PARIS

Process Attribution of Regional Emissions

GA 101081430, RIA

MS12 – Mobile-platform continuous methane isotope measurement campaigns

MS12			
Delivery due date Annex I	PM 6 | 30 June 2023		
Actual date of submission			
Lead beneficiary: UU	Work package: 4	Nature: Data	Dissemination level: PU
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Version: 1			

Horizon Europe Cluster 5: Climate, energy and mobility

"This project has received funding from the European Union's Horizon Europe Research and Innovation programme under HORIZON-CL5-2022-D1-02 Grant Agreement No 101081430 - PARIS".

Table of content

1. Changes with respect to the DoA (Description of the Action)

The milestone MS12 belongs to Task 4.2 of work package WP4 and has so far been executed as planned in the DoA. The means of verification are the initiation of several mobile measurement campaigns to determine the isotopic composition at a range of European locations.

2. Dissemination and uptake

This report gives an overview of the initial activities regarding the first campaigns and data compilation. It is a publicly available document. Data will be used to improve atmospheric models that are being developed by several partners, both within PARIS (e.g. EMPA, UNIVBRIS) and outside, e.g. for partners from related projects such as EYE-CLIMA and AVENGERS (e.g. NILU, TNO, NOAA). Data will also be used for intercomparisons and be made publicly available on the Integrated Carbon Observation System (ICOS) Carbon Portal.

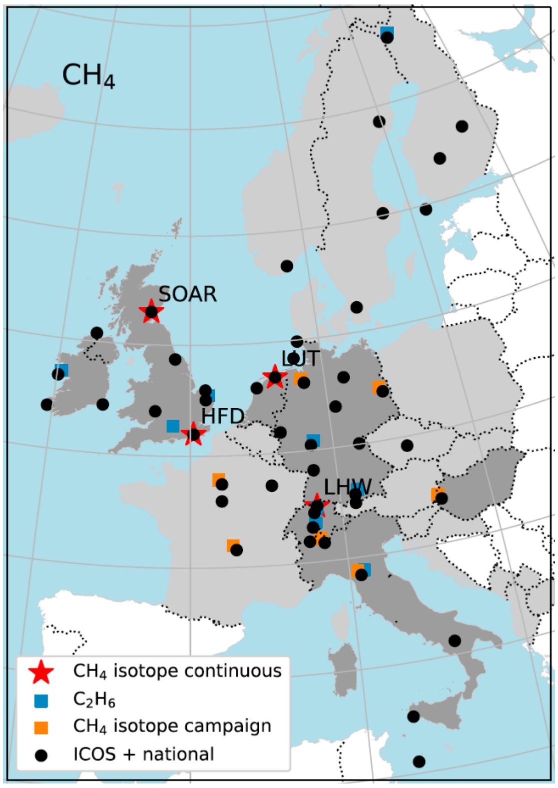

Fig. 1: CH_4 measurement sites.
Black dots: existing high-frequency measurement stations.
Red stars: proposed for high frequency CH_4 isotopologue measurements: Heathfield, UK (HFD), eastern Scotland, UK (SOAR) or Lutjewad (LUT), Lägern-Hochwacht (LHW)
Orange squares: proposed locations for deployment of the UU high-frequency GC-IRMS instruments.
Countries in light grey: national total emissions inference is likely to be possible.
Countries in dark grey: focus countries aimed to derive sector level CH_4 emissions.

3. Short Summary

Within WP4, measurement campaigns of 6-12 months duration will be executed, using mobile instruments to perform high frequency, high precision isotope observations. Campaigns are planned at five European sites between 2022 and 2024 (see Fig. 1). As part of the PARIS project, we use data from two mobile campaigns measuring CH_4 isotopes that were carried out before PARIS officially started, one at the ICOS station Lindenberg (Germany) and another one in the city of Hamburg. The data of both campaigns are currently being evaluated and prepared for publication.

