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## **Table of contents**

1. Overall summary	7
2. MEMO <sup>2</sup> – Explanation of work and overview of progress	
2.1 Objectives	7
2.2 General progress of the Work Packages	8
2.2.1 WP1 – Mobile measurements of CH₄ (Lead: Martina Schmidt, UHEI)	8
2.2.2 WP2 – Source identification by isotopic characterization	
(Lead: Dave Lowry, RHUL)	
2.2.3 WP3 – Modelling: A multi-scale interpretation framework for CH₄ observations (Lead: Maarten Krol, WU)	. 22
2.2.4 WP4 – Training (Lead: Philippe Bousquet, UVSQ)	. 27
2.2.5 WP5 – Project Management (Lead: Thomas Röckmann, UU)	. 28
2.2.6 WP6 – Ethics (Lead: Thomas Röckmann, UU)	. 28
2.3 Deliverables	.29
2.4 Milestones	.31
2.5 Impact	. 32
2.5.1 Impact on ESRs	. 32
2.5.2 Impact on training	
2.5.3 Impact on science	. 33
2.6 Evaluation of the Action by the external Supervisory Board (SB)	. 38
3. Communication, dissemination and exploitation	40
3.1 Scientific platforms	.40
3.2 Internet-based platforms	
3.2.1 MEMO <sup>2</sup> website	
3.2.2 Social media	
3.2.3 Data repositories	
3.3 General communication and dissemination activities	.44
4. Management of MEMO <sup>2</sup>	45
4.1 General overview of the management	.45
4.2 Recruitment	.45
4.3 Consortium	.46
4.4 Meetings	.46
4.5 Communication infrastructure	.48
4.5.1 Email, telephone	. 48
4.5.2 Web-based board meetings and tele-conferences	
4.5.3 Face-to-face meetings	
4.5.4 Participant Portal	
4.6 Risk assessment and faced difficulties	.49
5. Outlook 3 <sup>rd</sup> Reporting Period	49



D5	9	MEM	$O^2 -$	Midterm	Review	Report
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6. Individual ESR reports	50
6.1 ESR1 - Monitoring the methane emissions from different sources in Germany	50
6.1.1. Scientific progress	50
6.1.2 Deliverables	53
6.1.3 Training and network activities	53
6.1.4 Dissemination activities	55
6.2 ESR2 - Quantifying CH4 emissions using measurements on cars and UAVs in the	
Netherlands	56
6.2.1. Scientific progress	
6.2.2 Deliverables	
6.2.3 Training and network activities	62
6.2.4 Dissemination activities	64
6.3 ESR3 - Validating $CH_4$ inventories over intense mining area, natural and	
anthropogenic emissions	
6.3.1 Scientific progress	
6.3.2 Deliverables	
6.3.3 Training and network activities	
6.3.4 Dissemination activities	
6.4 ESR4 - Assessing CH <sub>4</sub> emission from wetlands and other sources by use of mob	
measurements	
6.4.1 Scientific progress	
6.4.2 Deliverables	
6.4.3 Training and network activities	
6.4.4 Dissemination activities	
6.5 ESR5 - Characterizing CH <sub>4</sub> emissions in urban environments (Paris)	
6.5.1 Scientific progress	
6.5.2 Deliverables	
6.5.3 Training and network activities	
6.5.4 Dissemination activities	99
References	99
6.6 ESR6 - Mid-infrared laser spectroscopy for three dimensional CH4 mapping by	
unmanned aerial vehicles (UAV)	
6.6.1 Scientific progress	
6.6.2 Deliverables	
6.6.3 Training and network activities	
6.6.4 Dissemination activities	
6.7 ESR7 - CH <sub>4</sub> from waste: constraints on captured and fugitive emissions from isot	
analysis	
6.7.1 Scientific progress	
6.7.2. Deliverables	109



MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

D5.9 MEMO<sup>2</sup> – Midterm Review Report

	6.7.3 Training and network activities	110
	6.7.4 Dissemination activities	112
	6.8 ESR8 – Isotopic characterisation of methane sources in Europe	113
	6.8.1 Scientific progress	113
	6.8.2 Deliverables	126
	6.8.3 Training and network activities	126
	6.8.4 Dissemination activities	128
	6.9 ESR9 - The isotopic signature of urban CH4 emissions	129
	6.9.1 Scientific progress	129
	6.9.2 Deliverables	133
	6.9.3 Training and network activities	134
	6.9.4 Dissemination activities	136
	6.10 ESR10 – Integration of mobile measurement data in monitoring, reporting, and	
	verification (MRV) of key CH <sub>4</sub> sources in GHG emission reporting across Europe	
	6.10.1 Scientific progress	
	6.10.2 Deliverables	
	6.10.3 Training and network activities	
	6.10.4 Dissemination activities	
	6.11 ESR11 – High-resolution modelling of CH4 dispersion	
	6.11.1 Scientific progress	
	6.11.2 Deliverables	
	6.11.3 Training and network activities	
	6.11.4 Dissemination activities (March 2017 – February 2019)	151
	6.12 ESR12 - Inverse modelling of CH <sub>4</sub> and its isotopic composition at European and point source scales	152
	6.12.1 Scientific progress	
	6.12.2 Deliverables	
	6.12.3 Training and network activities	
	6.12.4 Dissemination activities	
	6.13 ESR13 - Atmospheric monitoring of the CH <sub>4</sub> emissions at the European scale	
	6.13.1 Scientific progress	
	6.13.2 Deliverables	
	6.13.3 Training and network activities (March 2017 – February 2019)	
	6.13.4 Dissemination activities	
7	History of the deliverable	
		. 100

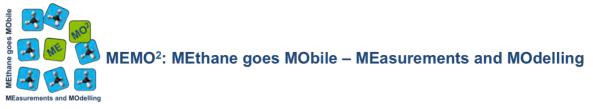


### Abbreviations

AirCore	Atmospheric Sampling System
AGH	Akademia Gorniczo-Hutnicza im. Stanislawa Staszics w Krakowie, Krakow, Poland
C <sub>2</sub> H <sub>6</sub>	Ethane
CH <sub>4</sub>	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
	Pan-European research infrastructure for quantifying and understanding the greenhouse gas balance
	of Europe and its neighbouring regions
InGOS	Research infrastructure targeted at improving and extending the European observation capacity for non-CO <sub>2</sub> 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 ( <u>https://cordis.europa.eu/project/rcn/91167/factsheet/en</u> )
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



Figures and Tables are numbered continuously per chapter.



## 1. Overall summary

This Midterm Review report is updating the 1<sup>st</sup> Progress Report and describes the outcome of work performed within MEMO<sup>2</sup> during the first two reporting periods of the project (1<sup>st</sup> Period March 2017 – February 2018, 2<sup>nd</sup> Period March 2018 – February 2019) and references to the overall progress accordingly to the Grant Agreement.

Up to now the project is running smoothly and successfully. All beneficiaries and partner organisations are engaged and committed to the project as initially planned or even more. The recruitment took slightly longer than expected, but all ESRs were starting within the first eleven months of the project. One ESR decided due to personal reasons to stop after 16 months, and the position has been re-opened in agreement with the project officer. The position has been refilled by an ESR who started on 1 December 2018.

In total 16 deliverables and 10 milestones were due in these reporting periods. All of them were submitted in time or just slightly delayed. Delay was mainly caused due to the delay in recruitment and the resulting shift of work executed.

Scientifically, the first year was dedicated to starting the project and its collaborations, to set up the management and infrastructure, and to design / execute first network activities such as the 1<sup>st</sup> MEMO<sup>2</sup> school and several (joint) measurement campaigns. In the second year we gained first results and intensified scientific collaborations within and outside the consortium. Up to now three additional partner organisations (ECCC, GEOMAR, and DTU) joined the network. The 2<sup>nd</sup> MEMO<sup>2</sup> school is scheduled at the end of the second year, dealing with "Methane and society.

The main scientific achievements during the first reporting periods are

- Intensive data collection during, participation in and organisation of several joint measurement campaigns (Table 2.14)
- Bevelopment and first tests of UVA devices carrying lightweight CH<sub>4</sub> sensors and AirCores
- Execution of intercomparison measurements to ensure common scales used within MEMO<sup>2</sup>
- Implementation of data and modelling devices such as MicroHH and CHIMERE
- Drganisation of several network wide training activities, such as workshops and MEMO<sup>2</sup> schools

MEMO<sup>2</sup> is actively disseminating results and network activities by using several platforms, such as the project website, scientific conferences and social media (see Chapter 3). All are getting more and more attention, and our ESRs are encouraged to present their projects to the broader public and scientific community.

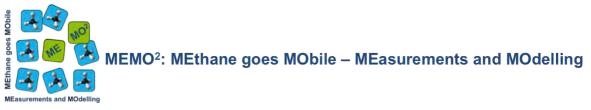
## 2. MEMO<sup>2</sup> – Explanation of work and overview of progress

## 2.1 Objectives

Within MEMO<sup>2</sup> we defined several objectives to target our main goals.

**The main scientific** goal of MEMO<sup>2</sup> is to I) 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 use these updated emissions to III) improve estimates at the European scale. These tools will 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** is to educate a generation of "cross–thinking" scientists that will be able to effectively develop and use novel measurement and modelling tools in an interdisciplinary and intersectoral context. A dedicated training program includes original actions to reinforce the autonomy (learning by doing approaches) and the maturity (student autonomous virtual network) of the MEMO<sup>2</sup> early stage researchers (ESRs). This ensemble of training actions helps them to refine their career plan, either within the scientific community, or in the non-academic sector.



The following **MEMO<sup>2</sup> objectives** were defined:

- Implementation of a mobile CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub>
- 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<sub>4</sub> emission map, including isotopic information, across Europe
- Derivation of top-down emission estimates over Europe exploiting the new information acquired in MEMO<sup>2</sup>.
- Fostering a close collaboration between the academic and the non-academic sector by joint network activities

Within the first two reporting periods the focus laid on I) the recruitment of the 13 Early Stage Researcher (ESRs) and their training at the host institutions, II) the implementation of measurement networks, III) the development of new measurement systems and networks, and modelling tools, and IV) the augmentation and implementation of training and collaboration activities inside and outside the consortium. The following chapters describe the progress towards the goals and objectives.

## 2.2 General progress of the Work Packages

MEMO<sup>2</sup> is organised in five work packages (WPs): three scientific WPs, one training WP and one management WP. Within the first reporting periods only small changes or adjustments were made, e.g. a change of location of the first MEMO<sup>2</sup> school or shifts in the secondments to ensure their usefulness for the ESRs. The changes had no significant impact on the project, neither scientifically nor administratively.

#### 2.2.1 WP1 – Mobile measurements of CH4 (Lead: Martina Schmidt, UHEI)

#### 2.2.1.1 General WP overview and contribution of involved beneficiaries

Within WP1 – Mobile measurements of CH<sub>4</sub> (including primarily the ESRs 1-7 from the beneficiaries UHEI, RUG, AGH, LU, UVSQ, EMPA, and RHUL) – we will identify, quantify and monitor CH<sub>4</sub> plumes of major anthropogenic and natural CH<sub>4</sub> emitters in Europe from mobile platforms. The ESRs in WP1 further developed the instrumentation needed for mobile measurements and executed several (joint) measurement campaigns to gather data (see Table 2.14).

One of the joint campaigns were the CoMet campaigns in Upper Silesia in May 2017 and June 2018, (co)organized by **AGH**. The participants, including 5 ESRs from MEMO<sup>2</sup>, performed measurements using mobile platforms (CRDS analyser in cars, planes and with AirCore payload of the drone). FTIR technique was applied with stationary and mobile platform. The mobile campaigns aimed at industrial emission of CH<sub>4</sub> (priority was given to mining activities) over Silesia Coal Basin, Belchatow open pit mine, Lublin Coal Basin and Mirocin gas fields. Additionally, a set of measurements were done in vicinity of closed mines and sealed shafts. The data are currently under evaluation.

During the first two years **EMPA** accomplished preliminary tests for the lightweight sensor on a benchbased system in order to characterize a dedicated custom-built, ring-shaped segmented multipass cell. A mechanically rugged and lightweight mobile measurement device was designed, with a very low overall weight of around 2 kg, which is suitable for the foreseen application on board a UAV. First test flights have been performed using a commercial drone (DJI Matrice 600). During those flights, no impact on mechanical stress was visible. However, some limitations have been observed in thermal control and



the available voltage of the power supply for the Peltier coolers. These aspects are currently being solved before proceeding with the final field characterization.

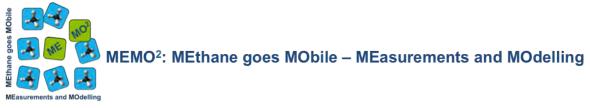
Next to EMPA **RUG** is using drones as a mobile platform to measure CH<sub>4</sub>. A prototype UAV AirCore has been successfully developed and tested by RUG. A validation flight that compares the UAV AirCore measurements and 60-m measurements was performed at the Lutjewad station. Three field campaigns at the Grijpskerk dairy cow farm have been accomplished, showing that the CH<sub>4</sub> enhancement downwind of the farm is significant compared to the background upwind of the farm, and a mass balance approach has been used to derive an estimated emission rate. Besides this, a cheap CH<sub>4</sub> sensor capable of reporting in situ CH<sub>4</sub> mole fractions during flight has been installed and tested.

**UHEI** is also performing mobile CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> measurements using an AirCore, in the region of Heidelberg and during the CoMet campaign in Upper Silesia (Poland). During 10 measurement days, UHEI monitored the CH<sub>4</sub> enhancements downwind of several locations such as a landfill, a biogas plant, two gas compressor stations, and two farms in the region of Baden Württemberg. The isotopic source signature has been calculated. The CH<sub>4</sub> mole fraction measurements will be used to derive CH<sub>4</sub> emission rates with Gaussian plume models during the next months. In Upper Silesia the CH<sub>4</sub> mole fraction and <sup>13</sup>CH<sub>4</sub> from coal mining was measured and the CH<sub>4</sub> source signature from coal mines have been determined. The data on mole fractions are under evaluation in cooperation with partner AGH and UU. In November 2018 UHEI organised the dispersion plume workshop in Heidelberg, bringing together 12 MEMO<sup>2</sup> ESRs and four teachers from EMPA, LSCE, WU and Shell.

The first airborne measurement campaign using the **LU** aircraft was planned over a wetland during July or August 2018. Preparations for the campaign (contact to the authorities, application for a permit to fly over the national park area) had been started at the end of the first year. Unfortunately, no airborne measurement campaign using the LU aircraft could be performed during 2018. This was due to technical problems with the CH<sub>4</sub> gas analyser which needed to be solved before new flights, as well as a late negative answer from the authorities regarding the flight permissions over the natural reserve where the main campaign in 2018 was planned. However, analyses of ground-based eddy-covariance measurements over different Swedish wetlands were performed and showed reduced CH<sub>4</sub> emissions due to the very dry weather conditions. ESR4 continued the analyses of aircraft data over various wetlands in northern Scandinavia from previous campaigns and substitutional activities are in preparation for 2019.

At UVSQ, the mobile platform (installation and measure) has been introduced to the ESRs as well as the analysis tools to handle the data from the mobile measurements (i.e. R, Python codes). Besides this several locations such as a landfill (October 2017), a gas compression site and a farm (January 2018) were sampled (see Table 2.14). Preliminary measurements in the region of Ile de France focused on gas compressor stations and took place in March 2018. First on 05.03.2018 at Limoges-Fourches and second on 13.03.2018 at Fontenay-Mauvoisn. These prior surveys gave the possibility to determine the best weather condition for further mobile measurement according to available infrastructure. Further measurement at these sites and also at another one in Ile de France region will be conducted. The goal of further surveys is to measure the isotopic composition of CH4 and estimate emissions from each source. In September and November 2018, 5 mobile surveys in the Paris urban area were conducted. The first Paris surveys focused on peripheral Paris area. Then mobile measurements were conducted at the main Paris pipeline located along the Seine, which belongs to GRT gas, the French natural gas transmission operator. The rest of Paris was also canvassed. Furthermore, 5 days are planned before end of January 2019 to cover Paris and its close suburbs. UVSQ also participated in measurement campaigns outside of France as the CoMet in Silesia region in Poland (May/June 2018) and the mobile measurement in London (June/July 2018).

The mobile measurement campaigns organised by **RHUL** and **UU** (see Table 2.14) contribute to WP1 by providing samples and data, and are described in chapter 2.2.2.



#### 2.2.1.2 Progress of WP tasks

#### Task 1.1 Surface-based mobile CH<sub>4</sub> measurements on vehicles (Lead: UHEI, Martina Schmidt)

ESRs 1-5 & 7 will use mobile CH<sub>4</sub> analysers on vehicles to monitor and quantify the main emitters in their respective hosting countries, (DE, NL, FR, PL, UK, SE), by performing regular measurement campaigns, in cooperation with non-academic partners. This includes tracer release experiments and multi-tracer analysis. Methodology, data and findings will be shared among the ESRs to assess the EU scale. Event samples will be collected and analysed for <sup>13</sup>CH<sub>4</sub> and CH<sub>3</sub>D at RHUL or UU (cooperation with WP2). ESRs from UVSQ, UHEI and AGH will also carry out in-situ <sup>13</sup>CH<sub>4</sub> source signature measurements.

During the first two years, all groups started their mobile measurements with focus on the region close by the institution. The first year of all ESR project focused mostly in instrumental training and test for the best setup, but also in finding the best measurement strategy to monitor the emission plumes. Herby it was important to investigate the suitability of available roads and the typical wind conditions. A data base of available roads for different wind conditions will help for further planning of measurement campaigns.

With D1.5 the consortium prepared a quideline for harmonisation of measurement technics and good practice. In the guideline we summarise the different equipment used and give advices how the measurements need to be performed and submitted to the ICOS database (see also chapter 3.2.3). Table 2.1 summarises the instrumentation used for CH<sub>4</sub> measurements in vehicles. All partners involved in this task, including RHUL and UU, performed several campaigns to measure CH<sub>4</sub> concentrations in the downwind of CH4 emitters (Table 2.14). UHEI focused on CH<sub>4</sub> sources

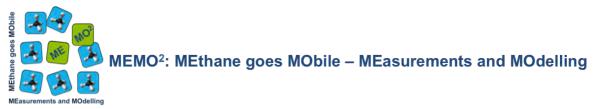
Table 2.1: Analysers used in the  $\mathsf{MEMO}^2$  project for mobile  $\mathsf{CH}_4$  measurements in vehicles.

Laboratory	Analyser Model	Manufacture	Measured species
UHEI	CRDS G2201-i	Picarro	CH <sub>4</sub> , CO <sub>2</sub> , <sup>13</sup> CH <sub>4</sub> , <sup>13</sup> CO <sub>2</sub> , C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> O
LSCE	CRDS G2201-i	Picarro	CH <sub>4</sub> , CO <sub>2</sub> , <sup>13</sup> CH <sub>4</sub> , <sup>13</sup> CO <sub>2</sub> , C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> O
LSCE	CRDS G2203	Picarro	CH <sub>4</sub> , C <sub>2</sub> H <sub>2</sub>
LSCE	CRDS G2402	Picarro	CO <sub>2</sub> , CH <sub>4</sub> , CO
AGU	CRDS G2201-1	Picarro	CH <sub>4</sub> , CO <sub>2</sub> , <sup>13</sup> CH <sub>4</sub> , <sup>13</sup> CO <sub>2</sub> , C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> O
RHUL	CRDS G2301	Picarro	CH4, CO2, H2O
RHUL	OA-ICOS UMEA	LGR	CH <sub>4</sub> , C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> O
RUG	CRDS G2401-m	Picarro	CH4, CO2, CO, H2O
UU	CRDS G2301	Picarro	CH4, CO2, H2O

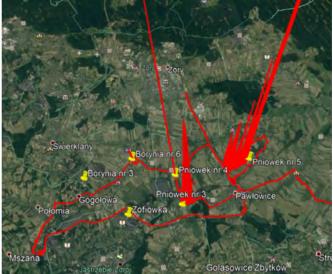
in the region around Heidelberg and monitored the CH<sub>4</sub> enhancements downwind of a landfill, a biogas plant, two gas compressor stations, and two farms in the region of Baden Württemberg. Fig. 2.1 shows a typical measurement campaign, performed in Heidelberg and its surrounding. From North to South the main CH<sub>4</sub> peaks have been detected at a gas compressor station, a farm in the city of Heidelberg, and at a biogas plant. When a peak was detected, several crossings were performed to get a better statistic. A second focus was the CoMet campaign in Upper Silesia (Fig. 2.2). Close to the mine shafts very high CH<sub>4</sub> concentrations of up to several hundred ppm were detected.

Mobile measurements of LSCE focused on gas compressor stations and took place in March 2018. In September and November 2018, 5 mobile surveys in Paris urban area were conducted. AGH focus the campaigns to industrial emission of methane with priority to coal mining activity in the Silesia Coal Basin. In addition, one open pit mine and also a gas field was part of the measurement campaigns.

All data are now under closer evaluation to produce a harmonised data product of the measurements.







**Fig. 2.1:** Typical transect of 6 hours of measurement in Heidelberg and surroundings with concentration peaks at a biogas plant, in the city of Heidelberg, close to a farm and a natural gas compressor station

Fig 2.2:  $CH_4$  concentration measurements in Upper Silesian coal mining region.

#### Task 1.2 Airborne measurements of CH<sub>4</sub> on airplanes and UAVs (Lead: LU, Jutta Holst)

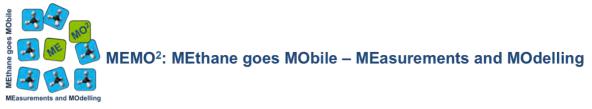
Natural CH<sub>4</sub> emissions from wetlands and lakes will be monitored in Sweden using a small research aircraft from LU. Inflow and outflow CH<sub>4</sub> concentrations of a major source area will be determined at different times throughout the year to derive total CH<sub>4</sub> emissions. Air samples and AirCore (see Task 1.3) samples will be collected using UAVs to map CH<sub>4</sub> plumes. The lightweight CH<sub>4</sub> spectrometer (Task 1.3) will be deployed on a UAV by Empa to study CH<sub>4</sub> emissions of rivers and lakes in Switzerland. Air samples will be collected on the aircraft and on UAVs for isotope analysis (cooperation with WP2).

The airborne measurements using the LU small research aircraft Sky Arrow have been delayed due to hardware problems with the CH<sub>4</sub> analyser and due to late negative reply by the authorities on the originally planned measurement campaign in summer 2018. For 2019 several flight campaigns are planned. Airborne ecosystem fluxes will be measured over a wetland in Uppland during several occasions throughout the year to measure CH<sub>4</sub> concentrations and flux variations over the year. Furthermore, a longer flight campaign is planned in collaboration with ecosystem researchers at the Swedish University of Agricultural Science (SLU) Umeå and invers modellers from Lund University. Aim of the campaign is to deliver both concentration and flux of CH<sub>4</sub> and CO<sub>2</sub> over at a regional scale with the target area being the county of Jämtland / Sweden.

#### Task 1.3 Development of lightweight sensors and AirCore (Lead: Empa, Lukas Emmenegger)

Partner Empa will develop a lightweight high-precision CH<sub>4</sub> sensor-based quantum cascade lasers (QCL) and interband cascade lasers (ICL) for UAV application. High-precision 3-D measurements of CH<sub>4</sub> will be developed at RUG using a lightweight active AirCore aboard a UAV. The lightweight spectrometer and the active AirCore will be simultaneously deployed on UAVs and compared during a joint measurement campaign.

Task 1.3 is focused on the development of a lightweight  $CH_4$  sensor (EMPA, ESR6) and an active AirCore system (RUG, ESR2). Both activities are strongly related to D1.1 (due month 24), i.e. the development of a lightweight  $CH_4$  sensor and AirCore and their deployment on a UAV. In the following the progress is discussed individually for the two topics.



#### Lightweight CH<sub>4</sub> sensor

The lightweight CH<sub>4</sub> sensor is based on open-path direct absorption spectrometry, using a single-mode quantum cascade laser (DFB-QCL) emitting around 7.83  $\mu$ m. The laser is encapsulated in a TO-3 package including a Peltier element and collimation optics. The absorption signal is enhanced by a circular, segmented multi-pass cell with an optical path length of 10 m (Fig. 2.3a).

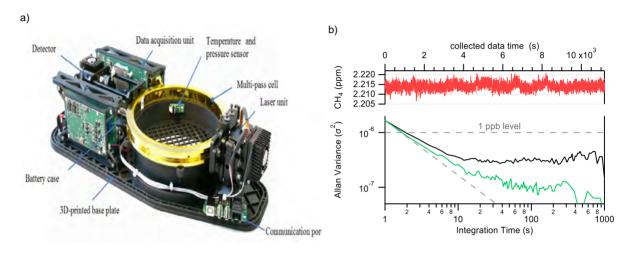


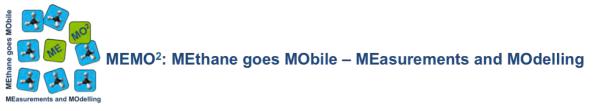
Fig. 2.3: a) photography of the methane sensor; b) typical (black) and best (green) Allan-Werle variance measurements.

This novel cell design yields low optical noise, increased stability against mechanical distortion and a compact footprint. The overall instrument weights 1.6 kg (excluding battery) and has an average power consumption of around 15 W. The low heat dissipation is achieved by intermittent continuous wave laser driving and a system-on-chip FPGA data acquisition module. The spectrometer is equipped with additional sensors for pressure, temperature, and relative humidity, as well as a GPS receiver and a dedicated data transceiver. Therefore, it is possible to use the sensor aboard any drone regardless of the available GPS sensor or specific communication protocol.

Laboratory validation measurements indicate excellent performance with a measurement precision at the low parts-per-billion (ppb) level (Fig. 2.3b). First test flights have been performed using a commercial drone (DJI Matrice 600). During those flights, no impact on mechanical stress was visible. However, we observed some limitations in thermal control due to insufficient aeration of the cover and the available voltage of the power supply for the Peltier coolers. These aspects are currently being solved before proceeding with the final field characterization. Overall, the task is well on track and we anticipate that deliverable D1.1. will be reached well within schedule.

#### UAV active AirCore

The active AirCore system was developed based on the concept of balloon AirCore. As opposed to the conventional concept of passively sampling air using the atmospheric pressure gradient during descent, the active AirCore collects atmospheric air samples using a pump to pull air through the tube during a flight. The active AirCore system consists of a ~ 50 m long stainless-steel tube, a dryer (small stainless-steel tube filled with magnesium perchlorate), a datalogger, a KNF pump, and a 45  $\mu$ m orifice working together to form a critical flow of dried atmospheric air through the active AirCore (Fig. 2.4a). It is placed in a carbon fibre box, and the total weight of the active AirCore system, including the AirCore box, is ~ 1.1 kg (Fig. 2.4b).



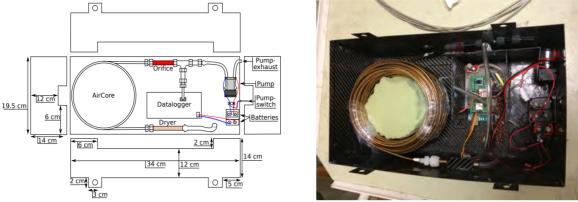


Fig. 2.4: a) a schematic design of the UAV AirCore system, b) an image of the UAV AirCore system

The UAV AirCore measurements have been validated against CH<sub>4</sub>, CO<sub>2</sub>, and CO measurements at a 60-m tall tower in the Netherlands. Due to the smearing of air samples in the cavity of the trace gas analyser and the air diffusion in the AirCore tubing, the spatial resolution of CH<sub>4</sub> measurements is 26 - 28 m at a typical flight speed of 1.5 m/s. To improve the spatial resolution, we plan to improve the response time of the trace gas analyser by reducing the cavity pressure from 140 Torr to 80 Torr. Furthermore, a direct comparison of the UAV AirCore CH<sub>4</sub> measurements with the in-situ spectrometer results of EMPA will help improve the accuracy of the profile retrieval.

#### The major challenges / problems are

1) the UAV-based measurements can only be made under fair weather conditions, and with weak and moderate wind; however, the mass balance approach requires sufficiently stable and moderate wind during the measurements. Therefore, the mass balance approach cannot be applied to those flights with very low wind speed or fast-varying wind directions.

2) the altitude measurements from the UAV contain large uncertainties, which affect the estimate emission rates when the mass balance approach is used. We are installing a new altitude meter to solve this problem.

3) the atmospheric conditions are turbulent, and the UAV flight only captures a snapshot of the downwind of the emission sources. It is therefore difficult to accurately estimate the strength of the emission sources. To overcome this, we will make observations with multiple UAVs to acquire sufficient data for statistical analysis and combine with model simulations to drive a robust estimate of the emissions.

#### Task 1.4 Intensive campaigns (Lead: RUG, Huilin Chen)

Two intensive campaigns involving all ESRs and numerous instruments will be carried out to train the ESRs and improve methodologies to quantify  $CH_4$  emissions. The first campaign (month 12) will be performed at an agricultural farm in Dronten, The Netherlands, with very simple topography and easy logistics. The second campaign (month 30) will be carried out at the European  $CH_4$  emission hotspot upper Silesia (Poland), an industrialized region with a dense network of coalmines.

Within MEMO<sup>2</sup>, two intensive campaigns to jointly obtain measurement data have been planned. The first intensive campaign (reported as milestone MS2) was associated with the 1<sup>st</sup> MEMO<sup>2</sup> school, and took place in Schoorl, the Netherlands from 5 to 16 February 2018. The camping was strongly supported by our local organizer ECN.

In total 8 teams from ECN, UU, AGH, UVSQ, LU, RHUL, UHEI, and RUG brought their own measurement instruments and gathered data during the joint fieldwork (see Table 2.2 for an overview of the instrumentation and measured species). During the first week the ESRs followed theoretical lectures meanwhile the instruments run simultaneously to obtain data for an intercomparison of instruments. After the theoretical part, the ESRs were introduced to the instruments and started first joint sampling along a dedicated transect. Also, a joint tracer release experiment (CH<sub>4</sub>, N<sub>2</sub>O, C<sub>2</sub>H<sub>2</sub>) and



a drone flight with AirCore sampling were conducted. All teams sampled together along several distinct routes with different sources, as farms, biogas plants, peak gas installation, or landfills. Beside the joint activities, the ESRs got the opportunity to choose transects which were interesting for their individual projects.

Partner	Mobile platform	Instrumentation	Measurement species
RUG	Drone DJI Inspire I	UAV AirCore & Picarro	CH4, CO2, CO, H2O
UHEI	Van VW	Picarro G2201i	CH <sub>4</sub> , CO <sub>2</sub> , <sup>13</sup> CH <sub>4</sub>
LSCE	Van	Picarro G2201i and G2203	CH <sub>4</sub> , CO <sub>2</sub> , <sup>13</sup> CH <sub>4</sub> , <sup>13</sup> CO <sub>2</sub> , C <sub>2</sub> H <sub>2</sub>
RHUL	Car 4WD	Picarro G2301, LGR UMEA and bag sampling	CH <sub>4</sub> , CO <sub>2</sub> , C <sub>2</sub> H <sub>6</sub> /CH <sub>4</sub> ratios + $^{13}$ CH <sub>4</sub> in lab
AGH	Car 4WD	Picarro G2201i, 2D wind, T,P,H, PM10, PM2.5	CH <sub>4</sub> , CO <sub>2</sub> , <sup>13</sup> CH <sub>4</sub>
ECN	Van	Aerodyne QCL & Ecophysics NOx & Lasx PM	CH <sub>4</sub> , C <sub>2</sub> H <sub>6</sub> , CO <sub>2</sub> , N <sub>2</sub> O, CO, (NH <sub>3</sub> ), NO, NO <sub>2</sub> , PM1-10
EMPA	Drone, not yet available	Homebuilt CH <sub>4</sub> QCLAS	CH <sub>4</sub> , H <sub>2</sub> O
LU	aircraft not available	Picarro <sup>13</sup> CH <sub>4</sub> available	CH4, <sup>13</sup> CH4
UU	Van	Picarro CO <sub>2</sub> & CH <sub>4</sub> and Picarro backpack. LGR	CH <sub>4</sub> , CO <sub>2</sub>
Other fiel	d equipment		
ECN	Meteo 1	wind profile 5 heights Gill 2 D, Vaiasla all weather station	ws, wd, rh, t, p, rain, H, u*
ECN	Meteo 2	sonic Gill - WMPro	ws, wd, H, u*
ECN	Trailer (mobile lab)	Vaisala all weather station and space for instruments	ws, wd, rh, t, p, rain
UU	Meteo		

Table 2.2: overview of used instrumentation and measured species during the campaign

Task 1.5 Emission factor estimates with dispersion models (together with WP3) (Lead: UVSQ, Camille Yver-Kwok)

Dispersion models will be evaluated and applied to derive emission rates from mobile CH<sub>4</sub> measurements performed in Task 1.1, 1.2 and 1.4 (cooperation with WP3). This top-down approach will used to improve national Emission factors used for UNFCC reporting.

Dispersion and Gaussian plume models are useful tools to derive emissions rates from mobile CH<sub>4</sub> measurements as performed within MEMO<sup>2</sup>. To introduce all ESRs to this topic, UHEI organised a workshop on "Gaussian plume and dispersion models", held 9-10 October 2018 in Heidelberg, Germany (reported as milestone MS3). The aim of the workshop was to teach theoretical basics on plume dispersion, practice with two exercises and to discuss future measurement strategies to ensure best model use.

ESR5 has been working on using a simple Gaussian model to estimate CH<sub>4</sub> emissions from different sites. The model has already been applied to a landfill in France and a gas compressor in the Netherlands. More measurements are needed to give a proper estimate. ESR5 has been working with ESR12 on the GRAM model to see if it was relatively applicable for our cases. It appeared that it needs information from a 3D sonic anemometer. We have acquired a 3D sonic anemometer and the model will be applied on the next campaign results.



#### 2.2.2 WP2 - Source identification by isotopic characterization (Lead: Dave Lowry, RHUL)

#### 2.2.1.1 General WP overview and contribution of involved beneficiaries

Within WP2 - Source identification by isotopic characterization (including primarily the ESRs 8 and 9 from UU and RHUL) - we aim on CH<sub>4</sub> source identification by isotope measurements and improved on an understanding of the temporal and spatial variability of isotopic signatures of CH4 emissions. In close collaboration with WP1 we investigate the local and regional sources. Together with WP3 we appoint sources by modelling activities and verify emission inventories. UK mobile campaigns that will feed data to WP2 have been organized by RHUL, ongoing since the start of the MEMO<sup>2</sup> project.

In the first year, a new LGR UMEA instrument, measuring  $CH_4$  and  $C_2H_6$  (ethane), has been tested in the vehicle. By this we can distinguish thermogenic gas from



**Fig. 2.5:** Zoom of mobile survey in North Yorkshire indicating the isotopic signatures calculated for identified plumes.

other CH<sub>4</sub> sources. Data comparison between this ultraportable instrument and the high-precision Picarro instrument, sampling from co-located air inlets, has been made. In the Netherlands several locations were sampled by UU.

Significant progress has been made in measuring the isotopic signatures of methane and reporting them to a common scale. Inter-comparison between RHUL and UU showed that the two IRMS groups are reporting to the same carbon isotopic scale (submitted as deliverable D2.1) and this is now being extended to the CRDS isotope groups. This inter-comparison is being pushed further by joint measurement and sampling campaigns by multiple partners (see Task 2.2 results), and by comparison of the results from co-located sampling and multi-laboratory measurement (milestone MS7). RHUL has successfully completed Task 2.6 / milestone MS8 (by month 20), which was the organisation of the 3-day methane isotope workshop. The workshop (reported as milestone MS8) from 17-19 Sept 2018 saw RHUL and UU speakers, plus invited guests discuss sampling, measurement, data correction and modelling, and gave the opportunity for ESRs working on isotopes to present some of their early results.

RHUL organised and prepared inter-comparison cylinders for exchange between RHUL and UU, resulting in an accepted C-isotope scale for methane in the range -61 to -38 ‰ (see deliverable D2.1). Progress has continued on WP2 tasks, particularly tasks 2.1, 2.2 and 2.3.

ESR7 and ESR9 have made measurements at waste, fossil fuel and biogenic sites across SE England. In particular the rapidly expanding biogas sector needs to be isotopically characterised and CH<sub>4</sub> plumes from some of these have been measured (Table 2.3).

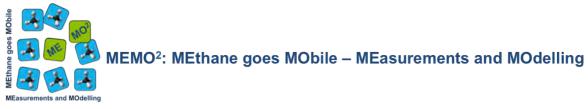
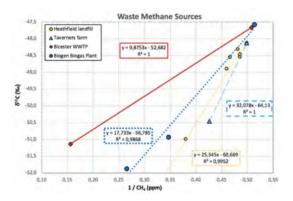


Table 2.3: Summary of Summer Campaigns 2018.

Sources	δ <sup>13</sup> C (‰)±
Cambridge Cow Barn	-58.0±1.1
Agriculture (Manure)	-56.9±1.1
Taverners Farm	-64.1±1.2
Heathfield Landfill	-60.7±1.3
Greatness Landfill	-55.6±0.9
Milton Landfill	-58.2±0.6
Brighton Viridor Landfill	-57.9±0.5
Horsham Landfill	-56.9±1.9
Bicester WWTP	-52.7±1.7
Chertsey WWTP	-50.7±0.3
Modgen WWTP	-50.7±0.5
Horsham WWTP	-53.9±0.1
Bygrave Biogas Plant (Food Waste)	-56.8±1.3
Milton Keynes Biogas Plant (Municipal Solid Wastes)	-61.6±0.6
Snodland Biogas Plant (Papermill Waste)	-50.8±0.2



**Table 2.4:** Comparison of previous studies and recent studies.

UK CH₄ Sources	Previous Studies* δ <sup>13</sup> C(‰)	Recent Survey Results δ <sup>13</sup> C(‰)
Agriculture (cows)	-66	-61±2
Agriculture (manure)	-58	-57±1
Landfills	-58	-57.8±0.8
Waste Water Treatment	-53	-52±0.8
Biogas Plant		-56±3

\* Fisher, R.E., 2000; Lowry, D., et al. 2001; Zazzeri, G., et al. 2017



**Fig. 2.7:** Isotopic signatures of sources sampled during surveys in SE England. The red lines are the high-pressure gas pipes along which are located major above ground infrastructure, such as compressor stations and offtake stations. The white circles indicating isotopic enrichment identify some of these stations which have significant plumes of  $CH_4$  emission.

Fig. 2.6: Isotopic signature of waste CH<sub>4</sub> sources in the UK.

Much of Task 2.3 work has focussed so far on London, UK, with a survey of the coastal city of Brighton, and on Alkmaar, the Netherlands, and surroundings during the 1<sup>st</sup> MEMO<sup>2</sup> school. ESR7 and ESR9 have collected urban samples for isotopic analysis during their secondment periods, in Groningen and Hamburg respectively, and the analyses of these samples is ongoing. The major sources identified are leaks in the gas distribution network, showing a very distinctive isotopic signature that is enriched in <sup>13</sup>C relative to atmospheric background.

Work is continuing to visit every 1km grid square in London with the mobile measurement and sampling system during the course of MEMO<sup>2</sup> (Fig. 2.8).

UHEI, AGH and UVSQ, although primarily belonging to WP1, organized also first campaigns (at e.g. a gas compressor station, dairy farm and a biogas plant, and at ventilation shafts located within the Southern Silesian Coal Basin) and investigated continuous measurement precision, shortterm and long-term repeatability, and temperature and atmospheric pressure influence on two isotopic Picarro instruments.

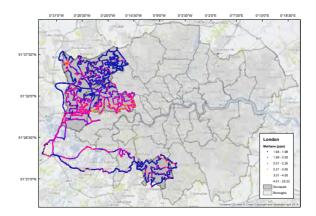


Fig. 2.8: London boroughs surveyed to date highlighting the  $\mathsf{CH}_4$  concentrations measured.



#### 2.2.2.2 Progress of WP tasks

#### Task 2.1: Isotope inter-calibration and calibration service (LEAD: RHUL, Dave Lowry)

Exchange of standards between isotope ratio mass spectrometry (IRMS) groups (UU, RHUL). UU and RHUL will provide calibration services to those ESRs that operate mobile CRDS isotopic analysers (UVSQ. UHEI, AGH-UST). This task will be undertaken by WP2-focussed students and lead to deliverable D2.1.

The task is ongoing. The associated deliverable D2.1 has been submitted, and milestone MS7 has been achieved.

Five tanks were prepared at RHUL and shipped to UU at the beginning of October, containing ambient air (1.96 ppm), natural gas diluted to 10 and 2 ppm with zero air, and landfill gas diluted to 10 and 2 ppm with zero air. These have allowed inter-comparison between RHUL and UU in the -60 to -40 ‰ range of  $\delta^{13}$ C, and a common scale for the MEMO<sup>2</sup> project agreed. There was excellent agreement between both laboratories and the existing laboratory corrections can continue to be used. This scale will be compared with co-sampled sources measured in both laboratories by ESR8 and ESR9 as part of ongoing milestone MS7 activities. The tanks have been shipped to the first of 3 partners with mobile laser-based isotopic instruments to extend the comparison and bring these instruments onto the common scale, ready for transfer of isotopic data to the MEMO<sup>2</sup> data repository.

Once the CRDS partners have measured the cylinders we will consider how to circulate a more expansive document to the wider CH<sub>4</sub> isotopic community.

#### Task 2.2: Isotopic characterisation of significant CH<sub>4</sub> sources (LEAD: RHUL, Dave Lowry)

Air samples will be collected and analysed for CH₄ isotopic composition by UU or RHUL. ESRs from WP1 will sample source plumes identified by mobile measurements for isotope analysis. Links to innovation in WP1 (UAV sampling and AirCore sampling) for vertical profiling of isotopic signature through plumes. Strong collaboration with mobile CRDS based isotope measurements in WP1.

This task is ongoing, with significant progress for large regions of the UK (ESR7, ESR9 and other projects) and Netherlands (ESR8 and ESR10), and smaller regions of France (ESR5), Germany (ESR1), Poland (ESR3) and Sweden (ESR4). This process has been aided by multi-partner isotopic campaigns

- **B** Silesia – Mav-June 2018 (AGH, UU, UHEI, UVSQ)
- Southern England – June-July 2018 (RHUL, UU, UVSQ) (Fig.2.5)
- Hamburg Oct 2018 (UU, RHUL) **B**

and ESR secondments: ESR5 and ESR8 to RHUL for 4 weeks (June-July 2018) - 5 days of mobile measurement and sampling: During 4 campaigns in situ isotopic composition was measured by Fig. 2.10: Setup for mobile measurement with monitoring / ESR5 using AirCore. This storage tube allows to obtain better time resolution and accuracy for

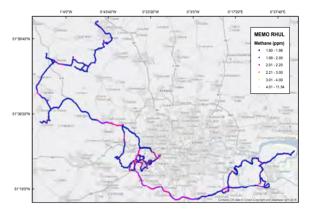
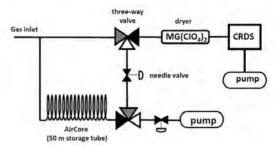
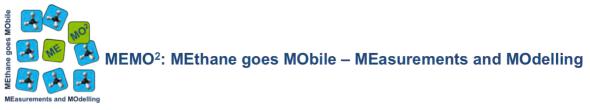


Fig. 2.9: Routes taken during joint SE England methane surveys for the MEMO<sup>2</sup> project.



replay mode

<sup>13</sup>CH<sub>4</sub> and measure in situ isotopic after observation of CH<sub>4</sub> peak (Rella et al., 2015). In Fig. 2.10, a scheme of an AirCore is presented. Fig. 2.11 shows a comparison of measurement in monitoring and replay mode using AirCore, and Fig. 2.12 presents a Keeling plot and Miller-Tans plot which allow to calculated isotopic composition (Pataki et al. 2003).



D5.9 MEMO<sup>2</sup> – Midterm Review Report

Based on the map of infrastructure there exist two probably sources of CH<sub>4</sub> in the measured urban area: gas leaks and wastewater industry. According to Zazerri et al.,2017, the value of the isotopic composition for natural gas distributed in this area is equal  $(-36 \pm 3)$ % and  $(-53 \pm 3)$ % for the wastewater sector. Results from the survey on 27.06.2018 obtained using an AirCore storage tube are presented in Table 2.6. A map of measured concentration during urban area survey is shown in Fig. 2.13.

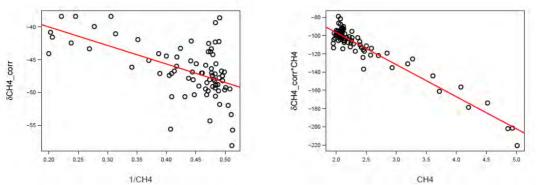


Fig. 2.11: Results obtained in replay mode, AirCore number 2 from 27.06.2018, Ashford Water Treatment Plant left: monitoring mode, right: replay mode

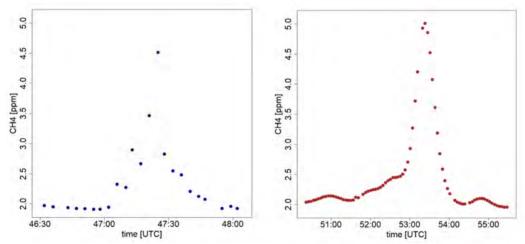


Fig. 2.12: Replay mode, AirCore number 2 from 27.06.2018 left: Keeling plot, right: Miller-Tans plot



N° Peak	Localization and probably source	CH₄ [ppm]	δ¹³CH₄ (Keeling plot)	δ¹³CH₄ (Miller- Transplot)
1.	Laleham, gas leak	2.60	-37.9 ± 3.8	-37.8 ± 3.7
2.	Ashford, Water Treatment Plant	5.01	-33.9± 1.9	-34.6 ± 1.1
3.	Feltham	6.71	-33.9 ± 1.0	-34.2 ± 0.6
4.	Stanwell	3.71	-42.1 ± 6.4	-42.9± 4.3
5.	Stanwell	3.84	-32.9± 2.1	-32.1 ± 1.5
6.	Egham	4.98	-33.0 ± 2.9	-35.4 ± 1.9

Fig. 2.13: Map of CH<sub>4</sub> concentration measured during urban area survey, 27.06.2018, white points and number indicate stops for changing to replay mode



The calculated iso-

## MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

D5.9 MEMO<sup>2</sup> – Midterm Review Report

topic composition determined that sources of measured peaks are connected with natural gas leaks Even in case of the observed peak close to the water treatment plant, the calculated matched to the isotopic composition of natural gas. In the case of peak nr. 4, the obtained value of  $\delta^{13}CH_4$ 

probably

Table 2.7: Isotopic composition of methane source, 26.06.2018

Air Core	Latitude	Longitude	CH₄ [ppm]	δ <sup>13</sup> CH₄ (Keeling plot)	δ <sup>13</sup> CH₄ (Miller-Tans plot)
1.	51°37'27.3381" N	1°13'22.2688" W	3.91	-34.8 ± 2.2	-35.1 ± 1.7
2.	51°52'45.9314" N	1°10'11.8802" W	4.58	-56.8 ± 1.9	-57.4 ± 1.1
3.	51°53'56.8505" N	1°0'23.5382" W	3.16	-54.1 ± 3.4	-54.3 ± 2.9

from the pipeline. Table2.8: Isotopic composition of methane source, 28.06.2018

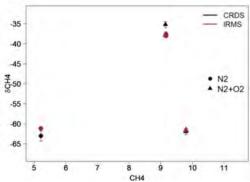
Air Core	Latitude	Longitude	CH4 [ppm]	δ <sup>13</sup> CH₄ (Keeling plot)	δ <sup>13</sup> CH₄ (Miller-Tans plot)
1.	51°27'37.118" N	0°43'1.996" W	2.70	-33.6 ± 3.6	-33.9 ± 3.5
2.	51°17'44.6234" N	0°11'40.8069" W	3.11	-56.3 ± 2.1	-56.3 ± 2.1

value of  $\delta^{13}$ CH<sub>4</sub> Table2.9: Isotopic composition of methane source, 05.07.2018

Air Core	Latitude	Longitude	CH₄ [ppm]	δ <sup>13</sup> CH₄ (Keeling plot)	δ <sup>13</sup> CH₄ (Miller-Tans plot)
1.	50°34'30.2693" N	3°36'38.9308" W	2.60	-57.0 ± 5.6	-57.5 ± 5.0
2.	50°34'22.9468" N	3°36'45.5954" W	2.68	-58.8 ± 2.8	-59.3 ± 2.5
3.	50°34'22.5197" N	3°36'36.7858" W	3.15	-59.7 ± 2.1	-60.0 ± 1.6
4.	50°40'13.0915" N	3°31'43.4637" W	3.24	-40.6 ± 2.0	-40.7 ± 1.6

correlated with the mixed isotopic signature from two peaks occurring close to each other. In Tables 2.7, 2.8 and 2.9 results obtained by ESR5 during surveys in Southern England are presented.

Secondments in RHUL were also the opportunity to compare results from the stationary measurement made by CRDS (LSCE, ESR5) and IRMS. One of the activities was measurement of diluted sample from landfill or natural gas. Dilution was made with  $N_2$  or mixed  $N_2$  with  $O_2$ . Obtained results are presented in Fig. 2.14 and Table 2.10 below.



is

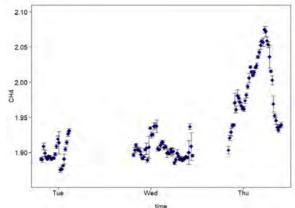
Table 2.10: Comparison of isotopic composition  $^{\delta13}CH_4$  [‰] measured by IRMS and CRDS

source of gas	dilution	IRMS	SD	CRDS	SD
landfill	N <sub>2</sub>	-61.24	0.03	-63.1	1.25
landfill	80%N <sub>2</sub> +20%O <sub>2</sub>	-61.63	0.04	-61.99	0.77
Geochem gas	N <sub>2</sub>	-38.04	0.01	-37.72	0.66
Geochem gas	80%N2+20%O2	-37.83	0.04	-35.21	0.74

Fig. 2.14: Comparison of isotopic composition measured by IRMS and CRDS, x axis CH<sub>4</sub> [ppm], y axis  $\delta^{13}$ CH<sub>4</sub> [‰]

Additionally, 3 nocturnal continuous measurements from the common inlet by CRDS (from LSCE, ESR5) and IRMS were conducted. This activity allowed to compare results from the continuous measurement obtained by two different instruments. CH<sub>4</sub> concentration obtained by CRDS average by 20 minutes and comparison of the calculated value of the isotopic composition is shown below.





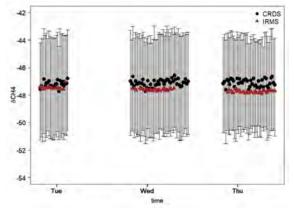


Fig. 2.15: CH<sub>4</sub> concentration measured by CRDS analyser during continuous measurements of ambient air, 20 minutes average

Fig. 2.16:  $\delta^{13}\text{CH}_4$  measured by CRDS and IRMS during continuous measurement of ambient air, for CRDS 20 minutes average

Several samples were collected during the following secondments:

- ESR7 spent 5 weeks at RUG (Sept-Oct 2018) for sampling of sources for isotopic analysis. The samples collected during this secondment are currently being analysed.
- ESR9 went to UU for 4 weeks (Sept-Oct 2018) and also joint a campaign of ESR10 and ESR8 in Hamburg for 4 days. The samples collected during this secondment are currently being analysed.
- ESR4 came to RHUL for 2 weeks (Nov 2018) to analyse Swedish wetland samples for isotopic signature and interpreting data.

#### Task 2.3: Deciphering mixed urban and industrial emissions (LEAD: RHUL, Dave Lowry)

Samples will be collected when CH<sub>4</sub> plumes are identified by grid-pattern mobile measurement surveys in complex areas with small to medium-sized urban / industrial sources. Isotope data will improve understanding of source contributions (D2.3). CRDS groups (WP1) will directly measure the <sup>13</sup>CH<sub>4</sub> during these surveys for direct comparison with IRMS technique.

This task has been significantly aided by the collaboration with EDF to study fossil fuel infrastructure emissions in cities. Combined measurements of CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub> and isotopic proxies can distinguish fossil fuel and combustion sources from biogenic sources during mobile surveys. Utrecht has been fully surveyed (ESR10), Hamburg is ongoing (ESR10, aided by ESR8 and ESR9), London is ongoing (ESR9 aided by ESR7), with continuing studies in Paris (ESR5), Heidelberg (ESR1) and Katowice (ESR3). Data currently are under evaluation. More details are given in the respective ESR reports.

#### Task 2.4: Isotope monitoring at fixed sites (LEAD: UU, Thomas Röckmann)

The IRMS and isotopic CRDS instruments from WP1 and WP2 will make co-located continuous isotope measurements at fixed sites. This will provide important information on temporal and meteorological variations in isotope source signatures that will be interpreted with regional models from WP3. This task will lead to deliverable D2.4.

In May 2018, a continuous measurement system for  $\delta D$  and  $\delta^{13}C$ -CH<sub>4</sub> in ambient air was set-up at AGH by ESR8. From the 16<sup>th</sup> to the 30<sup>th</sup> of May 2018, ESR8 stayed in Krakow to install the instrument for continuous CH<sub>4</sub> mole fractions,  $\delta D$  and  $\delta^{13}C$ -CH<sub>4</sub> measurements in ambient air. The system was prepared at UU and shipped to AGH. Due to a broken compressor the measurements could start only in mid-September 2018.

Fig. 2.17 shows the data that has been collected so far. Analyses and evaluation will start soon.



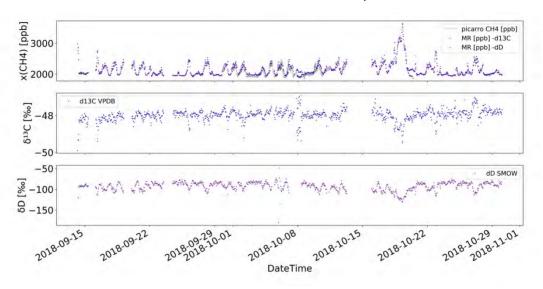


Fig. 2.17: Overview of the data being currently collected in Krakow

For ESR9 (RHUL) some diurnal monitoring at the RHUL site near London with more and longer measurement periods is planned for the next reporting period.

#### Task 2.5: Validation of inventories at grid scale (LEAD: UU, Thomas Röckmann)

The isotopic signature of each major source will be constrained at regional to EU-wide scale (D2.2) for use in models (WP3). ESRs from WP2 and WP3 will use data from Tasks 2.2-2.4 to produce maps of isotopic source signatures contributing to the validation of inventories (D2.5, due month 42).

This task has been commenced but need detailed rural and urban street-by-street surveys to be evaluated before this task can gather momentum. So far, the surveys that can be linked to emission inventories at 1 x 1 km resolution are those in the cities described under Task 2.3 above. UK surveys suggest that distribution of landfill and agricultural emissions by 5 x 5 km grids in the inventory could be better resolved as measured emissions are dominated by smaller source footprints such as cow barns or active landfill cells / gas engines. Distribution of gas leaks by population density does not accurately reflect the distribution of gas leaks, which tend to be located along a small number of pipelines or above-ground infrastructure. While isotopes can identify sources that are not seen during surveys they are most likely to be of use for assessing source proportions at a larger area scale.

#### Task 2.6: Isotope workshop at RHUL (LEAD: RHUL, Dave Lowry)

Sessions aimed at helping WP1 students interpret the isotopic measurements from their mobile campaigns, and in discussing with WP3 students the suitability of data to incorporate into their models.

This task was milestone MS8 and has been successfully completed. A report on the workshop was submitted to the Project Officer.



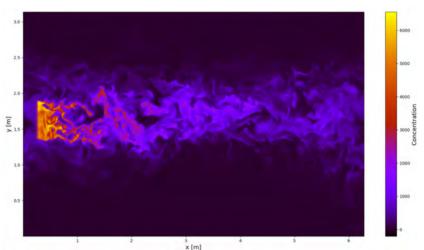
2.2.3 WP3 – Modelling: A multi-scale interpretation framework for CH<sub>4</sub> observations (Lead: Maarten Krol, WU)

#### 2.2.3.1 General WP overview and contribution of involved beneficiaries

In WP3 – Modelling: A multi-scale interpretation framework for CH<sub>4</sub> observations (including primarily the ESRs 10 - 13 from the beneficiaries UU, WU, EMPA, and UVSQ) – we develop and use a hierarchy of innovative modelling tools to quantify emissions based on the measurements of CH<sub>4</sub> concentrations and its isotopic composition obtained in WP1 and WP2. Good progress has been made, despite the fact that a large fraction of the time of the ESRs was dedicated to training and getting acquainted with their working environment. The division of tasks works well.

Tools are being developed at WU to optimally infer point / line source strengths from measurements made by MEMO<sup>2</sup> ESRs with drones (ESR2) or made available by non-academic partners (ECN, Afvalzorg Nederland) and other collaborators (Aerodyne). These tools form the basis of campaign planning, and the set-up of simplified tools (Gaussian Plume, RANS) for the evaluation of the measurements.

On long-term different modelling techniques will be used to estimate



**Fig. 2.18:** Simulation of a line source in MricoHH. Axis and scales arbitrary. One of the remaining problems that now has been solved is the inflow (left) due to periodic boundary conditions that are used for the flow simulation.

emission fluxes from different kinds of sources (point, line, diffuse) in order to link measured concentrations of pollutants with emission fluxes. These efforts go beyond the classical interpretation using well-known Gaussian plume models. In the special workshop on Gaussian Plume modelling the other ESRs were made aware of these developments. Also, the secondment of ERS12 helped to coordinate research efforts.

At the smallest geographical scale, ESR11 is implementing CH<sub>4</sub> point and line sources in the MicroHH model, a fluid dynamics simulation model. Good progress has been made in setting up the MicroHH model for the interpretation of atmospheric observations. Measurement data are available from a campaign in the Gulf of Mexico (Aerodyne, Scott Herndon, personal communication) and through ESR2 (University of Groningen) from drone flights close to a cow shed. In a next step, ship and drone-type model-sampling will be implement. Using these sampled data, it will be assessed how well a source can be calculated from sparse measurements in a turbulent flow field. Fig. 2.18 show an example of a simulated plume downwind of a cow-shed.

A first milestone was reached by the provision of a Large Eddy Simulation tool for the first campaign and the workshop (MS10, due month 12). A deliverable about new tools to estimate CH<sub>4</sub> source strengths from point sources including mobile measurements is due month 24 (D3.1).

The **UU**, responsible for the integration of mobile measurement data in the monitoring, reporting, and verification of key  $CH_4$  sources in the greenhouse gas emission reporting in Europe, focussed on gathering data to establish a solid data basis. ESR10 started the interpretation of recent measurements and will concentrate on the integration of data within the next reporting period.

On the somewhat larger scale, **Empa** (ESR12) is working on the implementation of a Lagrangian dispersion tools at the scale of individual point sources and, in a second step, at the scale of Europe. This step is "intermediate" between the work of ERS11 (WU) and ESR13 (UVSQ). ESR12 analyses a



well-documented tracer release experiment in Australia and applies tools such as the GRAL model to the first test flight of the CH<sub>4</sub>-UAV developed by ESR6. ESR12 made important steps to more efficiently use the GRAL model. For instance, GRAL has been adapted, such that it can now simulate dynamic situations with rapidly changing wind conditions. GRAL is currently being applied to additional tracer release experiments in situations of varying complexity in order to better understand the potential and limitations of the model and to optimize the setup. During the first secondment of ESR12, the model was applied to sources in France in collaboration with ESR5 (UVSQ).

On the European scale, **UVSQ** (ESR13) is modelling the European CH<sub>4</sub> distribution using the CHIMERE chemistry transport model with emission inventories from EDGAR 4.3.2 and TNO-MACC\_III. Multi-year simulations have been carried out (2011 to 2015), with focusing on estimating the so-called representation error: how well can a relatively coarse-grid model represent the local situation at the measurement site? Sensitivities for the boundary conditions, model grid resolution, and emission inventory (e.g. including natural wetlands) have been calculated, model output has been compared to station observations throughout Europe, and the effect of different emission inventories has been investigated. Error characterization is important, because at a later stage realistic "mismatch" errors are needed in atmospheric inversions.

