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 2

The study by Lampert et al. describes an approach by which air samples can be obtained from different altitudes reaching far into the atmospheric boundary layer (ABL) using an unmanned aerial system. Though the higher level ideas and potential benefits behind the determination of CH4 isotopic composition within the ABL are distributed throughout the manuscript, a concise summary of the higher level aims of this study in both abstract and introduction is missing.

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."

We added in the introduction: "The need to improve understanding of the heterogeneous methane source and the transition from the surface into the atmosphere in the Arctic motivated the development of a flexible airborne sampling system, which provides information on atmospheric stability."

In addition, there is inadequate use of terms with regard to isotopic compositions and isotope ratios, as well as imprecision in describing footprint sizes.

We checked the text for consistency in wording. Now the terms "isotopic composition" and "delta values" are used throughout the text.

Further, we now avoid the term "footprint", which is used as a specific technical term. We changed the interpretation section to:

"The isotopic composition of the two air samples taken on 5 September 2018 simultaneously but with a constant horizontal distance of 13 cm agree within 0.1‰ at the lowest altitude of 10 m for Flight 1 to 4 (Fig. **??**). Besides this strong locally and temporally related agreement of the isotopic composition, for other altitudes and flights this difference is larger than the uncertainty, and therefore the systematic differences are treated as features. There are several striking features in the profiles of isotopic composition, from lower to higher altitudes:

- On 5 September 2018 the difference in delta values between the two simultaneous samples is systematically smaller at 10 m altitude compared to the higher altitudes, except the last profile.
- On 5 September 2018 the differences in delta values at 100 m altitude increase during the course of the day. The profiles of differences in delta values exhibit similarities for parts of the profile between subsequent flights on both days.
- The delta values are more negative in the morning before vertical mixing starts, as long as a temperature inversion is present (first flight on 23 May 2018 below 150 m, and first flight on 5 September 2018 below 70 m). This is in agreement with methane from biologic processes emitted from the surface that are not vertically mixed.

As the order of analysing the air samples was chosen randomly, the differences of the delta values exceed the uncertainty and the differences in delta values correlate in section between subsequent flights, it is assumed that the differences in delta values are physically present in the air samples. Two aspects can be highlighted:

- An ideally vertically stratified delta value would not be sampled by the present system as the very dynamic circulation process around the copter does not result in a homogenously mixed air at the sample ports. On the contrary, this circulation process can even amplify natural inhomogeneity. It is assumed that the differences in delta values indicate natural inhomogeneity, but it is not possible to prove it based on the data set.
- Beside the buoyancy vertical turbulent mixing the natural inhomogeneity of delta values is not known for the measurement site. Small-scale horizontal variability can be induced by inhomogeneous sources. Episodic CH_4 outbursts on short time scales of few min have been observed by Schaller et al. (2018). The high spatial and temporal variability of methane concentration and isotopic composition reported here is in agreement with their observations. Such variability

of methane emissions at the field site as well as the potential upwind CH_4 sources cause the inhomogeneous character of the air samples.

Respecting that the air sample profiling gives a snapshot of a turbulent mixing process, a clear transition in the vertical distribution of the delta values can be seen."

After considering the below specified aspects, this publication could be reconsidered for publication in AMT.

We hope that we can answer all concerns satisfactorily.

1. At this stage, the abstract is not convincing and stops short of revealing the scientific benefit that may arise from profiles of methane isotopic composition reaching far into the atmospheric boundary layer.

We changed the beginning of the abstract to: "To determine the source of methane emissions at different altitudes and to study mixing processes, a quadrocopter air sampling system was developed. A proof of concept study was performed to demonstrate the capabilities of quadrocopter air sampling for analysing the methane isotopic composition δ^{13}C in the laboratory. The advantage of the system compared to classical sampling at ground 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."

There is a more or less recent publication by Roeckmann et al. in ACP that summarizes nicely the potential benefit of tall tower and, thus, probably also airborne measurements.

We would like to thank the referee for pointing out this interesting publication. We included the following sentence in the introduction: "Field measurements of methane isotopic composition have been performed at the Cabauw tower at a sampling altitude of 20 m, demonstrating the potential of isotopic analyses to determine contributions from isotopically different sources to the dominating source (Röckmann et al., 2016)." Further, we added "UAS can be operated in remote areas, requiring less infrastructure in comparison with permanent measurement stations, and they can be used more flexibly than manned aircraft, enabling fast reactions to environmental events like changes of emissions through rain, drought, construction, or fire."