Another additional measurement campaign was started, carried out and finalized in Cluj-Napoca, Romania. This campaign builds on the results and experiences of the previous project ROMEO (http://romeo-memo2.wikidot.com/home:scientificbackground, (Menoud et al., 2022b).

The methane isotope time series in Cluj-Napoca covers an 8-months' time high-frequency data series in 2023. Results will be available on the ICOS Carbon Portal after finishing the data analysis.

A fourth campaign, with continuous measurements planned for at least 6 months, started at Hegyhátsál, Hungary, a certified ICOS station. The mobile GC-IRMS system, operated by the UU, was installed and measurements started in October 2023, but paused due to technical issues with a key cryostat unit. The cryostat is currently in repair with the manufacturer, which will lead to a delay in this activity.

To identify the best location for future measurement series, EMPA Dübendorf executed two Observation Simulation System Experiment (OSSE) to select the most appropriate location (see Annex I). Based on the results, EMPA will conduct methane (CH_4) mole fraction, CH_4 isotope and ethane (C_2H_6, fossil tracer) measurements at the Swiss site, as the OSSE´s showed that the initial location (station Lägern Hochwacht), had infrequent sensitivity to fossil sources, while Dübendorf showed large increments. In November 2023, measurements of CH_4 and C_2H_6 concentrations were initiated and comparison measurements for CH_4 and C_2H_6 were conducted. CH_4 isotope measurements will commence in spring 2024 as foreseen. This station will not be part of the mobile campaign, but data will be used for comparison.

For the mobile measurements, UEDIN created forward models for several locations (see Annex I), which showed potential for the following ICOS stations: HUN, KRE, LIN, CBW and CMN. The Locations for future campaigns will be chosen later in the project, based on these OSSEs. A preparatory air sampling program has been started at Monte Cimone station in Italy, using ICOS flask sampler. Depending on location logistics, an isotope measurement system is planned to be deployed at the mountain station. Alternatively, the system will be deployed in the UK for intercomparison measurements with one of the laser systems from UEDIN/NPL (Stavert et al., 2019). The station of Cluj-Napoca was chosen because Romania lacks continuous measurements, even though studies found a variety of emissions (Stavert et al., 2019). An overview of all chosen stations is shown in chapter 4.3.2.

4. Evidence of accomplishment

4.1 Introduction | Background of the milestone

CH_4 is emitted in Europe from a mixture of sources, where the main contributor varies per country. Mobile measurements are a fast and accurate approach to observe and also identify even small amounts of methane emissions. Mobile measurement campaigns can provide extensive coverage of larger area, depending on the deployed platform and system. This improves the local and global methane budgets, and by this supports the implementation and integration of mitigation measures.

It is challenging to attribute emissions to the individual sectors, which is currently attempted by bottom-up models or inventories. An alternative source attribution technique uses atmospheric observations of methane isotopic composition ($\delta^{13}C$ and δD) since different sources emit methane with a specific stable isotopic composition. Sector specific fluxes can be estimated, and atmospheric transport models and inversions can be used to improve the atmospheric CH_4 budget.

The IRMS (Isotope-Ratio Mass Spectrometry) system of the UU offers the possibility to carry out high frequency isotope observations at field locations. Such "campaign-mode" deployments have been carried out previously to investigate the methane source mix in selected European regions.

PARIS will advance this approach with the goal to derive sector specific methane emission estimates by performing continuous measurements in important regions with different source mixes. Advanced process attribution will provide policymakers and inventory compilers with better information about the sectors on which to focus mitigation efforts.

High-frequency $\delta^{13}C$-CH_4 and δD-CH_4 observations at field sites have been pioneered by PARIS partners at UU, EMPA and UEDIN, using both IRMS (GC-IRMS) and laser spectrometry. Scientific evaluation has demonstrated that these measurements allow evaluation of regional emission inventories (Eyer et al., 2014), (Menoud et al., 2020), (Menoud et al., 2021), (Rennick et al., 2021), (Röckmann et al., 2016).