Fig. 2.19 shows statistics of the comparison. Biases are stronger in winter, and larger for the TNO-MACC-III inventory. Adding a description of wetland emissions resolves a small part of the bias.

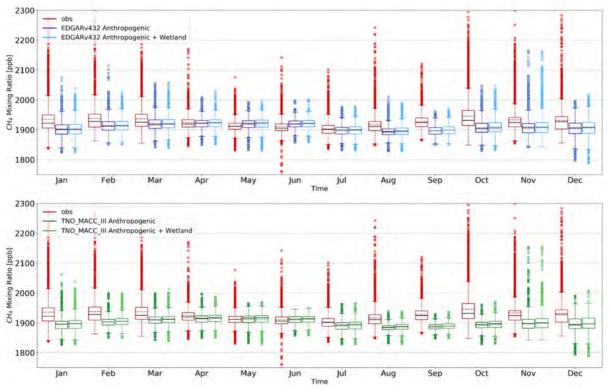


Fig. 2.19: Differences between concentrations using the two inventories with and without adding wetland emissions over Europe; top: EDGAR v4.3.2, bottom: TNO-MACC\_III.

During a secondment at WU, ESR13 learned from ESR11 how to use the high-resolution model MicroHH to assess representation errors.

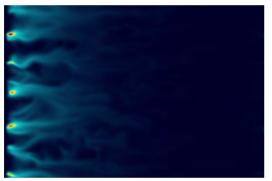


#### 2.2.3.2 Progress of WP tasks

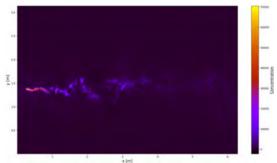
#### Task 3.1: Interpretation mobile observations (LEAD: WU, Maarten Krol)

Dispersion of emissions will be modelled using the newest Large Eddy Simulation tools, in which detailed land-use maps, roughness elements, and surface heterogeneity can be taken into account. This approach will be compared to the dispersion model approach (WP1). Modelling will also assist in planning of the mobile measurements and intensive campaigns.

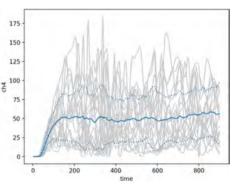
Fig. 2.20 and 2.21 show the results of MicroHH simulations of a line source. These simulations are more advanced than simple Gaussian plume models. By averaging (Fig. 2.21) you obtain a stationary state and statistical information about variability. MicroHH was only able to simulate surface sources. To better interpret measurements that are being conducted within MEMO<sup>2</sup>, it is important to be more flexible. Point and line sources are now added 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. 2.22, 2.23).



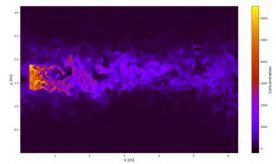
**Fig. 2.20:** DNS simulation of a plume from a line source in stationary homogeneous turbulence.



**Fig. 2.22:** MicroHH simulated dispersion from a point source (arbitrary scales).



**Fig. 2.21:** Ensemble average of CH<sub>4</sub> mixing ratios at one point.



**Fig. 2.23:** MicroHH simulated dispersion from a line source (arbitrary scales).

One remaining problem that has now also been solved is the circular boundary conditions. This is desirable for the flow field, but not for CH<sub>4</sub>. In Fig. 2.22 and 2.23 you clearly can spot concentrations that flow in from the left into the domain.

The model is now ready to interpret observations. There are already drone measurements from ESR2. Model output will be sampled according to drone sampling downwind of a cow barn. Thus, both the space and time in the model will change. Given stationary turbulence and dispersion, a large number of realisations will be made from the simulations, which allows to get statistics. An open question is how well we can determine the source strength from a particular drone flight path. In the model, fortunately, this relation can be made. Currently, we are exploring this sampling issue for a first publication.



**Task 3.2: Construction of new bottom-up emission maps, including isotopes (LEAD: TNO, Hugo Denier v. Goon)** The mobile measurements that become available in MEMO<sup>2</sup> will be used to construct updated high-resolution emission maps for the EU domain. For modelling purposes, the isotopic signature of the CH<sub>4</sub> sources will be included in the inventories.

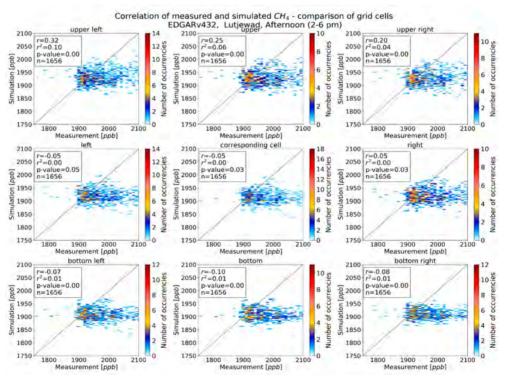
During the 1<sup>st</sup> reporting periods the focus was laid on measurement campaigns to ensure a solid data base. So, there are no particular outputs related to the inventories. The task of constructing high-resolution emission maps and include the isotopic signatures will be implemented in the upcoming reporting period.

#### Task 3.3: Forward simulations of CH<sub>4</sub> and CH<sub>4</sub> isotopes (LEAD: UVSQ, Philippe Bousquet)

Two meso-scale transport models with up to km-scale resolution will be used to simulate CH<sub>4</sub> and its isotopic composition over Europe with the aim to predict how different sources with different isotopic signatures (e.g. from an urban environment) blend into larger-scale concentration fields. Comparison to new and existing "background" observations will be performed.

New simulations of CH<sub>4</sub> mixing ratios have been performed at the European scale 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 have been carried out from 2011 to 2015 with a horizontal resolution of  $0.5^{\circ}x0.5^{\circ}$  (~50x50 km). Also, a large number of sensitivity experiments were performed. The comparison and the sensitivity tests aim a better understanding of the difference between modelled and measured CH<sub>4</sub>, and thus help reveal which part can be attributed to errors in inventories and serve the goal of estimating top-down CH<sub>4</sub> emissions on the European scale.

We checked the correlation between measurements and simulated values of the grid cell corresponding to the station location and its eight neighbouring cells. An example for the site Lutjewad in the Netherlands can be found in Fig. 2.24. On the basis of this analysis, we decided to use the values of model grid cells with the highest correlation coefficients for the comparison against the measurements.



**Fig. 2.24:** 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.



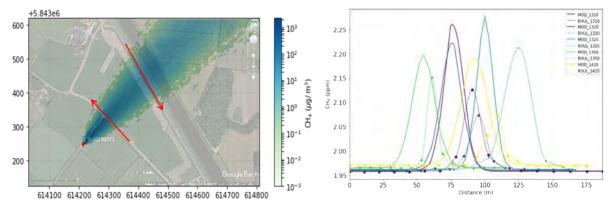
One of the sensitivity tests consisted of running the model with boundary conditions obtained from the CAMS-MACC reanalysis product (Inness et al. 2013) which is available for every three hours per day from July 2013 on. In contrast to this, the pre-optimized boundary conditions derived from the LMDz model are only available up until 2010 and so they follow the same pattern every year in 2011-2015, resulting in a uniform seasonal behaviour. The differences in the latitudinal gradients of the two products come from different assumptions used in the models, such as a higher latitudinal gradient in the MACC product. Simulations using the MACC boundary conditions compare better to the measurements. We decided to use the MACC boundary conditions for every other model run hereafter, including the other sensitivity runs.

To investigate the impact of the use of natural CH<sub>4</sub> emissions in addition to the anthropogenic emissions, we carried out a sensitivity run for which emissions from wetlands were included following Poulter et al. (2017). 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.

#### Task 3.4: Top-down estimates of EU emissions (LEAD: EMPA, Dominik Brunner)

Two meso-scale transport models with up to km-scale resolution will be used to infer EU-scale CH<sub>4</sub> emissions, and these estimates will be compared to products of the Copernicus services. The updated emission maps of task 3.2 will form the starting point of inverse modelling. EMPA will employ Lagrangian dispersion models to analyse the small-scale dispersion.

This task employs Lagrangian dispersion tools at the scale of individual point sources and, in a second step, at the scale of Europe. Since ESR12 working on this task was hired in December 2017, the current reporting period (Mar 2018 - Feb 2019) roughly corresponds to the first year of the PhD project. During this period, ESR12 implemented the GRAL (Graz Lagrangian Model) dispersion model to simulate the dispersion of CH<sub>4</sub> emitted from individual sources. An example simulation applied to a tracer release experiment conducted during the first MEMO<sup>2</sup> winter school in February 2018 is presented in Fig. 2.25.



**Fig. 2.25:** Left: GRAL simulated CH<sub>4</sub> 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<sub>4</sub> mole fractions along different transects sampled by the car of RHUL. Matching the areas below the curves allows estimating the strength of the source.



ESR12 modified and extended the GRAL model in several important ways in order to make it more efficient and easily applicable to any given situation. This included (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, (iii) development of a python package for post-processing and visualization of the output. Furthermore, ESR12 implemented a simple Gaussian plume model to compare the results obtained with GRAL, GRAL is currently being applied to additional tracer release experiments in situations of varying

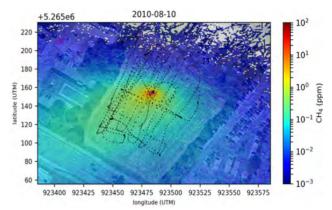


Fig. 2.26: Simulated CH<sub>4</sub> mixing ratio (at 5 m above surface) for a CH<sub>4</sub> release test at EMPA to analyse the performance of the new CH<sub>4</sub>-UAV. Black dots represent the UAV's path during the release test.

complexity in order to better understand the potential and limitations of the model and to optimize the setup. This includes a well-documented tracer release experiment in Australia and the first test flight of the CH<sub>4</sub>-UAV developed by ESR6 (Fig. 2.26). During the first secondment of ESR12, the model was applied to sources in France in collaboration with ESR5.

#### Task 3.5: Workshop on top-down emission estimates (LEAD: WU, Maarten Krol) This task is not active yet. A workshop will be organised in project month 30.

#### 2.2.4 WP4 – Training (Lead: Philippe Bousquet, UVSQ)

#### 2.2.4.1 General WP overview and contribution of involved beneficiaries

This WP involves all beneficiaries and non-academic partners of MEMO<sup>2</sup>. All activities scheduled in WP4 have started, i.e. all ESRs wrote their individual Career Development Plan (CDP, D4.1 and D4.2) and the consortium organized the first network training events (theoretical and practical lessons, co-supervisions, the 1<sup>st</sup> and 2<sup>nd</sup> MEMO<sup>2</sup> school and two workshops, joint field campaigns, networking, and international training as e.g. first secondments and participation in conferences). The main highlights of the periods were the organization of the 1<sup>st</sup> and 2<sup>nd</sup> MEMO<sup>2</sup> school, as described in the report on MS2 and (upcoming) D4.4, and the two workshops at RHUL (MS8) and UHEI (MS3).

#### 2.2.4.2 Progress of WP tasks

#### Task 4.1: Monitoring and quality assurance of training goals (LEAD: UVSQ, Philippe Bousquet)

Training of ESRs will be organized on an individual and a network level as described in detail in chapter 1.2 of the GA. The individual training will be specified in the CDPs for each ESR, which are updated on an annual basis. The progress of the ESRs will be monitored by the supervisors / co–supervisors. A more detailed description of the quality assurance procedure is given in chapter 3.2.4 of the GA.

The monitoring of training goals is performed both at local and project scales. At local scale, the supervisors of ESRs take care that their student i) has access to useful training courses in local universities/institutions to support his/her career development plan and ii) attend to the relevant international conferences and workshops. At the project scale, the career development plan (CDP) and the table of secondments are the key elements of the monitoring. The objectives and the detailed template to be filed by the ESRs and their supervisor has been explained during a dedicated session of the 1<sup>st</sup> MEMO<sup>2</sup> school. We particularly insist that filling the CDP is the occasion for the ESR to revisit with her / his supervisors present and future objectives and activities. Then all the filled CDPs have been gathered by the project manager and reviewed to build deliverable D4.1 on time. The update of the CDP, which is a living document adapting to the present and future activities and plans of the ESRs, is planned for the 2<sup>nd</sup> Annual Meeting, as scheduled in the project.



The secondments (Table 2.13) are an important part of the student work with a research (something useful for the research work) and/or a training (something new to learn) dimension. The project management closely monitor the secondments, when and where ESRs do their secondments, and ensures reporting about them. The ESRs may choose if they would like to report by a given template or by writing a blog about their secondments. The blogs are public available on the website. This option is clearly encouraged as ESRs by this option I) not only reflect the work done but also II) enhance their writing skills towards a public audience and III) disseminate the project and its outcomes. By end November 2018, all ESRs will have performed their first secondment and some ESRs have already performed two secondments. Secondment periods may have shifted a bit in time compared to the original plan due to the delay in recruitment, but the activity is launched for all ESRs.

Finally, the (at least) annual co-supervision committees are a way to monitor the general activities of the ESRs, including training ones.

The quality insurance of training goals is achieved by two ways. First in the construction of the training events such as the MEMO<sup>2</sup> school, we have kept a constant attention to follow the objective mentioned in the project about the learning by doing approach. Not only students follow lectures, but they are involved in their learning by doing by themselves activities: presenting their work to other supervisors and ESRs during the school (poster) and during the annual meetings (talk or poster), practising on concrete situations both for modelling (plume modelling during a dedicated workshop, box modelling during the 1<sup>st</sup> school) or terrain (joint field campaign organized with the 1<sup>st</sup> school, practice with instruments, joint data analysis). Several ESRs presented their first results already at scientific conferences such as the EGU2018 or the ICOS Science Conference. Some of the ESRs had also the opportunity to participate in another field campaign in Poland in May / June 2018. Quality insurance has also been estimated by the ESR evaluation of the activities proposed during the 1<sup>st</sup> MEMO<sup>2</sup> school as developed in the report for milestone MS2. The mean ranking of the lectures and lecturers is 3.7 out of 4. The overall evaluation of the 1<sup>st</sup> MEMO<sup>2</sup> School is 3.2 out of 4, the best mark being the interest of the activities and the less good is the length (too long) of some activities.

#### Task 4.2: Organisation of network training events (LEAD: UU, Thomas Röckmann)

The network training will be coordinated by the UU and locally organized by dedicated PI's and their groups. A list of the training events is given in Table 1.2.1c of the GA. The network training events are mandatory for the ESRs. The project meetings are part of the training (see WP5).

All network training events scheduled so far in the project have been organised: 1<sup>st</sup> MEMO2 school (5-16 February 2018 in the Netherlands), 1<sup>st</sup> annual meeting in Switzerland (22-23 March 2018), two workshops (17.-19.09.2018 at RHUL an 9.-10.10.2018 at UHEI) and joint field campaigns. For the school, networking has been reinforced by the joint field campaign activities organized in parallel, and by the discussions at the end of each day with school participants (including external partners) about intellectual property rights, career development planning, ethics and good practices, gender issues, and the usage of networking platforms.

#### 2.2.5 WP5 – Project Management (Lead: Thomas Röckmann, UU)

The Project Management and the work done are described in detail in Chapter 4.

#### 2.2.6 WP6 - Ethics (Lead: Thomas Röckmann, UU)

Within WP6 - Ethics – two deliverables (D6.1 and D6.2) have been submitted. The ethical aspects of the project, with focus on the use of drones, were addressed and will be applied throughout the project lifetime.



## 2.3 Deliverables

Within the first reporting periods 16 deliverables were due. Additional to the due deliverables, the consortium started preparation and execution of upcoming deliverables. Table 2.11 gives an overview of all project deliverables and their actual status.

Table 2.11:	overview	of project	deliverables	and their	actual status
		_			

Scientific	Deliverables

	Deliverables		D	
No.	Deliverable Title	Lead	Due Date	Status*
D1.1	Lightweight CH₄ sensor and AirCore developed and deployed on UAV	RUG	24	In progress Preliminary tests for the lightweight sensor have been accomplished at EMPA on a bench-based system in order to characterize the dedicated custom-built, ring-shaped segmented multipass cell. A mechanically rugged and lightweight mobile measurement device was designed for application on board a UAV. The prototype UAV AirCore has been successfully developed and tested by RUG. A validation flight comparing the UAV AirCore measurements and 60-m measurements was performed (Lutjewad, NL).
D1.2	Report / publication on CH <sub>4</sub> emissions from wetland and lakes in Sweden	LU	30	In progress The first airborne measurement campaign using the LU aircraft needed to be postponed due to technical and administrative problems.
D1.3	Report / publication of results from the campaign in Silesia, Poland	AGH	36	In progress Two CoMET campaigns in Silesia, Poland, have been executed and data are under evaluation. New campaigns are in discussion.
D1.4	Improved emission factors for different source categories from mobile measurements	UHEI	42	In progress Several measurement campaigns on regional CH <sub>4</sub> sources have been executed to ensure a solid data base.
D1.5	Report on harmonized method for mobile CH <sub>4</sub> and <sup>13</sup> CH <sub>4</sub>	UHEI	18	DELAYED The measurement techniques used by different groups have been inspected and compared during the first campaign in Petten. A draft on the harmonization of measurement methods is in preparation. The report will be used to update the D5.3.
D2.1	Isotopic measurements linked to common scale	RHUL	18	Submitted (month 20) An agreed common scale has been defined by UU and RHUL for C- isotopic measurement of CH <sub>4</sub> , and a correction factor derived so that all samples analysed so far on the MEMO <sup>2</sup> project can be retrospectively corrected to this scale, and likewise going forward for all new analyses on the project. Inter-comparison cylinders will be circulated additionally around the mobile laser-based mobile isotopic measurement groups to link then to the common scale in addition to UU and RHUL. Once the CRDS partners have measured the cylinders we will consider how to circulate a more expansive document to the wider methane isotopic community.
D2.2	Improved isotopic source signatures of local and regional CH <sub>4</sub> emissions	UU	36	In progress First measurements are executed, future measurements are in planning
D2.3	Publications on the use of isotopes for CH <sub>4</sub> source attribution in urban / industrial regions	RHUL	36	Not relevant yet, activities related to this deliverable are described in detail in task 2.3
D2.4	Publication on temporal / meteorological influences on CH <sub>4</sub> at fixed sites	RHUL	42	Not relevant yet, activities related to this deliverable are described in detail in task 2.4
D2.5	Report providing isotopic maps at grid scale from inventories and atmospheric measurements	UU	42	In progress First measurements are executed and results are evaluated, future measurements are in planning
D3.1	New tools to estimate CH <sub>4</sub> source strengths from point sources, including mobile measurements	WU	24	In progress The tools such as MicroHH have been further developed. Now it is possible to simulate multiple sources at arbitrary locations in the domain and the problem of circular boundary conditions has been solved. The model is now ready to interpret observations delivered by WP1 and WP2. First publications are planned.
D3.2	Improved bottom-up European CH <sub>4</sub> emissions	UU	30	In progress During the 1 <sup>st</sup> reporting periods the focus was laid on measurement campaigns to ensure a solid data base. So, there are no particular outputs related to the inventories. The task of constructing high- resolution emission maps and include the isotopic signatures will be implemented in the upcoming reporting period and it is planned to implement findings in the TNO MACC inventory.
D3.3	Forward modelling simulations of CH <sub>4</sub> and isotopologues	UVSQ	30	In progress New simulations of CH <sub>4</sub> mixing ratios have been performed at the European scale with the CHIMERE chemistry transport model using



MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

D3.4	Top-down estimates of EU-scale CH <sub>4</sub> emissions	Empa	42	the EDGAR version 4.3.2 and TNO-MACC_III emission inventories from the year 2011. Multi-year analyses have been carried out and also a large number of sensitivity experiments were performed. The comparison and the sensitivity tests aim a better understanding of the difference between modelled and measured CH <sub>4</sub> , and thus help reveal which part can be attributed to errors in inventories and serve the goal of estimating top-down CH <sub>4</sub> emissions on the European scale. In progress The GRAL model has been improved in order to make it more efficient and easily applicable to any given situation. This included (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, (iii) development of 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.
Manage	ement, Training, Recruitment an	d Dissem	ination Deli	
D4.1	Individual Career Development	UVSQ	12	Approved
	Plan for each ESR			
D4.2	Annual update of the CDP for each ESR	UVSQ	24	In progress ESRs discussed the CDPs at the 1 <sup>st</sup> Annual Meeting (month 13). The first update of the CDPs is scheduled for the 2 <sup>nd</sup> Annual Meeting (month 24).
D4.3	Two secondments for each ESR completed	UU	30	In progress First secondments have started, several are in preparation, see Table 2.13
D4.4	Two MEMO <sup>2</sup> schools organized	UU	30	In progress 1 <sup>st</sup> MEMO2 school has been organized from February 5 <sup>th</sup> to February 16 <sup>th</sup> in Schoorl (the Netherlands), see report on MS2, the 2 <sup>nd</sup> School is scheduled in project month 24, associated to the Mid-term Review Meeting (18 – 21 February 2018)
D4.5	Annual update of the CDP for each ESR	UVSQ	36	Not relevant yet The second update of the CDPs will be initiated at the end of 2019.
D4.6	Two conference participations for each ESR completed	UU	42	In progress Conference participations are given in Table 2.14
D5.1	MEMO <sup>2</sup> consortium agreement	UU	1	Approved
D5.2	Project Management Plan (PMP)	UU	6	Approved
D5.3	Data management, Dissemination & Exploitation Plan (DDEP)	UU	6	The DDEP was submitted in month 6, an update is in progress
D5.4	Recruitment of ESRs finished	UU	9	Approved
D5.5	Project meetings organized (UU, Empa, RUG, RHUL, UHEI)	UU	48	In progress First meetings organized: Kickoff (UU), 1 <sup>st</sup> Annual Meeting (Empa), 2 <sup>nd</sup> Annual Meeting (UVSQ)
D5.6	Reports approved by Supervisory Board and sent to EC	UU	48	In progress
D5.7	Communication infrastructure established – interactive website including public dissemination	UU	6	Approved
D5.8	Progress Report	UU	13	Approved
D5.9	Mid-term Review Report	UU	22	Submitted
D5.10	Supervisory Board of the network	UU	2	Approved
D6.1	NEC – Requirement No.1	UU	6	Approved
D6.2	EPQ – Requirement No.2	UU	6	Approved

D5.9 MEMO<sup>2</sup> – Midterm Review Report

\*Note: The status "In Progress" may differ between this table and the status given in the ESR reports (in the attachment). This is due to the differing involvement of ESRs into the planning and execution of deliverables.



## **2.4 Milestones**

Within the first reporting periods the consortium achieved all 10 envisaged milestones. At the 2<sup>nd</sup> Annual Meeting the consortium will discuss and plan the upcoming milestones for the third period.

Nr.	Milestone Title	Lead	Due date	Status / Report
M1	ESRs trained at host institute with mobile equipment	UHEI	10	Achieved (see D5.8)
M2	First intensive campaign with training in the Netherlands	RUG	12	Achieved (MS2 report submitted)
М3	Workshop on Gaussian plume and dispersion models	UHEI	15	Achieved (MS3 report submitted)
M4	Lightweight CH <sub>4</sub> sensor and AirCore developed and deployed on UAV	RUG	24	In progress Prototypes have been successfully developed and tested, and are under improvement.
M5	Second intensive campaign in Silesia (Poland)	AGH	30	In progress MEMO <sup>2</sup> participated in several campaigns, including 2 campaigns in Silesia, see Table 2.14. A third intensive campaign to Silesia is in discussion, eventually this campaign will be re-located to Rumania.
M6	ESRs trained at host institute to measure/interpret isotope data	RHUL	12	Achieved (see D5.8)
M7	Comparative isotopic scale for project groups established	UU	15	Achieved (see D2.1), however, activities are ongoing and a comparison of co-located sampling can be made
M8	Workshop on isotope measurement techniques and data interpretation	RHUL	20	Achieved (MS8 report submitted)
M9	Isotopic maps at grid scale produced from inventories and atmospheric measurements	RHUL	36	In progress (see D2.5) Has commenced but need detailed rural and urban street-by-street surveys to be evaluated before this task can gather momentum. So far, the surveys that can be linked to emission inventories are those in the cities described under Task 2.3 above.
M10	Large Eddy Simulation Tools ready for campaign & workshop	WU	12	Achieved (MS10 report submitted)
M11	First updated CH₄ emission map EU	UU	30	In progress Currently the focus has been on landfill emissions, and analysis now works on the quantification of land-fill emissions. Plans are to implement these new findings in the TNO MACC inventory. ESR10 is in close contact with WU to use suitable modelling tools to derive emission strength from concentration measurements.
M12	Workshop on top-down emission estimates	UVSQ	30	Not relevant yet
M13	Two secondments for each ESR completed	UU	30	In progress First secondments have started, several are in preparation (Table 2.13).
M14	Two MEMO <sup>2</sup> schools organized	UU	30	In progress 1 <sup>st</sup> MEMO2 school has been organized from February 5 <sup>th</sup> to February 16 <sup>th</sup> in Schoorl (the Netherlands), see report on MS2 2 <sup>nd</sup> School planned in project month 24, associated to the Mid-term Review Meeting
M15	Two conference participations for each ESR completed	UU	42	In progress Conference participations are given in Table 3.1
M16	Communication infrastructure established – interactive website including public dissemination	UU	6	Achieved (see D5.7)
M17	Planned recruitments completed and recruited fellows enrolled in PhD programme	UU	12	Achieved (see D5.4)
M18	Project meetings organized (UU, Empa, RUG, RHUL, UHEI)	UU	48	In progress First meetings organized: Kickoff (UU), 1 <sup>st</sup> Annual Meeting (Empa), 2 <sup>nd</sup> Annual Meeting (UVSQ)



## 2.5 Impact

MEMO<sup>2</sup> aims on impact on different levels such as personal levels for the ESRs (career perspective and employability), structural training levels (national / international training) and scientific levels (provision of data and contribution to the evaluation of EU greenhouse gas emission reduction policies). After the first two years of MEMO<sup>2</sup> all expected impacts are still relevant and the consortium is successfully working in the direction of them.

#### 2.5.1 Impact on ESRs

The project and its network activities have a positive impact on the career of the ESRs: they are introduced to a highly interdisciplinary training program and get acquainted with techniques to identify CH<sub>4</sub> emissions in the atmosphere (WP1), attributed emissions to various source categories (WP2) and quantified such emissions from the local to the European scale (WP3). They are performing state-of-the-art measurements and develop modelling approaches. All ESRs are involved in secondments and have already visited one or more consortium partners different from their host institution, which impact the career of the ESRs not only by increasing their professional knowledge, but also their networking and complementary skills. All ESRs have to report on their secondments, either by a template-based report or a contribution to the public blogs on the project website. The project management encourages the ESRs to report as a blog to enhance both the writing skills of the ESRs and the visibility of the project towards the public.

The secondment schedule as described in the Grant Agreement has been adjusted to ensure them as efficient as possible for the ESRs. This was necessary e.g. due to the late recruitment of several ESRs, due to the execution of several mobile measurement campaigns which often depend on circumstances such as weather conditions or unexpected delays in sample measurements. Table 2.13 gives an overview of planned and already executed secondments.

ESR	Secondments executed / ongoing	Secondments planned
ESR1: Piotr Korben (UHEI)	AGH (22.05.18 – 10.06.18, 31.10.18 – 9.11.18)	LSCE (February / March 2019)
ESR2: Katarina Vinkovic (RUG)	ECN (October 2018 – February 2019 (5 months)	EMPA (October 2019)
ESR3: Mila Stanisavljevic (AGH)	PGI (short preparation visit 18.10 20.10.2017)	UU
	UHEI (13.0110.02.2018)	
ESR4: Patryk Lakomiec (LU)	Afvall Sverige - Single preparation days	EMPA (2019)
	RHUL (19.1130.11.2018)	
ESR5: Sara Defratyka (UVSQ)	RHUL (17.0613.07.2018)	
ESR6: Badrudin Stanicki (EMPA)	No secondments due to resignation	New ESR started 01.12.2018
ESR6b: Jonas Ravelid (EMPA)	Start date planned 01.12.2018	
ESR7: Semra Bakkaloglu (RHUL)	RUG (17.0926.10.2018)	
ESR8: Malika Menoud (UU)	AGH (16.0530.052018, to be continued)	
	RHUL (18.0614.07.2018)	
ESR9: Julianne Fernandez (RHUL)	UU (24.0921.10.2018)	UVSQ (February / March 2019)
ESR10: Hossein Maazallahi (UU)	-	TNO (April 218)
ESR11: Anja Raznjevic (WU)	Empa (26.0327.04.2018)	
ESR12: Randulph Morales (EMPA)	UVSQ (29.1023.11.2018)	
ESR13: Barbara Szenasi (UVSQ)	WUR (19 February 2018 – 19 March 2018)	TNO (March / April 2019)

Table 2.13: Overview of executed and planned secondments

Besides their work on their individual projects, the ESRs started to work together as a group. This is clearly visible by the self-organisation of their Skype meetings, which are independent from the PIs (for more details see Chapter 4.4).



#### 2.5.2 Impact on training

The training activities of MEMO<sup>2</sup> are in principle open to interested students or employees of the beneficiaries and partner organisations. As the capacities such as host space, men power and finances are limited, the schools and workshops were only announced within the network and open for associated researchers and staff members.

All our ESRs participated in several scientific and complementary skills training activities, not only within the network, but also from their host institutions. As they are offered a lot of international experience, they by this interact, influence and transfer their experiences on different levels. For the upcoming MEMO<sup>2</sup> school we are planning to involve groups of master students.

#### 2.5.3 Impact on science

Within the first two years, the MEMO<sup>2</sup> consortium was quite active in participating in scientific activities as several measurement campaigns and presenting the project on scientific platforms. By this MEMO<sup>2</sup> fostered the intersectoral exchange inside the consortium and initiated several collaborations outside the consortium, which has been intensified during the reporting period. First results of the campaign activities and collaborations are presented either in the individual ESR project reports or will be shown in the next reporting period.

#### 2.5.3.1 Collaborations

#### I) Participation in the CoMet (Carbon Dioxide and Methane Mission) campaigns

In August 2017, 18 scientists from AGH (Poland), DLR (Germany), RUG (Netherlands), and KIT (Germany) performed the CoMet Silesia pre-campaign as a joined effort. This was the first external measurement campaign, where MEMO<sup>2</sup> participated. The aim of the campaign was to investigate CH<sub>4</sub> emissions from co-located mines in the area of Jastrzębie Zdrój city, Poland. Four mines with seven exhaust shafts were sampled, with several activities in parallel, e.g. FTIR analysis of XCH<sub>4</sub> concentration, Aircore CH<sub>4</sub> analyses with cross-sections of plumes, ground base mobile measurements with Picarro close to the exhaust shafts and in background area, and a dozen of drone flights downwind the Pniowek V coal-mining shaft. From 14.05. - 12.06.2018 the CoMet 1.0 campaign took place in the Upper Silesia Region in Poland. Five MEMO<sup>2</sup> students from AGH, UHEI, UVSQ and UU participated. Measurements were mostly focused on mine shafts and the observation of CH<sub>4</sub> concentration and isotopic composition close to these facilities (https://h2020-memo2.eu/2018/06/25/memo2-at-comet/).

#### II) Participation in the FOAM campaign

The FOAM campaign in October 2017, where also scientists of MEMO<sup>2</sup> were involved, was a EUFAR TNA campaign devoted to the measurements of plumes from mine shafts and other sources by using a small Cessna aircraft. The aircraft was equipped with Picarro and HySpex analysers. For more information see: <u>http://www.eufar.net/weblog/2018/01/05/eufar-funded-flight-campaign-over-silesian-coal-district-quantify-methane-emission-rates-urban-and-biogenic-sources/</u>

#### III) Build up collaboration with the Environmental Defence Fund (EDF), USA

A first contact was made with Daniel Zawalla from EDF USA. Several MEMO<sup>2</sup> participants (e.g. RHUL, USQV, and ECCC) were involved in collaborations with the Climate and Clean Air Coalition (CCAC) and EDF, which projects show an obvious and strong link with MEMO<sup>2</sup>. Due to limited resources as time and budget, a participation in a planned measurement campaign was unfortunately not possible, but the ESRs have the opportunity to collaborate by exchanging data and experiences. The collaboration will be intensified in the next reporting period by joint measurement campaigns together with EDF Europe. Besides this, Daniel Zawalla agreed to join the Scientific Advisory Board as an additional member and to lecture at the 2<sup>nd</sup> MEMO<sup>2</sup> school.

#### IV) Build up collaboration with the Environmental Defence Fund (EDF), Europe

During the Antwerp industrial methane conference, MEMO<sup>2</sup> made a first contact with EDF Europe (via Michael Donatti and William Dow). EDF Europe is interested to measure CH<sub>4</sub> emissions in Romania. As the reported methane emissions from oil and gas in Romania are higher than anywhere else in Europe, this might also be an interesting target area for the MEMO<sup>2</sup>. The consortium agreed on this topic and



contacts in this direction were intensified during the second project year. MEMO<sup>2</sup> also collaborated in the UNEP European cities project together with EDF to measure the CH<sub>4</sub> concentration e.g. in Hamburg, Germany and London, UK.

#### V) Collaboration with DRL

ESR1 is working together with Andreas Fix group (DLR) to analyse and compare results.

#### VI) Collaboration with WU

ESR2 will set up an external collaboration with Dr. Nico Ogink and Dr. Leon Sebek from the University of Wageningen, the Netherlands. A goal of this collaboration would be to relate our measurements of CH<sub>4</sub> emissions with food intake, with respect to the amount, the diurnal cycles, and the seasonal cycles. Hopefully, this will provide us with some additional information to help design our strategy to make the measurements, and what farms / times we focus on in the next years.

VII) Collaboration with the Sveriges lantbruksuniversitet (Swedish University of Agricultural Sciences, SLU) ESR4 will collaborate with SLU in Umeå to execute research flights over a chrono-sequence of wetlands in northern Sweden (along the coast between Umeå and Luleå), which are scheduled for 2019. Data from the campaign will enlarge the database for quantifying CH<sub>4</sub> wetland emissions from different wetlands.

#### VIII) Collaboration with the ETHZ

ESR6 set up a collaboration with the Aquatic Chemistry group of Prof. B. Wehrli (EAWAG/ETHZ). After the resignation of the ESR this collaboration needs to be reviewed.

#### VIIII) Collaboration with the TU Delft

ESR8 is collaborating with Dr. Julia Gebert (Faculty of Civil Engineering and Geosciences, TU Delft), who is aiming at the quantification of the isotopic fractionation factor from the diffusion of methane through landfill soils. The experiments were carried out by her master student Tijmen Blom, during August 2018. They consisted in different soils chambers filled with soil of a range of densities and water content. The results will be presented in Tijmen Blom's thesis, and will be written in a publication.

#### X) Collaboration with the Royal Netherlands Institute for Sea Research (NIOZ) and GEOMAR

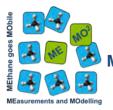
The collaboration with NIOZ started during the PELAGIA-439 cruise (June 2018), where samples were taken for the analysis of methane isotopes. More than 300 water samples, 37 atmospheric samples and 13 air samples from a gas seep were taken for isotopic analyses. The data will be analysed in collaboration with the research group at NIOZ, who collected water samples for drawing the depth profile of dissolved methane above the seep. During the cruise additional samples were taken for GEOMAR to also investigate the  $N_2O$  concentrations in the water column.

#### XI) Collaboration with Aerodyne

During the EGU conference in April, 2018, contact was made with Scott Herndon of Aerodyne who shared measurements from their campaigns in the Gulf of Mexico. ESR11 will compare their measurements with the simulations from MicroHH.

#### XII) Collaboration with Netherlands Earth System Science Center (NESSC, http://www.nessc.nl)

MEMO<sup>2</sup> will collaborate with the Netherlands Earth System Science Center (NESSC, <u>http://www.nessc.nl</u>) by providing input and data for developing a new Methane Module for school educational purpose.



MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

D5.9 MEMO<sup>2</sup> – Midterm Review Report

### 2.5.3.2 MEMO<sup>2</sup> measurement campaigns

#### Table 2.14: Overview of executed measurement campaigns within MEMO<sup>2</sup>

Host	Campaign	Start - End	Location	ESR	Description of work	Scientific objective	Samples nature/number	Future plans
RUG	1st Grijpskerk campaign	27.03.17	Grijpskerk, NL	ESR2	Drone measurements	Quantify CH <sub>4</sub> emissions	data	publication
LSCE	landfill	06.10.17	Butte-Bellot, FR	ESR5	Mobile measurement using acetylene as tracer	Estimation of emission from source	Mobile measurement with 20 transects	Estimated emission from landfill
AGH	Belhatow coal mine	19.10.17	Poland	ESR3	CH4 emission from non-point source			
Afvalzorg	Nauerna Iandfill	04.12.17 - 15.12.17	Amsterdam, NL	ESR10	Landfill Measurements, 2 days of measurements	Quantifying methane emission from landfills	Mobile measurement	
Afvalzorg	Zeeasterw eg landfill	29.11.17 - 05.12.17	Lelystad, NL	ESR10	Landfill Measurements, 2 days of measurements	Quantifying methane emission from landfills	Mobile measurement	
Afvalzorg	Braamber gen landfill	29.11.17 - 05.12.17	Almere, NL	ESR10	Landfill Measurements, 2 days of measurements	Quantifying methane emission from landfills	Mobile measurement	
RHUL	Yorkshire	Jan.18	Yorkshire, UK	ESR9	Survey of area to have fracking activity	Conduct and collect data for study on methane before, during and after	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], 3 bag samples for δ <sup>13</sup> C-CH4	
UHEI	l, Heidelberg	10.01.18	Heidelberg and surrounding area (Weinheim, Hähnlein, Kirchheim), DE	ESR1	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farm, gas compressor station, biogas plant / 10	Values of isotopic composition of methane from visited sources, calculate emission factor
LSCE	Gas compress or station	19.01.18	Beynes, France	ESR5	First attempt to measure site in Ile de France	Primary survey to optimize condition for further measurement	Mobile measurement	No further plans
RHUL	Yorkshire (KM5)	30.01.18 - 31.01.18	Yorkshire, UK	ESR7	Mobile car night measurement	Training on mobile methane measurement	25 bags were collected	
ECN	1st MEMO <sup>2</sup> school	09.02.18 - 11.02.18	Schoorl, NL	All ESRs	Mobile car measurements across the North Holland province, methane release experiment,	Training purpose, Intercomparison and harmonisation of data and methods,	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], from Landfill, dairy farms, gas compressor station, city, bag samples for 5 <sup>13</sup> C-CH4	Conference presentation of results,cooperation with other groups, Estimated emission from landfill, further work with Polyphemus model on obtained data
UU	Utrecht city		Utrecht	ESR10	City measurements	Quantifying and identifying methane emission sources across Utrecht city	Mobile measurement	Comparing the results with other cities.
LSCE	Gas compress or station	05.03.18	Limoges- Fourches, FR	ESR5	First attempt to measure site in Ile de France	Primary survey to optimize condition for further measurement	Mobile measurement	No further plans
LSCE	Gas compress or station	13.03.18	Fontenay- Mauvoisin, FR	ESR5	First attempt to measure site in Ile de France	Primary survey to optimize condition for further measurement	Mobile measurement	No further plans
UHEI	IV, Heidelberg	26.03.18 - 27.03.18	Heidelberg and surrounding area (Hähnlein, Kirchheim, Scheidt), DE	ESR1	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Gas compressor stations, biogas plant / 15	Values of isotopic composition of methane from visited sources, calculate emission factor
UHEI	V, Heidelberg	26.04.18	Heidelberg and surrounding area (Weinheim, Ladenburg, Sinsheim), DE	ESR1	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farms, landfill / 13	Values of isotopic composition of methane from visited sources, calculate emission factor
RUG	2nd Grijpskerk campaign	03.05.18	Grijpskerk, NL	ESR2	Drone measurements	Quantify CH <sub>4</sub> emissions.	bag samples / 3 aircores	publication
RHUL	UNC1, Sutton	03.05.18	Sutton, UK	ESR7, ESR9	Urban city survey, mobile car night measurement	To locate and quantify methane mole fractions and isotopic signatures	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], 3	Gas leaks have been found



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

							bag samples for δ13C-CH4	
RHUL	Cambridg e	13.05.18	Cambridge, UK	ESR7	Cow Barn	Quantify CH4 mole fractions and isotopic signatures	CAM 1,2,3,4,5,6,7,8,9, 10,11,12 13,14,15,16,17,1 8,19	
RHUL	Brighton (UNC2)	14.05.18	Brighton city, UK#	ESR7	Mobile car night measurement	Quantify CH <sub>4</sub> mole fractions and isotopic signatures	UNC2- 1,2,3,4,5,6,7,8,9, 10,11,12 13,14,15,16,17,1 8	
AGH	CoMet	23.05.18 - 10.06.18	Sielsia, Poland	ESR1, ESR5, ESR8, ESR10	Mobile measurement in mining area: around mining shafts and in urban area, sampling of plumes identified by mobile CRDS measurements	Better understanding of emission from mining industry in Poland, urban area source mapping, isotopic characterisation of methane from Polish coal mines	10 days of mobile measurement around mining shafts, 6 nighttime measurement of urban area, 43 air samples in aluminium bags	Urban source mapping, find probably emission source in Silesia urban area, study the formation processes and influencing parameters, Values of isotopic composition of methane from coal mines on Silesia area, vertical profile of methane concentration in cooperation with aicrafts
RHUL	UNC 2	12.06.18	Norfolk / Lincolnshire	ESR9	LNG terminals survey		Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ], 18 bag samples for $\delta^{13}$ C-CH <sub>4</sub>	
RHUL	secondme nts	17.06.18 - 13.07.18	South of United Kingdom	ESR5	Mobile campaigns with in situ measurement of isotopic composition, nightlime measurement from common inlet by CRDS and IRMS	Calculated isotopic composition of different source, comparison of results obtained by CRDS and IRMS	5 measurement campaign,4 with using storage tube, 3 nighttime measurement,	Plume mapping and source isotopic comparison, in the future - further comparison of obtained results
RHUL	MEMO2- RHUL 1	22.06.18 - 05.07.18	Egham/Staines, UK	ESR7, ESR8, ESR9	Local source survey, sampling of methane sources identified by mobile CRDS measurements, mobile car night measurement	Measure and locate urban CH4 sources with visiting UU & UVSQ student, isotopic characterisations of various sources in the UK	Continuous measurements of [CH4], [CO2], [H2O], [C2H4], 6 bag samples for $\delta^{13}$ C-CH4, 47 air samples in aluminium bags	The data shows clear agreement between the source types, and the potential of isotopes for source identification. It is now to be shared and combined with RHUL and LSCE data.
UU, NIOZ	PELAGIA 439	22.06.18 - 02.07.18	Northsea, ship cruise	ESR10 , ESR8	Continuous methane measurements from oil/gas platforms and natural seeps, CTD samples bucket samples for isotopic analyses	Understanding methane emissions from anthropogenic and natural sources	13 glass flasks, 37 tedlar bag samples, continuous monitoring	
RHUL	MEMO2- RHUL 2	26.06.18	Oxford, Bicester, Milton Keynes	ESR7, ESR9	Urban city survey, mobile car night measurement	Measure and locate CH4 sources of waste facilities with visiting UU & UVSQ student, to quantify methane mole fractions and isotopic signatures	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], 6 bag samples for $\delta^{13}$ C-CH4	Some samples for IRMS RHUL & UU comparison
RHUL	MEMO2- RHUL 3	27.06.18	Spelthorne, UK	ESR7, ESR9	Urban city survey, mobile car night measurement	Measure and locate CH4 sources of waste facilities with visiting UU & UVSQ student, to quantify methane mole fractions and isotopic signatures	$\begin{array}{l} \mbox{Continuous} \\ \mbox{measurements} \\ \mbox{of [CH4], [CO2],} \\ \mbox{[H2O], [C2H6], 9} \\ \mbox{bag samples for} \\ \mbox{$\delta^{13}$C-CH4} \end{array}$	Gas leaks have been found
RHUL	MEMO2- RHUL 4	28.06.18	Isle of Grain/Kent	ESR7, ESR9	Urban city survey, mobile car night measurement	Measure and locate CH4 sources of waste facilities with visiting UU & UVSQ student, to quantify methane mole fractions and isotopic signatures	$\begin{array}{l} \mbox{Continuous} \\ \mbox{measurements} \\ \mbox{of [CH4], [CO2],} \\ \mbox{[H2O], [C2H6], 12} \\ \mbox{bag samples for} \\ \mbox{$\delta^{13}$C-CH4} \end{array}$	Some samples for IRMS RHUL & UU comparison
UHEI	VI, Heidelberg	04.07.18 - 06.07.18	Heidelberg and surrounding	ESR1	Mobile measurements	Measurements isotopic composition	Dairy farms, landfill, gas	Values of isotopic composition of

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report



			area (Weinheim, Ladenburg, Ludwigshafen, Hähnlein)		using CRDS Analyzer Picarro G2201-I and release experiment	of methane from different sources to check seasonal variations	compressor station / 10	methane from visited sources, calculate emission factor
RHUL	MEMO2- RHUL 5	05.07.18	Devon, UK	ESR7, ESR9	Heathfield landfill survey, Mobile car night measurement	To quantify methane mole fractions and isotopic signatures for Heathfield Landfill, and Exeter region, visiting UU & UVSQ student	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], 14 bag samples for ŏ13C-CH4	Some samples for IRMS RHUL & UU comparison
TNO	NOGEPA			ESR10 , ESR8	Continuous methane measurements from oil/gas platforms	Understanding methane emissions from oil and gas extraction platforms	22 Bag samples, continious measurements	
Empa	Test Flight 1	10.08.18	Empa, Dübendorf	ESR6	Test flight with mobile methane spectrometer	First test for the assessment of measurement characteristics during flight conditions.	CH4 concentration with 2Hz resolution, additional: H2O conc., T, p, GPS pos.	Results can be used for quantifying and improving the stability of the spectrometer, further test flights are planned
UHEI	VII	24.08.18	Heidelberg and surrounding area (Weinheim, Sinsheim, Kirchheim)	ESR1	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farm, landfill / 17	Values of isotopic composition of methane from visited sources, calculate emission factor
AGH	Lublin coal Basin	28.08.18	Poland Silesia	ESR3				
RHUL	UNC 3, UNC 4	29.08.18	Brighton and Hillingdon, UK	ESR9	Urban city survey	Measure and locate urban CH4 sources	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], 25 bag samples for $\delta^{13}$ C-CH4	
LSCE	Paris urban area	07.09.18 - 26.09.18	Paris	ESR5	Mobile measurement in Paris urban area	Source mapping in Paris urban area	3 measurement days	Not significant sources detected
RUG	RUG-1	25.09.18 - 02.10.18	Groningen	ESR7	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures for Groningen city and Germany pit fire	GROG1- 1,2,3,4,5,6,7,8	Will be evaluated
UU, TUM*	Oktoberfe st		Munich, DE	ESR10	Stationary and mobile measurements from Oktoberfest festival	Understanding methane emissions from the citiy and a major local event	Continuous measurements	
Empa	Test Flight 2	10.10.18	Empa, Dübendorf	ESR6	Test flight with mobile methane spectrometer	Testing a slightly modified setup and the observation of artificial methane release.	CH4 concentration with 2Hz resolution, additional: H2O conc., T, p, GPS pos.	Results can be used for quantifying and improving the stability of the spectrometer, further test flights are planned
LU	Bag sampling	12.10.18	Skogaryd Mycklemosse, wetlands	ESR4	automatic chambers system on wetlands	Spatial distribution of isotopic composition	120 from 6 chambers	
UU, UHH*, MPI*	Ham 1, 2, & 3	18.10.18 - 09.11.18	Hamburg, Germany	ESR9, ESR8, ESR10	Urban city survey	Quantifying and identifying methane emission sources across the Hamburg Assist and learn protocol of UU surveys to measure and locate urban CH4 sources, Isotopic characterisation of urban methane sources	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], 104 bag samples for $\delta^{13}$ -CH4 & $\delta^{2}$ H-CH4 Air samples in aluminium bags	Analysis in process
RUG	RUG-5	18.10.18	Lutjeward	ESR7	Observation	Observation of sampling and measurement techniques, taking background samples for isotopic methane signatures	ROG5-1,2	Will be evaluated
RUG	3rd Grijpskerk campaign, RUG-6	19.10.18	Grijskerk Cow Farm	ESR2, ESR7	Farm- Drone measurement	Observing UAV Aircore Measurement Techniques to quantify methane mole fraction. Figure outing Duct Farm isotopic methane signatures	bag samples / 4 aircores	publication

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report



RHUL	UNC 5, UNC 6	25.10.18	Ealing and Harrow, UK	ESR9	Urban city survey	Measure and locate urban CH4 sources	Continuous measurements of [CH4], [CO2], [H2O], [C2H6], 4 bag samples for δ <sup>13</sup> C-CH4	Analysis in process
UHEI	VII	25.10.18	Heidelberg and surrounding area (Weinheim, Ladenburg, Hähnlein)	ESR1	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farms, gas compressor station / 18	Values of isotopic composition of methane from visited sources, calculate emission factor
UHEI	ll, Heidelberg	26.10.18	Heidelberg and surrounding area (Kirchheim, Sandhausen)	ESR1	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Biogas plant, gas compressor station, city / 12	Values of isotopic composition of methane from visited sources, calculate emission factor
AGH	Krakow	10.12.18 - 20.12.18	Krakow, PL	ESR8	Sampling of various methane sources in the surroundings of Krakow.	Isotopic characterisation of the main local sources influencing the methane elevations measured in the city.	Air samples in aluminium bags, quantity to be determined	The results will be combined with the continuous in-situ measurements performed through the winter, to potentially identify the sources of methane elevations.
AGH	Post mining emission	planned?	Poland Silesia	ESR3		Checking methane emission from closed mines		
AGH	Samplings from mine shafts	planned?	Poland Silesia	ESR3	Collecting of bag samples directly from mines shafts	10		

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

\*TUM: Technical University Munich, UHH: University of Hamburg, MPI: Max-Planck Institute

# 2.6 Evaluation of the Action by the external Supervisory Board (SB)

The external Supervisory Board (SB) was invited to participate to the 1<sup>st</sup> Annual Meeting. Martin Heimann (MPG-BGC, Jena), Colm Sweeney (NOAA, Boulder), and Alex Vermeulen (ICOS, Lund) were present during the two days. Gabrielle Petron (NOAA) could unfortunately not participate due to personal reasons.

All relevant documents, e.g. Grant Agreement and deliverables were provided in advance and individual progress of the project was demonstrated at the first day of the meeting. At the end of the meeting the SB gave an overview of its impression of the project and discussed future planning with the consortium. Based on the evaluation report template used by the EC, the Supervisory Board was asked to submit a detailed joint review report afterwards. The SB report is available on SURFdrive.

As a brief overview, the SB stated that the project shows good progress despite delays in ESR recruitment, with impressive results from the measurement campaign executed in February. The campaign provided a good groundwork; next steps for upcoming campaigns should be the formulation of clear scientific questions and a detailed method selection.

MEMO<sup>2</sup> includes many different technological and scientific advances covering the development and application of mobile platforms, isotope studies and modelling. While each of these activities represent state-of-the-art and are novel and exciting, the SB suggested that the project would profit by defining already early in the project lifetime a few (1-3) "flagship" projects which ties in all the different project WPs and project partners. Given the expertise of the entire project consortium, these projects could provide substantial visibility, not only in the scientific domain, but also in the wider climate mitigation policy arena, and could lead to a few key publications in major journals. Defining these early in the project lifetime would allow for sufficient planning. Examples could be a revisit of quantification of CH<sub>4</sub> emissions from a major Silesian coalmine or coalmine field using the full toolset as developed in the project. Alternative hot-spots could be the emissions from a particular city, an important wetland/lake complex, a poorly known geological source or a region with high emissions from ruminants. The consortium discussed this extensively not only during the 1<sup>st</sup> Annual Meeting but also afterwards during the regular tele-conferences and decided to intensify the city measurement campaigns and to explore opportunities for collaborations with Romanian research institutes.

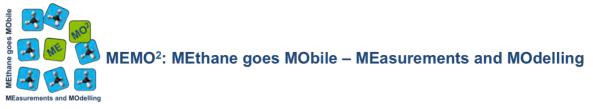


In the proposal, mapping the road to new top-down based pan-European CH<sub>4</sub> emissions estimates based on the new MEMO<sup>2</sup> information is envisaged. To reach this goal, quite a few steps have to be designed and planned out as it is a very long way from detailed CH<sub>4</sub> emissions from a few Dutch farms to the livestock CH<sub>4</sub> emissions from Europe as a whole. Especially the spatial and temporal representation of the emission drivers need to be carefully addressed. It does not suffice e.g. to claim that the EDGAR database is not good – what is the alternative? Since the up scaling of detailed source information and the complementary downscaling by the top-down method are major steps towards this goal the SB suggested to early develop a road map detailing the various steps of how this goal will be achieved later in the project. This document could also define the characteristics of a base case scenario (spatial/temporal resolution, domain, target time period) so that contributing work by the different groups can be focused. As the focus was laid on starting up the measurement campaigns, the set-up of a road map will be discussed on the 2<sup>nd</sup> Annual Meeting in Paris.

Regarding dissemination and outreach activities, the SB suggested progress / campaign / project blogs written by young scientists, which have proven to be very useful. So, the ESRs are encouraged to report their secondments and also campaign activities as a blog for the website (<u>https://h2020-memo2.eu/category/blog/</u>). Due to the general workload of the ESRs, regular periodic blogs are not planned. Also, extra writing exercises such as scientific reviews are only planned for the next reporting period. The website as one of the main outreach channels has been updated, and is continuously progressing.

The societal impacts of the project are a strong point of it and the SB suggested to intensify activities in this direction and also to strengthen the link to external projects and organisations, e.g. IG3IS or ICOS. A first step was made by storing MEMO<sup>2</sup> data at the ICOS Carbon Portal, data will be public available later via this platform. Besides this, MEMO<sup>2</sup> initiated and started several collaborations and continuously increased dissemination activities, which will increase the public awareness and societal impact over time (see Chapter 2.5.3.1 and Table 3.1).

Another issue mentioned by the SB was the update and adjustment of the DMP at some points. They suggested that the consortium should study data licences and prepare to choose one. Besides this discovery metadata are not well covered (ISO19115/INSPIRE/Dublin Core). Data formats are part of interoperability, proprietary formats are deprecated (e.g. excel). An internal data sharing solution is urgent and critical for success. As mentioned above, the storage of data in the ICOS repository is a first step towards this direction. Accordingly, the DMP will be updated based on the deliverable D1.5. However, this deliverable which is led by the UHEI has shown substantial delay.



# 3. Communication, dissemination and exploitation

MEMO<sup>2</sup> has been actively communicated and disseminated as EU project in general and by presenting results of the individual ESR projects. The consortium is using different platforms to approach different target groups.

# **3.1 Scientific platforms**

Commonly used scientific platforms are conferences such as EGU, AGU, PEFTEC-IMMC, NCGG or the ICOS Science Conference, targeting on scientists from relevant disciplines. At the conferences we are not only communicating the individual scientific projects within MEMO<sup>2</sup> and their results, but also MEMO<sup>2</sup> as itself. Aim is to increase the impact of the consortium as such and the understanding of the project type H2020-ITN-ETN in general.

Although the ESRs were recruited in the second half of the first project year in 2017 and therefore no or only preliminary scientific results were available, the project was presented already in April at the EGU 2017 (<u>https://egu2017.eu</u>) in Vienna and in November at the PEFTEC-IMMC 2017 in Antwerp (<u>https://www.youtube.com/watch?time\_continue=187&v=MmV9Dga6A58</u>). Especially during the PEFTEC-IMMC 2017, MEMO<sup>2</sup> was introduced intensively and mentioned in several talks and posters. Both two conferences were also used to advertise the non-filled ESR positions.

Since then the dissemination activities were intensified and are ongoing (see Table 3.1). At the EGU2018 (https://egu2018.eu) MEMO<sup>2</sup> was presented in several scientific and administrative orals and posters, but also during a workshop (SC3.13) and a public splinter session (SMP1). During the ICOS Science Conference (https://www.icos-ri.eu/3rd-icos-science-conference-2018) 6 of our ESRs presented first results by posters. Upcoming conferences are e.g. the EGU 2019 (https://egu2019.eu) were MEMO<sup>2</sup> results will be presented in a in Vienna. dedicated CH<sub>4</sub> session (https://meetingorganizer.copernicus.org/EGU2019/session/30576), the PEFTEC-IMMC 2019 (https://www.ilmexhibitions.com/methane/) in Rotterdam, or the NCGG 8 (https://www.ncgg.info/home) in Amsterdam, where we are planning to organise a dedicated MEMO<sup>2</sup> session. Contributions of MEMO<sup>2</sup> are also planned for the next ICOS Scientific conference, which will also be held in the Netherlands. Table 3.1 shows the dissemination activities of MEMO<sup>2</sup> at the scientific platforms so far.

