MEMO^2: MEthane goes MObile – MEasurements and MOdelling

Improved emission factors for different source categories from mobile measurements

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Table of contents

1. Executive Summary

Within the MEMO^2 project a new approach for innovative experimental and modelling tools was developed and applied, based on mobile analysers and Gaussian plume and dispersion models to identify and quantify methane (CH_4) emissions from local sources in Europe. ESRs received a training and performed measurements individually, in groups during secondments to other institutes or during the two intensive campaigns in Upper Silesia (2018) and Romania (2019). Within the MEMO^2 project, several methods were applied by the ESRs to transform the CH_4 concentration measurements to CH_4 emission rates: Gaussian plume modelling, other tracer method (OTM 33a), dual tracer method and mass balance method using drone measurement. The translation into emission factors is more challenging as it requires investigation into the most suitable activity factors, which was not possible since the majority of the data were only available in finalized form near the end of the project.

Nevertheless, MEMO^2 measurements present a significant extension of the database on CH_4 emission rate measurements in Europe. The results were thoroughly evaluated and interpreted, and disseminated in numerous PhD theses and scientific publications, which are summarized in this deliverable report.

CH_4 emissions from 120 oil and gas wells were determined in Romania (Korben et al., 2021, in preparation, Vincovic et al., 2021, in preparation). The measured CH_4 concentration varies between background and 1500 ppm resulting in CH_4 emissions between 0.01 to 100 g CH_4 s^{-1} for the different oil and gas wells or facilities. CH_4 emission rates from coal mining in the Upper Silesia Coal Basin (USCB) have been determined and evaluated by Stanisavljević et al., 2021 (in preparation). The measurements were performed during the CoMET/MEMO^2 campaign in May 2017 and during additional campaigns. Determined CH_4 emission rates range between 45.3 $g{\cdot}s^{-1}$ (IQR = 30.3 – 67.3) and 1772.6 $g{\cdot}s^{-1}$ (IQR = 1071.4 – 1476.4) and indicate strong variability between coal mine ventilation shafts. The rising number of operational biogas plants in the UK was the motivation of the study from Bakkaloglu et al., 2021. In total emissions of CH_4 from ten biogas plants with emission rates between 0.1 and 58.7 kg CH_4 hr^{-1}. were derived. This results in a percentage of losses relative to the calculated production rate between 0.02 and 8.1%.

The study of Vinkovic et al., 2021 (in preparation) focus on CH_4 emission rates from a dairy farm in the Netherland with a total of seventeen UAV flights (2017-2019) and several mobile van transect measurements downwind of the farm. Vinkovic et al., 2021, estimate of CH_4 emission rate from the farm to be between 2.6 – 4.7 $g{\cdot}s^{-1}$ using a mass balance approach and UAV flights. When analysing the van measurements the emission rate varies between 1.1 – 2.9 $g{\cdot}s^{-1}$ using an inverse Gaussian model.

These studies improved our knowledge on the actual emission rates of CH_4 emitters.

2. Introduction

2.1 Background

On global and continental scales, the scientific community has established in-situ greenhouse gas monitoring programs, like ICOS in Europe or WMO GAW (Global Atmospheric Watch) which provide “top-down” quantification of emissions on the country scale (Bergamaschi et al, 2018). However, emission reductions happen at the local scale where emission estimates usually rely on “bottom-up” assessments (e.g. cattle statistics, estimating leaks from landfills), which are aggregated to yield national emission inventories. Often large discrepancies occur between bottom-up and top-down estimates of emissions (Saunois et al., 2020).

Each country or region within the European Union has other major CH_4 sources as well as different management of these emitters. In the Upper Silesian Coal basin, the major CH_4 emitters are the ventilation of the coalmines, whereas in the German Ruhr Area, all coalmines are closed and the reported emissions dropped significantly. Landfill management, as well as the use of biogas plants and therefore the CH_4 emission rate differs in each country. The MEMO^2 project was ideal for identifying country-specific emission rates, but also for targeting larger CH_4 emitters such as the coal mines in Upper Silesia or oil and gas wells in Romania with coordinated campaigns.

2.2 Scope of the deliverable

Within the MEMO^2 project a new approach for innovative experimental and modelling tools was developed and applied, based on mobile analysers and Gaussian plume and dispersion models to identify and quantify CH_4 emissions from local sources in Europe. Several methods were applied by the ESRs to transform the CH_4 concentration measurements to CH_4 emission rates: Gaussian plume modelling, other tracer method (OTM 33a), dual tracer method and mass balance method using drone measurement.

