MEMO^2: MEthane goes MObile – MEasurements and MOdelling

Sylvia Walter, Thomas Röckmann, and the MEMO^2 team

Sylvia Walter, Utrecht University, s.walter@uu.nl
Thomas Röckmann, Utrecht University, t.roeckmann@uu.nl

Semra Bakkaloglu, Royal Holloway and Bedford New College, semra.bakkaloglu@rhul.ac.uk
Hermann W. Bange, Helmholtz Centre for Ocean Research Kiel, hbange@geomar.de
Philippe Bousquet, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), philippe.bousquet@lsce.ipsl.fr
Gregoire Broquet, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), gregoire.broquet@lsce.ipsl.fr
Dominik Brunner, Swiss Federal Laboratories for Materials Science and Technology, Dominik.Brunner@empa.ch
Huilin Chen, University of Groningen, huilin.chen@rug.nl
Sara Defratyka, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), sara.defratyka@lsce.ipsl.fr
Tim de Groot, Royal Netherlands Institute for Sea Research, tim.de.groot@nioz.nl
Hugo Denier van der Gon, Netherlands Organisation for applied scientific research, hugo.deniervandergon@tno.nl
Lukas Emmenegger, Swiss Federal Laboratories for Materials Science and Technology, Lukas.Emmenegger@empa.ch
Julianne Fernandez, Royal Holloway and Bedford New College, Julianne.Fernandez.2018@live.rhul.ac.uk
Rebecca Fisher, Royal Holloway and Bedford New College, r.fisher@es.rhul.ac.uk
Arjan Hensen, Netherlands Organisation for applied scientific research, arjan.hensen@tno.nl
Bill Hirst, Shell, bill.hirst@shell.com
Magdalena Hofman, Picarro, mhofmann@picarro.com
Jutta Holst, Lund University, jutta.holst@nateko.lu.se
Piotr Korbeń, Heidelberg University, pkorben@iup.uni-heidelberg.de
Maarten Krol, Wageningen University, maarten.krol@wur.nl
Patryk Łakomiec, Lund University, patryk.lakomiec@nateko.lu.se
David Lowry, Royal Holloway and Bedford New College, d.lowry@es.rhul.ac.uk
Hossein Maazallahi, Utrecht University, h.maazallahi@uu.nl
Malika Menoud, Utrecht University, m.menoud@uu.nl
Randulph Morales, Swiss Federal Laboratories for Materials Science and Technology, randulph.morales@empa.ch
Jaroslaw Necki, AGH University of Science and Technology, necki@agh.edu.pl
Euan Nisbet, Royal Holloway and Bedford New College, e.nisbet@es.rhul.ac.uk
Hans Oonk, OONKAY, hans@oonkay.nl
Jean-Daniel Paris, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), jean-daniel.paris@lsce.ipsl.fr
Isabelle Pison, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), isabelle.pison@lsce.ipsl.fr
Jonas Ravelid, Swiss Federal Laboratories for Materials Science and Technology, jonas.ravelid@empa.ch
Anja Raznjevic, Wageningen University, anja.raznjevic@wur.nl
Janne Rinne, Lund University, janne.rinne@nateko.lu.se
Rod Robinson, National Physical Laboratory, rod.robinson@npl.co.uk
Alessandro Sarno, Avfall Sverige,alessandro.sarno@gotland.se
Marielle Saunois, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), marielle.saunois@lsce.ipsl.fr
Martina Schmidt, Heidelberg University, Martina.schmidt@iup.uni-Heidelberg.de
Mila Stanisavljevic, AGH University of Science and Technology, mila.stanisavljevic@gmail.com
Barbara Szenasi, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), barbara.szenasi@lsce.ipsl.fr
Danielle van Dinther, Netherlands Organisation for applied scientific research, danielle.vandinther@tno.nl
Chiel van Heerwaarden, Wageningen University, chiel.vanheerwaarden@wur.nl
Ilona Velzeboer, Netherlands Organisation for applied scientific research, ilona.velzeboer@tno.nl
Katarina Vinkovic, University of Groningen, k.vinkovic@rug.nl
Felix Vogel, Environment and Climate Change Canada, felix.vogel@canada.ca
Wojciech Wolkowicz, Polish Geological Institute, wwol@pgi.gov.pl
Camille Yver-Kwok, University of Versailles Saint-Quentin-en-Yvelines (UVSQ), camille.yver@lsce.ipsl.fr

Keywords: methane, greenhouse gas emissions, H2020 MSCA ITN-ETN

Abstract

CH_4 emissions are a major contributor to Europe's global warming impact and emissions are not well quantified yet, although this is indispensable knowledge to reach the targets of 2015 United Nations Climate Change Conference in Paris (COP21) and the required massive reductions of greenhouse gas emissions. There are significant discrepancies between official inventories of emissions and estimates derived from direct atmospheric measurement, and effective emission reduction can only be achieved if sources are properly quantified and mitigation efforts are verified.

MEMO^2 is a H2020 MSCA European Training Network with more than 20 collaborators from 7 countries and will contribute to the targets of the EU with a focus on methane (CH_4). The goal of the project is to bridge the gap between large-scale scientific estimates from in situ monitoring programs and the 'bottom-up' estimates of emissions from local sources that are used in the national reporting by the combination of I) developing and deploying new and advanced mobile methane (CH_4) measurements tools and networks, II) isotopic source identification, and III) modelling at different scales. This paper will give a brief overview of the project and its results achieved during the first two years.

1. Introduction

Mitigation of climate change is a key scientific and societal challenge, and of pivotal societal and public interest. The 2015 United Nations Conference of the Parties in Paris (COP21) agreed to limit global warming "well below" 2ºC and, if possible, below 1.5ºC. Reaching this target requires massive reductions of greenhouse gas emissions, far beyond the intended Nationally Determined Contributions (NDCs). In this context, achieving significant reduction of greenhouse gas emissions is a logical headline target of the EU climate action [1], which envisages as one of the key targets for the year 2020 a “20 % cut in greenhouse gas emissions from 1990 levels”. In addition, the Sustainable Development Goal (SDG) nr. 13 of the 2030 Agenda for Sustainable Development, implemented in 2015 by the United Nations, aims to “take urgent action to combat climate change and its impact”. In this context of urgent required massive greenhouse gas emission reductions, CH_4 is a promising target. CH_4 is the second most important greenhouse gas after CO_2, its emissions are a major contributor to Europe's global warming impact and it is one of Europe’s most important sources of energy. With a global warming potential (cumulative forcing over 20 years) of 84, a rather short lifetime of 12.4 years [2] and several sources such as landfills, gas leaks and manure offering possibilities of “no-regret” emission reduction, a reduction of CH_4 can make a significant contribution to climate change mitigation actions. CH_4 emission reductions are more cost-effective than most CO_2 emission reduction measures and will lead to quicker gains in reduction of greenhouse gas radiative forcing.