Nr.	Conference name	Location	Date	Presentation [oral / poster]	Title of presentation	Authors / Conveners	Public	Link
1	EGU 2017	Vienna, Austria	24-28 April 2017	Poster	MEMO2: Methane goes MObile – MEsurements and MOdelling – Part 1	Walter, S., Röckmann, T., and the MEMO2 team:	yes	http://meetingorganizer.cop ernicus.org/EGU2017/EGU 2017-13442.pdf
2	EGU 2017	Vienna, Austria	24-28 April 2017	Poster	MEMO2: Methane goes MObile – MEsurements and MOdelling – Part 2	Röckmann, T., Walter, S., and the MEMO2 team	yes	http://meetingorganizer.cop ernicus.org/EGU2017/EGU 2017-15754.pdf
3	EGU 2017	Vienna, Austria	24-28 April 2017	Splinter meeting SMP6	MEMO2: Methane goes MObile – MEsurements and MOdelling	Walter, S. and Röckmann, T.	yes	http://meetingorganizer.cop ernicus.org/EGU2017/sessi on/25151
4	Industrial Methane Measuremen t Conference –PEFTEC 2017	Antwerp, Belgium	29-30 November 2017	oral	Short duration, high precision methane flux measurements: Implications for annual CH4 emission reporting and CH4 mitigation strategies	Denier van der Gon, H., Arzoumanian, E., Bouchet, C., Jonkers, S. Kelly, R., Morin, D.	yes	https://www.imexhibitions.c om/petec/abstracts/Short+d uration%2C+high+precision +methane+flux+measureme nts%38+tmplications+for+a nnual+CH4+emission+repor tine+an4-CH4+mitiation+s trategies/223/
5	Industrial Methane Measuremen t Conference –PEFTEC 2017	Antwerp, Belgium	29-30 November 2017	oral	Identification and validation of methane sources using carbon-13 measurements	Fisher, R., Lowry, D., Zazzeri, G., al- Shalaan, A., France, J., Brownlow, R.	yes	https://www.ilmexhibitions.c om/peftec/abstracts/identific ation+and+validation+of+m ethane+sources+using+car bon-13+measurements/210/
6	Industrial Methane Measuremen t Conference –PEFTEC 2017	Antwerp, Belgium	29-30 November 2017	oral	Validating methane measurement techniques	Robinson, R.	yes	https://www.imexhibitions.c om/peftec/abstracts/Validati ngt-methane+measurement +techniques/224/

Table 3.1: Dissemination activities of MEMO<sup>2</sup>



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

7	Industrial Methane Measuremen t Conference –PEFTEC	Antwerp, Belgium	29-30 November 2017	oral	Methane source attribution: Methane and ethane analysis using a portable battery-powerd Picarro Cavitiy Ring-	Winkler, R.	yes	https://www.ilmexhibitions.c om/peftec/abstracts/Methan e+source+attribution%3A+ Methane+and+Ethane+Anal ysis+Using+a+portable+Batt once powerdetPlocarro+Cavity+Ri
	2017				Down spectrometer			<u>powered+Picarro+Cavitv+Ri</u> <u>ng-</u> <u>down+Spectrometer/238/</u>
8	Industrial Methane Measuremen t Conference –PEFTEC 2017	Antwerp, Belgium	29-30 November 2017	oral	A new technique fro detecting gas emissions and estimating the locations and mass emission rates of sources	Hirst, B., Randell, D.	yes	https://www.ilmexhibitions.c om/oeftec/abstracts/A+new +technique+for+detecting+q as+emissions+and+estimati na+the+locations+and+mas s+emission+rates+of+sourc es./241/
9	Industrial Methane Measuremen t Conference –PEFTEC 2017	Antwerp, Belgium	29-30 November 2017	poster	Methane source distribution in the complex landscapes of the United Kingdom: isotopic characterisation, seasonal variation and inventory validation	Lowry, D.	yes	https://www.imexhibitions.c om/peftec/abstracts/Methan e+source+distribution+in+th e+complex+landscapes+of+ the+Unite1+Kingdom%2A+i solopic+characterization%2 C+seasonal+vaniation+an+ inventory+validation/284/
10	Industrial Methane Measuremen t Conference –PEFTEC 2017	Antwerp, Belgium	29-30 November 2017	poster	MEMO <sup>2</sup> : MEthane goes MObile - MEasurements and MOdelling	Walter, S., Röckmann, T.	yes	https://www.ilmexhibitions.c om/oeftec/abstracts/MEMO 2%3A+MEthane+qoes+MO <u>bile+-</u> <u>+MEasurements+and+Mod</u> <u>elling/214/</u>
11	Industrial Methane Measuremen t Conference –PEFTEC 2017	Antwerp, Belgium	29-30 November 2017	poster	Isotopic composition of methane from exhausts of mines and gas fields in South Poland	Necki, J., Zimnoch, M., Jasek, A., Chmura, L., Lakomiec, P., Korben, P., Wolkowicz, W.	yes	https://www.ilmexhibitions.c om/peftec/abstracts/sotopic t_comoosition+of+methanet+ from+exhausts+of+mines+a nd+qas+fields+in+South+Po land./229/
12	EGU 2018	Vienna, Austria	8-13 April 2018	poster	Starting an EU project – lessons learnt from the first year of MEMO <sup>2</sup>	Walter, S.	yes	https://presentations.coperni cus.org/EGU2018- 7406_presentation.pdf
13	EGU 2018	Vienna, Austria	8-13 April 2018	poster	MEMO <sup>2</sup> : MEthane goes MObile – MEsurements and MOdelling	Walter, S., Röckmann, T., and the MEMO <sup>2</sup> team	yes	https://meetingorganizer.co pernicus.org/EGU2018/post ers/26398
14	EGU 2018	Vienna, Austria	8-13 April 2018	Splinter meeting SMP1	MEMO <sup>2</sup> : Methane goes MObile – MEsurements and MOdelling	Walter, S. and Röckmann, T.	yes	https://meetingorganizer.co pernicus.org/EGU2018/sess ion/29051
15	EGU 2018	Vienna, Austria	8-13 April 2018	Short Course SC3.13	How to apply for the MSCA grants IF and ETN	Walter, S., Ingrin, J., Henkel, D., Padrón- Navarta, J.A.	yes	https://meetingorganizer.co pernicus.org/EGU2018/sess ion/28965
16	EGU 2018	07. – 12.04. 2018.	Vienna (Austria)	poster	Bottom – up methane budget estimation from the sources over Upper Silesian Coal Basin	Mila Stanisavljevic, Jaroslaw Necki, Miroslaw Zimnoch, Lukasz Chmura, Michal Galkowski, Wojciech Wolkowicz, and Patriyk Lakomiec	yes	https://meefingorganizer.co pernicus.org/EGU2018/EG U2018-14798.pdf
17	EGU 2019	Vienna, Austria	7-12 April 2019	Splinter meeting SMP7	MEMO <sup>2</sup> : MEthane goes MObile – MEsurements and MOdelling	Walter, S. and Röckmann, T.	yes	https://meetingorganizer.co pernicus.org/EGU2019/sess ion/33411
18	3 <sup>rd</sup> ICOS Science Conference	Prague (Czech Republic)	10. – 14.09. 2018	poster	Quantification of methane emissions from dairy cows in the Netherlands	K.Vinkovic + T.Andersen, M.de Vries, W. Peters, A. Hensen, H. Chen	Yes	https://conference.icos- ri.eu/wp- content/uploads/2018/08/IC OS-SC-Programme.pdf
19	3 <sup>rd</sup> ICOS Science Conference	Prague (Czech Republic)	10. – 14.09. 2018	poster	Using the PicarroG2301-m for airborne eddy covariance measurements of GHG fluxes	Lakomiec, P., Peltola, O., Holst, J., Rinne, J.	yes	https://conference.icos- ri.eu/wp- content/ubads/2018/08/IC OS-SC-Programme.pdf
20	3 <sup>rd</sup> ICOS Science Conference	Prague (Czech Republic)	10. – 14.09. 2018	poster	Mobile measurement of CHa isotopes in urban, mining and industrial environments	Sara Defratyka, Camille Yver Kwok, Arjan Hensen, Jaroslaw Necki, Dave Lowry, Jean- Daniel Paris, Pawel Jagoda, Philippe Bousquet	yes	https://conference.icos- ri.eu/wp- content/ubloads/2014/08/I/C OS-SC-Programme.pdf
21	Flair conference	12.09.201 8	Assisi, Italy	oral	A compact QCL absorption spectrometer for mobile, high-precision methane measurements aboard drones	Badrudin Stanicki, <u>Béla Tuzson</u> , Liu Chang, Manuel Graf, Philipp Scheidegger, Herbert Looser and Lukas Emmenegger	no	https://fox.ino.ti/filair/FLAIR %202018%20- %20Scientific%20Program, <u>pdf</u>

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report



22	3 <sup>rd</sup> ICOS Science Conference	Prague (Czech Republic)	10. – 14.09. 2018	poster	Waste Source in the UK	S. Bakkaloglu + D.Lowry, R. Fisher, E. Nisbet	https://conference.icos- ri.eu/wp- content/uploads/2018/09/IC OS2018SC Book of Abstr acts.pdf
23	3 <sup>rd</sup> ICOS Science Conference	Prague (Czech Republic)	10. – 14.09. 2018	poster	Isotopic characterization of methane from mine shafts in the Silesia region	Malika Menoud, Hossein Maazallahi, Mila Stanisavljevic, Thomas Röckmann, Jaroslaw Necki	https://www.researchqate.n et/oublication/327655309_ls otopic_characterisation_of_ methane_from_mine_shafts in_the_Silesia_region
24	EGU 2018	Vienna, Austria	8-13 April 2018	poster	Modeling CH4 dispersion using three modeling techniques to prepare a field campaign on methane emissions	Anja Ražnjević , Chiel van Heerwaarden , Maarten Krol	https://meetingorganizer.co pernicus.ora/EGU2018/EG U2018-13940.pdf
25	EGU 2018	Vienna, Austria	8-13 April 2018	poster	Atmospheric monitoring of methane emission at the European scale	B. Szénási, I. Pison, G. Broquet, M. Saunois, P. Bousquet, A. Berchet	https://presentations.coperni cus.org/EGU2018- 14964_presentation.pdf

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

### 3.2 Internet-based platforms

To reach out to a broader audience, we used internet-based platforms such a dedicated project website and social media. The coordinator takes responsibility on the more professional platforms such as LinkedIn and ResearchGate, the ESRs were encouraged to take responsibility of those social media where they are most active, e.g. Instagram, Facebook or Twitter.

#### 3.2.1 MEMO<sup>2</sup> website

The project website of MEMO<sup>2</sup> (<u>https://h2020-memo2.eu</u>) serves as a central communication platform within and outside the consortium, and for dissemination of project relevant information, including documents (e.g. GA, CA, deliverables, minutes), templates, and results. The website was launched two months before the official start of the project and is maintained by the coordinator, with input from all participants.

In November 2017, the website was transferred from a commercial host to the ICT service of the UU. Dropbox was used in the beginning to share documents and results, but as dropbox is limited for the purpose of data exchange and also with respect to data security, the project results are now stored on the ICOS data server (ICOS Carbon Portal) and the project documents in the UU SURFdrive (with a password protected link on the website).

The website is continuously updated and will be further expanded in the upcoming months. All participants are encouraged to use the website for information exchange. Visitors of the website will find general information about the project and its objectives, overviews of relevant scientific results and short report summaries, newsletters, blogs, and fact sheets.

Table 3.2: mean number of views / visitors of the website							
Recruitment period	After recruitment period						
Ø an month	a nu month						

	Ø nr. month	Ø nr. month
Total views	1505	692
Total visits	441	235
Unique visitors	255	129

The website has been visited more than 20.000 times, by almost 4000 individual users in the first reporting periods. In the first months of the project the website was mainly visited because of the vacancies advertised there (Fig. 3.1a, Table 3.2). In the meantime, we see a shift to the more general parts, with a clear increase of visits after posting news on the social media, publishing new blogs or announce events were MEMO<sup>2</sup> was presented (Fig. 3.1b, c).

The blogs published at the website are mainly written by the ESRs. Instead of writing a report about their secondments or measurement campaigns, they are encouraged to write blogs and addressing a broader public. This gives them the opportunity to present themselves and their projects, and also to practice their communication skills towards the public. An overview of blogs is given in Table 3.3 below.



#### 3.2.2 Social media

MEMO<sup>2</sup> is visible at several social media platforms. We are disseminating on platforms for professionals such as LinkedIn (<u>https://www.linkedin.com/groups/13506528/</u>, 23 members) and ResearchGate (<u>https://www.researchgate.net/project/MEMO2-MEthane-goes-MObile-MEsurements-and-Modelling</u>, 82 followers). The accounts were set up already 5 months before the official start of the project to introduce MEMO<sup>2</sup> and to use these platforms to advertise the open positions within MEMO<sup>2</sup>.

During the second year of the project, social media platforms such as Twitter (https://twitter.com/H2020 MEMO2, 32 followers). Facebook (MEMO<sup>2</sup> PhDs H2020. https://www.facebook.com/H2020MEMO2/) and Instagram (H2020 MEMO2, https://www.instagram.com/h2020 memo2/, 64 followers) were added to the portfolio, used as a general communication channel within a group (Facebook, Instagram) or towards the broader public (Twitter). The coordinator is responsible for the maintenance and update of the LinkedIn and ResearchGate accounts, the ESR group for the others. All participants are encouraged to contribute either by uploading posts themselves, or sending information or links to the coordinator and the ESRs.

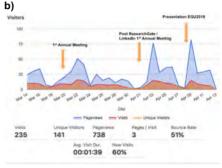
Fig. 3.2 shows a qualitative evaluation overview of the professional platforms LinkedIn and ResearchGate, the website and Twitter. Qualitative because platforms are organised differently, we cannot clearly distinguish between double visits and define the reached audience. It might also make a different if a platform is brought to the attention of the audience by notifications or tweets. All platforms are public, which is not the case for the Instagram and Facebook accounts and therefore the reason why no evaluation has been done on these platforms. Although the evaluation of the 4 platforms is not quantitatively, it gives a first impression about which platform might be more useful in reaching the envisaged target audience.

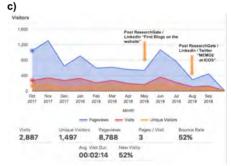
In a mean we have around 3300 views per months in total on these platforms. It can clearly be seen, that at this moment information about MEMO<sup>2</sup> activities is best visible on Twitter and the website. Within the next reporting periods we are planning to increase the visibility of MEMO<sup>2</sup>, not only on the other platforms but if possible also by producing a short video.

#### 3.2.3 Data repositories

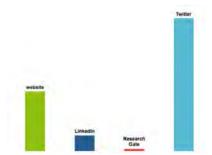
We are using SURFdrive and the ICOS Carbon Portal to ensure, that all MEMO<sup>2</sup> data are disseminated FAIR, which mean that they are easily findable and accessible and therefore usable in the end and beyond the project. SURFdrive, which is used for the storage of the project documents, is a cloud storage service for the Dutch research community, accessible via a link on the project website. Here the project documents are stored for internal use and those







**Fig. 3.1:** visit statistics MEMO<sup>2</sup> website; a) recruiting period, b) first posts and activities, c) launch of MEMO<sup>2</sup> blogs and start using Twitter, d) overview statistics during recruitment period (March – December 2017) and afterwards (January – December 2018)



**Fig. 3.2:** Qualitative overview of mean views of MEMO<sup>2</sup> activities on different platforms. The graphs show the mean views per month, starting at the launch of the platforms. The amount of activities is similar for LinkedIn and ResearchGate (n = 19), and some higher for Twitter (n = 28).



documents who are public are linked to the website. MEMO<sup>2</sup> data itself are stored at the ICOS Carbon Portal <u>https://fileshare.icos-cp.eu/login</u>. We set up a protocol which data have to be stored and how this should be structured. The protocol is based on D5.3, the Data Management, Dissemination and Exploitation Plan. Up to now both repositories are password-protected and only available for the MEMO<sup>2</sup> consortium. The MEMO<sup>2</sup> data will be public later on. The switch to use the server from ICOS and the UU will also ensure that both, data and website, and stored in long-term trusted repositories and that they will be accessible and available beyond the lifetime of the project.

## 3.3 General communication and dissemination activities

Next to the communication and dissemination activities mentioned above, we planned to produce newsletters and factsheets addressing the broader public. A first newsletter introducing the project and inform about campaign activities and network events was sent out to the consortium in December 2017. A second one was planned, but postponed due to all the other dissemination activities. Factsheets will be produced in the second half of the project, when first results are available.

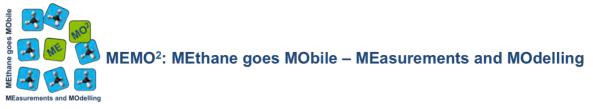
During conferences a MEMO<sup>2</sup> flyer is available and distributed e.g. to participants visiting poster sessions (<u>https://h2020-memo2.eu/general-dissemination-activities/</u>). The flyer informed about the project in general and will be updated towards including an overview of first results for the upcoming conferences next year.

The Netherlands Earth System Science Center (NESSC, <u>http://www.nessc.nl</u>) - coordinated by the Faculty of Geosciences at Utrecht University - is developing a new Methane Module for school educational purpose. MEMO<sup>2</sup> will contribute to this module by offering knowledge and presentation material. Depending on capacities, measurement demonstrations might be offered in the future.

Besides this, we strive to increase the general visibility of MEMO<sup>2</sup> e.g. by blogs, videos or during public events. The following table shows the general communication and dissemination activities during the first reporting periods.

Author / contact	Nature	Title	Link
Sylvia Walter	Blog	MEMO <sup>2</sup> goes marine!	https://h2020-memo2.eu/2018/06/14/memo2-goes-marine/
Sylvia Walter	Blog	Hidden secrets in the North Sea	https://www.nioz.nl/en/blog/hidden-secrets-in-the-north-sea-an- expedition-addressing-sea-level-rise-oxygen-loss-and-microbial- breakdown-of-methane
Sylvia Walter	Lecture	Methane lecture	Not yet, lecture in progress
Sara Defratyka	Blog	MEMO <sup>2</sup> at CoMet	https://h2020-memo2.eu/2018/06/25/memo2-at-comet/
Piotr Korben	Blog	My story from 3 weeks of measurements in Poland	https://h2020-memo2.eu/2018/08/16/1056/
Barbara Szenasi	Blog	My research visit at Wageningen University	https://h2020-memo2.eu/2018/09/05/barbary-szenasi-my- research-visit-at-wageningen-university/
Malika Menoud	Blog	Installation of a CF-IRMS and methane extraction system	https://h2020-memo2.eu/2018/09/21/installation-of-a-cf-irms-and- methane-extraction-system/
Anja Raznjevic	Blog	Modeling dispersion of methane	https://h2020-memo2.eu/2018/11/07/anja-raznjevic-modeling- dispersion-of-methane/
Sara Defratyka	Blog	AirCore: simple tool better than magical tricks	https://h2020-memo2.eu/2018/12/05/sara-defratyka-aircores/
Sylvia Walter	Blog	Workshop on Gaussian Plume and dispersion models	https://h2020-memo2.eu/2018/12/06/workshop-on-gaussian- plume-and-dispersion-models/
Sylvia Walter	Blog	How to identify sources of methane – workshop on isotopes	https://h2020-memo2.eu/2018/12/06/how-to-identify-sources-of- methane-workshop-on-isotopes/
Patryk Lakomiec	Blog	Swedish air goes Britain	https://h2020-memo2.eu/2018/12/06/patryk-lakomiec-swedish-air- goes-britain/
Thomas Röckmann	Public event	Methane emission contest: who wins?	https://www.uu.nl/en/research/sustainability/conference- 2019/impression-2018
Hossein Maazallahi	Interview	Wat is de rol van het MEMO <sup>2</sup> project? VLOG #9 Offshore Methaan Meetprogramma	https://www.youtube.com/watch?v=fcsXxEwIF6w&t=1s

Table 3.3: general MEMO<sup>2</sup> communication and dissemination activities



# 4. Management of MEMO<sup>2</sup>

## 4.1 General overview of the management

The management of MEMO<sup>2</sup> is organised based on the Grant Agreement and the Consortium Agreement (D5.1). The project structure and the management of it follow the approach as described in Chapter 3.2 of the GA, Part B, and the CA. The GA and the CA are available in digital form for all participants via the internal page of the website (re-directed to surf drive) or on request, they were also provided as a hardcopy to all participants of the Kickoff Meeting. These two documents provide, together with the deliverables D5.2 (Project Management Plan) and D5.3 (Data Management, Dissemination & Exploitation Plan) the basis for the management of MEMO<sup>2</sup>.

The project management - implemented by the UU as the coordinating beneficiary and organized within WP5 - provides scientific and administrative coordination of the project according to the EU requirements and facilitates communication within the consortium and also between the consortium and external stakeholders, e.g. the European Commission. The project management is responsible for regular reporting to both. In close collaboration with the consortium and also the maintenance of the website for internal and external exchange of information. The tasks as described for WP5 are:

- Task 5.1: administrative, contractual and financial project management
- Task 5.2: scientific project management
- Task 5.3: network meetings

All tasks started officially in project month 1 (March 2017) and are ongoing. The day-by-day task implementation is described in detail in the Project Management Plan (PM plan), submitted as deliverable D5.2 and the Data Management & Dissemination and Exploitation Plan (DMDE plan), submitted as deliverable D5.3. Both plans are living documents. During the annual meetings the consortium will decide, whether and which adjustments are necessary to ensure an effective and efficient project management over the course of the project and update the PM plan and the DMDE plan accordingly.

During the 1<sup>st</sup> Annual Meeting the consortium decided that no changes of the PM plan are necessary, and that the DMDE plan will be updated regarding some minor issues on data exchange and data quality, keywords and metadata. An updated version of the DMDE plan is in progress and will be send to the EC.

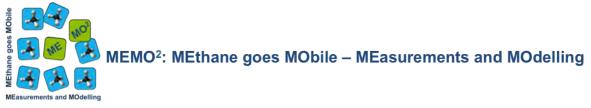
## 4.2 Recruitment

Within MEMO<sup>2</sup> 13 Early Stage Researcher were employed. Although MEMO<sup>2</sup> officially started in March 2017, the recruitment procedure already started end of November 2016 to ensure an efficient and timely recruitment. The recruitment was organised based on a decentralized strategy by the beneficiaries, but in close collaboration with the project management.

The aim was to recruit all ESRs ideally within the first six months of the project. The project is of high relevance and positions were advertised widely, but several positions attracted fewer applicants than expected and also the quality of applicants was beyond expectations. MEMO<sup>2</sup> is a highly complex project requiring candidates which are expected to have several characteristics, talents and skills, but a high number of applicants did not show the necessary experience and background for the project and those applications could not taken into account. Therefore, the recruitment required more time than proposed, and for some positions a second selection round was necessary. Finally, all ESRs started between project months 7 and 11 (September 2017 – January 2018).

During the second year of the project one ESR (ESR6, Badrudin Stanicki) decided to resign due to personal reasons. In agreement with the PO the ESR reduced his position during the last months to 0.6 fte, and the position has been re-opened. The position has been refilled by an ESR starting on 1 December 2018.

The initial recruitment is described in detail in deliverable D5.4 and the 1<sup>st</sup> Progress Report.



### 4.3 Consortium

The initial consortium has not changed during the first reporting period. All 9 beneficiaries and 13 partner organisations are actively involved and part of the consortium, and the project is running smoothly. In total about 60 researchers and staff members are involved in MEMO<sup>2</sup>.

Due to the commitment indicated in the proposal some partner organisations were more active in the first reporting period as others, e.g. by supervising ESRs, organising network events, giving advice or access to their properties. All partner organisations function as external mentors for the ESRs and are in contact to them.

Now in the second year we gained first results and intensified scientific collaborations within and outside the consortium. Up to now three additional partner organisations joined the network.

Environment and Climate Change Canada (ECCC) has been added as a new partner organization, as one PI from UVSQ has moved to it and would like to stay committed to the project. Additionally, GEOMAR was included as a partner organization, as oceans are significant contributors to the CH<sub>4</sub> budget but this topic was not implemented in the project. Just recently, the Technical University of Denmark joint the consortium. DTU Environment has a strong scientific record in development of measurement technologies for quantification of greenhouse gas emissions (in particular methane) covering various methods including surface flux chambers and eddy covariance. They are hosting two flux towers and are part of the ICOS / DK.

### **4.4 Meetings**

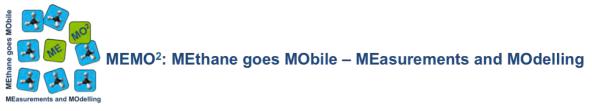
As MEMO<sup>2</sup> requires a high level of collaboration, regular meetings (for an overview see Table 4.1) were implemented right from the beginning and on all levels within the consortium. If possible, meeting dates were chosen by consensus (using the date finder "doodle") to ensure participation of as much partners as possible. In case this is not possible, the meeting organiser decides in agreement with the management. The meeting organisers were asked to prepare minutes from their meetings and provide them to the project management.

**I) Consortium Meetings:** Within the first reporting period one Consortium Meeting was held, the MEMO<sup>2</sup> Kickoff Meeting (23 and 24 March 2017), hosted by the coordinator (UU). All beneficiaries and 7 out of 13 partner organisations participated and showed a high level of commitment and interest in the project.

The first day focussed in the introduction of the beneficiaries and partner organizations, and the expertise and scientific profile of each participant. The leaders of the scientific WPs gave a general overview of the status of the WP and the involved partners, including first results and previous data that are relevant for the project. In addition to the scientific discussion all participants got a general overview of the main administrative tasks and responsibilities by the project manager and had the opportunity to discuss relevant questions with the financial administrative of the UU, Pieter Thijssen. As several non-academic partners join the consortium, they gave a brief introduction about their organisation and commitment in the project. A joint dinner closed the first day.

The second day focussed on planning concrete steps for the first year. The status of the recruitment procedure and the received applications was discussed in the consortium. Afterwards the training elements as defined in the GA were presented and discussed, also ideas about dissemination and exploitation strategies and the use of social media were exchanged. In addition, the consortium discussed the upcoming sampling campaigns and the organisation of them.

At the beginning of the second reporting period the 1<sup>st</sup> Annual Meeting of MEMO<sup>2</sup> was held (22 and 23 March 2018, <u>https://www.researchgate.net/project/MEMO2-MEthane-goes-MObile-MEsurements-and-MOdelling/update/5abc8ef24cde269658662413</u>), hosted by Empa in Dübendorf, Switzerland. 34 participants (all ESRs, all supervising PIs, 4 non-academic partner organisations (Shell, Picarro, Afalzorg, PGI), and 3 external scientific advisors (M. Heimann, A. Vermeulen, C. Sweeney)) were updated about the progress the project made within the first reporting period and discussed the planning of the second period. Besides this the ESRs discussed their CDPs face-to-face with supervisors and



co-supervisors, and if present, with their non-academic mentors. Before the official meeting, the local organisers offered the opportunity to visit the research station at Jungfraujoch. The station for e.g. meteorology, glaciology, spectrometry, or geophysics is one of the highest European measurement stations and frequently used by researchers from all over the world. The visit was highly appreciated by the ESRs.

At the end of the meeting the external scientific advisors evaluated the project and supported the consortium in its future planning. An overview of the evaluation is given in chapter 4.1.6.

To ensure that all ESRs are informed about their rights and obligations, a hard-copy of the notes for MSCA-ITN fellows were handed out to them.

The second annual meeting is associated to the 2<sup>nd</sup> MEMO<sup>2</sup> School (CH<sub>4</sub> and the society) and the Midterm Meeting, all scheduled in the week 18-22 February 2019.

**II) 3-monthly tele-conferences:** The Network Supervisory Board (NSB) held five tele-conferences, starting three months after the Kickoff Meeting. The NSB consists of the coordinators (scientific and administrative), a representative of each beneficiary and partner organisation, and the ESR representatives. The first tele-conference was held by using the telephone-based platform Powwownow. As this platform was not suitable due to technical problems for some partners (security reasons) and also its high costs, the consortium decided to switch to the internet-based platform WebEx. This platform has been proven to be suitable for all participants and will be used in the future. Dates were picked by using the date finder "doodle", with at least 5 suggestions regarding day and time.

During the tele-conferences the WP leader gave brief overviews about the status of their work packages and the involved partners, and the participants discussed the main relevant issues for the upcoming months, e.g. status of recruitment, planning of measurement campaigns, the organisation of the MEMO<sup>2</sup> school or upcoming administrative tasks as deliverables and their contributions. Minutes are available on the SurfDrive for all MEMO<sup>2</sup> participants.

**III) ESR skype meetings:** The ESRs were right from the beginning encouraged to hold regular Skype meetings. This is working well, and up to now they held 10 meetings. Independently from the PIs or the coordinator, they set up an ESR council, with two chairmen for a period of 5 months, responsible for organising the ESR meetings (envisaged one meeting per month, with at least 9 ESRs present). The chairmen are also responsible for making the minutes and send them to the management. During the meetings, which are announced to the management, the ESRs exchange information, and discuss relevant issues such as campaigns or data exchange. In case input or information is requested from the ESRs, e.g. for planning network events or reporting, this is communicated by the management to the respective ESR chairmen and discussed within the ESR meetings. The ESR group vice versa can request relevant information they would like to distribute or discuss from the management.

**IV) WP meetings:** All WP leaders were asked to organise regular WP meetings with the PIs involved in their WP. Initially this was planned bi-monthly. As the WPs internally have a close email contact, e.g. due to organising joint measurement campaigns, circulation of inter-comparison cylinders, or data exchange, and also in combination with the regular 3-monthly tele-conferences, the need for bi-monthly WP meetings is not given and the frequency is more on request.

Meeting	Meeting Date / Location		Work Package	Invited participants
Kickoff Meeting	23–24 March 2017, Utrecht, The Netherlands	UU	All	All project participants
1 <sup>st</sup> Annual Meeting	21 – 22 March 2018, Dübendorf, Switzerland	Empa	All	All project participants
2 <sup>nd</sup> Annual Meeting	21 – 22 February 2019, Paris, France	UVSQ	All	All project participants, Project Officer, external reviewer(s)
Tele-Conferences	15 June 2017 15 September 2017 13 December 2017 1 June 2018 12 October 2018	UU	All	Representatives of all beneficiaries and partner organisations, ESR representatives

Table 4.1: overview of MEMO<sup>2</sup> meetings during the reporting period 1 March 2017 - 28 February 2019



WP Tele-Conferences	On request of WP leader	Respective WP leader	Respective WP	WP participants (Pls + ESRs)
ESR Tele-Conferences	14 November 2017	ESRs	All	ESRs
	14 December 2017			
	13 March 2018			
	19 April 2018			
	23 May 2018			
	27 June 2018			
	16 July 2018			
	21 August 2018			
	22 November 2018			
	11 December 2018			

### 4.5 Communication infrastructure

The communication infrastructure has been described in detail in deliverable D5.7. According to the spatial distribution of the consortium, the day-by-day communication within the consortium is assured mainly remote by email. The chosen communication channels, structure and frequency worked well, all participants are responsive and engaged, and the management received requested information without problems. Within the consortium all communication channels are available for any participant, the communication lines are short and direct with dedicated responsibilities (see D5.2 and D5.3 for more details) but no formal restrictions. The direct communication with the Project Officer as a representative of the EU as funding agency is restricted to the coordinator.

#### 4.5.1 Email, telephone

The main communication channel according to the spatial distribution of the consortium is email. A dedicated project email has been created (<u>management@h2020-memo2.eu</u>) as contact for external requests, setup via the Dutch company GetHost. Individual exchange of information uses the institutional email addresses of the participants. Email is also the preferred channel for official project communication to ensure traceability of information and decisions. Besides emailing, phone calls are used as communication channel. Both channels work well.

#### 4.5.2 Web-based board meetings and tele-conferences

Direct and regular exchange between participating groups and boards increases collaboration and reduce the risk of failing of the project. High-frequent face-to-face meetings are not manageable due to geographically reasons. They are costly and also causing unnecessary environmental pollution, thus participants are invited to participate in regular remote board meetings. The meetings are planned by either using *Skype* as a web-based platform for meetings with only a few participants or WebEx for consortium meetings. The platform *powwownow* (<u>https://www.powwownow.co.uk</u>), a telephone-based platform has been proven as not suitable.

#### 4.5.3 Face-to-face meetings

For the consortium the mandatory face-to-face moments are the annual meetings (see Table 4.1). The meetings are organized by dedicated beneficiaries as indicated in the Grant Agreement, and communicated via email and the project website. At the annual meetings, which are mandatory for the beneficiaries and strongly encouraged for the partner organisations, the ESRs present the progress of their work, meet with (co)supervisors and mentors and update their CDPs, and the consortium discuss project related scientific and administrative issues including planning of the future project periods.

#### 4.5.4 Participant Portal

The Participant Portal (<u>https://ec.europa.eu/research/participants/portal/desktop/en/home.html</u>) is the web portal of DG Research & Innovation and the entry point for electronic administration of EU-funded projects. The Participant Portal hosts the services for managing projects throughout their lifecycle. The coordinator uses the Participant Portal for general project managing.

The beneficiaries have to use the EU Participant Portal for individual beneficiary related reporting or documentation issues, e.g. financial reporting or submission of researcher declarations.



### 4.6 Risk assessment and faced difficulties

A detailed overview of possible risks is listed and described in Chapter 7 of the Project Management Plan (submitted as deliverable D5.2).

In the first two years of MEMO<sup>2</sup> we faced some small issues, with no significant impact on the project.

Risk 1: The ESR recruitment took longer than expected, but was still on schedule. In project month 9 all ESRs were selected. However, the initial planning of the secondments started already from month 6 of the project, as this was the envisaged begin date of the ESRs in the proposal. Therefore, the secondment schedule will be structurally postponed. During the second year, one ESR resigned due to personal reasons. This could have been an issue for the progress of the individual ESR project and also regarding the general management budget in the case that no suitable candidate will be found in time. But as this ESR was recruited quite early and the position has been re-opened as soon as possible, a new candidate was found quite quickly. So, the ESR project will be continued with no significant impacts on the project.

Risk 11: One of the PIs (Felix Vogel, UVSQ) left the consortium to start at ECCC (Environment and Climate Change Canada). The ESR supervision was taken over by the former colleagues, and besides this ECCC was added as a new partner organisation to the consortium. Felix Vogel will still be involved in the supervision, and all ESRs of the consortium have the opportunity to take additional secondments to ECCC.

Some difficulties were faced on organisational administrative level:

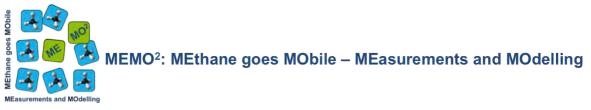
- At RHUL one of the ESRs (originally from Turkey) needed a Schengen Visa additional to the UK work visa, which caused extra travel and time. In the UK the medical support is more difficult due to different laws (between US and UK) regarding medication, resulting in long waiting time to get proper medication support.
- The recruitment procedure at AGH includes a PhD examination by the respective department. Without passing it officially, a student cannot be hired as a PhD or apply for a visa. As the examination is only once a year, this was a critical issue in the recruitment procedure. Besides the examination the ESR hired at AGH (originally from Serbia) faced the problem that all official documents including the working contract are only in Polish. Her supervisor translated all relevant contracts and instructions, as also most of the administrative staff only speaks polish. Also, the application for a visa caused problems. Here the National Contact Points (from Poland and also Serbia) were contacted and could help to solve the situation at least short-term. Up to now no visa for the whole project lifetime is available.
- The students at UVSQ faced similar problems regarding language. The contract was also only offered in French, and some general information given in English. Administrative documentation needs to be filled in only in French. Here also the supervisors helped with translations. However, it is desirable, that all official and legal documents which have to be signed by the ESRs would be at least in English to ensure a minimum of necessary understanding what have been signed.

On a more general level, another difficulty is to ensure the engagement of the partner organisations as ITN-ETNs do not have a dedicated budget to cover e.g. travel and accommodation costs at least to the annual meetings. Especially for partner organisation without a reserved budget - next to in-kind contributions such as staff hours - this is a hinder to participate in projects such as MEMO<sup>2</sup> and to actively participate to face-to-face discussions. For e.g. the 1<sup>st</sup> Annual Meeting the beneficiaries covered the catering costs for the partner organisations, but still travel and accommodation was on their own expense.

Further no risks have been monitored and the project is running smoothly.

# 5. Outlook 3<sup>rd</sup> Reporting Period

The next reporting period will be discussed during the 2<sup>nd</sup> Annual Meeting and based on the outcome and the evaluation during the Midterm Meeting this chapter will be updated for the 1<sup>st</sup> Periodic Report.



# 6. Individual ESR reports

# 6.1 ESR1 - Monitoring the methane emissions from different sources in Germany

#### ESR1

Monitoring the methane emissions from different sources in Germany

ESR	Piotr Korben, <u>pkorben@iup.uni-heidelberg.de</u>
Supervisor	Martina Schmidt, martina.schmidt@iup.uni-heidelberg.de
Co-supervisor	Thomas Roeckmann, <u>t.roeckmann@uu.nl</u>
Non-Academic mentor	Bill Hirst, <u>bill.hirst@shell.com</u>
Official start – end date	1.1.2018 – 31.12.2020

#### 6.1.1. Scientific progress

#### 6.1.1.1 Project introduction and objectives

Methane (CH<sub>4</sub>) is one of the greenhouse gases (GHG) like CO<sub>2</sub> or water vapor, but with a larger greenhouse gas potential. Methane has natural but also anthropogenic sources. The main natural sources are wetlands and wild ruminants. Anthropogenic sources are agriculture, landfills, biomass burning, gas fields, oilfields, coal mines and coal burning. In Germany the major methane sources are ruminants, waste treatment and transport and storage of natural gas. In this project we monitor different methane sources with a mobile CRDS analyser. This instrument is installed in a vehicle and during regular measurements campaigns we measure methane mole fraction and the isotopic composition (<sup>13</sup>CH<sub>4</sub>) to determine the temporal and spatial variability of emissions. To obtain emissions from individual methane sources we will use Gaussian plume models and analyse our measurements data with them. Results will be compared to regional emission inventories. This project is focused on EU emission from natural gas transport, but we would like also to measure different sources around Heidelberg to get more information about methane in Germany.

#### 6.1.1.2 Project results

#### 6.1.1.2.1 First year

Piotr Korben started his PhD contract at University Heidelberg in Januarv 2018. During the first two months he participated in different training activities. The training consists of use of mobile CRDS analyser (Fig. 6.1.1). calibration, data analysis and participate to two measurements campaigns region Heidelberg in



Fig. 6.1.1: Mobile CRDS analyser in vehicle.



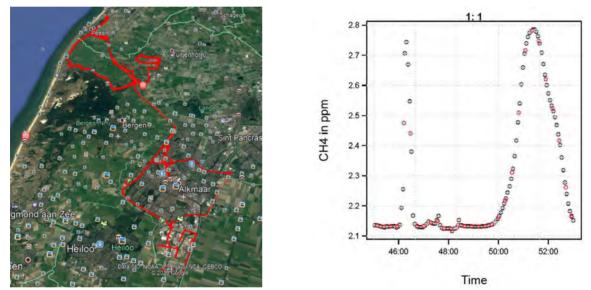
**Fig. 6.1.2:** Mobile CH<sub>4</sub> measurements in Heidelberg city, at a farm in Weinheim, and a compressor station at Hähnlein

(Southwest Germany). The first visited CH<sub>4</sub> sources were gas compressor station and biogas plant. The use of CRDS analyser and all equipment were trained directly on campaigns. One important aspect of the training period was to understand the method for data evaluation. This contains the application of calibration and different corrections factors, which consider cross sensitivity for <sup>13</sup>CH<sub>4</sub> measurements. All programs are written in R by students in Heidelberg and P. Korben learned to use and to improve these programs.



The second measurement campaign were performed at the same biogas plant and a landfill. In addition, a survey in the city of Heidelberg to detect CH<sub>4</sub> leakages from natural gas has been undertaken. Here we detected a gas leak, which shows a concentration enhancement of 40 ppm (see Fig. 6.1.2).

From 5<sup>th</sup> to 16<sup>th</sup> of February Piotr Korben participated at the1<sup>st</sup> MEMO<sup>2</sup> School and the associated first measurement campaign North of Alkmaar. The Heidelberg team participated with the institute van equipped with a CRDS analyser and mobile kit. Measurements were performed close to a biogas plant, several farms, landfill and a gas compressor station. Fig. 6.1.3 shows the CH<sub>4</sub> mole fraction measured in Alkmaar region. On the right panel an example of sampling of the emission plume in a 25m storage tube (AirCore) and the reanalysis is presented.



**Fig. 6.1.3:** Map of Alkmaar region including the measured CH<sub>4</sub> mole fraction (left panel) and direct vs. sampled plume (storage tube) measurement from a Farm near Petten.

Analysis of  ${}^{13}CH_4$  source signature in the region of Alkmaar have been carried out during the MEMO<sup>2</sup> campaign in the region around Alkmaar. The results are presented in Table 6.1.1.

#### 6.1.1.2.2 Second year

In the second year of project we focused on measurement campaigns around Heidelberg and a three-week campaign to coal mines in Upper Silesia (Poland).

**Table 6.1.1:** <sup>13</sup>CH<sub>4</sub> source signature of different CH4 emitters, measured during the first campaign in the region of Alkmaar.

Source	δ value (‰)	σ (±)	Nr of AirCores
lefjeshoeve Farm	-62.4	4.4	1
Farm (near to release test)	-74.4	4.5	3
Farm (near to release test)	-61.5	4.2	1
Landfill Alkmaar	-54	2.7	1
Gas compressor station Alkmaar	-28,7	3	1
Bag nr1 (release test)	-47.9	0.8	2
Bag nr 2 (natural gas)	-33.3	0.8	2

Piotr Korben planned to make each month 1-2 campaigns to visit different  $CH_4$  sources. Measurements have been performed at several farms, landfills, gas compressor stations and biogas plants. Results from these campaigns are presented in table below. These campaigns were focused on  $^{13}CH_4$  source signature with the goal to account for possible seasonal or temporal variation.

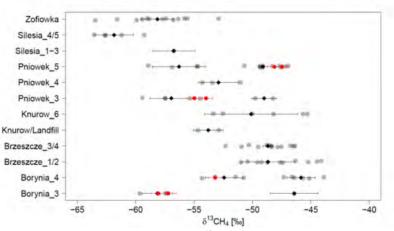
Prior to each campaign all equipment was prepared and weather conditions were checked. Especially wind direction and wind speed are important information for the campaign planning as due to available roads the CH<sub>4</sub> emitters can only be measured downwind under specific conditions.

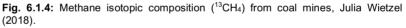


SOURCE	10.01 δ <sup>13</sup> CH₄ [‰]	26.01 δ <sup>13</sup> CH₄ [‰]	26-27.03 δ <sup>13</sup> CH₄ [‰]	26.04 δ <sup>13</sup> CH₄ [‰]	4-6.07 δ <sup>13</sup> CH₄ [‰]	24.08 δ <sup>13</sup> CH₄ [‰]	25.10 δ <sup>13</sup> CH₄ [‰]
Farm Weiheim	- 49,73 ± 1,1	-	-	- 49,35 ± 0,7	- 55,26 ± 0,9	- 51,48 ± 1,2	
Farm Ladenburg	-	-	-	-	- 49,00 ± 3,3	-	
Gas station Hahnlein	- 43,98 ± 0,7	-	- 39,03 ± 2,2	-	- 47,60 ± 3,9	-	
Landfill Sinsheim	-	-	-	- 56,56 ± 1,6	-	- 59,53 ± 2,0	
City of Heidelberg	-	- 45,71 ± 1,6	- 41,98 ± 0,8	-	-	-	
Biogas plant	- 61,98 ± 1,1	- 57,12 ± 2,0	- 61,52 ± 0,8	-	-	-61,45 ± 1,7	
Gas station Scheidt	-	-	- 41,26 ± 0,5	-	-	-	

#### Table 6.1.2: <sup>13</sup>CH<sub>4</sub> source signature measured during 7 campaigns in the Heidelberg region

The participation to the CoMet campaign in Upper Silesia in Poland was the second large achievement in the reporting period. Together with three other MEMO<sup>2</sup> partners (AGH, UU, LSCE)) mobile CH<sub>4</sub> measurements from vehicles were carried out in addition to aircrafts measurements from DLR München. This campaign took place from 23.05 to 10.06.2018. Seven coal mines and 12 mine shafts (include 1 in Czech Republic) were visited and measured with mobile CRDS analyzer and to cover Upper Silesia. Results





from this campaign were evaluated in the bachelor thesis of Julia Wietzel who participated in this campaign as support. Results of the derived <sup>13</sup>CH<sub>4</sub> source signature are presented in Fig. 6.1.4. The range of the <sup>13</sup>CH<sub>4</sub> isotopic source signature is larger than expected. Grey points are measurement points (from AirCores), black points are mean values and red points are values from bag samples.

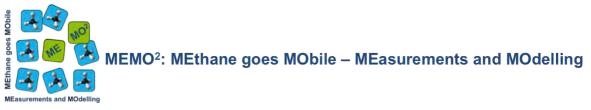
#### 6.1.1.3 Future plans and expected results

One of the next steps will be to calculate methane emission factor from different sources around Heidelberg. During the next few months Piotr Korben will focus in improving modelling skills using different Gaussian and Dispersion models (Polyphemus, GRALL, WindTrax). These models will be applied to data from campaigns to transfer the CH<sub>4</sub> measurements to emission rates.

Regular mobile measurements campaigns in the Heidelberg region will be continued. We plan to visit representative CH<sub>4</sub> emitters (dairy farms, biogas plant, compressor stations and landfills, city) several times a year to monitor CH<sub>4</sub> concentration and <sup>13</sup>CH<sub>4</sub> isotope ratios during different seasons. Every month we will carry out 1 - 2 measurements campaign around Heidelberg. With this frequency we will get more accuracy results for seasonal and temporal variations in <sup>13</sup>CH<sub>4</sub> isotopic composition. Piotr Korben will participate in campaigns during his secondment in Paris.

#### 6.1.1.4 Collaborations (internal / external)

We cooperated with others MEMO<sup>2</sup> research groups and PhDs on team campaigns (Schoorl) and meetings (Dübendorf)/workshops (London, Heidelberg). New collaborations were started in May during CoMet campaign (3 weeks) on Upper Silesia in Poland. We worked together with Andreas Fix group



(DLR) and another groups from Europe (Gronigen, Cracow, Warsaw, Paris, Utrecht). We are still in exchange with DLR group to analyze our results and compare them.

#### 6.1.1.5 Risks and difficulties

As the ESR1 started later than it was planned, we are slightly behind our work plan. Therefore, also the secondments are delayed. We changed the order of the secondments, and the first secondment was in Poland in May 2018. This secondment took place during MEMO<sup>2</sup> / CoMet campaign where Piotr Korben helped our partner AGH with organisation and logistic for the mobile surface measurements. The second secondment will be in month 24 in Paris, instead of month 19.

#### 6.1.2 Deliverables

ESR1 is involved in the following deliverables: D1.4 / D1.5 / D2.2 / D2.3. There is no contribution planned to D1.1, this is a typing error in the description of work.

D1.4 - Improved emission factors for different source categories from mobile measurements (month 42)

Every month I carried out one measurement campaign close to regional methane sources like dairy farms, gas compressor stations, landfills, biogas plant, city of Heidelberg and it depends on weather condition where is next campaign. Until now mobile measurement, campaigns were focused more on isotopic measurements. Next step will be to work with plume or dispersion models with the goal to transfer the concentration to emission flux.

D.1.5 - Report on harmonized methods for mobile CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> measurements (month 18)

During the MEMO training school in Schoorl and campaign, we inspected the different instrumental setups. At ECN a comparison of ambient air with all available analysers was performed. Further discussion on the first draft was scheduled during the first annual meeting. I participate on the discussion and writing of the report.

D.2.2 - Improved isotopic source signature of local and regional CH4 emissions (month 36)

Analysis with in-situ <sup>13</sup>CH<sub>4</sub> analyser started in January 2018 in the Heidelberg region and will be continued during the project duration. As was mentioned before, around Heidelberg is possible to measure different isotopic source signature.

**D.2.3** - Publications on the use of isotopes for CH<sub>4</sub> source attribution in urban / industrial regions (month 36)

Data collection started and it will be continued for next months. Data will be analysed and used for writing publications in cooperation with other students or groups.

### 6.1.3 Training and network activities

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
MEMO <sup>2</sup> TrainingSchool	5.02.2018 – 16.02.2018	Schoorl, Netherlands (UU)	Knowledge about chemistry of atmosphere,	6	Presenting a poster, and oral presentation, participating in measurements campaign	
I MEMO <sup>2</sup> Annual Meeting	22.03.2018 - 23.03.2018	Duebendorf, Switzerland (EMPA)	Progress of work from other PhD and other work package		Oral Presentation	
lsotope Workshop	17.09.2018 – 19.09.2018	Egham, UK (RHUL)	Improve knowledge about isotopes and measurements techniques		Oral presentation,participation to lecture and training	
Plume Workshop	9.10.2018 – 10.10.2018	Heidelberg, Germany (UHEI)	Getting knowledge about different models and how use them		Participation to lecture and exercises	
Institute seminar	11.01.2018 - now	Heidelberg, Germany (UHEI)	Lectures about environmental physics, knowledge about other topics from science / other groups	2 (SWS)	participating	

#### 6.1.3.1 General training events



#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

delberg, Meeting where problems and 2 Participa many progress of each member is (SWS) presenta IEI) discussed	
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#### 6.1.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
AGH	22.05.18 – 10.06.18, 31.10.18 – 9.11.18	Cracow, Poland	AGH UST in Krakow	Participation in CoMet campaign with other students and different research group on coal mines area on Silesia, Poland <i>I</i> .	Improving measurements campaign organization skills, measurements isotopic composition of methane from coal mines, cooperation with other groups and other instruments, creating scientific network	Work on data is still in process. Coal mines, landfill and dairy farm were visited during this campaign. Methane isotopic composition ( $\delta^{13}$ C) for coal mines showed values from – 64 to – 43 ‰. It is a big range, and during campaign days methane could come from different depth. Cooperation with aircrafts helped to construct vertical profile for methane on Silesia area.
LSCE	02.2019 – 03.2019	Paris, France	LSCE	Measurements campaign on urban area and work with Polyphemus model	Getting knowledge about using Polyphemus model to results from measurement campaigns	

### 6.1.3.3 Conferences

The ESR did not participated in scientific conferences yet.

#### 6.1.3.4 Measurement / sampling campaigns

Campa ign	Date (start – end, planned (when))	Location	Ho st	Description of work	Scientific objective	Samples (nature / number of AirCores)	Results and future plans
I	10.01.2018	Heidelberg and surrounding area (Weinheim, Hähnlein, Kirchheim)	UH El	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farm, gas compressor station, biogas plant / 10	Values of isotopic composition of methane from visited sources, calculate emission factor
Ι	26.10.2018	Heidelberg and surrounding area (Kirchheim, Sandhausen)	UH EI	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Biogas plant, gas compressor station, city / 12	Values of isotopic composition of methane from visited sources, calculate emission factor
ш	9.02.2018 – 12.02.2018	Schoorl, Netherlands	UU	Measurements campaign and release experiment in Schoorl and surrounding area	Learning about campaign organization, measurements different sources in Netherlands	Landfill, dairy farms, gas compressor station, city / 17	Values of isotopic composition of methane from visited sources, cooperation with other groups
IV	26.03.2018 - 27.03.2018	Heidelberg and surrounding area (Hähnlein, Kirchheim, Scheidt)	UH El	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Gas compressor stations, biogas plant / 15	Values of isotopic composition of methane from visited sources, calculate emission factor
V	26.04.2018	Heidelberg and surrounding	UH El	Mobile measurements	Measurements isotopic	Dairy farms, landfill / 13	Values of isotopic composition of

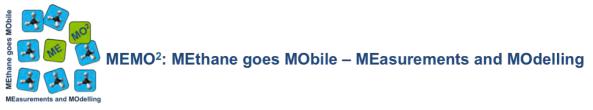


		area (Weinheim, Ladenburg, Sinsheim)		using CRDS Analyzer Picarro G2201-i	composition of methane from different sources to check seasonal variations		methane from visited sources, calculate emission factor
VI	22.05.2018 – 10.06.2018	Upper Silesia, coal mines area	AG H	Mobile measurements using CRDS Analyzer Picarro G2201-I / CoMet campaign	Methane and isotopic composition of methane measurements from coal mines	Coal mines, landfill, dairy farm / 75	Values of isotopic composition of methane from coal mines on Silesia area, vertical profile of methane concentration in cooperation with aicrafts
VII	4.07.2018 – 6.07.2018	Heidelberg and surrounding area (Weinheim, Ladenburg, Ludwigshafen, Hähnlein)	UH EI	Mobile measurements using CRDS Analyzer Picarro G2201-I and release experiment	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farms, landfill, gas compressor station / 10	Values of isotopic composition of methane from visited sources, calculate emission factor
VIII	24.08.2018	Heidelberg and surrounding area (Weinheim, Sinsheim, Kirchheim)	UH EI	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farm, landfill / 17	Values of isotopic composition of methane from visited sources, calculate emission factor
IX	25.10.2018	Heidelberg and surrounding area (Weinheim, Ladenburg, Hähnlein)	UH EI	Mobile measurements using CRDS Analyzer Picarro G2201-i	Measurements isotopic composition of methane from different sources to check seasonal variations	Dairy farms, gas compressor station / 18	Values of isotopic composition of methane from visited sources, calculate emission factor

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.1.4 Dissemination activities

So far no scientific publications or other dissemination activities from the ESR.



# 6.2 ESR2 - Quantifying CH<sub>4</sub> emissions using measurements on cars and UAVs in the Netherlands

#### ESR2

Quantifying  $\text{CH}_4$  emissions using measurements on cars and UAVs in the Netherlands

ESR	Katarina Vinkovic ( <u>k.vinkovic@rug.nl</u> )
Supervisor	Prof.dr. Huilin Chen (huilin.chen@rug.nl)
Co-supervisor	Prof.dr. Wouter Peters (wouter.peters@wur.nl)
Non-academic mentor	Dr. Arjan Hensen ( <u>arjan.hensen@tno.nl</u> )
Official start-end date	01.10.2017 – 30.09.2021

#### 6.2.1. Scientific progress

#### 6.2.1.1 Project introduction and objectives

The aim of the ESR2 project is to quantify  $CH_4$  emissions using atmospheric concentration and isotopic composition measurements, with a focus on the agriculture  $CH_4$  emissions (cattle and manure) that account for ~ 67% of the total emissions in the Netherlands in 2015 (Coenen et al., 2017). Spatial and temporal variations of  $CH_4$  concentrations near major sources will be obtained on two different mobile platforms, a vehicle (car/van) and unmanned aerial vehicle (UAV). Additional trace gas measurements (e.g.  $NH_3$ , CO,  $^{13}CH_4$  and  $CDH_3$ ) on the vehicle enable us to identify the source type, whereas 3D mapping of the  $CH_4$  plumes using an active UAV AirCore system will allow an accurate estimate of the source strength when combined with a Gaussian plume model as well as large eddy model simulations in collaboration with other researchers within the same project.

#### 6.2.1.2 Project results

#### 6.2.1.2.1 First year

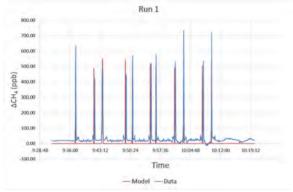
During the first year, ESR2 participated in the 1st MEMO2 School, which was held in Schoorl, the Netherlands, 5-16 February 2018. The 1st MEMO<sup>2</sup> School was a two-week school on methane practical includina courses. exercises, field campaigns, and data analysis. On the first day of our field campaign (9<sup>th</sup> Feb., 2018) CH<sub>4</sub> was measured from the lefjeshoeve farm, close to Petten (Fig. 6.2.1). In this particular case, the ECN's QCL Aerodyne was used for collecting the data, as well for measuring the concentration of trace gases.

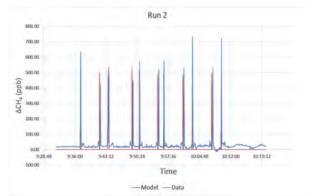


Fig. 6.2.1: The measured CH<sub>4</sub> spike near the lefjeshoeve farm.

The next step was to evaluate the acquired observations using ECN's Gaussian plume model. The model was run twice with different initial parameters, i.e. in the first case, the wind speed was set to 5 m/s and the wind direction to  $160^{\circ}$  (Fig. 6.2.2), while in the second case 4 m/s and  $170^{\circ}$  (Fig. 6.2.3), respectively. From meteorological data is known that on the 9th February wind speed was around 5 m/s, to observe the uncertainties, the model was run twice with wind speeds 4 m/s and 5 m/s.







**Fig. 6.2.2:** The first case, with wind speed 5 m/s and wind direction 160°. The red line presents simulation of Gaussian model, while blue real measurements.

**Fig. 6.2.3:** The second case, with wind speed 4 m/s and wind direction 170°. The red line presents simulation of Gaussian model, while blue real measurements.

To quantify the emissions, each source from each peak needs to be calculated separately. In both cases, emissions from first three peaks were calculated and the compared (Table 6.2.1). The lefjeshoeve farm has around 190 cows, of which 130 dairy cows and 60 young cows.

Table 6.2.1: CH<sub>4</sub> emissions from the lefjeshoeve farm.

	RUN 1		RUN 2				
Source (g/sec)	Emission (g/day)	Emission (kg/cow/year) <sup>1</sup>	Source (g/sec)	Emission (g/day)	Emission (kg/cow/year)		
1.62	139 968	319.30	1.67	144 288	329.16		
1.03	88 992	203.01	1.13	97 632	222.72		
1.34	115 776	264.11	1.40	120 960	275.94		

Calculated CH<sub>4</sub> emissions from cows are in the range 203.01 - 329.16 kg/cow/year (Table 6.2.1) implying a large discrepancy compared to the values proposed in the literature, 50 - 130 kg/cow/year (Johnson and Johnson, 1995). This leaves us space for further studies in the future.

Furthermore, for the PhD candidate, it was mandatory to write an Introductory essay within the first six months of their PhD programme. The Introductory essay gives the student's vision of the research project and detailed description of the research plan. It usually consists of a literature review, a description of the research questions to be addressed and a detailed project plan, including a schedule/timeline.

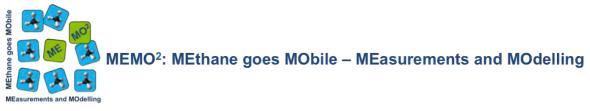
#### 6.2.1.2.2 Second year

Based on the instrumental setup that ESR 2 uses for her research, the progress and results of the second year of the project can be divided into mobile van measurements and UAV measurements.

#### 6.2.1.2.2.1 Mobile van measurements

During the first part of her secondment (August – September 2018), ESR2 continued to analyse the mobile van dairy cow farm (~18km North – West of Alkmaar, the Netherlands) measurements from the  $1^{st}$  MEMO<sup>2</sup> School (February 2018). The time lag of methane mole fraction measurements due to the travelling of air samples through the inlet tube was corrected. In Fig. 6.2.4b different colours indicate the magnitude of CH<sub>4</sub> mole fractions.

<sup>&</sup>lt;sup>1</sup> An equivalent of 160 cows was used in the conversion from g/day to kg/cow/year, i.e. 60 young cows emit the same amount of CH<sub>4</sub> as 30 dairy cows.



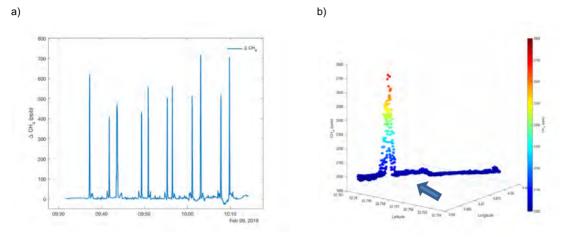


Fig. 6.2.4: CH<sub>4</sub> plume measurements as a function of time (a) and location (b).

These measurements were compared with a series of Gaussian plume model calculations to determine the  $CH_4$  emission rates from the dairy cow farm. The emission level is obtained in three steps (Fig. 6.2.5). **Step 1:** Run the model with different settings and a source of 1 g  $CH_4$ /sec. **Step 2:** Integrate both measured and modelled plumes. **Step 3:** Scale each model run result with the ratio of the modelled to the measured integration. **Final:** Decide on which modelled plume is in the best agreement with the measurements, and use other runs to get an uncertainty range.

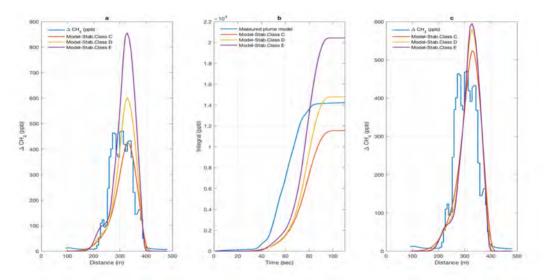


Fig. 6.2.5: Sensitivity test for Pasquill classes C, D, and E. (a) Step 1 (b) Step 2 (c) Step 3

Based on the procedure mentioned above, for this particular campaign, the model run with Pasquill class D agrees the best with the measurements. The other two runs (stability E and C) are used to get the uncertainty range.

Emission rates presented in Table 6.2.2 are representative only for that particular day, with the uncertainty not larger than  $\pm$  20 % in the meteorological data. It is quite important to have high-quality meteorological data instead continuing with further improvement of a methane sensor. Furthermore, it is not unlikely that the variability in the source strength, due to the change of activity over the year, is the leading uncertainty in the comparison measured estimate and inventory estimate.



Pasquill Class	E (min)	D	C (max)
Source (g/sec)*	$0.80\pm0.08$	$1.11 \pm 0.12$	$1.43 \pm 0.15$
Source (kgCH₄/day/(adult cow+manure))*	$0.46 \pm 0.05$	$0.63 \pm 0.07$	$0.82\pm0.09$

\* The non-enteric emissions are not subtracted.

ESR2 started to develop a quick analysis tool for large datasets with multiple farms during the second part of her secondment (October 2018 – January 2019). First step is to develop a methodology in an excel environment, which later will be transformed into a R script. And if possible, in the end to link the quick analysis tool with data acquisition system for online source assessment in the field.

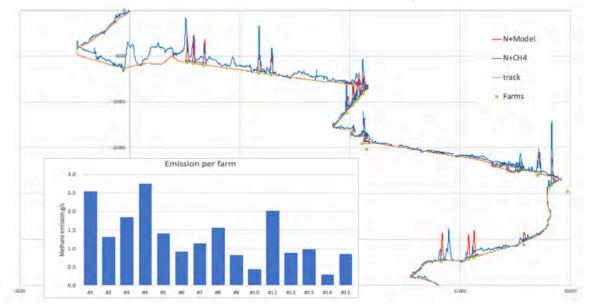
The analysis is performed on the mobile van measurements, that were obtained on 21<sup>st</sup> August 2018 in South - East part of the North Holland province, the Netherlands. The area is almost completely agricultural, it consists of dairy farms and arable land (Fig.6.2.6).

acquired The observations were evaluated using the ECN Gaussian plume model. The model was run once for each farm with one source, the wind speed was set to 3 m/s, the wind direction to 180° (Fig. 6.2.7) and a source of 1 g CH4/sec. The second model run will include multiple sources per each farm, and the comparison with the results from the first step. The final goal is to calculate emission rate per farm per year, and then compare with an emission inventory methodology.