I suggest using this overarching view to introduce the topic and made a suggestion in this direction in the abstract- section of this referee comment. Some of the ideas can also be found in the conclusions section.

We would like to thank the referee for the practical recommendation, which we took into account (see answer to comment above).

2. In the methods section, I am missing information on from where exactly air is sampled. There is quite some discussion on the adverse effect air parcel transport due to rotor downwash, so that this issue, and how it was tackled (if at all), should be mentioned already in the methods section.

We included three more co-authors who have done numerical simulations, and added a subsection about simulations with the following text: "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 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 $\mathrm{m\,s}^{-1}$ 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 was 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."

3. In the methods section, water samples are referred to as a proxy for spatial heterogeneity of the emitted methane isotopic composition. However, methane dissolved in water and the methane emitted to the atmosphere will have a different isotopic composition due to the fractionation effect of volatilization. I wonder why the authors have not taken air samples from a closed chamber several times and used the keeling-plot approach to determine the isotopic composition of the emitted methane? This would be a direct measurement of the isotopic composition of emitted methane.

We agree that it would be good to have closed chamber samples for comparison of the delta value. However, the aim of this study is to show the vertical distribution of the methane isotopic composition in dependence of atmospheric stability, which we interpret as related to different methane sources.

4. Possible influence from surrounding sources, i.e. surrounding land cover/land use, is not discussed at all, but is necessary in view of the highest altitudes at which measurements took place.

We agree that we cannot fully understand the processes without taking into account more information, like land cover. However, the aim of the manuscript is to show that it is possible to identify vertical layers of different isotopic composition with the experimental setup of the quadrocopter. Further, the case study shows that there is a high horizontal variability of methane sources, which requires specific tools to further understand the observations. We changed the introduction to: "The goal of the study is two-fold:

- Proof-of-concept for the experimental setup of the quadrocopter borne sampling system, and subsequent laboratory analyses, to identify vertical layers of different isotopic composition
- Identification of small-scale atmospheric methane inhomogeneities which require the development of new methods for understanding dynamic processes

In order to test the system's capabilities of providing reliable vertical profiles of the isotopic composition, measurements were performed at a rewetted peatland site, Polder Zarnekow (Zerbe et al., 2013), which is known as a source of biologically produced methane **?**. In the absence of turbulent mixing, local emissions of the wetlands produce a depleted delta value

compared to the atmospheric background above the temperatue inversion. During the morning transition, when the stable stratification is gradually replaced by a convectively mixed atmospheric boundary layer, the isotopic composition should adjust to a constant delta value throughout the profile within the uncertainties of laboratory air sample isotopic analyses. To support the hypothesis of the small-scale horizontal inhomogeneity at the study site, the methane concentration of water samples from locations within a radius of 100 m was analysed."

5. When on page 4, the design point is mentioned, and the reasoning behind, the introduction makes more sense. Please underline this connection already in the introduction.

We added earlier in the introduction: "The need to improve understanding of the heterogeneous methane source and the transition from the surface into the atmosphere in the Arctic motivated the development of a flexible airborne sampling system, which provides information on atmospheric stability."

6. There is a fundamental misunderstanding of isotope ratios. The isotope ratio (not isotopy ratios) cannot be negative. After transformation of the isotope ratios to the delta scale, negative values arise if the abundance of the heavy atom is lower than that of the reference material. The use of technical terms in a scientific journal has to be right.

We apologize for the sloppy use of the terms. Now the terms "isotopic composition" and "delta values" are used in the text accordingly.

7. There seems to be quite some imprecision when the discussion addresses footprints. I suggest consulting the literature, one example is given in the detailed comments. It is unclear why footprints for a measurement taken at 100 m should be similarly small as for a measurement at 10 m, as long as stable atmospheric conditions prevail.

We admit that we used the technical term "footprint" in an imprecise way, and rephrased the sentences without using it.

8. Especially the photographs in figure 2 are at least inadequate for a journal like AMT. Both the picture detail and the background makes it practically impossible to discern anything. In addition, the font color is illegible.

