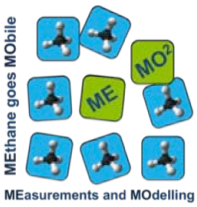

MEMO^2: MEthane goes MObile – MEasurements and MOdelling

MEMO^2 – Final Report

Period: 1 March 2017 – 28 February 2021

Lead authors

Dr. S. Walter

Prof. T. Röckmann

Utrecht University, Institute for Marine and Atmospheric Research Utrecht (IMAU)

Princetonplein 5

3584CC Utrecht

The Netherlands

Email : s.walter@uu.nl / t.roeckmann@uu.nl

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Abbreviations

AirCore	Atmospheric Sampling System
AGH	Akademia Gorniczo-Hutnicza im. Stanislawa Staszics w Krakowie, Krakow, Poland
C_2H_6	Ethane
CH_4	Methane
CHIMERE	Multi-scale chemistry-transport model for atmospheric composition analysis and forecast
CoMet	Carbon Dioxice and Methane Mission
CRDS	Cavity Ring-Down Spectroscopy
D	Deliverable
DFB-QCL	Single-Mode QCL
DTU	Technical University of Denmark
ECCC	Environment and Climate Change Canada
EDGAR	Emissions Database for Global Atmospheric Research
EMPA	Eidgenössische Materialprüfungs, und Forschungsanstalt, Dübendorf, Switzerland
ESR	Early Stage Researcher
FTIR	Fourier-Transform Infrared Spectroscopy
GEOMAR	Helmholtz Centre for Ocean Research Kiel
GPS	Global Positioning System
GRAL	Graz Lagrangian Model
ICL	Interband Cascade Laser
ICOS	Pan-European research infrastructure for quantifying and understanding the greenhouse gas balance of Europe and its neighbouring regions
INCAS	National Institute for Aerospace Research "Elie Carafoli"
InGOS	Research infrastructure targeted at improving and extending the European observation capacity for non-CO_2 greenhouse gases
IRMS	Isotope Ratio Mass Spectrometry
LGR	Los Gatos Research
LMDz	Laboratoire de Météorologie Dynamique zoom, general circulation / global climate model
LU	Lunds Universitet, Lund, Sweden
MicroHH	Large-Eddy Simulation code
MS	Milestone
PPB	Parts Per Billion
PPM	Parts Per Million
Python	Programming language
QCL	Quantum Cascade Laser
R	Programming language / software environment for statistical computing and graphics
RANS	Reynolds-averaged Navier–Stokes equations
RHUL	Royal Holloway and Bedford New college, London, United Kingdom
RUG	Rijksuniversiteit Groningen, Groningen, The Netherlands
SLU	Swedish University of Agricultural Science
TNO MACC	Monitoring Atmospheric Composition and Climate – Emission data set (https://cordis.europa.eu/project/rcn/91167/factsheet/en)
TO-3	Transistor Outline, standardized metal semiconductor package incl. transistors, rectifiers and circuits
UAV	Unmanned Aerial Vehicle
UHEI	Ruprecht-Karls-Universität Heidelberg, Heidelberg, Germany
UU	Universiteit Utrecht, Utrecht, The Netherlands
UVSQ	Université de Versailles Saint-Quentin-en-Yvelines, Paris, France
WDCGG	World Data Center for Greenhouse Gases
WP	Work Package
WU	Wageningen Universiteit, Wageningen, The Netherlands

1. Executive summary

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 the 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 measurements. Effective emission reduction can only be achieved if sources are properly quantified and mitigation efforts are verified.

MEMO² (https://h2020-memo2.eu) was a H2020 MSCA European Training Network with 25 collaborators from 8 countries. The project contributed to the climate targets of the EU with a focus on methane (CH_4). The goal of MEMO² was to bridge the gap between large-scale *top-down* estimates from atmospheric 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.

MEMO² ran smoothly and successfully, and ended on 28 February 2021. All tasks have been fulfilled, several were extended, in particular by the organisation of an additional large joint measurement campaign (ROMEO). In total, 17 milestones were achieved and 32 deliverables submitted. The quality and achievements of MEMO² resulted in the declaration of MEMO² as a REA Success Story.

This final report gives an overview about the project, the achieved results during its four-year lifetime and the socio-economic impact.

2. 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" 2°C and, if possible, below 1.5 °C. 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 headline target of the EU climate actions. 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 the context of the 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 (IPCC, 2014) and several sources offering possibilities of "no-regret" emission reduction such as landfills, gas leaks and manure, 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. Unfortunately, there are still 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.

MEMO² – Final Report (2017-2021)

The H2020 European Training Network (ETN) MEMO² (MEthane goes MObile – MEasurements and MOdelling, https://h2020-memo2.eu) aimed to bridge the gap between large-scale estimates from the established scientific monitoring programs and the 'bottom-up' estimates of emissions from local sources that are used in the national reporting. As an ETN, MEMO² aimed not only on scientific excellence but also on the training of early stage researchers (ESRs), which are capable of investigating CH_4 emissions in an interdisciplinary context related to climate change mitigation.

The main scientific goals of MEMO² were I) 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, II) to identify and quantify CH_4 emissions from local sources in Europe and III) to use these updated emissions improve estimates at the European scale. The MEMO² results enable the scientific and non-academic communities to improve the objective verification of CH_4 emission reduction strategies for specific source sectors.

The main training goal was to train qualified scientists in the use and implementation of interdisciplinary knowledge and techniques that are essential to meet and verify emission reduction goals. Due to the complexity and interdisciplinary character of detecting and quantifying CH_4 emissions, and the evaluation of climate mitigation measures, emission mitigation requires skilled scientists with a high-level of theoretical and practical competences that are able to cooperate in national and international networks. So MEMO² developed and implemented a dedicated research training program, which stimulated key competences and knowledge exchange, aiming at the education of a generation of “cross–thinking” scientists. The training included activities at the individual, local, network-wide and international levels. By this, MEMO² also contributed to associated targets of SDG 13, which focus e.g. on the improvement of education and awareness-raising (https://sustainabledevelopment.un.org/sdg13).

The following **MEMO² specific objectives** were defined:

- Implementation of a mobile CH_4 measurement network across Europe that can be used for detection and quantification of sources, verification of mitigation measures, and for developing refined emission estimates
- Development of innovative new mobile CH_4 measurement systems (e.g. by using unmanned aerial vehicles (UAVs))
- Training of researchers to utilize and develop methods / tools for detection, quantification, and verification of greenhouse gases such as CH_4
- Augmentation of the established training programs at individual institutions with an innovative network training that incorporates direct links with non-academic partners
- Development and application of novel modelling tools to refine local emission estimates from mobile and isotope measurements
- Derivation of a new bottom-up CH_4 emission map, including isotopic information, across Europe
- Derivation of top-down emission estimates over Europe exploiting the new information acquired in MEMO².
- Fostering a close collaboration between the academic and the non-academic sector by joint network activities

3. MEMO² background

3.1 General set-up

MEMO² as a H2020 MSCA-ITN European Training Network was an international and interdisciplinary project, with 9 academic and 16 non-academic partners. MEMO² synergistically used the highly specialized competencies and facilities of these partners from the disciplines of particularly atmospheric physics and chemistry, environmental sciences, meteorology, and metrology. This included 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 comprised of three scientific work packages (WPs), which were strongly interconnected (Fig. 1).

WP1 was dedicated to mobile measurements across Europe. In WP2 state-of-the-art isotope techniques were 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 was the task within WP3.

The scientific WPs shared a common objective and complemented each other by detecting (WP1), attributing (WP2) and quantifying (WP3) CH_4 emissions in Europe using measurements on mobile platforms.

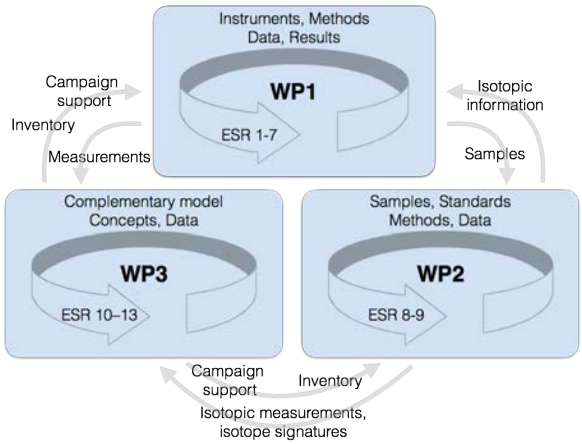

Fig. 1: Interconnection scheme of the 3 scientific Work Packages of MEMO²

The geographic locations of the partners provide excellent opportunities to characterize important natural and anthropogenic CH_4 source categories around Europe, such as e.g. agriculture and oil & gas industry, landfills, city emissions, coal mining or arctic wetlands.

3.2 General scientific and methodological approach

CH_4 measurements within MEMO² span the full range from high-precision flask samples for isotope analysis, to continuous time series using laser spectroscopy, and in situ measurements by vehicles, airplanes and drones in all three dimensions. The modelling activities allowed 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 was 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 (Kirschke et al. 2013; Dlugokencky et al. 2011). 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 (Bousquet et al. 2006; Bergamaschi et al. 2010; Henne et al. 2015). 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 (EDGAR database version 4.2, 2010, http://edgar.jrc.ec.europa.eu). Often large discrepancies occur between bottom-up and top-down estimates of emissions (Karion et al. 2013; Caulton et al. 2014). 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 estimate CH_4 sources at the EU-level use both bottom-up and top-down methods Henne et al. 2015; Karion et al. 2013). 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 (Dlugokencky et al 2011). 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 MEMO² research program was to use innovative measurements and modelling of CH_4 using mobile platforms as a principle tool to bridge the current scale gaps between local measurements, emission inventories, and European scale modelling.

3.3 Mobile measurements

Mobile measurements of CH_4 emissions became available only few years ago (Rella et al 2015). The interpretation of such measurement 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 were fast and accurate analysers on mobile platforms. Analysers used were 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 were cars, but also unmanned aerial vehicle (UAV) platforms and light aircrafts were used to investigate focus source types such as wetlands, landfills, city emissions, lakes, gas leaks, agricultural emissions, or mining emissions. By this, MEMO² mapped the small-scale distribution of CH_4 across Europe, identified and quantified CH_4 emissions at the local scale and provided emission factors for further modelling activities. The CH_4 source mix is different per country, and - based on the inventories - MEMO² targeted the largest uncertainties in the individual countries. A key advantage of the network was that due to close cooperation the regional/national scale but also the European scale was covered.

