdot SWC - 0.19 \pm 0.12 \cdot LAI$ $\alpha = 0.590; \beta = 0.160; \gamma = 0.160$
CH ₄	$= 3e^{-3} \pm 7e^{-3} \cdot T_a + 2.45 \pm 0.39 \cdot SWC - 0.09 \pm 0.04 \cdot LAI$ $\alpha = 0.990; \beta < 0.001; \gamma = 0.860$
CO ₂	$= 0.26 \pm 0.07 \cdot T_a + 7.96 \pm 3.71 \cdot SWC + 0.24 \pm 0.39 \cdot LAI$ $\alpha = 0.004; \beta = 0.057; \gamma = 0.557$

CH₄ at FRU in the years 2007, 2008 and 2009. However, their measurements were taken in generally drier soils, supporting the idea of regulating effects of SWC on CH₄ fluxes (RI of 89%). At the extensively managed AWS site, both Hartmann et al. (2011) and this study exclusively observed uptake of CH₄.

Annually, soil CH₄ fluxes were well predictable by SWC (Liebig et al., 2010; Schrier-Uijl et al., 2010; Hartmann et al., 2011). Furthermore, at the intensively managed site (CHA), LAI had comparable explanatory power to SWC. As mentioned before, step changes in LAI during the growing period reflect management activities (fertilization at low LAI). Although ammonium-based fertilizers (e.g., organic fertilizers) can inhibit CH₄ uptake (oxidation) by methanotrophs (Willison et al., 1995; Stiehl-Braun et al., 2011), SWC still had a highly significant impact on CH₄ fluxes at LAI < 1 (Fig. 7), indicating dominant environmental drivers.

4.1.3 CO₂ fluxes

Opaque soil chambers were used to exclusively measure respiratory fluxes of CO₂, which were in the expected range from close to zero in winter up to 15 μmol m⁻² s⁻¹ during summer, similar to, e.g., Myklebust et al. (2008). At FRU, chamber measurements agreed well with eddy covariance-based respiration data, simultaneously recorded at all three sites (Fig. 8). At CHA, eddy-covariance-based fluxes were systematically higher, which is likely due to the different scale of both measurement techniques (Wang et al., 2009). Except for one outlier of eddy-covariance-based soil respiration, chamber measurements agreed well with eddy covariance data at AWS. This outlier on August 25 may be an artifact of the gap-filling procedure for eddy covariance respiration data. Besides the expected importance of *T*_a and SWC for CO₂ efflux rates, LAI was once more a reasonable predictor (RI of 21.8%) at the intensively managed grassland (CHA). Keeping LAI constant at < 1 (Fig. 7) revealed a still highly significant effect of *T*_a on soil CO₂ fluxes, supporting the notion that management impacts on soil flux magnitudes of CO₂ are rather small compared to environmental drivers (Peng et al., 2011).

4.2 Diel variation of N₂O and CH₄ fluxes

In contrast to what we learned on the annual scale, highest emissions of N₂O of all three grasslands were observed at AWS, being twice the observed seasonal mean. As manure was applied to AWS pastures 10 days prior to sampling, we likely captured a “hot” moment, supporting our conclusion that management impacts are dominating soil N₂O flux variability. Already studies by Christensen (1983) and Flechard et al. (2007) have shown that peak emissions of N₂O appeared lagged to manure application (8 to 12 days).

In the literature, there is no consistent picture regarding the presence of significant diel patterns of N₂O and CH₄