The initial scope to translate CH4emission rates into improved emission factors turned out to be more challenging as it requires investigation into the most suitable activity factors, which was not possible since the majority of the data were only available in finalized form near the end of the project. Therefore, we report here on the quantification of CH4 emission rates, which can be used by the national agencies to revisit their CH4 emission factor for the corresponding sectors.

3. Methods

Within the MEMO^2 project, several methods were applied by the ESRs to transform the CH_4 concentration measurements to CH_4 emission rates. Below we give a short summary on the four methods used, namely Gaussian plume modelling, Other tracer Method (OTM 33a), Dual Tracer method and Mass balance method using drone measurement.

Most students applied methods based on **Gaussian Plume modelling** or other dispersion models. Several ESRs used a script for the Gaussian Plume approach, which was made available during our MEMO^2 workshop on plume modelling, others wrote their own program code. The model is based on a simple Gaussian equation together with Pasquill stability classes and Briggs equations for the plume reflected from the ground (Turner 1994). Gaussian plume models are simple and efficient in computation.

ESR 12 applied **a more complex model (GRAL),** which simulates the dispersion of a trace gas plume using a Gaussian plume model - formulated with a dispersion parameterization scheme based on the Monin-Obukhov Similarity Theory. This includes dispersion coefficients in the horizontal and vertical direction. On the other hand, Lagrangian forward dispersion simulations were also conducted using GRAL by prescribing the location of the source and the evolution of the plume was followed within the modelling domain with no obstacles and no topography.

CH_4 emission rates from oil and gas wells can be quantified using **the EPA other test method (OTM33A).** This method based on Gaussian dispersion and the measured concentration of methane and meteorological conditions at a stationary location in the emissions plume downwind of the emission location. The parallel CH_4 and 3-D wind measurements are performed stationary at one place during 20 - 40min, 20 - 200 m downwind a point source. During the ROMEO campaign ESR 1 and 10 applied this method to quantify the CH_4 emission rates from Romanian oil and gas wells (see section 4.1 and Korben et al., 2021).

Partner UVSQ has the equipment to apply a **tracer dispersion method**. More details can be found in the PhD Thesis of ESR5 Sara Defratyka (2021). This method was already succesfully applied to determine CH_4 emission rates from landfills (Mønster et al. 2014), gas facilities (Roscioli et al. 2015) or wastewater treatment plants (Yver Kwok et al. 2015). In the tracer dispersion method, an additional tracer gas is released with a known emission rate near to the suspected emission source. A good tracer for our studies is acetylene (C_2H_2). A mobile analyser can measure the resulting downwind enhancements of CH_4 and C_2H_2. Under the assumption that CH_4 and the tracer gas disperse in the same way, the ratio of the measured concentration and the known release rate of C_2H_2 can be used to determine the unknown CH_4 emission rate. The advantage of this method is that in contrast to dispersion methods, the dual tracer flux methods do not need knowledge of atmospheric stability and transport.

The dual tracer flux method was applied to the data collected during 1st MEMO² training school in the Netherlands. For the measurements at a compressor station ESR5 applied three different methods to calculate the emission rates, which agree well within the errors: 2.9 kg/h (tracer dispersion method), 2.6 kg/h (Polyphemus model) and 3.2 kg/h (GRAL model – Graz Lagrangian Model).

ESR2 applied a different approach, using **a mass balance approach in combinations with UAV measurements**. The CH_4 concentrations measured from UAV AirCore (8-15 minutes) were used to calculate the CH_4 difference between downwind and upwind of a CH_4 source. To integrate the CH_4 enhancements, data needs to be spatially interpolated to regularly spaced grids in a plane perpendicular to the prevailing wind. CH_4 emission rates from the dairy cow farm were determined using a mass balance approach.

4. Results

The following results are extracted and summarized from several publications and PhD thesis, which are drafted, under review, or published.

4.1 CH_4 emission rates from oil and gas wells in Romania

CH_4 emissions from oil and gas wells have been determined in Romania. The results are part of a paper draft prepared by ESR1 Piotr Korben at Heidelberg University (Korben et al., 2021 in preparation). The measurements were performed during the ROMEO campaign (ROmanian Methane Emissions from Oil and gas). The main campaign took place in October 2019 and covered the southern part of Romania around the cities Bucharest, Ploesti, Pitesti and Craiova. Fig. 1 shows a map of Romania, with 17 defined regions, corresponding to areas with a high density of gas and oil wells.