We replaced the picture with Fig. 1.

9. Figure 3 is not helpful for the manuscript.

We removed the figure and the reference in the text.

10. Figure 9: showing concentration until 10:00 would be enough, source isotopic composition may be best reflected during night. What is the increased concentration at approx. 12:00?

We corrected the plots and included them in the figures of the time series of meteorological parameters.

P1L1: A higher level rationale is missing in the abstract.

We changed the beginning of the abstract to: "To determine the source of methane emissions at different altitudes and to study mixing processes, a quadrocopter air sampling system was developed. 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 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."

In addition, the first sentence sounds a bit like quadrocopter air sampling influences laboratory analysis. I get the drift of the authors, but suggest something like "The determination of the methane isotopic composition at a tall tower and in high temporal resolution indicated a high potential to further constrain methane budgets at regional scales. However, tall towers are rare research infrastructures that may be supported by airborne measurement approaches. In this proof of concept study, we demonstrate the feasibility of using aquadrocopter to obtain air samples at heights between 10 and 600 m above ground. The methane isotopic composition of the air samples was subsequently determined in the laboratory."

We added in the introduction: "The need to improve understanding of the heterogeneous methane source and the transition from the surface into the atmosphere in the Arctic motivated the development of a flexible airborne sampling system, which provides information on atmospheric stability. In this context, unmanned aerial systems (UAS) fill an observational gap for methane mixing processes. They are able to sample small scales with a typical horizontal distance of 1 km, if they are required to be operated in the line of sight, and they reach the top of the atmospheric boundary layer, with a maximum altitude of typically around 1 km. UAS can be operated in remote areas, requiring less infrastructure in comparison with permanent measurement stations, and they can be used more flexibly than manned aircraft, enabling fast reactions to environmental events like changes of emissions through rain, drought, construction, or fire."

P2L1: I suggest changing to "32 times that of CO2"

We changed as suggested.

P2L6: Please elaborate on what exactly remains inadequate. The source categories are quite clear I think, but the relative contributions of the different categories need better constraints.

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

P2L20: please revise typo in background

We corrected the typo.

P2L21: I suggest changing to "indicates", since isotopic compositions of biological sources and fossil / thermogenic methane may overlap in the region of -55 per mil.

We changed the text as suggested.

Figure 2: please use a monochrome background for the figure, and change to "battery" on left picture

We replaced the figure and changed as suggested (see Fig. 1)

P3L17: please decide either for methane isotopic composition or for isotope ratio. Isotopic ratio is at least uncommon.

We now use the terms "isotopic composition" and "delta value" consistently throughout the manuscript.

In addition, line 16 starts naming the aim, and in line 17, the goals are listed. Please revise the section.

We revised the section, and changed to:

"The goal of the study is two-fold:

- Proof-of-concept for the experimental setup of the quadrocopter borne sampling system, and subsequent laboratory analyses, to identify vertical layers of different isotopic composition

– Identification of small-scale atmospheric methane inhomogeneities which require the development of new methods for understanding dynamic processes

"

P3L22: I suggest sharing your hypotheses that this is due to the more pronounced influence of the wetland producing depleted CH4 compared to the atmospheric background in absence of turbulent mixing.

We changed the text to: "In the absence of turbulent mixing, local emissions of the wetlands produce a depleted delta value compared to the atmospheric background above the temperature inversion."

See comment above with regard to isotopic ratio - isotope ratio

We now use the term "isotopic composition" throughout the text.

P4L2: it sounds like the quadrocopter was not based on a commercial chassis. If this applies, please state more clearly.

We changed the text to "The quadrocopter ALICE was designed as platform to carry meteorological sensors and 12 glass bottles for air sampling. The construction of the quadrocopter was calculated for the specific tasks and payload described in the following. Therefore, all relevant load cases that were expected during the flight were applied in analytical and numerical models to optimize the structure of the quadrocopter. Modern manufacturing methods like selective laser sintering and laser cutting where used to build the structure as light-weight as possible but as stable as necessary."

P4L3: unclear why the payload should alter the dimensions of the UAV. Please provide details.

We changed the sentence to "ALICE has dimensions of 1.56 m x 1.56 m x 0.38 m, not including the scientific payload."

