CH_4 release experiment

1. Materials and Methods

A methane (CH_4) release experiment was carried out between June 25th and 29th in 2014 at an experimental site in Posieux, Switzerland, using the measurement setup of Felber et al. [1]. The identical source grid as in the Witzwil release experiment in the present paper was used. The release grid was moved during the experiment from position NE to SW due to changes in the wind direction (WD). Pure bottled CH_4 (5.0, CarbaGas, Switzerland) was released at a constant rate of 10 L_n min^{-1}.

Two different CH_4 analyzers have been used. An Aerodyne Quantum Cascade laser (QCL) device and a Los Gatos Cavity Ring down (FGGA) device (details can be found in Felber et al. [1]). The QCL inlet was located approximately 20 m away from the grid centre, at a height of 2 m above ground level (a.g.l.). The FGGA was installed approximately 3 m downwind of the grid edge at the heights of 0.95 m a.g.l. (NE) and 0.98 m a.g.l. (SW), respectively. Two 3D ultrasonic anemometers were located 5 cm below each concentration measurement inlet and served to characterise 10 min averaged model input parameters for dispersion modelling.

Although the experimental site is located near agricultural facilities with livestock production, the investigated wind sectors did not have any upwind CH_4 sources (animals or barns) close by. Based on the 10 min intervals, the background concentration $\bar{c}_{bgd}$ was assumed to be the level of the lowest 2 % (2nd percentile) of the 10 Hz CH_4 time series for each instrument, evaluated according to WD when no CH_4 sources were active.

Emissions were calculated using the freely available bLS model WindTrax (WT-bLS) version 2.0.8.8. (http://www.thunderbeachscientific.com/) for cases when the mean WD was within defined wind sectors (Figure 1). Estimated fluxes were not subjected to extra quality selection.

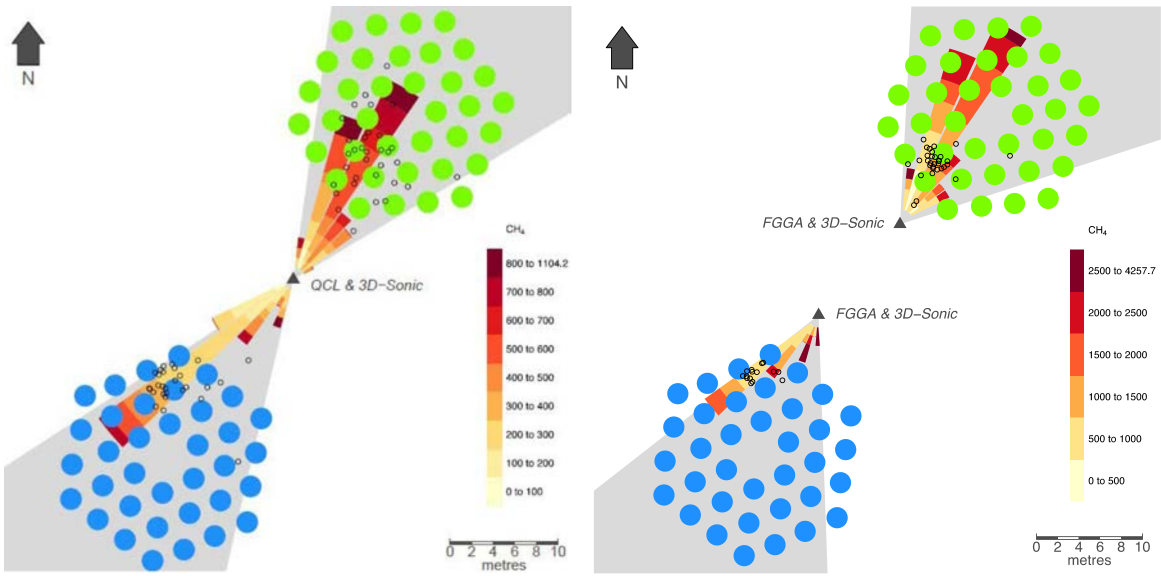

Figure 1. Overview on the gas release experiments. The release grid was moved from NE to SW due to changes in WD; coloured circles represent individual orifices with their circular representation for footprint modelling. grey: WD selection sector; black circles: (vertical flux) footprint function maxima using the Kormann-Meixner algorithm [2] for individual measurement intervals; concentration rose: $\Delta\bar{c}$ for CH_4 in ppb). Figure obtained from Häni et al. [3].

2. Results

During the release experiment, 61 to 79 valid 10-minute intervals were available due to wind sector and footprint model constraints. Figure 1 illustrates the distribution of the average CH_4 concentration increase above background $\Delta\bar{c} = \bar{c} - \bar{c}_{bgd}$, which ranged from 0.1 to 1.2 ppm at position QCL and from 0.5 to 5 ppm at position FGGA, reflecting the varying dispersion regimes and inlet positions within the emission plume. Estimated recovered fractions of CH_4 using WT-bLS were variable and differed slightly between QCL and FGGA (Figure 2). The average recoveries from FGGA and QCL were very similar and reasonably accurate ranging between 0.96 to 1.04 (Table 1). Given the uncertainty in the release rate as well as in $\bar{c}$, it is concluded that these recovered fractions are not significantly different from 1. The atmospheric stability was ranging from very unstable over near neutral to stable conditions. Although stratification of the data by L shows no systematic differences in the average recovery rates, Figure 2 suggests that the recovery deteriorates with systematically lower recoveries towards very stable conditions (i.e. towards larger values of $(z-d)/L$) coinciding with low values of u_*.