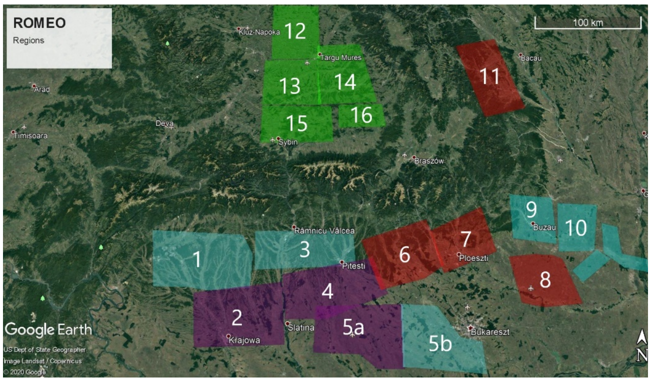

Fig. 1: Map of the interesting targeted regions for the ROMEO campaign. During the main campaign, MEMO² partners covered the regions 2, 4, 5a, 6, 7 and 8.

Mobile CH_4 measurements from regions 2, 4, 5a, 6, 7and 8 were carried out by 3 research groups: Institute of Environmental Physics of Heidelberg University (UHEI, Germany), Faculty of Physics and Applied Computer Science of AGH University of Science and Technology in Cracow (AGH, Poland) and Institute for Marine and Atmospheric research Utrecht of Utrecht University (IMAU, Netherland).

These teams were supported with cars and expertise from INCAS (National Institute for Aerospace Research "Elie Carafoli") and UBB (Babes-Bolyai University).

Emission rates were calculated from mobile CH_4 and wind measurements using the Other Test Method 33a (OTM-33a) from U.S. Environmental Protection Agency (EPA) and Gaussian Plume Method (GPM). These methods were used to determine emissions from 112 oil and gas wells and facilities. In total CH_4 emission rates from 120 oil and gas wells were quantified using OTM-33a and 20 using GPM. Using additional equipped mobile devices nearly 1050 facilities potential CH_4 emitters were previously screened. This large number of screening data was used to calculate the percentage of oil and gas facilities with emissions below detection limit, which account to 35 %. The measured CH_4 concentration varies between background and 1500 ppm resulting in CH_4 emissions between 0.01 to 100 g CH_4 / s.

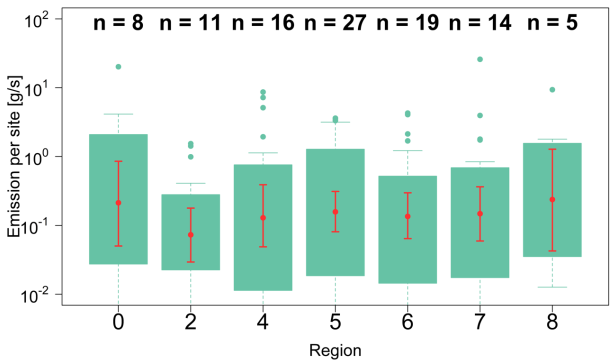

Fig. 2: Methane emission rates from oil and gas wells in different regions in Romania.

Fig. 2 shows averaged CH_4 emission rates by region. Boxes represents the first and third quartile of the data, while whiskers extend to the largest value that is within 1.5 times the interquartile range (IQR). Means and 95 % CIs are shown in red and were calculated using a nonparametric bootstrap technique. Data are presented on a logarithmic y-axis. The numbers of accepted quantifications per region are given as n. This graph includes already the non-detects taking into account the emission rates below the detection limit. More details can be found in the paper of Korben et al. (2021, in preparation) which will be submitted in May 2021.

4.2 CH_4 emission rates from coal mines in Upper Silesia (Poland)

CH_4 emission rates from coal mining ventilation shafts in the Upper Silesia Coal Basin (USCB) were determined from mobile measurements in the downwind emission plumes. The results are part of a paper draft prepared by ESR3 Mila Stanisavljević at AGH – University of Science and Technology, Krakow, Poland (Stanisavljević et al., 2021 in preparation). The measurements were performed during the CoMET/MEMO^2 campaign in Mai 2017 and during additional campaigns.

