MEMO²: MEthane goes MObile – MEasurements and MOdelling

Report / Publication of the results from the campaign in Silesia

Lead author

Jaroslaw Necki
Faculty of Physics and Applied Computer Science

Mickiewicza 30
30-059 Kraków
Poland

Telephone: +48 12 617 30 46
Email: necki@agh.edu.pl

Deliverable D1.3

Delivery month Annex I	36		
Actual delivery month	38		
Lead participant: AGH	Work package: 1	Nature: Report	Dissemination level: PU
Version: 1			

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 722479.

Table of contents

1. Executive Summary

In May and June 2018 several ESR's and PI's where involved in a large-scale field campaign co-organized by AGH - University at the Upper Silesian Coal Basin (USCB), together with German Space Agency (DLR). The CoMet (Carbon Dioxide and Methane Mission) campaign, coordinated by DLR, was oriented toward the application of airborne instruments for methane (CH_4) balancing of the entire USCB, as USCB belongs to the biggest area of anthropogenic CH_4 emissions in Europe. MEMO^2 was official partner of this campaign and provided ground-based measurements and UAV measurements delivering data from the bottom part of atmosphere. The objectives of MEMO^2 to participate in the campaign were both scientifically and educational.

Four ground teams (AGH, UU, UHEI, UVSQ) and one unmanned aerial vehicle (UAV) team (RUG) as well as a modeling team (EMPA) from MEMO^2 were active during two weeks of the campaign. Two of the MEMO^2 ESR were also involved in the Fourier-transform infrared spectroscopy (FTIR) measurements executed by DLR – as novelty technique of mobile FTIR was applied. The ground teams visited in particular coal mine shafts, aiming on completing data records necessary for a successful use of the Gaussian plume model. The encountered mixing ratios occasionally exceeded 100 ppm. During DLR aircraft flights, the ESRs performed measurements of methane mole fractions below the flight track of the aircraft, recording data that were essential for applying the mass balance approach at different scales (the whole USCB or single mine excavation areas). Usually, CH_4 mixing ratios didn't exceed 4 ppm along these routes. The UAV team was involved in the quantification of single shaft emissions, flying over the different exhaust shafts of coal mines that are the biggest emitters in the USCB. The active AirCore technique used revealed CH_4 mole fractions of up to ~300 ppm, at a few hundred meters downwind of the exhaust shafts. To support the ground measurements, Lagrangian model calculations were run by the EMPA group to provide information of the CH_4 plume location.

Data delivered by the mining authorities were compared with results of direct measurements done by the ESRs giving them opportunity to test the methodology of measurements.
Outcomes of the campaign are:

- Emission rates from individual ventilation shafts estimated and compared to the coal mine data.
- Emission from USCB estimated and compared to the existing inventories.
- Capability of a group activity during field campaign

2. Introduction

2.1 Background

The MEMO^2 project focusses on estimation of CH_4 emissions from different sources, particular anthropogenic origin. Hard coal mining excavation, still occurring at USCB is accompanied by extraordinary large emission of CH_4 released to the atmosphere during mining as a byproduct. Each of the 32 mines operating over the USCB area releases CH_4 to the atmosphere through ventilation shafts or degasification infrastructure, and the total emission rate is estimated at almost 500 kt CH_4 / year. This in turn is a good proxy to validate the field measurements.

The COMET campaign was organized over the USCB by DLR, Germany. Participants from MEMO^2 joined in to support CH_4 emission estimates on the regional scale performed by airplanes equipped with in-situ and remote analyzers. The aircraft could not fly at low altitudes and therefore could not probe the lowest part of the boundary layer. The role of the MEMO^2 team was to measure the spatial and temporal distribution of CH_4 at the ground level (UAV – up to first 100 m). ESRs from different groups formed a closely collaborating ground team which was working in parallel to achieve the data collection necessary for mass balance methodology.

Beside the scientific approach, the participation of the ESRs in this campaign followed also a trainng approach. Field experiments are very common in environmental sciences. Measurements under real conditions reveal new discoveries which can be easily overlooked in the laboratory. For ESRs practical experience is essential. In case of CH_4 budgeting, measurement campaigns open the eyes of young researchers for problems that are relevant on different scales. The young researchers can also experience how important it is to cooperate with all other campaign participants, as one person or even group is not capable to quantify emissions at all of these scales. Large field campaigns are great opportunities for the ESRs to improve their experimental skills and team skills.

