Answers to referees on "Studying boundary layer methane isotopy and vertical mixing processes at a rewetted peatland site by unmanned aircraft system"

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1 Answers to referees

The authors would like to thank the anonymous referee for the comments on the manuscript. In the following, the comments are given in *italic*. The answers are given in normal letters. The modified text in the manuscript is given in quotation marks.

2 Referee 1

The manuscript presents a quadrocopter equipped with a flask sampler in a proof of concept study looking at delta 13C in methane above a peatland on two mornings. Using unmanned aircraft for sampling in the lower atmosphere is not new. Comparable approaches are referred to in the introduction. The novelty here is perhaps the target parameter: delta 13C in methane.

We changed the beginning of the abstract to "The combination of two well-established methods, of quadrocopter-borne air sampling and of methane isotopic analyses, is applied to determine the origin of methane at different altitudes and to study mixing processes. A proof of concept study was performed to demonstrate the capabilities of quadrocopter air sampling for subsequently analysing the methane isotopic composition δ^{13}C in the laboratory. The advantage of the system compared to classical sampling at ground and at tall towers is the flexibility concerning sampling location, and in particular the flexible choice of sampling altitude, allowing to study layering and mixing of air masses with potentially different origin of methane."

Precision of the isotope measurement in the laboratory is indicated to be about 0.5 permill (page 5, line 28). Could this indication be more precise? Are the 0.5 permill one sigma for the same, repeatedly measured sample?

We changed the text to: "Precision, determined as the maximum difference of delta values during repeated analysis of ambient Bremerhaven air samples taken simultaneously and analysed consecutively many times per year, is better than 0.5 ‰. "

Considering the difference in isotopic signal between nocturnal boundary layer and the air above during the first flight of a day is only about 1 permill, the isotopic signal of the polder is barely significant.

We agree that the isotopic signature of the air surrounding the polder is similar. This can be expected as the rewetted area is larger than the polder. Further, the methane isotopic signature from ruminants is similar, as summarised by Röckmann et al. (2016). We added in the abstract: "The systematically more negative delta values occurred only as long as the nocturnal temperature inversion was present."

Its size may be limited by three factors. First, the difference may be small in isotopic signature between the polder and other, presumably also biogenic sources in the larger surroundings.

We agree with this point, and changed the text in the discussion to "The difference in delta values obtained for air samples near ground and above the temperature inversion during stable stratification is around 1.5 ‰, thus significantly higher than the uncertainties (flight 1 for each measurement day). This shows that the observed systematic differences are real measurement features, not measurement uncertainty. Other methane sources in the surroundings of the polder are presumably of biological origin as well, they may include larger areas of the rewetted peatland and ruminants, with similar isotopic composition (Röckmann et al., 2016)."

Second, the polder area may be too small to substantially alter atmospheric methane isotopic composition in the lower tens of metres, especially at wind speeds of several metres per second.

During the night, when the temperature inversion inhibits mixing and the locally emitted methane is trapped, the wind speed was below $2\,m\,s^{-1}$. Therefore, the effect of a different isotopic composition below the temperature inversion can be traced back to local emissions. We changed the discussion text to: "Under stable atmospheric conditions and low wind speed smaller than $2\,m\,s^{-1}$ during the night and in the early morning hours, the near-surface methane concentration is enhanced, as no vertical mixing is possible, in agreement with Emeis (2008); Brosy et al. (2017)."

A simple box model would suffice for an initial estimate.

We do not agree that a box model is helpful for this case. As underlined by the very different methane concentrations in the water, we assume local inhomogeneities of sources. On the contrary, a suitable method for qunatifying small and inhomogeneous sources is urgently needed. We changed the text in the conclusion section to: "The differences in delta values of water and air, the differences in delta values between both flight days and the development during each day emphasize the highly complex and inhomogeneous nature of methane processes on horizontal scales below 1 km in sediments, at the sediment-water and the water-atmosphere interface. Therefore, a suitable method is required for quantifying small-scale inhomogeneous methane sources."

