Aircraft based CH$_4$ flux estimates for validation of emissions from an agriculturally dominated area in Switzerland

Rebecca V. Hiller,$^{1,2,7}$ Bruno Neininger,$^3$ Dominik Brunner,$^2$

Christoph Gerbig,$^4$ Daniel Bretscher,$^5$ Thomas Künzle,$^6$

Nina Buchmann,$^1$ Werner Eugster$^1$
Corresponding author: Werner Eugster, Department of Environmental Systems Science, Institute of Agricultural Sciences, ETH Zurich, LFW C55.2, Universitaetsstrasse 2, CH–8092, Zurich, Switzerland. (eugsterw@ethz.ch)

1Institute of Agricultural Sciences, ETH Zurich, Zurich, Switzerland
2Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland
3MetAir AG, Airborne Observations, Airfield LSZN, Hausen am Albis, Switzerland
4Max Planck Institute for Biogeochemistry, Jena, Germany
5Agroscope Reckenholz-Tänikon Research Station ART, Zurich, Switzerland
6Meteotest, Bern, Switzerland
7Climate Services, Federal Office of Meteorology and Climatology (MeteoSwiss), Krähbühlstrasse 58, CH–8044 Zurich, Switzerland
Abstract. For regional-scale investigations of greenhouse gas budgets the spatially explicit information from local emission sources is needed, which then can be compared with flux measurements. Here we present the first validation of a section of a spatially explicit CH$_4$ emission inventory of Switzerland. The validation was done for the agriculturally dominated Reuss valley using measurements from a low-flying aircraft (50–500 m above ground level). We distributed national emission estimates to a grid with 500 m cell size using available geostatistical data. Validation flux measurements were obtained using the eddy covariance (EC) technique and the boundary-layer budgeting (BLB) approach that only uses the mean concentrations of the same aircraft transects. Inventory estimates for the flux footprint of the aircraft measurements were lowest (median 0.40 µg CH$_4$ m$^{-2}$ s$^{-1}$), and BLB fluxes were highest (1.02 µg CH$_4$ m$^{-2}$ s$^{-1}$) for the Reuss valley, with EC fluxes in-between (0.62 µg CH$_4$ m$^{-2}$ s$^{-1}$). Flux estimates from measurements and inventory are within the same order of magnitude, but measured fluxes were significantly larger than the inventory emission estimates. The differences are larger than the uncertainties associated with storage of manure, temperature dependence of emissions, diurnal cycle of enteric fermentation by cattle, and the limitations of the inventory that only covers ≥90% of all expected methane emissions. From
this we deduce that it is not unlikely that the Swiss CH$_4$ emission inventory estimates are too low.

**Keywords:** methane; spatially explicit inventory; regional flux; eddy covariance; boundary-layer budget
1. Introduction

Emission inventories are typically collected on a national basis and serve policy
makers to track greenhouse gas (GHG) emissions to their sources, to evaluate
the success and progress of emission reduction measures. Recently, the
credibility of these inventories was questioned because direct comparisons of
emission inventories against independent “top-down” estimates obtained from
atmospheric measurements are rare and sometimes disagree by a factor of two
or three [Nisbet and Weiss, 2010]. At the same time, non-CO$_2$ GHGs including
methane have gained increasing attention besides CO$_2$. This paper aims at
answering the question whether current CH$_4$ emission estimates can be
validated via field surveys with aircraft based flux measurements.

Methane is the second most important anthropogenic GHG after CO$_2$. At the
global scale, wetlands are thought to be the most important CH$_4$ source (30%),
followed by agriculture (rice cultivation 9% and ruminants 15%) [Denman
et al., 2007]. Under the absence of large wetland areas in Switzerland, the
agricultural sector becomes the dominant source of methane (79% of Swiss
emissions, mostly stemming from ruminants) whereas methane emissions from
wetlands and other natural sources are estimated to be small (<6%) and are not
included in the National Inventory Report [FOEN, 2012], which includes only
anthropogenic emissions. In order to compare the methane inventory to fluxes
calculated from measurements, agriculturally dominated regions are hence of
special interest. Biomass burning and wetlands are only very minor sources in Switzerland [see Hiller et al., 2014] and hence are not further addressed here.

Recent advancements in laser spectroscopy have brought instruments to the market that are also suitable for aircraft deployment. While numerous studies have been published on continuous airborne measurements of air pollutants and CO$_2$ in the planetary boundary layer (PBL) at the regional scale [e.g. Graber et al., 1998; Lehning et al., 1998; Barr et al., 1997; Desjardins et al., 1997, 1995; Mahrt et al., 1994; Gerbig et al., 2003], airborne observations of CH$_4$ are still rare. To the best of our knowledge, only three studies have addressed biosphere-atmosphere CH$_4$ fluxes using continuous airborne measurements. Ritter et al. [1992] presented eddy covariance CH$_4$ measurements from arctic Alaska, and the Canadian boreal forest and northern wetland regions [Ritter et al., 1994]. Mays et al. [2009] estimated the carbon footprint of Indianapolis with the help of a boundary-layer budget approach, intensively sampling the urban plume downwind of the city. Other studies, such as e.g. Wratt et al. [2001] who measured concentration profiles to estimate regional CH$_4$ emissions from agriculture, used grab sampling of air that was later analysed in the laboratory [see also Choularton et al., 1995; Pattey et al., 2006; Beswick et al., 1998; Kort et al., 2010]. All these airborne methane studies demonstrated the applicability of aircraft measurements to derive regional scale fluxes.
In this study, we present the first airborne CH$_4$ flux estimates for a valley dominated by agriculture that can be compared with a spatially explicit high-resolution CH$_4$ emission inventory. Fluxes were calculated with the eddy covariance method (EC) as well as with a boundary-layer budget (BLB) approach from a total of 58 flight legs on 16 days between June 2009 and late August 2010.