P4L4: I suggest: "At a tare weight of 4 kg, ALICE's maximum take-off weight is 25kg." Does the tare weight include batteries for driving the rotors, or are the LiPo batteries for both operating the scientific instrumentation and driving the rotors?

We changed the text to "At a tare weight of 4 kg, ALICE's maximum take-off weight is 25 kg. For the operations presented here, the total weight was 19 kg, which is composed of 4 kg the quadrocopter system itself, 7.2 kg of LiPo batteries with a total capacity of 21 Ah and a nominal voltage of 44.4 V for rotor power supply, and 7.8 kg payload including sensors, glass bottles, data acquisition, power supply for payload and a safety parachute of 12 m^2."

P4L27-31: I don't understand exactly, if there are both manual and magnetic valves necessary. Please explain in more detail. My first impression would be that only magnetic valves that are normally closed would suffice the purpose?

We replaced the figure of the ALICE system and included more information about the valves (see Fig. 1).

P4L34: From this sentence I guess that I am right that there are two sets of valves. However, the meaning of this sentence is unclear.

We hope that the setting of valves becomes clear with the new figure (Fig. 1).

P5L4-6: this section seems redundant to me. The electromagnetic valves were explained some lines above.

We agree that this is already mentioned before, and removed the sentence.

P5L16: I suggest starting with where the samples were analysed and that they were brought there directly after the end of the mission, and then go into detail. Otherwise the transport issue pops up out of the blue.

We changed the order of the sentences to "Following the quadrocopter mission, the sample containers (SC) were transported to the laboratory at the Alfred Wegener Institute in Bremerhaven, Germany for aanalysis of the isotopic composition. The $\delta^{13}C$ value of the air samples was analyzed using a Delta plus XP mass spectrometer combined with a combustion oven, a gas pressure interface and a pre-concentration device (PreCon) (ThermoFinnigan, Bremen, Germany)."

P6L11: I cannot follow that a restored peatland should become a net sink for methane?

This is indeed wrong. The intention 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)."

P6L25: I suggest including subsection heads "site description", "water sampling" and "flight strategy"

We modified the structure as suggested.

P6L35: Please provide rationale for sampling during descent only.

We added in the text: "During ascent, the temperature profiles were studied, as a base to plan sampling altitudes for the descent."

P7L5: Please explain why the first flights were after sunrise though the aim was to investigate the transition from night time stable conditions to daytime turbulence.

We added in the text: "Flights were permitted between sunrise and sunset. As it takes some hours until the nocturnal temperature inversion is heated away, it was possible to take the air samples during the transition from nocturnal stable boundary layer to the convectively mixed boundary layer. "

P7L11/12: please change typo to evidenced, suggest to change to "indicated"

We changed to "indicated" as suggested.

P7L22: Please indicate nocturnal temperature inversion in figure.

We indicate the height interval of the temperature inversion with a horizontal box in the figures of potential temperature and water vapour mixing ratio.

P8L25: please change to stratification

We changed the text as suggested.

P9L9: -48to -49 is not an isotope ratio, but the delta value. I suggest using the term isotopic composition throughout the manuscript.

We now use the terms "isotopic composition" and "delta value" consistently throughout the text.

P9L26: This assumption is erroneous. The footprint of a measurement at a given height is larger under stable conditions, because the effect of advective transport is much more distinct compared to a situation dominated by turbulence in which vertical mixing is stronger. Please refer to several papers for example by Kljun et al., e.g., "A three-dimensional backward

lagrangian footprint model for a wide range of boundary layer stratifications". In addition, the footprints of measurements at 10 m height will be much smaller than those at 100 m height.

We admit that we used the technical term "footprint" in an imprecise and misleading way, and rephrased the sentences without using it.