However, effective emission reduction can only be achieved if sources are properly quantified, and mitigation efforts are verified. Europe's CH_4 emissions are yet not well quantified. There are significant discrepancies between official inventories of emissions and estimates derived from direct atmospheric measurement. New advanced combinations of measurement and modelling are needed to achieve reliable emission quantification. The H2020 European Training Network (ETN) MEMO^2 (MEthane goes MObile – MEasurements and MOdelling, https://h2020-memo2.eu) aims to bridge the gap between large-scale scientific estimates from in situ monitoring programs and the 'bottom-up' estimates of emissions from local sources that are used in the national reporting. As an ETN, MEMO^2 aims not only on scientific excellence but also on the combination with training of early stage researchers (ESRs). So MEMO^2 has two goals:

The main scientific goal of MEMO^2 is to develop and apply innovative experimental and modelling tools, based on recently developed mobile analysers, on state-of-the-art isotope techniques, and on a hierarchy of models, including newly developed high-resolution dispersion models, to identify and quantify CH_4 emissions from local sources in Europe and use these updated emissions to improve estimates at the European scale. These tools will enable improved and objective verification of CH_4 emission reduction strategies for specific source sectors.

The second goal is based on the complexity and interdisciplinary character of detecting and quantifying CH_4 emissions, and the evaluation of climate mitigation measures. It requires skilled scientists with high-level of theoretical and practical competences that are able to cooperate in networks. So MEMO^2 developed and implemented a dedicated research training program, which follows a holistic approach to stimulate key competences and knowledge exchange, aiming at the education of a generation of “cross–thinking” scientists. The training includes activities on local, individual, network-wide and international level. As a training network, MEMO^2 fosters the education of qualified scientists in the use and implementation of interdisciplinary knowledge and techniques that are essential to meet and verify emission reduction goals.

By this, MEMO^2 also contributes to associated targets of SDG 13, which focus e.g. on the improvement of education and awareness-raising (https://sustainabledevelopment.un.org/sdg13).

2. MEMO2

2.1 General set-up

MEMO^2 is an international and interdisciplinary project, with 9 academic and 16 non-academic partners, and makes synergistic use of highly specialized competencies and facilities of these partners from the fields of atmospheric physics and chemistry, environmental sciences, meteorology, and metrology. This includes atmospheric and isotopic measurement facilities, mobile measurement equipment, UAV and

AirCores and several modelling facilities to increase the overall scientific quality and societal impact. The research program comprises of three scientific work packages (WPs), which are strongly interconnected (Fig. 1).

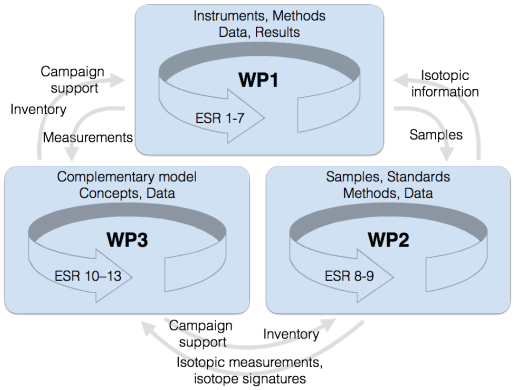

Fig. 1: Interconnection scheme of the 3 scientific Work Packages of MEMO^2

WP1 is dedicated to mobile measurements across Europe. In WP2 state-of-the-art isotope techniques are used to attribute observed CH_4 elevations to individual sources. The translation of these CH_4 elevations into emissions and to integrate local measurements from WP1 and WP2 to the European scale is the task within WP3. The scientific WPs share a common objective and complement each other by detecting (WP1), attributing (WP2) and quantifying (WP3) CH_4 emissions in Europe using measurements on mobile platforms.

The geographic locations of the partners provide excellent opportunities to characterize important CH_4 source categories around Europe, such as e.g. agriculture and gas industry in the Netherlands, landfills in UK, city emissions in France and Germany or coal mining in Poland.

2.2 General scientific and methodological approach

CH_4 measurements within MEMO^2 span the full range from high-precision flask samples for isotope analysis, to continuous time series using laser spectroscopy, and airborne measurements by vehicles, airplanes and drones, which will allow in-situ CH_4 monitoring in all three dimensions. The modelling activities allow the development of new modelling concepts, covering European, regional and local scales by such diverse techniques as inversion of Lagrangian Particle Dispersion Models and Large Eddy Simulations. Another benefit of the network is the opportunity to perform joint field campaigns and intercomparison campaigns.

On global and continental scales, the scientific community assesses atmospheric CH_4 by *in situ* monitoring programs, e.g., the ICOS ESFRI infrastructure in Europe and the UN's Global Atmosphere Watch [3, 4]. This provides “top-down” quantification of emissions on a large scale (e.g. Germany, France, UK), but is by design not sensitive to local emissions from individual sources [5, 6, 7]. In contrast, 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 [8]. Often large discrepancies occur between bottom-up and top-down estimates of emissions [9, 10]. Mitigation legislation drives reductions in reported emissions of CH_4, e.g. emissions from landfills. However, such reductions are mostly reported by bottom-up assessment, but not independently confirmed by top-down measurements and models. The concept “trust but verify” can only be applied if adequate verification tools are available, which is not the case yet for most greenhouse gases and ozone–depleting gases.