3.4 Isotopic measurements

Different sources emit CH_4 with slightly different isotopic composition (Zazzeri et al. 2015). So, measuring the isotopic composition of CH_4 helps to identify the sources responsible for observed elevations of CH_4 in the atmosphere. However, this source attribution can only be as good as the knowledge of the isotope signatures of the sources in the surrounding area, and in MEMO² we put emphasis on improving the understanding of the temporal and spatial variability of isotopic signatures of CH_4 emissions. This 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 in MEMO² allowed investigating the vertical as well as horizontal variability of the isotopic signature in emission plumes. The information provided important input for the use of isotope information in atmospheric models.

3.5 Modelling approaches

The quantification of emissions from concentration measurements requires complementary modelling tools on various scales, from the local scale of an individual oil well, a coal mining shaft, a leak in a gas distribution network or a farm, via the regional scale of fossil fuel production basins or cities, up to the EU and global scale (Bergamaschi et al. 2005). At the local scale, several variants of the "Gaussian plume" concept, as well as mass balance approaches were used in MEMO² for emission quantification. In addition, we developed Large Eddy Simulations (LES) as innovative tools to predict and analyse in a high level of detail, including turbulence, dispersion patterns from local sources. "Virtual vehicles" and UAVs were used to sample simulated 3D dispersion fields from CH_4 sources. On the regional scale, flow patterns that integrate mixed sources from e.g. a city were analysed using regional high-resolution modelling. Combined, these approaches allowed optimal interpretation and usage of the measurement results. At the European scale, forward simulations of CH_4 concentration fields were derived, using detailed and updated bottom-up emissions maps. Due to the necessary time temporal sequence between measurements and their inclusion in models (the measurements first have to be carried out and evaluated before they can be used in models), the modelling activities at the EU scale, which started at the beginning of MEMO², used largely previously available inventory information. At the end of MEMO², we started to use the first published results of the individual projects to improve the European CH_4 inventories. Modelling activities also assisted in the design of measurement strategies. The goal was to determine areas where measurements will have the largest benefit concerning both uncertainty of emissions and possibilities of mitigation measures.

4. Results

The results presented in this section show a summary of the results of MEMO². For more details we refer to the Periodic Reports including the respective ESR reports, deliverables and milestone reports as well as to the publications of the consortium. All documents are available on the project website: https://h2020-memo2.eu.

4.1 Mobile measurements of CH_4

The aim of MEMO² was to identify, quantify, and monitor CH_4 plumes of major anthropogenic and natural CH_4 emitters in Europe using mobile platforms such as cars, drones and airplanes. Almost 800 individual ESR measurement days and joint campaign days were performed. Fig. 2 gives an overview of (joint) measurement locations spread over Europe. Besides individual measurement locations such as agricultural facilities (Vinkovic et al. 2021, in preparation), biogas plants (Bakkaloglu et al. 2021), landfills, or subarctic mires (Łakomiec et al. 2021), specific highlights of MEMO² were the international measurement campaigns in Poland (CoMet) and Romania (ROMEO), and joint city campaigns which will be described in more detail below.

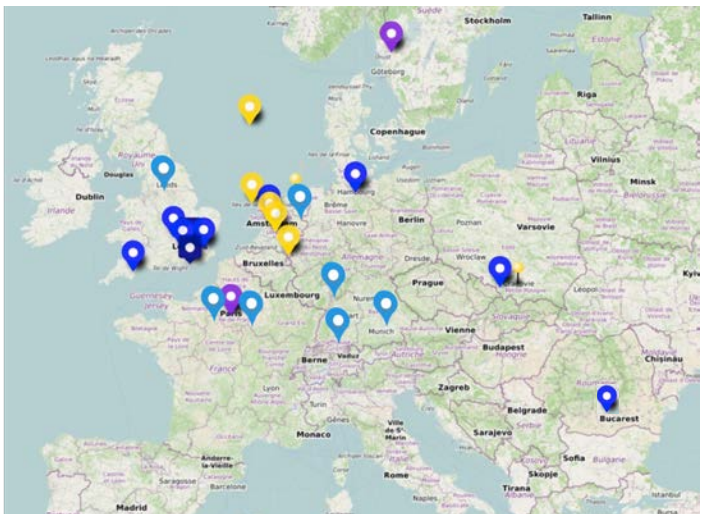

Fig. 2: Overview of (joint) measurement campaigns

4.1.1 Upper Silesian Coal Basin (USCB)

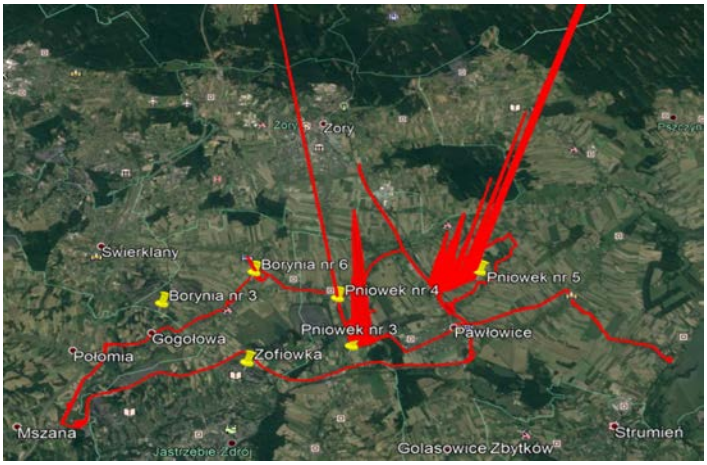

Fig. 3: Examples of mobile CH_4 concentration measurements in the Upper Silesian coal mining region.

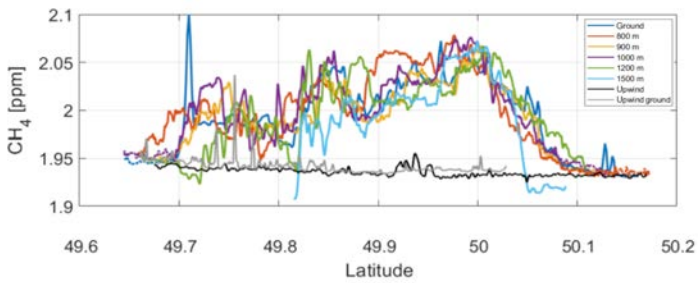

Fig. 4: Measurements of the mole fractions of CH_4 upwind and downwind the UCSB from aircraft and the MEMO² vehicle measurements. Modified from Fiehn et al., 2020.

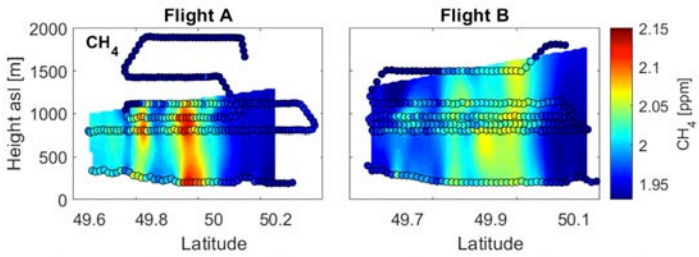

Fig. 5: Mole fractions of CH_4 in the downwind in situ "wall" from observations 450 (circles) and inter- and extrapolation with a kriging algorithm (shading). The CH4 wall incorporates ground-based measurements. Modified from Fiehn et al., 2020.

In May 2017 and June 2018, MEMO² 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 focus on mining activities over Silesia as one of the European anthropogenic CH_4 hotspots (Fig. 3). In this region 33 mines are active, but also additional methane sources are present: landfills, cities gas networks, farms, wetlands and agriculture.

During COMET, 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 (Andersen et al. 2018) and applied the FTIR technique with stationary and mobile platform. MEMO² particularly contributed with ground-based measurements using vehicles. One of the objectives was to add "ground based legs" during the days or aircraft mass balance flights. Three vehicles measured on streets along the aircraft transects during the COMET flight days (Fig. 4). The surface and aircraft measurements were combined and interpolated with a Kriging technique (Fig. 5). This allowed to constrain the vertical extent of the observed CH_4 plumes.

The combined aircraft and vehicle measurements from two flights were then used in a mass balance approach to estimate the total basin-wide emissions of CH_4 (and other trace gases) from the USCB (Fiehn et al., 2020). The airborne mass balance CH_4 emission estimates are in the lower range of inventory emissions (Fig. 6).

E-PRTR emission estimates are similar to our estimate. The CoMet emission inventory is higher than both mass balance estimates, but within the error range of flight B. Compared to E-PRTR from 2017, the CoMet inventory includes several mines in Poland that reported higher CH_4 emissions in 2016 than in 2017, three additional Czech mines and four landfills within the mass balance area. Scarpelli, CAMS and EDGAR CH4 estimates are also higher than our mass balance results. The GESAPUinventory states the lowest emissions, which may result from the missing results from Czech mines.

MEMO² vehicles also carried out measurements in the vicinity of coal mine ventilation shafts, and the results are being prepared for publication (Stanisavljević et al., in preparation). Fig. 7 shows an example of mole fraction and isotope measurements at the Bogdanka mine. We find a high variability in emission rates from different mining shafts, but also from repeated plume transects of the same shaft. Also, the isotope signatures of the emitted methane are surprisingly variable.

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. 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.

The emission rates of comparable shafts agreed only partly with reported emission estimates from the bottom-up E-PRTR and CoMet v4.0 inventories. The difference might be partly attributable to different reported time scales. Emissions on the minute to hour scale are quantified by the measurements, whereas monthly to annually numbers are provided by the operators. Thus, the discrepancy between our measurements and bottom-up inventories may be due to the temporal variability of emissions from individual shafts, which cancels out over the entire basin.

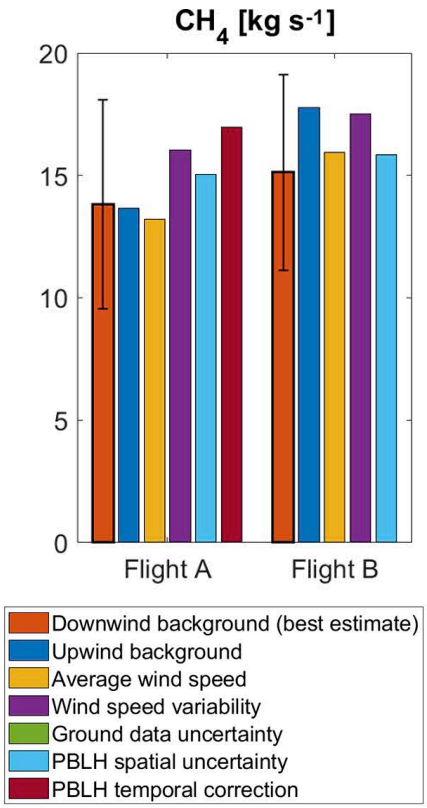

Fig. 6: USCB emission estimates from two aircraft flights in June 2018, using an airborne mass balance approach, compared to inventory estimates Figure modified from Fiehn et al., 2020.