Since ESR2 recently started to develop the quick analysis tool, the next report will provide more information.



**Fig. 6.2.6:** Google Maps image of 20 dairy farms, SE part of the North Holland province, the Netherlands. The red line presents the  $CH_4$  concentration, while the blue line gives the N<sub>2</sub>O concentration.



**Fig. 6.2.7:** First model run, with wind speed 3m/s and wind direction 180o. The red line presents simulation of the Gaussian model, while the blue one gives real measurements. The block diagram shows a preliminary emission rate per farm on 21<sup>st</sup> August 2018, SE part of the North Holland province, the Netherlands. The non-enteric emissions are not subtracted.

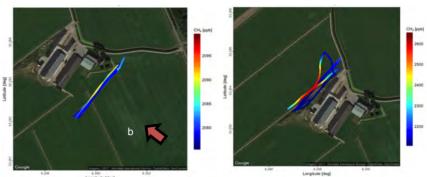


#### 6.2.1.2.2.2 UAV measurements

Furthermore, the ESR2 is working continuously on UAV measurements as well. The CH<sub>4</sub> mole fraction measurements from the UAV AirCore (8 – 15 minutes) were used to determine the CH<sub>4</sub> enhancement of the downwind against the upwind from a dairy cow farm,  $\sim$  20km North – West of the city of Groningen,

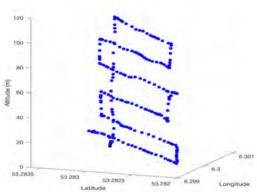
the Netherlands (Fig. 6.2.8). So far, three campaigns have been made (March 2017, May and October 2018), and the results of two campaigns have been derived and shown in this report.

To be able to integrate the enhanced CH<sub>4</sub> mole fraction, data needs to be interpolated since it is unequally distributed in the space. A threedimensional plot showing CH<sub>4</sub> concentration distributed in the



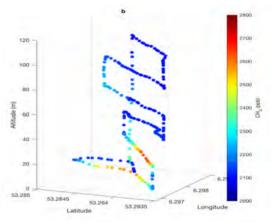
**Fig. 6.2.8:** CH<sub>4</sub> concentration, March 27<sup>th</sup> 2017. The wind came from the South – East. Left: upwind, right: downwind.

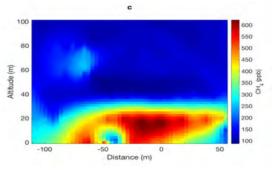
space (Fig. 6.2.9a, 6.2.9b), together with interpolated data (Fig. 6.2.9c), that were used to estimate a flux with a mass balance method.



CH<sub>4</sub> emission rates from the dairy cow farm is determined using a mass balance approach. The uncertainty is mostly caused by large uncertainties in the wind speed and wind direction measurements. Emission rates presented in Table 6.2.3 are representative for that particular day, 27<sup>th</sup> March 2017.

Table 6.2.3: Emiss Date	sion rates Source* (g/sec)	Source* (kgCH₄/day/(adult cow + manure))
27 <sup>th</sup> March 2017	$4.60 \pm 0.40$	$1.10 \pm 0.10$
3 <sup>rd</sup> May 2018	$3.70\pm0.70$	$0.90 \pm 0.20$
* The non-enteric emiss	ions are not subtracte	d





**Fig. 6.2.9:** 3D plot of CH<sub>4</sub> concentration, March  $27^{th}$  2017: a) upwind, b) downwind, c) interpolated data



#### 6.2.1.3 Future plans and expected results

In the next reporting period, the ESR 2 will improve her writing skills and organizational skills, as well complete the first manuscript on cow farm emissions.

The plan is to improve writing skills by attending the course "Publishing in English" from the 28th January to 26th March 2018, at the University of Groningen. The manuscript on CH<sub>4</sub> emissions from the Grijepskerk farm will be a result of a successfully co-organised measurement campaign in May and October 2018 (additional campaigns will be organised in spring 2019, if needed).

The general plans are listed first, followed by future plans related to mobile van measurements and UAV measurements, respectively.

- Analyse the MicroHH simulations of the Grijpskerk farm CH<sub>4</sub> mole fractions by ESR11 (1 month in 2018 or 2019)
  - o Learn more about the measurement strategy and apply that to future campaigns.
  - Perform more campaigns at the Grijpskerk farm (March 2019).
    - Fly simultaneously with two drones.
    - Solve GPS location and altitude issues.
- Draft of the 1st manuscript (2019)

**B** 

- Make an outline until the end of January 2019.
- Analyse the 3<sup>rd</sup> Grijpskerk campaign data (October November 2018).
- Secondment at ECN (29<sup>th</sup> October 2018 25<sup>th</sup> January 2019).

#### 6.2.1.3.1 Mobile van measurements

- B Further develop the modelling and the uncertainty analysis.
  - Develop quick analysis tool for large datasets with multiple farms.
  - Sensitivity analysis for critical parameters in dispersion model.
  - Comparison of Gaussian model with the backward Lagrangian model.
- Make drone and van CH<sub>4</sub> measurements simultaneously at one farm and compare the estimated CH<sub>4</sub> emission rates.

#### 6.2.1.3.2 UAV measurements

- Improve the accuracy of the wind speed measurements.
  - Deploy a 2D anemometer on the ground during the measurement campaign.
- Perform trace release experiments at a dairy farm to help develop the quantification method for both mobile van and UAV measurements.
- Later expand to different types of farms.
- Dotain more flights in different seasons.

#### 6.2.1.4 Collaborations (internal / external)

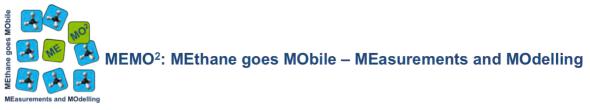
During the reported period the collaboration with the Energy research Centre of the Netherlands (ECN), has started in terms of Gaussian plume modelling. In addition, the collaboration will be continued through the secondment (October 2018 – January 2019).

Furthermore, two more collaborations are planned, with the University of Wageningen (WUR), the Netherlands, and the Royal Holloway University of London (RUHL), United Kingdom.

During the secondment (September – October 2018) of ESR 9 (RUHL) a drone-based field measurement campaign was performed at the Grijpskerk farm nearby Groningen. Four air samples were collected from the drone flight and one inside the Grijpskerk farm during the campaign, which will be analysed for isotopic compositions at RUHL.

In July 2018 we started a collaboration with ESR 11 (WUR) about possible model simulations of  $CH_4$  emissions at the Grijpskerk farm where we made drone-based  $CH_4$  measurements. Based on the campaigns, that we had so far, we realized that is also very important to design the measurement strategy and to combine with model simulations to derive accurate estimates of  $CH_4$  emissions.

Also, an external collaboration with Dr. Nico Ogink and Dr. Leon Sebek from the University of Wageningen, the Netherlands, is planned. A goal of this collaboration would be to relate our



measurements of CH<sub>4</sub> emissions with food intake, with respect to the amount, the diurnal cycles, and the seasonal cycles. Hopefully, this will provide us with some additional information to help design our strategy to make the measurements, and what farms/times we focus on in the next years.

#### 6.2.1.5 Risks and difficulties

Every measurement campaign is a potential risk. In a sense, we may fail to collect high quality data or any data at all. In our case, we depend very much on the weather conditions. Besides that, we need to consider possible technical failures of the instruments as well as the availability of our technicians. That is why we need to plan our measurement campaigns very thoroughly and on time.

One of possible difficulties is a failure of the RUG active AirCore measurements. In case of a failure, a new active AirCore system will be built.

#### 6.2.2 Deliverables

ESR 2 is involved in the following deliverables: D1.1 / D1.2 / D1.4 / D1.5 / D2.2 / D2.3.

**D1.1** – Report on harmonized method for mobile CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> measurements (month 18)

ESR 2 will be part of many mobile measurement campaigns, as well as responsible for the assessment of these data during her secondment at ECN (October 2018 – January 2019).

**D1.2** – Lightweight CH<sub>4</sub> sensor and AirCore developed and deployed on UAV (month 24)

A low-cost methane sensor (model TGS2600, Figaro USA, Inc.) is implemented to the active AirCore box to measure in-situ methane concentration. It is a low-power consumption gas sensor for the detection of air contaminants, which is sensitive to  $CH_4$  and other hydrocarbons. Unfortunately, measurements that were performed with a low-cost  $CH_4$  sensor before June 2108 with the active AirCore are not usable, because the batteries were not able to provide the  $CH_4$  sensor with sufficient power due to the power consumption of other electronic components. A low-cost  $CH_4$  sensor was tested and improved in the lab by adding two extra AA batteries to the system, and then later tested at the Grijpskerk dairy farm. Based on the improvement, the sensor should be able to measure enhanced methane concentrations on the active AirCore, aa well as to improve the spatial mapping of  $CH_4$  due to its quick response time. Above mentioned improvement was done by master student Sybren Couwenberg, as a part of his master thesis.

**D1.5** – Report and public on improved emission factors for different source categories from mobile measurements (month 42)

Nothing has been done.

**D2.2** – Improved isotopic signatures of local and regional CH<sub>4</sub> emissions (month 36)

ESR2 was part of mobile measurement campaign for one day in Groningen, the Netherlands, during the secondment of ESR7 (September – October 2018). During the 2<sup>nd</sup> Grijpskerk campaign, three air samples were collected from drone flights, and later analysed for isotopic compositions at UU by ESR8.

D2.3 – Publication on the use of isotopes for CH<sub>4</sub> source attribution in urban / industrial regions (month 36)

Nothing has been done.

#### 6.2.3 Training and network activities

#### 6.2.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
PhD Introductory Event	09. – 10.11. 2017.	University of Groningen (RUG)	To get acquainted with other PhD students, and the university.	1	attended	Training programme for a PhD students.
Mastering your PhD	1 <sup>st</sup> meeting: 19.02.2018. 2 <sup>nd</sup> meeting: 22.05.2018.	University of Groningen (RUG)	Project management, time management.	2	attended (2 meetings out of 6)	Training programme for a PhD students



#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

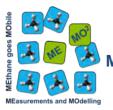
						(new meeting every 6 months).
1 <sup>st</sup> MEMO <sup>2</sup> School	05.02. – 16.02. 2018.	Schoorl (The Netherlands)	Two-week thematic school on CH <sub>4</sub> including courses, practical exercises, field campaigns, and data analysis.	6	poster, presentation	-
1 <sup>st</sup> MEMO <sup>2</sup> annual meeting	21.03. – 23.03. 2018.	EMPA (Switzerland)	Meet and update each other about the project, discuss and evaluate the progress of the first year and give an outlook to the second year.	2	poster, presentation	-
English Academic Writing Skills	01.05. – 21.06. 2018.	University of Groningen (RUG)	Focus on academic writing.	1	written essay	-
Gaussian Plume Modelling workshop	09. – 10.10. 2018.	University of Heidelberg (UHEI)	The workshop included lectures and hands-on practical exercises.	2	attended	-
Publishing in English	28.01. – 26.03. 2019.	University of Groningen (RUG)	Improve academic writing skills.	2	draft of 1 <sup>st</sup> manuscript	
Global Change course	February – April 2019	University of Groningen (RUG)	Various causes of climate change, global water and carbon cycle, stable isotope analysis methods, IPCC.	-	teaching assistant	-
2 <sup>nd</sup> MEMO <sup>2</sup> School	18.02. – 22.02. 2019.	University of Versailles-St- Quentin en Yvelines (UVSQ)	Meet and update each other about the project, discuss and evaluate the progress of the second year and give an outlook to the third year.	4	poster, presentation	-

#### 6.2.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
Energy research Centre of the Netherlands (ECN)	26.08. – 07.09. 2018.	Petten (The Netherlands)	ECN	Analyse data from 1 <sup>st</sup> MEMO <sup>2</sup> school (Feb. 2018) and make a poster for 3 <sup>rd</sup> ICOS conference.	Gaussian plume model.	Poster for 3 <sup>rd</sup> ICOS conference, and continue to work on Gaussian plume model.
Energy research Centre of the Netherlands (ECN)	29.10. 2018. – 25.01. 2019.	Petten (The Netherlands)	ECN	Modify Gaussian plume model for a drone, and participate in campaigns.	Gaussian plume model, CH₄ and N₂O emissions from cow farms in the Netherlands.	In progress.

### 6.2.3.3 Conferences

Conference name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation	Authors (main author + co- authors)	Public available (yes / no) / web link
BBOS Symposium 2017	25. – 27.10. 2017.	Berg en Dal (The Netherlands)	no	-	-	-
Industrial Methane Measurement Conference – PEFTEC 2017	29. – 30.11. 2017.	Antwerp (Belgium)	no	-	-	-
3 <sup>rd</sup> ICOS Science Conference	11. – 13.09. 2018.	Prague (Czech Republic)	poster	Quantification of methane emissions from dairy cows in the Netherlands	K.Vinkovic + T.Andersen, M.de Vries, W. Peters, A. Hensen, H. Chen	no



#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

BBOS Symposium 2018
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#### 6.2.3.4 Measurement / sampling campaigns

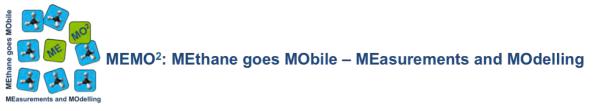
Campaign	Date (start – end, planned (when))	Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
1 <sup>st</sup> Grijpskerk campaign	27 <sup>th</sup> March 2017	Grijpskerk (The Netherlands)	RUG	Drone measurements at the Grijpskerk cow farm.	Quantify CH <sub>4</sub> emissions.	-	Material for 1 <sup>st</sup> publication.
1 <sup>st</sup> MEMO2 School	9 <sup>th</sup> – 11 <sup>th</sup> February 2018	North Holland (The Netherlands)	ECN	Van measurements across the North Holland province.	Design, plan and run field campaigns to measure atmospheric CH <sub>4</sub> at site scale.	-	Some of the results presented on 3 <sup>rd</sup> ICOS conference in Prague.
2 <sup>nd</sup> Grijpskerk campaign	3 <sup>rd</sup> May 2018	Grijpskerk (The Netherlands)	RUG	Drone measurements at the Grijpskerk cow farm.	Quantify CH₄ emissions.	bag samples / 3	Material for 1 <sup>st</sup> publication.
3 <sup>rd</sup> Grijpskerk campaign	19 <sup>th</sup> October 2018	Grijpskerk (The Netherlands)	RUG	Drone measurements at the Grijpskerk cow farm.	Quantify CH <sub>4</sub> emissions.	bag samples / 4	Material for 1 <sup>st</sup> publication.

#### 6.2.4 Dissemination activities

Except for the contributions to the conferences no scientific publications or other dissemination activities so far from the ESR.

#### References

Benjamin Poulter, e. a., 2017. Global wetland contribution to 2000-2012 atmospheric methane growth rate dynamics. Environ. Res. Lett. 12 094013.



# 6.3 ESR3 - Validating CH<sub>4</sub> inventories over intense mining area, natural and anthropogenic emissions

#### ESR3

Validating CH4 inventories over intense mining area, natural and anthropogenic emissions

ESR	Mila Stanisavljevic ; mila.stanisavljevic@gmail.com
Supervisor	Jaroslw Necki ; <u>necki@agh.edu.pl</u>
Co-supervisor	Martina Schmidt ; martina.schmidt@iup.uni-heidelberg.de
Non-Academic mentor	Wojciech Wolkowicz ; <u>wwola@pgi.gov.pl</u>
Official start – end date	16.10.2018 - 16.10.2020

#### 6.3.1 Scientific progress

#### 6.3.1.1 Project introduction and objectives

Main task of the project is to construct the methane balance and with most accurate approximations of particular fluxes, based on documented emissions and direct measurements done with mobile platforms. Besides, methane isotopic values should help with the problem of identification of sources and it's understanding.

The area of interest covers the Upper Silesian Coal Basin – the biggest hard active coal mining areas in Europe with high mechanized coal bed exploitation. Beside active mines there are abounded coal mines around Silesia and Germany (will be done during secondments) to be investigated. Additionally, Lublin coal Basin has been checked for methane emission and methane isotopic composition.

Beside mining industry emission there are a lot of other sources (landfills, city gas network leakages, peatlands and wetlands) to be inventoried. The expected results will consist of flux estimation together with its uncertainty for each type of the source together with their special distribution.

Available technique for mobile and stationary measurement: Picarro CRDS, FTIR (Fourier-transform infrared spectroscopy), LGR (Los Gatos Research), Static Chamber techniques, AirCore system, Bag samples (dilution techniques), 2D sonic anemometer, GPS and power system for 24h continuous measurement.

Three different secondments are planned during the project. The first one should be at Heidelberg University, Germany. The ESR will work for an in situ isotopic measurement using AirCore. During the second secondment, the ESR will spend 4 month at the partner organisation PGI (Polish Geology Institute) to work on quantifying mining emissions of methane using GS (gas chromatography mass spectrometry). The last one is planned at Utrecht University for isotope measurements.

#### 6.3.1.2 Project results

#### 6.3.1.2.1 First year

During November 2017 the ESR attended the PEFTEC conference (Industrial Methane Measurement Conference 2017) and presented a poster as co-author. Besides this, the ESR participated in several measurement campaigns

#### Bełchatów coal mine

Bełchatów coal mine belongs to a group of lignite coal mines with typical low methane emission. With an annual production of 41.2 million tonne of lignite coal, it takes a leadership role in lignite coal production in Poland. In case of methane emission to the atmosphere, well known databases such as EDGAR or EUROSTAT do not report any major emission from the source.

To validate databases reports we organized a first measurement campaign on 19.10.2017. The goal was to get familiar with the equipment and campaign organization as well as to check methane emissions coming from non-point sources. Fig. 6.3.1 presents a longitudinal transect around mine and



waste dump with methane concentrations observed in downwind direction. Different colours in Fig. 6.3.1 represent different methane concentrations.

Although the wind direction was northwest the highest methane plume concentrations was observed in downwind direction but from an additional source - waste dump. On the northwest side of the transect the highest methane concertation was observed (up to 2.9 [ppm], purple colour, Fig. 6.3.1) but in one point which might indicted additional source of methane.

A graph of methane concentrations and its isotopic composition observed over time are shown in Fig. 6.3.2. Fig. 6.3.2a shows the methane concertation on downwind direction (marked blue) and Fig. 6.3.2.b the methane isotopic composition around the mine (observed in downwind location, where methane plume has been identified - marked blue).



Fig. 6.3.1: Longitudinal transect around the Belchatów coal mine

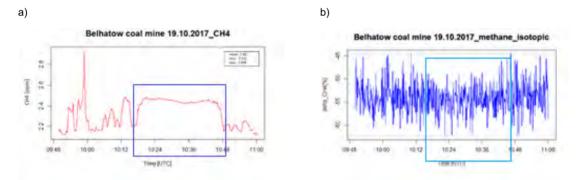


Fig. 6.3.2: a. Methane time dependence b. Isotopic composition over region

Conclusions: The maximum concertation measured in downwind direction was 2.93 [ppm]. Accompanying value of methane isotopic composition was in average – 54.44 ‰, which indicates a biogenic origin of methane. In future we might perform more measurement campaigns around Belchatów region to further investigate the origin of methane gas coming from Belchatów coal mine.

#### First measurement campaign Silesia region

A first measurement campaign took a place 21.12.2017. The goal was to get familiar with the equipment, be ready to perform own measurement campaigns, and collect data for the first data analysis (methane concentration and methane isotopic values). During a longitudinal transect (Fig. 6.3.3) we have visited 5 different mine shafts - Brzeszcze shaft III and V (shaft I and shaft V has the same position-one near the other), Silesia, Pnowek shafts IV and V. Data has been presented at the EGU 2018 conference.



Fig. 6.3.3: Transect through Silesian region





Fig. 6.3.4: Google Earth plot of methane plume elevation; left: Brzeszcze III mine shaft; right: Brzeszcze V mine shaft

During driving methane elevation up to 6 [ppm] has been identified near Brzeszce mine shafts III and V (Fig. 6.3.4). Methane isotopic composition has been analysed used Keeling plot approach with a goal to have the best estimate methane isotopic composition value over particular mine shafts (Fig. 6.3.5).

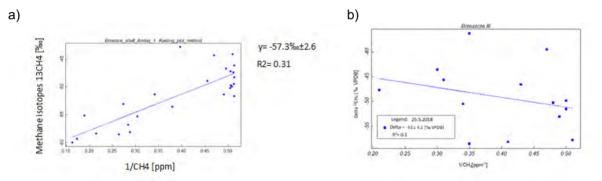


Fig. 6.3.5: Keeling plot approach of methane isotopes a. Brzeszcze mine V (I) b. Brzeszcze mine III

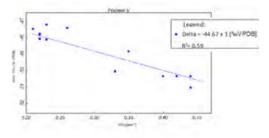
Even around one mine different isotopic values have been identified. It might indicate different origins of methane gas, different levels of coal excavation inside mine or different types of ventilation. The problem of different isotopic methane values around Silesian region will appear later on during investigation.

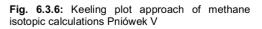
30 km far from Brzeszcze coal mine we performed a transect around the Pniowek V mine shaft. Methane concentration from this source was up to 20 [ppm]. The isotopic composition value (Fig. 6.3.6) is close to Brzeszcze III mine shaft – an indicator for the complex geology of Silesian region.

Table 6.3.1 shows an overview of methane concentrations and methane isotopic values. During later investigation, bag sample taken directly from source has been analysed and compared with results from first measurement campaign.

Table 6.3.1: CH4 isotopic composition at mine shafts - Silesian region

Name of mine shaft		Max CH <sub>4</sub> concentration [ppm]	CH₄ isotopic Value [‰]	Linear fit
	Brzescze III	10	-43.0±4.1	R <sup>2</sup> = 0.1
	Brzescze V	6	-57.3±2.6	R <sup>2</sup> =0.5
	Silesia	4	-59.0±5.1	R <sup>2</sup> =0.4
	Pniówek V	10	-44.6±1	R <sup>2</sup> =0.6
	Pniówek IV	20	-47.6±0.9	R <sup>2</sup> = 0.8







D5.9 MEMO<sup>2</sup> – Midterm Review Report

Conclusion: the obtained methane isotopic values of 5 different mine shafts differs from -43.00 ‰ to - 59.00 ‰, which fits more to mixing origins of methane gas. Linear fit to Brzeszcze coal mine is quite low. The same problem will show up again during later measurements. The maximum concentration of methane was 20 [ppm] Pnowek coal mine due the high miners' activities.

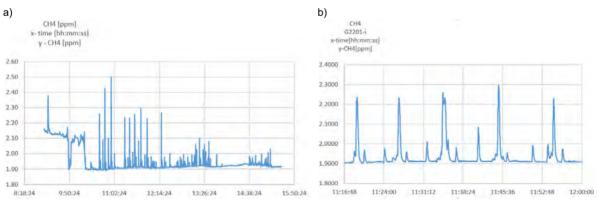
Also, during the reported period the PhD candidate participated in the 1<sup>st</sup> MEMO<sup>2</sup> school, held in Schoorl, the Netherlands, 5 – 16 February 2018. The 1<sup>st</sup> MEMO<sup>2</sup> school was a two-week school on methane including courses, practical exercises, field campaigns, and data analysis. On the second day of our field campaign (10<sup>th</sup> Feb.2018) methane emission from the release test was measured (Fig. 6.3.7, Fig. 6.3.8). In this particular case Picarro G2201-i CRDS was used for collecting data. The equipment was installed in a car, and connected to GPS and one anemometer for the better comparison of results. Beside cylinder filed with methane gas, additional sources have been identified: 2 farms. Values from farm will be calculate as background value.

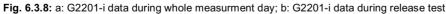
Conclusion: As we have driven the car all the time in one direction, it might it be better to change direction from time to time, for a better comparison of the results. Determine the exact position of a plume depends on the precision of the GPS equipment. Although GPS instrument are highly sensitive and precise, in case of slow driving (as slowest as possible) the equipment is less sensitive.





**Fig. 6.3.7:** Identification of methane plume, the left figure represents all measurement day- release test and data from 2 farms. The right figure represent only data obtained during release test – without values obtained from farms.

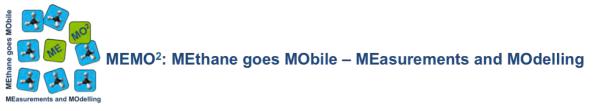




#### 6.3.1.2.2 Second year

During the second project year the ESR attended the EGU conference in April 2018 and presented results from the first year as a poster: Bottom – up methane budget estimation from the sources over Upper Silesian Coal Basin

From 23 May to 14 Jun the ESR attended the CoMet - Coal Mine Methane (CMM) measurements organized by DLR institute and  $MEMO^2$  colleagues. The ESR was involved in the FTIR stacionary measurements at the downwind location, the data are currently processed and evaluated. Additional



FTIR measurements were performed from March 2018 to 22 May 2018 at Faculty of Physics and Applied Computer Science building's roof.

#### Coal Mine Methane (CMM) measurements during CoMet

The CoMet campaign was organized by the DLR institute, Germany. Three measurement vans have been involved for 15 days (UU, UHEI) / 24 day AGH of survey. The area of interest was the Upper Silesian Basin - region with mining activities. Fig. 6.3.9 shows all paths of the AGH van during the measurement's days.

In this region 33 mines are active with additional methane sources: landfills, cities gas network, cow farms, wetlands and agricultures. During the CoMet measurements only the active mines have been checked. Before and after every measurements calibration gas has been flushed into instruments - to validate observed values and escape potential drifts of instruments. Additionally, a new instrument LGR has been installed together with AGH Picarro G2201-i, connected to the same inlet.

During the night instruments have been run at the hotel location to investigate the night inversion and the potential influence from coal mines. To harmonize instruments, the time shift and differences between measured concentration need to be apply.

The total time shift between instruments is 114s (response time between instruments is also different due the different frequency). Fig. 6.3.10 represents the records from instruments during observed 4h. between Different measured concentration is  $(0.1122 \pm 0.004)$ [ppm]. Later on, during longitudinal transect the same difference will appear. The CH<sub>4</sub> record (blue graph)

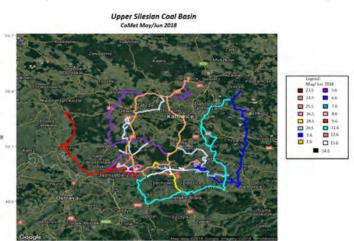


Fig. 6.3.9: Path of AGH van during CoMet measurements

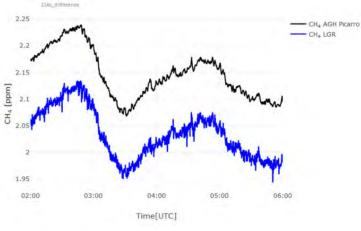


Fig. 6.3.10: 114s time difference LGR- Picarro G2201-i CRDS

presents more noisy record, again due the different frequency between instruments.

During Longitudinal transect instruments mentioned above checked concentration near source, as well as isotopic compositions. Fig. 6.3.11 represents the biggest elevation identified during driving (Upper Silesian Coal Basin). The maximum concertation identified near Pnowek coal mines- up to 350 [ppm]. But near others mines, as Brszeszce, methane elevation was up to 20 [ppm] has been identified.



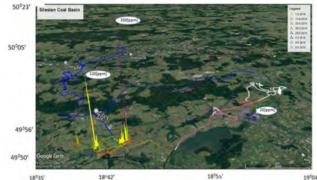


Fig. 6.3.11: Methane elevation during observed days

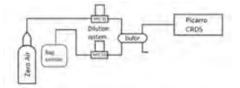


Fig. 6.3.12: Experimental setup used to determine methane isotopic value during dilution test

Conclusion: Collection of bag samples need to be continued. as it's the best method for validation of origin of methane gas. The best method to origin understand of methane qas is to understand isotopic compositions of methane. During secondments ESR will learn how to use other technique such AirCore and IRMS technique in propose to obtain more

 Table 6.3.2: Overview of methane isotopic values over Upper Silesian region (collected during CoMet measurement)

Name of mine		Bag sample	Picarro record	Literature value
			δ <sup>13</sup> CH₄ [‰]	
Pniówek	Pniówek V	Na	-47.2±2.1	-69.2 to -71.9
	Pniówek III	-53.7±2.8	-50.7±1.0	-79.1 to -69.2
	Pniówek IV	-47	-49.3±2.3	Na
Silesia Silesia V		Na	-59.8±1.5	-61
	Silesia III	Na	-58.8±1.6	
Boryna Boryna III		-50.9±3.4	-55.9±3.4	Na
Boryna IV		-53.8±3.7	-51.6±3.2	
Jastrzebie		Na	-55.2	-52.6
Knurów		Na	-35.4	Na
Brzeszce III		Na	-48.5	-47.1 to -48.9

\* Literature values has compared with data from Kotarba et al 2001 (Composition and origin of coalbed gases in the Upper Silesian and Lublin basins, Poland)

precise results. In a way of processing of data collected during CoMet measurement next step is validate emission using Gaussian model and compare all obtain data with databases EDGAR, EPRIT, EUROSTAT. ESR will work more on Large uncertainties has been identification during literature comparison between databases.

#### Measurements - Lublin coal Basin

The second active hard coal basin has been checked for methane concentration, source identification. Databases such EPRIT do not report methane emission from Lublin Basin. Otherwise basin consist from one active mine- Bogdanka (only 3 ventilation shafts).

Measurement campaign has been performed 29.8.2018. All 3 mine shafts were checked. The highest methane concertation was identified near main shaft in downwind direction- 12.54 [ppm] (Fig.

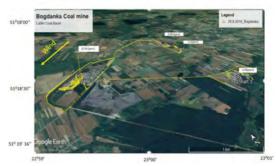


Fig. 6.3.13: Bogdanka Coal mine

To validate the measurements and the recorded plumes, air bag samples has been taken directly from the shafts. In the laboratory a conditional dilution test was performed measuring the methane isotopic values and compare them with results obtained during driving (Picarro CRDS record).

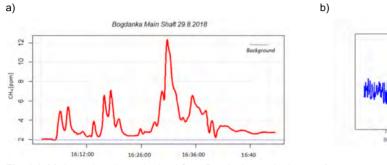
To perform measurements of high  $CH_4$  concentrations from sources (up to 1000 [ppm]) with our equipment gas from bag samples need to be near atmospheric value. Our laboratory experimental set up is shown in Fig. 6.3.12.

Bag sample are connected to one line, other line is zero air cylinder. After proper choosing of flow (from sample and cylinder) Picarro CRDS is able to measured proper methane isotopic value. Table 6.3.2 represent results of measurement air bag samples, Picarro CRDS record and comparison with literature.



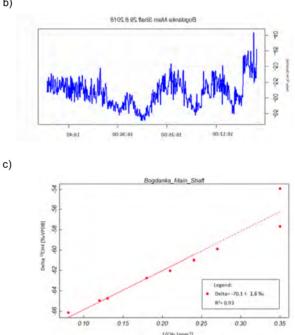
6.3.13). Near another two shafts only slightly higher concentration than background were identified. Additionally, air bag samples have been collected and tested in laboratory conditionals using experimental set up from Fig. 6.3.12.

Methane isotopic values from Picarro CRDS record has been compared values from bag sample (Fig. 6.3.14). Values fits more to biogenic record (-  $70.1 \pm 1.6$ %).



**Fig. 6.3.14:** Isotopic values obtained from Bogdanka coal mine a. record from Picarro G2201-i analyser b. isotopic record from Picarro G2201-i analyser c. Keeling plot from data from a. and b.

Conclusion: In future, more miner's activities are planned for Lublin region. Coal reserved are the same as in Silesian area. More measurements campaigns need to be organized with goal of monitoring methane emission coming from mining activities.



#### Measurement methane emission from abandoned mine and waste dump

Into regular databases reports from post mining emission take between 4 to 10 % of total CH<sub>4</sub> emission from coal to atmosphere. Otherwise, no precise data quantifying emissions from abandoned (Polish) mines are currently available. Characteristic of CH<sub>4</sub> emission from abandoned mines during time: emission is high few months, then emission start to be constant due the long period of time.

On the Fig. 6.3.15 are first results of measurement campaign over few Silesian abandoned mines. We have performed longitudinal transect near closed mines, and collected bag samples from plume.



Fig. 6.3.15: a. Piast (closed 2012) b. Krupinski (closed 2018) c. Warszowice (closed 2000) abandoned coal mines

Near Krupinski abandoned coal mine, methane elevation reaches values up to 12 [ppm] (Fig. 6.3.16). Krupinski coal mine is recently closed, hence still high methane concentration coming out from shaft. Observation near Pisat coal mine indicate higher evaluation above background of 2.4 [ppm] (Fig. 6.3.17). The mine is closed since 2012, hence expectation is constant methane emission. Additionally, bag sample from plume are collected (Krupinski). In laboratory conditions, samples are tested.



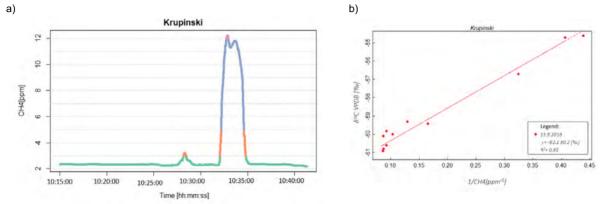


Fig. 6.3.16: a. Krupinski abandoned coal mine- methane elevation b. isotopic characterisation

Conclusion: Observed value indicate more biogenic value (methane isotopic values from Krupinski coal mine reach -62.1±0.2 ‰). The open question is: do bacterial come into mine and produce methane? In the moment of reporting there are no experimental results about origin of methane from abandoned coal mine (Silesian Basin), hence additional investigations are necessary. ESR3 is working on measurement campaign as well as obtained knowledge of origin of methane from abboned coal mine.

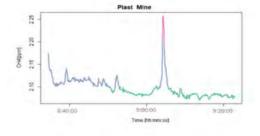


Fig. 6.3.17: Piast Coal abandoned coal mine

#### 6.3.1.3 Future plans and expected results

For the next year, ESR plans to finish secondments at UHEI (January 2019), UU (April 2019), and PGI (June / October 2019)

First manuscript of result from CoMet campaign and additional measurement camping will be prepared during 2019.

Additional measurement camping will be organized: regular checking of particular mine shafts Upper Silesia Coal Basin, post mining- abandoned mining campaign, city gas network measurement, and methane flux from agriculture. Additionally, ESR3 will try to collect water samples from mines and test them during secondments.

Before next reporting period ESR3 will attend NCGG conference.

ESR3 will work on Gaussian modelling. Skill is partly improved by attending Gaussian modelling workshop October 2019.

#### 6.3.1.4 Collaborations (internal / external)

During the reported period is started a collaboration with German DLR institute (organization of CoMet campaign). ESR cooperate in team of working with FTIR equipment (Krakow measurement as well as measurement during campaign). The idea was to use new, complex, expensive technique -total column in team of observation methane emission.

Future more cooperation with industrial (mining facilities) has been planed. ESR try to get permission for sample collecting. As ESR is not high level Polish speaker work is still in progress.

#### 6.3.1.5 Risks and difficulties

ESR has language issues during daily work, e.g administration in Poland do not speak English, mining companies do not want to cooperate (foreign student), attending lectures recommended for PhD



students is partly possible, as lectures are mostly in Polish language. Teaching duties is not possible to fully performed – not enough English classes.

ESR start to learn new language to avoid problems, but still giving classes to Polish students in Polish language is not possible.

#### 6.3.2 Deliverables

ESR 3 is involved in the following deliverables: D1.1 / D1.4 / D1.5 / D2.2 / D2.3 / D2.5.

**D.1.1** – Report on harmonized method for mobile CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> measurements (month 18)

ESR 3 will be part of many mobile measurement campaigns, as well as responsible for the assessment of these data. Harmonized method from CoMet camping is still in progress.

D1.4 – Report and publication of the results from the campaign in Silesia (month 36)

The preparation for this deliverable is ongoing. ESR will work as co-author or 1<sup>st</sup> author in particular future publications

**D1.5** – Report and public on improved emission factors for different source categories from mobile measurements (month 42)

Ongoing. ESR has performed measurements around post mining area, which is a poorly investigated area from a emission factor perspective. An idea is to perform measurement around post-mining area, but in moment of reporting there is no progress as coal companies do not wat to cooperate

D.2.2 – Improved isotopic signatures of local and regional CH<sub>4</sub> emissions (month 36)

The ESR is working on measurement of methane plume from mining area and methane isotopic composition. First step has been done during CoMet measurement. In future ESR will perform more measurement with goal to see potential changing from isotopic signature

D.2.3 – Publication on the use of isotopes for CH4 source attribution in urban/industrial Regions (month 36)

First publication will be prepared with cooperation of other groups at AGH University .

**D.2.5** – Report providing isotopic maps at grid scale from inventories and atmospheric measurements ESR collected data necessary for improvement of maps at grid scale

## 6.3.3 Training and network activities

#### 6.3.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
Applied geology	16.10.2017 – 20.1. 2018.	AGH University	Be familiar with complex Polish coal geology	6	attended	Courses are obligator for PhD students in Poland.
Ecological economics in global change	16.10.2017 – 20.1. 2018	AGH University	Get a knowledge about Ecological economics in global change	3	attended	Courses are obligator for PhD students in Poland.
Physics	16.10.2017 – 20.1. 2018	AGH University	Improve physics skills	5	attended	Courses are obligator for PhD students in Poland.
Introduction to statistics and data handling	1.3- 1.7. 2018	AGH University				Courses are obligator for PhD students in Poland
1 <sup>st</sup> MEMO <sup>2</sup> School	05.02. – 16.02. 2018.	Schoorl (The Netherlands)	Two-week thematic school on CH <sub>4</sub> including courses, practical exercises, field campaigns, and data analysis.	6	poster, presentation	-



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

## D5.9 MEMO<sup>2</sup> – Midterm Review Report

1 <sup>st</sup> MEMO <sup>2</sup> annual meeting	21.03. – 23.03. 2018.	EMPA (Switzerland)	Meet and update each other about the project, discuss and evaluate the progress of the first year and give an outlook to the second year.		poster, presentation	-
Isotopic workshop	17. – 19.09. 2018.	Royal Holloway University of London	Learning about isotopic skills and methods	-	presentation	-
Gaussian Plume Modelling workshop	09. – 10.10. 2018.	University of Heidelberg (UHEI)	The workshop included lectures and hands-on practical exercises.	-	attended	-
Air Pollution	October 2018– January 2019	AGH University	Get a knowledge about non methane air pollution substance	-	In progress	-
2 <sup>nd</sup> MEMO <sup>2</sup> School	18.02. – 22.02. 2019.	University of Versailles- St-Quentin en Yvelines (UVSQ)	Meet and update each other about the project, discuss and evaluate the progress of the second year and give an outlook to the third year.	??	poster, presentation	-

#### 6.3.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
University of Heidelberg	13.1. – 10.02. 2019.	Heidelberg Germany	UHEI	Laboratory teste (calibration), field transect (AirCore, post mining emission)	New system: AirCore system Cross Calibration Ethane	ESR will be able to perform ethane calibration in Poland

# 6.3.3.3 Conferences

C	Conference name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation/poster	Authors (main author + co-authors)	Public available (yes / no) / web link
	Industrial Methane Measurement Conference – PEFTEC 2017	29. – 30.11. 2017.	Antwerp (Belgium)	Poster	Isotopic composition of methane from exhausts of mines and gas fields in Southern Poland" -	Jaroslaw Necki, Miroslaw Zimnoch, Alina Jasek, Luksz Chmura, Michal Galkowski, Wojciech Wolkowicz, Patryk Lakomiec, Piotr Korben, Mila Stanisavljevic	no
	EGU 2018	07. – 12.04. 2018.	Vienna (Austria)	poster	Bottom – up methane budget estimation from the sources over Upper Silesian Coal Basin		no

### 6.3.3.4 Measurement / sampling campaigns

Campaign	Date (start – end, planned (when))	Location	Host	Description work	of	Scientific objective	Samples (nature / number)	Results and future plans
Belhatow coal mine	19.10.2017	Poland		Checking methane emissio	on			



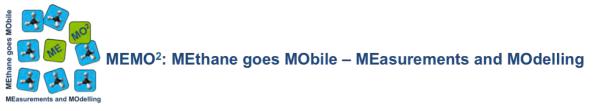
# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

				from non-point source			
1 <sup>st</sup> MEMO2 School	9 <sup>th</sup> – 11 <sup>th</sup> February 2018	North Holland (The Netherlands)	ECN	A van measurements across the North Holland province.	Design, plan and run field campaigns to measure atmospheric CH <sub>4</sub> at site scale.	-	-
Lublin coal Basin	28.8.2018	Poland Silesia					
Post mining emission	13.9.2018 1.10.2018	Poland Silesia		Checking methane emission from closed mines			
Samplings from mine shafts		Poland Silesia		Collecting of bag samples directly from mines shafts		10	

### D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.3.4 Dissemination activities

No further dissemination activities except for the above mentioned conference contributions have been executed.



# 6.4 ESR4 - Assessing CH<sub>4</sub> emission from wetlands and other sources by use of mobile measurements

#### ESR4

Assessing CH $_4$  emission from wetlands and other sources by use of mobile measurements

ESR	Patryk Lakomiec, patryk.lakomiec@nateko.lu.se
Supervisor	Janne Rinne, janne.rinne@nateko.lu.se
	Jutta Holst, jutta.holst@nateko.lu.se
Co-supervisor	Dominik Brunner, <u>dominik.brunner@empa.ch</u>
Non-academic mentor	Johan Fagerqvist, johan.fagerqvist@avfallsverige.se
Official start-end date	2017-08-01-2021-07-31

#### 6.4.1 Scientific progress

#### 6.4.1.1 Project introduction and objectives

The ESR4 project will quantify CH<sub>4</sub> emissions in Sweden with focus on wetlands and lakes by use of mobile measurements from a small research aircraft and vehicle. Airborne measurements will allow for direct assessment of vertical CH<sub>4</sub> exchange between surface and atmosphere as well as the vertical structure of CH<sub>4</sub> concentration within the atmospheric boundary layer. As the isotopic signatures of CH<sub>4</sub> varies depending on the origin, air samples for isotopic analyses in the lab and in situ field measurements will be taken at specific points of interest to be able to identify sources of measured CH<sub>4</sub>. Isotopic signals from these sources will be assessed by comparison with the analysis of samples from these source categories collected by WP1 students in trans-European mobile measurement campaigns.

The planned project work includes:

- Analysis of existing airborne flux data
- Airborne flux measurement campaign over wetland
- Mobile car measurement campaign to observe <sup>13</sup>C signature of methane enhancements
- Field measurement to specify <sup>13</sup>C signatures of methane emission from wetlands

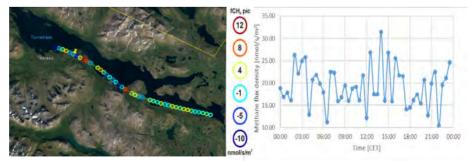
#### 6.4.1.2 Project results

#### 6.4.1.2.1 First year

The project is focused on wetland ecosystems. Until this moment, the ESR started to analyse data from previous campaigns, in which the small research aircraft owned by Lund University has been used. In total, data from around 20 measurement flights are available. The analysis of the airborne data consists of two steps: (1) Understanding the variables in the airborne raw dataset and (2) calculating the airborne greenhouse gas fluxes using the WURMFP toolbox (Matlab toolbox designed for calculating eddy

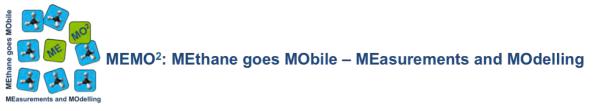
covariance from aircraft WU by (Vellinga et.al 2013)). In summer 2015, the aircraft took off from Kiruna airport for measurements along the Torneträsk valley and the ICOS site Abisko-Stordalen. High-frequency EC data for comparison

of temporal variabi-



**Fig. 6.4.1:** Spatial distribution of methane flux density measured on way through valley from 2015-07-13 8:45-9:02 (CET), measured at an average height of 130 m height above the ground, pin shows location of ICOS Sweden station Abisko-Stordalen.

lity at the Stordalen mire and spatial variability from the airborne measurements have also been started



to be analysed by the ESR. The fluxes showed a high variability with generally lower fluxes over the lake than over the mire (Fig. 6.4.1).

#### 6.4.1.2.2 Second year

During the second year, the ESR focused on 3 topics. The first problem was connected with hardware problem of the used Picarro gas analyser. The second problem was connected with data analyses and software understanding. The third topic was connected to gathering information about isotopic signatures from wetlands and spatial heterogeneity of isotopic signatures in wetlands.

#### 6.4.1.2.2.1 Analyses of cavity pressure instabilities of the gas analyser

During the analyses of the existing data, inconsistencies in methane concentration data and cavity pressure arose, where the analyser showed a high variability in cavity pressure while moving. To find out, whether mechanical adjustments could avoid the problem, a series of tests was performed. During the tests, the Picarro analyser was mounted on a heavy-duty trolley which acted as moving platform. Tedlar bags were connected to Picarro analyser to have the same air composition throughout the experiment (Fig. 6.4.2).

The idea was to either find appropriate vibration damping material that would dampen the vibrations of the analyser or to find a position of the analyser where the cavity pressure would react less sensitive to vibrations of the moving platform. However, no appropriate material could

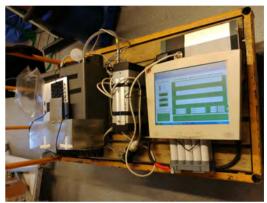


Fig. 6.4.2: Moving platform designed to test vibration damping material

be found to eliminate then cavity pressure variations (Fig. 6.4.3). Finally, ESR and supervisors contacted Picarro support who connected to the analyser remotely. As the error could not be solved on this way, the analyser was finally sent to USA for repair.

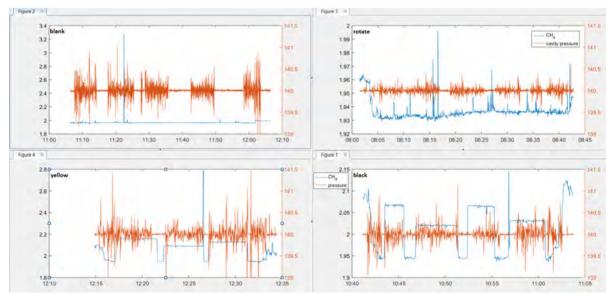


Fig. 6.4.3: Test of vibration damping materials while platform was moving. Blank: no additional damping material, yellow, black: different additional damping materials, rotate: rotated mounting of the analyser on the trolley.



#### 6.4.1.2.2.2 Spectral analyses of existing airborne measurement data

To investigate the feasibility to use the used set-up for airborne eddy-covariance flux calculations, the spectral characteristics of the measurement data has been analysed with the aim to investigate the time and length scales of the eddies contributing to the eddy-covariance flux. For this, it was necessary to implement a new function in the Matlab toolbox WURMFP suite.

The functions used for the spectral a) analyses were adapted from the eddyUH software to meet the requirements for airborne eddy-covariance data. For flux calculations, the WURMFP toolbox creates high frequency (50 Hz) time series from all data measured at lower frequency (e.g. Picarro gas concentrations at 1 Hz, LiCor gas concentrations at 20 Hz) by using a stepwise algorithm, following some linear interpolation. From the power spectra (one example output is given in Fig. 6.4.4) it is obvious, that the frequency response follows the expected b) slope in the frequency range, which was covered by the original data set.  $CH_4$ However. for the frequency response shows the need for corrections using transfer functions and noise introduced by the artificially created higher time resolution of the time series. Investigations on the co-spectra of vertical wind component and air temperature, resp. gas concentrations (example output shown in Fig. 6.4.5) show that only part of the high-frequency noise is reflected in the co-spectra, which in general follow the expected frequency c) response. The co-spectrum frequency response of vertical wind speed and 50 Hz Licor CO<sub>2</sub> signal which is not used for flux calculations shows a flatter slope while for CH<sub>4</sub> the slope is stepper than expected. In the next step, transfer functions will be used to correct for this. The ogives (Fig. 6.4.6) show that not all contributions from the bigger eddies can be captured by the chosen averaging

path length using the uncorrected Picarro

gas concentration data. The ogive of the

high frequency co-spectrum (<w'T'>)

however, indicate, that 80% of the flux is

contributed on time scales smaller than 2

sec. Assuming an average air speed of

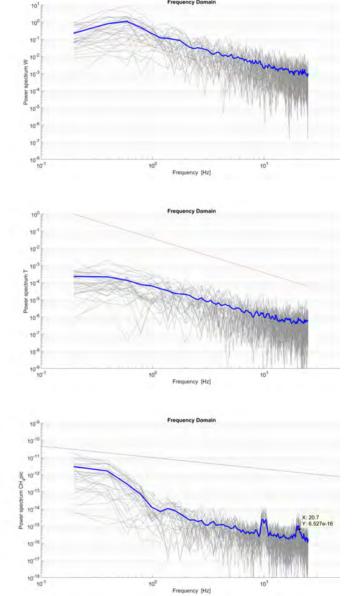


Fig. 6.4.4: Average power spectra for (a) vertical wind component w (50 Hz sampling frequency), (b) temperature T (50 Hz sampling frequency), and (c) CH<sub>4</sub> concentration (1 Hz sampling frequency).

85 kt during measurement flights, this this translates into a length scale of 90 m. The analyses on the spectral analyses and flux calculations are ongoing and will be updated during the next progress report.



D5.9 MEMO<sup>2</sup> – Midterm Review Report

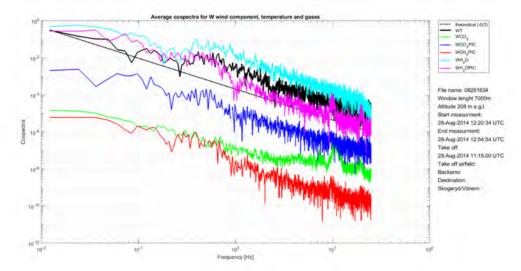


Fig. 6.4.5: Average co-spectra for vertical wind component and temperature, resp. gas concentrations during one example measurement flight.

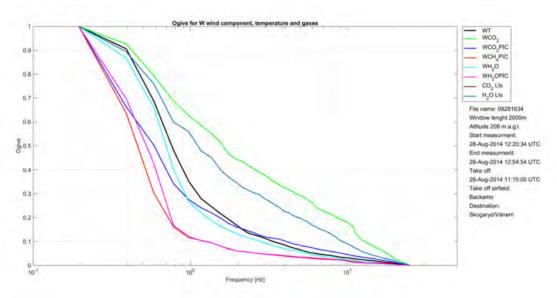


Fig. 6.4.6: Ogives for vertical wind component and temperature, resp. gas concentrations during one example measurement flight.

#### 6.4.1.2.2.3 Horizontal variability of isotopic signatures in wetlands

Air from an automatic chamber system at the wetland site Skogaryd Mycklemossen (58°21'52.85"N, 12°10'16.71"E) was sampled into bags on October, 12, 2018. The measurement system at Mycklemossen contains six different chambers. Each pair of chambers has different water level conditions, (dry, semi-wet, wet). Each chamber is measured half hour. The continuous measurements are done using a Picarro isotope analyser. Data from the manual sampling will be compared to the data from the continuous sampling (8 sample bags per chamber).



#### 6.4.1.3 Future plans and expected results

The ESR is planning to finish more courses connected with his topic to deepen the knowledge in this field and collect the required 60 ECTS points.

The spectral analyses, as well as the final flux calculation and interpretation of results will be continued in the coming weeks. Information about the temporal variability of CH<sub>4</sub> emissions will be gathered by further analyses of eddy-covariance data from wetlands which are part of the ICOS Sweden network as well as through repeated flight campaigns over the same wetland area in Sweden.

#### 6.4.1.4 Collaborations (internal / external)

The collaboration with the Avfall Sverige has started. Few joint measurements campaigns have been done. Within this collaboration, the ESR is focused to develop a better method to monitor leaks on landfills.

In collaboration with the Sveriges lantbruksuniversitet (Swedish University of Agricultural Sciences, SLU) in Umeå, research flights over a chrono-sequence of wetlands in northern Sweden (along the coast between Umeå and Luleå) is scheduled for 2019. Data from the campaign will enlarge the database for quantifying CH<sub>4</sub> wetland emissions from different wetlands.

#### 6.4.1.5 Risks and difficulties

ESR haven't met any difficulties to this moment.

#### 6.4.2 Deliverables

D1.2 - Report & publication on seasonal variation of  $CH_4$  emissions from wetland and lakes in Sweden (month 30)

Literature review was started, measurement campaigns have been planned and are scheduled for summer 2019. Data from previous flight campaigns as well as continuous data from ICOS Sweden site Abisko-Stordalen, a subarctic fen, has been started to be analysed.

D1.4 - Improved emission factors for different source categories from mobile measurements (month 42)

In progress. Especially the planned measurement campaigns in collaboration with SLU and LU, but also the ongoing measurements in collaboration with Avfall Sverige / SWECO on landfills will provide additional data material to receive improved emission factors.

**D1.5** - Report on harmonized method for mobile CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> (month 18)

The ESR is providing data and information on his measurement methods to this report.

The mobile flux platform (MFP) for measuring methane concentrations and fluxes on board the Sky Arrow ERA consists of two major parts: the wind measurements and gas concentration measurements. A GPS/INS system is used to determine the aircraft's position. Atmospheric turbulence measurements are made with the "Best-Aircraft-Turbulence Probe" (BAT-probe), developed by NOAA's Atmospheric Turbulence and Diffusion Division (NOAA-ATDD) and Airborne Research Australia (ARA). The BAT-probe measures the velocity of air with respect to the aircraft using a hemispheric 9-hole pressure sphere that records static and dynamic pressures by means of four differential pressure transducers. The actual 3-dimensional wind components relative to the surface can be derived from a blend of the GPS/INS data, BAT-probe data, and corrections for the aircraft's movements (heading, roll and pitch). A fast temperature sensor at the nose of the measurement probe. CH4 measurements are done using a close path gas analyser (G2301-m, Picarro Inc., Santa Clara, CA, USA). The analyser's 1 Hz measurements are oversampled with 10 Hz. The T shaped tubing's inlet (length 3.1 m) is placed below the measurement probe at the nose of the aircraft. CH4 analyser and pump are battery driven, while the other components of the MFP are powered by the aircraft's power supply.

The cruise speed of the Sky Arrow ERA is about 85 knts, the maximum total flight path length during research flights is about 200 km. The operating altitudes range from 50 m to 2000 m a.s.l. Flight plans include straight level flights and profile flights. Straight level flights, are performed straight and at constant pressure level and at rather low levels (< 100 m a.g.l.), and are most favourable to determine ecosystem energy and greenhouse-gas fluxes. Measurements from flights at low levels are influenced



mostly by relatively small areas, which are more likely to be homogeneous than larger areas. Spiral profile flights provide vertical profiles of concentrations and meteorological parameters up to 2000 m a.g.l., which give information vertically across the atmospheric boundary layer.

D2.2 - Improved isotopic source signatures of local and regional CH4 emissions - month 36

The ESR started to take bag samples from wetlands in western Sweden for isotopic analyses. Additional samples are planned for 2019 in connection to the flight campaigns in Jämtland (Northern Sweden).

#### D3.4 - Top-down estimates of EU-scale CH<sub>4</sub> emissions – Month 42

In collaboration with the invers modelers Marko Scholze and Guillaume Monteil (LU University), research flights over the county Jämtland in northern Sweden are planned for 2019 with the main goal to measure regional scale CH<sub>4</sub> concentrations. The data will help to generate improved model process parameter estimates, with quantified posterior uncertainties by assimilating observed CH<sub>4</sub> fluxes from wetlands. The study is connected to an ongoing research project at SLU Umeå (Matthias Peichl) on landscape carbon balance of the Krycklan area. The measured data can also be used in the inversion models used within MEMO<sup>2</sup>.

#### 6.4.3 Training and network activities

#### 6.4.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
Biosphere- Atmosphere interaction	28.08.2017- 27.10.2017	Lund University and Hyltemossa ICOS station, Sweden	Extend knowledge about biosphere-atmosphere interactions	15	participating	
Greenhouse gases GHG - biogeochemistry and measurement techniques in ecosystems and landscapes	3.09.2017- 9.09.2017	Örtagaarden and Skogaryd, Sweden	Extend knowledge about biogeochemistry and measurement techniques in ecosystems scale	5	participating	
PhD Introduction Course	9.01.2018- 19.01.2018	Lund University, Sweden	Gain knowledge about phd studies at Lund University	3	participating	
MEMO <sup>2</sup> Summer School	5.02.2018- 16.02.2018	Schoorl, Nederlands	Extend knowledge about methane mobile measurements	6	participating	
Advanced Analysis of Atmosphere-Surface Interactions and Feedbacks	5.03.2018- 16.03.2018	Hyytiälä, Finland	Extend knowledge about atmosphere-surface interactions	5	participating	
MEMO <sup>2</sup> Workshop on Methane Isotopes	17.09.2018- 19.09.2018	RHUL, United Kingdom	Extend knowledge about methane isotopes measurements techniques		participating	
MEMO <sup>2</sup> Workshop on Gaussian plum modelling	9.09.2018- 10.09.2018	UHEI, Germany	Gain knowledge about Gaussian plum modelling		participating	

#### 6.4.3.2 Secondments

Secondmen t	Date (start – end, planned (when))	Locati on	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
RHUL	19.11.2018- 30.11.2018	RHUL, Egham	RHUL	Analysing isotopes composition air from wetlands	Mass spectrometer training	
Afvall Sverige	Single preparation days	Lund	Avfall Sverige	Single preparation days for measurements	Practical skills	



#### 6.4.3.3 Conferences

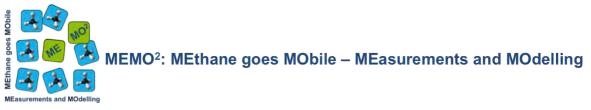
Conference name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation	Authors (main author + co-authors)	Public available (yes / no) / web link
3 <sup>rd</sup> ICOS Science conference	10-13 Sep 2018	Prague, CZ	poster	Using the PicarroG2301-m for airborne eddy covariance measurements of GHG fluxes	Lakomiec, P., Peltola, O., Holst, J., Rinne, J.	no
MEMO annual meeting	22-23 Mar 2018	Zurich, CH	oral + poster	Airborne methane fluxes from wetlands	Lakomiec, P.	

## 6.4.3.4 Measurement / sampling campaigns

Campaign	Date (start – end, planned (when))	Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
Bag sampling	12.10.2018	Skogaryd Mycklemosse, wetlands, 58°21'52.85"N, 12°10'16.71"E.		Samples were taken from automatic chambers system on wetlands	Spatial distribution of isotopic composition	Wetlands/ around 120 from 6 chambers	

#### 6.4.4 Dissemination activities

No scientific publications or other dissemination activities except for the above mentioned conference contributions so far from the ESR.