Within PARIS, the isotope monitoring capabilities are planned to be extended across focus countries during several campaigns (Fig. 1).

4.2 Scope of the milestone

The milestone MS12 has been defined as the initial step to plan and set-up the mobile measurement campaigns within PARIS. MS12 belongs to Task 4.2 which is an ongoing task in the project.

Task 4.2 is a joint activity of 6 PARIS partners and contains new, high-frequency $\delta^{13}C$ and δD methane isotope measurements, established at SOAR (UK) or LUT (NL), Heathfield (UK) and Zürich (CH). Isotope monitoring campaigns of 6-12 month duration using a mobile platform will be conducted at five sites. One campaign will take place at Hegyhátsál in focus country Hungary and others are chosen based on the OSSEs in Task 4.1. The isotopic composition for both $\delta^{13}C$-CH_4 and δD-CH_4 of the background air will be determined by regular (~weekly) analysis at UU on flask samples collected from Mace Head. Involved partners in Tasks 4.1 and 4.2 are UU, UEDIN, UNIVBRIS, ATMOKI, EMPA, NUIG and MO. This document gives an overview of the actual status of the milestone and the future planning for Task 4.2.

4.3 Content of the milestone

4.3.1 Methodology

4.3.1.1 Selection of sampling locations

As measurement campaigns are costly and time-consuming, sampling locations for the campaigns need to be selected carefully. Observation Simulation System Experiments (OSSEs) are used to investigate the potential performance and usefulness of selected locations for dedicated measurement parameters.

An OSSE was conducted by modelling methane and the isotopic ratios to determine the most useful sites for high frequency methane isotope monitoring campaigns in Europe. The effect of uncertainties in the isotopic source signatures was included in this assessment.

Other criteria for the selection of campaign location and sampling strategy are related to logistics, since the systems require a certain environment and technical support. For ICOS stations or station with continuous methane measurements, the χ(CH_4) is evaluated to determine whether there are sufficient peaks of interest and which season would be optimal.

4.3.1.2 Facilities

Some CH_4 isotope measurement campaigns are planned at tall tower stations of the ICOS network. ICOS is providing standardised and open data from more than 170 measurement stations across 16 European countries. The stations are exposed to atmospheric transport and processes covering larger areas to receive integral information on sources and sinks of greenhouse gases. An overview of the available ICOS stations is given on the ICOS website https://www.icos-cp.eu/observations/atmosphere/stations.

Besides using the ICOS tall tower facilities, isotope monitoring instruments were or will be placed at other interesting locations to accompany mobile surveys, as e.g. in Cluj-Napoca.

4.3.1.3 Mobile surveys

Mobile surveys follow in principle the set-up as e.g. described in Fernandez et al., 2022 and Maazallahi et al., 2020. A mobile detector is mounted in a car and the χ(CH_4) is monitored while the vehicle is operated. When enhancements above a certain threshold are detected, discrete samples are collected for further lab investigation.

4.3.2 Measurement campaign planning

The status of the measurements is shown in Table 1. For the ICOS stations the methane mole fraction of the IRMS will be compared with the ICOS methane measurements.

Table 1: Overview of campaigns, executed and planned with their status. Some parameters are to be determined (-). The parameters are: Methane mole fraction (χCH_4), ethane mole fraction (χC_2H_6), Carbon 13 isotopic signature on the international scale VPDB ($\delta^{13}C$), Deuterium isotopic signature on the international scale V-SMOW (δD).