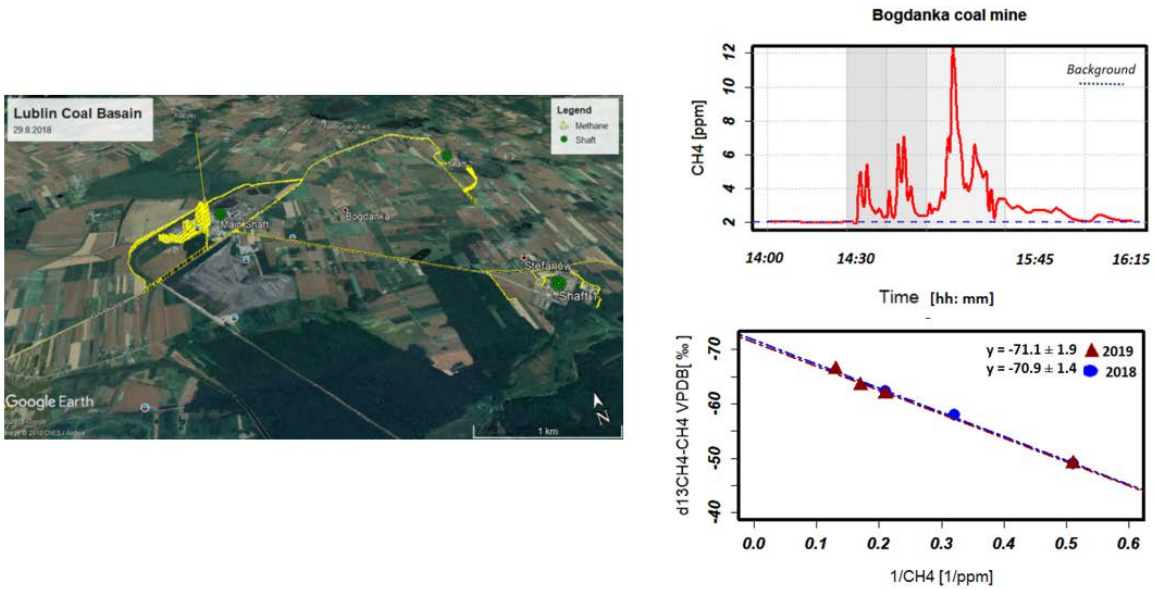

Fig. 7: Bogdanka mine investigation. Left: CH_4 concentrations around the ventilation shafts. Right: methane concentration record (above) and a Keeling plot (below) to investigate the source signature.

An alternative explanation is that the plumes, which are released from the mining shafts at 10 m above ground level, and with considerable vertical speed, may not have mixed fully down to ground level where we carry out the ground-based measurements from vehicles. More frequent measurements are needed to fully understand the variability and errors related to the temporary variability of sources and finally provide in-depth emission inventories.

In parallel with the CoMet campaign, a long-term isotope monitoring-campaign was carried out in Krakow. The goal was to get a clearer picture of the main CH_4 sources in the Krakow region, and especially to evaluate the influence of emissions from the Silesian coal mines (see chapter 4.2).

4.1.2 Fossil, biogenic and combustion emissions from cities

Cities, with their large and complex infrastructures, are significant sources of CH_4 emissions and MEMO² carried out mobile measurements in several European cities in cooperation with a UNEP CCAC project

Systematic screening of city emissions was performed in Paris (Fig. 8, left), London, Hamburg, Utrecht and Bucharest (Fig. 8, right).

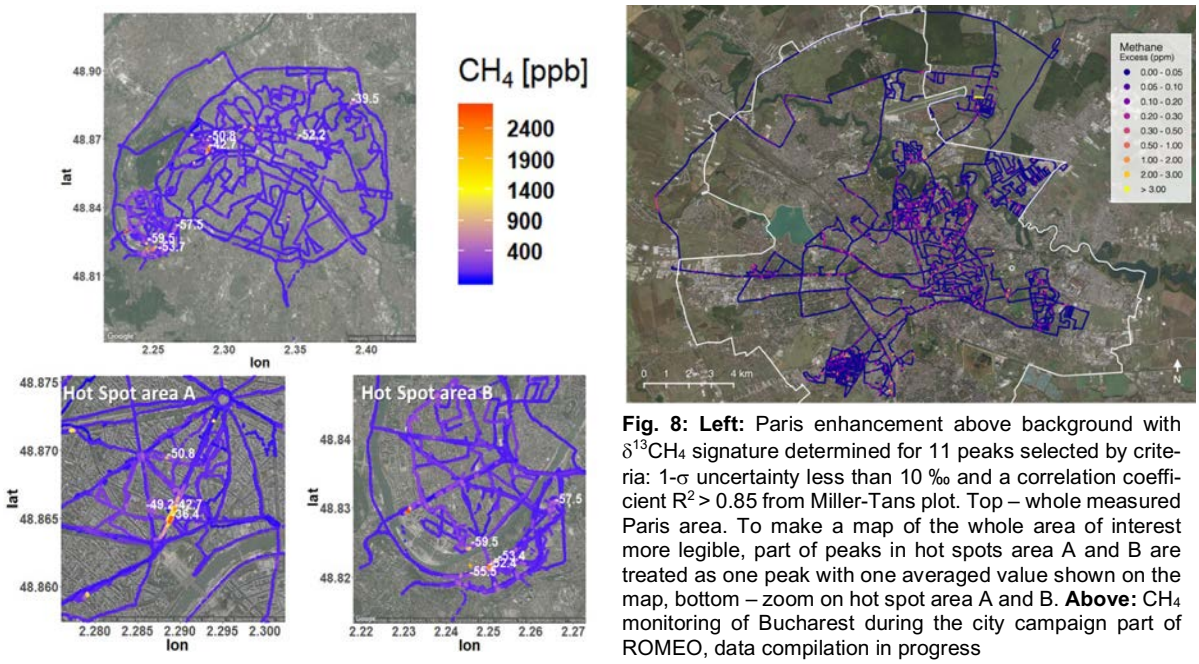

Fig. 8: Left: Paris enhancement above background with $\delta^{13}CH_4$ signature determined for 11 peaks selected by criteria: 1-σ uncertainty less than 10 ‰ and a correlation coefficient $R^2 > 0.85$ from Miller-Tans plot. Top – whole measured Paris area. To make a map of the whole area of interest more legible, part of peaks in hot spots area A and B are treated as one peak with one averaged value shown on the map, bottom – zoom on hot spot area A and B. **Above:** CH_4 monitoring of Bucharest during the city campaign part of ROMEO, data compilation in progress

In Paris (Defratyka et al., in preparation), initial surveys were used to identify areas of the greatest elevated CH_4 concentrations, designated as 'hot spots'. Two hot spot areas were identified: 1) on the west-south part of inner Paris (Fig. 8, area A), and 2) hot spot area B on west-south suburbs (Fig. 8, area B). These two areas of clusters counted for 22% and 56% of the total potential emissions of Paris. The leaks detected sorted into three leak categories: natural gas distribution network emissions (63%), sewage network emissions (33%), and emissions from heating furnaces of buildings (4%). The latter category has not previously been reported in urban methane studies. Accounting for the detectable emissions from the ground, the total estimated CH_4 emission rate of Paris was 5000 L/min (190 t/yr), with the largest contribution from gas leaks (56%). This ranks Paris as a city with medium CH_4 emissions. The findings suggest that the natural gas distribution network, the sewage system, and furnaces of buildings are ideal targets for street-level CH_4 emission reduction efforts for Paris.

Data were also used to determine the precision of instruments and improve methods, which in the end can help to develop simple and low-cost methodologies to quantify CH_4 emissions from cities. Defratyka et al. (2020) investigated the capability of CRDS G2201-i to measure C_2H_6:CH_4 ratio under field conditions in Paris.

Even though the instrument is not dedicated for C_2H_6:CH_4 ratio measurements, after applying correction and calibration factors, and adapt the sampling methodology, the CRDS G2201-i instrument can contribute to better constrain methane sources deploying only one instrument.

Street-level measurements in Bucharest (Fernandez et al. in preparation) were part of the ROMEO project (see chapter 4.1.3). A large part of the city of Bucharest was screened to investigate the street level enhancements, flux rates, and potential source origins (using also carbon and hydrogen stable isotopic composition along with ethane:methane ratios). Results for Bucharest (Fernandez et al., in preparation) confirmed almost 1000 locations with CH_4 enhancements (Fig. 9). The total estimated city emission rate is around (1832 ± 66) metric tonnes CH_4 per year. Based on isotopic data, more than half (58% to 63%) of the CH_4 elevations were attributed to biogenic wastewater, mostly from leaking sewer pipelines or sewer vents, whereas thermogenic fossil fuels accounted for 32% to 42%.

4.1.3 Romanian Methane emissions from Oil and Gas (ROMEO)

The ROMEO campaign (ROmanian Methane Emissions from Oil and gas) was an international campaign that made heavy use of the MEMO² infrastructure. In total, there were 80 participants from 18 institutes and universities involved. ROMEO was organised and executed by UU, funded through the Climate and Clean Air Coalition (CCAC) international methane science studies (https://www.ccacoalition.org/en/activity/oil-and-gas-methane-science-studies), administered through the United Nations Environment Program and MEMO².

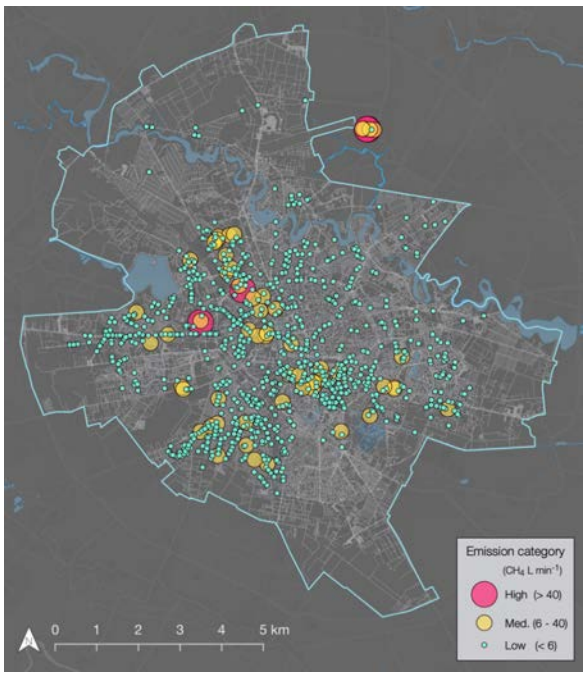

Fig. 9: CH_4 emission rate categories of Bucharest. Bucharest has 969 CH_4 emission localities that were identified through clustering a total of 2482 CH_4 indicators. The major Drumul Potcoavei leaks (northeast of the Bucharest boundary) include 7 emission locations which were clustered from 89 CH_4 indicators. Magnitude categories defined in von Fisher et al., 2017.