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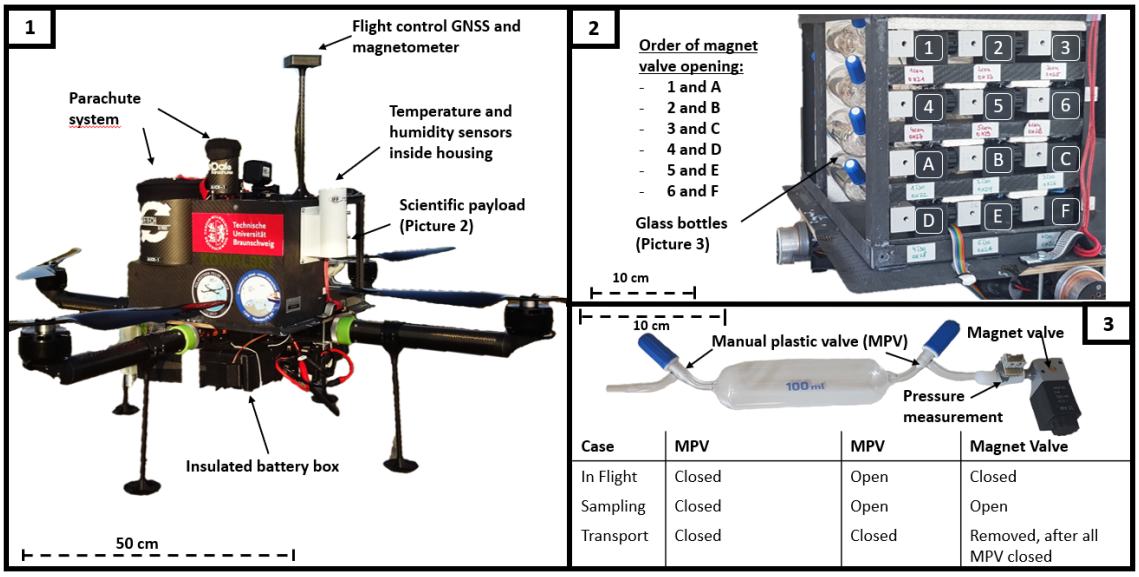

Case	MPV	MPV	Magnet Valve
In Flight	Closed	Open	Closed
Sampling	Closed	Open	Open
Transport	Closed	Closed	Removed, after all MPV closed

Figure 1. ALICE vital components.

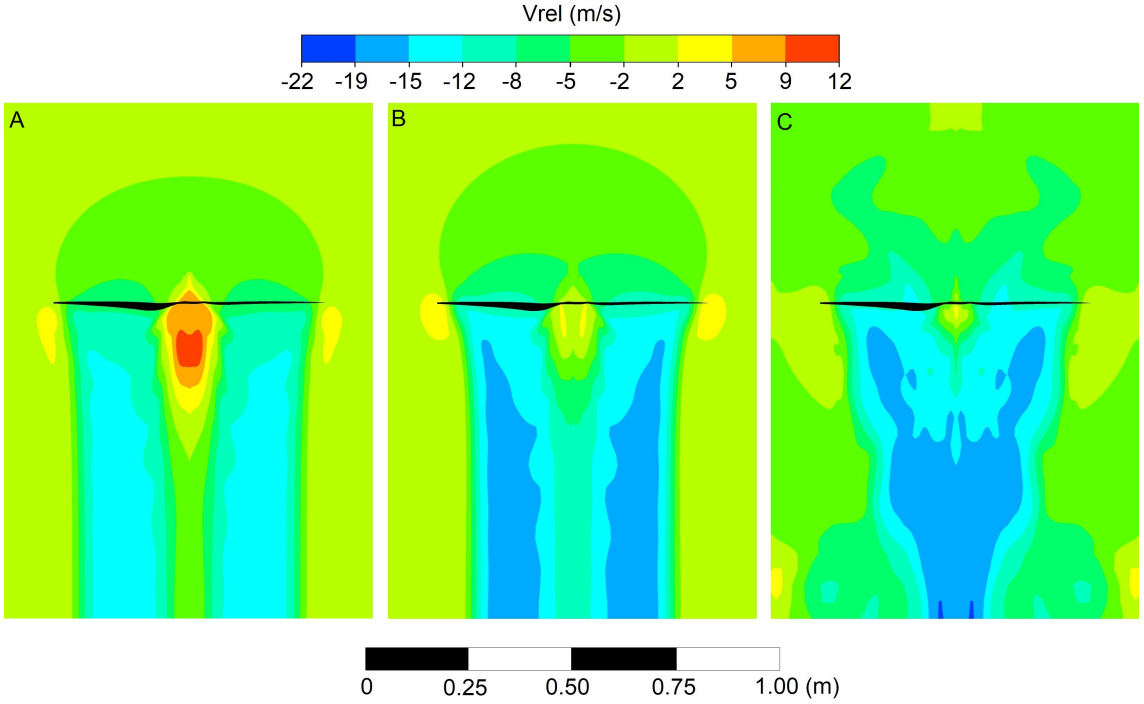

Figure 2. Simulation of the flow induced by a propeller blade during hover, climb and descent.