2.2 Scope of the deliverable

The campaign was originally planned for February 2019. According to the invitation of DLR to participate to the CoMet project, the initial schedule was shifted so that MEMO^2 ESRs got the opportunity to participate in a joint campaign with DLR. The MEMO^2 objectives to join the campaign were to:

- Experimentally determine the Silesian emission of methane (scientific goal)
- Test and improve experimental abilities of ESR in field conditions (educational goal)
- Realize a small project at very local scale (scientific goal)
- Enhance the cooperative skills of ESR under demanding conditions and time pressure (social skills)

This deliverable report will describe the contribution of MEMO^2 to the campaign and give a brief overview of the outcomes. Scientific results have been published or are in preparation of publication, whereas at least 4 publications are envisaged.

3. Content

The MEMO^2 campaign associated to the CoMet campaing targeted to estimate the CH_4 emissions from hard coal mining excavation over the USCB. It was organized from 24th of May till 10th of June 2018. All “ground base” teams were stationed in the village Wisla Mala, located in the southern part of USCB, in the vicinity of 4 large mining facilities: Pniowek, Silesia, Zofiowka and Borynia.

Fig. 1: Ground base team performing first parallel measurements at USCB during the MEMO^2 campaign in 2018.

Beside mobile Cavity Ring-Down Spectroscopy (CRDS) measurements and air sampling realized exclusively by the MEMO^2 team, MEMO^2 also supported groups that performed stationary FTIR and mobile FTIR measurements, wind profile observation and an inter-airport coordination team. Input of the MEMO^2 project to these activities allowed ESRs to learn different techniques and understand problems of determining the vertical column distribution of CH_4, which is different from “in situ” measurements. During occasional briefing meetings the ESRs had also contact with the airborne measurements group stationed close to the Katowice airport (Northern USCB). The MEMO^2 team consisted of 5 groups distributed over 3 cars (Fig. 1) operating all days and some nights with only 2 days off during the campaign. To ensure safe working conditions, ESRs were working in pairs with other persons separating driving from observation of instruments and navigation in terrain. The activities of the individual groups were related to 6 particular tasks:

T1: Collaboration with the airborne group
T2: Single ventilation shaft observations by mobile vans
T3: Identifying CH_4 sources over the USCB with mobile platforms
T4: Mapping city emission
T5: Isotope sampling of methane from its different sources
T6: Drone (UAV)-based measurements

The tasks will be described in more detail in the following chapters 3.1 to 3.6.

3.1 Task 1 - Collaboration with the airborne group

The first task required the coordination between all groups to support the airplane trajectory using roads. Airborne measurements were performed usually during midday hours when the boundary layer included the lower 1.5 km of the troposphere. To apply the mass balance technique, the airplane performed so called “wall” measurements on the downwind part of the region to integrate the whole plume of methane coming from the different ventilation shafts spread over the USCB. The length of the wall usually exceeded 50 km.

To account for possible CH_4 plumes from other regions bringing CH_4 into the USCB, airplanes needed also to perform “wall” measurement along the upwind area. For this reason, the trajectory of the airplane was divided into one upwind and two downwind parts. Cars from MEMO² were covering streets below the flight track (Fig. 2) as fast and as frequent as it was possible. The upwind “leg” of the flight track was usually much longer as it was not necessary to repeat this “transects” more than one time during the unchanged wind conditions. The downwind “legs” were shorter and the measurements were repeated at least 4 times. The data were corrected for calibration, checked by inter-comparing analyzers in all of the cars and delivered to the database. The airborne measurement group used the ground data for closure of the unknown part of the plume below the lowest flight level (approx. 500 m) by interpolation of CH_4 mixing ratio between the airplane and ground-based measurement results. To ensure the compatibility of the results the airborne and ground-based teams organized common measurements of calibration cylinders that were used as reference scale for analyzers installed in the aircraft. The estimated CH_4 budget of the USCB obtained from this task ranges between 400 and 550 kt CH_4 per year. The bottom-up estimates are pointing exactly at the same range (350 – 700 kt CH_4 / year).

Fig. 2: Example result of the USCB transects realized by different groups the MEMO² team. The UU group followed leg AB, the UHEI group followed leg CD and the AGH group followed leg DE (upwind).