According to UNFCCC statistics, Romania is one of the European countries that reported highest emissions of CH_4 from the oil and gas sector to the atmosphere, in particular related to CH_4 production and end use. However, the estimates reported by Romania are derived using non-country specific emission factors and there are only very few observations which investigate whether the reported emissions are realistic.

The ROMEO campaign aimed at better experimental quantification of CH_4 emissions from the oil and gas sector in Romania with the goal to devising emission mitigation strategies. The combined bottom-up and top-down approach to quantify the CH_4 emissions were executed on three different scales - the exploration basin scale, the well scale and the city scale – by using different mobile platforms such as aircrafts, drones, or cars. On basin and well scale, Fig. 10 gives an overview about measurement areas identified as scientifically interesting, of which 9 were chosen based on scientific but also logistical constrains. The initial measurement areas were defined on basis of public and confidential available facility locations, and a-priori bottom-up inventories.

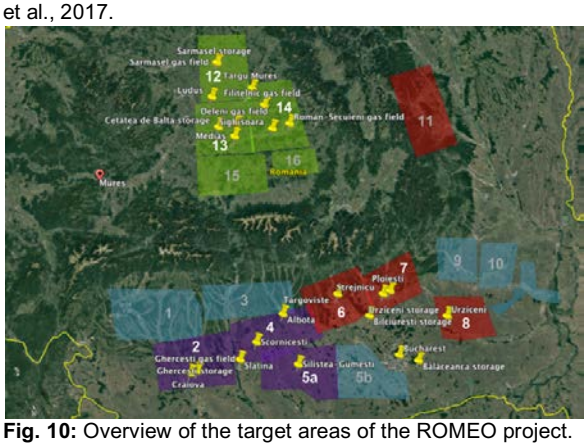

Fig. 10: Overview of the target areas of the ROMEO project.

During the ROMEO campaign, in total more than 1000 facilities were visited. Data show that the concentration elevations vary between one and several thousand ppm at distances between several meters to several ten meters away from a facility. There was almost no gas flaring in the regions visited, and often the visited methane collection infra-structure at production / separation / storage sites were not in good state. Observations at the smaller facilities such as oil and gas wells and compressor stations showed, that emissions were often due to operation (e.g. open pipes, not properly closed valves) and partly leaking equipment (flanges, pressure gauges).

At several facilities simultaneous measurements using different quantification methods were executed to facilitate intercomparison and integration of the entire dataset. All ground-based teams made joint calibration measurements that were used in the data evaluation for correction and intercomparison of the data.

The component level measurements show detectable methane emissions with the FLIR camera (Fig. 11) at roughly half of the individual wells, in agreement with the facility scale estimates.

Fig. 11: leak detection and quantification using a FLIR camera (location 229, 6 October 2019)

Approximately 200 wells and facilities out of the visited >1000 were quantified. Used methods were mass balance and Gaussian plume approaches, OTM33A (EPA 2014, Brantley et al. 2014, Omara et al. 2018), and Tracer gas dispersion models. Fig. 12 shows an example of quantifications using the OTM-33A technique. The distribution shows the typical “heavy-tail” characteristics, with a few very high emitters and the majority of sites with lower emission rates.

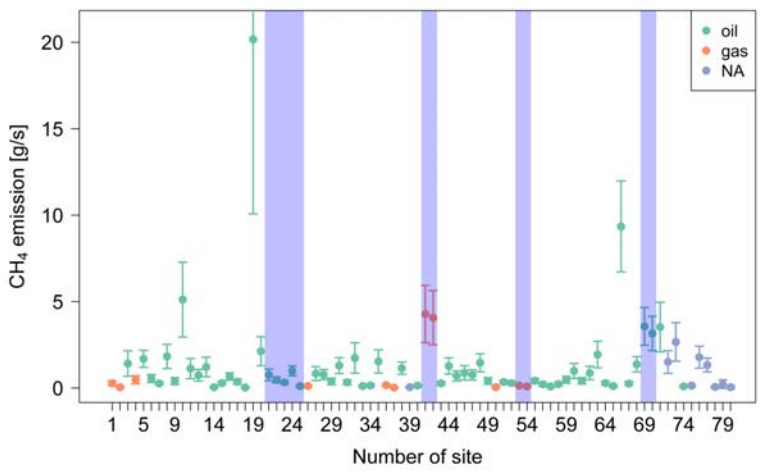

Fig. 12: Calculated CH_4 emission rates using OTM–33a. Green, orange and blue dots represent CH_4 emissions for oil, gas and unknown types of the source sites, respectively. The shaded areas mark sites with several simultaneous application of OTM33a (from Korben et al., in preparation, 2021).

In addition, two drone teams performed more than 120 flights to quantify sources CH_4 emission fluxes were mainly determined using the mass balance method. Fig. 13 shows typical layouts of the flight patterns during ROMEO.

Fig. 14 shows an example of CH_4 measurements performed by flying the QCLAS-drone system downwind of a given source, perpendicular to the main wind direction at different altitudes above ground. Geostatistical interpolation (Kriging) of the measured CH_4 molar fractions was performed to spatially fill the gaps as illustrated in Fig. 15.

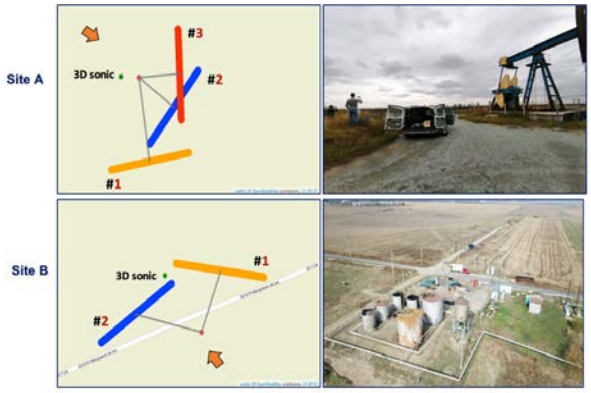

Fig. 13: The flight location of downwind flights with its surroundings, conducted on the site A and B during ROMEO. Red dots: location of the source(area), green dots: location of the 3D sonic anemometer, orange arrows: wind direction for each site.

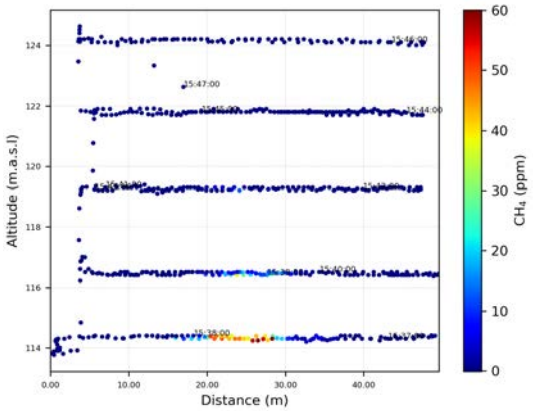

Fig. 14: Gridded measured CH_4 molar fractions (by drone)

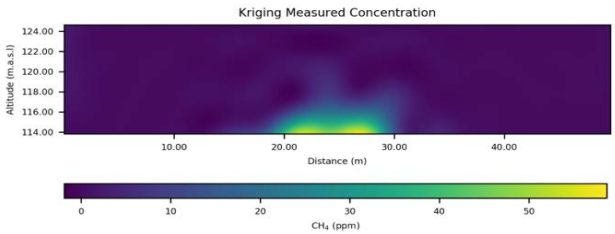

Fig. 15: Spatially filled measured CH_4 molar fractions (Kriging)

A clear benefit from joint campaigns such as CoMet and ROMEO is that results of different participants, methods, and methodologies can be compared and that they also complement each other. E.g., preliminary data of CH_4 emission measurements during ROMEO show, that in general the application of the Gaussian Plume Model (GPM) significantly underestimated the CH_4 emissions quantified using the Mobile Tracer gas Dispersion Method (MTDM) (Delre et al., in preparation). This underestimation could clearly be reduced by implementing information about atmospheric dispersion of tracers in the model. Another example is the combined use of δ^2H-CH_4 source signatures and C_2H_6:CH_4 ratios, which were more effective for source apportionment compared to δ^{13}C-CH_4, due to ^{13}C depleted natural gas in the region (Fernandez et al, in preparation). Fostering collaboration and taking those findings into account for future campaigns can clearly improve the impact of these joint measurement efforts.

The ground-based data from ROMEO are presently being compiled in three publications, one describing the tracer release measurements (Delre et al., in preparation, 2021), one the OTM-33A and Gaussian plume modelling results (Korben et al., in preparation, 2021). The third publication presents the drone-based data, the entire combined dataset, and an upscaling approach (Vinkovic et al., in preparation, 2021).

The airplane teams conducted more than 20 upwind and downwind flights, including regional mass balance and facility-cluster quantification. In several cases, the aircraft measurements could directly be linked to the underlying facility clusters and show significant methane enhancements at altitudes of hundred to several hundred meters close to production clusters, in qualitative agreement with the facility scale measurements. Preliminary results show that significant leakage of facilities can been identified.

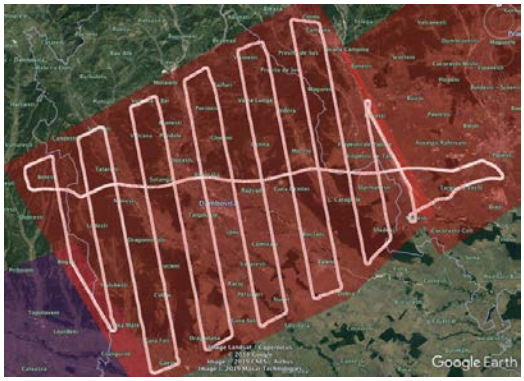

Fig. 16: Example of flight pattern, executed in area 6.

Isotopic analysis of air samples collected on board allows to identify the source category. Generally, the isotopic signals confirmed the oil and gas sector as the main source of CH_4 in the target areas because the isotopic signatures observed on the aircraft are similar to those measured on the ground at the oil and gas wells and facilities. This means that the elevations observed during the flights can provide reliable emission estimates for this sector of activity.

The aircraft from Scientific Aviation included an in-situ wind sensor, which enabled direct calculation of surface fluxes from an encircled area using the mass balance approach. Quantitative estimates were derived for 7 emission clusters and 3 larger regions (Fig. 10). The Romanian BN2 aircraft operated by our partner INCAS did not include on board wind measurements, therefore the aircraft was mainly used for raster flights in order to map potential emission spots in the target regions. Fig. 16 gives an example of flight patterns; the quantitative evaluation is not straightforward, in particular due to the low wind speeds that prevailed during the campaign. Therefore, we decided to compare the aircraft measurements to results of three different models (Flexpart-COSMO, MECON and WRF). The results are presently under investigation.