# 6.5 ESR5 - Characterizing CH<sub>4</sub> emissions in urban environments (Paris)

ESR5

Characterizing CH<sub>4</sub> emissions in urban environments (Paris)

ESR	Sara Defratyka <u>sara.defratyka@gmail.com</u>
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	Jean-Daniel Paris jean-daniel.paris@lsce.ipsl.fr
Co-supervisor	David Lowry D.Lowry@rhul.ac.uk
Non-academic mentor	Rod Robinson Rod.Robinson@npl.co.uk
Official start-end date	01.10.2017 – 30.09.2020

#### 6.5.1 Scientific progress

#### 6.5.1.1 Project introduction and objectives

On a global scale, CH<sub>4</sub> emissions are relatively well estimated. However, characterization of local scale CH<sub>4</sub> sources is still not clear and require further analysis (Dlugokencky et al., 2011). According to the IPCC report, the anthropogenic CH<sub>4</sub> emission is partly associated with urban areas (IPCC, 2006). Urban and sub-urban areas contribute from 30% to 40% anthropogenic greenhouse gas emission and concentrate more than 50% of the global population. However, those areas cover 2% of the Earth's surface. According to the predictions, urban population will double by 2050 (Satterthwaite, 2008; Duren and Miller, 2012). Moreover, an urban ecosystem is a complex case, where many different sources coexist: oil and natural gas networks, heating/cooling system, landfills and waste treatment, wastewater and road transport (Gioli et al., 2012; Townsend-Small et al., 2012; Zazzeri et al., 2017). In case of different cities in the United States like Los Angeles, Boston and Washington, the dominant CH<sub>4</sub> sources are leakages of fossil fuels (Townsend-Small et al., 2012; Jackson et al., 2014; McKain et al., 2015). The similar situation has been observed in Florence, in Italy (Gioli et al., 2012). However, in the case of London, landfills and the waste treatment sector are the major sources of CH<sub>4</sub> (Lowry et al., 2001; Fisher at al., 2006). The significant but not well-determined contribution to global emissions of urban CH<sub>4</sub> requires detailed measurement.

One of the significant urban CH<sub>4</sub> sources can be the IIe-de-France (IDF) region (number of inhabitants:12,14 million, Paris contributing to 18% population). Due to this, methane emissions in IDF region need independent estimations, source by source, from atmospheric measurements. The main approach of this project is focused on field mobile campaigns using cavity ring-down spectrometer (CRDS analyser). Measurements focused on Paris city and suburbs and other anthropogenic sources like gas compressor stations or landfills allow to better understand urban fugitive emissions in a metropolitan area. To achieve this, mobile measurements are made using the trace release method. This method combined with Gaussian models let not only measure concentration but also estimate emission from source (Ars et al., 2017; Mønster et al., 2014). Moreover, during campaigns the isotopic composition of methane is also measured, which can extend knowledge about methane isotopic composition of European anthropogenic sources (Townsend-Small et al., 2012; Zazzeri et al., 2015; Zazzeri et al., 2017). This will allow characterizing finely the spatio-temporal variations of mole fraction and isotopic signature of CH<sub>4</sub> in this region. Measurement data obtained during campaigns will help to determine significant methane sources in the IDF region. Moreover, this data will be used to create an urban scale <sup>13</sup>CH<sub>4</sub> emission model. Thanks to this, it will be also possible to create new urban CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> modelling framework. To get a wider picture of CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> emission in urban scale, field methane measurements may be performed in others urban regions.



#### 6.5.1.2 Project results

#### 6.5.1.2.1 First year

To achieve my PhD goals, the first months of the study were used for instrument testing and first mobile measurement campaigns. Those actions allowed me to get the knowledge and skills connected with GHG measurement, planning and conducting campaigns and also data treatment with R package.

First, an initial test for two isotopic CRDS analysers was conducted. According to ICOS ATC metrology laboratory protocol (Yver Kwok et al., 2015), from 18.10.2017 to 27.11.2017 initial test was conducted on Picarro CFIDS 2067 and CFIDS 2072. Both CRDS analysers are G2201-i models. They can measure the isotopic signature of  $CO_2$  and  $CH_4$ . They also have the possibility to measure  $C_2H_6$  concentrations. The initial test consists of the following tests:

- Internal leak test (internal analyser test)
- Precision test (continuous measurement of the target gas during 25 h)
- Calibration test (2 measurement cycles of three calibration gases with different CO<sub>2</sub>, CH<sub>4</sub> mole fraction and isotopic composition)
- Repeatability, short-term test (10 measurement cycles 30 minutes target, 10 minutes ambient air)
- Reproducibility, long-term test (tested for wet air 17 cycles and dried air 7 cycles 30 minutes target, 600 minutes ambient air)
- Atmospheric pressure test
- Outside temperature test (22 °C first day, 18 °C first night and second day, 32 °C second night and third day, 22 °C third night).

To conduct experiments, natural or dried ambient air, and tanks filled with different target gases were used. For ambient air measurements, an inlet located on the roof of the LSCE building in Gif-sur-Yvette was used. To dry the air, ambient air was passed through a glass trap placed in an ethanol bath kept at about -70 °C by using an immersion cooler. Multi-position valve was used to switch automatically from a gas sample to another one. Examples of results are presented below.

#### **Precision test**

Table 6.5.1: Summary of precision test for CFIDS 2067

Average time	CO <sub>2</sub>		<sup>13</sup> CO <sub>2</sub>		$\delta^{13}CO_2$		CH4		<sup>13</sup> CH <sub>4</sub>		$\delta^{13}CH_4$	
	mean (ppm)	1 σ (ppm)	mean (ppm)	1 σ (ppm)	mean (%)	1σ (%)	mean (ppb)	1 σ (ppb)	mean (ppb)	1 σ (ppb)	mean (%)	1σ (%)
10 sec	399.607	0.088	4.4423	0.0024	-10.709	0.543	1903.64	0.27	20.136	0.058	-50.3	2.7
1 min	399.607	0.049	4.44235	0.0012	-10.709	0.260	1903.64	0.21	20.136	0.035	-50.3	1.7
60 min	399.607	0.032	4.44235	0.0005	-10.709	0.083	1903.64	0.14	20.136	0.029	-50.3	1.3

#### Table 6.5.2: Summary of precision test for CFIDS 2072

	Average time	CO <sub>2</sub>		<sup>13</sup> CO <sub>2</sub>		δ <sup>13</sup> CO <sub>2</sub>		CH4		<sup>13</sup> CH <sub>4</sub>		δ¹³CH₄	
		mean (ppm)	1 σ (ppm)	mean (ppm)	1 σ (ppm)	mean (%)	1σ (%)	mean (ppb)	1 σ (ppb)	mean (ppb)	1 σ (ppb)	mean (%)	1 σ (%)
	10 sec	399.361	0.073	4.3747	0.0022	-11.142	0.504	1902.16	0.27	20.3304	0.0466	-48.84	2.18
	1 min	399.361	0.034	4.4374	0.0010	-11.142	0.229	1902.16	0.20	20.3303	0.0194	-48.84	0.91
	60 min	399.361	0.012	4.4374	0.0003	-11.142	0.054	1902.16	0.14	20.3303	0.0025	-48.84	0.11

#### **Calibration test**

 Table 6.5.3:
 Summary of calibration test methane

Table 6.5.4: Summary of calibration test  $\delta^{13}CH_4$ 

Target gas name	Reference value [ppb]	CFIDS 2067 [ppb]	CFIDS 2072 [ppb]	Target gas name	Reference value [ <sup>½</sup> ]	CFIDS 2067 [ <sup>%</sup> ]	CFIDS 2072 [ <sup>½</sup> ]
D482677	2557.2	2543.64 ± 0.07	2538.07 ± 0.35	D482677	-25.86	-26.46 ± 0.01	-25.33 ± 0.09
D481332	2176.2	2157.72 ± 0.01	2157.75 ± 0.53	D481332	-34.73	-36.44 ± 0.49	-35.71 ± 0.51
D481323	2388.4	2371.58 ± 0.19	2368.90 ± 0.37	D481323	-56.02	-59.55 ± 0.09	-58.5 ± 0.46



#### **Reproducibility long term test**

In Fig. 6.5.1 results from 7 measurement cycles of the target gas are presented. Between 30 minutes of target gas measurement, the dried air was measurement 600 minutes. The whole test lasted 69 hours. For this period, for measurement of target gas, max drift, calculated as difference between the lowest and the highest measured value, for CRDS analysers were 2.22% and 0.39%, for CFID2067 and

CFIDS2072 respectively. The  $\delta^{13}$ CH<sub>4</sub> target values, assessed after dried air measurement, obtained by using both Picarro are compatible within uncertainty with rate (-48.37 ± 0.76) ‰ and (-48.77 ± 0.15) ‰, correspondingly.

The results obtained in the test show that the analysers are precise and stable enough for our purpose. The dried air values obtained by using both Picarro are compatible. Therefore, one of them can be used during dynamic mobile campaigns and second simultaneously during static measurements. Afterwards obtained results can be compared.

The previous measurement (e.g. Rella et al., 2015, Assan et al., 2017) showed significant cross sensitivities between  $C_2H_6$  and  $\delta^{13}CH_4$  measured by CRDS analyser. As ethane is one of the most important components of natural gas, it

can have a big influence for measurement of methane isotopic composition in an urban environment. In January 2018, part of the test described in Assan et al., 2017 was repeated. Ethane was diluted with ambient air with different proportion and then the concentration of  $\delta^{13}$ CH<sub>4</sub> was measured. Obtained results are shown in Fig 6.5.2.

The results obtained in 2015 and 2018 for Picarro CFIDS 2072 within measurement uncertainty are compatible. For CFIDS 2067 bigger difference was observed but under expanded uncertainty they are still compatible.

 $C_2H_6$  measurement is an additional option of Picarro CFIDS 2072 and 2067. One of the ways of calibration is to compare Picarro's results with other calibrated instrument (e. g. gas chromatograph).

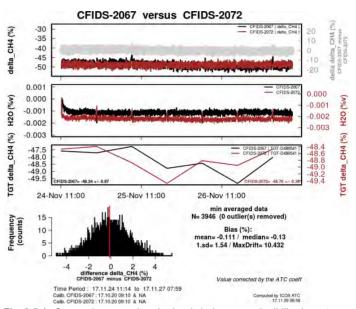
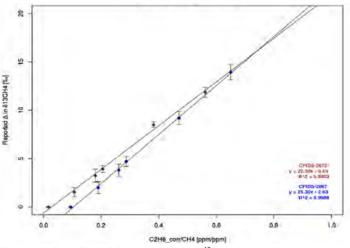
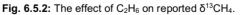
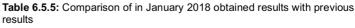


Fig 6.5.1. Comparison results obtained during reproducibility long term test for both instrument, results for  $\delta^{13}CH_4$ , dry air measurement







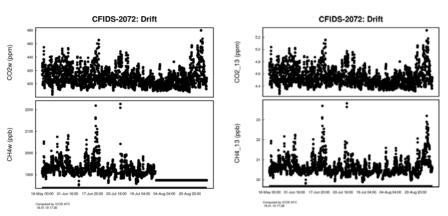
δ <sup>13</sup> CH₄	CFIDS	2072	<b>CFIDS 2067</b>			
Correction	Slope (‰ CH₄/C₂H₀)	Intercept (‰)	Slope (‰ CH₄/C₂H <sub>6</sub> )	Intercept (‰)		
July,15	24 ± 2	0.5 ± 0.6	(/// C114/C2116) -	( /00) -		
Nov,15	23 ± 1	0.2 ± 0.6	23 ± 1	-2.3 ± 0.7		
Jan,18	23 ± 1	-0.6 ± 0.2	25 ± 1	-2.6 ± 0.2		



The calibration factors received using gas chromatograph in 2015 for CFIDS 2072 and CFIDS 2067 are  $(0.505 \pm 0.007)$  and  $(0.52 \pm 0.01)$  (Assan et al., 2017). For tests conducted in January 2018 the equation from Hoheisel 2017 was used to determine the calibration factor. Calculated in this way the calibration factor is a slope of plot C<sub>2</sub>H<sub>6</sub> measured versus C<sub>2</sub>H<sub>6</sub> theoretical with a calculated value of  $(0.508 \pm 0.001)$  for CFIDS 2072 and  $(0.509 \pm 0.100)$  for CFIDS 2067. Subsequently, corrected and calibrated ethane factor, calculated as a slope of linear regression fitted to  $\delta^{13}$ CH<sub>4</sub> versus C<sub>2</sub>H<sub>6</sub> calibrated/CH<sub>4</sub>, is equal (44.5 ± 1.5) and (49.74 ± 0.80) for CFIDS 2072 and CFIDS 2067, respectively. This factor is used to correct the value of isotopic composition measured by CRDS analyser. The factor is used in equation

$$\delta^{13}CH_{4 \text{ corrected}} = \delta^{13}CH_{4 \text{ raw}} - \frac{EC_2H_6 \text{ calib}}{CH_4} + F$$

Where Е is the correction factor calculated as slope and F is the intercept of linear regression fitted to  $\delta^{13}CH_4$  versus C<sub>2</sub>H<sub>6</sub> calibrated/CH<sub>4</sub>. However, if  $\delta^{13}CH_4$  is calibrated to the common scale, the intercept (F value in the equation above) which represent instrumental offset. can be neglected (Assan et al., 2017).



**Fig. 6.5.3:** Results obtained from 16 May to 31 August in Gif-sur-Yvette using CFIDS2072, Left: isotope <sup>12</sup>C, Right: isotope <sup>13</sup>C, Above: CO<sub>2</sub>, Below: CH<sub>4</sub>

During the first months, the data from CFIDS 2072 from 16 May to 31 August 2017 was analysed. During this period instruments was located in Gif-sur-Yvette. This long-term measurement let to see how  $CH_4$  and  $CO_2$  background looks.

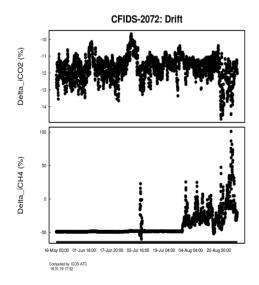


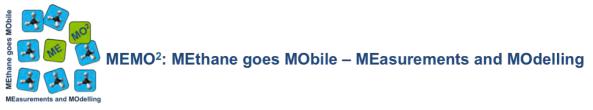
Fig. 6.5.4: Results obtained from 16 May to 31 August in Gif-sur-Yvette using CFIDS2072 Above  $\delta^{13}CO_2$  Below  $\delta^{13}CH_4$ 

Table 6.5.6: Long term measurement in Gif-sur-Yvette from 16 May to 31 August by CFIDS 2072, results for  $CO_2$ 

month	<sup>12</sup> CO <sub>2</sub> [ppm]	<sup>13</sup> CO <sub>2</sub> [ppm]	δ <sup>13</sup> CO <sub>2</sub> [‰]
16.05-30.05	414 ± 13	4.60 ± 0.15	-11.97 ±0.57
01.06-30.06	411 ± 13	4.57 ± 0.14	-11.53 ±0.74
01.07-31.07	406 ± 12	4.51 ± 0.13	-11.52 ±0.58
01.08-31.08	408 ± 16	4.53 ± 0.17	-11.85 ±0.76

Table 6.5.7: Long term measurement in Gif-sur-Yvette from 16 May to 31 August by CFIDS 2072, results for  $CH_4$ 

month	<sup>12</sup> CH₄ [ppb]	<sup>13</sup> CH₄ [ppb]	δ <sup>13</sup> CH₄ [‰]
16.05-30.05	1933 ± 29	20.66 ± 0.31	-49.00 ± 0.19
01.06-30.06	1930 ± 42	$20.62 \pm 0.44$	-48.84 ± 0.39
01.07-31.07	1913 ± 30	20.51 ± 0.33	-46.55 ± 7.48
01.08-31.08	1874 ± 0	20.66 ± 0.47	-18.83 ± 22.46



From May to July results for both CO<sub>2</sub> and CH<sub>4</sub> within uncertainty were stable and compatible. However, in whole August <sup>12</sup>CH<sub>4</sub> was the same without any fluctuation. It shows that this isotope was not measured properly by the instrument. This issue has significant influence on  $\delta^{13}$ CH<sub>4</sub>. Due to this to further analyses from August data cannot be used.

In the first period of PhD not only tests were conducted but also mobile measurements. The first mobile measurement was executed 06.10.2017 on landfill Butte Bellot in Ile de France region. During this campaign, the trace release method was used. Acetylene was used as a gas with known emission. After methane and acetylene measurement, using simple proportion, methane emission was calculated (Ars et al., 2017).

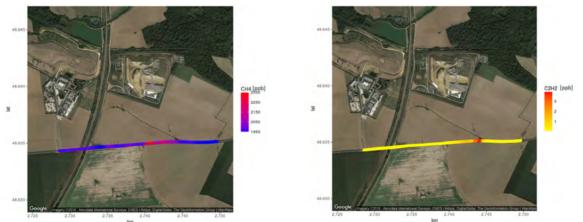
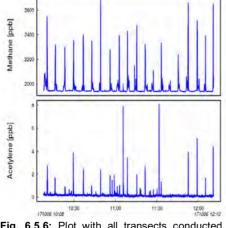


Fig. 6.5.5: Maps with measurements data for fourth transect from Field campaign Butte Bellot 06.10.2017, left: methane measurement right: acetylene measurement



N° Peak	1	2	3	4	5	6	7	8	9	10
Emission s of CH₄ (kg/d)	117 1	120 6	225 8	54 2	98 0	222 0	162 7	94 2	228 0	72 2

Table 6.5.9: Estimated emission using trace release method for peaks 11-20

N° Peak	11	12	13	14	15	16	17	18	19	20
Emission s of CH4 (kg/d)	139 1	73 0	136 8	24 3	172 4	674 8	92 7	57 0	139 4	81 7

Fig. 6.5.6: Plot with all transects conducted during campaign 06.10.2017 in Butte Bellot

After estimation of emission for each peak, daily emission from landfill Butte Bellot was calculated as average from each transect. The daily estimated emission is equal ( $1232 \pm 596$ ) kg/d. On 19.01.2018 preliminary measurement near the Grignon farm and gas compressor in Beynes were conducted Local map with concentration is showed in Fig. 6.5.7. These locations will be measured in more detail in the future.



D5.9 MEMO<sup>2</sup> – Midterm Review Report

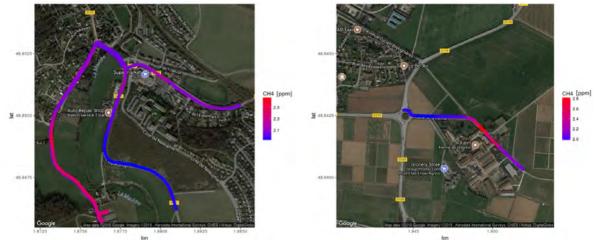


Fig. 6.5.7: Map with concentration, Left: near the gas compressor in Beynes, Right: farm in Grignon

Additionally, measurements were organized during the first MEMO<sup>2</sup> school in February 2018 in the Netherlands. Measurement campaigns were conducted on 08.02., 09.02., and 11.02. During these surveys, the Picarro which measures methane and acetylene was used. The best condition to successfully execute the trace release method was on 12.02. Using acetylene as additional tracer allowed us to estimate the emission from this source. This survey was focused on Kodeiijk gas station. Fig. 6.5.8 shows the maps with measured methane and acetylene concentration from the third transect.

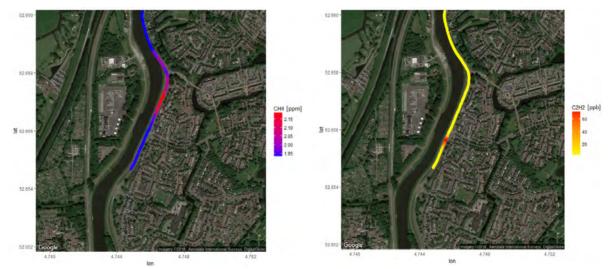
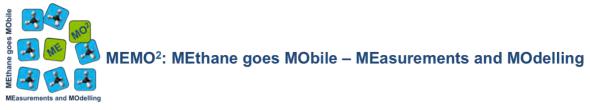


Fig. 6.5.8: Map with concentration Left: methane concentration Right: acetylene concentration

The measuring route was close to the methane source. Moreover, the cylinder with target gas (acetylene) was situated a few meters away from the source. Due to this, the peak from acetylene and methane appear in different places. According to this, the acetylene concentration peaks could not be a perfect representation of methane peaks. This can cause inaccuracy in estimated emission value from the gas station.



Emissions from the gas station were estimated by using the dispersion trace release method (Ars et al., 2017). Another way to estimate the emissions from concentration is using a model. In this case, the Polyphemus model, a Gaussian Eulerian model, was used <u>http://cerea.enpc.fr/polyphemus/</u>. Fig. 6.5.10 shows the concentration simulated by using the Polyphemus model for acetylene and methane. Fig. 6.5.11 and 6.5.12 present modelled and measured peaks for two different transects, for acetylene and methane respectively. Estimated emissions for particular transects obtained using these two methods are presented in Table 6.5.10.

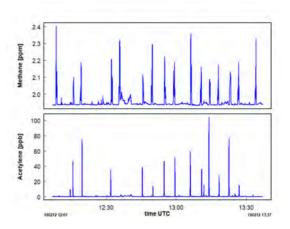
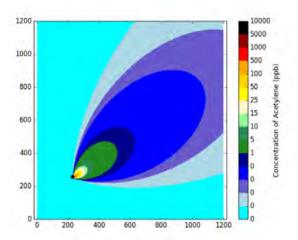


Fig. 6.5.9: Plot with all transects conducted during campaign 11.02.2018 in  $\text{memo}^2$  school



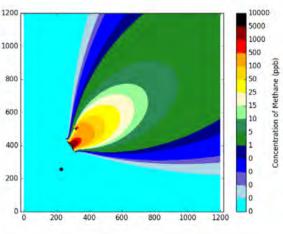


Fig. 6.5.10: Concentration from Polyphemus model, left acetylene, right methane

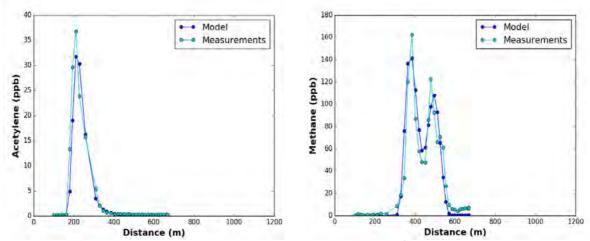


Fig. 6.5.11: Comparison of results obtained from model and measurements for the 1st transect, left: acetylene, right: methane



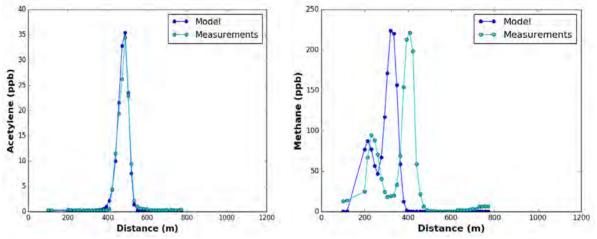


Fig. 6.5.12: Comparison of results obtained from model and measurement for 9th transect left acetylene, right methane

Table 6.5.10: estimated emission for particular transect during measurement on Kodejiik gas station

N° Peak	1	2	3	4	5	6	7	8	9	10
Trace release method	39	192	54	77	50	60	20	57	33	107
Gaussian plume model	71	328	80	105	87	80	22	110	37	216

Finally, estimated emission from gas station was calculated as an average from estimated emission for each transect. Due to difficulties with fitting modelled peaks to measurement, in the case of the 10<sup>th</sup> transect, for this

transect modelled emission is rejected. Estimated emission from the gas station is equal (69  $\pm$  16) kg/day for trace release method and (102  $\pm$  9) kg/day for Polyphemus model. On an annual scale, it gives (25  $\pm$  6) tons / year and (37  $\pm$  3) tons / year, respectively.

#### 6.5.1.2.2 Second year

In March 2018 preliminary measurements in the IIe de France region were organized. Two surveys, focused on a gas compressor station, were conducted. The first one on 05.03.2018 at Limoges-Fourches and the second on 13.03.2018 at Fontenay-Mauvoisn. These prior surveys gave the possibility to find the best weather condition for further mobile measurement according to available infrastructure. Further measurement at these sites and also another one in the IIe de France region will be conducted. Aim is to measure the isotopic composition and estimate emission from each source.

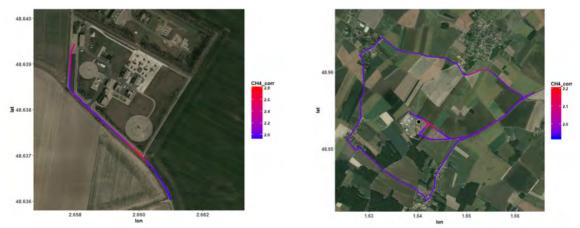
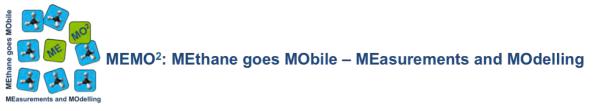


Fig. 6.5.13: Map with concentration near the gas compressor, left: in Limoges-Fourches, right: Fontenay-Mauvoisn



In September 2018, three mobile surveys in the urban area of Paris were conducted. A map with marked tracks is presented in Fig. 6.5.14. First Paris surveys were focused on peripheral Paris area. Moreover, according to GRT gas, which is natural gas transmission operator, the main Paris pipeline is located along the Seine. Due to this, mobile measurements were also conducted in this area. Measured mole fraction and isotopic composition during surveys on 07.09.2018, 25.09.2018 and 26.09.2018 are presented in Fig. 6.5.15 – 6.5.17.

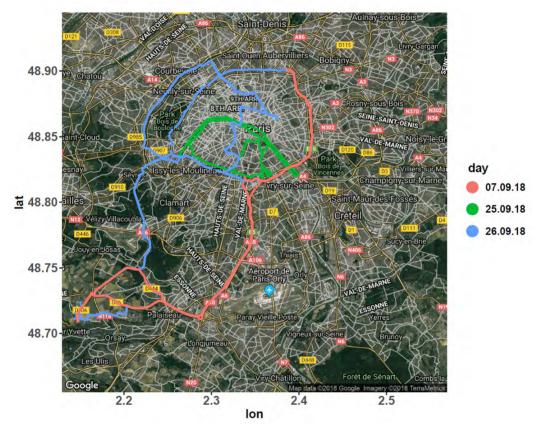


Fig. 6.5.14: Transects of Paris urban area surveys on September 2018

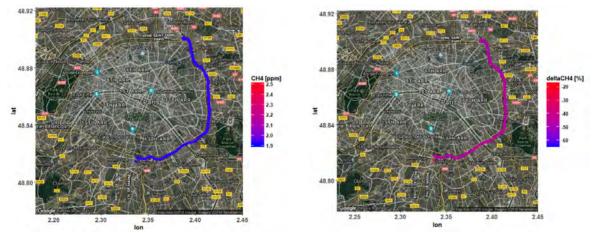


Fig. 6.5.15: Paris urban area survey, 07.09.2018, left: methane concertation, right: methane isotopic composition



D5.9 MEMO<sup>2</sup> – Midterm Review Report

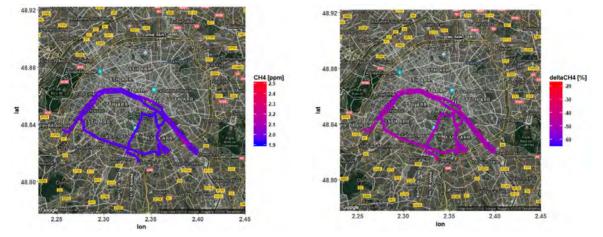


Fig. 6.5.16: Paris urban area survey, 25.09.2018, left: methane concertation, right: methane isotopic composition

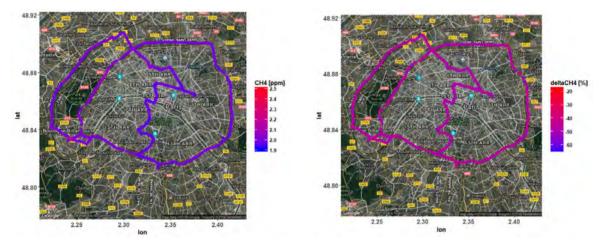


Fig. 6.5.17: Paris urban area survey, 26.09.2018, left: methane concertation, right: methane isotopic composition

Also, in the second year, measurement campaigns were conducted outside of France. The first campaign was CoMet in the Silesia region, Poland, in May / June 2018. The other opportunity to organize mobile measurements out of France was during the secondment at Royal Holloway University of London in June / July 2018.

The CoMet campaign took place between 14.05.2018 and 12.06.2018. Due to being a coal mines region, Silesia is considered as one of the biggest methane sources in Europe. According to EDGAR v4.2 FT2010, annual emission from this region is equal 1.49 Tg/year, where 1.35 Tg/year comes from the mining sector. As part of MEMO<sup>2</sup>, I had also the possibility to participate in this campaign. One of my main jobs was taking care of the FTIR instrument and participate in daily measurements around mines 'shafts. To make this campaign more useful for my PhD thesis (Characterization of CH<sub>4</sub> emission in urban environments), I also conducted night measurements in the urban area in Silesia Region. Moreover, calibration of isotopic Picarro (CFIDS 2072) was conducted by measuring gas from other MEMO<sup>2</sup> groups, from Heidelberg University and AGH University.

On 30<sup>th</sup> May, two calibration gases were used to calibrate Picarro CFIDS 2072. CH<sub>4</sub> concentration in one cylinder was equal to 8.8 ppm and in second 3.0 ppm. To obtain different gas concentration, target gas was diluted with synthetic air. This activity allowed to see instrument response for different value of measured concertation. Isotopic composition calibration requires further works. Obtained values for measured mole fraction are presented in the Fig. 6.5.18 and 6.5.19.



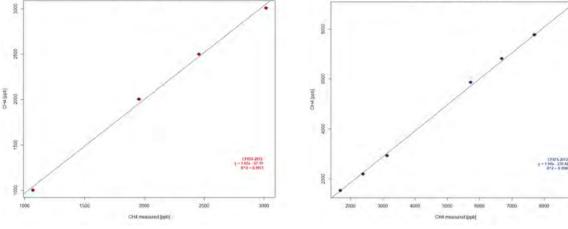


Fig. 6.5.18: Calibration curve calculated for gas with concertation 3010 ppb

Fig. 6.5.19: Calibration curve calculated for gas with concertation 8800 ppb

Implemented calibration shows that the raw value from CFIDS 2072 is in good agreement with the real value of calibration gas. Additionally, at the end of campaign a calibration using different gases was performed. In this case, measured value was equal ( $20775.2 \pm 0.32$ ) ppb where the real value was 2072 ppb and measured isotopic composition was (-49.8 ± 3.2) ‰ where real value was equal -50 ‰. In view of my PhD thesis, the most important part of the campaign was focused on night measurement in the urban area. The night surveys were conducted six times. The measurement area included four cities: Katowice, Chorzów, Bytom and Ruda Śląska. Three times measurements were started from Katowice and three times from Ruda Śląska. This approach allows looking closer for the hourly difference in measured concentration. Fig. 6.5.20 shows maps with marked concentration from every night in the measured urban area.

As a further treatment, based on Silesia inventtory, obtained data will be analysed. Additionally. the measurement of the isotopic composition can help to determine probable sources of methane emission in this area. Moreover, during nighttime surveys, 9 times measurements on highway A1 were conducted.

A map with an observed concentration higher than 4000 ppb during all transects is presented in Fig. 6.5.21.

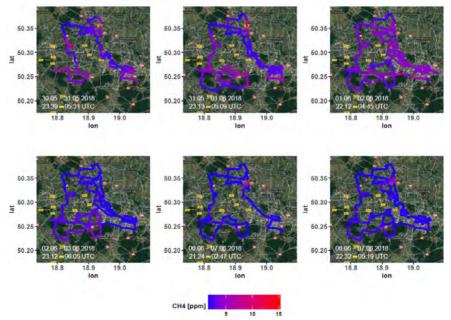


Fig 6.5.20: Maps of CH<sub>4</sub> concentration in cites during night transect CoMet in Silesia Region

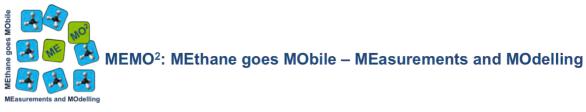
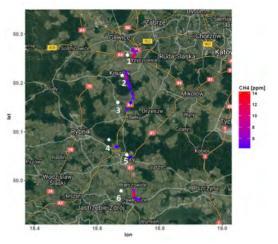


Table6.5.11:ProbablymethanesourceduringmeasurementonhighwayA1duringnight-timemeasurementinCoMet

N° Peak	Probably source	Source latitude [°N]	Source longitude [°E]		
1.	KWK Sosnica shaft VI	50.257099	18.696855		
2.	KWK Knurow	50.215403	18.679976		
3.	KWK Debiensko	50.160034	18.666003 18.630531		
4.	Ponds and wetland	50.083518			
5.	Wastewater Treatment Plant	50.053075	18.694740		
6.	KWK Pniówek shaft IV	49.979892	18.676645		

This allowed determining probably methane source along this road, which is presented in Table 6.5.11. For further treatment, the isotopic composition of observed peaks will be analysed. Secondments in the Royal Holloway University of London were the opportunity to conduct 5 measurement surveys. Four of them were made around London city. The last one was conducted in Devon, on the west south part of United Kingdom and was focused on landfill measurement. The isotopic composition was measured in two ways. First of all, it was measured in situ by Picarro CFIDS 2072 using storage tube called AirCore. This storage tube allows to obtain better time resolution and accuracy for <sup>13</sup>CH<sub>4</sub> and measure in situ isotopic after observation of CH<sub>4</sub> peak (Rella et al., 2015). During measurement air from the CH<sub>4</sub> concentration peaks were collected to the sampling bags. Afterwards, there were measured in the laboratory using IRMS. In this report results from the



**Fig. 6.5.21:** Map of CH<sub>4</sub> concentration above 4 ppm measured observed during all night-time surveys, highway A1 Wisla Mala – Ruda Slaska; numbers are numbers of peaks starting from Ruda Slaska, white points – assigned sources

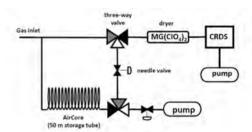


Fig. 6.5.22: Setup for mobile measurement with monitoring/replay mode

survey conducted 27.06.2018 in the urban area are shown. Fig. 6.5.22 shows the scheme of an AirCore. Fig. 6.5.23 shows a comparison of measurement in monitoring and replay mode using AirCore. In Fig. 6.5.24 a Keeling plot and a Miller-Tans plot are presented which allow to calculated isotopic composition (Pataki et al. 2003).

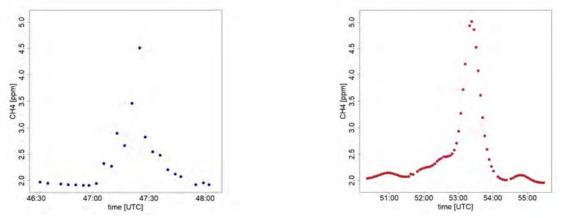


Fig. 6.5.23: Results obtained in replay mode, AirCore number 2 from 27.06.2018, Ashford Water Treatment Plant left: monitoring mode, right: replay mode



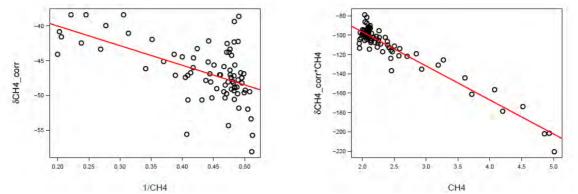


Fig. 6.5.24: Replay mode, AirCore number 2 from 27.06.2018 left: Keeling plot, right: Miller-Tans plot

Based on the map of infrastructure, during the survey on 27.06.2018, there exist two probable sources of methane in measured that day urban area: gas leaks and wastewater industry. According to Zazerri et al.,2017, the value of isotopic composition for natural gas distrubuted in this area is equal  $(-36 \pm 3)$  <sup>//.</sup> and  $(-53 \pm 3)$  <sup>//.</sup> for wastewater sector. Results from survey 27.06.2018 obtained using AirCore storage tube are presented in Table 6.5.12. A map of measured concentrations during urban area survey is shown in Fig. 6.5.25.

Calculated isotopic composition determined that sources of measured that day peaks are connected with natural gas leaks from the pipeline. Even in case of the peak observed close to the water treatment plant, the calculated value of  $\delta^{13}CH_4$ matched to isotopic composition of natural gas. Example of this peak shows a big role of knowing isotopic composition of observed CH<sub>4</sub> concentration peaks in correct identification of methane source. In the case of peak number 4, obtained value of  $\delta^{13}CH_4$  probably is correlated with the mixed isotopic signature from two peaks occur close to each other.

In the next step, results from the other



Fig. 6.5.25: Map of  $CH_4$  concentration measured during urban area survey, 27.06.2018, white points and number indicate stops for changing to replay mode

Table 6.5.12: Probably methane source during urban survey, 27.06.2018

N° Peak	Localization and probably source	CH₄ [ppm]	δ¹³CH₄ (Keeling plot)	δ¹³CH₄ (Miller- Transplot)
1.	Laleham, gas leak	2.60	-38.8 ± 4.2	-38.7 ± 4.0
2.	Ashford, Water Treatment Plant	5.01	-34.4 ± 2.0	-35.2 ± 1.2
3.	Feltham	6.71	-34.43 ± 1.1	-34.7 ± 0.6
4.	Stanwell	3.71	-43.4 ± 7	-44.2 ± 4.7
5.	Stanwell	3.84	-33.3 ± 2.3	-32.5 ± 1.6
6	Egham	4.98	-33.4 ± 3.2	-36.0 ± 2.0

surveys will be analysed. Results obtained using CRDS analyser will be compared with results obtained by IRMS. Secondments in RHUL were also the opportunity to compare results from the stationary measurement made by CRDs and IRMS. One of the activity was measurement of diluted sample from landfill or natural gas. Dilution was made with N<sub>2</sub> or mixed N<sub>2</sub> with O<sub>2</sub>. Obtained results are presented in Fig. 6.5.26 and in Table 6.5.13.



D5.9 MEMO<sup>2</sup> – Midterm Review Report

According to data from table and plot, a shift in value for  $\delta^{13}CH_4$  was Isotopic observed. composition measured by CRDS is lower about 5 compare to IRMS. One of the reasons of this situation can be fact, that in measured gas CO<sub>2</sub> value was about 20 ppm and CRDS analyser is dedicated for measurement of ambient air which in CO<sub>2</sub> concertation is about 400 ppm. The other possible reason is nonlinear response of CRDS for measurement of isotopic composition. This issue requires further analysis. However, this activity was one of the preliminary steps to making calibration gas for MEMO<sup>2</sup> students by Royall Holloway University of London and Utrecht University.

Additionally, 3 nightly continuously measurements from the common inlet by CRDS and IRMS were conducted. This activity allowed to compare results from the continuous

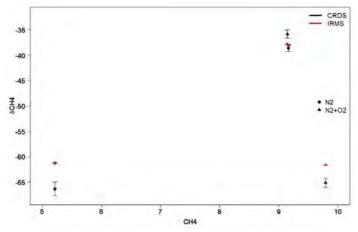
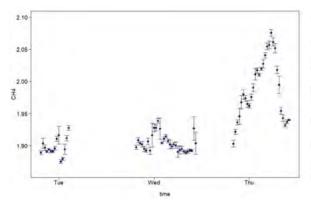


Fig. 6.5.26: Comparison of isotopic composition measured by IRMS and CRDS, x axis CH4 [ppm], y axis  $\delta^{13}CH_4$  [ $\not\!\!\!\!/$ ]

Table 6.5.13: Comparison of isotopic composition  $^{\delta 13}CH_4$  [  $\not\!$ ] measured by IRMS and CRDS

source of gas	dilution	IRMS	SD	CRDS	SD
landfill	N2	-61.24	0.03	-66.38	1.37
landfill	80%N <sub>2</sub> +20%O <sub>2</sub>	-61.63	0.04	-65.16	0.84
geochem_gas	N <sub>2</sub>	-38.04	0.01	-38.59	0.72
geochem_gas	80%N <sub>2</sub> +20%O <sub>2</sub>	-37.83	0.04	-35.84	0.81

measurement obtained by two different instruments. CH<sub>4</sub> concentration obtained by CRDS average by 20 minutes is presented in Fig. 6.5.27. In Fig. 6.5.28 a comparison of the calculated values of the isotopic composition is shown.



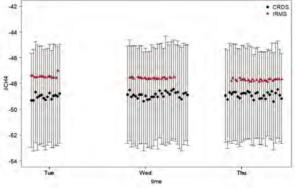


Fig. 6.5.27:  $\text{CH}_4$  concentration measured by CRDS analyser during continuous measurements of ambient air, 20 minutes average

Fig. 6.5.28:  $\delta^{13}CH_4$  measured by CRDS and IRMS during continuous measurement of ambient air, for CRDS 20 minutes average

The  $\delta^{13}$ CH<sub>4</sub> value measured by CRDS analyser is lower about 2 than obtained by IRMS. These results confirm nonlinear tendency of CRDS analyser response. However, this hypothesis requires further measurement.

#### 6.5.1.3 Future plans and expected results

The main goal for the next reporting period is connected with mobile measurement in Paris and the IIe de France region. This time will be destined to collect enough data to create database which allow making PhD thesis and articles. Further work with Polyphemus model is planned Additionally, in the



next reporting period secondment in National Physical Laboratory will be organized. During the next reporting period, I plan to participate at least two conferences: EGU General Assembly 2019 in Vienna and NCGG8 in Amsterdam.

#### 6.5.1.4 Collaborations (internal / external)

Due to common measurements, data treatment and sharing during MEMO<sup>2</sup> school, real collaboration with other MEMO<sup>2</sup> students have started during 1<sup>st</sup> MEMO<sup>2</sup> school. Additionally, I participated in CoMet campaign in Silesia region in Poland. This project was conducted by AGH University in collaboration with German Aerospace Center (DLR). During the 1<sup>st</sup> period secondments in the Royal Holloway University of London were made.

#### 6.5.1.5 Risks and difficulties

After overcoming the initial administrative issues, probable risks and difficulties are more connected with the experimental aspect of measurements. After MEMO<sup>2</sup> school, one of measurement instrument was broken and could not be used for a few weeks. After that, the other one was broken and it was in repair until the end of July. This situation can also happen in the future. Unsuitable weather condition can also delay work plan. Difficulties to get access to survey sites are the other probable issues which can influence for project realization.

#### 6.5.2 Deliverables

**D1.1** - Report on harmonized method for mobile CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub> (month 24)

Campaigns conducted during MEMO<sup>2</sup> school allow verifying compatibility of measuring instruments used in the different MEMO<sup>2</sup> institutions. Both stationary and mobile measurements were arranged. It was the first step to harmonize method for mobile CH<sub>4</sub> and <sup>13</sup>CH<sub>4</sub>. The next occasions to harmonize method were during CoMet in Silesia and during secondments in RHUL. Additionally, I took part in the discussion about data format from mobile campaigns during MEMO<sup>2</sup> school and plume modelling workshop in Heidelberg.

**D1.5** - Improved emission factors for different source categories from mobile measurement (month 18) Get the knowledge and first steps in using Polyphemus model, participation in plume modelling workshop

D2.1 - Isotopic measurements linked to common scale (month 18)

Based on literature (Rella et al.2015 and Hoheisel 2017) build setup to storage air in the tube so-called AirCore which gives the possibility to conduct mobile measurement in monitoring and replay mode, first using this setup during secondments and applied Keeling and Miller-Tans plot to obtained data. Measurement of diluted gas from natural gas distributor and landfill by CRDS and IRMS during secondments in RHUL.

**D2.3** - Publications on the use of isotopes for CH<sub>4</sub> source attribution in urban / industrial regions (month 36)

Due to the early stage of the project no tangible progress has been made for this deliverable.

**D2.5** - Report providing isotopic maps at grid scale from inventories and atmospheric measurements (month 42)

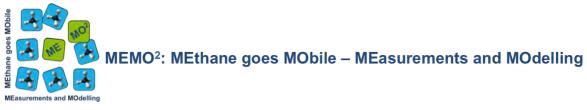
Due to the early stage of the project no tangible progress has been made for this deliverable.

D3.2 - Improved bottom – up European emissions (month 30)

First measurements in IIe de France region and in Paris urban area were conducted. Trace release method and Polyphemus model was used to estimate emission from part of measurement sites.

**D3.3** - Forward modelling simulations of CH<sub>4</sub> and isotopologues (month 30)

Due to the early stage of the project no tangible progress has been made for this deliverable.



# 6.5.3 Training and network activities

## 6.5.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
course	7.11.2017- 30.01.2018	Universite Paris-Sud	Practical Data Analysis	30 h	participating	
Language course	Whole time since november2017	LSCE	French course		Participating	
1 <sup>st</sup> MEMO2 school	05.02.2018 - 16.02.2018		physics and chemistry connected with greenhouses gases	160 h/ 6 ECTS	Participating	
Isotopic workshop	17.09.2018- 19.09.2018	RHUL	Measurement and data treatment of isotopic composition		Participating	
Plume modeling workshop	09.10.2018- 10.10.2018	UH	Plume modeling		Participating	
2 <sup>nd</sup> MEMO2 school	18.02.2018- 22.02.2018	LSCE	Methane and society		Participating	

#### 6.5.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
	17.06.2018- 13.07.2018	Egham	RHUL	campaign for isotope measurements of CRDS (UVSQ) and IRMS (RHUL)/ comparison of measurements done by CRDs and IRMS	Build and using storage tube AirCore, calculation isotopic composition from in situ measurements	Plume mapping and source isotopic comparison, in the future - further comparison of obtained results

#### 6.5.3.3 Conferences

Conference name	Date (start – end, planned (when))	Location	Presentation (oral/poster)	Title of presentation	Authors (main author + co-authors)	Public available (yes / no) / web link
3 <sup>rd</sup> ICOS science conference	11.09.2018- 13.09.2018	Prague	poster	Mobile measurement of CH₄ isotopes in urban, mining and industrial environments	Sara Defratyka, Camille Yver Kwok, Arjan Hensen, Jaroslaw Necki, Dave Lowry, Jean-Daniel Paris, Pawel Jagoda, Philippe Bousquet	no

# 6.5.3.4 Measurement / sampling campaigns

Campaign	Date (start – end, planned (when))	Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
landfill	06.102017	Butte- Bellot, France	LSCE	Mobile measurement using acetylene as tracer	Estimation of emission from source	Mobile measurement with 20 transects	Estimated emission from landfill
MEMO <sup>2</sup> school	05.02.2018 - 16.02.2018	Netherlan ds		Continuous measurement from common inlet by CRDS analyzers from different PhD students, 3 mobile campaigns	Comparison of obtained value, first attempt to harmonize different methods	3 days of mobile surveys, one with using acetylene, continuously measurement 05.02-09.02	Estimated emission from landfill, further work with Polyphemus model on obtained data



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

Gas compressor station	19.01.2018	Beynes, France	LSCE	First attempt to measure site in Ile de France	Primary survey to optimize condition for further measurement	Mobile measurement	No further plans
Gas compressor station	05.03.2018	Limoges- Fourches, France	LSCE	First attempt to measure site in Ile de France	Primary survey to optimize condition for further measurement	Mobile measurement	No further plans
Gas compressor station	13.03.2018	Fontenay- Mauvoisin , France	LSCE	First attempt to measure site in Ile de France	Primary survey to optimize condition for further measurement	Mobile measurement	No further plans
CoMet	23.05.2018- 10.06.2018	Sielsia, Poland	DLR, AGH	Mobile measurement in mining area: around mining shafts and in urban area	Better understanding of emission from mining industry in Poland, urban area source mapping	10 days of mobile measurement around mining shafts, 6 nighttime measurement of urban area	Urban source mapping, future plans – find probably emission source in Silesia urban area
secondments	17.06.2018- 13.07.2018	South of United Kingdom	RHUL	Mobile campaigns with in situ measurement of isotopic composition, nighttime measurement from common inlet by CRDS and IRMS	Calculated isotopic composition of different source, comparison of results obtained by CRDS and IRMS	5 measurement campaign,4 with using storage tube, 3 nighttime measurement,	Plume mapping and source isotopic comparison, in the future - further comparison of obtained results
Paris urban area	07.09.2018, 25.09.2018, 26.09.2018	Paris	LSCE	Mobile measurement in Paris urban area	Source mapping in Paris urban area	3 measurement days	Not significant sources detected

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.5.4 Dissemination activities

Dissemination activity	Name	Date	Location	Type of audience	Size of audience
Blog note on MEMO <sup>2</sup> website	MEMO <sup>2</sup> at CoMet	25.06.2018	https://h2020- memo2.eu/category/blog/	general	

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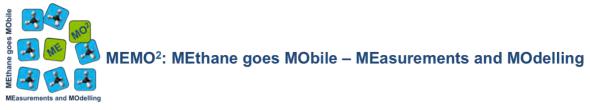
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# 6.6 ESR6 - Mid-infrared laser spectroscopy for three dimensional CH4 mapping by unmanned aerial vehicles (UAV)

#### ESR6

Mid-infrared laser spectroscopy for three dimensional CH<sub>4</sub> mapping by unmanned aerial vehicles (UAV)

ESR	Badrudin Stanicki, badrudin.stanicki@empa.ch					
Supervisor	Lukas Emmenegger, lukas.emmenegger@empa.ch					
Co-supervisor	Huilin Chen, <u>huilin.chen@rug.nl</u>					
Non-academic mentor	Michael Strogies, michael.strogies@uba.de					
Official start-end date	01.09.2017 – 31.12.2018					

#### 6.6.1 Scientific progress

#### 6.6.1.1 Project introduction and objectives

The project aims to develop a rugged and lightweight mid-IR spectrometer based on quantum cascade lasers (QCL) for measuring methane aboard of a mobile platform.

The system will be used on an unmanned aerial system (UAS) for 3D mapping of CH<sub>4</sub> plumes as a unique tool for dynamically detecting local-scale emissions of methane sources. In combination with inverse transport modelling, this should allow drawing conclusions about the location and strength of those methane sources. Applications may include field experiments on natural and anthropogenic sources in collaborations within MEMO<sup>2</sup>, and studies of surface water emissions in collaboration with the Swiss Federal Institute of Aquatic Science and Technology (EAWAG).

#### 6.6.1.2 Project results

#### 6.6.1.2.1 First year

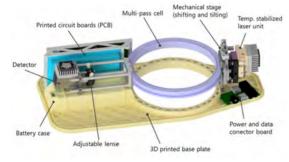


Fig. 6.6.1: CAD model of the laser spectrometer

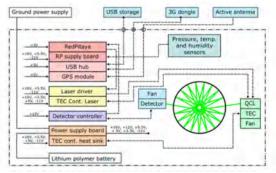
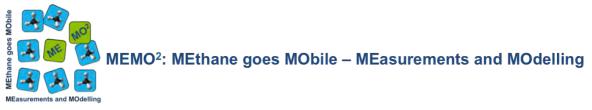


Fig. 6.6.2: Block diagram of the electronical and optical components

A specially designed circular, segmented optical multipass cell was manufactured and characterised with a laboratory bench measurement setup. Furthermore, the laser, the detector, the thermoelectric cooling system and the other main components were successfully tested. The results showed a very good overall performance with a measurement precision of around 1 ppb.

Based on these first results, a mechanically rugged and lightweight mobile measurement device was designed, which is shown in Fig. 6.6.1. The construction excels with a very low overall weight below 2 kg and compact dimensions, and it is thus suitable for the foreseen application on board of a UAV. It includes a new design of the laser temperature stabilization and improved heat insulation. Detailed technical drawings were derived from this conceptual design, and the manufacturing process was planned in collaboration with the in-house mechanics workshop.



Furthermore, the required electronical components such as power supply, laser driver, TEC controller, and data acquisition boards were developed by the Laboratory's electronics engineer. A scheme of the electronic components, needed for autonomous operation, can be seen in Fig. 6.6.2.

#### 6.6.1.2.2 Second year

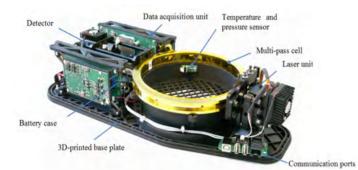


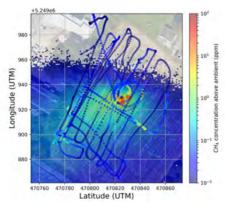
Fig. 6.6.3: Photograph of the spectrometer for methane measurements

The optical setup and the printed circuits were completed and integrated into a lightweight 3D printed housing, as shown in Fig. 6.6.3 and 6.6.4. All components were successfully tested, and a high stability of the laser frequency was demonstrated. Additional software was implemented for collecting and logging the temperature, the relative humidity, the pressure and the GPS position. The device was tested in a purpose-built measurement chamber under controlled temperature conditions. The achieved precision lies well below 1 ppb, as can be seen in the Allan plot in Fig. 6.6.5.

A Matrice 600 unmanned aerial system (UAS) from DJI was purchased (Fig. 6.6.4) and training was completed on UAS operation and safety. A special holder was constructed to

carrv

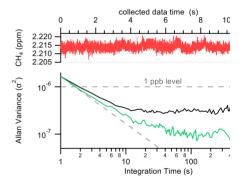
the



**Fig. 6.6.6:** Simulated footprint of a methane source (10 l/min, simulated by the Graz Lagrangian Model, GRAL) and the concentrations obtained by the drone measurements (dots). The colour indicates the increase of the local CH<sub>4</sub> concentration with respect to the ambient concentration (all values of  $10^{-2}$  ppm and below are shown in the same colour).



Fig. 6.6.4: DJI Matrice 600 drone equipped with the methane sensor



**Fig. 6.6.5:** Allan plot of the methane concentration measured in the temperature stabilized test chamber.

methane sensor on the UAS while decupling it from vibrations produced by the drone.

Various test flights were conducted, including flying next to an ambient air measurement site to determine the stability of the retrieved methane concentration, as well as flying near an artificial methane source. Figure 6 shows the result of a measurement flight above an artificial source, which was emitting methane with a rate of 10 l/min.

#### 6.6.1.3 Future plans and expected results

The precision of the spectrometer is already now beyond the targeted performance. The limiting factors will, therefore, be slow drifts and the absolute accuracy. These aspects are strongly related to changes in ambient temperature. This aspect will be investigated in detail through further tests in a climate chamber. Additional test flights for characterizing the performance of the overall



system are planned. The final specifications will then be the starting point for field experiments targeting specific methane sources. This part of the work will be done in close collaboration with modelling activities and may include a field validation with an artificial methane source.

#### 6.6.1.4 Collaborations (internal / external)

Internal collaborations are planned with K. Vinkovic (PhD student RUG) and Prof. H. Chen in Groningen on common measurement campaigns in the Netherlands. Furthermore, collaborations are planned with LU (ESR 4) for wetland related methane emissions and with the ETHZ Aquatic Chemistry group of Prof. B. Wehrli (EAWAG/ETHZ) on aquatic systems.

#### 6.6.1.5 Risks and difficulties

The instrumental developments are currently advancing as planned. We anticipate challenges mainly in three fields (i) long-term stability and accuracy of the measurements, (ii) mixing of the air masses due to the UAS' propellers and corresponding difficulties in data interpretation, and (iii) combination of inverse modelling and measurements at the available time and spacial resolution.

The current PhD student (Badrudin Stanicki) is leaving Empa by the end of 2018. The position has been advertised, and it is foreseen to continue the activities within MEMO2 as planned. Due to these circumstances it was decided to postpone the first secondment as described in chapter 3.2. Depending on the success in recruiting a successor, the schedule for the UAV based activities may have to be adapted.

#### 6.6.2 Deliverables

**D1.1** - Lightweight CH<sub>4</sub> sensor and AirCore developed and deployed on UAV (month 24)

The lightweight methane sensor has been developed and its functionality was demonstrated successfully in laboratory experiments as well as aboard of a UAV. The achieved precision is well within the requirements. The stability regarding changes of the surrounding temperature might be increased trough further improvements of the design of the device.

D1.2 - Report/publication on CH4 emissions from wetland and lakes in Sweden (month 24)

This deliverable is pending because it requires the development and characterization of the instrument as described in D1.1.

**D1.4** - Improved emission factors for different source categories from mobile measurements (month 24) This deliverable is pending because it requires the development and characterization of the instrument as described in D1.1.

#### 6.6.3 Training and network activities

#### 6.6.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
Laboratory seminar	13.09.2017 – 14.09.2017	Sigriswil	Team building event	-		
Aquatic Chemistry group retreat	29.11.2017 – 31.11. 2017	Waltensburg	networking activities and general PhD training course, workshop on scientific writing	-	Short presentation of project	
Drone training (theory)	07.12.2017	Empa, Dübendorf	Theoretical introduction for using a UAS	-		Realised by Koller Engineering
Drone training (practice)	15.12.2017	Zwillikon	Practical training on how to fly a drone	-		Realised by Koller Engineering
MEMO <sup>2</sup> School	05.02.2018 – 16.02.2018	Schoorl, Netherlands	Methane measurements and modeling	6	preparation of a poster and the analyses of data	



MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

Annual Memo2	22.03.2018	Empa, Dübendorf	Networking event	Presentation poster	n,				
PhD congress	06.04.2018	Institute of Biogeochemistry and Pollution Dynamics at ETH Zurich	Phd congress for networking	Poster					
Drone training	07.06.2018	Empa, Dübendorf	Practical training with the Matrice 600 drone		Realised by Koller Engineering				

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.6.3.2 Secondments

No secondments have been done so far. The contract of the current ESR 6 is ending by the end of 2018 and the position has been advertised, Due to these circumstances, it was decided to postpone the first secondment and adapt the schedule when a new ESR 6 has been hired.

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
Secondment 1	Mid 2019 (planned)	Groningen	RUG	UAS based methane measurements with QCL spectrometer and aircore	Characterisation of natural and anthropogenic methane sources	
Secondment 2	End 2019 (planned)	Lund	LU	Mobile methane measurements aboard of an airplane	Emissions from wetland and lakes in Sweden	

#### 6.6.3.3 Conferences

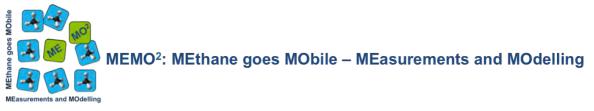
Conference name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation	Authors (main author + co-authors)	Public available (yes / no) / web link
Flair conference	12.09.2018	Assisi, Italy	oral	A compact QCL absorption spectrometer for mobile, high-precision methane measurements aboard drones	Badrudin Stanicki, <u>Béla</u> <u>Tuzson</u> , Liu Chang, Manuel Graf, Philipp Scheidegger, Herbert Looser and Lukas Emmenegger	no

#### 6.6.3.4 Measurement / sampling campaigns

Campaign	Date (start – end, planned (when))	Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
MEMO <sup>2</sup> School	05.02.2018 – 16.02.2018	Schoorl, Netherlands	RUG	Mobile measurements with cars, methane release experiment	For training purpose		
Test Flight 1	2018.08.10	Empa, Dübendorf	Empa	Test flight with mobile methane spectrometer	First test for the assessment of measurement characteristics during flight conditions.	CH4 concentration with 2Hz resolution, additional: H2O conc., T, p, GPS pos.	Results can be used for quantifying and improving the stability of the spectrometer, further test flights are planned
Test Flight 2	2018.10.10	Empa, Dübendorf	Empa	Test flight with mobile methane spectrometer	Testing a slightly modified setup and the observation of artificial methane release.	CH4 concentration with 2Hz resolution, additional: H2O conc., T, p, GPS pos.	Results can be used for quantifying and improving the stability of the spectrometer, further test flights are planned

#### 6.6.4 Dissemination activities

No scientific publications or other dissemination activities so far from the ESR.



# 6.7 ESR7 - CH<sub>4</sub> from waste: constraints on captured and fugitive emissions from isotopic analysis

#### ESR7

 $\ensuremath{\mathsf{CH}_4}\xspace$  from waste: constraints on captured and fugitive emissions from isotopic analysis

ESR	Semra Bakkaloglu ( <u>semra.bakkaloglu@rhul.ac.uk</u> )			
Supervisor	Dave Lowry ( <u>d.lowry@rhul.ac.uk</u> )			
Co-supervisor	Huilin Chen ( <u>huilin.chen@rug.nl</u> )			
Non-academic mentor	Stuart Davies ( <u>SDavies@viridor.co.uk</u> )			
Official start-end date	08/01/2018- 31/12/2020			

#### 6.7.1 Scientific progress

#### 6.7.1.1 Project introduction and objectives

As mitigation of climate change is a key scientific and societal challenge, CH<sub>4</sub> emissions are a major contributor to Europe's global warming impact and emissions are not well quantified yet. There are significant discrepancies between official inventories of emissions and estimates derived from direct atmospheric measurement. Effective emission reduction can only be achieved if sources are properly quantified, and mitigation efforts are verified. Globally, human activities produce over 60% of total CH<sub>4</sub> emissions, with 22% of emissions from the energy sector and 10% from the waste sector.