In the study of Stanisavljević et al. (2021) an improved understanding of CH_4 emission rates from the coal mining industry is presented. The analysis covers two main aspects: Quantification of CH_4 emission rates and the determination of CH_4 isotopic signatures at the source level. In this deliverable report we focus on the CH_4 emission rates from mine shafts.

CH_4 concentration was measured with a car equipped with a CRDS (cavity ring-down spectroscopy) analyser, and a GPS receiver. Basic meteorological data were simulated by the WRF- Chem v3.9 model (Weather Research Forecast Greenhouse Gas simulation). A Gaussian plume model was used to estimate the CH_4 emission rates from the different coal mine ventilation shafts (see Fig. 3, red circles). The emission rates with corresponding uncertainties of 12 surveyed coal mine ventilation shafts are shown in Fig. 4. The study covered part of the coal mine ventilation shafts operating in the Polish part of the USCB. The derived emission rates are presented as median values with corresponding uncertainties. Determined CH_4 emission rates range between 45.3 $g \cdot s^{-1}$ (IQR = 30.3 – 67.3) and 1772.6 $g \cdot s^{-1}$ (IQR = 1071.4 – 1790) and indicate strong variability between coal mine ventilation shafts.

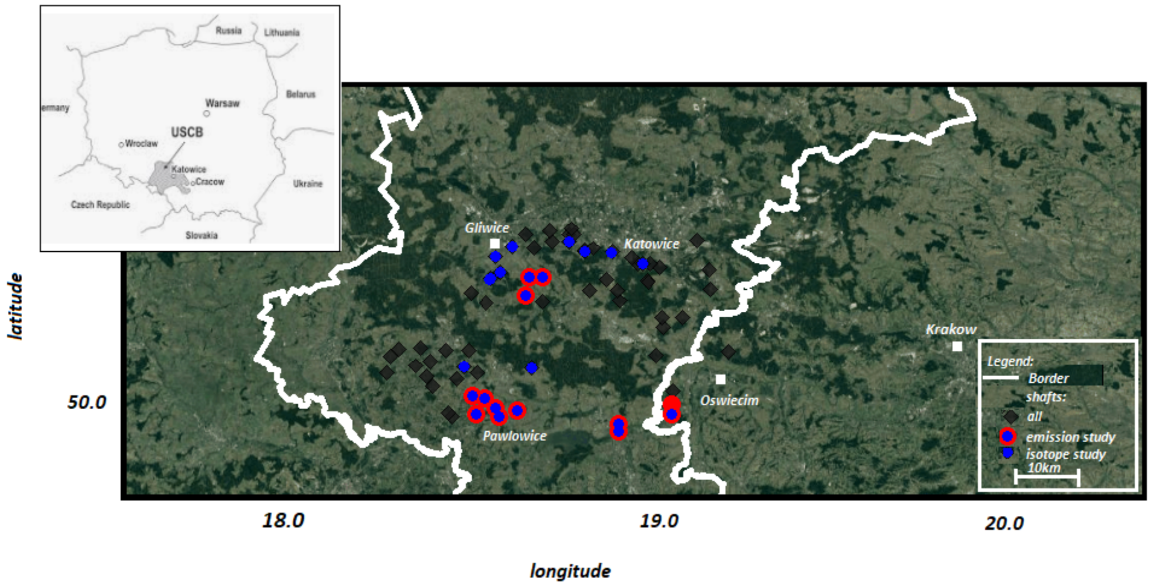

Fig. 3: Map of the USCB in southern Poland (see small upper left panel). The coal shafts are shown as red (studied shafts) or blue (active but not studied shaft) dots on the map (Figure from Stanisavljević et al., 2021).

Variations in the CH_4 emission rates can also be observed within the same coal mine facilities. The Pniowek coal mine has three ventilation shafts with CH_4 emission rates of 57.8 (IQR = 46.5 – 77.1) g·s^{-1}, 86.37 (IQR = 57.6 – 130.1) g·s^{-1}, and 180.4 (IQR = 115.2 – 340.1) g·s^{-1}, respectively.

This variability within a facility, and even from a certain shaft during different times, illustrates that emissions from coal mining shafts are not continuous, which implies that a comparison to monthly or annual average numbers reported in the inventories is not straightforward.