4.1.4 New sensor development

Next to reliable methodologies for sampling and measurements, successful campaigns depend also on excellent and suitable equipment, and MEMO² contributed to the development of new instrumentation such as a lightweight high-precision mid-IR methane laser spectrometer for unmanned aerial vehicles (UAV) (Tuzson et al. 2020, Fig.17). 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 (Graf et al. 2018). 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 (Liu et al. 2018). 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.

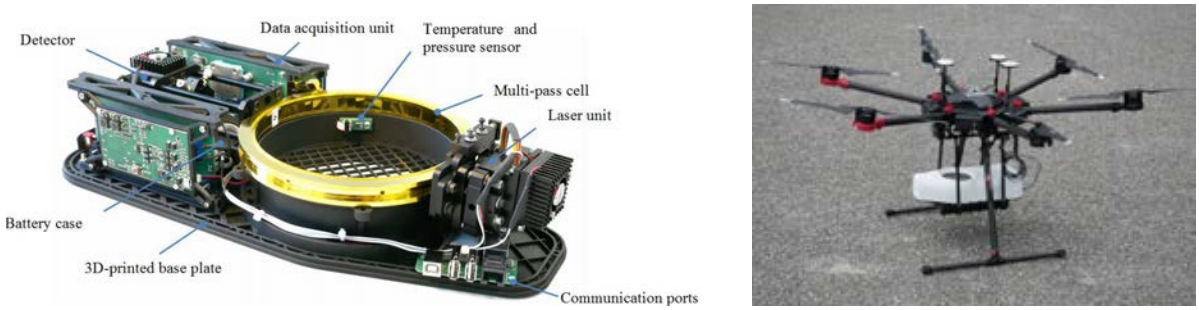

Fig. 17: Photography of the high-precision methane sensor (left side) and its mounting on a UAV (right side)

4.2 Source identification by isotopic characterization

CH_4 originates from several sources, such as biogenic (e.g. cows), thermogenic (e.g. natural gas) and pyrogenic (combustion) sources, which can be identified by their isotopic composition. Isotopic characterization and mapping of CH_4 sources requires all laboratories measuring the isotopes of CH_4 being 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 near instantaneous field measurements of isotopes. However, the measurement precision is lower than for the standard method, isotope ratio mass spectrometry (IRMS). In order to make the results from MEMO² internally consistent, an isotope intercomparison using prepared calibration tanks to align the isotope scale of all instruments was conducted. Therefore, the results obtained within MEMO² can be integrated, and directly compared with other results obtained globally.

With the intercomparison as a basic step, long-term monitoring experiments were intensified and new ones started. Fig. 18 shows a 6-months' time series at Lutjewad, the Netherlands (Menoud et al. 2020), similar long-term monitoring experiments were executed in Krakow from October 2018 till March 2019.

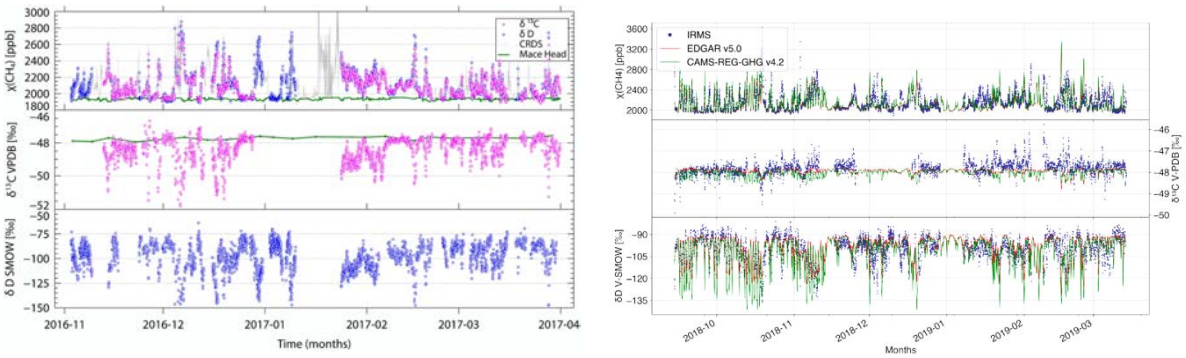

Fig. 18 left: 6-month high-resolution time series of δD, $\delta^{13}C$, and the CH_4 mole fraction at Lutjewad, NL (Menoud et al. 2020, left), **right**: high-resolution δD, $\delta^{13}C$, and the CH_4 mole fraction time series in Krakow, PL, and related modeling, using two different emission inventories. The differences between measurements and model results point to discrepancies in the inventories. For Krakow, this suggests a mis-representation of sources from coal mining waste in the category "waste" in the CAMS-REG-GHG v4.2 inventory.

When these long-time measurement series are evaluated with a modified Keeling plot approach, the source signatures clearly illustrate that the CH_4 in Krakow mainly originates from thermogenic sources, whereas CH_4 at the Dutch stations is mainly biogenic in the Netherlands (Fig. 19). This is not clear a priori since the measurement location Lutjewad is near Europe´s largest gas reservoirs in the North Sea and close to the Groningen reservoir. 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 also used to compare global and meso-scale models, which in the end helps to improve the model predictions and inventories (see also chapter 4.3).

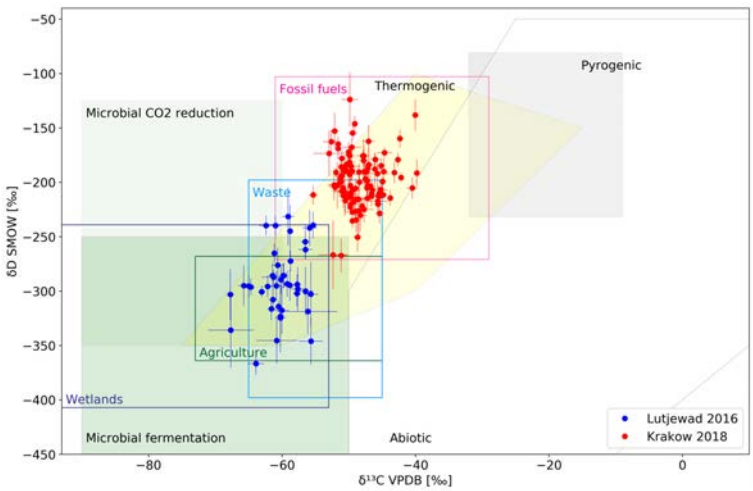

Fig. 19: Keeling plot intercepts for the measurements in Lutjewad (blue) and Krakow (red), confirming that in Krakow the observed elevations are mainly due to thermogenic sources, and in Lutjewad due to biogenic sources.

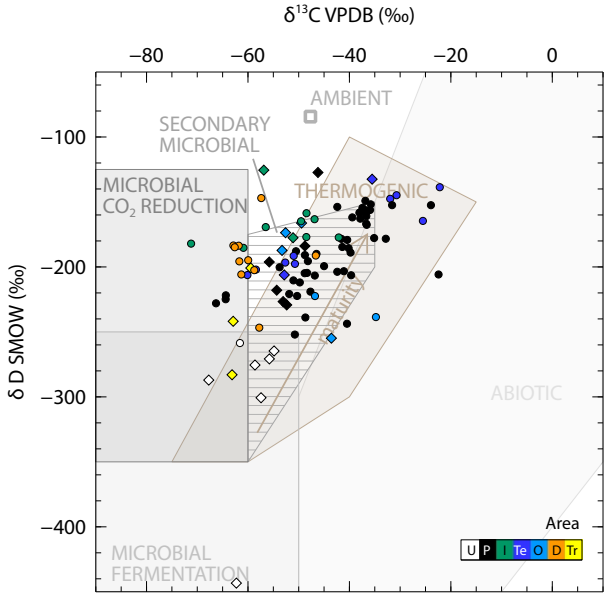

Fig. 20: Dual isotope plot of the resulting source signatures, for the geographical areas we visited using both ground vehicles and aircraft.

During ROMEO, more than 300 discrete air samples for isotope analyses were taken (Menoud et al., in preparation). 18 plumes could be linked to oil and gas extraction activities, and 6 plumes were of unknown origin (Fig. 20). The isotopic signals of the unknown plumes are typical of CH_4 from microbial fermentation, for example enteric fermentation (cattle farming) or waste degradation (landfills, sewage treatment, etc). Detailed analysis of aircraft isotopic signals and corresponding wind directions allowed us to directly link at least 3 plumes from the aircraft to the underlying oil and gas facilities.

The source signatures obtained for 80 individual oil and gas operation sites range from -71.2 to -22.2 ‰ V-PDB, and from -252 to -139 ‰ V-SMOW, for $\delta^{13}C$ and δD respectively (Fig. 20). The distinction of emissions from fossil fuel operations is mainly possible using δD signatures, because the wide range of $\delta^{13}C$ values overlap with typical signatures of other types of sources (biogenic, pyrogenic). In several regions, the lowest $\delta^{13}C$ values are associated with gas and condensed gas extraction plants, suggesting the presence of microbial gas formations. To our knowledge, CH_4 of microbial origin was not reported in Wallachian plain deposits until now. The results generally help understanding the geology of the region, as well as significantly improving the knowledge of the isotopic signatures of CH_4 emissions in Romania, especially from oil and gas installations.

Isotopic measurements do not only increase the knowledge of local sources and conditions. Measurements across the Upper Silesian Coal Basin (USCB) aimed on a better understanding of CH_4 origin (since different coal mine shafts can have different CH_4 isotopic signatures) and to assign an isotopic signature to the USCB region and particularly coal mine shafts (Stanisavljević et al., in preparation). The δD signature found in the USCB is distinct from emissions in other main world sources, such as coal deposits in Australia (Surat Basin), where δD values are much lower (-202 to -238 ‰) than those determined in the USCB during this study (range: (-143 ± 1.5) to (-267 ± 60) ‰). The δD values found in the USCB region also differ from Chinese coal mines (Liangping region), where isotopic signatures are more enriched than at the USCB, revealing a clearly dominant thermogenic origin in China.

In the UK, the isotopic signatures of biogas plants (Bakkaloglu et al. 2021), composting facilities and landfills were investigated (Bakkaloglu et al., in preparation). Results reveal greater ^{13}C depletion than in other waste sources. As the Net Zero Commission recommends that all biodegradable waste being sent to landfill sites in the UK will be banned within the next five years and more waste will be diverted to biogas and composting facilities (CCC, 2020), the isotopic signatures of waste emissions may change in coming years due to variation in waste management strategies.

This shows, that it is not only useful to distinguish signatures of different regions or sources, but that continuously measuring and monitoring is necessary for future modelling studies and up-to-date emission inventories.

Beside atmospheric measurements, MEMO² made an excursion to the marine environment. During a measurement campaign to the North Sea in 2018, the origin of CH4 above an active cold seep at the Doggerbank was investigated (de Groot et al. 2019). Fig. 21 shows depth profiles of $\delta^{13}C$ and δD taken during a 2-days' time series at the Doggerbank.