3.2 Task 2 - Single ventilation shaft observations by mobile vans

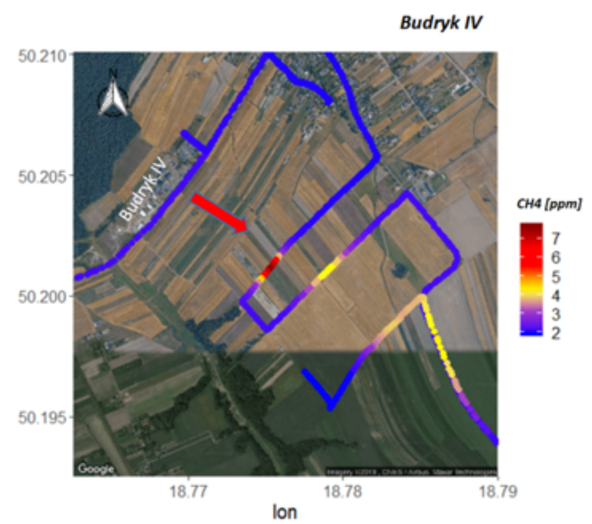

Fig. 3: Example of CH_4 plume crossing at different distances from the exhaust ventilation shaft of the Budryk coal mine

The second task was realized when no flight activity was performed by DLR. In the mornings and evenings under appropriate meteorological conditions (stable wind and no rain), the cars equipped with analyzers drove along the roads circulating the exhaust ventilation shafts of coal mines.

The idea of the quantification of CH_4 release relies on the application of the Gaussian plume dispersion model. It requires not only the CH_4 concentration distribution along the cross-section of the plume but also wind direction and speed, stability class of the atmosphere and parameters of the source itself. All of this information was collected by the cars cruising in the vicinity of the shafts. Each car was equipped with sonic 2D anemometer, GPS device and Picarro CRDS laser analyzer. Usually, it was not possible to drive full circles around the shafts. Where possible, cars tried to cross a plume at different distances from the shafts (Fig. 3).

During the campaign, the MEMO² team visited in total 21 shafts, acquiring estimates of CH_4 emission connected to mining activity. For individual shafts, the Gaussian plume modeling has a relatively large uncertainty of the calculated release in an order of magnitude of +- 50 %. The total emission from the mining sector can then also be estimated with an accuracy of about 50%. The CH_4 emissions in most cases agree with the emission factors reported by mining facilities to official databases.

3.3 Task 3 - Identifying CH_4 sources over USCB with mobile platforms

Aim of this task was to investigate if and with what accuracy mobile screening is able to reliably locate single sources of CH_4 as in some cases single plumes might be missed by a car, even if it is equipped in very sensitive equipment.

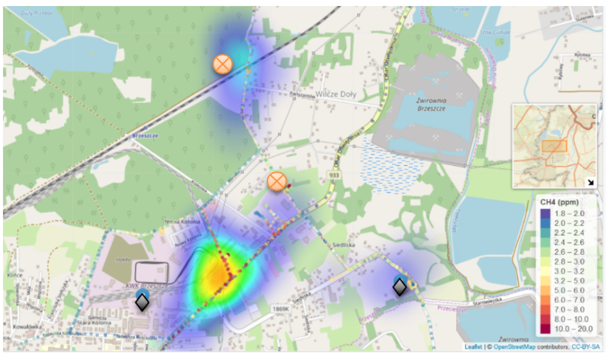

Fig. 4: Analysis of methane spatial distribution in vicinity of the Brzeszcze hard coal mine. Plumes related to mining activity (black diamond) and natural gas distribution (orange crossed circles) were spotted.

For this task teams focused e.g. on single mines. Cars drove a dedicated transect, where all CH_4 enhancements were recorded and subsequently they returned to this location trying to localize and quantify the source of CH_4. This exercise was performed simultaneously by all 3 groups to validate the comparability of the equipment and to enhance awareness of a plume behavior under real field conditions. Usually, a single mine has 3 different exhaust shafts for ventilation, and not all of them release the same amount of CH_4. Next to the mine shaft also city gas network leakages, landfills, wetlands and farms were identified as CH_4 sources in this region.

3.4 Task 4 - Mapping city emissions

The USCB covers several medium size cities and the Katowice agglomeration is inhabited by 3 million people. The mining activity is not the only source of CH_4 there. The city gas network is leaking, multiple landfills are releasing CH_4 to the atmosphere at rates of few kilotons of CH_4 per year and also abandoned mining grounds are sources of CH_4.