Methane from waste is dominantly of biogenic origin and can vary with temperature and production process, which results in variation of emissions with time of day and time of year. In addition, the waste sites now commonly produce and combust this biogas, and emissions from each component can be identified by analysing the methane isotopic composition, as different source types are characterized clearly by distinct  $\delta^{13}$ C signatures. For landfill sites in particular a percentage of the methane produced is oxidised by soil cap or oxygen in upper-levels of less-compacted waste and this results in a different isotopic signature to non-oxidised methane in the gas extraction system.

This project includes measurement and modelling of CH<sub>4</sub> plumes originating from waste processing sites at different times of year under different meteorological conditions, using mobile equipment. Moreover, off-site plume samples will be collected for isotopic characterization. Selected sites will be studied in more detail using isotopic characterization of emission plumes from individual site components such as active and remediated landfill cells, gas combustion plants, anaerobic digestion cells. Gas wells will be sampled to characterize unoxidized CH<sub>4</sub> to aid in the understanding of on-site oxidation rates.

The main objectives of this project are:

- To measure plumes of methane downwind of waste processing sites at different times of year under different meteorological conditions using mobile instruments and model the emissions.
- To collect off-site plume samples for isotopic characterization from individual site components such as sewage emission, active and remediated landfill cells, biogas plants for selected sites.
- To sample methane plumes and gas wells on sites of project partner Viridor, and use the isotopic difference to assess the role of oxidation of fugitive emission.

#### 6.7.1.2 Project results

#### 6.7.1.2.1 First year

The first set of results were from the MEMO<sup>2</sup> school campaign during February 2018. Collected air samples were analysed for isotopic analysis at RHUL. Calculated  $\delta^{13}$ C signatures for the sources investigated are given in Table 6.7.1. Moreover, ESR7 worked on comparison of different mobile methane sensors and vehicle speed impact on plume shape during the MEMO<sup>2</sup> school

Source	Typical δ <sup>13</sup> C Signature (‰)
Alkmaar Landfill	-52.8
Biogas Production	-58.0
Agriculture (Farming)	-66.8
NW Alkmaar Gas Plant	-31.5

Table 6.7.1: NL campaign results



D5.9 MEMO<sup>2</sup> – Midterm Review Report

campaign as given in the figures below. As seen in Fig. 6.7.1 and 6.7.2 different mobile sensors have different time delay due to length of sample line, instrument flow rate and measurement frequency.

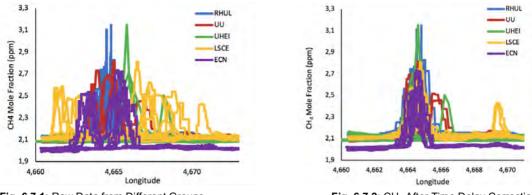


Fig. 6.7.1: Raw Data from Different Groups



Moreover, as seen in Fig. 6.7.3, when the car speed is low, the plume shape is narrower with steep sides. The optimum car speed to be used in campaigns should be determined by all groups, but needs to account for the flow of traffic when roads are busy. Furthermore, the measurements produced by the smaller LGR (Los Gatos Research) portable instrument has been compared with the larger, high precision Picarro instrument in Fig. 6.7.4.

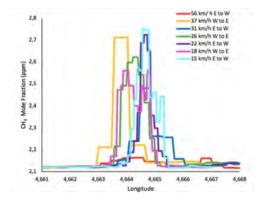
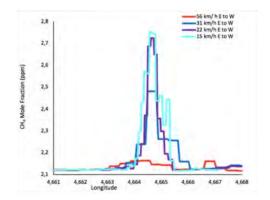


Fig. 6.7.3: Car Speed Impact on Plume Shape

As seen in Fig. 6.7.4 the LGR baseline mole fraction shifts during measurement due to temperature and calibration differences. The LGR is more susceptible to temperature changes, because it is not temperature controlled. The LGR measures every second and had a 20 second time delay with respect to Picarro due to differences in the computer time during the campaign. To conclude, the LGR measurements should be corrected based on temperature and calibration standards.



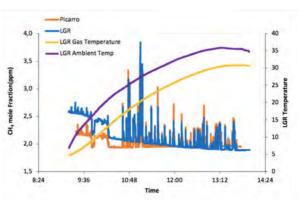


Fig. 6.7.4: LGR and Picarro Mobile Sensor Comparison



#### 6.7.1.2.2 Second year

ESR7 has been part of seven measurement and sampling campaigns during summer 2018. The collected air bag samples have measured by GC-IRMS to characterize isotopic signature. The figures are listed below.

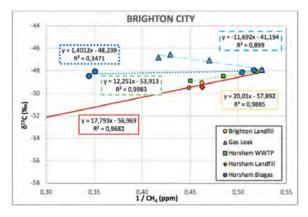


Fig. 6.7.5: Isotopic Signature of Methane from Brighton Campaign

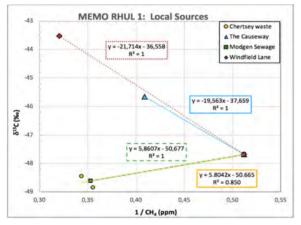


Fig. 6.7.7: Isotopic Signature of Methane from RHUL Local Sources

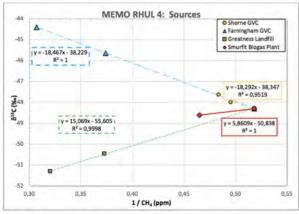


Fig. 6.7.9: Isotopic Signature of Methane from Isle of Grain/Kent Campaign

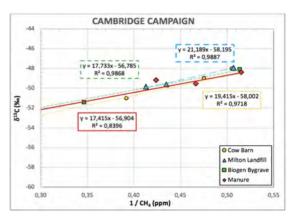


Fig. 6.7.6: Isotopic Signature of Methane from Cambridge Campaign

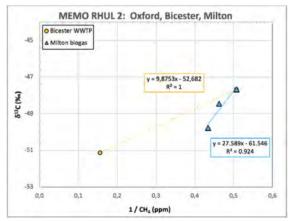
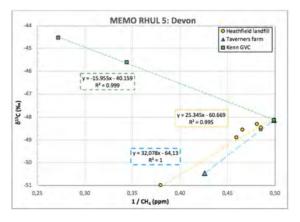


Fig. 6.7.8: Isotopic Signature of Methane from Oxford, Bicester and Milton Regions



 $\label{eq:Fig. 6.7.10: Isotopic Signature of Methane from Devon Campaign$ 



The MEMO<sup>2</sup> RHUL 1, 2, 4, 5 campaigns have been performed with together ESR5, ESR8 and ESR9. The results were same and shared with others.

Isotopic signatures of waste sources in the UK measured so far are summarised in Fig. 6.7.11. Due to different atmospheric background conditions on different days, the regression lines do not intersect at the same background point.

During campaigns in summer 2018, various methane sources have been identified; farms, landfills, WWTPs and biogas plants. Once identified the plumes have been sampled by filling Tedlar bags for later carbon isotopic analysis by high-precision IRMS.

Average isotopic signatures are (-52  $\pm$  0.8) % for WWTP, (-56  $\pm$  3) % for biogas plants, (-58  $\pm$  0.8) %

for landfills,  $(-61 \pm 2)$  ‰ for agriculture, and  $(-57 \pm 1)$ ‰ for manure (Table 6.7.2). Moreover, there is 10 ‰ spread between biogas plants. The reason might be biomass type and process differences between biogas plants.

Conclusions:

Recent studies in the UK have identified source signatures of methane. Agriculture values are heavier than previous studies, possibly linked to a larger manure to

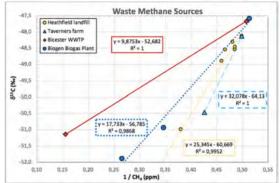


Fig. 6.7.11: Isotopic Signature of Waste Methane Sources in the UK

Table 6.7.3: Standard Gas Cylinder Preparation Results (see D2.1 report for more details)

Sources	Concentration (ppm)	Peak (nA)	Height	Date	δ <sup>13</sup> C corrected (‰)	STD
Landfill Gas	1.770	5.1		27/07/2018	-59.68	0.00
	1.770	5.2		28/07/2018	-59.79	0.02
	1.770	5.5		31/07/2018	-59.71	0.09
	10.14	7.2		31/07/2018	-60.89	0.02
	10.14	10.6		28/08/2018	-60.94	0.02
	10.14	5.6		29/08/2018	-60.99	0.06
	10.14	9.2		31/08/2018	-60.90	0.03
RHUL Gas	1.967	5.6		27/07/2018	-39.65	0.01
	1.967	5.8		28/07/2018	-39.67	0.04
	1.967	5.9		28/07/2018	-39.61	0.04
	10.22	11.6		23/07/2018	-38.22	0.05
	10.22	10.7		26/07/2018	-38.21	0.04
	10.22	11.2		31/07/2018	-38.22	0.05
	10.22	5.7		31/8/2018	-38.20	0.02

UK CH4

Sources

breath ratio which gives a more enriched <sup>13</sup>C mix.

- Landfill average values are similar to previous studies.
- WWTP isotopic signatures have a narrow range.
- Biogas emissions have a varied isotopic signature.
- Biogas isotopic signatures depend on waste type.

ESR 7 has also helped in preparation and analysis of standard gas tanks for inter-comparison (see D2.1 for more details). The results are given in Table 6.7.3.

#### 6.7.1.3 Future plans and expected results

ESR 7 future plans are listed below to enhance methane inventory from waste:

lysis of Agriculture -66 -61 ± 2

	(cows)		
	Agriculture (manure)	-58	-57 ± 1
	Landfills	-58	$\textbf{-57.8} \pm \textbf{0.8}$
	Waste Water Treatment	-53	$\textbf{-52}\pm0.8$
1	Biogas Plant		$-56 \pm 3$

Table 6.7.2: Comparison of previous and recent studies

Previous Studies<sup>[1,2,3]</sup>

δ<sup>13</sup>C(‰)

**Recent Survey** 

Results

δ<sup>13</sup>C(‰)

Variation in emissions by season and diurnal temperature will be assessed by mobile methane measurement of plumes at a selected number of waste sites in the UK with samples collected for isotopic analysis: Landfills from UK and Netherlands



- Mobile methane measurements of plume and isotopic signatures of Groningen, NL area will be evaluated.
- ESR7 will be trained for using Python programme or R.
- Methane oxidation rate changes with age of waste and temperature will be calculated by utilizing additional gas well samples at selected landfill sites.
- Isotopic differences between waste sources and fossil fuel and ruminant sources will be assessed by mobile measurement and sampling at key sites.
- The significance of fugitive CH<sub>4</sub> from combustion plants at landfills, biogas plants and wastewater treatment plants to the total CH<sub>4</sub> emitted will also be assessed.

The landfill sites, wastewater plants and biogas composting plants to be studied have not been selected yet. This will be done after meeting with partner Viridor in the next months. Moreover, to get access to selected waste treatment sites, related correspondence will be initiated.

## 6.7.1.4 Collaborations (internal / external)

ESR7 has cooperated with ESR5, ESR8 and ESR9 during ESR5 and ESR8 secondments. They have participated five days campaign in the UK. They have also analysed Egham air for diurnal measurement on GC-IRMS and isotopic CRDS to compare the results.

ESR7 has completed her secondment at University of Groningen. She attended six days campaigns at Groningen and collaborated with Prof. Huilin Chen and ESR2. She had an experience on drone measurement with air core at cow farm.

ESR7 will work with the non-academic partner Viridor on landfill monitoring, site access and combined measurement and training in the waste industry. Also, ESR7 will have a 2-week secondment at Utrecht University to learn deuterium isotope measurement techniques.

#### 6.7.1.5 Risks and difficulties

One week of working time was lost in January 2018 to return to Turkey to acquire Schengen visa for MEMO<sup>2</sup> school in the Netherlands and annual meeting in Switzerland. After first three employment months, future Schengen visas can be obtained directly in UK.

ESR7 lost her biometric residence permit during the ICOS Conference in Prague and could not turn back to the UK for methane isotope workshop, but logged in by Skype for the presentations. She had to start her secondment in Groningen one week early and expend the secondment period to five weeks.

#### 6.7.2. Deliverables

**D1.1** - Lightweight CH<sub>4</sub> sensor & AirCore development & deployed on UAV (month 24)

ESR 7 has done her secondment at University of Groningen. She had an experience on UAV-based active AirCore mobile measurement at cow farm during her secondment. She collected the gas samples from AirCore to measure their isotopic signatures.

D1.4 - Improved emission factors for different source categories from mobile measurement (month 42)

ESR 7 has been part of 19 days of mobile measurement campaigns and responsible for assessing the data for 18 of these. She has analysed isotopic signatures of waste sources. She has evaluated the isotopic signatures and mole fraction of different waste sources, such as landfill, waste water treatment plant and biogas plants.

**D1.5** - Report on harmonized method for mobile CH<sub>4</sub> and  $\delta^{13}$ C-CH<sub>4</sub> (month 18)

Martina Schmidt has reported the harmonized method by for mobile CH<sub>4</sub> and  $\delta^{13}$ C- CH<sub>4</sub>. ESR7 has contributed the part of RHUL instrument setup and measurement methods.

D2.1 - Isotopic measurements linked to common scale (month 18)

ESR7 has prepared the standard tanks by diluting from original sources and measured the standards for inter-calibration of isotopic measurements on the GC-IRMS at RHUL.

**D2.2** Improved isotopic source signatures of local and regional CH<sub>4</sub> (month 36)



ESR7 has collected samples of waste sources, landfill, wastewater treatment plant and biogas plant on eight days in UK and 6 days in the Netherlands. ESR7 has also calculated isotopic signature for different waste sources on all but very recent Groningen campaigns. These will be characterised by November 2018.

**D2.3** - Publication on the use of isotopes for CH<sub>4</sub> source attribution in urban/industrial regions (month 36)

ESR 7 has been a part of seven days of urban surveys where waste emission hotspots occur in the UK inventory. She has identified methane emissions from wastewater treatments, biogas plants and old closed landfill sites and collected air samples from methane plume.

#### 6.7.3 Training and network activities

#### 6.7.3.1 General training events

ESR 7 has attended or will attend the training events listed below.

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
Mobile Measurement Training	30.01.2018- 31.01.2018	RHUL	To get the skills of using mobile measurement equipment such as CRDS, LGR and sample collection.	2	Participating	
Sample Measurement Training	12.01.2018	RHUL	To learn to how air bag samples are measured on CRDS instrument and mobile LGR	0.5	Participating	
IRMS Analysis Training	02.02.2018	RHUL	To get the skill of laboratory analysis of carbon isotopes using GS-IRMS and interpret the data	0.5	Participating	
MEMO <sup>2</sup> School	05.02.2018- 16.02.2018	Holland	Two-week thematic school on methane including courses, practical exercises, field campaigns and data analysis.	6	Presenting a poster and oral presentation	
MEMO <sup>2</sup> annual MEMO <sup>2</sup> Annual Meeting	22.03.2018- 23.03.2018	EMPA	To be updated about the project, discuss and evaluate the progress of the first year and give an outlook to the second year.		Presenting a poster and presentation	
Academic Grammar and Vocabulary Course	21.02.2018- 14.03.2018	RHUL	Four-week course to enhance basic knowledge of grammar more effectively in writing, to express ideas more clearly and academic in sentences	2	Participating	
Clarity in Academic Writing Course	26.02.2018	RHUL	It is about the importance of logically grouping and sequencing ideas; how to construct clear, cohesion paragraph; how to use sentence/clause connectors; improving clarity though simplified language.	0.5	Participating	
Managing of Research Data and Publication Course	01.03.2018	RHUL	Getting knowledge about open accesses for research publication, introduction to research management	0.5	Participating	
Volunteering in the Science Festival	11.03.2018	RHUL	Guide and show public to how mobile methane measurements are made, make cow mask with kids	0.5	Participating, teaching	
Writing a Literature Review Course	14.03.2018	RHUL	Tips about writing literature review; how to organize and connect to different articles related to sub-subjects.	0.5	Participating	
Networking Workshop	27.02.2018	RHUL	It is about to understanding how to remain your authentic self, whilst building relationships and making helpful contact; giving practical tips and techniques for creating own networking strategy and plan; and Getting the most from digital and online networking	0.5	Participating	
Mentoring and Cultural Awareness Workshop	08.05.2018	RHUL	Couse help to understand cultural differences and the impact on the mentoring relationship; and it gives a chance to work together to identify strategies to manage differences.	0.5	Participating	



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

# D5.9 MEMO<sup>2</sup> – Midterm Review Report

MEMO <sup>2</sup> Methane Isotope Workshop	17.09.2018- 19.09.2018	RHUL	To learn much more detail about methane isotopes such as global trends. Identification of sources, sampling analysis, measurement techniques and regional /global /temporal modelling of methane isotopes	0	Participating on Skype	
MEMO <sup>2</sup> Plume Workshop Modelling	09.10.2018- 10.10.2018	UHEL	To learn how to model plume, make a practice by writing a code on Phyton.	2	Participating	
GIS Course	01.11.2018- 06.11.2018	RHUL	To learn ArcGIS programme more effectively to draw methane emission maps	4	Participating	
Introduction to R course	09.11.2018	RHUL	Introduction of some basics of R.	1	Participating	
Emotional Intelligence	08.11.2018	RHUL	To develop my own intelligence profile, using the Emotional Quotient Inventory, understand how to maximize on own emotional strength and improve it.	0.5	Participating	
CV writing and interview Skills Training Course	27.11.2018	RHUL	To learn writing effective CV and interviewing techniques	0.5	Participating	
MEMO <sup>2</sup> School	18.02.2019- 22.02.2019	France	To be updated about the project, discuss and evaluate the progress and have a meeting with EU representative.		Presenting a poster and presentation	

#### 6.7.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
1.	17.09.2018- 26.10.2018	Holland	RUG	MobilemeasuringofGroningenemissionsource.CharacterizationofGroningenMethaneEmissionSources	UAV Air core working Principle	Isotopic results and mobile measurement characterization will be done.

# 6.7.3.3 Conferences

Conferen ce name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation	Authors (main author + co-authors)	Public available (yes / no) / web link
ICOS	11.09.2018- 14.09.2018	Czech Republic	Poster	Waste Source in the UK	S. Bakkaloglu + D.Lowry, R. Fisher, E. Nisbet	https://conference.icos- ri.eu/wp- content/uploads/2018/09/ICO S2018SC Book of Abstract s.pdf
BBOS	25.10.2018- 26.10.2018	Holland	Poster	Waste Source in the UK	S. Bakkaloglu, + D.Lowry, R. Fisher, E. Nisbet	NO

# 6.7.3.4 Measurement / sampling campaigns

Campai gn	Date (start – end, planned (when))	Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
Yorkshir e (KM5)	30.01.2018- 31.01.2018	Yorkshire	RHUL	Mobile car night measurement	Training on mobile methane measurement	25 bags were collected	
MEMO <sup>2</sup> School	09.01.2018- 12.02.2018	School		Mobile car night measurement	Training on mobile methane measurement	MEMO NL- 35 bags	
Sutton (UNC1)	03.05.2018	Sutton	RHUL	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures	UNC1-1,2,3	Gas leaks have been found
Cambrid ge	13.05.2018	Cambridg e	RHUL	Cow Barn	To quantify methane mole fractions and isotopic signatures	CAM 1,2,3,4,5,6,7,8,9, 10,11,12	See 1.2.2



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

		_			-		_
						13,14,15,16,17,1 8,19	
Brighton (UNC2)	14.05.2018	Brighton city	RHUL	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures	UNC2- 1,2,3,4,5,6,7,8,9, 10,11,12 13,14,15,16,17,1 8	See 1.2.2
MEMO-1	22.06.2018	Local Sources	RHUL	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures	MEMO RHUL1- 1,2,3,4,5,6	See 1.2.2
MEMO-2	26.06.2018	Oxford, Bicester, Milton Keynes	RHUL	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures	MEMO RHUL2- 1,2,3,4	See 1.2.2
MEMO-3	27.06.2018	Local RHUL Sources	RHUL	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures	MEMO RHUL3- 1,2,3,4,5	Gas leaks have been found
MEMO-4	28.06.2018	Isle of Grain/Ke nt	RHUL	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures	MEMO RHUL4- 1,2,3,4,5,6,7,8	See 1.2.2
MEMO-5	05.07.2018	Devon	RHUL	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures for Heathfield Landfill, and Exeter region	MEMO RHUL1- 1,2,3,4,5,6,7,8,9, 10,11,12	See 1.2.2
RUG-1	25.09.2018	Groninge n	RUG	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures for Groningen city and Germany pit fire	GROG1- 1,2,3,4,5,6,7,8	Will be evaluated
RUG-2	26.09.2018	Groninge n	RUG	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures for Groningen city	GROG2- 1,2,3,4,5,6,7,8,9	Will be evaluated
RUG-3	27.09.2018	Groninge n	RUG	Mobile car night measurement	To quantify methane mole fractions and isotopic signatures for Groningen city	GROG3-1,2,3,4	Will be evaluated
RUG-4	2.10.2018	Groninge n	RUG	Mobile car night measurement	To see the methane emissions and isotopic signatures change during night measurement.	GROG4- 1,2,3,4,5,6,7,8,9	Will be evaluated
RUG-5	18.10.2018	Lutjeward	RUG	Observation	Observation of sampling and measurement techniques, taking background samples for isotopic methane signatures	ROG5-1,2	Will be evaluated
RUG-6	19.10.2018	Grijskerk Cow Farm	RUG	Farm- Drone measurement	Observing UAV Aircore Measurement Techniques to quantify methane mole fraction. Figure outing Duct Farm isotopic methane signatures	GROG6- 1,2,3,4,5	Will be evaluated

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.7.4 Dissemination activities

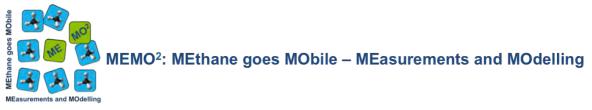
Except for the contributions to the conferences no scientific publications or other dissemination activities so far from the ESR.

#### Referencesc

Fisher, R.E. (2000) Development and application of continuous gas chromatography isotope ratio mass spectrometry for atmospheric methane and carbon dioxide studies, PhD thesis, Royal Holloway University of London, p:369.

Lowry, D., Holmes, C.W., Rata, N.D., O'Brien, P. & Nisbet, E.G. London methane emissions: Use of diurnal changes in concentration and δ13C to identify urban sources and verify inventories. J Geophys Res-Atmos 106, p. 7427–7448 (2001). Zazzeri, G., Lowry, D., Fisher, R.E., France, J.L., Lanoisellé, M., Grimmond, C.S.B. & Nisbet, E.G. (2017) Evaluating methane

inventories by isotopic analysis in the London region, Scientific Reports 7: 4854



# 6.8 ESR8 – Isotopic characterisation of methane sources in Europe

#### ESR8

#### Isotopic characterisation of methane sources in Europe

ESR	Malika Menoud, m.menoud@uu.nl
Supervisor	Thomas Röckmann, T.Roeckmann@uu.nl
Co-supervisor	David Lowry, <u>d.lowry@es.rhul.ac.uk</u>
Non-Academic mentor	Renato Winkler, <u>rwinkler@picarro.com</u>
Official start – end date	15/11/2017 – 15/11/2021

#### 6.8.1 Scientific progress

#### 6.8.1.1 Project introduction and objectives

Methane is a greenhouse gas of major importance, for which the interest increased recently. With a GWP of 25, the increase in atmospheric mixing ratios of methane makes it a cause for concern. From pre-industrial times, atmospheric methane mole fraction has been increased by 150%. On a closer scale, the growth rate in the atmosphere slowed down from the beginning of the 1990's, until the recent rise of methane emission rate in 2007. These variations confirm the need for more investigations on methane source strengths and partitioning.

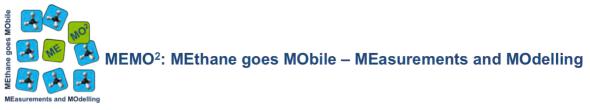
Methane is emitted from three main origins: fossil fuel extraction (natural gas, coal, oil) and use (leaks), biogenic formation in wetlands, animals (ruminants) and oceans, and biomass burning (wild or anthropogenic). These pathways produce methane with different isotopic ratios in the emitted gas, due to different isotopic fractionations. Measuring carbon and hydrogen isotope ratios in sampled air methane provides useful information about the origin of the methane found at a certain place. These measurements also help to calculate the partitioning of the different methane sources on a larger scale. Isotopic measurements have therefore an important role in reducing the uncertainties in the global methane budget.

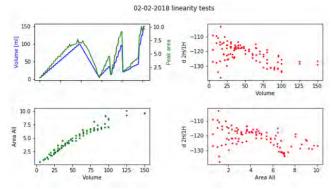
This project aims at the characterization of methane sources in Europe using isotopic measurements. The data is collected through a high-precision measurement system for both carbon and hydrogen isotopes of methane in atmospheric air samples. Through the MEMO<sup>2</sup> network, samples from various locations and diverse sources can be collected and measured. The system can also measure the ambient air continuously at a fixed location for several months. Such measurements will be performed at a European scale, in order to better identify the main methane sources, as well as potential temporal and spatial variability. This data would allow to map the methane source signatures on the continent, as well as being used as input for the validation of isotopic models. By collecting more specific samples, these measurements will also be used to investigate the processes that lead to variations in the isotopic ratios of emitted methane.

#### 6.8.1.2 Project results

#### 6.8.1.2.1 First year

Testing the linearity of the measurements made by the main instrument that will be used during the project In Fig. 6.8.1 and 6.8.2, the results of the linearity assessment of our instrument are shown. It appears that a certain range of methane volume injected into the IRMS leads to more precise results. According to Fig. 6.8.2, the optimal peak size would be of 4 Vs, which corresponds to an accuracy of ~2 ‰ for values of  $\delta D$ . This value represents the precision we can currently achieve for the measurements of  $\delta D$ -CH<sub>4</sub> with the measurement system as its current state. The amount of methane to be injected in the measurement system when analysing a sample is therefore to be adjusted to this value, as much as possible. This information is used to better understand the generated data and optimize the measurements procedure to get results as precise as possible for the samples. Therefore, such tests will be performed regularly, and also for the measurement of <sup>13</sup>C isotopes.





D5.9 MEMO<sup>2</sup> – Midterm Review Report

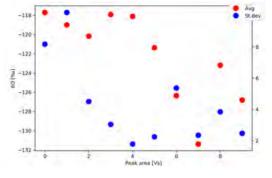


Fig. 6.8.1: Overview of the linearity tests performed for  $\delta D\text{-}CH_4$  measurements

**Fig. 6.8.2:** Average and spread of the isotopic measurements in relation with the amount of methane measured

#### Regular sampling at nearby sources: cow farms

In January 2018, 8 samples were collected every week during 3 weeks, at the same times of the day and same locations around the farm (Fig. 6.8.2). They were measured for methane mole fraction and hydrogen isotopic signature. The corresponding Keeling plots are shown in Fig. 6.8.4, and the resulting signatures in Table 6.8.1. The values correspond to what can be expected for a biogenic source.



Fig. 6.8.3: Sampling locations around the farm

**Table 6.8.1:** Resulting source isotopic signatures from the Keeling plot analysis

Date	δD [‰]	err δD [‰]
11/01/2018	-277.70	34.54
18/01/2018	-285.56	27.52
25/01/2018	-378.66	53.71

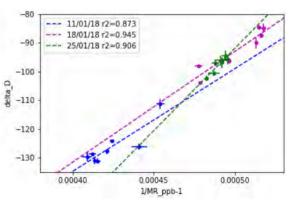


Fig. 6.8.4: Keeling plot of the  $\delta D$  results for January 2018 at the farm



More data is needed to interpret the differences, which can be related to weather conditions or change in the farm (feeding, living configuration, manure management. Two additional ...). sampling campaigns were done at another farm, in the same area. The method was the same: the samples were collected in Tedlar bags, using a hand pump and a sampler box for creating a vacuum around the bag so that it is not filled through the pump. The bags were measured at the lab within a week. From the first sampling day to the second (23rd of January and 29th of March 2018), the sampling locations were the same, as well as the approximate time in the day. The results are presented in the

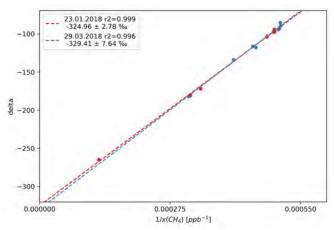


Fig. 6.8.5: Keeling plot of the  $\delta D$  results for another farm nearby, based on 2 days of survey

Keeling plot in Fig. 6.8.5. The x-axis shows that a wider range of methane mole fraction was captured at that farm, which significantly decreases the uncertainty in the  $\delta D$ -CH<sub>4</sub> source estimates (see the values in the legend of the graph).

#### Analysis of long-term time series of methane mixing ratio, carbon and hydrogen isotopic signatures

The data was collected during 5 months in 2016 and 2017 at Lutjewad, Netherlands. It concerns air CH<sub>4</sub> mole fractions,  $\delta D$  and  $\delta^{13}C$ . CH<sub>4</sub> mole fractions were simultaneously measured with a Picarro CRDS instrument, in order to check the data provided by the IRMS. The measurements were continuous (about 1 every 30 min) from the 3<sup>rd</sup> of November 2016 to the 31<sup>st</sup> of March 2017, except when technical maintenance was performed. Fig. 6.8.6 shows an overview of the raw data.

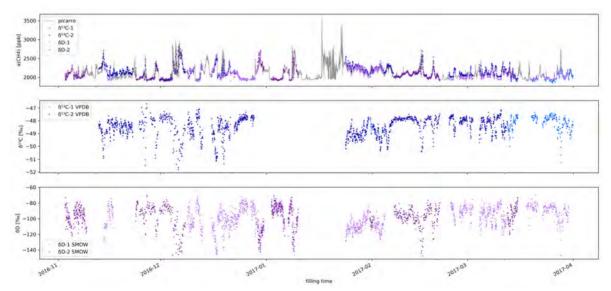


Fig. 6.8.6: Overview of the data taken at Lutjewad



#### 6.8.1.2.2 Second year

#### Analysis of long-term time series of methane mixing ratio, carbon and hydrogen isotopic signatures

I've been analysing the dataset taken at Lutjewad in more details. First, I have corrected the CH<sub>4</sub> mole fractions from the IRMS according to the Picarro measurements. The difference was a constant offset during a certain time period. When technical manipulations were performed and the system restarted, the offset was also changing, and sometimes was inexistent. Fig. 6.8.7 shows the data after correction.

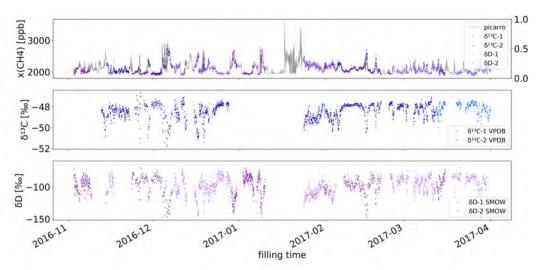


Fig. 6.8.7: Overview of the entire dataset, after correcting the [CH4] values

The next step was to extract isotopic signatures of the emission source that caused the peaks in the data. The methods that we're using most of the time are the Keeling plot and Miller-Tans approaches. When applied on the entire dataset, it shows that the sources are relatively constant. However, there are potentially other sources that we're missing due to the large number of [CH<sub>4</sub>] peaks that are in the dataset.

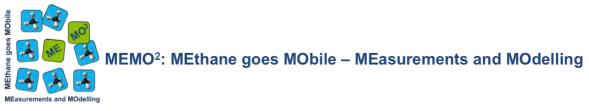
To have a closer look at each of these events, the Keeling plot and Miller-Tans plots were applied to a moving window on the dataset.

Several parameters can then be tuned:

- the time extend of the window, the minimal number of points that have to be aligned,
- the minimal range of mixing ratio the data points must cover
- a background value that at least one point must have (for the Keeling plot only).

For more visibility, the data was cut in 9 parts, and the routine applied for each of them. Fig. 6.8.8 and 6.8.9 are showing the resulting Keeling plots for one of the subsets.

We can see that a clearly different source isotopic signatures are calculated for the different elevations in [CH<sub>4</sub>] during that period. We can visualize them on the graph in Fig. 6.8.10. We can see that the differences can be clear from one peak to another, but there are still some ambiguities. They might be reduced by applying different parameters for the moving window Keeling plot. The same graphs were created for Miller-Tans plots, and lead to similar results.



D5.9 MEMO<sup>2</sup> – Midterm Review Report

Moving window Keeling plot, n=8, min MR window = 150 ppb, min MR value = 1800 ppb

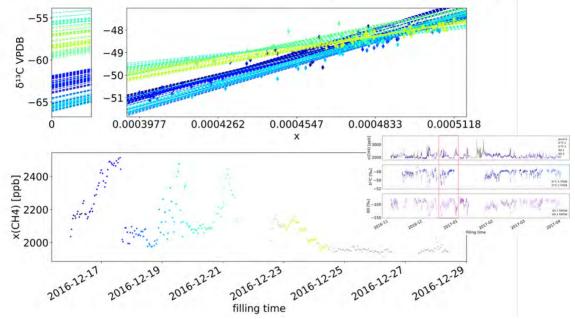
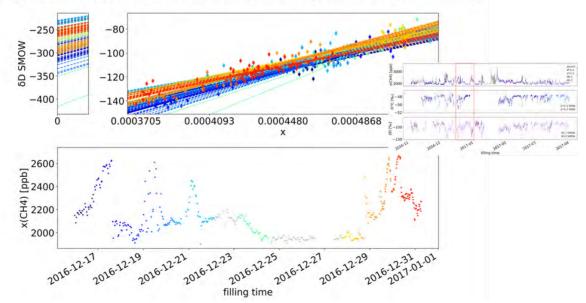


Fig. 6.8.8: Moving window Keeling plot approach for  $\delta^{13}$ C on a subset from the 16/12/2016 to 29/12/2016



Moving window Keeling plot, n=8, min MR window = 150 ppb, min MR value = 1800 ppb

Fig. 6.8.9: Moving window Keeling plot approach for  $\delta D$  on a subset from the 16/12/2016 to 01/01/2017



D5.9 MEMO<sup>2</sup> – Midterm Review Report dt = 24 h, spread > 150 ( $\delta^{13}$ C), 150 ( $\delta$ D) [ppb], n>8, r2>0.8

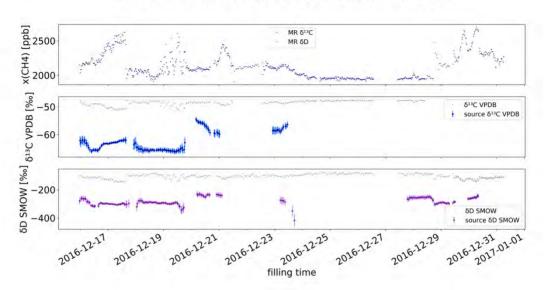
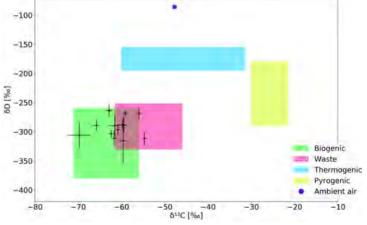


Fig. 6.8.10: Source isotopic signatures resulting from the moving window Keeling plot analysis on the subset from the 16/12/2016 and 01/01/2017

Another analytical approach was to manually select the highest and most clear peaks of CH<sub>4</sub> mole fractions in the entire dataset, and calculate the source signatures with a Keeling plot. 20 peaks were selected that way. The source signatures values resulting from the Keeling plots intercepts – which had an R-value larger than 0.8 - were plotted in a  $\delta D$  vs.  $\delta^{13}C$  graph. It allows to compare with previously reported signatures for the different types of methane sources. We can also see compare with the ambient air signature. This was calculated from a 3 days period of constant background [CH<sub>4</sub>] observed in December 2016 in the Lutjewad dataset ( $\delta^{13}C_{background} = (-47.74 \pm 0.20)$ , n=43 and  $\delta D_{background} = (-85.79 \pm 4.37)$ , n=40).

This kind of plot allows a clear interpretation of the main sources. Further work will include getting more signatures from smaller events, with the help of the moving window approach, and visualize them on such plot. The goal is to identify the source of as many methane peak events as possible. Other aspects that could be analysed is the influence of a potential day/night cycle, and the wind data that was also recorded at the same location.



Set-up of a continuous measurement system for  $\delta D$  and  $\delta^{13}\text{C-CH}_4$  in ambient air, at AGH University of Science and Technology of Krakow, Poland

Fig. 6.8.11: Methane source isotopic signatures of 20 high peaks in the Lutjewad dataset (black points)

From the 16<sup>th</sup> to the 30<sup>th</sup> of May 2018, I've stayed in Krakow together with Carina van der Veen, in order to install the instrument for continuous CH<sub>4</sub> mole fractions,  $\delta D$  and  $\delta^{13}C$ -CH<sub>4</sub> measurements in ambient air. The system was prepared at UU and shipped to AGH University. It took about 10 days to have all



D5.9 MEMO<sup>2</sup> – Midterm Review Report

the components installed and working. However, the compressor broke just before I left, and getting a new one took a long time. The measurements could start only in mid-September 2018.

Fig. 6.8.12 shows the data that has been collected so far. My work in analysing the data has not started yet, and will start by correcting it for a mixing ratio offset and some irregularities due to environmental disturbances.

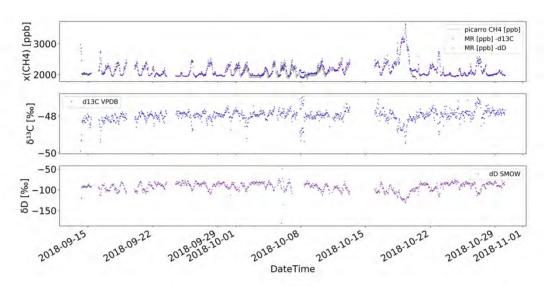


Fig. 6.8.12: Overview of the data being currently collected in Krakow

# Participation in the CoMet campaign (Silesia, Poland): isotopic measurements of methane from mine shafts exhaust gas

From the 27<sup>th</sup> to the 29<sup>th</sup> of May 2018, I could join the CoMet campaign in the region of Silesia, Poland. This campaign was organized by DLR (Germany) and aimed at the calibration of measurements from an aircraft. Other students from the MEMO<sup>2</sup> network joined the ground-based teams for measuring the methane concentration in the mining area of the upper Silesia. During my stay, I collected 43 samples from mobile surveys in a van, using a Picarro analyser on board for detecting the methane plumes. 9 mine shafts were visited, from 6 different mines. 3 additional samples were taken directly in 3 shafts of the Pniówek mine during visits in cooperation with the mining company. 7 samples are from the hotel Pustelnik, and were taken at regular times during a period of 24 hours. Finally, 6 background samples were taken (2 per days) to provide a robust isotopic value for ambient air. Fig. 6.8.13 and 6.8.14 provide an overview of the sample locations and methane mole fractions.



Fig. 6.8.13: Sampling locations



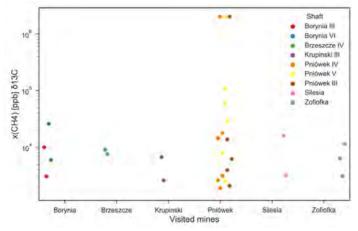


Fig. 6.8.14: Methane mixing ratios in the collected samples (all atmospheric, except the 3 taken inside the Pniówek shafts)

After measuring all the samples for  $\delta D$  and  $\delta^{13}C$ -CH<sub>4</sub> at the lab in Utrecht, the data was analysed with the Keeling plot approach to calculate the source isotopic signature for each shaft. The results are presented in Table 6.8.2. The r-values show that the method was adapted for most of the shafts, except Pniówek V.

Indeed, the bad correlation is due to the fact that the high concentration sample taken in the shaft didn't align with the atmospheric samples taken in the plume outside. The reason in probably because they were not collected the same day, which also points out the fact that the isotopic signals are changing rapidly in time, apparently even in an hourly scale.

The results show also important variations in methane isotopic ratios between different shafts, even within the same mine. This heterogeneity is probably due to changes in the depth where the mines are ventilated.

Table 6.8.2: Resulting intercepts of the Keeling plots, with the errors of estimate and the r-values of the linear fits, for the samples from Silesia

	δ <sup>13</sup> C VPDB [‰]	err_δ <sup>13</sup> C [‰]	r-value - δ <sup>13</sup> C	δD SMOW [‰]	err_δD [‰]	r-value - δD
Borynia III	-56.37	0.11	1.00	-210.96	4.16	1.00
Brzeszcze IV	-47.02	0.09	-0.97	-144.29	1.63	1.00
Silesia	-58.41	1.36	0.96	-189.73	2.99	1.00
Pniówek III	-53.79	0.32	0.98	-195.47	6.97	0.98
Zofiofka	-56.54	0.08	1.00	-198.02	10.56	0.97
Pniówek IV	-48.86	0.09	0.96	-190.84	2.29	1.00
Pustelnik	-57.13	1.26	0.91	-195.13	9.47	0.96
Pniówek V	-48.13	0.66	0.11	-209.49	9.92	0.95
Krupinski III	-57.02	0.21	1.00	-155.81	3.48	0.99
Borynia VI	-50.41	0.43	0.93	-193.68	8.88	0.98
All mines	-53.37	4.33		-188.34	21.56	

The diagram in Fig. 6.8.15 provides an overview of the mines' isotopic signatures, related to other methane sources previously reported. As we can expect, the points are in or close to the thermogenic zone. However, it seems that both  $\delta D$  and  $\delta^{13}C$  are rather depleted for this kind of source. This diagram shows how useful it is to measure both isotopes in this case, since only  $\delta^{13}C$  wouldn't be enough to characterize the source.

The samples from Hotel Pustelnik taken at regular time intervals during 24 h allow to identify a diurnal cycle in the methane concentrations.

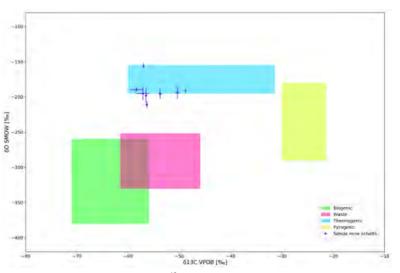
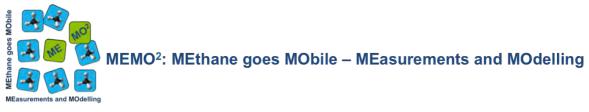


Fig. 6.8.15: Diagram of  $\delta D$  versus  $\delta^{13}C$  of previously reported methane sources, together with the signatures of the mines from this campaign



Indeed, the location was not affected by a direct plume from any shaft, so an elevation in the background methane mole fractions could be measured. The isotopic signatures for these samples are shown in Fig. 6.8.16. The source signatures calculated from the Keeling plots of these values are in Table 6.8.2 and also show a clear prevalence of a thermogenic source in this diurnal methane cycle.

#### Isotopic characterization of various methane sources in the UK during my secondment at the Royal Holloway University of London

I have stayed in Egham (UK) during 4 weeks, from the 18<sup>th</sup> of June to the 14<sup>th</sup> of July, to complete my secondment at the Royal Holloway University of London (RHUL). Together with 3 other MEMO<sup>2</sup> students, Sara Defratyka, Semra Bakkaloglu and Julianne Fernandez, we went on 5 surveys in different parts of England. The goal for me was to sample various sources of methane from the UK, in order to characterize their isotopic composition. Table 6.8.3 summarizes information on each campaign.



Fig. 6.8.16: Diurnal cycle observed at Hotel Pustelnik

Table 6.8.3: Information of the surveys done during my secondment at RHUL

Date	Location	Targeted site	Num. of collected samples (incl. 1 background)
22/06/2018	Surroundings of Egham, West London	Mix	6
26/06/2018	N-E of Oxford, Oxfordshire and Buckinghamshire	Mix	8
27/06/2018	East of Egham, West London	Gas leaks	7
28/06/2018	Kent county	North Sea gas terminal on the Isle of Grain	11
05/07/2018	Devon county	Greatness landfill	14

We went surveying with 2 cars: in the first car were a Los Gatos ethane / methane analyser and a Picarro CRDS analyser, together with the sampling material; in the second car were another Picarro CRDS, that is also measuring  $\delta^{13}\text{C-CH}_4$ , together with a newly developed air core system. The instruments inlets were located on top of each car, and they were following each-other closely. The Picarro data from the first car is shown in Fig. 6.8.17.

I took a total of 47 atmospheric samples from the first car, in FlexiFoil Tedlar bags (3I) and SUPELCO aluminum bags (2I). We used the Picarro measurements to guide us in the methane plumes in order to sample significantly higher concentrations and get the source isotopic signature as precisely

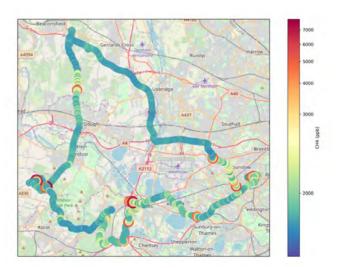
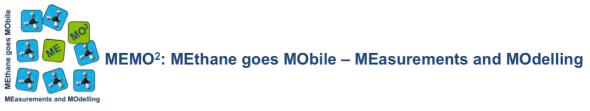
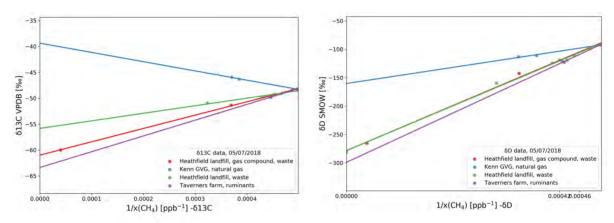


Fig. 6.817: Picarro records from the first survey (22/06/2018)



as possible. In addition, 2 high concentration samples were taken directly in the biogas plant and in a gas well of the Greatness landfill, during our visit on the 5<sup>th</sup> of July 2018. The source signature for each site was calculated using the Keeling plot approach. Fig. 6.8.18 shows example Keeling plots for one of the campaigns. All the results are summarized in Table 6.8.4.



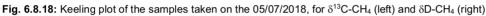


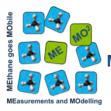
Table 6.8.4: Resulting intercepts of the Keeling plots, with the errors of estimate and the r-values of the linear fits, for the samples from the UK

Sites	Source	Date	δ <sup>13</sup> C VPDB [‰]	err_δ <sup>13</sup> C VPDB [‰]	r-value - δ <sup>13</sup> C	δD SMOW [‰]	err_δD SMOW [‰]	r-value - δD
Chertsey STW	STW	22/06/2018	-49.95	0.54	0.96	-292.19	24.83	0.99
Isleworth WWTP	WWTP	22/06/2018	-48.95	0.00	1.00	-302.32	0.00	1.00
The Causeway	Gas leak	22/06/2018	-37.71	0.00	-1.00	-131.03	0.00	1.00
Winkfield	Gas leak	22/06/2018	-38.95	0.00	-1.00	-128.23	0.00	1.00
Bicester	WWTP	26/06/2018	-52.02	0.11	1.00	-275.28	3.85	1.00
Calvert landfill	Landfill	26/06/2018	-53.84	0.00	1.00	-172.46	0.00	1.00
Milton biogas	Biogas plant	26/06/2018	-56.90	0.64	0.99	-239.91	20.03	0.98
Felthem, Sunbury rd	Gas leak	27/06/2018	-40.84	0.00	-1.00	-161.89	0.00	1.00
Laleham, Ashford rd	Gas leak	27/06/2018	-42.63	0.00	-1.00	-160.97	0.00	1.00
Shepperton community landfill	Landfill	27/06/2018	-39.46	3.03	-0.93	-154.02	10.27	0.99
Farningham GVC	Gas compound	28/06/2018	-37.05	0.50	-1.00	-137.65	1.23	1.00
Greatness quarry	Landfill	28/06/2018	-55.82	0.69	0.97	-273.32	7.33	0.99
Leybourne lakes	WWTP	28/06/2018	-51.99	1.21	0.95	-255.37	19.45	0.99
Heathfield landfill	Landfill	05/07/2018	-55.81	0.82	0.97	-278.39	4.15	1.00
Kenn GVG	Gas governor	05/07/2018	-39.36	0.33	-1.00	-160.20	6.00	0.99
Taverners farm	Ruminants	05/07/2018	-63.36	0.02	1.00	-298.98	14.69	1.00

Overall, the signatures correspond to what we can expect from the sources we sampled. However, we can identify the plume from Shepperton community landfill as being from a thermogenic source even though we thought of sampling from the landfill. The gas leaks sampled in Felthem and Laleham have quite a depleted isotopic signature, for both isotopes. Combining these results with the data from RHUL could help to go deeper in their interpretation.

#### Inter-comparison measurements of $\delta^{13}\mbox{C-CH}_4$ between RHUL and UU labs

The time of my secondment at RHUL was also dedicated to lab work. The goal was to perform an intercomparison of the  $\delta^{13}$ C-CH<sub>4</sub> measurements made in RHUL and UU. Therefore, I have brought a set of samples from UU, after being measured there. I took back to UU another set of samples from the surveys in England, after measuring them at RHUL lab. We then got a dataset for the same samples from the 2 labs, and could compare the values to potentially identify differences. The list of selected samples is shown in Table 6.8.5.



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### Table 6.8.5: Samples used for the inter-comparison between RHUL and UU labs

ID	Sampling date	Location/campaign CH₄ sou		[CH₄] (ppb)	err [CH₄] (ppb)
W-B5 1	06/06/2018	Experimental farm, Wageningen (NL)	Ruminants	984.75	1.21
W-B2 2	06/06/2018	Experimental farm, Wageningen (NL)	Ruminants	1172.12	0.13
Wa-ba 2	06/06/2018	Experimental farm, Wageningen (NL)	Ruminants	42131.38	1611.11
W-bg 1	06/06/2018	Wageningen (NL)	Background	2047.66	0.66
PN-IV S	28/05/2018	Silesia (PL) - Pniówek IV	Coal mine	2062.41	5.68
PN-V S	28/05/2018	Silesia (PL) - Pniówek V	Coal mine	949.30	4.18
U-1 4	09/05/2018	Utrecht city (NL)	Urban	2341.21	2.90
U-1 7	09/05/2018	Utrecht city (NL)	Urban	2880.01	2.38
U-1 8	09/05/2018	Utrecht city (NL)	Urban	2312.57	0.66
U-1 9	09/05/2018	Utrecht city (NL)	Urban	2238.27	0.40
NG U	13/06/2018	Natural gas (NL)	Fossil fuel	2607.84	33.13
HOTEL-DAK- 7:30 (E)	10/08/2017	Stratoclim, Geophysica (NO)		2616.53	15.02
FL3-5	2017	Stratoclim, Geophysica (NO)	Low mole fraction	1884.18	8.35
FL5-20	2017	Stratoclim, Geophysica (NO)	Low mole fraction	1558.83	15.05
PP19SS	2017	Pipers (NZ)	Low mole fraction	1773.32	2.96
PP95SS	2017	Pipers (NZ)	Low mole fraction	1788.50	3.98
Dome 12	1998/1999	Dome Concordia (Antartica)	Low mole fraction	1485.10	
UEA 15	28/07/2009	NEEM (Greenland), surface	Low mole fraction	1861.87	7.03
ER	14/02/2018	Krakow area (PL)	Unknown	15113.29	339.03
GD	14/02/2018	Krakow area (PL)	Unknown	11293.59	95.56
NN	14/02/2018	Krakow area (PL)	Unknown	3254.35	14.95
MO 1	22/06/2018	Isleworth (UK)	Waste	2404.55	7.53
WI 1	22/06/2018	Winkfield (UK)	Winkfield	2507.41	12.53
CA 1	22/06/2018	Causeway, Egham (UK)	Fossil fuel	2189.99	13.87
BG I	22/06/2018	Winkfield Lane (UK)	Background	1936.85	0.08
COV 1	26/07/2018	Calvert landfill (UK)	Waste	2066.29	3.34
MIL 2 - a	26/07/2018	Milton Keynes (UK)	Waste	2115.49	17.3
LA 1	27/06/2018	Laleham, Ashford rd (UK)	Fossil fuel	2263.42	17.32
LIT 1 - a	27/06/2018	Shepperton community landfill (UK)	Waste	2267.82	3.6
LIT 2 - a	27/06/2018	Shepperton community landfill, Charlton road (UK)	Waste	2241.54	6.37
FEL 1	27/06/2018	Felthem, Sunbury road (UK)	Fossil fuel	2334.61	1.39
A 228 - a	28/06/2018	Leybourne lakes, Snoland (UK)	Waste	2222.99	14.58
SHO 1	28/06/2018	Shorne gas governor (UK)	Fossil fuel	2153.1	6.4
FAR 1 - a	28/06/2018	Farningham GVC (UK)	Fossil fuel	2685.77	32.65
GRE 2 - a	28/06/2018	Greatness quarry, Vestry rd (UK)	Waste	3013.18	11.57
HEA 1	05/07/2018	Heathfield landfill (UK)	Waste	2333.12	18.25
ICE 1 - a	05/07/2018	Taverners farm (UK)	Ruminants	2243.49	48.12
Ambient EGH air	-	RHUL (UK)	Background	1967.57	2.9
Lab gas Low	-	RHUL Lab natural gas supply (UK)	Fossil fuel	1960.14	5.21
Lab Gas High	-	RHUL Lab natural gas supply (UK)	Fossil fuel	10261.3	19.4
Landfill Gas Low	-	Heathfield landfill (UK)	Biogas	1758.27	6.24
Landfill Gas High	-	Heathfield landfill (UK)	Biogas	10136.06	25.73

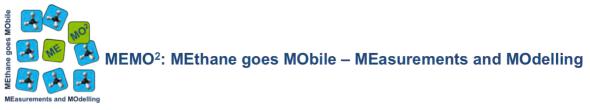
The last 5 samples are calibration tanks that were prepared at RHUL in order to be sent to the other MEMO<sup>2</sup> partners who are using an isotopic Picarro. The results of the measurements from each lab are presented in Table 6.8.6, Fig. 6.8.19 and 6.8.20. The large differences that we can observe for some of the samples is explained by sub-optimal measurements conditions such as:



- a low or high methane mole fraction, outside of the optimal measurement range of 1800 to 2500 ppb, which can require to dilute the sample prior to the isotopic measurement.
- a high or low enrichment compared to the reference  $\delta^{13}$ C -CH<sub>4</sub> (similar to ambient values of -47 ‰, as it corresponds to our working standard gas), which increases the uncertainty of the measurements
- different sample holders (snailless steel cans or aluminium foil bag), which can be more or less hazardous: the bags are less stable and a contamination can occur during the measurement

Table 6.8.6: Results of the inter-comparison measurements between R	RHUL and UU labs
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		UU		RHUL				
ID	Meas. date	n	δ <sup>13</sup> C	err_δ <sup>13</sup> C	Meas. date	n	d <sup>13</sup> C	err_δ¹³C
		rep.	<b>VPDB</b> [‰]	VPDB [%]		rep.	VPDB [‰]	VPDB [‰]
W-B5 1	15/06/2018	2	-59.24	0.00	25/06/2018	3	-57.69	0.04
W-B2 2	15/06/2018	2	-61.26	0.08	25/06/2018	3	-60.40	0.00
Wa-ba 2	15/06/2018	2	-56.6	0.07	02/07/2018	3	-56.85	0.03
W-bg 1	15/06/2018	2	-48.61	0.13	25/06/2018	3	-48.55	0.06
PN-IV S	15/06/2018	2	-48.84	0.07	25/06/2018	4	-48.98	0.02
PN-V S	15/06/2018	2	-51.51	0.05	25/06/2018	4	-50.93	0.01
U-14	14/06/2018	2	-49.88	0.02	25/06/2018	3	-49.65	0.04
U-17	14/06/2018	2	-47.38	0.02	25/06/2018	4	-47.73	0.01
U-1 8	14/06/2018	2	-49.47	0.09	25/06/2018	3	-49.43	0.02
U-1 9	14/06/2018	2	-49.47	0.00	25/06/2018	4	-49.45	0.05
NG U	15/06/2018	2	-39.31	0.16	25/06/2018	3	-39.87	0.02
HOTEL-DAK-		2	-49.81	0.06	02/07/2018	3	-49.91	0.02
7:30 (E)		_				-		
FL3-5		2	-47.56	0.01	02/07/2018	3	-47.45	0.01
FL5-20		2	-45.70	0.05	02/07/2018	3	-45.53	0.01
PP19SS		2	-47.23	0.08	02/07/2018	3	-47.04	0.03
PP95SS		2	-47.41	0.12	02/07/2018	3	-47.24	0.02
Dome 12		1	-49.83		02/07/2018	3	-47.77	0.03
UEA 15		2	-47.59	0.14	02/07/2018	3	-47.36	0.02
ER		2	-48.03	0.34	10/07/2018	3	-48.33	0.05
GD		2	-69.30	0.10	10/07/2018	3	-68.63	0.01
NN		2	-45.37	0.01	10/07/2018	3	-45.66	0.07
MO 1	11/07/2018	3	-48.38	0.02	31/07/2018	2	-48.12	0
WI 1	11/07/2018	3	-44.71	0.05	31/07/2018	2	-45.88	0.03
CA 1	11/07/2018	3	-45.99	0.08	31/07/2018	2	-46.74	0.06
BGI	11/07/2018	3	-47.71	0.06	13/08/2018	2	-47.92	0.02
COV 1	11/07/2018	3	-48.41	0.01	31/07/2018	2	-48.39	0.01
MIL 2-a	13/07/2018	3	-48.6	0.03	06/08/2018	2	-48.72	0.08
LA 1	11/07/2018	3	-46.01	0.01	31/07/2018	2	-46.88	0.17
LIT 1-a	16/07/2018	3	-46.09	0.03	09/08/2018	2	-46.17	0.03
LIT 1-b	16/07/2018	3	-46.19	0.07	08/08/2018	2	-46.79	0.04
LIT 2-a	16/07/2018	3	-45.77	0.07	09/08/2018	2	-45.73	0.05
FEL 1	11/07/2018	3	-46.05	0.02	31/07/2018	2	-46.73	0.04
A228-a	17/07/2018	3	-48.11	0.07	09/08/2018	2	-48.22	0.13
FAR 1-a	17/07/2018	3	-44.49	0.04	08/08/2018	2	-44.76	0.09
GRE 2-a	17/07/2018	3	-51.02	0.02	08/08/2018	2	-50.52	0.05
HEA 2	19/07/2018	3	-49.72	0.01	09/08/2018	2	-49.82	0.16
ICE 1-a	19/07/2018	3	-49.87	0.01	08/08/2018	2	-49.78	0.05
Ambient EGH air	25/09/2018	3	-48.07	0.01	05/10/2018	6	-48.09	0.03
Lab gas Low	29/07/2018	3	-39.54	0.02	05/10/2018	6	-39.61	0.07
Lab Gas High	31/08/2018	4	-38.21	0.01	05/10/2018	6	-38.14	0.04
Landfill Gas Low	31/07/2018	3	-59.73	0.05	05/10/2018	6	-59.8	0.1
Landfill Gas High	30/08/2018	3	-60.93	0.04	05/10/2018	6	-61.02	0.09
Landin Gas Flight	00/00/2010	0	-00.00	0.04	00/10/2010	0	-01.02	0.00



1.00

0.50

0.00

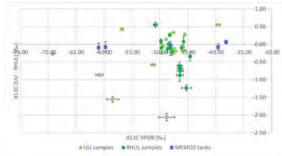
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3 -1.00

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-2.00

-2.50



PHI II cam Fig. 6.8.20: Differences between UU and RHUL measurements of  $\delta^{13}$ C-CH<sub>4</sub>, in function of [CH<sub>4</sub>] values of the samples

x(CH4) [ppb]

4000.0 5000.0

Litt samples.

8000.0 10000.0 \$2000.0 14000.0

MEMO2 tanks

15000 0 18000.0

Fig. 6.8.19: Differences between UU and RHUL measurements of  $\delta^{13}$ C-CH<sub>4</sub>, in function of  $\delta^{13}$ C-CH<sub>4</sub> values in the samples

For the calibration tanks, the differences remain within the measurement uncertainties. The results of this inter-comparison allowed us to conclude that the  $\delta^{13}$ C-CH<sub>4</sub> measurements at RHUL and UU labs are in good agreement, as long as the samples are carefully manipulated.