Current approaches to estimating CH_4 sources at the EU-level use both bottom-up and top-down methods [7, 9]. Bottom-up estimates rely on emission reporting, in which various sources are integrated into emission totals per country based on emission factors and activity magnitudes. These estimates are uncertain, partly because of a lack of observations to constrain the emission factors. The top-down approach usually starts with the bottom-up emission inventories as a prior estimate, and optimally adjusts the sources to make the emissions consistent with CH_4 observations. This approach requires a transport model to translate emissions into atmospheric concentration fields that can be compared to observations. Top-down approaches are limited by the density of atmospheric observations, by the quality of the transport model, but also by the quality of the prior estimate of emissions [4]. Here, scale issues become important. Local measurements close to sources are hard to reproduce by coarse-scale (> 10 km) models. Therefore, top-down approaches normally employ only “background” measurements that are considered representative for larger geographical domains. On the other hand, the model-

calculated concentrations cannot be attributed to individual sources at the (local) scale of the emissions. As a result, the information exchange is partial and mainly one-way: from the (uncertain) inventories to the atmospheric concentrations. Feedbacks from the larger-scale model calculations to the emission inventories, and integration of local scale emission factors into inventories remain both limited.

The approach of the MEMO^2 research program is to use innovative measurements and modelling of CH_4 using mobile platforms as principle tool to bridge the current scale gaps between local measurements, emission inventories, and European scale modelling.

2.3 Mobile measurements

Mobile measurements of CH_4 emissions are available since a few years [11]. The interpretation of such campaign results is challenging due to several factors which could impact the measurements, e.g. the spatial distribution of sources, measurement conditions (e.g. distance to the source, speed of the vehicles), changing emission rates and emissions-weighted distributions, or plume diluting atmospheric conditions.

The key measurement components are fast and accurate analysers on mobile platforms. Analysers used are various CRDS models and OA-ICOS to measure CH_4, CO_2, $^{13}CH_4$, $^{13}CO_2$, C_2H_6, H_2O, C_2H_2 or CO. The main experimental platform are cars, but also unmanned aerial vehicle (UAV) platforms and light aircrafts are used to investigate focus source types such as wetlands, landfills, city emissions, lakes, gas leaks, agricultural emissions, and mining emissions. By this MEMO^2 maps the small-scale distribution of CH_4 across Europe, and identify and quantify CH_4 emissions at the local scale [11] and provide emission factors for further modelling activities. The CH_4 source mix is different per country, and - based on the inventories - MEMO^2 targets the largest uncertainties in the individual countries. A key advantage of the network is that due to close cooperation the regional/national scale but also the European scale is covered.

2.4 Isotopic measurements

Different sources emit CH_4 with slightly different isotopic composition [12]. So, measuring the isotopic composition of CH_4 helps to identify the sources responsible for observed elevations of CH_4 in the atmosphere and improves the understanding of the temporal and spatial variability of isotopic signatures of CH_4 emissions. Its helps to verify emission inventories and to distinguish CH_4 sources in complex environments with many overlapping sources, such as cities. The link to the development of UAV sampling methods and modelling allows identifying possible vertical as well as horizontal variability in isotopic signature in emission plumes. The information can be used to provide novel EU-wide “isotopic source signature maps” of the most important CH_4 sources, and give important input for the use of isotope information in atmospheric models.

2.5 Modelling approaches

The quantification of emissions from concentration measurements and isotopic compositions requires complementary modelling tools applicable on various scales, from the local scale of an intensive measurements campaign up to the EU and global scale [13]. At the local scale, Large Eddy Simulations (LES) are employed to predict and analyse detailed dispersion patterns from local sources. Virtual vehicles and UAVs are used to sample simulated 3D dispersion fields from CH_4 sources. On the regional scale, flow patterns integrating mixed sources from e.g. a city are analysed using regional high-resolution modelling. Combined, these approaches allow optimal interpretation and usage of the measurement results. At the European scale, forward simulations of CH_4 concentrations and top-down emission estimates are derived, using detailed and updated bottom-up emissions maps. This joint bottom-up and top-down activity links the modelling and the measurements, and specifically includes the improvement of European CH_4 inventories. Modelling activities also assist in the design of measurement strategies. The goal is to determine areas where measurements will have the largest benefit concerning both uncertainty of emissions and possibilities of mitigation measures.

3. Results

3.1 Mobile measurements of CH_4

Measurement campaigns are an essential component of projects such as MEMO[2]. Within the first two years of the project more than 150 days of measurement campaigns were performed. Fig. 2 gives an overview of (joint) sampling locations.

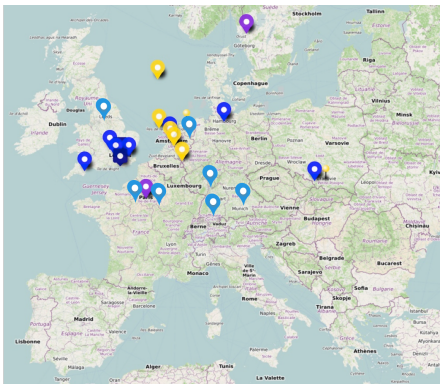

Fig. 2: Overview of (joint) measurement campaigns, the colour codes indicate the type of samples

As one example, in May 2017 and June 2018, we participated in the CoMet (Carbon dioxide and Methane mission) campaigns in Upper Silesia, which were (co)organized by the German Aerospace Center (DLR) and the University of Science and Technology (AGH). CoMet aimed at industrial emission of CH_4 with a priority on mining activities over Silesia as one of the European anthropogenic CH_4 hotspots. In this region 33 mines are active, but also additional methane sources are present: landfills, cities gas networks, cow farms, wetlands and agriculture. Several teams deployed in-situ and remote sensing instruments on aircraft as well as on ground, performed measurements using mobile platforms (CRDS analyser in cars, planes and an active AirCore system on a drone [14]) and applied FTIR technique with stationary and mobile platform. Preliminary results show, that CH_4 in general mainly originates from thermogenic sources, but with significant differences between the mine shafts, not only regarding mole fractions but also the isotopic composition of CH_4. (Fig. 3, Fig. 4). This might indicate either different origins of methane gas, different levels of coal excavation inside the mines or different types of ventilation.

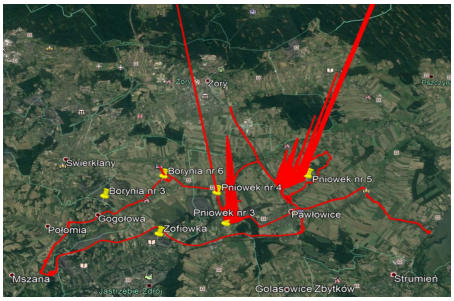

Fig. 3: CH_4 concentration measurements in Upper Silesian coal mining region.