2. Methods

2.1. Measurement site and flight pattern

The Reuss Valley is situated in central Switzerland at the southern border of the Swiss Plateau (see Figure 1). Before the leveeing in the 19th century the Reuss river wended through the wide valley, and the plain was flooded on a regular basis. The leveeing has strongly reduced flooding and combined with drainage made the land suitable for agriculture [AGR, 1982]. The national soil suitability map classifies the area into suitable to very suitable for fodder production and suitable for crop production [GEOSTAT, 1980]. Today, 74% of our study area is used for agriculture and 18% is covered by forests and semi-natural areas [CORINE land cover, GEOSTAT, 1990], while the remainders are artificial surfaces (4%), wetlands (2%) and water bodies (2%). CORINE is the acronym of the “Coordination of information on the environment” project of the European Union (http://www.eea.europa.eu/publications/COR0-landcover).
Under fair weather conditions, the valley wind controls the local wind system. During the day, air moves towards the Alps (up-valley winds from NNW), whereas during the night, cold-air drainage flow prevails (down-valley winds from SSE). Flight legs, approximately 14 km long, were flown along the valley axis at constant flight levels (50 m to 500 m a.g.l.). During 16 flight days the flight pattern was repeated two to three times per day, namely in the late morning, around noon, and in the afternoon, to cover different times of day.

The aircraft measurements (Section 2.2) were complemented with ground-based energy flux measurements (not shown) and micrometeorological observations at the ETH research station Chamau (47° 12′ 37″ N, 8° 24′ 38″ E, 400 m a.s.l.), situated at the southern end of the flight legs. As a measure of cloudiness, a clear-sky fraction was introduced that represents the ratio between the measured incoming shortwave radiation ($SW_{\text{in}}$, at 2 m, CNR1, Kipp & Zonen B.V., Delft, The Netherlands) and the maximal expected incoming shortwave radiation calculated after Allen [1996]. More details on the ground-based measurements can be found in Zeeman et al. [2010].

### 2.2. Aircraft measurements

Aircraft measurements were performed on fair weather days in the warm season from June 2009 to late August 2010 (Table 1). We used a small research aircraft of the type DIMO HK36 TTC-ECO (Diamond aircraft, Wiener-Neustadt, Austria) that was equipped and operated by a private company...
(MetAir AG, Hausen, Switzerland). The instruments, listed in Table 2, were situated in the fuselage, in two underwing pods, and in the cockpit.

Meteorological variables including air temperature, atmospheric pressure, and 3D turbulence and trace gas concentrations of CO\textsubscript{2} and CO were measured continuously. The 3D wind and turbulence measurements were derived from the five-hole-probe and the IMU (Inertial Measurement Unit combining GPS and motion sensors to accurately record the movements of the aircraft). The wind is defined as the difference of the flow impinging on the sensor and the movement of the sensor in the earth fixed system). The absolute accuracy for the three components ($u$, $v$, $w$) $<0.5 \text{ m s}^{-1}$. The relative precision for the vertical component $w'$ at 10 Hz, which is relevant for the vertical turbulent fluxes, was on the order of 0.1 m s\textsuperscript{-1} (see Table 2 for more details).

### 2.2.1. Airborne fast methane analyzer

Additionally, CH\textsubscript{4} concentrations were measured with a fast methane analyzer (FMA, Los Gatos Research Inc., Mountain View, CA, USA) at 5 Hz. This is a commercially available integrated off-axis cavity output spectrometer which was modified to reduce weight and size to fit into one of the underwing pods (Fig. 2). The original case was replaced by an isolated aluminum case to minimize temperature fluctuations in the instrument during flight. The internal pump was replaced by an external pump to increase the flow rate (Vacuubrand MZ2C Vario SP, Vacuubrand GmbH + Co KG, Wertheim, Germany). The
pump was regulated to keep the cell pressure of the FMA in the automatically regulated range, irrespective of the varying atmospheric pressure [Schneider, 2009]. The instrument in the pod was connected to the pump in the cockpit through a 1/2” outer diameter teflon tube through the wing. The inlet outside the pod, a 33 cm long tube with 6 mm inner diameter (Synflex-1300, Eaton Performance Plastics, Cleveland, OH, USA) was followed by a particle filter and droplet separator (SMC, Japan, model AF20-F03 with 0.3 µm filter). The FMA is an instrument that employs direct-absorption-spectroscopy techniques to yield absolute gas mole-fraction measurements [Baer et al., 2002]. This means, that theoretically no calibration is necessary. In practice, however, measurements had to be corrected for spectroscopic water interferences [Hiller et al., 2012] based on the water vapor measurements of the LI-7500 (Li-Cor Inc., Lincoln, NB, USA) that was referenced to the dew point mirror (TP3, Meteolabor, Wetzikon, Switzerland). This correction typically increases CH₄ concentrations measured by the FMA by 1% [Tuzson et al., 2010]. Since our goal was to deploy this analyser in a lightweight aircraft with low payload, with the purpose to quantify fluxes (not primarily absolute concentrations), we used independent flask sampling (Section 2.2.3) to assure the quality of the measurements. For deployments in larger aircrafts, more sophisticated calibration procedures would be possible, as O’Shea et al. [2014] have shown.

2.2.2. Data acquisition
Data acquisition was done with two independent industrial compact computers and one standard laptop computer, one of which was equipped with a 10-channel counter and two 16-bit analog-to-digital converter boards controlled by the TurboLab software (MDZ Buehrer & Partner, Germany). Data from the fast methane analyser were transferred via a RS-232 serial data link to one of the computers. The addition of a CH$_4$ analyser was the largest difference compared to the configuration of the same aircraft as it was used during the investigation of the Eyjafjallajökull volcano erruption [Kristiansen et al., 2012].

2.2.3. Flasks samples

Complementary to the continuous measurements, 4–17 grab samples were filled into 1 liter glass flasks throughout each flight day. The air was later analyzed for concentrations of CH$_4$, CO, CO$_2$, N$_2$O and SF$_6$ at the Max Planck Institute for Biogeochemistry in Jena, Germany. For analysis of CH$_4$, CO$_2$ and N$_2$O, an Agilent 6890 gas chromatograph equipped with an electron capture detector (ECD), a CO$_2$ converter (“methanizer”) and a flame ionisation detector (FID) was used. SF$_6$ and CO were measured using a second Agilent 6890 gas chromatograph equipped with an electron capture detector (ECD) and a Trace Analytical Reduction Gas Analyser (RGA). All flask measurements are traceable to the respective WMO scale within the recommended compatibility levels [WMO, 2012]. The continuous CH$_4$ measurements were compared
against the flask concentrations. The comparison was based on the weighting
function proposed by Chen et al. [2012] using averages over the flask flushing
and filling period. Flasks that showed unusual pressure fluctuations during the
filling and flushing period (12 flasks) were excluded from the comparison as no
weighting function could be determined. Additionally, flasks for which the
difference to the averaged continuous measurements exceeded 25 ppb were
discarded (3 data points). These outliers were assumed to be caused by
problems either during flask sampling, storage, or analysis. The comparison
between the continuous CH$_4$ and 197 good quality flask measurements is
presented in Section 3.2.
2.2.4. Flight tracks

Flights were only carried out under fair weather conditions and the flight tracks were chosen along the main valley axis such that the transect flights could be considered representative for the yellow box shown in Figure 3. Along these transects, a linear increase in CH$_4$ concentrations in the direction of the mean wind speed is expected under ideal stationary conditions with homogeneously distributed CH$_4$ emission sources at the ground surface. In reality, this linear increase was quite prominently seen at all low-level transect flights up to 300 m agl (Figure 4).