Campaign location	type	Lead PARIS partner	Date (planned or executed)	Parameters / metadata measured	Instrumentation	Actual status
Lindenberg (LIN)	ICOS class 1	UU	May 2022 – November 2022	χCH_4, $\delta^{13}C$, δD	GC-IRMS	Executed, under Evaluation
Hamburg	High building	UU	July 2021 – April 2022	χCH_4, $\delta^{13}C$, δD	GC-IRMS	Executed
ROMEO – Cluj-Napoca (RO)	University	UU	Jan 2023 – September 2023	χCH_4, $\delta^{13}C$, δD	GC-IRMS	Executed, data under evaluation
Hegyhátsál (HUN)	ICOS class 2	UU	October 2023– likely: April 2024	χCH_4, $\delta^{13}C$, δD	GC-IRMS	Interrupted
Empa, Dübendof	unknown	EMPA	2024	χCH_4, $\delta^{13}C$, δD, χC_2H_6	TREX-QCLAS	In preparation
Monte Cimone(CMN)	ICOS class 2	UU	May 2024 – September 2023	χCH_4, $\delta^{13}C$, δD	GC-IRMS	Checking requirements
Heathfield	Tall Tower	UU	September 2023 – unknown	χCH_4, $\delta^{13}C$, δD, χC_2H_6	GC-IRMS & laser	First contact started
Saclay	ICOS class 1	-	-	-	-	OSSE – listed*

Puy de Dome	ICOS class 2	-	-	-	-	OSSE – listed*
ISPRA	ICOS class 2	-	-	-	-	OSSE – listed*
SOAR	-	-	-	-	-	OSSE – listed*

*** OSSE listed:** Stations are of potential interest for measurement campaigns, decision depending on capacities during the project

4.3.3 Preliminary results

4.3.3.1 Cluj-Napoca (Romania)

Continuous measurements were performed for the CH_4 mole fractions, δD and $\delta^{13}C$ from January till September 2023 (Fig. 2). Gaps in the data are due to malfunctioning of the system. Peaks were analysed via the Keeling plot method (Keeling, 1961) to determine the source signature. The enhancements in the time series originate from a mixture of sources, including leaks from natural gas pipelines, sources with microbial fermentation and a thermogenic source.