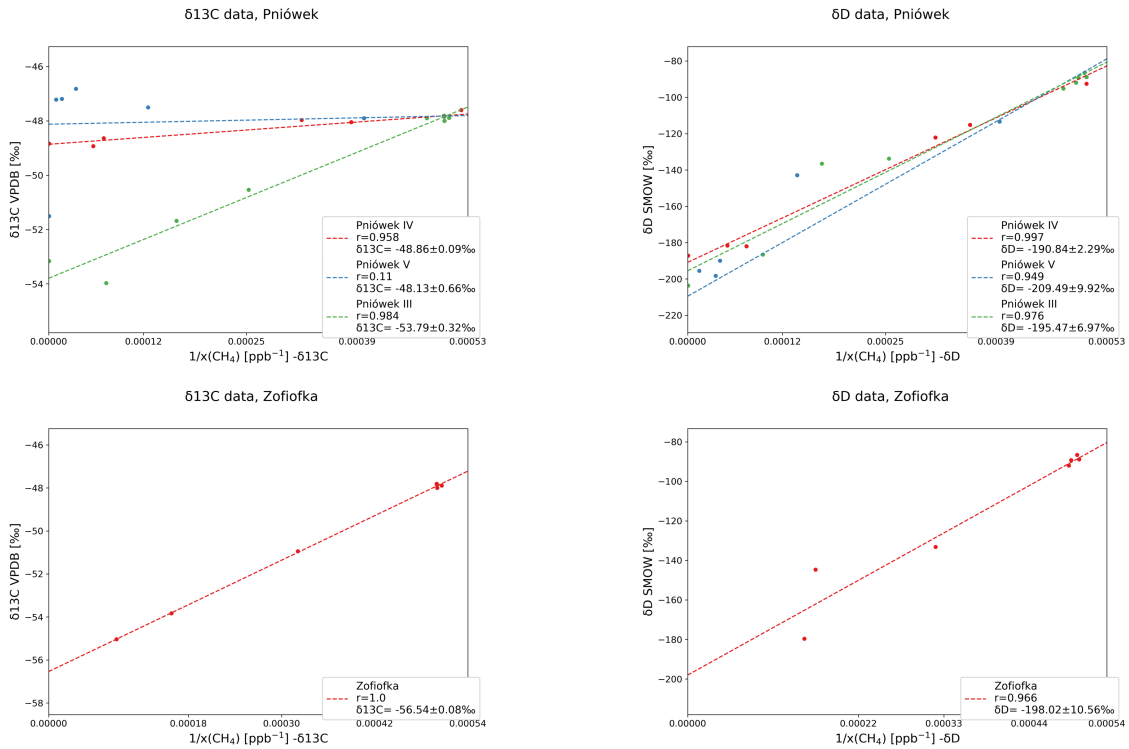

Fig. 4: δD and $\delta^{13}C$ of shafts in the Silesian region

The exhaust shafts of the mines are delivering air from different levels of the mines to the atmosphere, where CH_4 can have different origins. According to the geological history of the coal beds and the layers above them, the coal also has less or more methane accumulated. Thus, the same exhaust shaft can provide different amounts of CH_4 with different isotopic signatures of it during our observations. The amount of CH_4 released depends on the distance from the ridge (crack of rock bed) and the isotopic composition strongly depends on the depth of the coal bed excavated.

As successful measurement campaigns depend on excellent and suitable equipment, MEMO^2 also develops new instrumentation such as a lightweight high-precision mid-IR methane laser spectrometer for unmanned aerial vehicles (UAV) (Fig. 5). The spectrometer is based on a single-mode quantum cascade laser (DFB-QCL) and a circular, segmented multi-pass cell with an optical path length of 10 m [15]. This novel cell design has a compact footprint, and it achieves low optical noise and high stability against mechanical distortion. The overall instrument weighs 1.6 kg (excluding battery) and has an average power consumption of 15 W which is achieved by optimized laser driving and a system-on-chip FPGA data acquisition module [16]. The spectrometer is equipped with additional sensors for pressure, temperature, and relative humidity, as well as a GPS receiver and an optional module for real-time data transmission. Therefore, it is possible to use the device aboard any drone, regardless of its specific communication protocol.

The spectrometer reaches a precision of few ppb at 1 s time resolution and significantly below 1 ppb after 10 – 1000 s integration. It has been regularly flown on a commercial drone (DJI Matrice 600). The open-path design allows very fast sampling, and absorption spectra are measured at > 10 kHz. This gives a wide flexibility in terms of the required precision and time resolution. Ongoing field experiments explore the potential of this unique instrument for the identification, characterization and quantification of natural and anthropogenic methane sources.

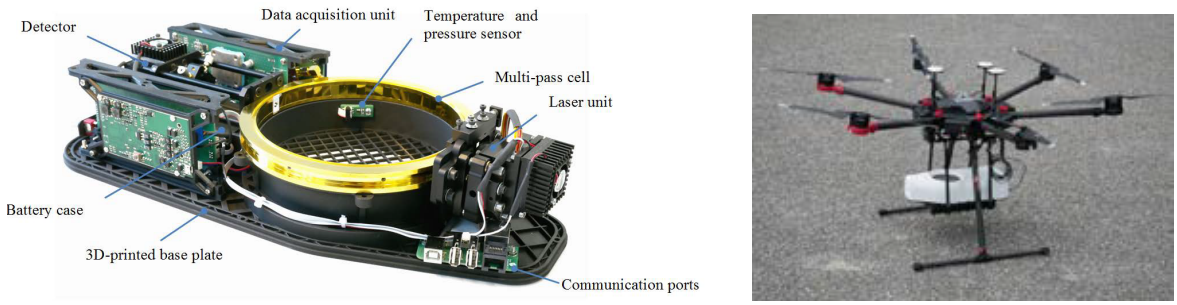

Fig. 5: Photography of the high-precision methane sensor (left side) and its mounting on a UAV (right side)