2.3. Flux calculations

The high frequency measurements of CH$_4$ and the wind components allowed the determination of the CH$_4$ fluxes in the Reuss Valley by two different methods, (1) the EC method that considers vertical transport by turbulent eddies (Section 2.3.1) and (2) a simplified BLB approach (Section 2.3.2).

2.3.1. Eddy covariance approach

Fluxes for the individual 14 km flight legs were calculated with the EC method from the 5 Hz aircraft data of CH$_4$ and the vertical wind speed $w$. Prior to the flux calculation, trends in the CH$_4$ concentration and $w$ were subtracted by a running mean with a window size of 120 s that corresponds to a flight distance of $\approx 6$ km at the typical travel speed of 50 m s$^{-1}$. To determine the optimal averaging period for eddy covariance flux measurements, we used the ogive
method as recommended by Moncrieff et al. [2004]. The term ogive is used for the cumulative cospectrum of eddy covariance flux measurements. We used the original procedure by Desjardins et al. [1989], which indicated that a window size of 120 s ensured the inclusion of all relevant flux contributing wavelengths up to a flight height of 200 m a.g.l. Transects flown above 200 m a.g.l. were excluded from the analysis of the EC fluxes. In principle, a linear decrease in fluxes with height is expected across the PBL. In our case, no systematic dependence on flight level was seen for flight levels below 200 m, whereas legs flown at higher altitude were not very consistent with this theoretic assumption, and hence only flights below 200 m were further analysed. The actual flux \[ \mu g CH_4 m^{-2} s^{-1} \] was computed as

\[
F_{CH_4} = \frac{M_{CH_4}}{M_{air}} \cdot \rho_{air} \cdot \overline{w'c'} ,
\]

where \( w' [m s^{-1}] \) is the deviation from the running mean of the vertical wind speed and \( c' [\mu mol mol^{-1}] \) is the deviation from the running mean \( CH_4 \) concentration over one flight leg, and the overline indicates averaging over the respective flight leg. \( M_{CH_4} \) and \( M_{air} \) are the molar masses of methane and air, respectively, and \( \rho_{air} \) is the air density. To prevent smearing of data from before and after the transect into the running mean, the flight legs were cut by 3 km on each side for the calculation of the flux. Fluxes were only computed during times when the standard deviation of the wind direction was <50° to assertain quasi-stationary turbulence conditions. This subset of data is classified as
“good quality” data in what follows. To further narrow in the conditions that
correspond with along-valley wind conditions, we selected all good quality data
for which the mean wind direction was either 150–180° (down-valley winds),
or 220–360° (up-valley winds). Selection criteria were always applied for an
entire flight leg. Results are then reported for both “good quality data”
conditions and the subset of “good quality and along-valley wind conditions”
data.

Spectra and cospectra of CH4, CO2 and H2O concentrations and fluxes were
computed to check proper operation of the instruments and data acquisition
(Fig. 5). Spectroscopic corrections and corrections for density fluctuations
inside the FMA sample cell were applied to raw data, so that fluxes did not
require additional corrections. Due to the high variability of methane fluxes as
seen in the cospectra (Fig. 5), no robust correction for high-frequency losses
could be applied. This means that measured CH4 emissions reported here may
be slightly biased low.

2.3.2. Boundary-layer budget approach

Simple one-box models have been used in many urban air pollution studies to
estimate atmospheric trace gas or pollutant emissions from a known source as a
function of time [Arya, 1999; Oke, 1987; Hanna et al., 1982]. We employed
such a model to estimate the CH4 emissions from the Reuss Valley. The
relevant flows into and out of an imaginary box with along-wind length \(a\),
width $b$ and the actual height of the planetary boundary layer $h_{\text{PBL}}$ were quantified (see Figure 6). Exchange of air contained in this box with the air above the boundary layer (i.e., entrainment and detrainment) was considered to be negligibly small compared to the horizontal fluxes across the vertical walls of the box, and to the surface fluxes. The main axis of the box was oriented along the valley axis and hence followed the main flow in the valley during days with a pronounced valley wind system (up-valley during daytime, down-valley at night). This additional simplification allowed us to also neglect the flow through the sidewalls. Hence, the only considered walls of the box were the upwind and downwind walls and the surface area. The fluxes through the upwind ($F_{\text{in}}$) and downwind wall ($F_{\text{out}}$) were defined by the area $b \times h_{\text{PBL}}$ [m$^2$] times the mean wind speed $\bar{u}$ [m s$^{-1}$] and the mean concentration at the respective walls ($\bar{\rho}_{\text{CH}_4,\text{in}}, \bar{\rho}_{\text{CH}_4,\text{out}}$ [µg m$^{-3}$]). The height of the PBL was assumed to be constant along this relatively broad valley, but was varied with time of day: $h_{\text{PBL}}$ was determined via integration of the PBL growth rate computed from the sensible heat flux measured on the ground [Lyra et al., 1992] which then was scaled to the actual height obtained from aircraft profiles of CO, CO$_2$, CH$_4$, aerosol, H$_2$O, and temperature measured once a day. Because flights were only carried out during daytime with well-mixed conditions, the atmosphere was neutral to unstable in the PBL, sometimes with indications of residual layers in progress being incorporated in the growing
PBL. The step change of concentrations, temperature or humidity across the upper boundary of the PBL was used to determine actual PBL height.