The isotopic signature from the remaining methane, that is not directly oxidized by methanotrophs during transport trough the water column indicates that at lower concentrations of methane (less seep activity) a shift occurs in δD as well as δ^{13}C that was linked to mixing with atmosphere. In contrast with high seep activity a clear isotopic source signal dominated the water column and was linked to biogenic methanogenesis. The results are close to publication and indicate the variability in activity of a cold seep and the importance of high frequency sampling above such dynamic systems.

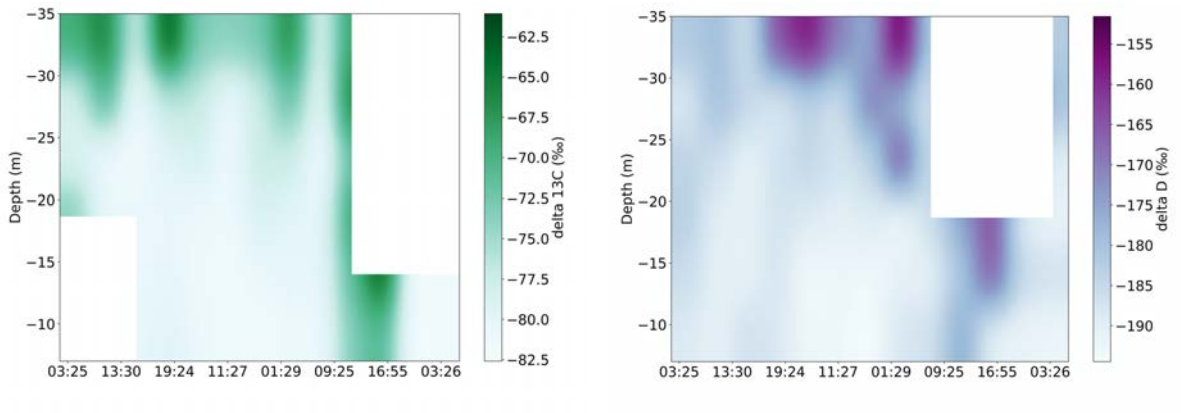

Fig. 21: δ^{13}C (left) and δD (right) during a 3-days' time series at the Doggerbank, North Sea

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

A multi-scale framework for interpreting CH_4 observations has been used in the modelling part within MEMO², aiming at the interpretation of mobile observations as well as on the estimation of emission fluxes at different scales. A variety of models were used, compared, and improved.

On the smallest scale, targeting dispersion of plumes right after emissions were investigated. Close to a source, variability of the observations (and simulations) are mostly due to meandering wind patterns that sweep the plume over a measurement device. Thus, observations are characterized by a strong "on-off" nature: either the plume is sampled or not. Further away from the source, atmospheric turbulence has had the time to disperse the concentrated plume patches further. This behaviour can be well quantified using numerical models, and this basic understanding is important for the interpretation of field measurements. To help the interpretation of mobile measurements, the CFD code MicroHH (www.microhh.org, van Heerwaarden et al. 2017) was used, which is more advanced than a simple Gaussian Plume Model to get information about source variability.

The aim was to validate the Gaussian Plume Model (GPM) approaches, such as OTM33a and drive-by´s. The numerical tools have been applied to measurements made during the ROMEO campaign (Ražnjevic et al., a & b, in preparation). By constraining the local meteorological situation (wind speed, direction) to observations, the tool provides a useful addition to more conventional models to estimate the source strength, such as Gaussian Plume models.

Fig. 22 shows an hourly averaged top view of a plume that should be comparable to the GPM. However, we found that lateral dispersion of the GPM is much faster. This is due to the larger dispersion coefficients, which are based on Briggs (1973) (according to the OTM33a protocol). In our analysis we found that dispersion of a plume can be described by two processes: turbulent mixing and plume meandering. The first process is the "classical" turbulent mixing, in which eddies slowly mix down the plume in a diffusion-like process (when averaged).

The second process represents the movement of the whole plume by the larger eddies in the flow. Near the source the meandering seems to dominate. Thus, close to the source the plume is keeping its shape

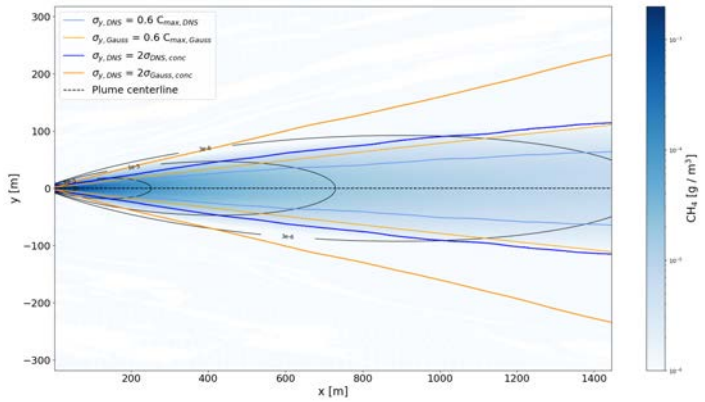

Fig. 22: One-hour time-averaged DNS plume at 60 m height. Isopleths of concentration show the GPM result at the same height with the mean wind at the release height. Straight lines denote the edges of the two plumes.

dictated by the shape of the source itself and the dispersion of the plume might be too small to use methods which rely on the GPM (Fig. 22).

The drive-by strategy was also tested. From the results it is clear that integrating the downwind concentration pattern can result in a correct source estimate. However, at least five drive-by experiments should be averaged to correctly infer the source strength.

As MicroHH simulations in the above described set-up were not able to simulate observed plume meandering, on which the OTM33a technique relies, the use of MicroHH in LES mode has been investigated. The use of these tools in interpreting atmospheric plumes is an emerging science field, because more and more sensors become available that can sample atmospheric concentrations downwind of point sources with high accuracy. The ROMEO campaign was therefore a perfect target. During the campaign, about 40 plume transects were measured downwind of oil wells. To verify the methane emissions, also tracer release experiments using a known source of N_2O were implemented. Moreover, the wind characteristics were measured.

The difficulty in estimating source strength in the field is illustrated in Fig. 23, which shows snapshots of LES simulations. Due to a lack of large-scale forcing (i.e. low wind speed), the simulated plumes behave in a rather chaotic way.

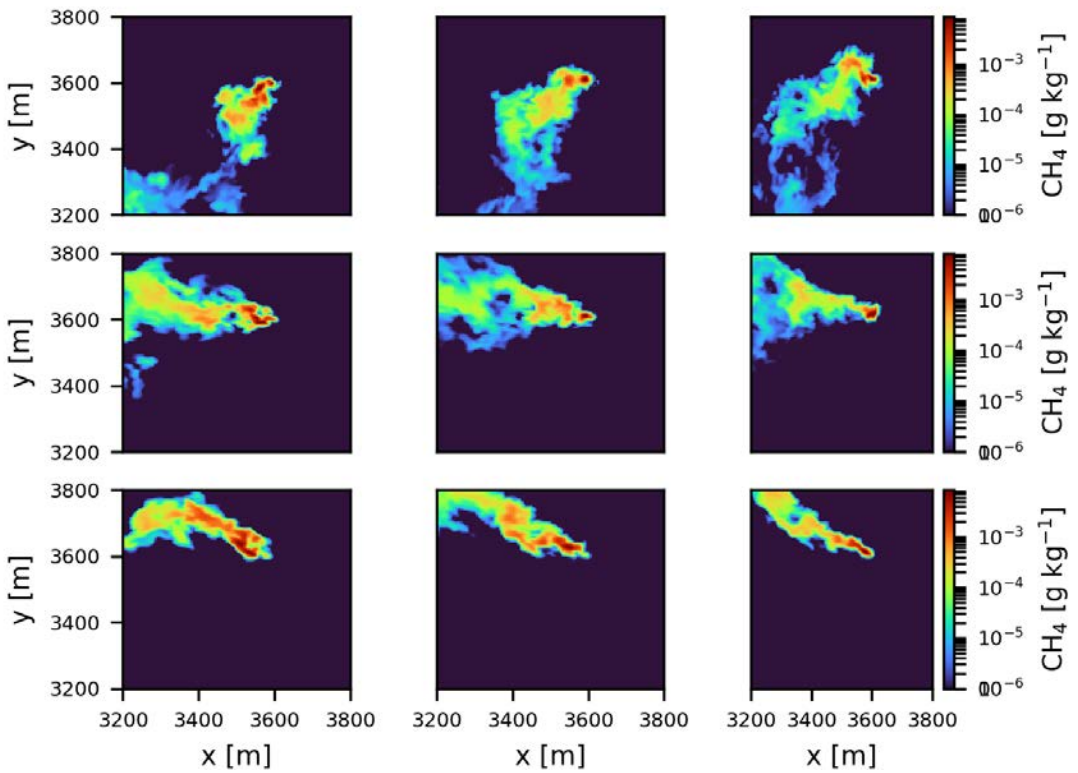

Fig. 23: LES simulation (resolution 5m x 5m x 2m) of the dispersion of methane from a gas well point source located at (x,y) = (3600m,3600m). Plumes were sample at z = 3m.

To solve this issue, plumes calculated with fixed background wind were rotated in order to match the observed wind speed and direction. After this, the source strength is determined using the mass-balance approach. Fig. 24 shows the half-hourly averages of the measured N_2O and CH_4 transects, as well as the corresponding LES results for passive tracer dispersion (here there is no difference between N_2O and CH_4).

The red line is the average of the four half-hourly values. The insets show the measured and modelled wind-speed and direction. Note here that specifically the wind direction is different (by design of the numerical experiment in which the background wind direction was forced to 270 degrees).

Using mass-balance, the source strength of the N_2O source was estimated as 0.53 g s-1, which is ~10 % smaller than the true source strength of 0.59 g s-1. The mass-balance estimate for the CH_4 source amounts to 1.11 g s-1 (corrected for the N_2O-based underestimate: QCH_4 = 1.23 g s-1). This example from the ROMEO campaign shows that LES simulations can be used to quantify source strengths, even when no co-emitted tracer is available.