3.2 Source identification by isotopic characterization

Isotopic characterization and mapping of CH_4 sources requires that all laboratories measuring the isotopes of CH_4 be on the same scale across the range of values commonly encountered in emissions from European sources. The use of newly-developed cavity ring-down laser spectroscopy (CRDS) techniques for measurements of the $^{13}C/^{12}C$ ratio of methane ($\delta^{13}C$) allows field measurements of isotopes to a much lower precision than by IRMS, but gives near instantaneous measurements rather than later laboratory analyses to discriminate CH_4 produced by biogenic (e.g. cows), thermogenic (e.g. natural gas) and pyrogenic (combustion) sources. This can be measured on emissions directly from source where the CH_4 % is very high, such as a gas supply (95 %) or a landfill gas (50 %), but for the MEMO^2 studies many of the sources are unknown and sometimes sampled at many hundreds of meters downwind of the point of emission and measured CH_4 varies from the ambient global background (around 1.9 ppm CH_4) to about 10 ppm or even more CH_4. International isotopic standards are not available for CH_4 in air at these concentrations, however. Given that the CRDS instruments have inherently poor precision at the 2 ppm ambient background levels of CH_4, but with an improvement of an order of magnitude at 10 ppm CH_4 (from ±4 ‰ to ±0.4 ‰), we first prepared an inter-calibration with the aim of bringing these instruments onto the common isotopic scale. Results were in the range of analytical error so that the results obtained within the MEMO^2 project can be confidently interpreted, and directly compared with other results obtained globally.

Next to the intercomparison, long-term monitoring experiments were intensified and new ones started. Fig. 6 shows a 6-months' time series at Cabauw, the Netherlands [17], similar long-term monitoring experiments were executed in Krakow from October 2018 till March 2019. First results show, that the CH_4 in Krakow mainly originates from thermogenic sources, whereas CH_4 at the Dutch stations is mainly biogenic even at Lutjewad, which is near Europe´s largest gas reservoirs in the North Sea and close to Groningen. The long-term monitoring allows the identification of specific events with elevated contributions from more enriched sources such as natural gas and landfills. The results are used to compare models such as the global TM5 model and the mesoscale model FLEXPART-COSMO.

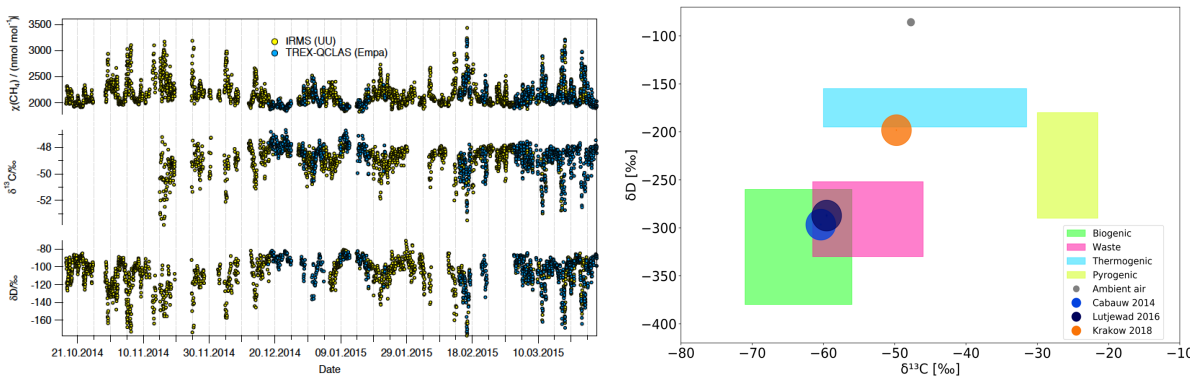

Fig. 6: 6-month time series of δD, $\delta^{13}C$, and the CH_4 mole fraction at Cabauw [16] (left), mean results of long-term campaigns in Krakow (PL), Lutjewad and Cabouw (NL) (right)

Next to the atmospheric measurements, MEMO^2 made an excursion to the marine environment. During a campaign to the North Sea in 2018, the origin of CH_4 above an active cold seep at the Doggerbank was investigated [18]. Fig. 7 shows depth profiles of $\delta^{13}C$ and δD taken during a 3-days' time series at the Doggerbank. The isotopic signature from the left-over methane that is not directly oxidized by methanotrophs during transport trough the water column indicates that at lower concentrations of methane a shift occurs in δD as well as $\delta^{13}C$. Both isotopic signals indicate that methane was from biogenic methanogenesis. Furthermore, the increase in heavy isotopes at lower concentrations is an indication that microbial methane oxidation occurs.

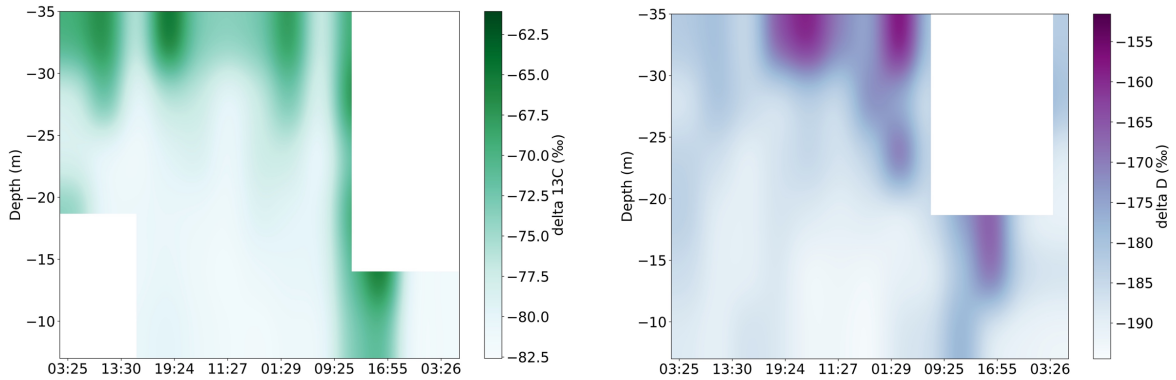

Fig. 7: $\delta^{13}C$ (left) and δD (right) during a 3-days' time series at the Doggerbank, North Sea

3.3 Modelling: A multi-scale interpretation framework for CH_4 observations

As the modelling part within MEMO^2 aims on e.g. the interpretation of mobile observations as well as on the estimation of emission fluxes at different scales, a variety of models are used, compared, and improved.