While the measured mole fractions $c$ [ppm] were relatively constant with altitude, values in density units change. Air density almost linearly decreased with altitude in the lowest part of the atmosphere and hence the measured $\bar{\rho}_{\text{air}}$ at the different flight levels were linearly interpolated throughout the boundary layer. The air density at $0.5 \times h_{\text{PBL}}$ was used for the unit conversions of $c$ to $\rho_{\text{CH}_4}$. The mean wind speed did not show a clear height dependency and hence the average wind speed along the valley axis was used for the flux calculations.

The total flux across the surface area $F_{\text{source}}$ was defined by the box area ($a \times b$) times the source strength per unit area ($f$).

Assuming steady state (no accumulation of methane inside the box over time), mass conservation requires that the outflow equals the sum of the inflow and the total flux across the surface. Hence,

$$F_{\text{in}} + F_{\text{source}} = F_{\text{out}};$$

which is

$$b \ h_{\text{PBL}} \ \bar{u} \ \bar{\rho}_{\text{CH}_4,\text{in}} + a \ b \ f = b \ h_{\text{PBL}} \ \bar{u} \ \bar{\rho}_{\text{CH}_4,\text{out}} .$$

Solving for $f$, equation (3) yields

$$f = h_{\text{PBL}} \ \bar{u} \ \frac{\bar{\rho}_{\text{CH}_4,\text{out}} - \bar{\rho}_{\text{CH}_4,\text{in}}}{a} .$$
For homogeneously distributed constant sources, a linear concentration increase along the flight leg is expected (see Figure 6). Hence, $\frac{\bar{\rho}_{\text{CH}_4, \text{out}} - \bar{\rho}_{\text{CH}_4, \text{in}}}{a}$ is equal to the observed linear concentration increase along the transect $\left(\frac{d\bar{\rho}_{\text{CH}_4}}{dx}\right)$ and is replaced therewith in the flux calculations.

Total mass balance was tested to assess possible leaks in the model. Therefore, the air density at $0.5 \times h_{\text{PBL}}$ was determined for the upwind and downwind walls from a height-depending model, using the measurements of the first and last kilometer of the box. The air density change along the transect $\frac{\bar{\rho}_{\text{air, out}} - \bar{\rho}_{\text{air, in}}}{a}$ was multiplied by $\bar{u}$ and $h_{\text{PBL}}$ to determine the mass imbalance. The imbalance indicates a missed flux and hence, the calculated methane flux had to be adjusted by the methane flux introduced by the mass imbalance. This correction was however in all cases less than 10% of the measured (uncorrected) methane flux and hence will not be discussed in more detail.

As for the EC method, values were rejected when the standard deviation of the wind direction exceeded 50°. Additionally, a separate analysis was performed for the subset of fluxes from periods with wind directions within ±15° along the valley axis (i.e. valley wind conditions), the preferred conditions for the BLB method.

### 2.4. Spatially explicit CH₄ inventory

To compare our measurement-based CH₄ emission estimates with the emissions reported in the National Inventory Report [NIR; FOEN, 2012], CH₄
sources from the year 2007 were spatially distributed over Switzerland in a 500 m grid. This emission inventory was published by Hiller et al. [2014] and was made available in the PANGAEA repository via a digital object identifier (http://doi.pangaea.de/10.1594/PANGAEA.828262). Thus, we only give a short summary of the key figures of the emission inventory. Total anthropogenic CH$_4$ emissions were estimated at 180,000 t CH$_4$ yr$^{-1}$. The NIR lists about 620 different CH$_4$ sources.

Of the 620 CH$_4$ sources, the eight most important ones, contributing $\geq 90\%$ of all emissions, were quantified for each grid cell. These processes are (ranked by their importance): enteric fermentation of dairy cattle (43.5%) and of young cattle (16%), manure of dairy cattle (9.5%), landfills (6%), grid losses in gas distribution (5%), enteric fermentation of non-dairy cattle, namely suckler-cows (3.5%), manure of swine (3.5%), and enteric fermentation of sheep (2.5%).

Increases in total Swiss CH$_4$ emissions between 2007 (inventory) and the years of the measurements are +0.8% (2009) to +1.7% (2010) [FOEN, 2012].

For the creation of the inventory, geostatistical data were not always available at the high resolution required for the inventory. Hence, the available information had to be distributed to geographical areas with similar land use characteristics with the help of additional information (e.g. emissions from cattle were distributed over the land surface areas where cattle potentially can graze).
2.5. Footprint calculation

For every flight leg, the footprint of the turbulent flux was estimated with the Kljun et al. [2004] flux footprint model. This simple 2-d cross-wind integrating footprint model uses $\sigma_w$, $u_*$, height of flight $z_m$, $h_{PBL}$ and roughness length $z_0$ to predict the spatial extent of the upwind land surface area that controls EC flux measurements. The friction velocity $u_*$ and the standard deviation of the vertical wind speed $\sigma_w$ were calculated from the in-flight measurements for each flight leg, and $h_{PBL}$ was derived as described in Section 2.3.2. The surface roughness $z_0$ in the model was set to 0.08 m which is representative for farmland with many hedges [Stull, 1988, p. 380]. To calculate the corresponding flux from the emission inventory, all grid cells covered by the 10%–90% range of the crosswind-integrated footprint along the flight leg were averaged. Each grid cell’s emission was weighted according to its footprint contribution. The width of the footprint strongly depended on the flight height and the wind direction. Footprint widths for low flight heights started at 0.3 km and ranged up to 3.9 km at higher flight heights under conditions that yielded good data quality.
3. Results

3.1. Spatial distribution of Swiss methane emissions

Swiss methane emissions (Figure 1) are highest in the pre-alpine areas, the south-eastern part of the Swiss Plateau. The boundary towards the Alps is relatively sharp and only the agriculturally relevant valley floors of larger valleys show considerable methane emissions (pink and dark violet color in Figure 1). The Reuss Valley (black rectangle in Figure 1) belongs to the areas with relatively high methane emissions.

To assess the variability of CH$_4$ emissions seen by an aircraft traveling over our region of interest, the following experiment was performed: The mean flux within a rectangle covering the Reuss Valley was calculated from the emission inventory. Then, this rectangle was moved along a 60 km north-south transect in 1 km increments and along a 100 km west–east transect with the same increments (Figure 7). In both directions, the Reuss Valley coincides with the location with highest methane emissions confirming that the valley is a hot-spot of agricultural CH$_4$ emissions in Switzerland. Variations in north-south direction are greater than in west-east direction. This can be explained by more pronounced land use variations in combination with a higher share of built-up populated areas in the northern part of the investigation area.