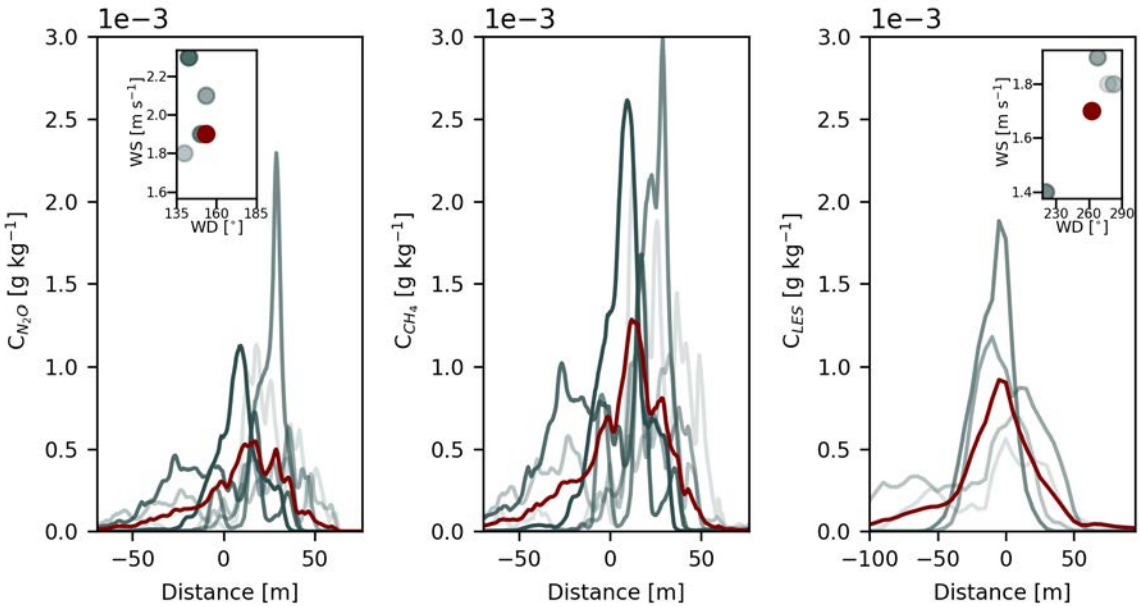

Fig. 24: Averages of instantaneous plumes over periods of half hour from (left) measured N_2O, (middle) measured CH_4 and (right) simulated LES tracer. LES transects were taken at 3 m height and 78 m downwind from the source. Plumes are shown with a color gradient corresponding to the half-hour increments i.e. lightest gray plume is the average of plumes measured in 11.30 - 12.00 UTC, dark grey is the average over 14.00 - 14.30 UTC. The insets show horizontal wind speed and direction for the corresponding half-hour averages in (left) measurements and (right) LES. Over-plotted in red are the averages of all the plumes.

To optimally exploit measurements with unmanned aerial vehicles, the GRAL (Graz Lagrangian Model) dispersion model has been applied. It aimed to improve or validate bottom-up emission inventories using high-resolution dispersion simulations of methane for the investigation of single sources. Moreover, a novel strategy of methane flux quantification with the use of unmanned aerial vehicle (UAV) equipped with a methane sensor has been developed. The methods were evaluated in tracer release experiments with known emission rates and applied to quantify emissions from oil and gas wells in Romania as measured during the ROMEO campaign

Unmanned vehicles allow a complete mapping of the spatial and temporal variability of emission plumes within a short period. A drone equipped with a QCLAS system was employed to sample a controlled release downwind. In a next step, the measured mole fractions were gridded in a single plane, and geostatistical interpolation (Kriging) was applied (Fig. 14, 15), to spatially fill the gaps within the plane.

After filling in the gaps the source strength was estimated, using the cross-sectional area (m^2) and mean streamwise wind profile (m/s) obtained from the 3D sonic anemometer.

To simulate effects of point sources at European scale, it was proposed to use Lagrangian Particle Dispersion Models (LPDM) and to perform atmospheric inversions. Concerning the use of LPDMs, simulation results from FLEXPART-COSMO were compared with observed ambient methane isotopic composition (e.g. at Lutjewad, NL, Fig. 20). FLEXPART-COSMO is a model version of FLEXPART, an offline LPDM, which uses the output of the mesoscale numerical weather prediction model COSMO as the driving meteorology. In this model version, all meteorological fields are preserved on the original COSMO vertical grid which, compared to other versions of FLEXPART, strongly reduces interpolation errors.

Using this model, continuous timeseries of CH_4 mole fraction as well as the isotopologues $\delta^{13}C$-CH_4 and δD-CH_4 in ambient air at the Lutjewad station located in the Netherlands were simulated. Fig. 25 shows an example in which measurements are compared to simulation results using three different emission inventories.

The results indicate a qualitative agreement between in-situ measurements of isotopic composition of CH_4 in ambient air and simulated CH_4 isotopic composition using FLEXPART-COSMO. The agreement between the measured and simulated values is a proof-of-concept that continuous high-resolution isotopic data can be used to better constrain source type information on a regional scale.

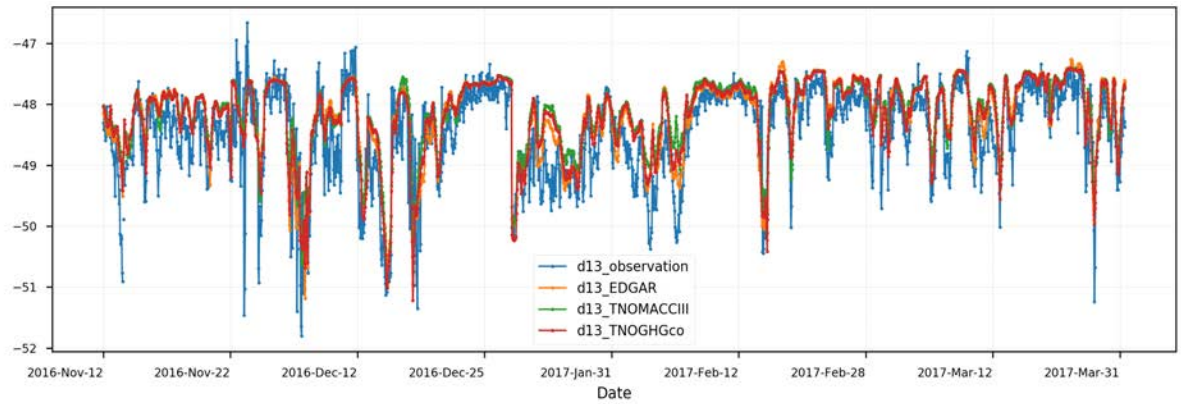

Fig. 25: Simulated isotopic composition of methane ($\delta^{13}C$-CH4) compared to observed values obtained from the Lutjewad station in the Netherlands.

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. With this model, time series of atmospheric $\delta^{13}C$ and $\delta^{2}H$ isotope ratios are computed with an hourly temporal resolution based on simulated CH_4 mixing ratios for the four main CH_4 source categories: agriculture, waste, fossil fuel related emissions and wetland emissions.

Atmospheric inversions make it possible to obtain top-down emissions which represent the best knowledge, including the information from both bottom-up emission inventories and atmospheric measurements. To prepare for atmospheric inversions, much effort has been dedicated to the error estimates. These errors are important, because they will finally determine how much information can sensibly be extracted from measurements, depending on the sampling location, and the capability of the model to simulate the measurement at that location. Using a dedicated set of simulations, the following errors are determined:

- The representation error ε_{repr}: error of a model not perfectly representing the measured values due to the difference between a grid cell in the model and the actual scale at which a measurement is representative.

- The boundary condition error ε_{LBC}: the background error associated with the lateral boundary conditions (LBCs), due to the choice of the lateral (sides and top of the domain) and initial conditions.
- The transport error ε_{transp}: error that is due to discretizing the model of the fundamental equations of the atmospheric transport used in a model.
- The emission induced error ε_b: error that is due to the misrepresentation of emissions on the spatial and temporal grid of the model.

Fig. 26 shows a simulation of the atmospheric mixing ratio of CH_4 over Europe using several bottom-up anthropogenic emission inventories of CH_4. The simulations were used for estimating errors for atmospheric inversions of CH_4 emissions over Europe.

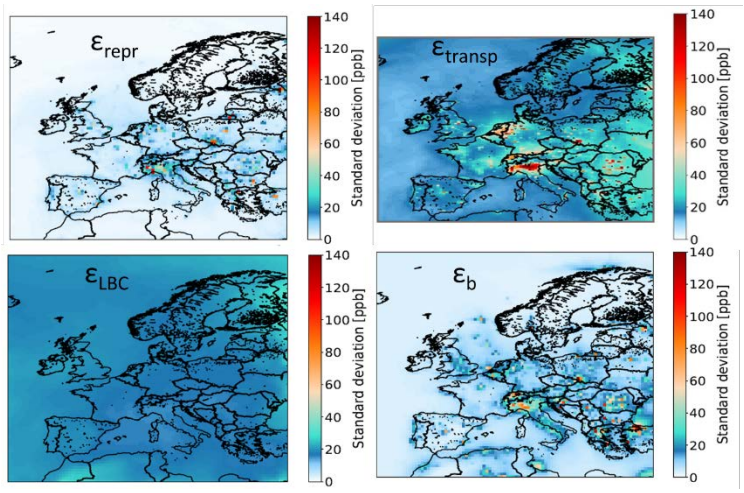

Fig. 26: Annual mean standard deviation of the estimated errors over the European domain for 2015

Also, other possible causes for misfits between measured and simulated atmospheric CH_4 mixing ratios, as well as isotopic ratios $\delta^{13}C$ and $\delta^{2}H$ and its required precision, were investigated to constrain European methane sources.

As an example, Fig. 27 shows the results of the sectoral discrimination at Lutjewad (Szénási et al. 2021, in preparation). Simulated CH_4 mixing ratios were taken from the anthropogenic and wetland emission sources, as well as the total CH_4 mixing ratios without taking the background into account. The contribution of each emission sector was compared to the total contribution in relative terms, which showed that 40 – 60 % of the simulated mixing ratios above the background is due to the agriculture sector.

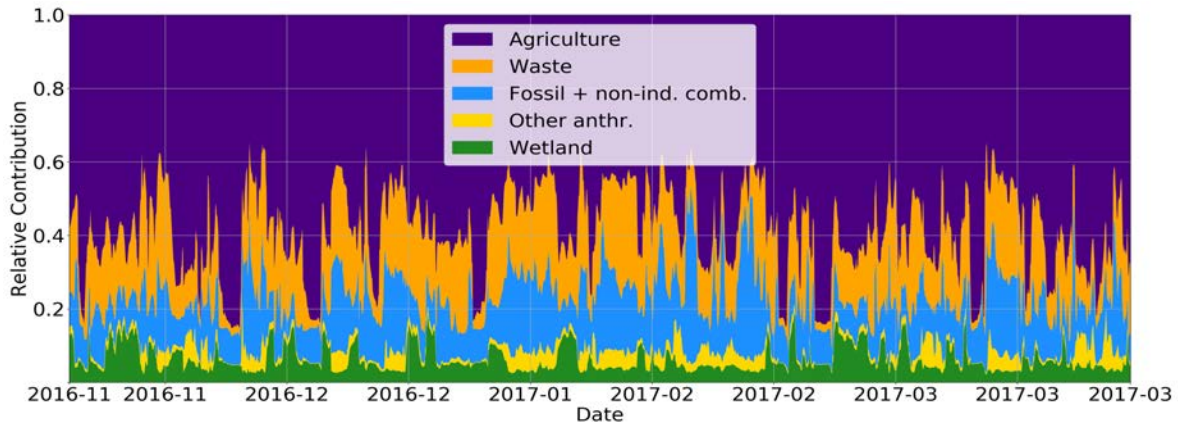

Fig. 27: Sectoral discrimination of CH_4 mixing ratios above background at Lutjewad. The emissions from TNO-MACC_III were used to simulate the sectoral contributions of CH_4 by CHIMERE that are shown in this figure.