The MEMO^2 team performed CH_4 mapping over some selected cities (Nowa Ruda, Chorzów and Katowice), revealing numerous individual CH_4 sources. The attribution of CH_4 enhanced concentrations (up to 20 ppm) is not possible using only the mole fraction measurement as sources mix. Therefore air samples were taken to ascribe the CH_4 origin according to its isotopic composition.

Some groups were travelling to hard coal mines, where shafts are located close to the city or even in city centers. In such cases, the average city level of the CH_4 mixing ratio was enhanced by a few ppm (double the background concentration) during the night time. Air that was strongly enriched in CH_4 was encountered also in some cities (e.g. Katowice, Fig. 5), although there are no current mining activities.

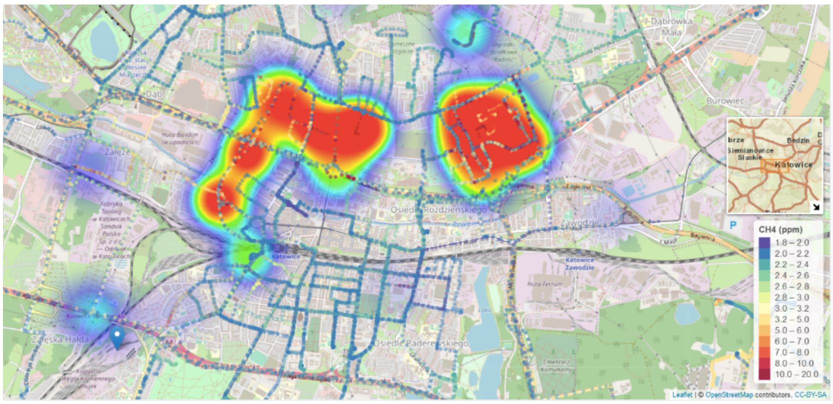

Fig. 5: Result of methane mapping activity in Katowice city with the marked heatmap of methane sources not related to the ventilation of mines

We found piles of excavated bedrock in which parts of coal were burning. The estimated CH_4 release from such burning heaps was in range 0.1 – 0.6 g CH_4 / s*m^2. Fires can also occur in formerly excavated coal seams, which are nowadays abandoned. These seems are relatively shallow and local (approximately 60 m below ground). Such fires were not included in available methane budgets by now. Unfortunately, the campaign was too short to cover all the sources.

3.5 Task 5 - Isotope sampling of methane from its different sources

The goal of this task was to use isotope information for source attribution. The analysis of isotopologues is a widely used tool in carbon cycle studies. The USCB coal mines release CH_4 which is often depleted in ^{13}C isotopes due to the migration processes occurring in coal seams and bedrock. Also, the geological history of coal at USCB is connected with secondary biogenic activity, shifting the isotope ratio towards lighter values. Air samples were collected while passing the CH_4 plumes from the ventilation shafts, directly at the shafts (in cooperation with ventilation officers of the mines), close to the burning heaps, next to the natural gas pressure reduction units, and in vicinity of landfills. All of them were analyzed with the IRMS technique at Utrecht University.

The average methane isotopic composition of particular sources encountered in the USCB are summarized in Table 1.

Table 1: Isotopic composition of methane sources at USCB

Source of CH_4	$\delta\ ^{13}C$
Coal mining	-48
City gas network	-53
Landfill	-68
Coal heap fire	-28

3.6 Task 6 - Drone (UAV)-based measurements

The drone team of the University of Groningen flew a recently developed active AirCore system (Andersen et al., 2018) aboard an unmanned aerial vehicle (UAV) to obtain CH_4 mole fractions downwind of individual coal mining ventilation shafts. In addition to CH_4 mole fraction measurements, CO_2, CO, ^{13}C, and deuterium in CH_4, atmospheric temperature, pressure, and relative humidity were also measured. Wind-speed and wind-direction measurements were made using a 2-D anemometer.