A modelling tool to assist in planning of mobile measurements and campaigns is MicroHH [19], a model which is more advanced than a simple Gaussian plume model to get information about source variability. As the interpretation of measurements as done within MEMO^2 requires some more flexibility, the MicroHH has been improved by adding a point and a line source in form of a Gaussian "ball" or "pipe"

that spans over multiple grid points and is limited by four standard deviations in order to avoid unwanted numerical behaviour of the simulation which would happen if all the mass was injected at a single grid point. The Gaussian function is normalized in a way that preserved the prescribed source strength. It is now possible to simulate multiple sources at arbitrary locations in the domain (Fig. 8). After solving the circular boundary conditions, which are desirable for the flow field, but not for CH_4, the model is ready to interpret observations.

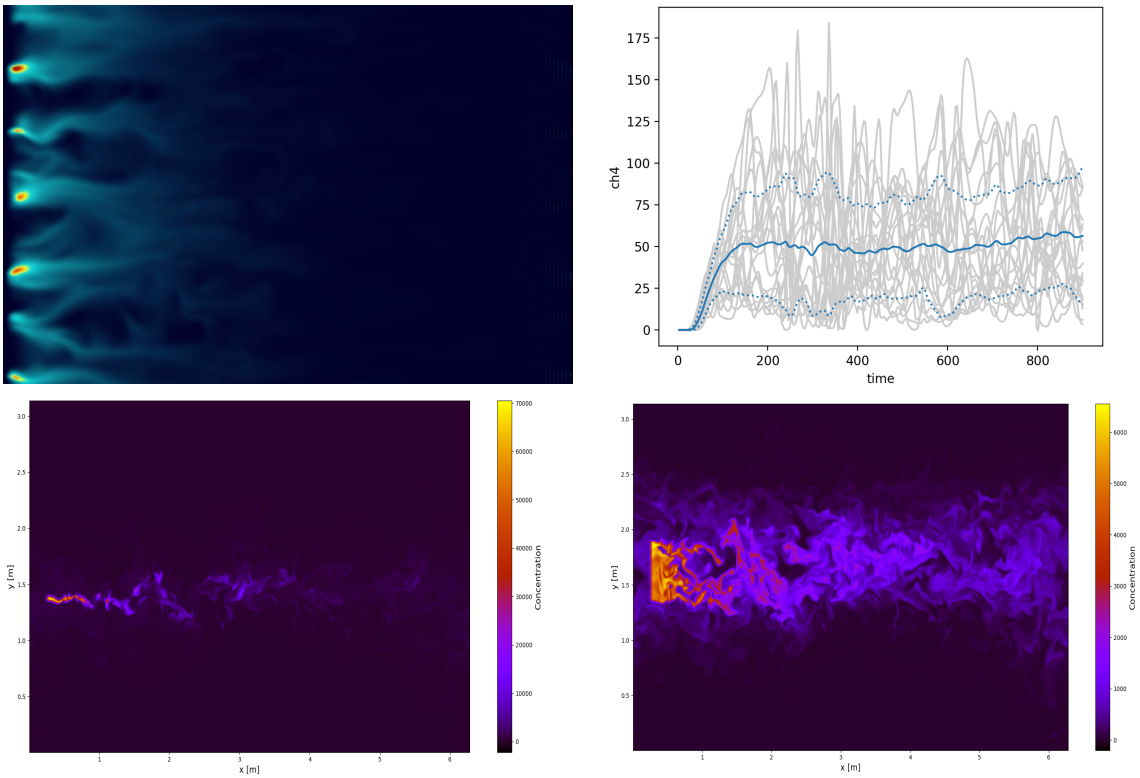

Fig. 8: DNS simulation of a plume from a line source in stationary homogeneous turbulence (above left); ensemble average of CH_4 mixing ratios at one point (above right); MicroHH simulated dispersion from a point source (arbitrary scales) (below left); MicroHH simulated dispersion from a line source (arbitrary scales) (below right).

On a meso-scale or European scale new simulations of CH_4 mixing ratios have been performed with the CHIMERE chemistry transport model using the EDGAR version 4.3.2 and TNO-MACC_III emission inventories from the year 2011. Multi-year simulations have been carried out from 2011 to 2015 with a horizontal resolution of 0.5°x0.5° (~50x50 km). Also, a large number of sensitivity experiments were performed. The comparison and the sensitivity tests aim at a better understanding of the differences between modelled and measured CH_4 concentrations and thus help reveal which part can be attributed to errors in inventories and serve the goal of estimating top-down CH_4 emissions on the European scale. An example for the site Lutjewad in the Netherlands shows the correlation between measurements and simulated values of the grid cell corresponding to the station location and its eight neighbouring cells was checked (Fig. 9), and the values of those model grid cells with the highest correlation coefficients can be used for the comparison against the measurements.

One of the sensitivity tests consisted of running the model with boundary conditions obtained from the CAMS-MACC reanalysis product [20] and in contrast to this using the pre-optimized boundary conditions derived from the LMDz model. The comparison showed, that simulations using the MACC boundary conditions compared clearly better to the measurements. To investigate the impact of the use of natural CH_4 emissions in addition to the anthropogenic emissions, we carried out a sensitivity run for which emissions from wetlands were included [21]. The inclusion of wetland emissions increased the mixing ratio especially over the wetland areas and coasts by up to about 36 ppb. Compared to the measurements, the addition of wetlands makes a slight positive difference to the simulation results, which seems advantageous as the measurements are mostly underestimated by the model.