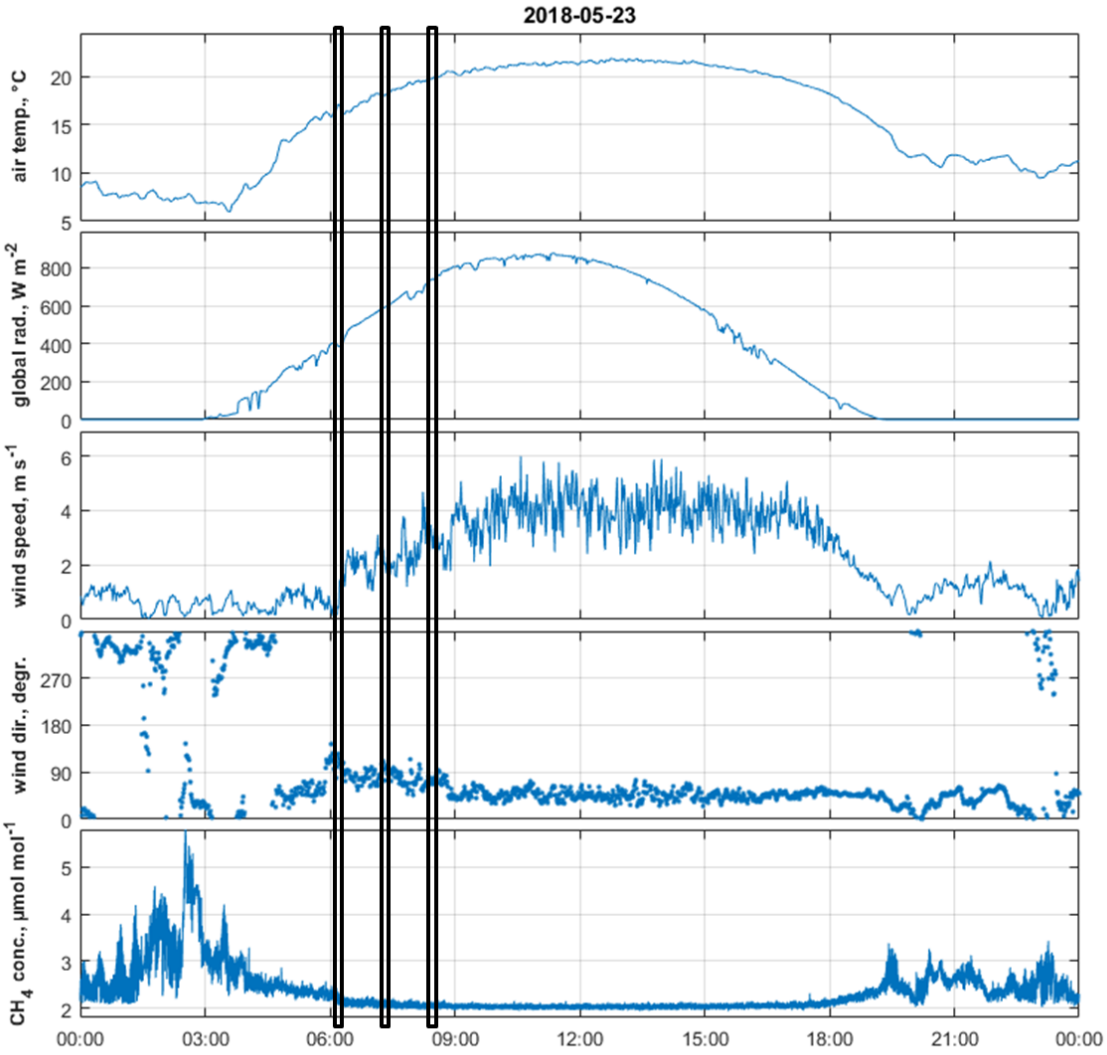

Figure 3. Diurnal course of the main meteorological parameters air temperature, global radiation, wind speed, wind direction, and methane concentration recorded at the meteorological mast at Zarnekow on 23 May 2018.