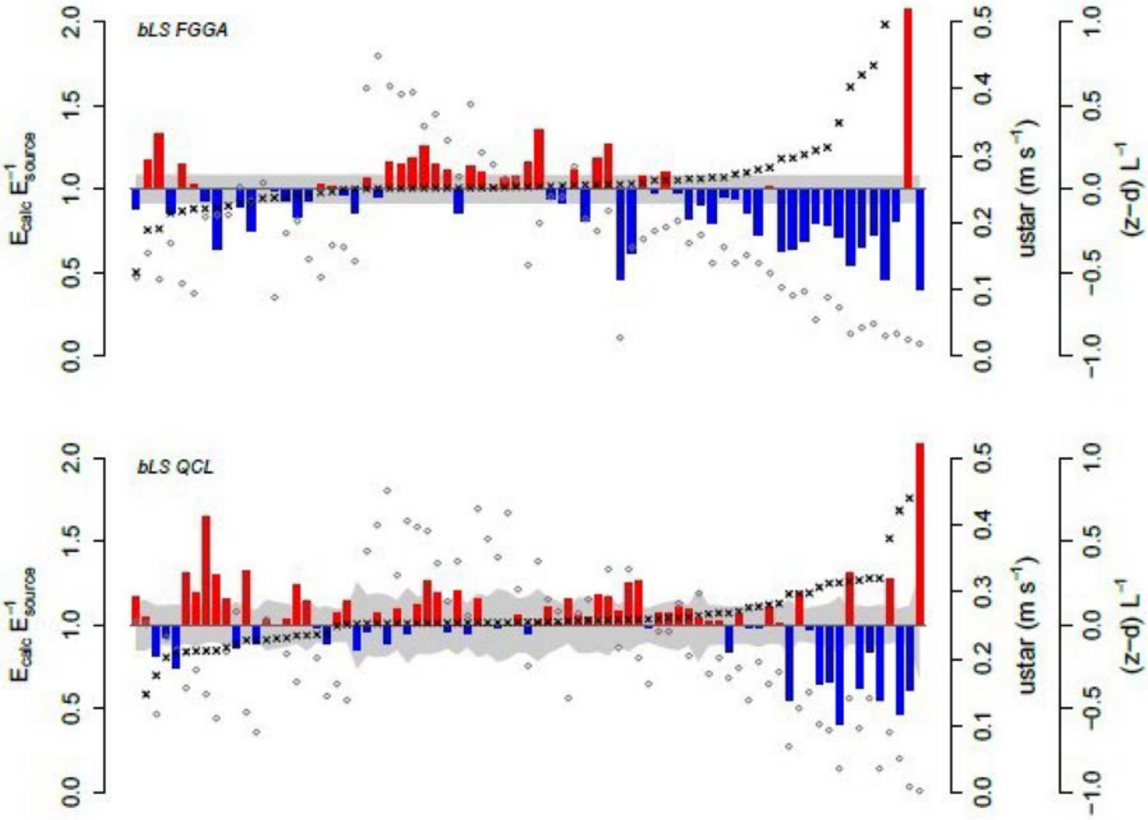

Figure 2. Recovery rates (E_{calc}/E_{Source}) as estimated by the two instruments FGGA (top) and QCL (bottom). The bars indicate deviations of the calculated flux from unity recovery (red = upward, blue = downward). The results are sorted from left to right by increasing stability of the atmosphere. Black dots: stability measure given as the aerodynamic measurement height (z-d) divided by the Obukhov-Length (L). Grey dots: friction velocity u_* (ustar). The shaded area illustrates the estimated effect of the uncertainty of the gas release rate itself, combined with the uncertainty of $\bar{c}$, mainly determined by the uncertainty in $\bar{c}_{bgd}$. Figure obtained from Häni et al. [3].

Table 1. Median CH_4 recoveries with median absolute deviation in parenthesis. Italic: number of 10-minute estimates.

Instrument	All Intervals	L > 0	L < 0
QCL	1.04 (0.16) *79*	1.03 (0.12) *58*	1.05 (0.21) *21*
FGGA	0.96 (0.21) *69*	0.95 (0.24) *50*	0.96 (0.10) *19*

References

1. Felber, R.; Münger, A.; Neftel, A.; Ammann, C. Eddy covariance methane flux measurements over a grazed pasture: effect of cows as moving point sources. *Biogeosciences Discuss.* **2015**, *12*, 3419–3468, doi:10.5194/bgd-12-3419-2015.

2. Kormann, R.; Meixner, F.X. An analytical footprint model for non-neutral stratification. *Boundary-Layer Meteorology* **2001**, *99*, 207–224.
3. Häni, C.; Sintermann, J.; Kupper, T.; Jocher, M.; Neftel, A. *Ammoniak-Emissionen nach Ausbringung von Gülle.*: CH-3052 Zollikofen, 2016. https://www.agrammon.ch/assets/Downloads/SchlussberichtInklAnh20160728subm.pdf (accessed on 3 May 2017).