3.6.1 Mass balance approach

The first step with this approach is to take the sampled 'curtain' of mole fractions obtained from the active AirCore, and inter- and extrapolate the data into a full two-dimensional grid using the statistical kriging method. Based on this mole fraction curtain, a flux corresponding to each flight was estimated by applying a mass balance approach:

$$Q = \frac{v \cdot \Delta X \cdot M_{CH4}}{R \cdot T} \sum_{i}^{k_i} \sum_{j}^{k_j} C_{i,j} \cdot P_{i,j} \tag{1}$$

where the output of the emission rate Q is in $kg\ s^{-1}$, v is the wind speed in $m\ s^{-1}$ and assumed to be constant throughout the flights, k_i is the number of horizontal grid boxes in the kriged plane, k_j is the number of vertical grid boxes in the kriged plane, M_{CH_4} is the molecular mass of methane in $kg\ mol^{-1}$, $C_{i,j}$ is the mole fraction of grid box i,j in $mol\ mol^{-1}$, ΔX is the area of each grid box in m^2, R is the universal gas constant in $kg\ m^2\ s^{-2}\ K^{-1}\ mol^{-1}$, T is the temperature in K, and $P_{i,j}$ is the pressure at each grid box in Pa.

3.6.2 Inverse Gaussian approach

The inverse Gaussian approach utilizes the sampled three-dimensional mole fraction data from the active AirCore together with a Gaussian dispersion model for point sources (Fig. 6). The AirCore flight data is paired with the Gaussian dispersion model, where the emission rate (Q), the mixing of pollutants in the horizontal- and vertical direction (σ_y and σ_z), and the center height of the plume (h) are free parameters. A best fit for equation (1) to the data can be found for these four parameters by finding a minimum of the sum of squared errors.

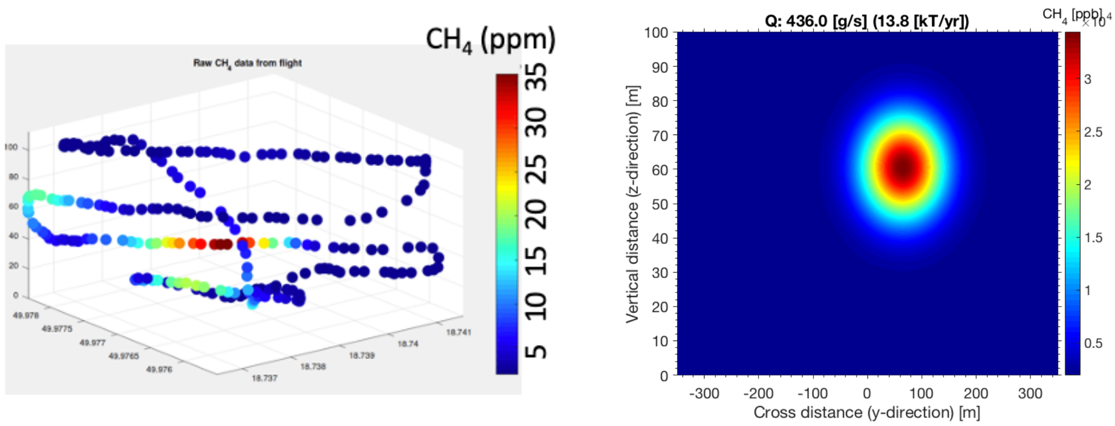

Fig. 6: The 3-D CH_4 concentration profiles of a UAV flight downwind of a shaft (left) and the Gaussian plume inversions to derive the emission rates and related dispersion parameters (right).

3.6.3 Estimated emission results

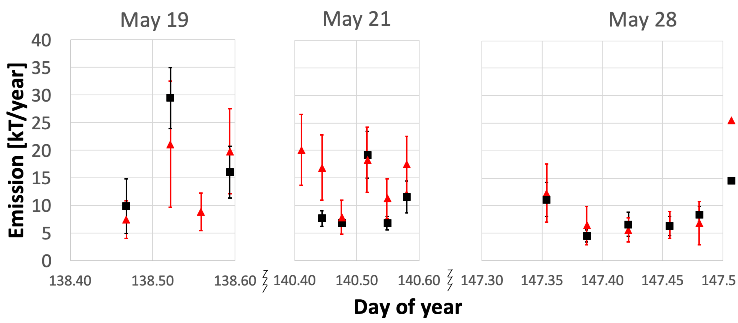

Fig. 7: The estimated emission rates of a single shaft on three different days using both mass balance and Gaussian plume inversion approaches; red: Mass balance approach, black: inverse Gaussian approach

As an example, we show the estimated CH_4 emission rates from a single shaft on three different days in May 2018 (Fig. 7). The emission rates were estimated using both mass balance and Gaussian plume inversion approaches. Most of the estimates from the two approaches agree within their uncertainties. The estimated magnitude change over different days suggests that there are significant temporal variations in the shaft emissions.