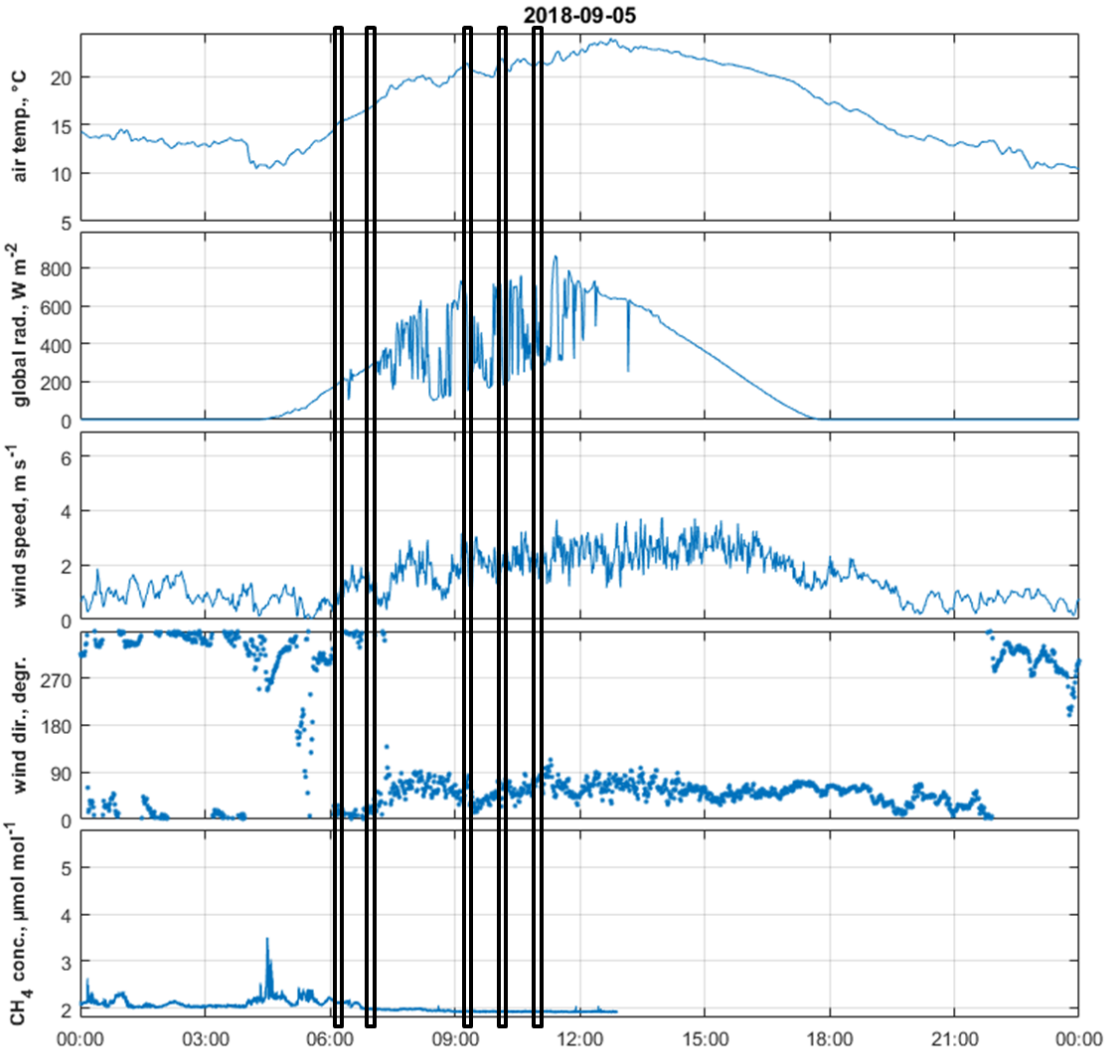

Figure 4. Diurnal course of the main meteorological parameters air temperature, global radiation, wind speed, wind direction, and methane concentration recorded at the meteorological mast at Zarnekow on 05 September 2018.

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