3.2. Performance of the methane analyzer
The FMA showed excellent performance in comparison with the flask samples (Figure 8). A linear regression between the flask concentrations and the continuous measurements of the FMA resulted in $[\text{CH}_4]_{\text{flask}} = (-17 \pm 9) \text{ ppb} + (0.989 \pm 0.005) \cdot [\text{CH}_4]_{\text{FMA}}$ ($R^2 = 0.9952$). On average, the continuous measurements were $4.0 \pm 5.1$ ppb (mean $\pm$ SD) lower than the flask reference samples.

In order to investigate whether the FMA measurements were sensitive to environmental variables, we compared the differences between the continuous and flask CH$_4$ concentrations with relative humidity, specific humidity, atmospheric pressure, and air temperature. Temporal drifts and day-to-day variations in the performance of the FMA were investigated as well, but were small (data not shown). These variables together explain only 26% of the total variations and the remaining 74% can not be attributed to environmental variables or temporal trends of the FMA. Most likely, the vast share of total variance is primarily due to random errors in the continuous and flask measurements as well as uncertainties associated with the flask sampling procedure and the corresponding weighting function. Overall, the difference between FMA and flask sample concentrations is very small, indicating that the FMA is very well suited for airborne observations. For EC flux measurements such random variations are unimportant (covariances are robust against true random noise in each of the two variables involved), but must be kept in mind.
when using the data with the BLB method. Still, our experience shows that spatial and temporal variations are large enough to provide a good signal-to-noise ratio.

### 3.3. Methane fluxes

Flux estimates from 58 flight legs, representing 11 of the 16 measurement days in 2009 and 2010, were classified as good quality data (Figure 9). Because data were not normally distributed, the non-parametric Wilcoxon test was used to test for differences between approaches. EC fluxes for the Reuss Valley (median 0.62 µg CH$_4$ m$^{-2}$ s$^{-1}$ / mean 0.84 µg CH$_4$ m$^{-2}$ s$^{-1}$) are significantly higher than the inventory based flux estimates (0.40 µg CH$_4$ m$^{-2}$ s$^{-1}$ / 0.43 µg CH$_4$ m$^{-2}$ s$^{-1}$) (p<0.005). The BLB approach yields fluxes (1.02 µg CH$_4$ m$^{-2}$ s$^{-1}$ / 1.61 µg CH$_4$ m$^{-2}$ s$^{-1}$) that are significantly higher than both the inventory and the EC fluxes (p<0.05).

Restricting the data to conditions when the mean wind followed the valley axis within ±15° led to slightly higher values, but this increase was not significant (p>0.3). The variability of BLB fluxes increased to a SD of 3.15 µg CH$_4$ m$^{-2}$ s$^{-1}$, whereas this data selection only had a marginal effect on EC fluxes (SD 0.87 µg CH$_4$ m$^{-2}$ s$^{-1}$) (Figure 9, Table 3). The inventory based estimates varied only slightly with changing size and position of the footprints due to the widespread agricultural activity in the region (SD 0.13 µg CH$_4$ m$^{-2}$ s$^{-1}$).
4. Discussion

4.1. Uncertainty of the methane emission inventory

The uncertainty of the inventory emissions is of relevance to address the question whether measurements are statistically different from the inventory values. The uncertainty of the emission inventory was addressed in a separate publication by Hiller et al. [2014], who consider the combined effects of (i) spatial uncertainty of allocation of sources to grid cells, and (ii) temporal variability of emission sources. For the comparison with measurements, we add a third aspect, the uncertainty of the emission factors for enteric fermentation and manure management.

Uncertainty due to spatial allocation (see Section 3.1) was estimated to be approximately 5%. Uncertainty due to temporal variability was estimated to be on the order of ±6.4% [ART, 2008], assuming that the uncertainty in livestock census data reflects the seasonal variability of the number of cattle and their emissions.

More difficult was the assessment of the uncertainty of emission factors for enteric fermentation and manure management, since they involve both, (a) an aspect related to the feed composition and quality, which may be subject to both spatial and seasonal variation, and (b) the question of diel variations in ruminant activity, which may introduce a bias if average conditions represented by an inventory are compared to daytime measurements. In the present study, simulation of error propagation of the 95% confidence intervals of individual
sources as applied in the NIR [ART, 2008] in the footprint area of the aircraft fluxes was used to quantify the uncertainties in emission estimates associated with spatial allocation inaccuracies. This yielded a ±17% (95% confidence interval) uncertainty for the combination of enteric fermentation and management, which corresponds well with the IPCC [2000] default value of ±20%. The methane conversion rate for enteric fermentation (Ym) and the methane conversion factor for manure management (MCF) contribute to the overall uncertainty. Currently, the NIR applies the IPCC [2000] default conversion rates for Ym and MCF. Recent studies however indicate that these values are not fully appropriate for Switzerland [Zeitz et al., 2012], since Ym depends on the diet and the husbandry type. The Swiss cattle diet contains less feed concentrate than the IPCC assumes [Hiller et al., 2014]. Hence Ym is expected to be slightly higher in Switzerland, perhaps leading to higher emissions than currently reported. Emissions would increase by 10% if the new IPCC [2006] guidelines instead of IPCC [2000] were used. In contrast to Ym, Zeitz et al. [2012] found much lower than default values for MCF, especially in winter. They suggest that the effect of higher Ym and lower MCF should compensate each other in Switzerland [Zeitz et al., 2012]. Seasonal variation due to summer grazing are 0.01 µg CH₄ m⁻² s⁻¹ of the mean 0.43 µg CH₄ m⁻² s⁻¹ (Figure 9) and therefore can be neglected. We also neglected seasonal variations in emissions from manure storage, since Zeitz
et al. [2012] only found a very minor dependence of emissions on temperature. This aspect however remains controversial: the estimates based on suggestions by Mangino et al. [2001] recommended by the IPCC guidelines for National Greenhouse Gas Inventories result in a strong seasonality of manure storage with summer emissions that are three times the rates expected during winter at low temperatures. CH$_4$ emissions also depend on the daily rhythm of ruminants for which Kinsman et al. [1995] presented a diurnal cycle in CH$_4$ emissions from dairy cows. They showed that daytime emissions were about 20% higher than during nighttime. Since all flights were performed during the day, our observations might be biased towards higher than average emissions from ruminants.