Third, the flights were performed well after sunrise. Much of the methane trapped near the surface during the night had already been mixed to greater altitude, as can be seen in the methane concentration in Figure 9.

We agree with this point, the methane concentration was higher during the night, as indicated in Fig. 9. We changed the text in the discussion to: "Under stable atmospheric conditions and low wind speed smaller than $2\,\mathrm{m\,s^{-1}}$ during the night and in the early morning hours, the near-surface methane concentration is enhanced, as no vertical mixing is possible, in agreement with Emeis (2008); Brosy et al. (2017). This can be seen in the time series of the methane concentrations for both days as well (Figs. 9 and 10). During the flights, the methane concentration was already smaller again, which can be explained by vertical mixing up to the temperature inversion of around 100 m. This dilution also influences the isotopic composition."

The data in Figures 9 and 10 seems to include erroneous measurements (e.g. zero methane around 18:00 on 5 September and a large number of spikes going above 0.3 mmol/m3 on both days).

Originally, we used only the data set of the LICOR 7700. Now we corrected the data with the quality controlled closed path data set of the Los Gatos sensor. We further included the time series in the figure of the diurnal cycle of the meteorological parameters.

Page 9, last paragraph: "The hypothesis ..." is not plausible at all. Horizontal gradients in air decrease with altitude because of higher windspeeds and more efficient mixing with increasing height above ground. Any remaining gradient will certainly not be large enough to cause measurable differences in samples taken simultaneously "only 13 cm apart". The sampling container is filled "within less than 2 s" (page 4, line 32). A horizontal wind speed of 2 m/s already results in a sample integrating air over a few metres in the horizontal direction.

We added in the text information about the sample integration:

"Assuming that in the worst case the sampling takes place within the downwash of the rotor blades of not more than $-19\,\mathrm{m\,s^{-1}}$, the sampling time of 1.3 s duration results in a vertical resolution of around 25 m. Sampling during descent with a speed of $-2.5\,\mathrm{m\,s^{-1}}$ adds an uncertainty in the altitude of 5 m. Altogether, the sampling is influenced by air in a height interval of 30 m. This is the sufficient for the sampling intervals of around 100 m, and for determining that the sampling was done below or above the temperature inversion."

Further, we added in the discussion: "In the presented case, the parallel samples obtained at 10 m altitude seem to be mostly of the same origin with small differences in the delta value. Above, differences in the delta values are higher. A wind speed of less than $2\,\mathrm{m\,s^{-1}}$ during the sampling results in horizontal sample integration over not more than 4 m. Small-scale differences in methane isotopic composition can be introduced by mixing processes, and may be reinforced by mixing induced by the quadrocopter system. A high variability of methane sources is in agreement with the highly variable methane water concentrations measured within a radius of 100 m on 5 September 2018." A visualisation of such small-scale mixing processes is shown in Figs. 5 and 6. In this case, colour was released artificially near ground (inhomogeneous sources). The homogeneous layer building up due to stable stratification was disturbed by turbulence, in this case induced by the aircraft, and large horizontal gradients are visible as a result.

In addition, the turbulence caused by the rotors will add several metres across which the sample integrates in the vertical dimension.

We included three more co-authors who have done numerical simulations, and added a section about simulations of the flow induced by a propeller blade:

"In order to quantify the effect of the vertical flow induced by the quadrocpter, numerical simulations were performed with the software ANSYS CFX. The simulations were transient in nature using a Reynolds-Averaged Navier Stokes (RANS) approach with the Shear Stress Transport (SST) turbulence model (Menter, 1994). A simplified model of the propeller blade was used, with a multidomain approach: The blade is enclosed in a rotating domain, surrounded by a static domain. Simulations were performed for hover with a propeller rotation speed of 3167 min^{-1}, for vertical climb at a speed of 6.5 $m\,s^{-1}$ with a rotation speed of 3913 $^{-1}$ and of vertical descent at a speed of -2.5 $m\,s^{-1}$ with a rotation speed of 2880 min^{-1}. An ambient temperature of 0° C and pressure of 1023 hPa were considered. Contours of relative vertical velocity show a core region of positive relative velocity directly below the center of the blade, and a negative relative velocity up to 19 $m\,s^{-1}$ below the blade for a distance exceeding 0.75 m (Fig. 2). Additionally, zones of recirculation can be seen around the tips of the propeller, especially for the descent case. The air sampling system is contained in the middle of the copter, and is less affected by artificial turbulence than the areas below the rotor blades.