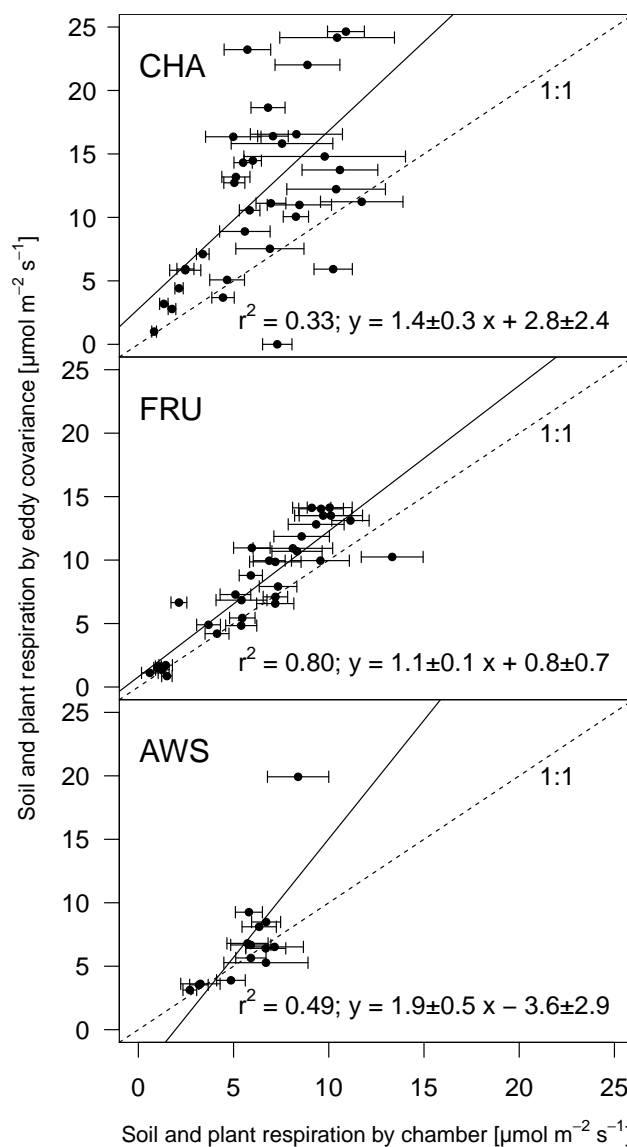


Fig. 8. Soil and plant CO₂ respiration rates calculated from eddy covariance and corresponding chamber-derived values. Mean chamber fluxes are shown for all sampling campaigns at Chamau (CHA), Frübül (FRU) and Alp Weissenstein (AWS); *p* values of the linear regressions were < 0.001 at CHA and FRU, and 0.003 at AWS. Uncertainties in the model equations are standard errors.

fluxes (Christensen, 1983; Skiba et al., 1996; Maljanen et al., 2002; Duan et al., 2005; Hendriks et al., 2008; Baldocchi et al., 2012). Duan et al. (2005) suggested that the type of ecosystem might have an influence on the presence of diel soil N₂O and CH₄ flux variations. However, our study exclusively investigated grasslands, and we found significant diel variations of N₂O fluxes for one out of three sites (FRU). At CHA, changes in air temperature affected soil N₂O fluxes less strongly, and thus no significant diel patterns in N₂O fluxes were observed. This might be attributable to the fact

that the last slurry application at CHA was more than four weeks prior to the intensive campaign (25.08.2010; slurry 30 kg N ha⁻¹), and that the vegetation composition at FRU exhibits a larger fraction of legumes. This suggests that not only ecosystem type, but also site specifics (e.g., management intensity or vegetation composition) might influence diel variations in soil N₂O fluxes.

In contrast to what we observed on the annual scale (Table 2), CH₄ fluxes were not constrained by changes in SWC over the course of 48 h, probably because changes were too small (decrease of less than 2 % vol) to significantly affect the magnitude of CH₄ fluxes.

4.3 Spatial patterns and autocorrelation

Working with soil chambers requires information on the spatial distribution of GHG fluxes at ecosystem scale to design appropriate experiments and to be able to correct mean soil fluxes for potential biases. In relation to the magnitude of mean chamber fluxes of N₂O and CH₄, individual chamber fluxes are highly variable in space (e.g., Matthias et al., 1980; Folorunso and Rolston, 1984; Mosier et al., 1991), as they are largely determined by small-scale biochemical processes (Ambus and Christensen, 1994; Dalal and Allen, 2008).

We observed that out of our three sites, only AWS exhibited permanent spots where CH₄ uptake was significantly smaller than at the rest of the transect. This corresponded well with local microtopographical conditions, i.e., the inclination of the terrain (Fig. 9). Chambers placed in terrain with greater inclination systematically exhibited lower SWC values (data not shown). This in turn corresponded well with what was observed at the annual scale, where lower CH₄ uptake rates correlated significantly with higher SWC. At CHA and FRU, we observed that flux magnitudes varied spatially; spots of reduced CH₄ uptake were, however, not permanent. Thus, the situation at CHA and FRU represents the ideal case when trying to sample a representative mean of an ecosystem. Omitting permanent hot spots may lead to a systematic bias in GHG flux budgets. In our case, omitting chambers one to four at AWS would have led to an underestimation of annual CH₄ uptake of roughly 5 % and an overestimation of annual N₂O emissions of 56 % (both regarding annual budgets). Thus, all aspects of exposition and slope should be covered when assessing flux estimates of CH₄ and N₂O in sloping terrain.