In summary, the overall systematic uncertainty was estimated at $\pm$18.8%.

4.2. Validation of the regional CH$_4$ flux measurements

4.2.1. Eddy covariance method

The validation of inventory emissions via the EC method was more robust than the BLB approach and EC fluxes were less dependent on wind direction relative to the valley axis. Moreover, each transect represents an independent flux sample while for the BLB approach, all transects from one overflight were compiled into one single flux value. Hence, more good quality EC than BLB fluxes were obtained.

4.2.2. Boundary-layer budget approach
The BLB approach relies on many assumptions. The fluxes across the sidewalls and the lid of the box were assumed to be negligible and the boundary layer height to be constant. For the sidewalls this assumption only holds for along-valley winds. Hence, even weak crosswinds could have a considerable impact on the budget.

Uncertainty in the estimates of $h_{PBL}$ directly translates to uncertainty in BLB flux estimates. Fluxes calculated from morning overflights are more susceptible to errors in $h_{PBL}$ because the relative uncertainty in $h_{PBL}$ is larger when PBL is shallow. Even more problematic is the assumption of a constant $h_{PBL}$ along a given flight leg. A change in $h_{PBL}$ between the start and end of the leg by only 20 m, which is of the same order of magnitude as the change in orography along the valley, would change the estimated flux by as much as 65% for a typical boundary layer height of 1000 m. Moreover, a growing $h_{PBL}$ also involves entrainment of air from above the PBL. Especially in the morning, air with low CH$_4$ concentration is mixed from this residual layer down into the box where concentrations are still high due to nocturnal accumulation.

Consequently, the true flux is underestimated under such conditions. This should be less of a problem in the afternoon, when the CH$_4$ concentration within the boundary layer is more comparable to the background concentration.
5. Conclusions

To the best of our knowledge, this is the first attempt to directly compare a spatially explicit CH₄ inventory with regional scale flux measurements. We were able to show that aircraft based flux estimates provide a useful tool to determine CH₄ emission rates from an agriculturally dominated region. The differences between bottom-up (inventory) and top-down (EC and BLB) flux estimates are statistically significant and larger than the uncertainties associated with storage of manure, temperature dependence of emissions, diurnal cycle of enteric fermentation by cattle and the limitation of the inventory that only covers ≥90% of all expected methane emissions. From this we deduce that it is not unlikely that the CH₄ emission inventory estimates are too low. To increase our ability to validate fluxes at regional scale via aircraft measurements, not only improvements on the experimental side, but also an and improved representation of short-term variability in emission inventories will be needed which explicitly includes diel and seasonal variations in source strengths.

Acknowledgments. We thank the MetAir crew (Moritz Isler, Lorenz Müller, Dave Oldani, Boris Schneider and Yvonne Schwarz) for their great effort during the measurement campaigns, Silas Hobi and Elke Hodson for their contribution to the spatially explicit CH₄ inventory of Switzerland, Hans-Rudolf Wettstein and his team for their support at the ETH Research Station Chamau, and Susanne Burri for her invaluable comments on the
manuscript. This project was funded in part by the Maiolica project of the Competence Center Environment and Sustainability (CCES) of ETH. Data are available free of charge from the corresponding author.
References

AGR (Ed.) (1982), Sanierung der Reusstalebene: ein Partnerschaftswerk, AT
Verlag, Aarau, Aargauer Regierungsrat (AGR), 159 pp., ISBN
3-85502-050-7.

Allen, R. (1996), Assessing integrity of weather data for reference
 evapotranspiration estimation, Journal of Irrigation and Drainage
 Engineering, 122(2), 97–106.

ART (2008), Uncertainty in agricultural CH$_4$ and N$_2$O emissions of
 Switzerland, Internal documentation by Bretscher, D. and Leifeld, J.,
 agroscope Reckenholz-Tänikon Research Station, Zürich,

Arya, S. (1999), Air Pollution Meteorology and Dispersion, 310 pp., Oxford
 University Press, USA, 310 pp.

Baer, D., J. Paul, M. Gupta, and A. O’Keefe (2002), Sensitive absorption
 measurements in the near-infrared region using off-axis
 integrated-cavity-output spectroscopy, Applied Physics B: Lasers and Optics,
 75(2), 261–265.

 Comparison of regional surface fluxes from boundary-layer budgets and
 aircraft measurements above boreal forest, Journal of Geophysical Research,


FOEN (2012), Switzerland’s greenhouse gas inventory 1990–2012 – national inventory report 2012, Swiss Federal Office for the Environment (FOEN), Bern, Switzerland, 483 pp.,

GEOSTAT (1980), Soil aptitude maps, Digital data, Swiss Statistical Office (BfS), Neuchâtel, Switzerland,

GEOSTAT (1990), Corine landuse map, Digital data, Swiss Statistical Office (BfS), Neuchâtel, Switzerland,

GEOSTAT (1992/97), Land use statistics, Digital data, Swiss Statistical Office (BfS), Neuchâtel, Switzerland,


IPCC (2000), Good practice guidance and uncertainty management in national greenhouse gas inventories (IPCC GPG), Intergovernmental Panel on Climate Change.


Figures and tables

Figure 1. High resolution (500 m × 500 m) CH₄ emission inventory for Switzerland for the year 2007. Over 90% of the total anthropogenic emissions, including the eight most important sources from the categories agriculture, landfills, and gas distribution as well as emissions from wetlands and lakes are included. The black rectangle locates the Reuss Valley where this inventory was cross-validated with direct regional-scale flux measurements from a small research aircraft. The black boundaries indicate the summarized biogeographical regions of Switzerland.
**Figure 2.** Modified fast methane analyzer mounted into left wingpod on aircraft.
Figure 3. Flight tracks (red lines) from 7 April 2010 along the Reuss Valley. The yellow box indicates the box used in the boundary-layer budget approach. The view is towards North-West. Base map: ©2012 Google Earth, ©2012 Geo Content, ©2012 TerraMetrics, ©Cnes/Spot.
Figure 4. CH$_4$ concentration increase along the transect for different flight heights.