#### 6.8.1.3 Future plans and expected results

For the next reporting period, I am planning to:

- finalize the analysis of the data from Lutjewad, and to write an article about it
- perform the detailed analysis of the data that is currently being collected in Krakow. I plan to go to Krakow for another secondment, when I could take samples of the surrounding methane sources and get their precise source isotopic signatures. The goal is to combine these with the long-term data, in order to relate the recorded methane elevations with the local sources.
- measure the water samples from the North Sea, collected in June 2018. The goal is to investigate marine methane sources, and their temporal variability.
- measure more samples from other sampling campaigns in Europe, that will be sent by other MEMO<sup>2</sup> students. I will then start to build a dataset of various methane source isotopic signatures on a larger scale.

#### 6.8.1.4 Collaborations (internal / external)

#### **TU Delft**

A project started by Dr. Julia Gebert (Faculty of Civil Engineering and Geosciences, TU Delft) is aiming at the quantification of the isotopic fractionation factor from the diffusion of methane through landfill soils. The experiments were carried out by her master student Tijmen Blom, during August 2018. They consisted in different soils chambers filled with soil of a range of densities and water content. Landfill gas (50 % CH4) was flown in these chambers and allow to diffuse in the soil. Any oxidation reaction was inhibited. Samples were taken when the methane concentration decreased to 40, 35, 30, 25 and 20%. I analysed the samples at UU lab for  $\delta^{13}$ C and  $\delta$ D -CH<sub>4</sub>. The results for  $\delta^{13}$ C showed a clear fractionation, whereas  $\delta D$  data follows an irregular pattern. Compared to what was expected, the influence of the soil parameters was not present. The results will be presented in Tijmen Blom's thesis, and will be written in a publication.

#### **Royal Netherlands Institute for Sea Research (NIOZ)**

The collaboration with NIOZ started during the PELAGIA cruise (June 2018), where samples were taken for the analysis of methane isotopes. More than 300 water samples, 37 atmospheric samples and 13 air samples from a gas seep were taken. The analysis of all air samples is achieved, but the water samples remain to be measured. The data will be analysed in collaboration with the research group at NIOZ, who collected the same water samples for drawing the depth profile of dissolved methane above a seep. The daily cycle was also captured, as the sampling was done regularly in the same column. The isotopic data will help in investigating the microbial processes behind this emission source.



#### 6.8.1.5 Risks and difficulties

No particular difficulty encountered this year.

#### 6.8.2 Deliverables

D2.1 - Isotopic measurements linked to common scale (month 18)

Progress: achieved, the report is being finalized.

Contribution: I participated in the preparation of the calibration tanks during my secondment at RHUL. I took care of their measurements in UU, and of their shipping to UHEI. I've also performed an intercomparison of our measurement systems of  $\delta^{13}$ C-CH<sub>4</sub> using 37 samples.

Problem faced: Finding samples to be measured in both labs can be challenging. They need to answer many criteria such as a sufficient volume, not being contaminated after measurement, being in the appropriate mole fraction range to avoid being dilute at one of the lab, and the sample holder to guarantee a stability of its content through a period long enough for the exchange and measurements to happen.

Deviations from the grant agreement: the final report will be delivered with a delay of 2 to 3 months.

**D2.2** - Improved isotopic source signatures of local and regional CH<sub>4</sub> emissions (month 36)

Progress: the data is being gathered through different campaigns and from previous measurements

Contribution: I have visited local sources by myself and measured a certain number of samples. We are also collecting samples from other partners in the EU, especially from RUG our other partner in the Netherlands.

Problem faced: Collecting samples from the other MEMO<sup>2</sup> students is a challenge, as it is not always part of their project and they might face technical issues.

Planning for the next reporting period: Put together the data we already have, to identify the areas and source types we should focus on.

**D2.3** - Publications on the use of isotopes for CH<sub>4</sub> source attribution in urban/industrial regions (month 36)

Progress: the data is being gathered through mobile measurements, mainly by RHUL.

Contribution: since I collected samples in the same locations during my secondment at RHUL, I have shared the data on  $\delta$ D-CH<sub>4</sub>, to provide more information for the source characterization using isotopes. In the future, further samples will be selected to be sent and measured at UU for having more data on  $\delta$ D-CH<sub>4</sub>.

Planning for the next reporting period: Sharing the data we are also collecting in other urban areas outside of the UK.

D2.4 - Publication on temporal and meteorological influences on CH<sub>4</sub> at fixed sites (month 42)

I am not involved yet in the achievement of this deliverable.

**D2.5** - Report providing isotopic maps at grid scale from inventories and atmospheric measurements (month 42)

This work has therefore has not being started yet.

#### 6.8.3 Training and network activities

#### 6.8.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
The art of scientific writing	05/11/2018 – 03/12/2018	Utrecht (UU)	Learning how to write scientific articles in a more effective way	-	Just participating	Still ongoing
Workshop on dispersion modelling	09/10/2018 – 10/10/2018	Heidelberg (UHEI)	Learning how to implement a Gaussian	?	Just participating	Successful training



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

## D5.9 MEMO<sup>2</sup> – Midterm Review Report

Workshop on isotopes	17/09/2018 – 19/09/2018	Egham (RHUL)	Learning the techniques for the measurements of CH <sub>4</sub> isotopes, from the sampling to the data interpretation	?	Presenting a poster and giving one presentation	Successful training
"Climate change in context" bachelor course	09/02/2018 – 12/04/2018	Utrecht (UU)	Experience in teaching	-	Teaching assistant	Positive feedback from the students. I will continue in 2019
1 <sup>st</sup> summer school	5/02/2018 – 16/02/2018	Schoorl, NL	Theoretical training on atmospheric processes and methane. Practical training on mobile measurements, sampling, tracer release, and data analysis	6	Just participating	Successful training

#### 6.8.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
RHUL	18/06/2018 – 14/07/2018	Egham, UK	RHUL	Isotopic measurement technique, inter- comparison between the labs and mobile surveys	Learning other techniques for isotopic measurements, and for sampling on the field.	The inter-comparison is done, together with the calibration tanks. A dataset of isotopic signatures of various sources in the UK was created. We will work further on the share and use of theis data.
AGH	16/05/2018 – 30/05/2018 (2 weeks, to be continued)	Krakow, PL	AGH	Installation of an in- situ measurement system Participation in the CoMet campaign in Silesia	Learning to install the IRMS and methane extraction system Getting data for isotopic characterisation of methane from the mine exhaust	Continuous data on ambient air is being collected. Other stays in Krakow are planned in December and February, for the sampling of local sources and the de- installation of the system. The results of the CoMet campaign are now available, and will be compared with the data gathered by other groups.

#### 6.8.3.3 Conferences

Conference name	Date (start – end, planned (when))	Locati on	Presentation (oral / poster)	Title of presentation	Authors (main author + co-authors)	Public available (yes / no) / web link
ICOS	10/09/2018 – 14/09/2018	Prague, (CZ)	Poster	Isotopic characterizatio n of methane from mine shafts in the Silesia region	Malika Menoud, Hossein Maazallahi, Mila Stanisavljevic, Thomas Röckmann, Jaroslaw Necki	Yes https://www.researchgate.n et/publication/ 327655309_Isotopic_charac terisation_ of_methane_from_mine_sh afts_in_the_Silesia_region
BBOS	25/10/2018 – 26/10/2018	Soester berg (NL)	Presentation	Continuous isotopic measurement of atmospheric methane	Malika Menoud, Carina vand der Veen, Thomas Röckmann, Bert Scheeren, Huilin Chen, Jaroslaw Necki	No

# 6.8.3.4 Measurement / sampling campaigns

Campaign	Date (start – end, planned (when))	Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
Krakow	10/12/2018 – 20/12/2018	Krakow, PL	AGH	Sampling of various methane sources in the surroundings of Krakow.	Isotopic characterisation of the main local sources influencing the methane	Air samples in aluminium bags, quantity to	The results will be combined with the continuous in-situ measurements performed through the winter, to



# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

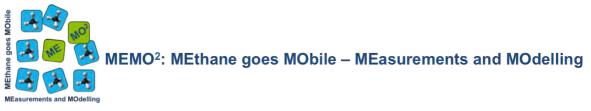
					elevations measured in the city.	be determined	potentially identify the sources of methane elevations.
Hamburg	07/11/2018 – 09/11/2018	Hamburg, DE	UU	Sampling of methane sources identified by mobile CRDS measurements	Isotopic characterisation of urban methane sources	Air samples in aluminium bags, 60	The measurements are to be performed.
MEMO <sup>2</sup> - RHUL	22/06/2018 – 05/07/2018	England, UK	RHUL	Sampling of methane sources identified by mobile CRDS measurements	Isotopic characterisations of various sources in the UK	Air samples in aluminium bags, 47	The data shows clear agreement between the source types, and the potential of isotopes for source identification. It is now to be shared and combined with RHUL and LSCE data.
CoMet	27/05/2018 – 29/05/2018	Silesia, PL	AGH, DLR	Sampling of plumes from coal mine shafts, identified by mobile CRDS measurements	Isotopic characterisation of methane from Polish coal mines	Air samples in aluminium bags, 43	The data shows a large heterogeneity in the isotopic signatures. It is now to be combined with the data from the other groups to better investigate the formation processes and influencing parameters.

## D5.9 MEMO<sup>2</sup> – Midterm Review Report

**6.8.4 Dissemination activities** So far, I did not publish my work within this project in a scientific journal. However, I've been doing more dissemination for the general public through social medias.

Other dissemination activities

Dissemination activity	Name	Date	Location	Type of audience	Size o audience	of
Blog on the MEMO <sup>2</sup> website	My secondment at RHUL	To be published	RHUL, UK	Public	100+	
Video on 2 social media platforms (Facebook and Instagram)	MEMO2 in England trailer	24/07/2018	RHUL, UK	Public	100+	
Blog on the MEMO <sup>2</sup> website	Installation of a CF- IRMS and methane extraction system	21/09/2018	AGH, PL	Public	100+	



# 6.9 ESR9 - The isotopic signature of urban CH<sub>4</sub> emissions

#### ESR9

The isotopic signature of urban CH<sub>4</sub> emissions

ESR	Julianne M. Fernandez, Julianne.Fernandez@rhul.ac.uk
Supervisor	Dave Lowry, <u>d.lowry@rhul.ac.uk</u>
Co-supervisor	Thomas Rockmann, <u>t.roeckmann@uu.nl</u>
Non-academic mentor	NA (Elementar)
Official start-end date	08/01/2018- 31/12/2020

#### 6.9.1 Scientific progress

#### 6.9.1.1 Project introduction and objectives

Urban methane emissions are poorly quantified fugitive emissions from gas leaks in the distribution network, old landfill sites and wastewater treatment plants. Mobile methane measurements will be made in three UK cities with different expected source contributions on a seasonal basis to assess the roles of the sources. The proposed cities are London, Birmingham, and a city in a location of Atlantic Ocean background air (possibly in south Wales). This will be coupled with continuous isotopic measurement campaigns at the RHUL site on the western edge of London during city outflow to assess the transfer of the emissions. Results will be compared to the UK city inventories, assessing source distribution and intensity.

Isotope measurements will be used to distinguish different components of urban CH<sub>4</sub> emissions. Such emissions include poorly quantified fugitive emissions from gas leaks in the distribution network, old landfill sites and wastewater treatment plants. The locations are normally not identified in inventories and the plumes from sources often merge, eventually producing a citywide plume of CH<sub>4</sub> that is transported some distance downwind before dispersing.

Samples collected in city campaigns by WP 2 students across Europe will be analysed for their  $\delta^{13}$ C-CH<sub>4</sub> and  $\delta^{2}$ H-CH<sub>4</sub> signatures. Results will be compared to the UK city inventories, assessing source distribution and intensity. Temporal distribution of sources will be assessed.

#### 6.9.1.2 Project results

#### 6.9.1.2.1 First year

During the 2018 Netherlands MEMO<sup>2</sup> School three mobile surveying campaigns took place (9<sup>th</sup>, 10<sup>th</sup>, and 12<sup>th</sup> of February 2018). Air samples were collected in FlexFoil bags via an air pump in the RHUL survey vehicle by the ESR9 student.

Background air samples were collected for every mobile surveying campaign. During these surveys, CH<sub>4</sub>, CO<sub>2</sub>, H<sub>2</sub>O, and C<sub>2</sub>H<sub>6</sub> concentrations were continuously monitored via a Picarro G2301 CRDS analyser with a A0941 Mobile Module and a LGR Ultra-portable methane / ethane analyser. Samples were collected when methane elevations were detected, at a range of concentrations between background and the peak of the plume.

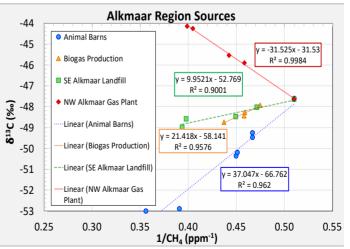
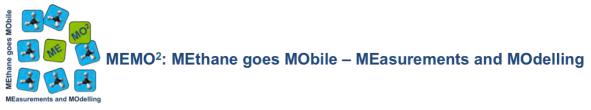


Fig. 6.9.1: Keeling plot of sampled sources during the February  $12^{th} 2018$  MEMO<sup>2</sup> School campaign.



Analysis of CH<sub>4</sub> concentration and  $\delta^{13}$ C-CH<sub>4</sub> were completed using a Picarro G2301 analyser and an IsoPrime Trace Gas continuous flow gas chromatograph isotope ratio mass spectrometer (CF GC- IRMS) at RHUL during the week following the campaigns. Keeling plot analysis was used to calculate the final isotopic signatures (Fig. 6.9.1) of the regional sources using measured background values. The calculated  $\delta^{13}$ C-CH<sub>4</sub> signatures for all the sources investigated are given in Table 6.9.1. The spatial differences can be seen in Fig. 6.9.2.

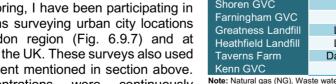
Table 6.9.1: 2018 MEMO<sup>2</sup> campaign results. Standard errors were calculated from the standard deviation of IRMS measurements of each sample divided by the square root of n.

Source	δ <sup>13</sup> C-CH <sub>4</sub> signature (‰)
Lefjeshoeve Farm	-67.19 ± 0.65
Biogas Production (Dome 1)	-58.14 ± 0.17
Biogas Production (Dome 2)	-58.19 ± 0.41
Biogas Farm Barn	-62.85 ± 0.56
Compost Farm	-69.24 ± 0.54
Animal Barns	-66.76 ± 0.52
NW Alkmaar Gas Plant	-31.53 ± 0.54
S Alkmaar Gas Plant	-38.99 ± 0.08
SE Alkmaar Landfill	-52.77 ± 0.22
Gas Release Test	$-44.49 \pm 0.06$

#### 6.9.1.2.2 Second year

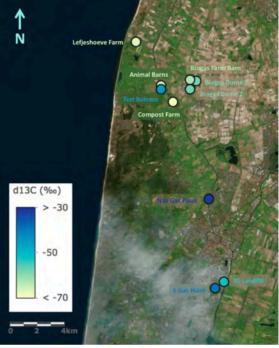
Technically this is not my second year, it is basically the 2<sup>nd</sup> semester of my first year. During the summer of 2018, students from UU (Malika Menoud) and USVQ (Sara Defratyka) joined the RHUL lab on 5 different surveys. These were conducted in UK areas of Staines, Oxford, Spelthorne, Kent, and Devon (Fig. 6.9.3). Continuous concentrations of [CH<sub>4</sub>] (Fig. 6.9.4), [CO<sub>2</sub>], [H<sub>2</sub>O], [C<sub>2</sub>H<sub>6</sub>] were measured and collected bag samples for isotopic analysis using the equipment and protocols listed under section 6.9.1.2.1. Results are seen in Fig. 6.9.5 and Table 6.9.2.

Since this past spring, I have been participating in various campaigns surveying urban city locations around the London region (Fig. 6.9.7) and at coastal regions of the UK. These surveys also used the same equipment mentioned in section above. Methane concentrations were continuously measured (Fig. 6.9.7 and 6.9.8). In the near future more of the boroughs in Fig. 6.9.7 will be survey for a larger coverage. Plans are to also sample



Note: Natural gas (NG), Waste water treatment plant (WWTP), Gas valve compound (GVC)

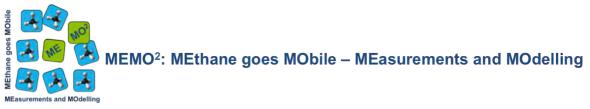
regions of Birmingham and an additional coastal location.



**Fig. 6.9.2:** Regional  $\delta^{13}$ C-CH<sub>4</sub> source signatures. Lighter shades indicate less enriched (more negative)  $\delta^{13}$ C-CH<sub>4</sub> values. Darker shades indicated more enriched  $\delta^{13}\text{C-CH}_4$ values (less negative). Signatures correlate to the values listed in Table 1.

**Table 6.9.2:** Regional  $\delta^{13}$ C-CH<sub>4</sub> source signatures of the MEMO2 RHUL summer surveys. Standard errors were calculated from the standard deviation of IRMS measurements of each sample divided by the square root of

Source	Source type	δ <sup>13</sup> C-CH <sub>4</sub> signature (‰)
Chertsey Waste	WWTP	-50.7 ± 0.3
The Causeway	NG pipeline	-37.6 ± 1.0
Modgen Sewage	WWTP	-50.7 ± 0.5
Windfield Lane	NG pipeline	-36.6 ± 2.1
Bicester Waste	WWTP	-52.7 ± 1.8
Milton Biogas	Biogas plant	-61.5 ± 0.6
Shoren GVC	NG	-38.3 ± 0.2
Farningham GVC	NG	-38.2 ± 1.2
Greatness Landfill	Landfill	-53.5 ± 0.7
Heathfield Landfill	Landfill	-60.7 ± 0.2
Taverns Farm	Dairy farm	-64.1 ± 1.2
Kenn GVC	NG	-40.2 ± 0.7



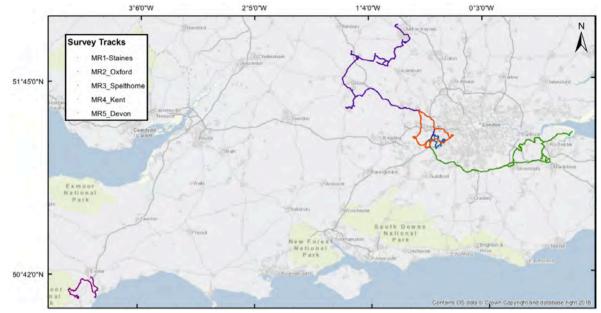
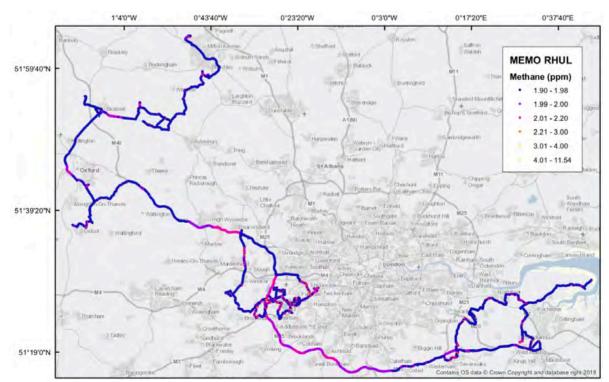


Fig. 6.9.3: Map of MEMO<sup>2</sup> RHUL 1-5 mobile tracks. Each colour represents a different survey.



**Fig. 6.9.4:** Methane concentrations of 1-4 surveys. The colour gradient represents the different  $CH_4$  in ppm. The lighter the colour the higher concentration and the darker indicate lower concentrations.



D5.9 MEMO<sup>2</sup> – Midterm Review Report



**Fig. 6.9.5:** Regional  $\delta^{13}$ C-CH<sub>4</sub> source signatures. Darker shades indicate less enriched (more negative)  $\delta^{13}$ C-CH<sub>4</sub> values. Lighter shades indicated more enriched  $\delta^{13}$ C-CH<sub>4</sub> values (less negative). Signatures correlate to the values listed in Table 6.9.1. High pressure natural gas pipelines are shown in red.

**Fig. 6.9.6:** UNC survey routes. Each survey is presented in a different colour.

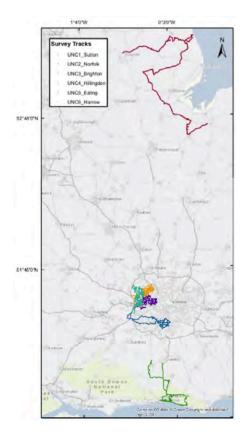
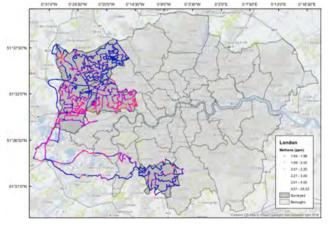
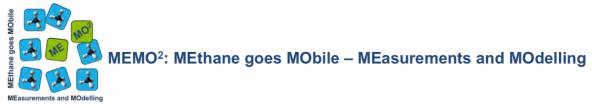


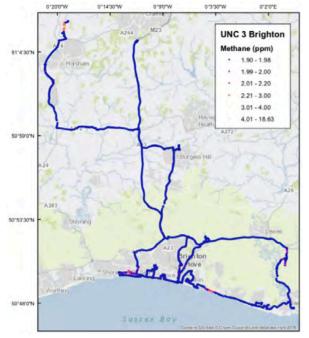
Fig. 6.9.7: London Borough surveys. The colour gradient represents the different  $CH_4$  in ppm. The lighter the colour the higher concentration and the darker indicate lower concentrations. Boroughs are outlined in grey. Hillingdon, Harrow, Ealing, and Sutton are in shaded.



#### 6.9.1.3 Future plans and expected results

My future plans are to finish my secondments at UVSQ and catch up on the mobile survey data and determine the methane ethane ratios. I also plan to have an established outline for a paper on UK city sources, which will partially contribute to D2.3 (Publications on the use of isotopes for CH<sub>4</sub> source attribution in urban / industrial regions). Data for this is ongoing from surveys that will be complete before next fall.





#### 6.9.1.4 Collaborations (internal / external)

Early spring, I joined a meeting with Elementar (assigned industry mentor). We discussed the progress of their IRMS system for hydrogen analysis. Though the company is very keen to work on the project, the development of this instrument may not fit into my postgraduate study.

This summer I collaborated with UU (Malika Menoud) and UVSQ (Sara Defratyka) during their secondments at RHUL. During this time, I shared field sampling and in lab isotopic measurement protocols, and learned more about sampling with LSCE's air core. Gas tanks were filled and analysed at RHUL to be passed to the other isotope institutions and field samples were collected, which both contribute to the intercomparing deliverable.

During this past fall I attended UU for my secondments. I worked closely with Malika and the lab engineer, Carina van der Veen, to learn measure both carbon and hydrogen using their CF IRMS. In the last weeks of my secondments, I joined Hossein Maazallahi for a campaign in Hamburg, Germany. We

Fig. 6.9.8: UNC 3 (Brighton, UK) survey. The colour gradient represents the different  $CH_4$  in ppm. The lighter the colour the higher the concentration, the darker indicate lower concentrations

exchanged and compared field sampling methods and shared how bag samples are collected for isotopic analysis. Bags to be analysed for isotopic ratios were collected for both UU and RHUL.

Starting late February, I will be joining Sara and Piotr Korbeń at UVSQ for another secondment. This will consist of 2 weeks of measurements and 2 weeks on modelling. Bag samples for isotopic measurements will be collected and measured at RHUL.

#### 6.9.1.5 Risks and difficulties

Some difficulties I have are finding adequate resources for ADHD and dyslexia, specifically finding counsellors / study mentors specializing on adults and postgraduates.

#### 6.9.2 Deliverables

ESR 9 is involved in the following deliverables: D1.2 / D1.4 / D2.1 / D2.2 / D2.3 / D2.4 / D2.5

**D1.2** - Report/publication on CH<sub>4</sub> emissions from wetland and lakes in Sweden (month 30)

This summer I helped a former masters student measure  $CH_4$  emissions from a freshwater lake at RHUL using our mobile instruments and a new chamber. The protocols for this project can possibly contribute to this deliverable.

**D1.4** - Improve emissions factors for different source categories from mobile measurements (month 42) I have been contributing to surveys collecting mobile data that will be compiled for this deliverable. There are still many more surveys to be conducted for this.

D2.1 - Isotopic measurements linked to common scale (month 18)

Standard tanks have been made to be measured between the universities responsible for isotopic measurements. I have assisted in the analysis of  $\delta^{13}$ C-CH<sub>4</sub> at UU during my secondments in October.

**D2.2** - Improved isotopic source signatures of local and regional CH<sub>4</sub> emissions (month 36)

I have been contributing to surveys collecting data mobile data that will be compiled for this deliverable. There are still many more surveys to be conducted for this.



**D2.3** - Publications on the use of isotopes for  $CH_4$  source attribution in urban / industrial regions (month 36)

The majority of the survey I have been participating in are focused around urban areas.

D2.4 - Publication on temporal and meteorological influences on CH<sub>4</sub> at fixed sites (month 42)

Over the past summer I helped set up and analyse diurnal isotopic measurements on the GC-CG-IRMS while it ran parallel with Sara's (UVSQ) air core.

**D2.5** - Report providing isotopic maps at grid scale from inventories and atmospheric measurements (month 42)

So far, I have been learning most of the skills to do this. For example, learning ArcGIS and all the isotopic training will aid me to contribute to this deliverable in the near future

#### 6.9.3 Training and network activities

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
Yorkshire campaign training survey	2018-01-29	RHUL	Lab Field Training		Learned how to prepare and use mobile instruments, collect bag samples, navigate to find methane plumes.	NA
Greenhouse Gas Laboratory training	2018-02-02	RHUL	Laboratory Training		Basic safety training. Introduced to lab instruments to measure [CH <sub>4</sub> ] and $\delta^{13}$ C- CH <sub>4</sub> using Yorkshire samples. Learned how to calibrate mobile instruments.	NA
1st MEMO2 school – Methane measurements and modelling	2018-02-05	MEMO2	External Training Session		The school is part of the European H2020 ITN-ETN project MEMO <sup>2</sup> , GA No. 722479. It was a two-weeks school, associated with an intensive measurement campaign, including preparation, lectures and practical.	NA
Greenhouse Gas Laboratory training	2018-02-20	RHUL	Lab Training		Hands on training for air bag samples measuring [CH <sub>4</sub> ] and $\delta^{13}$ C-CH <sub>4</sub> .	NA
Data correction training	2018-02-22	RHUL	Lab Training		Data corrections, data management & organization. Introduced to data software and Filezilla and Teamshare.	NA
Department of Earth Sciences Summer Seminar	2018-03-07	RHUL	Poster Presentation		Presented a poster on current research plans.	NA
Writing Literature Reviews	2018-03-14	RHUL	RDP session		Overview of basic steps on writing a literature reviews.	NA
1st MEMO2 Annual Meeting, Dübendorf, Switzerland	2018-03-21	MEMO2	Oral Presentation Poster Presentation		MEMO <sup>2</sup> Oral presentation on sample results collected during the MEMO <sup>2</sup> school. Poster presentation summarizing the same results.	NA
FAAM Facility Safety Training	2018-04-11	FAAM	External Training Session		Introduced to North Sea flight research and preparation for flight surveys. Took part in Safety training involving the facility, equipment.	NA
Department of Earth Sciences Postgraduate Research Seminar	2018-06-05	RHUL	Presentation		Presented current research results, progress, and plans for next few months	NA

#### 6.9.3.1 General training events



lsotope workshop	2018-09-17 to 2018-09-19	RHUL	Understand the principles behind mass spectrometry and other isotope measurement techniques. Learn how to collect, analyse and interpret the isotopes of methane		NA
Ploom modeling Workshop	2018-10-08 to 2018-10-10	UH			NA

## 6.9.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
UU - IMAU	2018-09-24 to 2018-10-21	Utrecht, NL	UU	Training on IRMS system, protocols, surveying		NA
UVSQ	2019-02-25 to 2019-03-22		UVSQ	Mobile survey sampling		NA

#### 6.9.3.3 Conferences

Conference name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation	Authors (main author + co-authors)	Public available (yes / no) / web link
1st MEMO2 Annual Meeting	2018-03-21 to 2018-03-23	Dübendorf, Switzerland	Oral & Poster	Isotopic signatures of the 2018 MEMO <sup>2</sup> school campaign	Fernandez, J.M.	no

# 6.9.3.4 Measurement / sampling campaigns

Campaig n	Date (start – end, planned (when))	Locatio n	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
Yorkshire	2018-01-xx to 2018-01-xx	Yorkshir e, UK	RHUL training	Survey of area to have fracking activity	Conduct and collect data for study on methane before, during and after	3 bag samples for δ <sup>13</sup> C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	
Schoorl, NL	2018-02-09 to 2018-02-12	Schoorl, NL	MEMO2 1 <sup>st</sup> school	Training survey with MEMO2 group and trace gas release test	Train on working on surveys with all MEMO2 survey vehicles	35 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	See above section 1.2.1
UNC 1	2018-05-03	Sutton, UK	RHUL	Urban city survey	Measure and locate urban CH₄ sources	3 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	
UNC 2	2018-06-12	Norfolk / Lincolns hire Termina Is, UK	RHUL	LNG terminals survey		18 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	



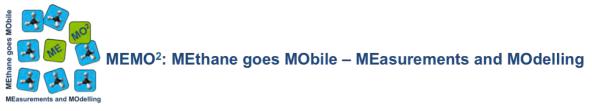
# MEMO<sup>2</sup>: MEthane goes MObile – MEasurements and MOdelling

UNC 3	2018-08-29	Brighton , UK	RHUL	Urban city survey	Measure and locate urban CH₄ sources	15 bag samples for δ <sup>13</sup> C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	
UNC 4	2018-08-29	Hillingdo n, UK	RHUL	Urban city survey	Measure and locate urban CH <sub>4</sub> sources	10 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	
UNC 5	2018-10-25	Ealing, UK	RHUL	Urban city survey	Measure and locate urban CH₄ sources	4 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	Analysis in process
UNC 6	2018-10-25	Harrow, UK	RHUL	Urban city survey	Measure and locate urban CH <sub>4</sub> sources	2 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	Analysis in process
MEMO RHUL 1	2018-0622	Egham/ Staines, UK	RHUL	Local source survey	Measure and locate urban CH <sub>4</sub> sources with visiting UU & UVSQ student	6 bag samples for $δ^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	
MEMO RHUL 2	2018-06-26	Oxford, UK	RHUL	Urban city survey	Measure and locate CH <sub>4</sub> sources of waste facilities with visiting UU & UVSQ student	6 bag samples for $δ^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	Some samples for IRMS RHUL & UU comparison
MEMO RHUL 3	2018-06-27	Spelthor ne, UK	RHUL	Urban city survey	Measure and locate urban CH <sub>4</sub> sources with visiting UU & UVSQ student	9 bag samples for δ <sup>13</sup> C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	Some samples for IRMS RHUL & UU comparison
MEMO RHUL 4	2018-06-28	Kent, UK	RHUL	Urban city survey	Measure and locate urban & waste CH <sub>4</sub> sources with visiting UU & UVSQ student	12 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	Some samples for IRMS RHUL & UU comparison
MEMO RHUL 5	2018-07-05	Devon, UK	RHUL	Heathfield landfill survey	Measure landfill CH4 with visiting UU & UVSQ student	14 bag samples for $\delta^{13}$ C- CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	Some samples for IRMS RHUL & UU comparison
Ham 1, 2, & 3	2018-10-18 to 2018-10-20	Hambur g, German y	UU	Urban city survey	Assist and learn protocol of UU surveys to measure and locate urban CH <sub>4</sub> sources.	17 bag samples for $δ^{13}$ C- CH <sub>4</sub> & $δ^2$ H-CH <sub>4</sub> Continuous measurements of [CH <sub>4</sub> ], [CO <sub>2</sub> ], [H <sub>2</sub> O], [C <sub>2</sub> H <sub>6</sub> ]	Analysis in process

### D5.9 MEMO<sup>2</sup> – Midterm Review Report

## 6.9.4 Dissemination activities

Except for the contributions to the conferences no scientific publications or other dissemination activities so far from the ESR.



# 6.10 ESR10 – Integration of mobile measurement data in monitoring, reporting, and verification (MRV) of key CH<sub>4</sub> sources in GHG emission reporting across Europe

#### ESR10

Integration of mobile measurement data in monitoring, reporting, and verification (MRV) of key CH<sub>4</sub> sources in GHG emission reporting across Europe

ESR	Hossein Maazallahi, <u>h.maazallahi@uu.nl</u>
Supervisor	Prof. dr. Thomas Röckmann, t.roeckmann@uu.nl
Co-supervisor	Dr. Hugo Denier van der Gon, hugo.deniervandergon@tno.nl
Non-Academic mentor	Ir. Heijo Scharff, <u>h.scharff@afvalzorg.nl</u>
Official start – end date	01.09.2017 – 31.08.2021

#### 6.10.1 Scientific progress

#### 6.10.1.1 Project introduction and objectives

The ESR in this WP will utilize the data from other MEMO<sup>2</sup> partners to link their results to emission reporting at various scales (site-national-European). This is implemented by an intensive 30% secondment to the non-academic partner TNO, who is responsible for emission registration and reporting in the Netherlands, and provides the European scale CH<sub>4</sub> emission inventories for the Copernicus Atmospheric Monitoring Services. The ESR will also carry out mobile measurements together with the non-academic partners OONKAY and AD in order to develop (new) monitoring, reporting and verification (MRV) protocols for emissions from landfills. We will focus in particular on novel low-cost solutions using mobile instrumentation, which could unlock the reduction potential and contribute to transparent more accurate reporting. The goal of this project is to achieve an integration of the national and facility level MRV that ultimately results in contributing to GHG emission reduction measures being implemented and their effectiveness being quantified. Once the proof of concept is achieved, the method will be applied in several data-poor locations (Eastern Europe) to provide baseline emission data and quantify the emission reduction potential. The results of the MEMO<sup>2</sup> project will flow into better and updated CH<sub>4</sub> emission inventories from the InGOS project and the TNO-CAMS European emission inventory.

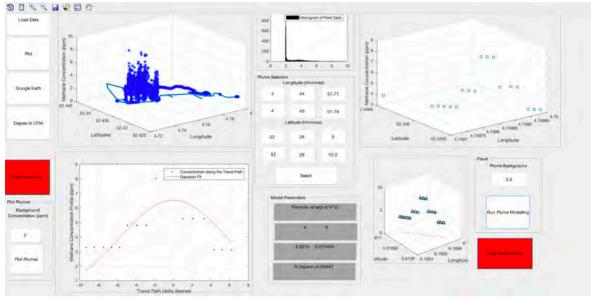
#### 6.10.1.2 Project results

#### 6.10.1.2.1 First year (March2017-March2018)

My PhD project is involved in improving the bottom-up inventory of MEMO<sup>2</sup>. For this purpose, I need to work in close collaboration with Netherlands Organisation for Applied Scientific Research (TNO), where I have already started my secondment since January 2018. However, acting on the discussion I had with my supervisors, we decided to put more focus on holding campaigns mostly in the first part of my PhD. For this report there is has not been particular outputs out my works related to the inventories.

Acting on the fact that each campaign generates large amount of data, first I noticed about a need to have a Graphical User Interface (GUI) in which it would be easy to pick a plume and quantify emission based on Gaussian plume model (Fig. 6.10.1). We use Picarro instrument for mobile measurements, and the Picarro outputs along with geographical dataset are the input for this GUI. This MATLAB enable users to investigate the data in a 3-D plot and find areas with concentration higher than a specific value. It is also possible to export the measurement rout into Google Earth, and also convert the data to Universal Transverse Mercator (UTM) system. The user is also capable to input geographical coordinates of a box around a specific area and model the methane concentration elevation using specific model. At the moment, the modelling is based on Gaussian model, however, it is possible to model the plume based on other models.





D5.9 MEMO<sup>2</sup> – Midterm Review Report

Fig. 6.10.1: A MATLAB tool for modelling plume using MATLAB

Initially I had mostly campaigns on landfills (Table 6.10.1), but there were also some field works during the first MEMO<sup>2</sup> to measure emissions from farm. There have been several driving tours inside the Utrecht city to with high precision methane analyser on board (Picarro).

#### Landfill Measurements

Methane elevations were measured using instrument on-board the UU van (Fig. 6.10.2). In Fig. 6.10.3, distribution of methane, carbon dioxide and water vapour using inverse distance weighted (IDW) interpolation across the landfill was produced.

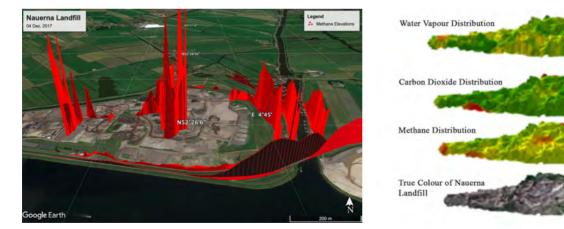
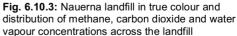


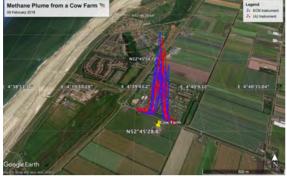
Fig. 6.10.2: Nauerna landfill campaign, 4 Dec. 2017



#### **MEMO<sup>2</sup>** School Campaigns

During the 1<sup>st</sup> MEMO<sup>2</sup> school, there were three days of campaigns. During the campaign on Friday, 09 February 2018, there were several transects in front of Lefjeshoeve farm and a biogas plant (yellow pins in Fig. 6.10.4 and 6.10.5). The methane elevations from the farm and the biogas plant are shown in Fig. 6.10.5, and Fig. 6.10.6 respectively.





**Fig. 6.10.4:** Methane Elevations emitted from Lefjeshoeve farm (yellow pin); measured by ECN (blue) and UU (red) instruments

Methano Plume from a Bio-Gas Plant to Howary 2018 Particular 2018 Parti

Fig. 6.10.5: Methane Elevations emitted from the biogas plant (yellow pin); measured by ECN (blue) and UU (red) instruments

The average over the transects and later the Gaussian plume were applied over the average lines from farm and biogas areas (Fig. 6.10.6).

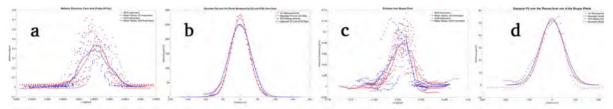


Fig. 6.10.6: Averaging over the transects in front of the farm (a) and the biogas plant (c); applying Gaussian model over the average lines for farm (b) and biogas area (d)

#### **City measurements:**

In the first year, city measurements were performed in Utrecht (NL) in February, March and April 2018. The whole city was covered using Picarro G2301 instrument. Completing measurements in Utrecht and translating concentration to emission rate using an empirical equation (von Fischer et al., 2017).

Translating concentrations toe mission rates in Utrecht (Fig. 6.10.8)

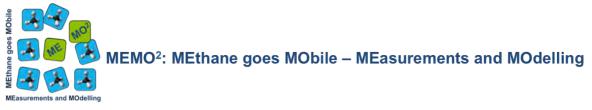


Fig. 6.10.7: Methane mobile measurements across Utrecht,  $\ensuremath{\mathsf{NL}}$ 



Fig. 6.10.8: Three emission categories in Utrecht; High (red), Medium (orange), Low (Yellow)

ane Elevations emitted from the



#### 6.10.1.2.2 Second year

#### **COMET Campaign, PL:**

Participating in this campaign which was considered as my secondment at AGH university, was in May and June 2018. I with 4 other MEMO<sup>2</sup> PhDs from UU, AGH, Heidelberg, and LSCE were contributing in mobile measurements and isotopic analysis with other scientific groups with main coordination from DLR, Germany. In September 2018, UU, UHEI, and AGH university got together in Heidelberg to work on the mobile measurements and also instrument calibrations. In Fig. 6.10.10, which shows the UU measurements on 6<sup>th</sup> of June, 2018, it is displayed that in all the four transects downwind the mining area in south-west part of the Poland, the enhancement was observed. The higher concentration within the urban area in Ostrava n Czech Republic is also depicted.

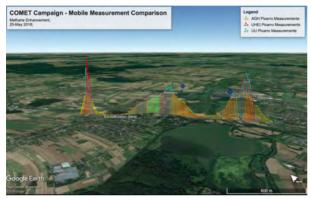


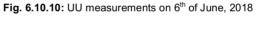
Fig. 6.10.9: A figure showing instruments measurements during the COMET campaign

#### Ship cruises, North Sea

From 21<sup>st</sup> June until 2<sup>nd</sup> July 2018 I participated in a scientific group form Royal Netherlands Institute for Sea Research (NIOZ) onboard research vessel (RV) Pelagia. During this campaign I was measuring continuously with two picarro (G2301, G4302) and also took atmospheric samples for further lab isotopic measurements at IMAU, Utrecht University. You may read stories about this campaign on MEMO<sup>2</sup> website: https://h2020-memo2.eu/2018/06/14/memo2-goes-marine/. Further results will be provided later.

From 17<sup>th</sup> of July until 20<sup>th</sup> of July there was a cruise to the North Sea to study methane emissions from the oil/gas platforms. I participated with the two picarro (G2301, G4302) to collaborate with the collagues from TNO. This cruise was funded by Netherlands Oil and Gas Exploration and Production Association (NOGEPA) and coordinated by TNO. I had a vlog interview about this cruise which is accessible on this link:

https://www.youtube.com/watch?v=fcsXxEwIF6w Further results will be provided later.



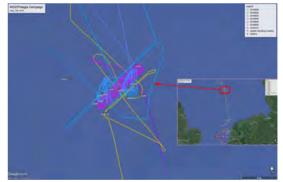


Fig. 6.10.11: NIOZ/Pelagia Cruise routs and sampling locations



Fig. 6.10.12: TNO/NOGEPA Cruise routs and sampling locations



#### **Oktoberfest, Munich, DE:**

From 24<sup>th</sup> October until 2<sup>nd</sup> September. I participated in a campaign in collaboration with Technical University of Munich (TUM): Prof. Jia Chen, to understand the methane emissions from the Oktoberfest event in the city. I had two picarro (G2301, G4302) and TUM were measuring with Fouriertransform infrared spectroscopy (FTIR) instruments to understand the total column of methane concentration around the festival. Further results will be provided later.

#### Plume Modelling Workshop:

In September, MEMO<sup>2</sup> students had plume modelling workshop hosted by Heidelberg University, Heidelberg, Germany. In this workshop, we had lectures on how to model the plume in different situations, and we also had an extensive exercise on implementing Gaussian model on a release test, to estimate the release test rate. As it is shown in Fig. 6.10.14, the release test was held in the middle of a waste water treatment plant.

The plumes were measured, smoothed and peaks were extracted out of the plumes. At the end, the release rate was identified, but we noticed that the real release rate was three times more than the gaussian model output.



Fig. 6.10.13: Measurements with Picarro G4302, Oktoberfest 2018



Fig. 6.10.14: The release test; release locations and plumes

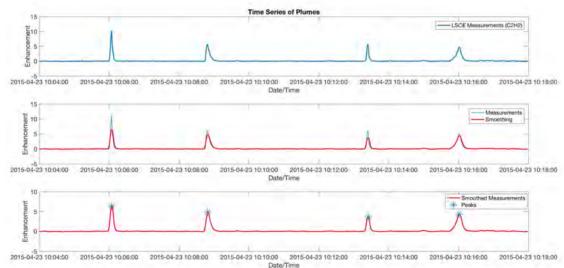
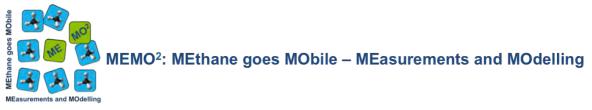


Fig. 6.10.15: Peaks time series



#### Hamburg, DE

The main purpose of the Hamburg campaign (Fig. 6.10.16) - in collaboration with the Environmental Defense Fund (EDF, <u>https://www.edf.org</u>) – was to understand the contribution of mid/downstream of oil and gas sector. Hamburg is one a very good examples of European cities which has not only the mid/downstream oil and gas sector but also some wells in south east part of the city (upstream of oil and gas industry). In this campaign, two Picarro instruments; G2301 and G4302 were used, which allowed to cover the Hamburg with the following species measurements; CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, and H<sub>2</sub>O. Further results will be provided later. In addition to the mobile measurements, isotopic bag samples were taken for further lab measurements.

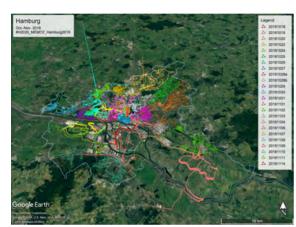


Fig. 6.10.16: Hamburg campaign, Oct. Nov. 2018



Fig. 6.10.17: Isotopic bag samples locations, Hamburg, DE

#### 6.10.1.3 Future plans and expected results

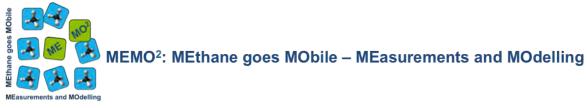
Training: I'll participate in the second MEMO<sup>2</sup> school which will be held in at LSCE, Paris. This workshop is mainly about how relationships of researcher and policy makers in global warming framework.

Scientific: In the second year I had campaigns in different field environments. In the coming months, I'll work on the data evaluation. E.g. about the COMET campaign, we have already started the comparison between the measurement analysers from AGH, UHEI, LSCE, and UU. We will continue to understand the plume shape by comparing the flight measurements from DLR (<u>https://www.dlr.de</u>). Regarding the cruise, I'll put my time to understand the emission rates from oil/gas platforms and also natural gas seeps. Regarding the city measurements, there would be comparison between the measurement held in Utrecht and Hamburg, and evaluation on methane emission will be discussed. Regarding the Oktoberfest, there would be estimates on the emission from this festival and measurement from TUM will be compared with measurements from UU.

#### 6.10.1.4 Collaborations (internal / external)

a) During the EGU, 2018, Vienna, Austria, I met people from different companies, Picarro, Aerodyne, wind sensor producer companies, etc. I have been in contact with Picarro support team couple of times regarding the instruments we have (picarro G2301 and picarro G4302). The campaigns hold in Munich was also with participation of Picarro, as they sent a demo instrument (Picarro G4302) to the Technical University of Munich (TUM), and I introduced the instrument to the TUM as I had been already familiar with the instrument. I also have been in contact with people from Aerodyne especially for the cruise we had to the North Sea; TNO/NOGEPA, and learned from the experiences Aerodyne got in their own cruise. After discussing with different wind sensor companies, and also having discussions with other MEMO<sup>2</sup> PhDs, we decided to purchase 2-D Gill GMX200 anemometer.

b) During the ICOS conference, Prague, 11-14 Sep. I met people from Picarro, and MIRICO who I met also at the EGU, 2018, Vienna, Austria. We updated each other about the recent activities we had. I had a conversation with MIRICO and explained them the situation in cruise. Since last year, we have



been trying to find an opportunity to collaborate with each other (having MIRICO instruments in MEMO<sup>2</sup> campaigns), but according to the logistics it has not been happened yet.

c) During the NIOZ/Pelagia cruise, I mostly was in collaboration with new researcher from TNO, NIOZ, and Vrije Universiteit (VU) Amsterdam. The NIOZ/Pelagia cruise was really interesting as different groups with variety of specialties got together on a research vessel. E.g. the colleagues from TNO were busy with the research related to geology while people from VU Amsterdam were researching the DNA, and I was measuring the air samples. On the other hand, researchers from NIOZ and Utrecht University (UU) were busy with water sampling. The water samples are currently in measurements progress at UU, NIOZ, and GEOMAR.

d) During the TNO/NOGEPA cruise, I collaborated with people from TNO/ECN, TOTAL, and NOGEPA. It was a cruise with order of NOGEPA. After the cruise we had a meeting in Den Haag, about the first cruise and discussed about the data evaluation. The collaboration is continuing and we continue to work on understanding the emission factors from each platform.

e) During the measurements in Munich, I collaborated with people form Technical University of Munich (TUM) to understand the total emission from Oktoberfest activity. We are now working to write a scientific paper for the PEFTEC, 2019 conference.

f) Regarding the Hamburg campaign:

I met people from Environmental Defense Fund (EDF), we have been in contact with each other, and discussed about the Hamburg several times either online or in person.

I got connected to people at Meteorological Institute (MI) at the University of Hamburg where I had access to the facilitates to hold the Hamburg campaign. I got information from Bilwilder mast of Hamburg from the MI, which will be useful in further evaluation of Hamburg measurements.

I got connected Max-Planck Institute for Meteorology (MPI-MET) in Hamburg where I also had access to the facility of the institute. The people at MPI-MET), are involved in computational modelling. Having a connection with the MPI-MET, I was introduced to Helmholtz-Zentrum Geesthacht (HZG) where I discussed about the difficulties in understanding the emissions from city measurements.

I had a meeting with GazNet company in Hamburg, where my supervisor and I discussed the Hamburg campaign in a meeting with them and we will work together to reduce the fugitive emissions from gas distribution system and understand the correlation between natural gas distribution system and amount of gas escape from the system.

One day I went to the GEOMAR and delivered NIOZ/Pelagia water samples and also presented MEMO<sup>2</sup> and Hamburg campaign at GEOMAR. I brought my analysers to GEOMAR and we had a measurements with a cylinder from GEOMAR to compare our instruments with each other (Picarro and Los Gatos Research (LGR)).

#### 6.10.1.5 Risks and difficulties

There is no risk or difficulty faced at the moment.

#### 6.10.2 Deliverables

Within MEMO<sup>2</sup>, ESR 10 is involved in the deliverables D1.4, D1.5, D2.5, D3.2, and D3.4

**D1.4** - Improved emission factors for different source categories from mobile measurements (month 42) Acting on the group work we had at Heidelberg University related to the COMET campaign, and the other campaign and my preliminary studies over the inventories.

D1.5 - Report on harmonized method for mobile CH<sub>4</sub> and 13CH<sub>4</sub> (month 18)

No contribution by ESR 10.

**D2.5** - Report providing isotopic maps at grid scale from inventories and atmospheric measurements (month 42)

No contribution by ESR 10.

D3.2 - Improved bottom-up European CH<sub>4</sub> emissions (month 30)



The ESR just started to contribute, this will be intensified in the next reporting period. **D3.4** - Top-down estimates of EU-scale CH<sub>4</sub> emissions (month 42) No contribution by ESR 10.

# 6.10.3 Training and network activities

#### 6.10.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
Art of Presenting Science	Nov. 2017	UU, Utrecht, NL	Developing scientific presentation skills	2	Participant	
Art of Scientific Writing	Nov. 2017	UU, Utrecht, NL	Developing scientific writing skills	2	Participant	
MEMO2 first summer school	5th–16th February 2018	Schagen, NL	Understanding theoretical background of the plumes, team-work measurements, increasing synergy between students, meeting MEMO2 people for the first time	6	Participant	
Climate change in context	Feb. – April 2018	UU, Utrecht, NL	Assisting in a BSc course	10hr per week	Teaching Assistant	
MEMO2 Annual Meeting	22-23 March, 2018	EMPA, Zurich, CH	Presenting works in the first month of the MEMO2, discussions over the results and how to improve the progress quality and pace.		Participant	
National Iranian Gas Company	August 2018	National Iranian Gas Company, Tehran, IR	Introducing MEMO2, with focus on the projects and campaigns held within the MEMO2		Giving a Talk	
Plume Modelling Workshop	17-19 Sep., 2018	UHEI, Heidelberg, DE	Understanding the theoretical background of the plume, we also had a programming exercise related to gaussian plume modelling	-	Participant	
Talk at Max-Planck Institute	29 Oct., 2018	MPI-MET, Hamburg, DE	Introducing MEMO2 and the Hamburg campaign		Giving a Talk	
Talk at GEOMAR	05 Nov., 2018	GEOMAR, Kiel, DE	Introducing MEMO2 and the Hamburg campaign		Giving a Talk	MEMO2 partner
Talk at Meteorological Institute of Hamburg University	13 Nov., 2018	MI, UHH, Hamburg, DE	Introducing MEMO2 and the Hamburg campaign with the first preliminary results		Giving a Talk	

## 6.10.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
TNO	January 2018	Utrecht	TNO	Working on bottom-up inventories	In progress	In progress
AGH	May-June 2018	Krakow, PL	AGH	Mobile methane measurements, COMET campaign	Improving synergy between MEMO <sup>2</sup> PhDs	Started collaboration with DLR and see how ground base measurements and flights get along with each other

#### 6.10.3.3 Conferences



D5.9 MEMO <sup>2</sup> – Midterm	Review	Report
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Conference name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation	Authors (main author + co- authors)	Public available (yes / no) / web link
BBOS	25/10/2017- 27/10/2017	Hotel Erica in Berg en Dal	Poster	Methane Goes Mobile, Measurements and Modelling (MEMO <sup>2</sup> )	A. Raznjevic, H. Maazallahi, M. Krol, C. van Heerwaarden, D. Brunner, S. Walter, H. Denier van der Gon, T. Röckmann	No
Industrial Methane Measurements	29/11/2017- 30/11/2017	Antwerp, Belgium				
EGU	08/04/2018- 13/04/2018	Vienna, Austria	MEMO <sup>2</sup> Session	Integration of mobile measurement data in monitoring, reporting and verification (MRV) of key methane sources in GHG emission reporting across Europe	H. Maazallahi, H. Denier van der Gon, T. Röckmann	No
ICOS	11/09/2018- 14/09/2018	Prague, Czech Republic	Poster	Methane Emission Mapping and Evaluation across Utrecht City, the Netherlands	H. Maazallahi, M. Menoud, C. van der Veen, H. Denier van der Gon, T. Röckmann	No

### 6.10.3.4 Measurement / sampling campaigns

Campaign	Date (start – end, planned (when))	Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
Nauerna Landfill	4 and 15 December 2017	Amsterdam, NL	Afvalzorg	Landfill Measurements	Quantifying methane emission from landfills		
Zeeasterweg Landfill	29 November 2017, 5 December 2017	Lelystad, NL	Afvalzorg	Landfill Measurements	Quantifying methane emission from landfills		
Braambergen Landfill	29 November 2017, 5 December 2017	Almere, NL	Afvalzorg	Landfill Measurements	Quantifying methane emission from landfills		
Campaigns during MEMO2 School	09, 10, 12 February 2018	Schagen, NL	ECN	Farm, Biogas, city measurements	Quantifying and identifying methane emission from different sources		
Utrecht	Feb-Apr 2018	Utrecht	UU	City Measurements	Quantifying and identifying methane emission sources across the Hamburg		Comparing the results with other cities.
COMET	May-June 2018	Poland	AGH	Measurements from Coal mining activities	Finding methane emission rate form different shaft in the area		



NIOZ/Pelagia	June-July 2018	North Sea	NIOZ	Continuous methane measurements from oil/gas platforms and natural seeps	Understanding methane emissions from anthropogenic and natural sources	13 Glass flask and 37 Bag Samples	
TNO/NOGEPA	July 2018	North Sea	TNO/NOGEPA	Continuous methane measurements from oil/gas platforms	Understanding methane emissions from oil and gas extraction platforms	22 Bag samples	
Oktoberfest	Sep., Oct. 2018	Munich	Technical University of Munich (TUM)	Stationary and mobile measurements from Oktoberfest festival	Understanding methane emissions from the Oktoberfest		
Hamburg	Oct. Nov. 2018	Hamburg	University of Hamburg and Max-Planck Institute	City Measurements	Quantifying and identifying methane emission sources across the Hamburg	104 bag samples (81 UU and 23 RHUL)	

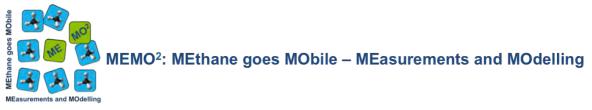
#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.10.4 Dissemination activities

Dissemination activity	Name	Date	Location	Type of audienc e	Size of audienc e
Interview regarding the TNO/NOGEPA cruise, which was broadcasted on youtube	Wat is de rol van het MEMO2 project? - VLOG #9 Offshore Methaan Meetprogramma	7 Oct. 2018	Interview at IMAU, UU, online on youtube: https://www.youtube.com/watch?v=fcsXxE wIF6w&t=1s	General Public	

#### References

J. C. von Fischer et al., "Rapid, vehicle-based identification of location and magnitude of urban natural gas pipeline leaks," Environmental Science & Technology, vol. 51, no. 7, pp. 4091–4099, Mar. 2017.



## 6.11 ESR11 – High-resolution modelling of CH4 dispersion

#### ESR11

#### High-resolution modelling of CH<sub>4</sub> dispersion

ESR	Anja Ražnjević, <u>anja.raznjevic@wur.nl</u>
Supervisor	Chiel van Heerwaarden, chiel.vanheerwaarden@wur.nl
Co-supervisor	Maarten Krol, maarten.krol@wur.nl
Non-Academic mentor	Harm Jonker H.J.J.Jonker@TUDelft.nl
Official start – end date	01/09/2017 – 01/09/2021

#### 6.11.1 Scientific progress

#### 6.11.1.1 Project introduction and objectives

In this project, direct numerical simulations (DNS) obtained from the MicroHH model will be used to simulate emissions of methane from various sources (point, line, diffuse) under different meteorological conditions and over surfaces with different heterogeneities and surface roughness's and compared against other techniques such as RANS. The simulations will then be used together with the measurements in estimation of the source strength. Virtual vehicles and unmanned aerial vehicles (UAVs) will also be simulated within 3D dispersion fields in order to help plan measurement campaigns. In estimation of the sources modeling and measuring techniques can be combined. Simulations with numerical models can help with interpreting measurements and planning campaigns.

The goal of this project is to use DNS in conjunction with the measurements and other modelling techniques to further advance our understanding of atmospheric dispersion

#### 6.11.1.2 Project results

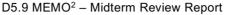
#### 6.11.1.2.1 First year

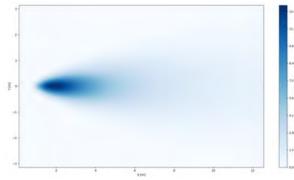
The beginning of the PhD project was dedicated to literature research on different dispersion modelling techniques in general and with special attention to current development with simulations of pollutant dispersion using DNS and different RANS approaches. The goal was, apart from familiarizing with the field, to write a research proposal for the SENSE graduate school of Wageningen, where PhD students are a part of.

Most widely used models for describing dispersion of a scalar are of Gaussian model type. They are computationally cheap and easy to use, but their ability to describe the structure of the plume close to the source is questionable (Tominaga & Stathopoulos, 2013). Nevertheless, a simple Gaussian plume model was built using Phyton with the goal of better understanding dispersion processes and having a simple model at hand for comparison with the DNS results (Fig. 6.11.1). Since the ESR was only familiar with Matlab, the exercise was used for familiarization with the Python programming language. Model simulates dispersion of a non-reactive scalar from a continuous point source (elevated or on the ground) with the uniform wind forcing in the longitudinal direction. The model is based on Pasquill stability classes and with Pasquill-Gifford formula for calculating dispersion coefficients in y and z direction. It is assumed that the advection with the mean wind dominates dispersion in the x direction.

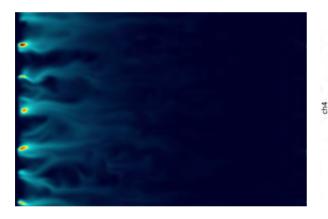
In preparation for the first MEMO<sup>2</sup> school, a first simple DNS case was set up in MicroHH, after Moser et al. (1999). The simulation mimics the flow in the atmosphere close to the surface in very windy conditions (> 7 m/s) which is a typical condition on the coast of the Netherlands where the school was held and a release experiment was performed. We simulated the emission of methane from a point (Fig. 6.11.2) and a line (Fig. 6.11.3) source. From the presented figures, the difference between a simple Gaussian plume model and plumes from DNS is obvious. Another advantage of using DNS is that we can have continuous recording of concentrations at a single location or that we can do multiple repetitions of the experiment and that way obtain an ensemble of plumes needed for the statistical analysis of the problem and potentially observations (Fig. 6.11.4).