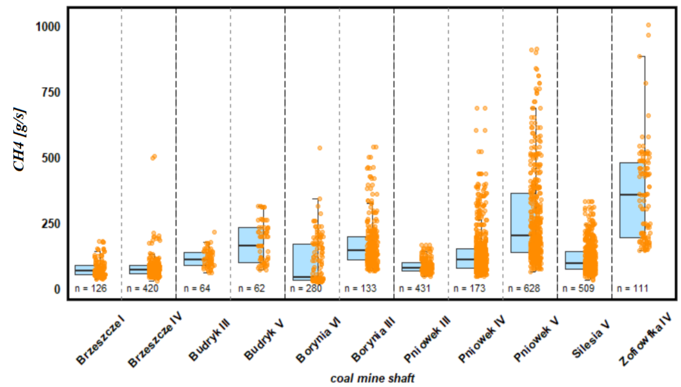

Fig. 4: Distribution of CH_4 emission rates with corresponding uncertainties for 11 individual coal mining shafts in the USCB. Due to better visibility, one studied coal mine shaft is not illustrated here. Vertical grey dashed lines indicate separation between different coal mine shafts, while n denotes the number of GPM simulations per site (Figure from Stanisavljević et al., 2021).

4.3 CH_4 emissions rates from one landfill and two gas compressor stations close to Paris

The study of Defratyka (2021) quantified the CH_4 emission rate of two gas compressor stations and one landfill in the Ile de France region (Paris, France).

The CH_4 emission rates from two gas compressor stations varies between (0.55 ± 0.2) kg CH_4 h^{-1} for site C and (2.5 ± 0.5) kg CH_4 h^{-1} for site A. Both gas compressor stations have two compressors and use the same technology. For gas compressor stations, estimated emission rates vary depending on maintenance work inside facilities (Zavala-Araiza et al. 2015; Saunois et al. 2020). Based on the study made by Subramanian et al. (2015) gas facilities emit methane both during standby and operating modes. Additionally, more CH_4 is emitted during irregular events like uncontrolled f venting, which can change over a year and usually is not included in inventories

Defratyka (2021) determined the emission rate of landfill D using the racer release method with (62 ± 13) kg CH_4 h^{-1}. Compared to previous studies (Ars 2017), the emission rate is lower. It seems to be in agreement with the operating company's which reported to reduce CH_4 emissions from the landfill. Compared to the first survey, made in 2016, the estimated emission rate has been divided by four. Observed reduction is likely an effect extension of the gas collection system (installed between September and November 2016) and a coverage with geomembranes (installed between November and December 2016). The study of Defratyka (2021) conclude that more campaigns are necessary to confirm this finding.

4.4 CH_4 emission rates from biogas plants in UK

ESR 7 Semra Bakkaloglu from RHUL London measured and quantified CH_4 emission rates from 10 biogas plants in UK. This study is published in the journal Waste Management (Bakkaloglu et al., 2021). The main results are summarized below.

The rising number of operational biogas plants in the UK brings a new emissions category to consider for CH_4 monitoring, quantification and reduction. Mobile greenhouse gas surveys were conducted to detect plumes of CH_4 emissions from the biogas plants in southern England that varied in their size, waste feed input materials and biogas utilization. This data was analysed using Gaussian plume modelling. In total emissions of CH_4 from ten biogas plants based on repeat passes through the plumes were derived. CH_4 emission rates ranged from 0.1 to 58.7 kg CH_4 hr^{-1}. This results in a percentage of losses relative to the calculated production rate between 0.02 and 8.1 %. The average emission rate was 15.9 kg CH_4 hr^{-1}, and the average loss was 3.7 %. In general, CH_4 emission rates from smaller biogas plants at individual farms were higher than from larger food waste biogas plants.

The authors suggest that biogas CH_4 emissions may account for between 0.4 and 3.8 %, with an average being 1.9 % of the total CH_4 emissions in the UK excluding the sewage sludge biogas plants.

Table 1: Estimated CH_4 emissions rates obtained from Gaussian plume modelling, CH_4 losses relative to calculated production rates and emission factors calculated as annual emission rates divided by annual feedstock amount (Bakkaloglu et al., 2021)