Assuming that in the worst case the sampling takes place within the downwash of the rotor blades of not more than -19 $m\,s^{-1}$, the sampling time of 1.3 s duration results in a vertical resolution of around 25 m. Sampling during descent with a speed of -2.5 $m\,s^{-1}$ adds an uncertainty in the altitude of 5 m. Altogether, the sampling is influenced by air in a height interval of 30 m. This is the sufficient for the sampling intervals of around 100 m, and for determining that the sampling was done below or above the temperature inversion."

Further, we added in the discussion section: "In the presented case, the parallel samples obtained at 10 m altitude seem to be mostly of the same origin with small differences in the delta value. Above, differences in the delta values are higher. A wind speed of less than 2 $m\,s^{-1}$ during the sampling results in horizontal sample integration over not more than 4 m at the sampling altitude. Small-scale differences in methane isotopic composition can be introduced by mixing processes, and may be reinforced by mixing induced by the quadrocopter system. A high variability of methane sources is in agreement with the highly variable methane water concentrations measured within a radius of 100 m on 5 September 2018."

Hence, the "difference" between samples taken in parallel is pure noise caused by differences in the tightness of the sample containers, in handling, and in the analysis in the laboratory. I wonder how this issue can have escaped a group of nine authors.

We do not agree with the referee. The tightness of each of the sample containers was controlled by a pressure sensor, and the pressure data were both provided in real-time to the operators and recorded as well. Sample containers were air tight until being filled with air. The smaller pressure due to sampling above ground level was checked before closing the manual valves. Further, it cannot be assumed that the methane concentration was homogeneous. As an illustration, Fig. 5 and 6 show artificially released colour in a stably stratified morning atmosphere. Large horizontal concentration gradients can exist, and it is particularly difficult to quantify such non-stationary cases, as described by Schaller et al. (2018).

Finally, the difference between the delta values of the samples is height dependent, with much lower differences at the lowest sampling altitude, and an apparent temporal development of the difference for the sampling at higher altitudes. Pure noise should follow a more stochastic distribution of the differences in delta values.

Abstract Line 10: isotopic signature of what? delta13C or D/H ?

We changed the text to "methane isotopic composition $\delta^{13}C$"

Page 2, line 6: "Yet, current knowledge of CH4 sources remains inadequate." What do you mean by "inadequate"? Inadequate to justify or guide mitigation measures? I do not think so. We know very well where methane is produced and what could be done to reduce emissions.

We totally agree with the referee. The text is now changed to "Yet, current knowledge of CH_4 biogeochemical processes, transport and small-scale distribution remains inadequate."

What is the size of the sample containers (flasks)? Also, more detail about the valves and their connection with the glass flasks is necessary.

We added a figure explaining details of the valves and included in the text: "The air sampling system consists of 12 glass flasks (sample containers) of 100 ml content"

I do not understand the sentence on page 4, line 33-34 ("The most critical point were the manual plastic valves deployed routinely for the glass flasks in open position.")