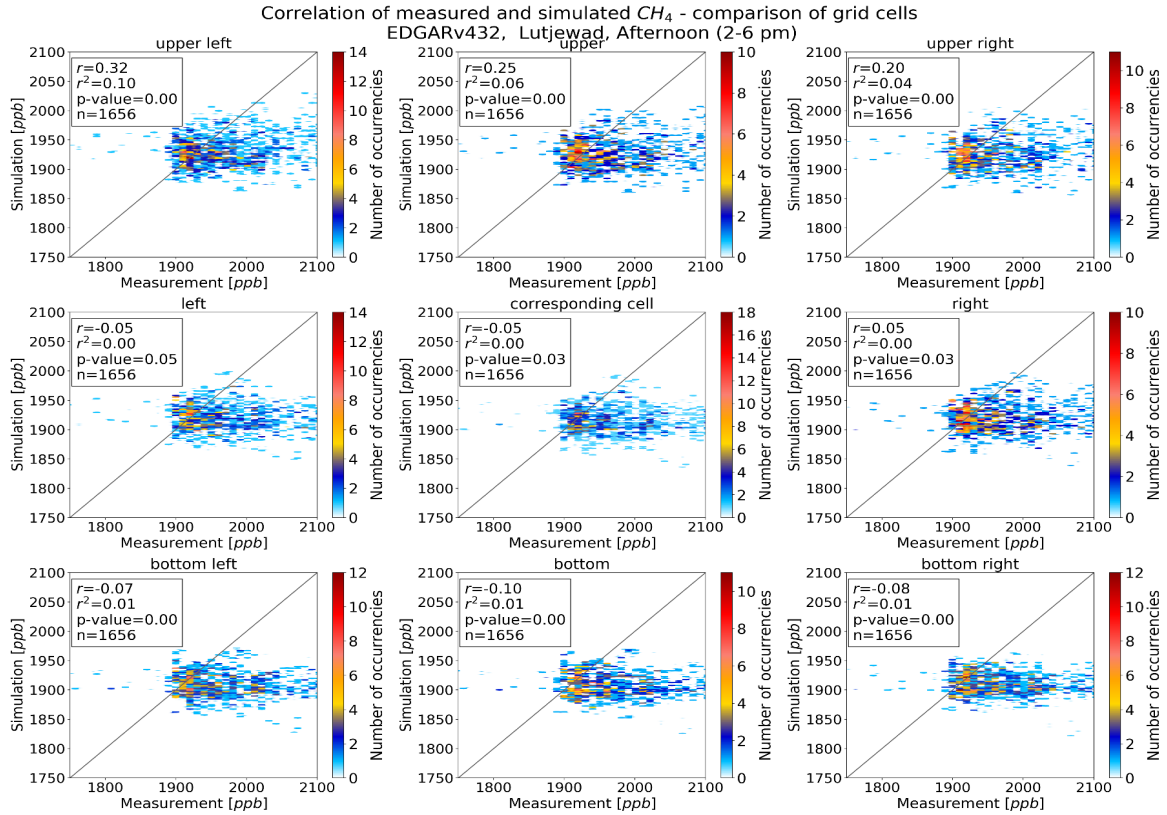

Fig. 9: Comparison of the simulated concentrations in the model grid cell corresponding to the measurement site's location and in its eight neighbouring cells. It is an example of mixing ratios simulated using EDGARv4.3.2 for the site Lutjewad. The analysis is based on hourly afternoon values from 2015.

To simulate the dispersion of CH_4 emitted from individual sources, the GRAL (Graz Lagrangian Model) dispersion model as implemented. An example simulation applied to a tracer release experiment conducted during the first MEMO^2 winter school in February 2018 is presented in Fig. 10. To improve the efficiency and applicability, the model has been improved by I) replacing the GRAL Graphical User Interface by a python module preparing all input data for a simulation (land cover, 3D obstacles, topography, etc.) and launching the computation jobs, (II) implementing the option to run dynamic (rather than static) simulations allowing to account for rapidly changing winds and turbulence, and (III) developing a python package for post-processing and visualization of the output. Furthermore, a simple Gaussian plume model has been implemented to compare the results obtained with GRAL.

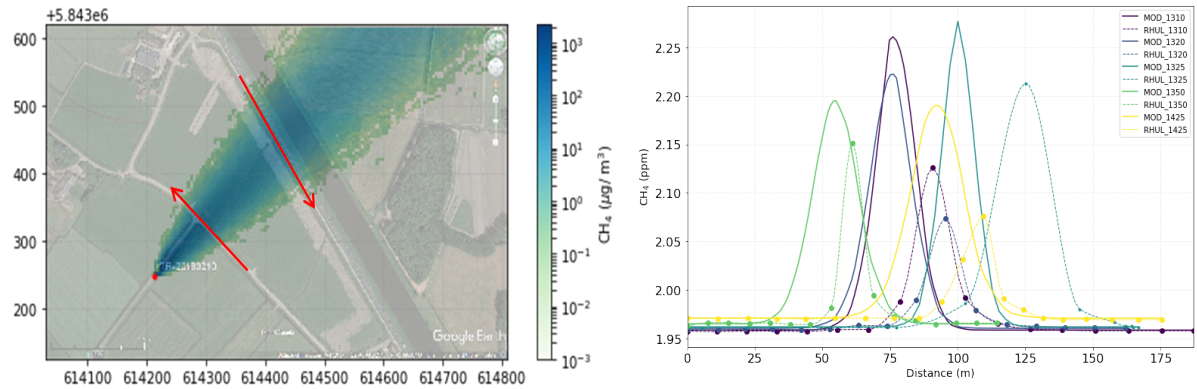

Fig. 10: Left: GRAL simulated CH_4 concentration (5-minute average) during a tracer release experiment in February 2018. The red arrows denote the paths of the mobile measurement platforms crossing the plume multiple times at two distances from the source. Right: Simulated (solid lines) and measured (dotted lines with symbols) CH_4 mole fractions along different transects sampled by the car of RHUL. Matching the areas below the curves allows estimating the strength of the source.

4. Summary and discussion

Effective emission reduction of any atmospheric relevant gas can only be achieved if sources are properly quantified, and mitigation efforts are verified. MEMO^2 as a European H2020 project focus on CH_4, and aims to bridge the gap between large-scale scientific estimates from in situ monitoring programs. The main benefit of MEMO^2 is the interdisciplinary character, the inclusion of many different technological and scientific advances covering the development and application of mobile platforms, isotope studies and modelling. The combination of measurement and modelling approaches as done by MEMO^2 are highly needed to achieve better and more reliable emission quantification and 'bottom-up' estimates of emissions from local sources that are used in the national reporting.

MEMO^2 provides a unique opportunity of data sets, especially including isotopic data which will go beyond individual measurements or forward and backward modelling alone to get realistic emission estimates. By focusing on the combination of interdisciplinary expertise and skills, MEMO^2 ensures that the interdisciplinary expertise will be capitalized such that MEMO^2 offers added value to the scientific community and creates a high societal impact.

Acknowledgements

The authors would like to acknowledge the support of all colleagues during the campaigns, measurements, and data evaluation. 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.

References

[1] EU climate action: http://ec.europa.eu/clima/policies/strategies/2020/indexen.htm

[2] IPCC, 2014: Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Core Writing Team, R.K. Pachauri and L.A. Meyer (eds.)]. IPCC, Geneva, Switzerland, 151 pp.