**Fig. 6.11.1:** Gaussian plume model for a stationary point source inconstant wind and C stability class, based on Pasquill stability classes



**Fig. 6.11.3:** DNS simulation of a plume from a line source in stationary homogeneous turbulence, following the simulation by Moser et al. (1999)

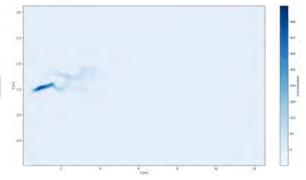


Fig. 6.11.2: DNS simulation of a plume from a point source in stationary homogeneous turbulence

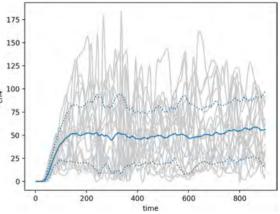


Fig. 6.11.4: Ensemble average of CH 4 concentrations at one point

#### 6.11.1.2.2 Second year

At the beginning of the second reporting period the ESR spent a month visiting EMPA in the Switzerland for her secondment. During that time plans for setting up an Observation system simulation experiment (OSSE) which will mimic the release test experiment and be used in estimation of uncertainties which are affecting the measurements. Due to the time constraints and the fact that ESR was still getting familiarized with the MicroHH model the OSSE was not performed but it will be done in the future, possibly when ESR12 will be doing his secondment at Wageningen.

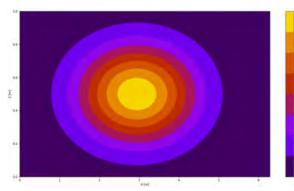
In preparation for the ESRs first experiments in MicroHH model, considerable part of the second reporting period was spent in learning C++ programming language in which the model is written. Reason for this is that the model was only able to simulate ground sources of methane so it needed to be adapted to be able to simulate multiple point or line sources at arbitrary positions in the domain. The adaption was needed to be able to reproduce situations other ESRs are encountering on the field to perform simulations which are as close to real conditions as possible. The main idea was to add the point and line sources 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. First step was to normalize Gaussian function in a way that keeps the source strength with the value that is prescribed by the user. Theoretical form of the normalization constant is given in the equation (6.11.1) as



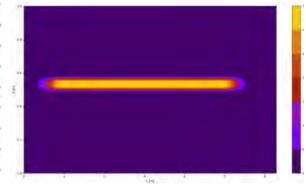
$$a = \frac{Q}{\pi^{\frac{3}{2}} \sigma^3 \operatorname{erf}(3)^3}$$

(6.11.1)

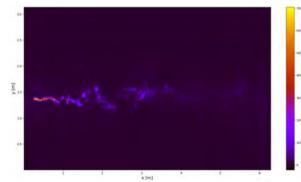
where a is the normalization constant, Q is the source strength,  $\sigma$  is the standard deviation. In the code normalization is done numerically because the grid is discretized and the value can never be the same as the theoretical one. Shapes of the point and line source that are being added into the MicroHH code are shown in Figures 6.11.5 and 6.11.6. After addition of the source class into the model the ESR spent some time on optimizing her code to run as fast as possible. That step was necessary because the size of the grid needed for high resolution simulations is such that the run time of the simulations is considerable and experiments can be very computationally expensive. Snapshots of simulations can be seen in the Figures 6.11.7 and 6.11.8.



**Fig. 6.11.5**: Illustration of how the point source is added into MicroHH. Integral over the whole Gaussian ball will give the exact prescribed source strength.



**Fig. 6.11.6**: Illustration of how the line source is added into MicroHH. Integral over the whole Gaussian pipe will give the exact prescribed source strength. Asymmetry comes from the grid resolution and chosen coordinates of the centerline of the pipe.



**Fig. 6.11.7**: Example of a high-resolution simulation of an elevated point source release in MicroHH. Mass re-entering domain is due to the periodic boundary condition.

**Fig. 6.11.8**: Example of a high-resolution simulation of an elevated line source release in MicroHH. Mass re-entering domain is due to the periodic boundary condition.

All of the preparations have been made with the goal of performing simulations of emissions of methane from a cow farm in the Netherlands. ESR2 has been measuring methane concentrations around a farm using UAV. The goal of the simulation will be to evaluate current approach to measurements by simulating the flight path of the UAV. Furthermore, assessment of different flight approaches as well as multiple drone approach will be made with the goal of devising the optimal measurement strategy given the current state of UAV technology and its limitations.



#### 6.11.1.3 Future plans and expected results

In the coming reporting period the ESR will work on her first scientific paper which will focus on the uncertainties in the measurements that arise from the wind fluctuations. From this an evaluation of the accuracy of estimations of methane source strengths using Gaussian plume model will be made.

The next step, and the second paper, will be to concentrate on dispersion with the presence of different orography. First on simple cubic building like shapes uniformly spaced over the whole domain and later with different complexities.

In the coming period ESR will also start with teaching in the course "Introduction Atmosphere" for first year bachelor students. The course is the introduction to basic physical and chemical processes in the atmosphere, weather systems across scales and mechanisms behind the climate change.

#### 6.11.1.4 Collaborations (internal / external)

In this reporting period collaboration with RUG has begun. ESR11 has adapted MicroHH code in order to do the simulations of a farm measurement campaign ESR2 has done. Katarina and Huilin visited WUR where it was agreed that Katarina will do analyses of the drone measurements using the MicroHH simulations which Anja has prepared.

During the EGU conference in April, 2018, Maarten Krol has had contact with Scott Herndon of Aerodyne who shared measurement data from their campaigns in the Gulf of Mexico. Measurement data will be compared with the simulations from MicroHH.

Collaboration with Bill Hirst and his group at Shell will also be made. The main objectives will be to explore the use of MicroHH for some of their measurement strategies.

#### 6.11.1.5 Risks and difficulties

No risks or difficulties were encountered in the reporting period.

#### 6.11.2 Deliverables

Within the project ESR12 is involved in the deliverables D1.3, D1.4, D3.1, D3.2, D3.3, and D3.4.

**D1.3** - Report and publication of the results from the campaign in Silesia (month 36)

Contribution to this deliverable is planned for the next reporting period. The work on the simulations of dispersion of CH<sub>4</sub> will help us preparing the measurement strategy in Silesia.

**D1.4** - Report and public on improved emission factors for different source categories from mobile measurements (month 42)

Contribution to this deliverable is planned for the next reporting period.

D3.1 - New tools to estimate CH\_4 source strengths from point sources, including mobile measurements (month 24)

We further improved the tool MicroHH and performed simulations in MicroHH, which can mimic release of methane from point and line sources. So MicroHH has been adapted to simulate different sources at arbitrary positions in the domain. The MicroHHs boundary conditions have adapted to have boundary condition that prevents the scalar from re-entering the domain. Besides this we estimated the accuracy of emission estimations using Gaussian plume models for UAV and mobile vehicles measurements.

D3.2 - Improved bottom-up European CH<sub>4</sub> emissions (month 30)

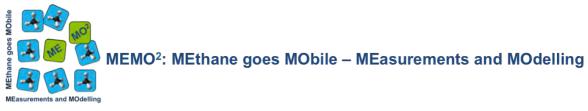
We prepared the MicroHH model to perform the required simulations.

D3.3 - Forward modelling simulations of CH<sub>4</sub> and isotopologues (month 30)

Contribution to this deliverable is planned for the next reporting period.

**D3.4** - Top-down estimates of EU-scale CH<sub>4</sub> emissions (month 42)

Contribution to this deliverable is planned for the next reporting period.



#### 6.11.3 Training and network activities

#### 6.11.3.1 General training events

Event	nt Date (start – Location Objective / expected skills end) (Host)		ECTS point s	Contribution	Comment s	
Memo <sup>2</sup> Gaussian plume workshop	9 – 10 October 2018	Heidelberg	Using Gaussian plume models in estimating source strength of methane	١	ESR helped with the DNS exercise	
SENSE A1 course	17 – 19 October 2018	Soest, the Netherlands	Introduction of the SENSE graduate school to the new PhDs	2	Poster presentation	

#### 6.11.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
1st	26/03/2018 – 27/04/2018	Dubendorf, the Switzerland	Empa	Planning an OSSE. Looking at the measurements from the first MEMO <sup>2</sup> school.	Familiarizing with the GRAL dispersion model.	Plans to perform OSSE simulations have been made in order to evaluate performance of both MicroHH and GRAL models.

#### 6.11.3.3 Conferences

Conferenc e name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation Authors (main author + co- authors)		Public available (yes / no) / web link
EGU	8 – 13 April 2018	Vienna	Poster	Modeling methane dispersion using three modeling techniques to prepare a field campaign on methane emissions	Anja Ražnjević, Chiel van Heerwaarden , Maarten Krol	Abstract available https://meetingorgani zer.copernicus.org/E GU2018/EGU2018- 13940.pdf

#### 6.11.3.4 Measurement / sampling campaigns

No participation in measurement or sampling campaigns during this reporting period except for the campaign associated to the 1<sup>st</sup> MEMO<sup>2</sup> school.

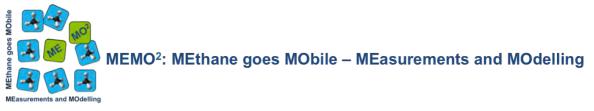
#### 6.11.4 Dissemination activities (March 2017 – February 2019)

Dissemination activity	Name	Date	Location	Type of audience	Size audience	of
MEMO <sup>2</sup> blog	Modeling dispersion of methane	07.11.2018	MEMO <sup>2</sup> website	scientific, industry, civil society, general public, media	100>	

#### References

1. Moser, R.D., Kim, J., Mansour, N.N. (1999): Direct numerical simulation of turbulent channel flow up to  $Re(\tau)$ =590. Physics of fluids, Vol.11, No.4, 943-945.

2. Van Heerwaarden, C., van Stratum, B. J. H., Heus, T., Gibbs, J.A., Fedorovich E., Mellado, J.P. (2017): MicroHH 1.0: A computational fluid dynamics code for direct numerical simulation and largeeddy simulation of atmospheric boundary layer flows. Geosci. Model Dev.



# 6.12 ESR12 - Inverse modelling of CH<sub>4</sub> and its isotopic composition at European and point source scales

#### ESR12

Inverse modelling of CH<sub>4</sub> and its isotopic composition at European and point source scales

ESR	Randulph Paulo Morales ( <u>randulph.morales@empa.ch</u> )
Supervisor	Dominik Brunner (dominik.brunner@empa.ch)
Co-supervisor	Felix Vogel ( <u>felix.vogel@canada.ca</u> )
Non-academic mentor	Arjan Hensen ( <u>hensen@ecn.nl</u> )
Official start-end date	1.12.2017 – 31.01.2021

#### 6.12.1 Scientific progress

#### 6.12.1.1 Project introduction and objectives

Dispersion models have been widely used in atmospheric science in determining the sources and sinks of emissions, such as  $CO_2$ ,  $CH_4$ , and  $N_2O$ . By coupling atmospheric observations of the target species with a suitable transport model, dispersion models may be used to validate emission inventories of greenhouse gases.

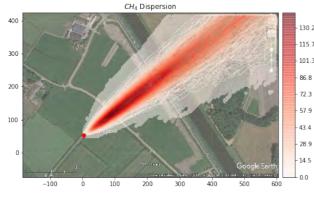
As one of the projects under MEMO<sup>2</sup>, this PhD study aims to better understand methane emissions from the level of localized sources up to the level of continental scale. With the ultimate goal of providing better estimates of local sources and validating emission inventories of methane, this project will develop high-resolution dispersion model of methane for the investigation of single sources. Moreover, a forward and inverse modelling system for methane will be implemented to qualitatively and quantitatively identify the spatial distribution of methane emissions across Europe.

#### 6.12.1.2 Project results

#### 6.12.1.2.1 First year

Gaussian plume models are the most attractive dispersion simulations because they are computationally cheap. However, one major disadvantage of a Gaussian plume model is that it needs to assume that atmospheric conditions must be uniform across the whole modelling domain and dispersion conditions must remain unchanged over a long period of time.

In order to address the problem brought about by Gaussian plume models, the multi-scale hybrid Eulerian-Lagrangian dispersion model GRAL (Graz Lagrangian Model) was adapted in this project. GRAL is a dispersion model developed at TU Graz adapted by EMPA which was specifically designed to study dispersion in complex orography and to properly deal with dispersion under low-wind conditions.



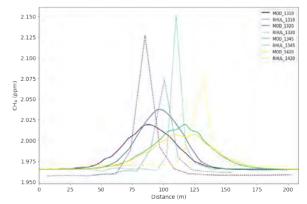


Fig. 6.12.1:  $\mbox{CH}_4$  concentration field at a height of 2 meters above surface

Fig. 6.12.2: Comparison of  $\mathsf{CH}_4$  plume peaks between measurements and the model.



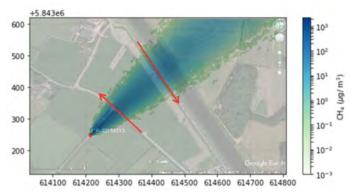
The first year of the PhD study was solely dedicated in learning how the dispersion model works. At the time of the first reporting period, the PhD had just started his study and during that time he was able to set-up a very simple dispersion scenario using the model from the measurement gathered from the MEMO<sup>2</sup> winter school held last February 2018.

#### 6.12.1.2.2 Second year

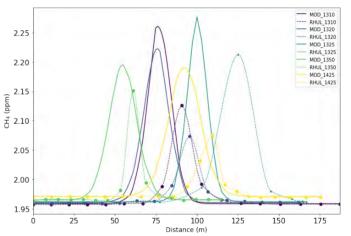
After the last reporting period, further refinements in the model were done. The tracer release experiment performed during the MEMO<sup>2</sup> winter school was reanalyzed with a much better scheme. In rerun of the same scenario. the meteorological conditions which were originally at an interval of 10 minutes were down sampled to a time resolution of 5 minutes. Down sampling the time interval into 5 minutes allowed the model to capture a better temporal variability of the CH<sub>4</sub> plumes.

In order to better compare the measurement data and the model results, methane mixing ratios were collected from two intersecting transects downwind of the plume. Fig. 6.12.3 shows the rerun of the model as well as the location of the downwind transects that were used to sample the CH<sub>4</sub> plume. The model was run with a domain size of 785 x 500 m<sup>2</sup> with an arbitrarily chosen release rate of 0.2 g CH<sub>4</sub>/s.

Furthermore, better interpolation schemes were implemented in the model to be able to capture the spatial properties of CH<sub>4</sub> dispersion from the source. A bilinear interpolation was used across the whole concentration field in order to precisely compare the mixing ratios between measurement values and modelled values in terms of their spatial properties. Since the number of points being sampled by the



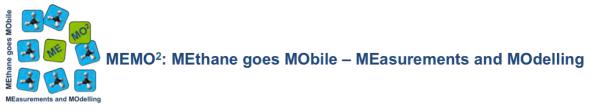
**Fig. 6.12.3:** Rerun of the model using a higher temporal resolution. Two red arrows indicating the position and direction of the mobile picarro transection the plume.



**Fig. 6.12.4:** Comparison of CH<sub>4</sub> plume peaks between measurements and the model with better interpolation schemes. Solid lines refer to the plume from the model while dotted lines refer to the plume from the measurements. Solid dots refer to the actual measurements from Picarro

mobile Picarro is limited and not continuous, a cubic spline interpolation was implemented in the measurement data in order to preserve the 'parabolic' shape of the plume. Fig. 6.12.4 shows the results of implementing a bilinear interpolation and a cubic-spline interpolation in the concentration field and the measurement data, respectively. Comparing Fig. 6.12.2 and Fig. 6.12.4, it can be seen that the spatial agreement between the model and measured data of the latter figure is better than the former. Moreover, because of the implementation of the cubic spline interpolation, the 'parabolic' shape of the measured data was preserved.

After implementing the new interpolation scheme, a quantification technique for CH<sub>4</sub> point sources was done. Modelled and measured CH<sub>4</sub> peaks obtained from Fig. 6.12.4 as well as other CH<sub>4</sub> peak transects from other mobile Picarro measurements were integrated with respect to the length of the plume. Areas under the curve obtained after the integration were then compared using a scatter plot. The resulting scatter plot from all the CH<sub>4</sub> peaks obtained during the MEMO<sup>2</sup> winter school is summarized in Fig.



6.12.5. An orthogonal regression was performed to fit a line along the measured values and modelled values. The resulting slope from the regression analysis was then used to scale the initial flux of the source which is 0.2 g CH<sub>4</sub> / s. Scaling the arbitrarily chosen initial flux, the estimate source strength of the source was computed to be 0.155 [0.139 – 0.471] g CH<sub>4</sub> / s.

A new run of the same scenario was performed but this time, a Gaussian plume model was used for computing the dispersion of the methane source. With the same meteorology conditions and the same initial arbitrary point source flux, CH<sub>4</sub> peaks obtained from the measurement campaign can also be compared to CH<sub>4</sub> peaks produced by the Gaussian plume model. Performing the same analysis as before, the initial point source flux can also be scaled using the slope of the regression analysis, as shown in Fig. 5. After scaling the initial flux, computed source strength of 0.429 [0.39300.471] g CH<sub>4</sub> / s was obtained using the Gaussian plume model. A significant difference between the computed source

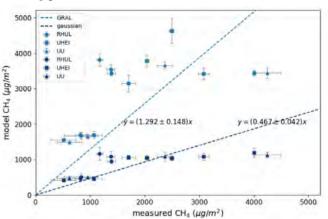


Fig. 5: Scatter plot comparing the areas under the curve from the  $CH_4$  peaks obtained from the measurement campaign and the  $CH_4$  peaks from the model.

strengths can be seen between the two models noting that the quantified source strength using GRAL is around 3 orders of magnitude lower than what the Gaussian plume gives. Factors which affected the difference in the result are still being investigated, and will be reported on the next period.

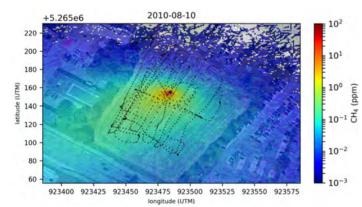


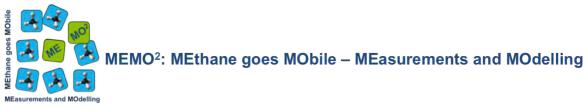
Fig. 6.12.6: Simulated methane mixing ratio for a  $CH_4$  release test done at EMPA to test the performance of the UAV. Footprint map was taken at an altitude of 5 meters above the surface. Black dots represent the UAV's path during the release test.

An inter- comparison between 1) the model results, 2) measured value from UAV, and 3) continuous measurements taken from the sampling tower in Beromünster will be performed in order to assess the performance of the model and the UAV.

In order to benchmark the performance of the GRAL model, a CH<sub>4</sub> mobile measurement dataset from the publication of Feitz et. al., 2018 regarding mobile measurement campaign from a tracer release of methane done in Ginnindera, Australia was requested from the authors. Fig. 6.12.7 shows the timeseries of the mixing ratio of methane for the aforementioned release test. A GRAL simulation was created for the said release test. Dispersion parameters such as meteorology conditions and source strength were also requested from the authors of the paper. Fig. 6.12.8 shows the obtained footprint map of methane mixing ratio. The analysis done for the dataset from the MEMO<sup>2</sup> winter school will be

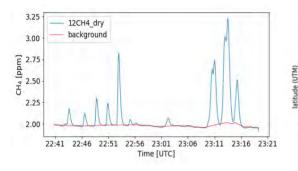
Preliminary run of the GRAL model was also used for mobile measurement devices, specifically for UAVs. A tracer release experiment at EMPA was performed by the ESR together with ESR 6 who is responsible in developing the UAV system for measuring ambient CH<sub>4</sub> mixing ratio. Fig. 6.12.6 shows the footprint map of CH<sub>4</sub> from the release experiment performed. Analysis of both the measurements and model results are still being done and will be reported at the next reporting period.

A planned campaign on a controlled farm in Beromünster, Switzerland is planned within the next few months. The aim of the controlled campaign is to test the sensitivity of both the model and the UAV.



D5.9 MEMO<sup>2</sup> – Midterm Review Report

adapted to analyse the Ginnindera dataset. Moreover, the results from this analysis will also be compared to other quantification techniques that were stated in the publication. Analysis is currently being done and is scheduled to be finished within the next few weeks.



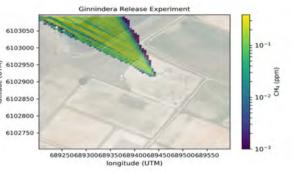
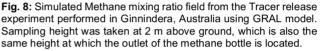


Fig. 7: Measured CH<sub>4</sub> mixing ratio from the tracer release test done in Ginnindera, Australia on May 18,2018



#### 6.12.1.3 Future plans and expected results

The beginning of the next reporting period is intended to finalize the use of the GRAL model. A culmination of GRAL and its application shall be done in a form of publication and a detailed documentation of the model. Moving forward, the ESR plans to focus more on the forward and inverse simulation of CH<sub>4</sub> and its isotopologues during the next reporting period. The second secondment is planned at TNO is scheduled in the middle of 2019 to work on the creation of emission inventories across Europe.

#### 6.12.1.4 Collaborations (internal / external)

#### Dispersion of $\ensuremath{\mathsf{CH}}_4$ at Point Source Scale

Data from the publication of Feitz et. al., 2018 was requested from the authors for reanalysis using the GRAL model. The aim of the reanalysis is to benchmark the performance of GRAL with respect to other models that was used in the publication.

Collaboration between the ESR and ESR 5 has been initiated during his secondment at LSCE, France. During this period, the two ESR will try to set-up a dispersion of CH<sub>4</sub> from a wastewater treatment plant in the city of Paris. The results will then be compared to a Gaussian plume model which was originally used in the study of this wastewater treatment plant.

#### Simulation of CH4 and its Isotopologues at European Scale

Inverse simulations of CH<sub>4</sub> together as well as its isotopologues started during the secondment of the ESR at LSCE, France. Inverse simulations using FLEXPART-COSMO were simulated for the period of November 2016 to March 2017. Emission inventory used in the simulation is the TNO-MACC-III. The results of the simulation will be used to validate the measurements made by ESR 8 in the tower of Lutjewad located in the Netherlands.

#### 6.12.1.5 Risks and difficulties

No administrative problems or difficulties have occurred within the reporting period. In terms of project results, availability of reliable dataset with proper meteorology parameters for testing the performance of GRAL model is limited.

#### 6.12.2 Deliverables

**D1.3** - Report/publication on CH<sub>4</sub> emissions from wetland and lakes in Sweden (month 30) No significant progress has been made for this deliverable



 $\mbox{D3.1}$  - New tools to estimate  $\mbox{CH}_4$  source strengths from point sources, including mobile measurements (month 24)

Running the model under different test cases is currently being done to benchmark the performance of the model. A Python tool that will be used to analyze the output of the dispersion model is almost finished and is also being tested in other test cases.

D3.2 - Improved bottom-up European CH<sub>4</sub> emissions (month 30)

No significant progress has been made for this deliverable. Initiation of this project is scheduled at the next reporting period.

D3.3 - Forward modelling simulations of CH<sub>4</sub> and isotopologues (month 36)

Initial forward and inverse simulations were started during the start of the secondment at LSCE (November 2018). Simulation period covers November 2016 - March 2017. The simulation done will be compared to the continuous isotope measurements of ESR 8 at Lutjeward Station located in the Netherlands.

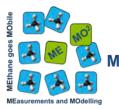
D3.4 - Top-down estimates of EU-scale CH<sub>4</sub> emissions (month 42)

No significant progress has been made for this deliverable.

#### 6.12.3 Training and network activities

#### 6.12.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
MEMO2 Winter School	5-16 February 2018	Schoorl, Netherlands	Lectures on atmospheric modelling specifically on modelling methane species using large eddy simulations. Lectures on the fundamental of atmospheric physics and chemistry, atmospheric isotopes, and methane measurement techniques Practical lectures on methane measurements	6	Participation in classes, Poster presentatiions, and modelling exercises	
Tropospheric Chemistry	Spring Semester 2018	ETH Zurich, Switzerland	Aims to provide an overview on tropospheric chemistry, which includes laboratory studies and numerical modelling. The focus of the course covers the sources and sinks of aerosols and oxidants at different scales.	3	Participation in classes as well as written exercise and oral exam	
Basic German A1.1	Spring Semester 2018	University of Zurich, Switzerland	Aims to teach students how to communicate in German at a basic level.	2	Participation in classes with written and oral exercises	
Basic German A1.2	Summer School 2018	University of Zurich, Switzerland	Continuation of the course basic German A1.1 which aims to teach students how to communicate in German at a conversational level.	1	Participation in classes with written and oral exercises	
Reproducibility in Computational Sciences	9-13 September (Summer School)	École polytechnique fédérale de Lausanne, Switzerland	Introduced the best practices and tools for reproducing research in computational sciences. Strategies on data management as well as maintenance of code and software were discussed in the session.	1	Participation in classes, computational exercises, and oral presentation	
MEMO2 Isotope Workshop	17-19 September 2018	Royal Holloway, University of London, UK	Aims to introduce the fundamental knowledge of methane isotopologues and its significance in the apportionment of methane ources and sinks in the atmosphere.		Participation in classes and data analysis	
MEMO2 Modelling Workshop	9-10 October 2018	Institut fuer Umweltphysik, Universitaet Heidelberg, Germany	Aims to introduce the different type of models whic are typically used in the field of atmospheric sciences. Different models ranging from a high resolution small scale simulations up to regional scale models were discussed during the workshop.		Participation, computational exercises, and presentation	
Boundary Layer Meteorology	Fall Semester 2018	ETH Zurich, Switzerland	The aim of the course is to acquire basic knowledge on atmospheric turbulence. It offers theoretical as well as practical approaches to treat atmospheric boundary layer flows.	4	Participation in classes. Written exercises, and oral examination	



D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.12.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
	29.10.2018- 23.11.2018	Paris, France	LSCE- UVSQ	The work of the ESR during the duration of his secondment is divided into 2 main tasks. The firs one is to be able to set-up GRAL model for a wastewater treatment plant in Paris. The secondment is to be able to be able to set-up FLEXPART=COSMO simulations	The objective of the first task is to be able to compare measured CH4 peaks with a local source scale model such as GRAL. The second task is to be able to do an inter- comparison between PYVAR-CHIMERE model and FLEXPART-COSMO	Future plans include the application of GRAL model in all the measurement campaigns made by ESR3. A joint publication with ESR 13 about the results of comparing the two models has also been planned.

#### 6.12.3.3 Conferences

No conference participation within this reporting periods.

#### 6.12.3.4 Measurement / sampling campaigns

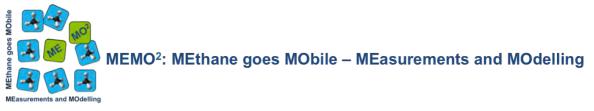
No participation in measurement or sampling campaigns during this reporting period except for the campaign associated to the 1<sup>st</sup> MEMO2 school.

#### 6.12.4 Dissemination activities

No dissemination activities within this reporting periods.

#### References

Feitz, A., Schroder, I., Phillips, F., Coates, T., Neghandhi, K., Day, S., ... Griffith, D. (2018). The Ginninderra CH4and CO2release experiment: An evaluation of gas detection and quantification techniques. International Journal of Greenhouse Gas Control, 70(November 2017), 202–224. https://doi.org/10.1016/j.ijggc.2017.11.018



#### 6.13 ESR13 - Atmospheric monitoring of the CH<sub>4</sub> emissions at the **European scale**

#### ESR13

Atmospheric monitoring of the CH<sub>4</sub> emissions at the European scale

ESR	Barbara Szénási, barbara.szenasi@lsce.ipsl.fr
Supervisor	Philippe Bousquet, philippe.bousquet@lsce.ipsl.fr
Co-supervisor	Maarten Krol, maarten.krol@wur.nl
Non-academic mentor	Renato Winkler, rwinkler@picarro.com
Official start-end date	01.10.2017 – 30.09.2020

#### 6.13.1 Scientific progress

#### 6.13.1.1 Project introduction and objectives

In the MEMO<sup>2</sup> project, local scale measurement campaigns should help improve a reference European CH<sub>4</sub> inventory and characterize the <sup>13</sup>C signature of the main source of CH<sub>4</sub> in Europe. In this PhD project, <sup>13</sup>CH<sub>4</sub> inventories will be derived from different versions of the CH<sub>4</sub> emission inventory, based on the current knowledge of the <sup>13</sup>C signature of sources in Europe and potentially including new knowledge from the measurement campaigns. The objective of this PhD project is to evaluate the accuracy of these different versions of <sup>13</sup>CH<sub>4</sub>/CH<sub>4</sub> inventories and potentially improve them using data assimilation. In addition to the information gained from bottom-up inventories, we use the information from atmospheric measurements. Through so-called inversions, this makes it possible to obtain topdown emissions that represent the best knowledge including the information from both bottom-up emissions and atmospheric measurements. The national and sectorial budgets of CH<sub>4</sub> emissions for Europe should be analyzed based on the inversions and the assessment of the inventories.

#### 6.13.1.2 Project results

#### 6.13.1.2.1 First year

been dedicated to starting to handle the PYVAR-CHIMERE modelling and data assimilation framework, and in particular the CHIMERE chemistrytransport model and its configurations that will be used during the whole project for simulating <sup>13</sup>CH<sub>4</sub> and CH<sub>4</sub> \*see table in Annex I on http://www.ceip.at/ms/ceip home1/ceip home/reporting instructions/annexes to guidelines/ mole fractions in Europe.

First simulations of CH<sub>4</sub> mixing ratios have been performed at the European Table 6.13.2: Model setup scale with the CHIMERE configuration described in Table 6.13.2, using the EDGARv4.3.2 and TNO-MACC III emission inventories described in Table 6.13.1. The simulations were carried out for the year 2011. As an input for the model, annual anthropogenic total CH<sub>4</sub> emissions were used. Fig. 6.13.1 shows the resulting annual mixing ratios using the two inventories.

The beginning of the PhD thesis has Table 6.13.1: Description of the emission inventories used in this project

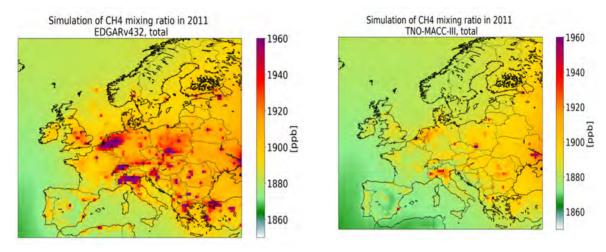
Inventory	EDGAR version 4.3.2	TNO-MACC_III
Coverage	Global	Europe
Spatial resolution	0.1° x 0.1°	0.125° x 0.0625°
Time resolution	Monthly and yearly	Yearly
Available years	1970-2012	2000-2011
Available	Sector- and country-	Sector- and country-
specifications	specific (NFR* code)	specific (SNAP** code)

\*\* see https://www.eea.europa.eu/publications/EMEPCORINAIR4/page009-a.html

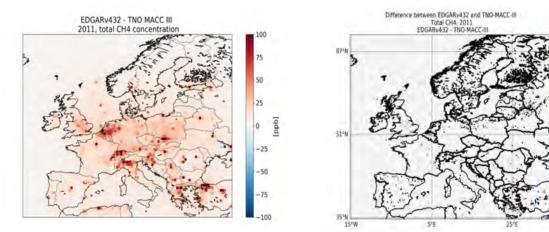
Horizontal resolution	50 km
Vertical resolution: Nr. of levels	29
- Top pressure	300 hPa
- Appr. thickness of layers	from about 10 m at the surface level
	to about 1700 m at the top of the domain
Species	Total CH <sub>4</sub>
Period simulated	1 year (2011)
Meteorology	ECMWF
Boundary and initial	LMDz
conditions	
Type of emissions	Anthropogenic (see Table 6.13.1)



As the simulated annual mixing ratios differ when using the two inventories, the differences between the simulated mixing ratios were calculated as a first overview (Fig. 6.13.2): the mixing ratios simulated with EDGARv4.3.2 are up to 100 ppb higher than those with TNO-MACC\_III, with larger differences at the hotspots, e.g. over the Po Valley or the area over Belgium, the Netherlands and North-western Germany. The differences between the emissions of the two inventories (Fig. 6.13.3) explain well the differences in the simulated mixing ratios (Fig. 6.13.2).



**Fig. 6.13.1:** Simulated annual mean values of CH<sub>4</sub> mixing ratios at the surface level over Europe with the CHIMERE chemistry-transport model using the EDGARv4.3.2 (left) and the TNO-MACC\_III (right) emission inventories.



**Fig.6.13.2:** Differences between 2011 averaged simulated CH<sub>4</sub> mixing ratios using the EDGARv432 and the TNO-MACC\_III emission inventories.

Fig.6.13.3: Differences between the total anthropogenic CH<sub>4</sub> emissions of the EDGARv4.3.2 and the TNO-MACC\_III inventories made for 2011 over Europe.

As the error statistics of the difference between modelled and measured concentrations are necessary for performing inverse modelling, the simulated hourly mixing ratios have been compared to ground based hourly measurements. A first comparison uses the data of 14 measurement sites, available through the World Data Center for Greenhouse Gases (WDCGG), and a measurement site managed by LSCE. The main characteristics of these sites are listed in Table 6.13.3 and their locations are shown in Fig. 6.13.4.

2.500 1.944 1.389 0.833

0.278

-0.278 5

0.833

-1.369



#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

Table 6.13.3: Description of the measurement sites used for analysis, * mountain stations									
Trigram	Name of site	Contributor	ontributor Country Co (latitud		Altitude above sea level [m]				
BAL	Baltic Sea	NOAA/ESRL	Poland	55.35, 17.22	28				
BSC	Black Sea	NOAA/ESRL	Romania	44.17, 28.67	3				
CMN	Monte Cimone	ISAC	Italy	44.18, 10.7	2165*				
HPB	Hohenpeissen-berg	NOAA/ESRL	Germany	47.80, 11.02	985*				
HHS	Hegyhatsal	NOAA/ESRL	Hungary	46.95, 16.65	248				
JFJ	Jungfraujoch	EMPA	Switzerland	46.54, 7.987	3580*				
LMP	Lampedusa	NOAA/ESRL	Italy	35.52, 12.63	45				
OXK	Ochsenkopf	NOAA/ESRL	Germany	50.03, 11.8	1185*				
MHD	Mace Head	NOAA/ESRL	Ireland	53.33, -9.9	8				
NGL	Neuglobsow	UBA	Germany	53.17, 13.03	65				
PAL	Pallas-Sammaltunturi	NOAA/ESRL	Finland	67.97, 24.12	560				
PRS	Plateau Rosa	RSE	Italy	45.93, 7.70	3480*				
SSL	Schauinsland	UBA	Germany	47.92, 7.92	1205*				
ZSF	Zugspitze / Schneeferner-haus	UBA	Germany	47.42, 10.98	2656*				
GIF	Gif-sur-Yvette	LSCE	France	48.71, 2.1475	160				
Table 6.1	3.3: Description of the mea	asurement sites us	ed for analysis, * r	nountain stations					
Trigram	Name of site	Contributor	Country	Coordinates	Altitude above sea				
Ū				(latitude, longitude)	level [m]				
BAL	Baltic Sea	NOAA/ESRL	Poland	55.35, 17.22	28				
BSC	Black Sea	NOAA/ESRL	Romania	44.17, 28.67	3				
CMN	Monte Cimone	ISAC	Italy	44.18, 10.7	2165*				
HPB	Hohenpeissen-berg	NOAA/ESRL	Germany	47.80, 11.02	985*				
HHS	Hegyhatsal	NOAA/ESRL	Hungary	46.95, 16.65	248				
JFJ	Jungfraujoch	EMPA	Switzerland	46.54, 7.987	3580*				
LMP	Lampedusa	NOAA/ESRL	Italy	35.52, 12.63	45				
OXK	Ochsenkopf	NOAA/ESRL	Germany	50.03, 11.8	1185*				
MHD	Mace Head	NOAA/ESRL	Ireland	53.33, -9.9	8				
NGL	Neuglobsow	UBA	Germany	53.17, 13.03	65				
PAL	Pallas-Sammaltunturi	NOAA/ESRL	Finland	67.97, 24.12	560				
PRS	Plateau Rosa	RSE	Italy	45.93, 7.70	3480*				
SSL	Schauinsland	UBA	Germany	47.92, 7.92	1205*				
ZSF	Zugspitze / Schneeferner-haus	UBA	Germany	47.42, 10.98	2656*				

In this first step, the data of the mountain stations are not used, leaving only 8 sites. Fig. 6.13.5 illustrates the comparison between the measured and the simulated mixing ratios. The values simulated with EDGARv4.3.2 are on average higher by about 4 ppb than the measurements, those simulated with TNO-MACC\_III are on average lower by about 14 ppb than the measurements (Fig. 6.13.6).

None of the two simulations reproduces the large range of variation of the measurements of 1851-1960 ppb but the simulation using the EDGARv4.3.2 inventory, with a range of 1878-1926 ppb, has a slightly larger spread than the simulation with TNO-MACC\_III (1873-1879 ppb).

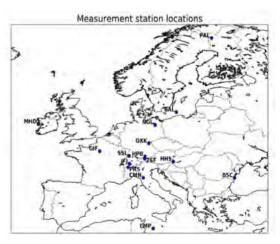


Fig. 6.13.4: Locations of the measurement sites used for analysis.



Simulations and measurements of CH4 concentration in 2011

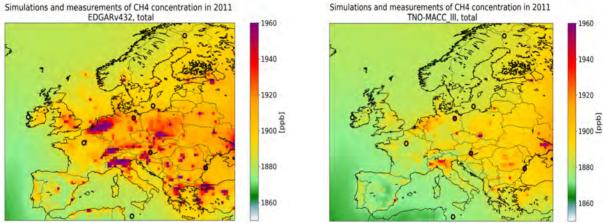


Fig. 6.13.5: Comparison of yearly averages of measured (circles) and simulated (background color) CH<sub>4</sub> mixing ratios using the EDGARv432 (left) and the TNO-MACC III (right) inventories.

#### 6.13.1.2.2 Second year

#### Multi-vear simulations and sensitivity tests using CHIMERE forward simulations

Since the end of the last reporting period, new simulations of CH<sub>4</sub> mixing ratios have been performed at the European scale 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° x 0.5° (~50x50 km). Furthermore, I have performed several sensitivity tests for 2015 (a summary of the simulations is

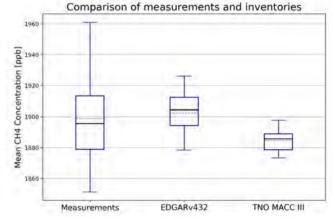


Fig. 6.13.6: Comparison between measured and simulated CH4 mixing ratios using the EDGARv432 and the TNO-MACC\_III inventory based on hourly modelled and measured values from 2011

presented in Table 6.13.5). The sensitivity tests are important means of error estimation as they can help determine several components of the errors, such as the transport model error or the representation error. The latter is the 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. Since the start of this reporting period, I have included data of additional measurement sites to the ones listed in Table 6.13.3. Information about these is given in Table 6.13.4. The data of all the measurement sites listed in both tables have been compared to simulated data when observations were available and flagged for good quality. The comparison to measurements is necessary for performing inverse modelling as we have to assess the errors of the difference between measured and modelled mixing ratios. The comparison and the sensitivity tests aim a better understanding of the difference between modelled and measured CH<sub>4</sub>, and thus help reveal which part can be attributed to errors in inventories and serve the goal of estimating top-down CH<sub>4</sub> emissions on the European scale.



Trigram	Name of site	Contributor	Country	Coordinates (latitude, longitude)	Altitude above sea level [m]
BEO	Beromuenster	UBERN	Switzerland	47.19, 8.18	1009*
BIS	Biscarrosse	LSCE	France	44.38, -1.23	120
CBW	Cabauw	InGOS	Netherlands	51.9703, 4.9264	0
CGR	Capo Granitola	WDCGG	Italy	37.6667, 12.65	5
DEC	Deltadel'Ebre	WDCGG/IC3	Spain	40.74, 0.79	15
ECO	Lecce Environmental- Climate Observatory	WDCGG	Italy	40.3358, 18.1245	36
ERS	Ersa	LSCE	France	42.9692, 9.3801	533
FKL	Finokalia	ECPL <sup>a</sup>	Greece	35.3378, 25.6694	150
GIC	Gredos	WDCGG/IC3	Spain	40.35, -5.18	1456*
GLH	Giordan Lighthouse	WDCGG	Malta	36.07, 14.22	160
PR	Ispra	InGOS/JRC	Italy	45.8147, 8.636	210
AE	LaegernHochwacht	UBERN	Switzerland	47.82, 8.4	872
_MT	Lamezia Terme	WDCGG	Italy	38.8763, 16.2322	6
LUT	Lutjewad	InGOS	Netherlands	53.4036, 6.3528	1
OHP	Observatoire de Haute Provence	OSU <sup>b</sup>	France	43.931, 5.712	650
OPE	Observatoire pérenne de l'environnement	LSCE	France	48.5619, 5.5036	390
OVS	OVSQ	LSCE	France	48.7779, 2.0486	150
PDM	Pic du Midi	LSCE	France	42.9372, 0.1411	2877*
PUI	Puijo	FMI <sup>c</sup>	Finland	62.9096, 27.6549	232
PUY	Puy de Dôme	InGOS/LSCE	France	45.7719, 2.9658	1465*
RGL	Ridge Hill	University of Bristol	UK	51.9974, -2.5398	199
SAC	Saclay	CEAd	France	48.7227, 2.142	160
SMR	Hyytiälä	University of Helsinki <sup>e</sup>	Finland	61.8474, 24.2947	181
SNB	Sonnblick	WDCGG/UBA	Austria	47.05, 12.95	3106*
ГАС	Tacolneston	University of Bristol	UK	52.5177, 1.1388	56
TRN	Trainou	InGOS/LSCE	France	47.9647, 2.1125	131
ΓΤΑ	Angus	University of Bristol	UK	56.555, -2.9864	313
/AC	Valderejo	WDCGG/IC3	Spain	42.88, -3.21	1122*
NAO	Weybourne	NILU <sup>f</sup>	Norway	52.95, 1.121	31

#### D5.9 MEMO<sup>2</sup> – Midterm Review Report

<sup>a</sup> ECPL: Environmental Chemical Process Laboratory

<sup>b</sup> OSU: Observatoire des Sciences de l'Univers Institut Pythéas

<sup>c</sup> FMI: Finnish Meteorological Institute

alternatives

<sup>e</sup> Division of Atmospheric Sciences, Department of Physics

<sup>f</sup> NILU: Norwegian Institute for Air Research

Table 1: Summary of the simulation runs carried out with CHIMERE over the first two reporting periods Horizontal 0.5° x 0.5° 0.25° x 0.25° 0.1° x 0.1° - 1- - 41

resolution			
Simulated year	2011-2015	2015	2015
Boundary condition	LMDz & MACC	MACC	MACC
Emission inventory	EDGARv4.3.2 & TNO-MACC-III (yearly values from 2011)	EDGARv4.3.2 & TNO-MACC-III (yearly values from 2011)	EDGARv4.3.2 & TNO-MACC-III (yearly values from 2011)
Emission sector	Total & sectors	Total	Total
Type of emission	Anthropogenic & Anthrpogenic+Wetland	Anthropogenic	Anthropogenic

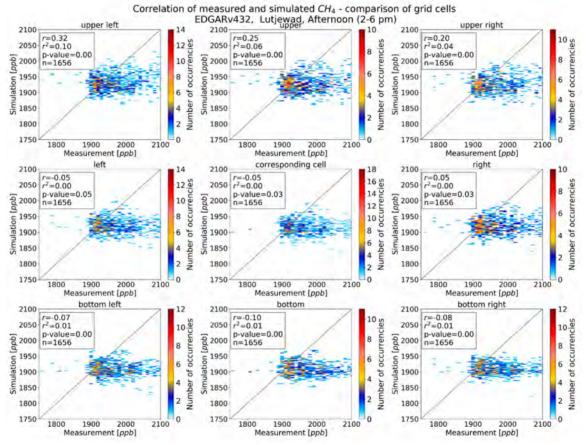
#### Representation error at the measurement sites

The above-mentioned representation error occurs because a model grid cell is large compared to a point measurement. Indeed, when we compare values of a model grid cell to measured values, we compare point measurements with simulated data calculated over a grid cell of about e.g. 2500 km<sup>2</sup>. For example, if a station lies on the coast and the corresponding grid cell covers a larger portion of the sea than of the land, then the model value may not be a good representation of the site's measurement.



The same misrepresentation can occur with topographical features, such as mountains, so that the value of the corresponding model grid cell for a mountain site rather represents the conditions of e.g. the valley. Hence, it can happen that the simulated values in a neighbouring model cell match the measured ones better.

We therefore checked the correlation between measurements and simulated values of the grid cell corresponding to the station location and its eight neighbouring cells. An example for the site Lutjewad in the Netherlands can be found in Fig. 6.13.7. On the basis of this analysis, we decided to use the values of model grid cells with the highest correlation coefficients for the comparison against the measurements.



**Fig. 6.13.7:** 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.

#### Assessment of the impact of the boundary conditions

One of the sensitivity tests consisted of running the model with boundary conditions obtained from the CAMS-MACC Reanalysis product (Inness et al. 2013) which is available for every three hours per day from July 2013 on. In contrast to this, the pre-optimized boundary conditions derived from the LMDz model are only available up until 2010 and so they follow the same pattern every year in 2011-2015, resulting in a uniform seasonal behavior. The differences in the latitudinal gradients of the two products (Fig. 6.13.8) come from different assumptions used in the models, such as a higher latitudinal gradient in the MACC product. Simulations using the MACC boundary conditions compare better to the measurements (illustrated in Fig. 6.13.9).

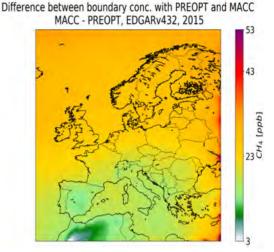


We decided to use the MACC boundary conditions for every other model run hereafter, including the other sensitivity runs. However, the LMDz boundary conditions have been planned to be updated to more recent years soon so that we will compare the up-to-date boundary conditions to the MACC ones.

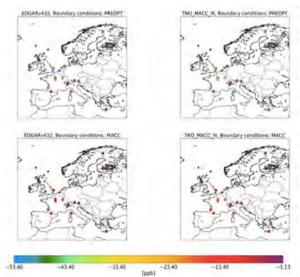
#### Impact of model resolutions

In order to evaluate the transport model skills and to identify and separate sources of errors due to model resolution and inventory uncertainties at different scales, model simulations with higher spatial resolutions have been carried out;  $0.25^{\circ} \times$  $0.25^{\circ}$  (~25x25 km) on the same domain and  $0.1^{\circ} \times$  $0.1^{\circ}$  (~10x10 km) on a smaller domain covering the BENELUX, Germany and Poland, i.e. the areas with the highest emissions in Europe. A smaller domain was necessary due to computational costs.

The simulation results have been compared to each other and to the measurements applying several statistical methods, one of which is displayed in Fig. 6.13.10. In this method, we calculated the seasonal cycle of the model bias compared to measurements using afternoon data from 12-18 UTC for all three horizontal resolutions. As already shown in the first reporting period, the modelled CH<sub>4</sub> values using the EDGARv4.3.2 inventory are in general higher than the ones driven by the TNO-MACC\_III inventory. The bias of the model using EDGARv4.3.2 compared to the measurements is thus smaller than in the case of the simulated CH<sub>4</sub> using TNO-MACC III. Moreover, the higher the horizontal resolution is, the smaller the bias gets. The bias is negative all year round but during the spring months, which means that the model mainly underestimates the measurements whatever the resolution and emission inventory. Also, the springtime overestimation mostly occurs with the horizontal resolution of 0.1° x 0.1°. The modelled values may get higher with a finer horizontal resolution as the emissions from point sources are not diluted in large grid cells but may lead to high concentrations in the smaller grid cells.



**Fig. 6.13.8:** Differences between the CH<sub>4</sub> mixing ratios resulting from using two sets of boundary concentrations over Europe. The mixing ratios resulted from the pre-optimized LMDz boundary concentrations (PREOPT) were subtracted from the ones from the MACC boundary concentrations. For this simulation run, the emissions of the EDGAR v4.3.2 inventory were used. (The result is the same in case of the TNO-MACC\_III emissions)



**Fig. 6.13.9:** The bias of simulation data using the two emission inventories (left: EDGAR v4.3.2, right: TNO\_MACC\_III) and boundary concentrations from PREOPT (upper row) and MACC (bottom row) compared to measurements



Bias for 2015 - hourly afternoon data (12-18 UTC)

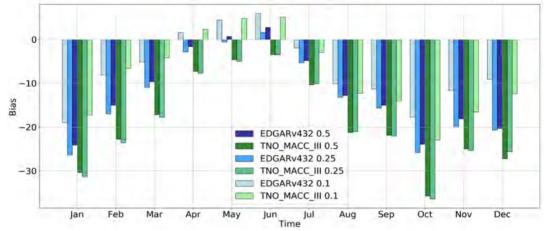


Fig. 6.13.10: Seasonal cycle of model bias compared to measurements for the horizontal resolutions of 0.5° (darkest inner bars), 0.25° (middle bars) and 0.1° (lightest outer bars); *blue bars*: EDGAR v4.3.2, *green bars*: TNO-MACC\_III

#### Analysis of the impact of natural CH<sub>4</sub> emissions

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 following Poulter et al. (2017). 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 (see Fig. 6.13.11), which seems advantageous as the measurements are mostly underestimated by the model.

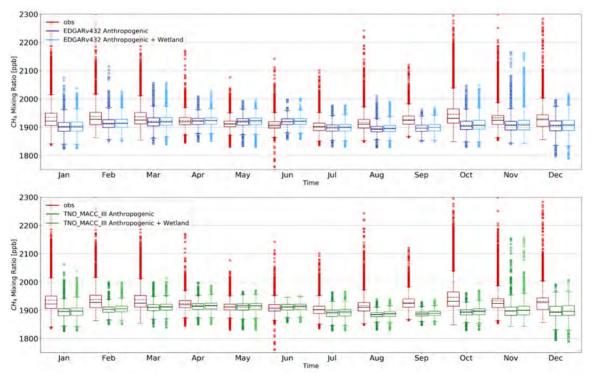
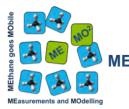


Fig. 6.13.11: Differences between concentrations using the two inventories *with* and *without* adding wetland emissions over Europe; *top*: EDGAR v4.3.2, *bottom*: TNO-MACC\_III



D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### Next steps

In the remaining time of the current reporting period, it is planned to analyse the simulation runs using sectorial emissions in order to define the contribution of the different emission sectors. A sensitivity test should be performed with monthly time profiles for the EDGARv4.3.2 inventory.

The data simulated by the LOTOS-EUROS chemistry-transport model provided by TNO will be compared to those simulated by the CHIMERE chemistry-transport model. The information obtained by this comparison is useful for estimating the transport model error as the differences in the simulation results can reveal the sources of errors that occur due to a different model parametrization.

The simulations with the horizontal resolutions of  $0.5^{\circ} \times 0.5^{\circ}$  and  $0.25^{\circ} \times 0.25^{\circ}$  will be repeated using the emissions from the domain of the  $0.1^{\circ} \times 0.1^{\circ}$  simulations, i.e. the emissions will be set to zero outside of the  $0.1^{\circ} \times 0.1^{\circ}$  simulation domain. Doing so helps further determining the representation error, as we will be sure that we analyse concentrations resulting from the same emissions.

Simulations were run using MicroHH, a computational fluid dynamics code for the simulation of turbulent flows in the atmosphere, during the first secondment at WUR for the location of the Cabauw measurement site on the chosen date of June 23, 2015. In this reporting period, we are going to resolve whether using the simulations by MicroHH is a real asset for the error estimation without taking excessive time for the analysis. However, simulations run by MicroHH are costly and comparisons of one day only might not provide statistically significant results.

Moreover, we will start including isotopes in the forward simulations with CHIMERE when ESR12 is absolving his secondment at LSCE in November 2018 and comparing the simulation results with continuous isotopic measurements of the measurement site Lutjewad.

#### 6.13.1.3 Future plans and expected results

The beginning of the next reporting period will be dedicated to the calculation of the prior uncertainty covariance and error covariance matrices to be able to perform inverse modelling based on the statistical error analysis from the current reporting period. Subsequently, it is planned to perform inversions using CHIMERE and the PYVAR variational data assimilation tool and obtain top-down estimates of CH<sub>4</sub> emissions on the European scale.

The second secondment is planned at TNO to work on the emissions.

Furthermore, scientific publication(s) of results obtained so far are planned.

#### 6.13.1.4 Collaborations (internal / external)

Collaboration with TNO: I am going to use the data simulated by the LOTOS-EUROS chemistrytransport model to compare them to the ones simulated by the CHIMERE chemistry-transport model. This is beneficial for assessing the transport model error.

#### 6.13.1.5 Risks and difficulties

There are only very few continuous isotopic measurements available so far that could be used in this project for the evaluation of modelled isotopic values. Other than this, there have not been any problems encountered.

#### 6.13.2 Deliverables

**D2.5** - Report providing isotopic maps at grid scale from inventories and atmospheric measurements (month 42)

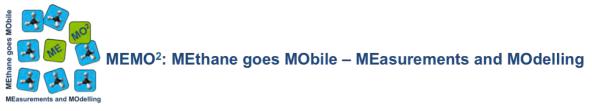
The isotopes have not been included in the modelling work yet but it is planned to start the work in November 2018 when ESR12 is absolving his secondment at LSCE.

D3.1 - New tools to estimate CH\_4 source strengths from point sources, including mobile measurements (month 24)

In this project, no work has been done for this deliverable.

**D3.2** - Improved bottom-up European CH<sub>4</sub> emissions (month 30)

This work has not been started yet. It is planned to start it in the next reporting period.



D3.3 - Forward modelling simulations of CH<sub>4</sub> and isotopologues (month 30)

Forward modelling simulations of  $CH_4$  mixing ratios have been carried out with CHIMERE using the EDGARv4.3.2 and the TNO-MACC\_III emission inventories. The simulation of isotopologues should be carried out by the end of this reporting period.

D3.4 - Top-down estimates of EU-scale CH<sub>4</sub> emissions, (month 42)

This work has not been started yet.

#### 6.13.3 Training and network activities (March 2017 – February 2019)

#### 6.13.3.1 General training events

Event	Date (start – end)	Location (Host)	Objective / expected skills	ECTS points	Contribution	Comments
Greenhouse gases (GHG) challenges and observations	October– December 2017	Ecole Polytechnique, Palaiseau, France	The course makes one aware of the main climate effects of the increase of the atmospheric burden of GHGs, as well as of the various emission scenarios, including their ties to regional policy and economy, and the link between emissions and atmospheric concentration. The methodologies used to improve the knowledge on GHG sources and sinks were detailed and various observation techniques were presented.	4	Participation in classes and written exam	
Biogeochemical cycles and interactions with the biosphere at global scale	October– December 2017	AgroParisTech, Paris, France	The course aims the understanding of pollutant and greenhouse gas global budgets but also to establish relevant climate scenarios for the future. This teaching unit gives a basic knowledge on the major global biogeochemical cycles (carbon, nitrogen,) and to provide highlights on few biosphere - atmosphere interactions from regional scale to global scale.	3	Participation in classes and writing of two reports for the exam	
1 <sup>st</sup> MEMO <sup>2</sup> school	5-16 February 2018	Schoorl, Netherlands	The school's objective was to present the basics of atmospheric physics and chemistry, greenhouse gases (especially methane), isotopes and several measurement techniques. The latter did not only include theoretical training but it was demonstrated in a measurement campaign. The gathered data from the measurements were analysed.	6	Participation in classes, helping in modelling exercise, preparation of a poster, data analyses	
Isotope workshop	17-19 September 2018	Royal Holloway, University of London, UK	The workshop was dedicated to the fundamentals in detail on methane isotopes. It included theoretical lessons, an experimental training in the laboratory and analysis of the collected data. Furthermore, the training included the modelling concepts of isotopes.		Participation and data analysis	
Dispersion modelling workshop	9-10 October 2018	Institut fuer Umweltphysik, Universitaet Heidelberg, Germany	This workshop was designed to understand the essential know-how on dispersion modelling and to be able to use Gaussian plume models.		Participation	

#### 6.13.3.2 Secondments

Secondment	Date (start – end, planned (when))	Location	Host	Description of work / deviations	Scientific / training (skills) objective	Results and future plans
WUR	19 February 2018 – 19 March 2018	Wageningen, the Netherlands	WUR	The goal of the secondment was an inter- model comparison using the MicroHH model, a computational fluid dynamics code for the simulation of turbulent flows in the atmosphere, and the CHIMERE chemistry-transport model.	For inverse modelling, we need to obtain the representation error of the vertical mixing and the transport error. These errors can be assessed when comparing the simulation results to those of another model (MicroHH). To learn more about MicroHH, I spent a month at WUR.	I have learnt to run MicroHH. Simulations of CH <sub>4</sub> have been performed for one day only as the simulations are costly. Comparisons of one day only does not provide statistically significant results and thus we are thinking about whether it is advisable to use the simulation results.
TNO (planned)	March/April 2019	Utrecht, the Netherlands	TNO Utrecht	It is planned to work on the emissions during this secondment		



D5.9 MEMO<sup>2</sup> – Midterm Review Report

#### 6.13.3.3 Conferences

Conference name	Date (start – end, planned (when))	Location	Presentation (oral / poster)	Title of presentation	Authors (main author + co-authors)	Public available (yes / no) / web link
EGU	9-13 April 2018	Vienna, Austria	Poster presentation	Atmospheric monitoring of methane emission at the European scale	B. Szénási, I. Pison, G. Broquet, M. Saunois, P. Bousquet, A. Berchet	https://presentations.copernicus.org/EGU2018- 14964_presentation.pdf
EGU	9-13 April 2018	Vienna, Austria	Presentation during the MEMO <sup>2</sup> session	Atmospheric monitoring of methane emission at the European scale	B. Szénási, I. Pison, G. Broquet, M. Saunois, P. Bousquet, A. Berchet	

#### 6.13.3.4 Measurement / sampling campaigns

Campaign	Date (sta planned		Location	Host	Description of work	Scientific objective	Samples (nature / number)	Results and future plans
MEMO <sup>2</sup> school	5-16 2018	February	Schoorl, NL	ECN, Petten, NL	Mobile measurements of methane and methane isotopes in and around Schrool. The targeted areas were around farms and agricultural fields.	It was within the framework of the MEMO <sup>2</sup> school with the objective of acquiring some measurement skills and techniques.	The measurements of our team were carried out using a Picarro analyser. It measures CH4 and CO <sub>2</sub> .	These measurements might be included in later model evaluation work.

#### 6.13.4 Dissemination activities

Dissemination activity	Name	Date	Location	Type of audience	Size of audience
Blog entry on the MEMO <sup>2</sup> website		05 September 2018	https://h2020- memo2.eu/category/blog/	General public interested in science	

#### References

Benjamin Poulter, e. a., 2017. Global wetland contribution to 2000-2012 atmospheric methane growth rate dynamics. *Environ. Res. Lett.* 12 094013.

Inness, e. a., 2013. The MACC reanalysis: an 8 yr data set of atmospheric composition. *Atmos. Chem. Phys. 13, 4073-4109.* 

# 7. History of the deliverable

#### Table 3: Deliverable history

Version	Author(s)	Date	Changes
1	Sylvia Walter	26 September 2018	Template of ESR report sent to ESRs and PIs, start writing report and updating parts of the 1 <sup>st</sup> Progress Report
	Sylvia Walter	8 October 2018	Template of WP report sent to WP leader, cc beneficiaries
	Sylvia Walter	7 November 2018	Deadline ESR and WP leader contributions
	Sylvia Walter	30 November 2018	Send to the coordinator and the consortium for a first review, planned submission date 21 December 2018
	Sylvia Walter	20 December	No further comments received, final Midterm Report submitted to the PO 20 December 2018