We explain now more in detail and added Fig. 1. The text has been changed to: "The air sampling system consists of 12 glass flasks (sample containers) of 100 ml content, which are evacuated before take-off. They are equipped with two manual valves, one on each side, and additionally one electromagnetic valve, which is applied only during the flight (Fig. 1). Directly before the mission, each glass flask is linked with a vacuum pump RE5 of Vacuubrand, Germany. One valve is left open, and an electromagnetic valve is connected, which is normally closed. Then the flask is evacuated, and the pressure is controlled by a pressure sensor integrated in the electromagnetic valve. The flasks are opened during the flight with magnet valves, that are triggered either manually by remote control or automatically at altitudes predefined by the operator. After triggering, ambient pressure is reached within less than 2 s. The pressure sensors integrated in the valves are used to monitor air tightness. The most delicate component are the manual plastic valves, used to close the glass flasks for transport, which are designed to be air tight when closed, but not when open. They had to be treated individually and controlled to make sure that no leakage occurred during the mission. For quality control and redundance, two glass flasks were filled simultaneously, resulting in six possible sampling altitudes during one flight."

What is the vertical resolution of sampling, given the turbulence caused by four rotors keeping 19 kg of vehicle and payload afloat?

We added a figure of simulations showing the vertical velocity induced by the propeller blades (Fig. 2) and added in the text: "Assuming that in the worst case the sampling takes place within the downwash of the rotor blades of not more than -19 $m\,s^{-1}$, the sampling time of not more than 2 results in a vertical resolution of around 40 m. Sampling during descent with a speed of -2.5 $m\,s^{-1}$ adds an uncertainty in the altitude of 5 m. Altogether, the sampling is influenced by air in a height interval of 45 m. This is the sufficient for the sampling intervals of around 100 m, and for determining that the sampling was done below or above the temperature inversion."

Page 6, line 11: "The restoration of the peatland area towards a net sink of greenhouse gases, and in particular CH4, ..." Why should a rewetted peatland turn into a sink of methane?

This is indeed wrong. The idea was to point out that the restoration of a peatland area takes some time to act as CO_2 sink. Further, the restoration is usually accompanied by enhanced methane emissions, and it takes some years until the system is not such a strong methane source any more. We changed the text to:

"The restoration of the peatland area towards a net sink of the greenhouse gas CO_2 is a process of several years to decades. Initially, the restoration is accompanied by a strong increase in CH_4 emissions, which depend on vegetation and the water level (Couwenberg et al., 2011; Zak et al., 2015)."

What is the size of the rewetted Polder Zarnekow and other peatlands in its vicinity?

We included in the text: " The total rewetted area is 421 ha in size (**?**)."

The number of Figures is too large for a short article.

We removed the original Fig. 3 and Fig. 14. We further suggest to combine Fig. 4 and Fig. 13 in the final publication.

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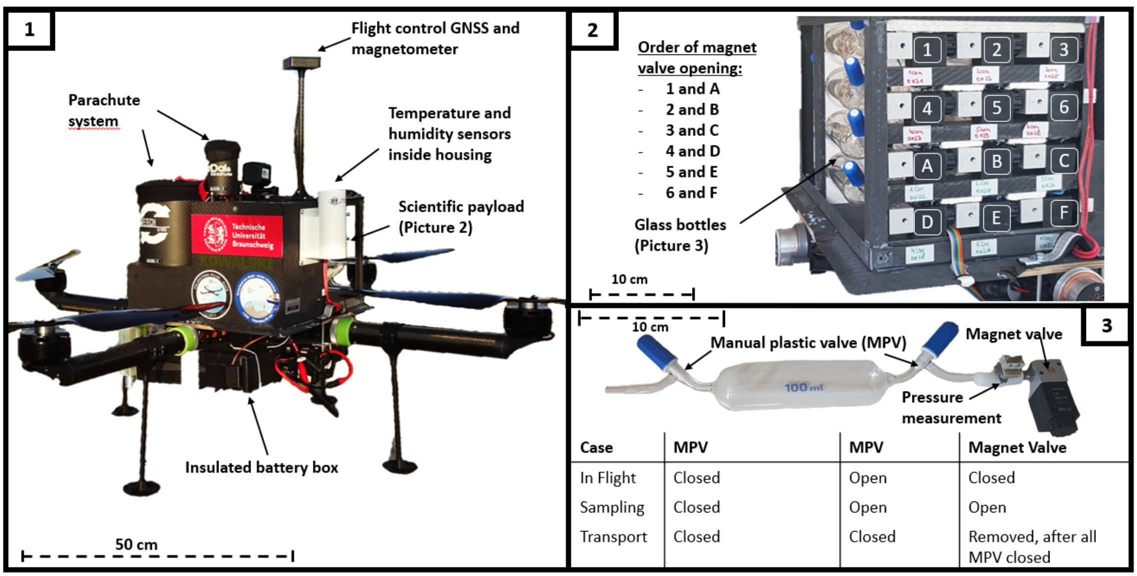