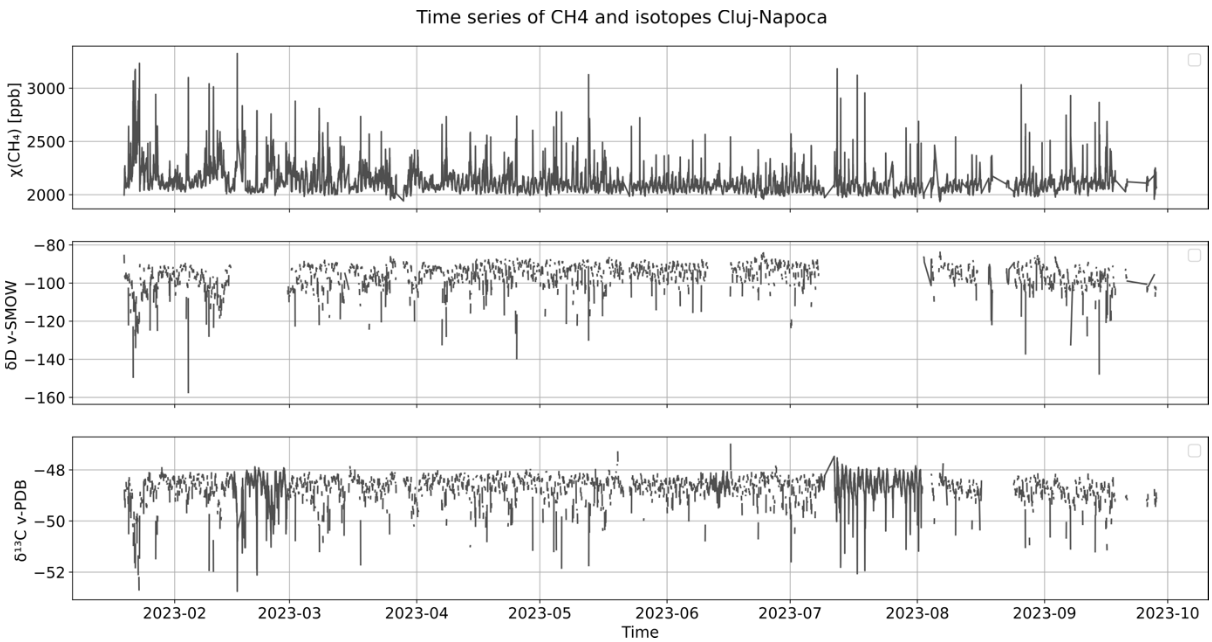

Fig. 2: Preliminary timeseries of methane mole fraction and isotope measurements in Cluj-Napoca (Romania). Gaps in this series are the result of system problems.

Four mobile campaigns have been conducted in Cluj-Napoca, Romania, to explain source signatures found in the continuous measurements. Samples were taken and analysed for δD-CH_4 and $\delta^{13}C$-CH_4 at Utrecht University. The samples for the 1st campaign are analysed and show from several microbial sources. Two samples from the gas network show that the gas network in Cluj contains methane originating from microbial reduction, similar to the Romeo campaign, which makes separation between biogenic sources and the gas network difficult (Menoud et al., 2022b). The network gas is similar in $\delta^{13}C$, but more enriched in δD compared to wetlands, agriculture, and waste, so network gas can be separated from these sources by the δD measurements. The measured samples partly explain the source signatures found during the continuous measurements, but some source signatures found in the continuous measurements are enriched in both δD and $\delta^{13}C$ which cannot be explained by these samples. Literature states that this is likely a thermogenic or pyrogenic source, which we attempted to samples during the follow-up campaigns (Menoud et al., 2022a).