The CH$_4$ concentration of an air parcel increases relatively linear as it travels along the Reuss Valley. The data show an example of 1 Hz averages obtained during an afternoon flight on 4 June 2010. Arrows in the legend show the direction of flight with tailwind (300 m, 200 m a.g.l.) and headwind (100 m a.g.l.).
Figure 5. Example composite cospectra of CO$_2$, CH$_4$, and H$_2$O fluxes (thick lines, filtered with a Gaussian running average) from six afternoon flight legs from 24 June, 6 July, and 8 September 2009. Thin lines show idealized cospectra without damping (according to Kaimal et al. [1972]; solid lines) and with high-frequency damping (Eugster and Senn [1995]; broken lines) for comparison. A clear effect of high-frequency damping loss is seen for all fluxes with damping constants of 0.1 s$^{-1}$ for CO$_2$ and H$_2$O fluxes, and 0.3 s$^{-1}$ for CH$_4$ cospectra. The gray band shows the inter-quartile range of bandwidth-averaged individual cospectra.
Figure 6. Scheme of the simplified box model. The imaginary box with a length $a$, width $b$, and height $h_{PBL}$ encloses the air volume of interest. The gray arrows indicate the methane transport in and out of the box that is forced by the wind speed $\bar{u}$ (black arrow) along the main box axis. The small arrows at the surface of the box depict a homogeneous CH$_4$ source. As the air travels over the source, the CH$_4$ concentration increases gradually. The resulting idealized linear CH$_4$ gradient is indicated by the white-black dashed line.
Figure 7. Spatially explicit emission inventory, shown as an overlay over a geographic map, with the study area as a black box in its center. This box was moved south-north and west-east along the indicated lines to simulate regional flux changes. The panels to the left and below the map show area plots of the emissions averaged along the north-south direction and west-east direction, respectively. The emissions are aggregated into the categories agriculture, landfills, gas distribution, wetlands and lakes. The boxplots at the top and the right of the two panels summarize the total emissions by indicating the median (solid line), the interquartile range (box), and the lowest and highest observed values (whiskers). Map units are Swiss coordinates in meters. Base map reproduced with the authorization of swisstopo (JA100120).
Figure 8. Scatterplot of flask CH$_4$ concentrations and weight averaged continuous measurements of the Fast Methane Analyzer (FMA). The grey area represents the 95% confidence interval and the solid line is the linear regression fit. The dashed line indicates the 1:1 relationship. The inset shows the histogram of the differences between in-situ and flask CH$_4$ measurements.
Figure 9. Probability density functions (PDF) of good quality data (dashed lines) for averaged inventory emissions within the footprint of the eddy covariance fluxes (left), the fluxes calculated by the eddy covariance method (EC, center), and the boundary-layer budget approach (BLB, right). In addition, periods when wind directions followed the valley axis ±15° were analyzed separately (solid line). Mean and median values of the respective PDF are given for “Good quality” data followed by the “Valley wind” data (in italics). For the EC fluxes, a 120 s moving average was used. BLB fluxes corrected for mass balance closure are given together with values before correction in parentheses. BLB fluxes were significantly higher than EC fluxes, and both were significantly greater than inventory estimates (p<0.05). CH₄ emissions (positive fluxes), namely from ruminants, dominate.
<table>
<thead>
<tr>
<th>Date</th>
<th>Weather situation</th>
<th>CSF [%]</th>
<th>$T_{\text{air}}$ [°C]</th>
<th>$T_{\text{soil}}$ [%]</th>
<th>SWC [%]</th>
<th>RH [%]</th>
<th>$u$ [m s$^{-1}$]</th>
<th>$h_{PBL}$ [m a.g.l.]</th>
</tr>
</thead>
<tbody>
<tr>
<td>24 June 2009</td>
<td>Flat pressure pattern</td>
<td>79</td>
<td>19</td>
<td>15.4</td>
<td>96</td>
<td>69</td>
<td>2.4</td>
<td>1130</td>
</tr>
<tr>
<td>02 July 2009</td>
<td>Flat pressure pattern</td>
<td>91</td>
<td>29</td>
<td>18.6</td>
<td>96</td>
<td>50</td>
<td>4.3</td>
<td>900</td>
</tr>
<tr>
<td>06 July 2009</td>
<td>Low pressure moves to Scandinavia, and direct humid-warm air towards</td>
<td>58</td>
<td>23</td>
<td>14.6</td>
<td>96</td>
<td>55</td>
<td>3.7</td>
<td>510</td>
</tr>
<tr>
<td>13 July 2009</td>
<td>High pressure</td>
<td>64</td>
<td>26</td>
<td>17.3</td>
<td>96</td>
<td>3.4</td>
<td>1.8</td>
<td>510</td>
</tr>
<tr>
<td>07 September 2009</td>
<td>Ridge of high pressure from Spain</td>
<td>98</td>
<td>20</td>
<td>15.3</td>
<td>314</td>
<td>3.5</td>
<td>0.1</td>
<td>680</td>
</tr>
<tr>
<td>18 March 2010</td>
<td>High pressure</td>
<td>90</td>
<td>13</td>
<td>4.3</td>
<td>273</td>
<td>44</td>
<td>11</td>
<td>910</td>
</tr>
<tr>
<td>06 April 2010</td>
<td>High pressure over Scandinavia, weak front over France</td>
<td>100</td>
<td>16</td>
<td>8.2</td>
<td>281</td>
<td>45</td>
<td>2.8</td>
<td>900</td>
</tr>
<tr>
<td>01 June 2010</td>
<td>Humid air from N</td>
<td>51</td>
<td>15</td>
<td>14.6</td>
<td>6</td>
<td>42</td>
<td>1.0</td>
<td>680</td>
</tr>
<tr>
<td>04 June 2010</td>
<td>High pressure over the North Sea</td>
<td>96</td>
<td>21</td>
<td>14.4</td>
<td>42</td>
<td>55</td>
<td>2.5</td>
<td>200</td>
</tr>
<tr>
<td>19 August 2010</td>
<td>High pressure development</td>
<td>62</td>
<td>20</td>
<td>17.8</td>
<td>41</td>
<td>32</td>
<td>1.8</td>
<td>510</td>
</tr>
<tr>
<td>20 August 2010</td>
<td>Flat pressure pattern</td>
<td>79</td>
<td>24</td>
<td>18.2</td>
<td>41</td>
<td>70</td>
<td>2.1</td>
<td>710</td>
</tr>
<tr>
<td>26 August 2010</td>
<td>Ridge of high pressure from Spain</td>
<td>96</td>
<td>27</td>
<td>20.0</td>
<td>41</td>
<td>72</td>
<td>2.1</td>
<td>710</td>
</tr>
</tbody>
</table>

Table 1: General weather situation compiled from Meteoswiss and meteoexwirfe (2009, 2010) and observed meteorological variables at the Chamau field station. The simulated boundary layer height ($h_{PBL}$) is reported for 15:00 CET.