Case	MPV	MPV	Magnet Valve
In Flight	Closed	Open	Closed
Sampling	Closed	Open	Open
Transport	Closed	Closed	Removed, after all MPV closed

Figure 1. ALICE vital components.

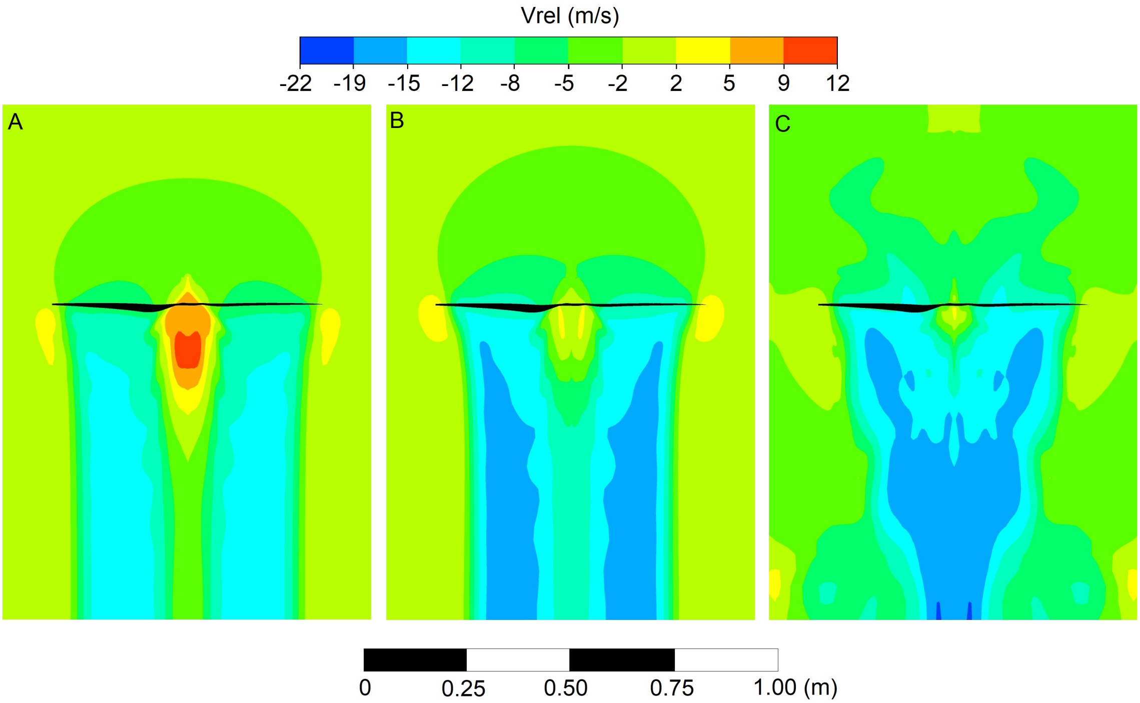

Figure 2. Simulation of the flow induced by a propeller blade during hover, climb and descent.