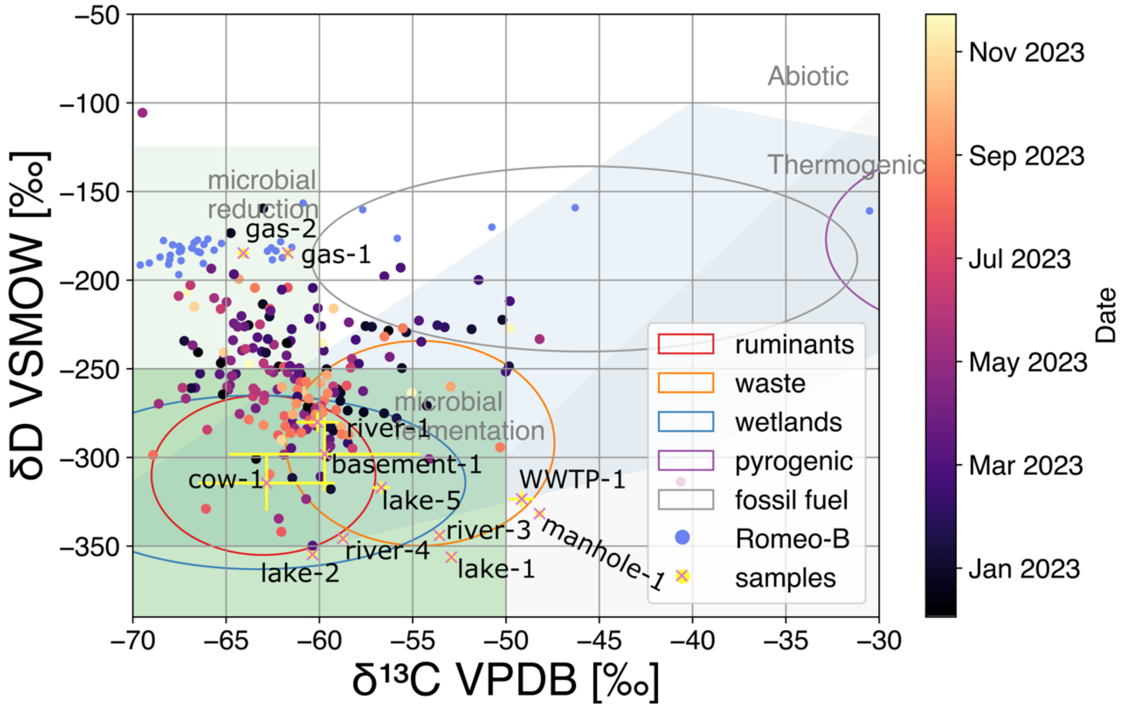

Fig. 3: Dual isotope plot of detected CH4 peaks from several measurement techniques. The green dots represent samples taken during the Romeo campaign. The dots coloured on dates represent the source signatures of the enhancements from the times series obtained from the continuous measurements. The red crosses with yellow errorbars and a label are the source signatures of the analysed samples that were taken during sampling campaigns.

4.4 Conclusion and possible impact

Mobile measurement campaigns have the potential to significantly improve local and global methane budgets, and by this support the implementation and integration of mitigation measures. This requires careful campaign planning and execution.

Current results show that continuous isotopic methane measurements can identify e.g. the diurnal cycles in methane mixing ratio and the isotopes. The data from these measurements can be used to further investigate the sources of these methane emissions. Mobile measurement campaigns can therefor help to identify the main emitters of methane emissions in Europe.

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5. History of the document

Version	*Author(s)*	*Date*	*Changes*
1	J.D. van Es	June 2023	First set-up
	J.D. van Es	October 2023	Draft for feedback sent
	J.D. van Es, S. Walter	November 2023 – February 2024	Feedback round
	J.D. van Es, S. Walter	March 2024	Finalising report and submission

Annex I

This Annex summarises work carried out by various groups to decide on the best placement for new methane isotope instruments as part of the PARIS project. The table gives a list of ICOS sites, the simulations run for each site and each group's recommendations.

I) Bristol/Met Office

Forward-modelled methane mole fractions were created using monthly transport footprints from NAME III v7.2 (the UK Met Office's Numerical Atmospheric Modelling Environment) for 26 ICOS and DECC sites for all of 2021.

Monthly fluxes were taken from the UK NAEI for the UK and EDGAR v6.0 for European lat/lons. Background mole fractions were modelled at the domain edges using the monthly ECMWF CAMS v19 CH4 volume mixing ratio dataset.

Sites were selected based on the mix of sources detected and the magnitude of modelled mole fractions.

Similarly to EMPA, they found that the modelled mole fractions matched the variation in observations well. Although there was some underestimation by the modelled values at some sites (notably LUT and PAL).

II) Empa

The FLEXPART-COSMO/IFS model was used to simulate methane mole fractions from two continuous Swiss Plateau sites (BRM and LHW), a background site (JFJ), two new sites (Breite and Empa) and Hamburg.

COSMO-1 analysis was used for the Alpine domain and ECMWF-HRES for the rest of the domain. Emissions were taken from Meteotest EKAT (2015) for Switzerland and from TNO for the rest of Europe. Background mole fractions were taken from the JFJ site.

They found that the modelled values matched the variability of the observations in time, but not in amplitude (modelled values were about 40% too small).

III) UU

ICOS sites were evaluated using three methods: information content (based on the method shown in Thompson et al., 2023, which used FLEXPART simulated values); posterior error covariance matrix; and source detection threshold methods (Szenasi 2020), using FLEXPART-COSMO and CHIMERE).

All these methods were tested with both EDGAR and TNO fluxes.

Sites were also ranked for priority based on the types of sources most detectable from these locations.