4. Conclusion and possible impact

The campaign organized in May / June 2018 was attended by 7 MEMO² ESRs, another 3 were involved by contributing model input or provided with samples. Estimations of CH_4 release from coal mine excavation were approached by different methodologies. It was the first large scale international campaign connected with this area. The ESRs had very good opportunity to train and develop their experimental skills as well as gain the field experience during the campaign. It was a very successful experience also regarding cooperation between the ESRs and other scientists. There were 5 other teams operating over USCB at the same time: Aircarft team, stationary FTIR, mobile FTIR, lidar team and wind profiling team. The results of the campaign will lead to several publications that will be completed by the end of the MEMO² project.

5. Dissemination & Exploitation

The results of the campaign were already used in many presentations at various conferences and meetings, e.g. at

- HALO SPP Kolloquium, Munich, Germany 2018
- IG3IS/TRANSCOM workshop “Inverse modelling of greenhouse gas fluxes from atmosphere observations”, Lund, Sweden 2018
- 3rd ICOS Science Conference, Prague, Czech Republic, 2018
- 24th COP, Katowice, Poland, 2018
- AGU 2018 / AGU 2019, Washington DC / San Francisco, USA
- CoMet meeting, Zakopane, 2019
- EGU 2019, Vienna, Austria, 2019
- NCGG 2019, Amsterdam, Netherlands, 2019

The following publications are either published or in preparation:

- Luther, A., Kleinschek, R., Scheidweiler, L., Defratyka, S., Stanisavljevic, M., Forstmaier, A., Dandocsi, A., Wolff, S., Dubravica, D., Wildmann, N., Kostinek, J., Jöckel, P., Nickl, A.-L., Klausner, T., Hase, F., Frey, M., Chen, J., Dietrich, F., Nęcki, J., Swolkień, J., Fix, A., Roiger, A., and Butz, A.: Quantifying CH4 emissions from hard coal mines using mobile sun-viewing Fourier transform spectrometry, Atmos. Meas. Tech., 12, 5217–5230 https://doi.org/10.5194/amt-12-5217-2019, 2019

- Alina Fiehn, Julian Kostinek, Maximilian Eckl, Theresa Klausner, Michał Gałkowski, Jinxuan Chen, Christoph Gerbig, Thomas Röckmann, Hossein Maazallahi, Martina Schmidt, Piotr Korben, Jarosław Nęcki, Pawel Jagoda, Norman Wildmann, Christian Mallaun, Rostyslav Bun, Anna-Leah Nickl, Patrick Jöckel, Andreas Fix, and Anke Roiger: Estimating CH_4, CO_2, and CO emissions from coal mining and industrial activities in the Upper Silesian Coal Basin using an aircraft-based mass balance approach; submitted to Atmospheric Chemistry and Physics, acp-2020-282, 2020.
- M. Stanisavljević, J. Nęcki, P. Korbeń, H. Maazallahi, M. Menoud, S. Defratyka, K. Vinković, J. Wietzel, C. van der Veen, *Ł.* Chmura, D. Zieba, M. Schmidt, W.Wołkowicz, T. Röckmann, J. Swolkień; Methane emissions from coal mines ventilation shafts in Silesia, Poland; in preparation
- Truls Andersen, Marcel de Vries, Bert Kers, Wouter Peters, Jaroslaw Necki, Justna Swolkien, Anke Roiger, and Huilin Chen: Quantifying methane emissions from coal mining shafts using a small Unmanned Aerial Vehicle (UAV)- based system; in preparation
- M. Menoud et al., Article about the isotope composition of methane in Krakow and over USCB, in preparation

6. References

Andersen, T., Scheeren, B., Peters, W., & Chen, H. (2018). A UAV-based active AirCore system for measurements of greenhouse gases. *Atmospheric Measurement Techniques*, *11*(5). https://doi.org/10.5194/amt-11-2683-2018

7. History of the document

Version	Author(s)	Date	Changes
	J. Necki	02/03/2020	Draft
	T. Röckmann	25/03/2020	Corrections
	H. Chen	30/03/2020	Section 3.6 & editing
	J. Necki	30.03.2020	Final Corrections
	S. Walter	16.04.2020	Final corrections, editing