[3] Kirschke, S., et al.: Three decades of global methane sources and sinks, Nat. Geosci., 2013. 6: 813-823, DOI:10.1038/NGEO1955

[4] Dlugokencky, E.J., Nisbet, E.G., Fisher, R., Lowry, D.: Global atmospheric methane: budget, changes and dangers; Philos. T. R. Soc. A, 2011, 369, 2058-2072, DOI:10.1098/rsta.2010.0341

[5] Bousquet, P., et al.: Contribution of anthropogenic and natural sources to atmospheric methane variability, Nature, 2006, 443, 439-443, DOI:10.1038/nature05132

[6] Bergamaschi, P., et al.: Inverse modeling of European CH_4 emissions 2001–2006, J. Geophys. Res., 2010, 115, D22309, DOI:10.1029/2010JD014180.

[7] Henne, S., et al.: Validation of the Swiss methane emission inventory by atmospheric observations and inverse modelling, Atmos. Chem. Phys. Discuss., 2015, 35417-35484, doi:10.5194/acpd-15-35417-2015

[8] European Commission Joint Research Centre, Netherlands Environmental Assessment Agency, Emission Database for Global Atmospheric Research (EDGAR), Version 4.2., 2010: Available at http://edgar.jrc.ec.europa.eu

[9] Karion, A., et al.: Methane emissions estimate from airborne measurements over a western United States natural gas field, Geophys. Res. Lett., 2013, 40, 4393-4397, doi:10.1002/grl.50811

[10] Caulton, D.R., et al.: Toward a better understanding and quantification of methane emissions from shale gas development, Proc. Nat. Acad. Sci. USA, 2014, 111, 6237-6242, DOI:10.1073/pnas.1316546111

[11] Rella, C.W., Hoffnagle, J., He, Y., Tajima, S.: Local- and regional-scale measurements of CH_4, $\delta^{13}CH_4$, and C_2H_6 in the Uintah Basin using mobile stable isotope analyzer, Atmos. Meas. Tech., 2015, 8, 4539–4559, DOI:10.5194/amt-8-4539-2015

[12] Zazzeri, G., et al.: Plume mapping and isotopic characterisation of anthropogenic methane sources, Atmos. Environ., 2015, 110, 151-162, DOI: 10.1016/j.atmosenv.2015.03.029

[13] Bergamaschi, P., et al.: Inverse modelling of national and European CH_4 emissions using the atmospheric zoom model TM5, Atmos. Chem. Phys., 2005, 5, 2431-2460, doi.org/10.5194/acp-5-2431-2005

[14] Andersen, T., Scheeren, B., Peters, W., and Chen, H.: A UAV-based active AirCore system for measurements of greenhouse gases, Atmos. Meas. Tech., 11, 2683-2699, https://doi.org/10.5194/amt-11-2683-2018

[15] Graf, M., Emmenegger, L, Tuzson, B.: Compact, circular, and optically stable multipass cell for mobile laser absorption spectroscopy, Opt. Lett. Optics Letters, 2018, 43(11), DOI:10.1364/OL.43.002434

[16] Liu, C., et al.: Laser driving and data processing concept for mobile trace gas sensing: Design and implementation, Review of Scientific Instruments, 2018, 89(6), DOI:10.1063/1.5026546

[17] Röckmann, T., et al.: In situ observations of the isotopic composition of methane at the Cabauw tall tower site, Atmos. Chem. Phys., 2016, 16, 10469-10487, DOI:10.5194/acp-16-10469-2016

[18] de Groot, T., et al.: Tidal dynamics control microbial methane oxidation in the water column above an active cold seep (Doggerbank, North Sea), EGU General Assembly, 2019, Vol. 21, EGU2019-5523-1

[19] van Heerwaarden, C. C., et al.: MicroHH 1.0: a computational fluid dynamics code for direct numerical simulation and large-eddy simulation of atmospheric boundary layer flows, Geosci. Model Dev., 2017, 10, 3145-3165, DOI:10.5194/gmd-10-3145-2017

[20] Inness, A., et al.: The CAMS reanalysis of atmospheric composition, Atmos.Chem.Phys., 2013, 19, 3515-3556, https://doi.org/10.5194/acp-19-3515-2019

[21] Poulter, B., et al.: Global wetland contributions to 2000-2012 atmospheric methane growth rate dynamics, Environ. Res. Lett., 2017, 12, 094013, https://doi.org/10.1088/1748-9326/aa8391

Figure captions

Fig. 1: Interconnection scheme of the 3 scientific Work Packages of MEMO^2

Fig. 2: Overview of (joint) measurement campaigns, the colour codes indicate the type of samples

Fig. 3: CH_4 concentration measurements in Upper Silesian coal mining region.

Fig. 4: δD and $\delta^{13}C$ of shafts in the Silesian region

Fig. 5: Photography of the high-precision methane sensor (left side) and its mounting on a UAV (right side)

Fig. 6: 6-month time series of δD, $\delta^{13}C$, and the CH_4 mole fraction at Cabauw [16] (left), mean results of long-term campaigns in Krakow (PL), Lutjewad and Cabouw (NL) (right)

Fig. 7: $\delta^{13}C$ (left) and δD (right) during a 3-days' time series at the Doggerbank, North Sea

Fig. 8: DNS simulation of a plume from a line source in stationary homogeneous turbulence (above left); nsemble average of CH_4 mixing ratios at one point (above right); MicroHH simulated dispersion from a point source (arbitrary scales) (below left); MicroHH simulated dispersion from a line source (arbitrary scales) (below right).

Fig. 9: Comparison of the simulated concentrations in the model grid cell corresponding to the measurement site's location and in its eight neighbouring cells. It is an example of mixing ratios simulated using EDGARv4.3.2 for the site Lutjewad. The analysis is based on hourly afternoon values from 2015.

Fig. 10: Left: GRAL simulated CH_4 concentration (5-minute average) during a tracer release experiment in February 2018. The red arrows denote the paths of the mobile measurement platforms crossing the plume multiple times at two distances from the source. Right: Simulated (solid lines) and measured (dotted lines with symbols) CH_4 mole fractions along different transects sampled by the car of RHUL. Matching the areas below the curves allows estimating the strength of the source.