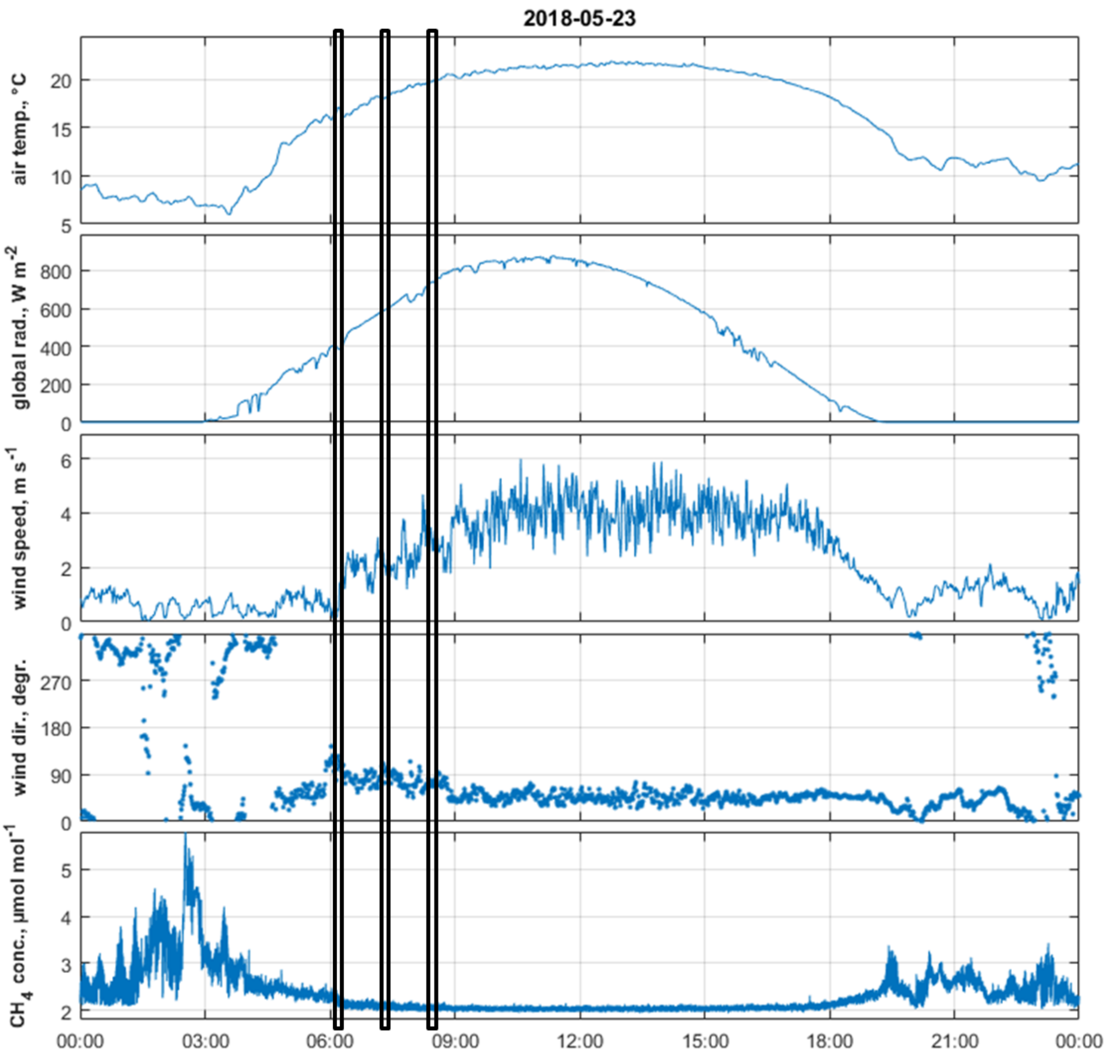

Figure 3. Diurnal course of the main meteorological parameters air temperature, global radiation, wind speed, wind direction, and methane concentration recorded at the meteorological mast at Zarnekow on 23 May 2018.