Name	Biomethane capacity (Nm^3/hr)	Calculated average CH_4 production rate (kg CH_4 h^{-1})	Estimated total CH_4 emissions (kg CH_4 h^{-1})	CH_4 loss relative to calculated production rates (%)	Emission factors (kg CH_4 emitted/ tonnes of feedstock)
A	N/A	970[b,c]	12.6 ± 3.8	1.3 ± 0.4	2.5 ± 0.7
B	N/A	861 [a,c]	58.7 ± 25	6.8 ± 2.9	10.3 ± 4.4
C	N/A	654 [a,c]	0.1 ± 0.02	0.02 ± 0.003	0.02 ± 0.004
D	990	709	2.8 ± 0.8	0.4 ± 0.1	0.5 ± 0.1
Plant average CH_4 loss and EF, **food waste**: 2.1% and 3.3, respectively					
Production weighted average CH_4 loss and EF, **food waste**: 2.3% and 3.4, respectively					
E	550	394	21.9 ± 6.2	5.6 ± 1.6	10.0 ± 2.8
F	N/A	425 [a]	14.3 ± 4.2	3.4 ± 1.0	6.3 ± 1.8
G	N/A	215 [a,c]	17.5 ± 3.7	8.1 ± 1.7	15.3 ± 3.2
H	N/A	198 [a]	0.5 ± 0.1	0.3 ± 0.1	1.5 ± 0.3
I	N/A	439 [a,c]	14.0 ± 3.9	3.2 ± 0.9	2.2 ± 0.6
J	N/A	209 [a,c]	16.6 ± 4.1	7.9 ± 0.02	11.4 ± 2.8
Plant average CH_4 loss and EF, **farm waste**: 4.8% and 7.8, respectively					
Production weighted average CH_4 loss and EF, **farm waste**: 4.5% and 6.1, respectively					
All biogas plants					
Plant average CH_4 loss and EF, all: 3.7% and 6.0, respectively					
Production weighted average CH_4 loss and EF, all: 3.1% and 4.4, respectively					

4.5 CH_4 emission rates from dairy cows using an UAV based active Air-Core system and mobile van

The study of Vinkovic et al., 2021 aimed to quantify the agricultural CH_4 emissions, especially from dairy cattle. In the Netherlands, these account for 69 % of the total emissions in the Netherlands, in 2017. From March 2017 to March 2019, four measurement campaigns were carried out on a dairy cow farm

~20 km north-west of the city of Groningen, in the northern part of the Netherlands. In March 2019, CH_4 and N_2O mole fractions were obtained using two different mobile platforms, a mobile van and an unmanned vehicle (UAV) based active AirCore system.

Fig. 5 shows the dairy farm with different stables and its surroundings. A tracer release experiment was performed on the farm in March 2019, using a pure N_2O cylinder with a release rate of 0.08 g/s. Furthermore, CO_2, CO, H_2O, C_2H_6, relative humidity, ambient temperature and pressure were measured in addition to CH_4 and N_2O mole fraction measurements. A total of seventeen UAV flights (2017-2019) and several mobile van transect measurements were performed downwind of the farm at a distance between 108 and 298 metres. Vinkovic et al.,(2021) estimate of CH_4 emission rate from the farm between 2.6 – 4.7 g/s using a mass balance approach and UAV flights. When analysing the van measurements the emission rate varies between 1.1 – 2.9 g/s using an inverse Gaussian model. The N_2O release rate was estimated using mass balance approach and an inverse Gaussian model to be 0.088 g/s and 0.086 g/s, respectively.

Fig. 5: The dairy farm and its surroundings. Stable (1) for adult lactating cows, stable (2) for young cows. The flight location of the downwind flights performed on 29 March 2019. The red arrow indicates the wind direction, the red circles specify the location of two 3D sonic anemometers, and the green circle the N_2O release location. The green lines indicate the projected flight tracks of the UAV, and the blue line indicates the driving path of the mobile van.

The final analysis of this data is still in progress and will be submitted before the end of the PhD thesis of ESR2 Katarina Vinkovic.

5. Conclusion and possible impact

The studies performed in the frame of MEMO^2 improved significantly our knowledge on CH_4 emission rates from several anthropogenic emitters. Intensive campaigns with 4-5 devices enable a representative coverage of many individual point sources in a larger region. The studies described in section 4 are either already published (Bakkaloglu et al., 2021, Defratyka (2021) or close to submission (Korben et al, 2021, Stanisavljević et al. 2021, Vinkovic et al., 2021) to international journals.

After the papers are accepted, the quantified CH_4 emission rates can be used by the national agencies to revisit their CH_4 emission factor for the corresponding sectors.

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Vinkovic, Katarina, Truls Andersen, Marcel de Vries, Bert Kers, Steven van Heuven, Wouter Peters, Arjan Hensen, Huilin Chen (2021) Quantification of methane emissions from dairy cows using an Unmanned Aerial Vehicle (UAV) based active AirCore system and mobile van (paper draft in preparation)