When the flight measurements were performed, the simulated boundary layer height ($h_{PBL}$) is reported for 15:00 CET.

The variables are averaged for the period 10:00–17:00 CET, since ($T_{\text{soil}}$) and soil water content (SWC) at 0.15 m in depth. The variables are averaged for the period 10:00–17:00 CET. Relative humidity ($RH$), wind direction ($W_{\text{dir}}$), and wind speed ($u$) were measured at 2 m above ground, soil temperature ($T_{\text{soil}}$) and soil water content (SWC) at 0.15 m in depth. The variables are averaged for the period 10:00–17:00 CET. The simulated boundary layer height ($h_{PBL}$) is reported for 15:00 CET.
Table 2. Instruments operated on the aircraft, variables measured, resolution of data acquisition, and estimated precision of measurements (modified after Neininger et al. [2001] and http://www.metair.ch/).

<table>
<thead>
<tr>
<th>Variable</th>
<th>Instrument / Method</th>
<th>Resolution</th>
<th>Precision</th>
</tr>
</thead>
<tbody>
<tr>
<td>Position</td>
<td>GPS TANS Vector</td>
<td>1 m</td>
<td>0.1 s</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1 s</td>
<td>5...20 m</td>
</tr>
<tr>
<td>Ground speed</td>
<td>GPS TANS Vector</td>
<td>0.1 m s$^{-1}$</td>
<td>1 s</td>
</tr>
<tr>
<td>Attitude (azimuth, pitch, roll)</td>
<td>GPS TANS Vector</td>
<td>0.1$^°$</td>
<td>1 s</td>
</tr>
<tr>
<td>Acceleration 3-d</td>
<td>Kistler/DLR</td>
<td>0.01 m s$^{-2}$</td>
<td>1 s</td>
</tr>
<tr>
<td>Air temperature</td>
<td>Thermocouple (Meteolabor)</td>
<td>0.1$^°$C</td>
<td>1 s</td>
</tr>
<tr>
<td>Dewpoint</td>
<td>Dewpoint mirror (Meteolabor)</td>
<td>0.1$^°$C</td>
<td>1 s</td>
</tr>
<tr>
<td>Flow angles</td>
<td>Five-hole probe using Keller capacitive sensors</td>
<td>0.1$^°$</td>
<td>1 s</td>
</tr>
<tr>
<td>Wind vector 3-d</td>
<td>Post flight processing</td>
<td>0.5 m s$^{-1}$</td>
<td>1 s</td>
</tr>
<tr>
<td>Aerosols (&lt;0.3 and &lt;0.5 mm)</td>
<td>MetOne laser particle counter</td>
<td>0.02 cm$^{-3}$</td>
<td>1 s</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>Licor Li6262 and Li7500$^a$</td>
<td>0.05 ppm</td>
<td>1 s</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>Licor Li6262 and Li7500$^a$</td>
<td>0.01 g kg$^{-1}$</td>
<td>1 s</td>
</tr>
<tr>
<td>CH$_4$</td>
<td>Los Gatos Research DLT-100$^b$</td>
<td>0.1 ppb</td>
<td>0.2 s</td>
</tr>
<tr>
<td>NO$_2$, NO$_x$, NO$_y$, HNO$_3$, PAN, O$_x$</td>
<td>NOxTOy 6-channel instrument$^d$</td>
<td>0.1 ppb</td>
<td>1 s</td>
</tr>
<tr>
<td>CO</td>
<td>Aerolaser AL-5003 fast vacuum UV fluorescence</td>
<td>0.5 ppb</td>
<td>0.2 s</td>
</tr>
<tr>
<td>O$_3$</td>
<td>single cell UV photometer$^e$</td>
<td>0.5 ppb</td>
<td>10 s</td>
</tr>
<tr>
<td>CO$_2$, CO, CH$_4$, N$_2$O, H$_2$, SF$_6$, $\delta^{18}$O, $\delta^{13}$C</td>
<td>grab samples in glass flasks</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td></td>
<td>analyzed at MPI Jena</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

$^a$ both modified and combined to achieve the short-term precision specified here with frequent calibration where the slower Li6262 provides the baseline concentration and the Li7500 the turbulent fluctuations

$^b$ modified by ETH Zurich to reduce size and weight, and by MetAir to improve cell pressure stability during aircraft operation

$^c$ determined from standard deviation of continuous measurements in this study, see Section 3.2

$^d$ built at PSI, based on a Monitorlabs instrument

$^e$ instrument using Luminol chemoluminescence and chemical converters, developed and built by Paul Scherrer Institute (PSI) and Metair
Table 3. Best estimates from methane emission inventory, aircraft-derived eddy covariance (EC) and boundary-layer budget (BLB) fluxes obtained from 58 flight legs of which 25 (or 43%) were carried out when the mean wind reflected along-valley flow conditions.

<table>
<thead>
<tr>
<th>Methane Flux Estimate</th>
<th>95% Confidence</th>
<th>Mean</th>
<th>Median</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Good quality data</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Inventory</td>
<td>0.28 ... 0.62</td>
<td>0.43</td>
<td>0.40</td>
<td>µg CH$_4$ m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>EC</td>
<td>-0.5 ... 2.89</td>
<td>0.84</td>
<td>0.98</td>
<td>µg CH$_4$ m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>BLB</td>
<td>-1.57 ... 8.14</td>
<td>1.61</td>
<td>2.47</td>
<td>µg CH$_4$ m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td><strong>thereof: with valley wind conditions</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Inventory</td>
<td>0.26 ... 0.92</td>
<td>0.46</td>
<td>0.46</td>
<td>µg CH$_4$ m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>EC</td>
<td>-0.28 ... 4.04</td>
<td>0.62</td>
<td>0.82</td>
<td>µg CH$_4$ m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>BLB</td>
<td>-2.81 ... 9.26</td>
<td>1.02</td>
<td>2.81</td>
<td>µg CH$_4$ m$^{-2}$ s$^{-1}$</td>
</tr>
</tbody>
</table>