5 Conclusions

Highest mean annual emissions of N₂O were observed at the intensively managed site, whereas highest uptake rates of CH₄ were measured at the extensively managed site. This clearly illustrates the impact that management intensity has on the magnitude of soil N₂O and CH₄ fluxes in grasslands. This clearly illustrates that management acts as a major co-

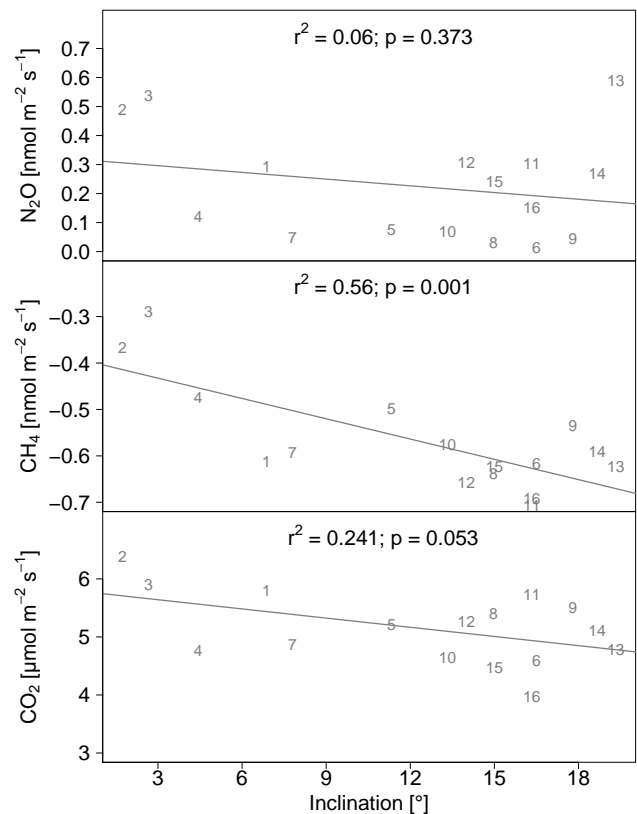


Fig. 9. Annually averaged chamber fluxes of N₂O, CH₄ and CO₂ at Alp Weissenstein (AWS) plotted against inclination at the respective chambers. The numbers indicate the chamber position along the transect. Regression coefficients as well as *p* values are given.

driver of CH₄ and N₂O fluxes in grasslands in addition to the commonly shown influences of environmental variables.

We identified the known set of drivers for fluxes of CO₂, CH₄ and N₂O fluxes (*T_a* and SWC for N₂O and CH₄, respectively). At the intensively managed site (CHA), LAI proved to be a good proxy for management influence on fluxes of all three GHGs.

Spatial variability, especially of soil CH₄ and N₂O fluxes was as high as expected. Permanent spots with lower CH₄ uptake coincided with smaller inclination of the terrain on which chambers were placed. Thus, on sloping terrain, mean chamber fluxes of CH₄ should be estimated from an ensemble that is (a) sufficient in size, (b) represents the common species composition including hot spots occurring due to grazing and (c) representative for the terrain of the site. This is important since SWC is one of the major environmental drivers of CH₄ exchange. For soil fluxes of N₂O, we suggest the use of portable chambers in conjunction with recently developed laser spectrometers allowing for much shorter sampling times and therefore sampling of additional hot spots occurring during grazing and hot moments after fertilization.

Acknowledgements. We acknowledge the help of all Grassland Sciences Group Members from ETH Zurich who participated in the intensive observation campaign. We also thank our technicians Peter Pluess and Thomas Baur for station maintenance and technical support. This study was partially funded by the European Union Seventh Framework Programme (FP7/2007–2013) under grant agreement no. 244122 (GHG-Europe). This study was further supported by COST Action ES0804 – Advancing the integrated monitoring of trace gas exchange Between Biosphere and Atmosphere (ABBA).

Edited by: K. Pilegaard

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