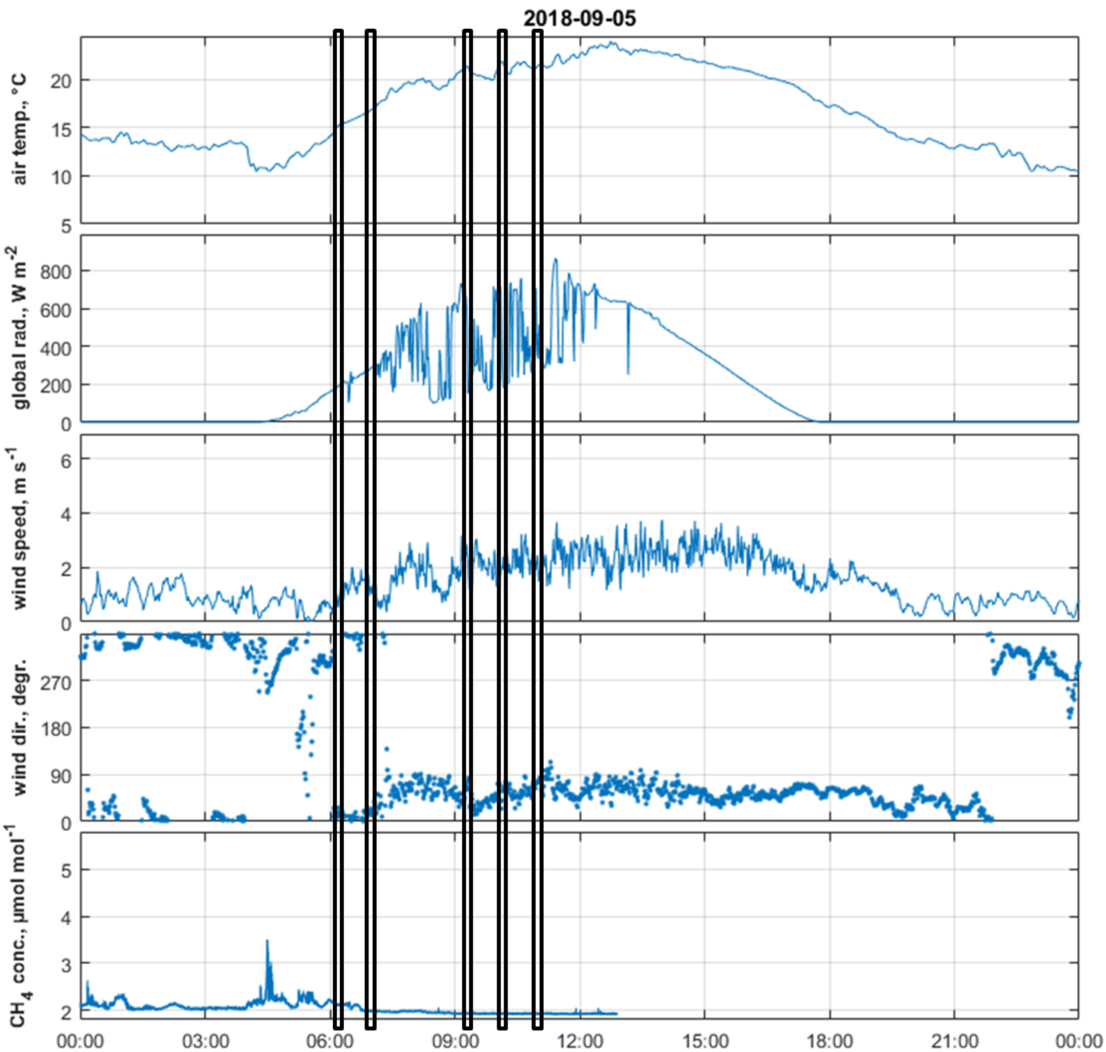

Figure 4. Diurnal course of the main meteorological parameters air temperature, global radiation, wind speed, wind direction, and methane concentration recorded at the meteorological mast at Zarnekow on 05 September 2018.

Figure 5. Illustration of concentration during stable stratification. The photo was taken at the air field Aue/Hattorf, Germany, on 2 October 2011 at 16:15 UTC, copyright Institute of Flight Guidance, TU Braunschweig

Figure 6. Illustration of small-scale concentration variability induced by aircraft. The photo was taken at the air field Aue/Hattorf, Germany, on 2 October 2011 at 16:15 UTC, copyright Institute of Flight Guidance, TU Braunschweig