PARIS OSSE results:

x	Model run completed
x (grey)	Selected based on fossil/non-fossil mix and ch4 excursion size
x (blue)	Selected, based on information content
x (green)	Selected, based on error covariance
x (light yellow)	Selected, based on detection thresholds

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Site	Location	Network	Bristol/MO (NAME)	NAME details	Empa (FLEXPART)	FLEXPART details	STILT	STILT details	UU (FLEXPART, information content, error covariance and threshold	Other considerations (site feasability, modelling issues etc.)	Intitutes involved in isotope measurements	Methane isotope capability/previous campaigns	More information
BIR	Birkenes		x						x				
BRM	Beromünter				x								
BSD	Bilsdale	DECC	x								Bristol		
CBW	Cabauw	ICOS	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME			x		Utrecht	5 months in situ measurements (at low height 25 m inlet) across 2014-2015, and continuously since Oct 2010 with a G2210-i	https://acp.copernicus.org/articles/16/10469/2016/
CMN	Monte Cimone	ICOS	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME			x				
GAT	Gartow								x				
HEI	Heidelburg	None	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME					Heidelburg uni	Running a Picarro for d13C	
HEL	Helgoland		x						x				
HFD	Heathfield	DECC	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME					NPL/Edinburgh	Semi continuous operation of dual laser spectrometer for dD and d13C	
HPB	Hohenpeissenberg		x						x				
HTM	Hyltemossa		x						x				
HUN	Hegyhátsál	ICOS	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME					ATOMKI		
IPR	Ispra	ICOS			x	FLEXPART-COSMO at 1 km resolution since 2017			x				
IVG	Invergowrie	New site									NPL/Edinburgh		Yet to be commissioned
JFJ	Jungfraujoch		x		x				x				
JUE	Jülich												
KIT	Karlsruhe								x				
KRE	Křešín u Pacova	ICOS	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME			x				
LHW	Lägern-Hochwacht				x								
LIN	Lindenburg	ICOS	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME			x				
LMP	Lampedusa								x				
LUT	Lutjewad	ICOS	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME			x		Utrecht	6 months 2016-2017 UU-IRMS	https://www.tandfonline.com/doi/full/10.1080/16000889.2020.1823733
MHD	Mace Head	DECC/ICOS/AGAGE	x		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME					Bristol		Part of the NOAA/INSTAAR flask network for d13C

NOR	Norunda		x			x			
OPE	Obervatoire…		x			x			
OXK	Ochsenkopf		x			x			
PAL	Pallas		x			x			
PRS	Plateau Rosa					x			
PUI	Puijo								
PUY	Puy de Dome	ICOS		x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME				
RGL	Ridge Hill	DECC/ICOS	x	x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME	x	Bristol		
SAC	Saclay	ICOS	x	x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME	x			
SMR	Hyytiälä					x			
SSL	Schauisinland		x			x			
STE	Steinkimmen	ISOS	x	x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME	x			
SVB	Svartberget					x			
TAC	Tacolneston	DECC	x	x	FLEXPART-IFS with 0.1°x0.1° input, output as UKMO NAME		Bristol		
TOH	Torfhaus		x			x			
TRN	Trainou		x			x			
UTO	Uto		x			x			
WAO	Weybourne		x			x			
WES	Westerland		x			x			
ZSF	Zugspitze					x			
Breite	Breite			x					
Empa, Dübendorf	Empa, Dübendorf	None		x	FLEXPART-COSMO at 1 km resolution		Empa	1 month June 2014 UU-IRMS	https://amt.copernicus.org/articles/9/263/2016/
Hamburg, Geomatikum	Hamburg, Geomatikum	None		x	FLEXPART-IFS with high-resolution output		Utrecht		
Krakow	Krakow	None					Utrecht	6 months 2018-2019 UU-IRMS	https://acp.copernicus.org/articles/21/13167/2021/acp-21-13167-2021.html
Ny-Ålesund	Ny-Ålesund	AGAGE		x			Stockholm uni	Running a dual laser fr multiple years	
Romania (Cluj Napoca)	Romania (Cluj Napoca)	None					Utrecht	4 months of data by UU-IRMS 2023	