In a further study it was investigated, through experiments with synthetic data, what could be the added value of high-frequency $\delta^{13}C$ and $\delta^{2}H$ data to be assimilated in atmospheric inversions, alongside with CH_4 mixing ratios, to improve CH_4 emission estimates. The analysis reveals that the 0.2 ‰ and 5 ‰ precisions for $\delta^{13}C$ and $\delta^{2}H$ (WMO recommended targets) are sufficient for detecting sources only from large emitting areas, such as the Po-Valley in Italy or the Silesian coal basin in Poland, during about six months of the year.

The analyses demonstrate that mainly background and agricultural sources would be detectable e.g. at most ICOS sites, long-term greenhouse gas monitoring stations in Europe. The sites Steinkimmen (DE), Cabauw (NL), Lutjewad (NL) and Ispra (IT) have been implied by simulations with both inventories as potential sites for signal detection during at least six months in a year. However, areas around the first three sites are known for large emissions originating from agricultural activities and thus the main detectable confirmed source at these sites is agriculture. At Ispra, waste sources are the dominant detectable source. The sites Krešín u Pacova (CZ) and Lindenberg (DE) appear to be promising for the detection of fossil fuel related sources for about a month.

In conclusion, δ^2H and $\delta^{13}C$ data sets can be implemented in atmospheric inversions of CH_4 emission over Europe, once a sufficient amount of δ^2H and $\delta^{13}C$ data with sufficient instrument precision are available. However, even higher precisions than 0.02 ‰ for $\delta^{13}C$ and 0.5 ‰ for δ^2H may be necessary to detect sources. This may be the case especially for wetland and fossil fuel related sources.

This work demonstrates that information gained from isotopic measurements within MEMO², in addition to measurements of CH_4 mixing ratios, can be valuable for evaluating emission inventories and estimating emissions by atmospheric inversions. Isotopologues are useful to discriminate sector contributions of CH_4 emissions as the isotopic composition varies highly depending on the source type. For instance, the signature is different depending on whether CH_4 is produced by thermogenic (e.g. natural gas), biogenic (e.g. domestic animals, landfills, wetlands) or pyrogenic (combustion) sources. As a result, comparing the measured and simulated CH_4 mixing ratios and isotopic composition can provide additional information about the sources that contribute to a particular measurement.

5. Dissemination and exploitation

The consortium was actively communicating and disseminating MEMO² as an EU project in general and by presenting scientific results of the individual ESR projects (https://h2020-memo2.eu/dissemination/). The consortium used different platforms to approach different target groups, mainly scientific conferences and several social media channels, aiming on both communicating the individual scientific projects within MEMO², and also MEMO² as itself. By this MEMO² increased the impact of the consortium as such and the understanding of the project type H2020-ITN-ETN in general.

Project related documents such as deliverable and milestone reports (https://h2020-memo2.eu/memo2-deliverables/) were prepared in a way that they create added value for the scientific community. Examples are the development of harmonised methods for mobile measurements, isotopic measurements linked to common scale or compare forward simulations by regional models.

With more than 100 conference contributions of only the ESRs (as author or co-author) the project was highly visible within the scientific community, which lead to several interactions and collaborations.

25 scientific publications have been published already, or are close to submission (see list below, MEMO² participants in bold, * mark datasets):

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8. Hoheisel, A., Yeman, C., Dinger, F., Eckhardt, H., and **Schmidt, M.:** *An improved method for mobile characterisation of δ13CH4 source signatures and its application in Germany*, Atmos. Meas. Tech., 12(2), 1123-1139, https://doi.org/10.5194/amt-12-1123-2019, 2019
9. **Korben, P.,** Jagoda, P., **Maazallahi, H.,** Kammerer, J., **Necki, J.M.,** Bartyzel, J., Radovici, A., **Schmidt, M., Röckmann, T.,** and other ROMEO team members: Quantification of methane emission from oil and gas wells using Other Test Method - 33a and Gaussian Plume Model during ROMEO campaign, Romania, in preparation
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As usual, the publication of scientific results lasts longer than the lifetime of a project and the list of publications is expected to extend over the next months. Although MEMO² has officially ended in February 2021, the website will remain active and maintained, respectively updated.

6. Conclusions and socio-economic impact on the project

Despite a few adjustments of the initial planning, which is common for innovative scientific projects, MEMO² delivered a high number of verified data from almost 800 individual field measurement days, improved methodologies and technologies, and resulted in a broad and stable long-term scientific network. These achievements have a high impact on several levels.

6.1 Impact of MEMO² on personal level for the ESRs

Participation in MEMO² had a beneficial impact on the career perspectives and employability of the ESRs, and on their professional skills including networking, interdisciplinary and intercultural collaboration. They were trained on cutting-edge research in an international consortium.

ESRs were obliged to execute at least two secondments and participate in two scientific conferences. By this they improved their presentation skills and broadened their scientific network. They had to report about their experiences by public blogs on the project website (https://h2020-memo2.eu/category/blog/), which did not only increase the visibility of their projects but also enhanced their writing skills towards a public audience.

Despite the fact that secondments were time consuming and required quite some effort, the majority of secondments, both at academic and non-academic partners, where experienced useful, inspiring, and highly appreciated. Two out of three ESRs who recently finished their thesis started already a follow-up contract at one of their former secondment partners.

6.2 Impact of MEMO² on training and community level

All ESRs had to follow individual training at their host institutions, on network level and on international level. This included the above-mentioned secondments and scientific conferences, but also MEMO² workshops and schools, or courses required from their host institutions.

Altogether, the ESRs got a broad spectrum of opportunities to learn not only about their own topic but to place their research in a broader context and at the same time collaborate with experienced researchers in their field. These opportunities and particularly the associated scientific network were additional benefits compared to traditional stand-alone PhD projects. Several training events of MEMO² were also available for students outside the consortium, which offered additional networking options.

An additional benefit in the context of network wide training events was the fact that MEMO² training was giving in English. Topic related courses were often only offered in national languages which is an obstacle for international students. By following these network training events the ESRs at the same time improved their language skills.

MEMO² was not only beneficial for the networks of the ESRs. It also brought experienced scientists of different disciplines together. It helped to better combine practical field work with modelling, preparing data for different scientific communities and foster understanding for discipline related methodologies and approaches. The MEMO² network has collaborated closely during the lifetime of the project and is currently preparing several follow-up activities. Based on the MEMO² achievement, we want to further invest in the combination of scientific measurements in the atmosphere and the 'bottom-up' estimates of emissions from local sources that are used in the national reporting. The long-term collaborations build in projects as MEMO² will support the EU in finding the required mitigation measures to massively reduce greenhouse gas emissions.

6.3 Impact of MEMO² on scientific level

MEMO² was the first large-scale project in the EU that investigated the use of novel fast high precision sensors on mobile platform at various locations throughout Europe and in large international campaigns. The results are highly positive and demonstrate that added value of such measurements that can be carried out close to sources, rather than at larger distances where the traditional atmospheric monitoring programs operate. In this respect, we successfully used ground-based vehicle, drone and aircraft measurement for emission quantification at various scales, from local to regional.

MEMO² provided a huge amount of new, high-quality data sets. The consortium not only carefully compared data (e.g. isotopic data linked to common scale, D2.1), but produced also detailed error estimates for parts of the modelling (Szenasi et al., in preparation) and prepared guidelines for harmonised measurements (D1.5).

The MEMO² activities allowed in-depth studies of CH_4 emissions from various sources in Europe by using different platforms and methodologies, e.g. in the agriculture sector, the coal, oil and gas industry, cities, landfills and wastewater, or natural environments such as arctic wetlands. The MEMO² data made a significant contribution to improving the data situation and foster a better understanding of CH_4 emissions from these sources. In this context particularly, the joint measurement campaigns have a high impact as they offered unique opportunities to not only collect data, but also to directly compare sampling and evaluation methodologies. The overall activities of data collection are listed in the individual ESR reports in the 2nd Periodic Report. The reports and guidelines are public available on the website (https://h2020-memo2.eu/memo2-deliverables/).

MEMO² contributes also to the idea of open access as data, collected on the ICOS data portal, will be publicly available soon after the project has ended. The isotopic characterization of methane sources in Europe performed within MEMO² is already gathered into an openly available database (Menoud et al. 2020, https://doi.org/10.5281/zenodo.3975543). The results, a joint effort of several ESRs, represent a substantial new contribution to the existing isotope datasets and improve significantly our understanding of the isotopic composition CH_4 sources in Europe. This will help in the development or improvement of mitigation strategies and recommendations to decrease CH_4 emissions from distinct sources. Better availability of integrated isotopic data such as $\delta^{13}C$ and δD may be e.g. effectively used to evaluate specific waste sources or emissions from city gas networks at regional level, improve on a national level the emission inventories and at global scale contribute to better atmospheric models of the CH_4 budget.

MEMO² innovated in the use of UAVs in combination with different estimation and interpolation methods for emission quantification, and different wind assumptions clearly show that wind conditions and sampling distance are crucial parameters for drone quantification methods to be successful. The development of a high-precision analyser that can be operated on drones is another key scientific achievement of MEMO².

Comparisons between Direct Numerical Simulations and Large Eddy Simulations targeting plume dispersion right after emissions revealed that LES with realistic atmospheric conditions has provided key new insight into the limitations of the standard tools that are used for emission quantification in the field.

Regarding the use of isotope data for improving methane budgets, emissions errors were quantified by using two anthropogenic emission inventories and investigating their impact on mixing ratios and isotope ratios. These simulations demonstrate that isotopologue measurements of CH_4 are a great asset for determining the origin of atmospheric CH_4. It was also shown, through experiments with synthetic data, that (potentially new) high-frequency measurements of $\delta^{13}C$ and δD are important to better constrain CH_4 emission estimates. Although these high-frequency measurements are not yet available at sufficient locations, this study clearly show the need of a precision of at least 1 ‰ for δD analyses and 0.05 ‰ for $\delta^{13}C$ analyses to discriminate methane sources at most ICOS sites, which form the backbone of the European greenhouse gas monitoring infrastructure.

In conclusion, regarding results and general output, MEMO² was a highly successful project. We note that the Research Executive Agency (REA) declared MEMO² officially a “Success Story”.

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