

# Current European in-situ atmospheric measurement capacity

Julia Marshall

che-project.eu





## D4.1 Current European insitu atmospheric measurement capacity

Dissemination Level:	Public
Author(s):	Julia Marshall (Max Planck Institute for Biogeochemistry)
Date:	29/06/2018
Version:	1.0
Contractual Delivery Date:	30/06/2018
Work Package/ Task:	4/ 4.1
Document Owner:	MPI-BGC
Contributors:	MPI-BGC
Status:	Final





## **CHE: CO2 Human Emissions Project**

Coordination and Support Action (CSA) H2020-EO-3-2017 Preparation for a European capacity to monitor CO2 anthropogenic emissions

Project Coordinator:Dr Gianpaolo Balsamo (ECMWF)Project Start Date:01/10/2017Project Duration:39 months

Published by the CHE Consortium

**Contact:** ECMWF, Shinfield Park, Reading, RG2 9AX, gianpaolo.balsamo@ecmwf.int



The CHE project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 776186.



## **Table of Contents**

1	Exe	Executive Summary6						
2	Intro	oduction	6					
	2.1	Background	6					
	2.2	Scope of this deliverable	8					
	2.2.	1 Objectives of this deliverables	8					
	2.2.	2 Work performed in this deliverable	8					
	2.2.	3 Deviations and counter measures	8					
3	Atm	nospheric CO <sub>2</sub> and CO measurements	8					
	3.1	Surface-based observations	9					
	3.2	Ground-based remote sensing	13					
	3.3	Operational aircraft-based measurements	15					
4	Atm	hospheric $\Delta^{14}$ CO <sub>2</sub> measurements	15					
	4.1	$\Delta^{14}CO_2$ measurements made within ICOS	15					
	4.2	European $\Delta^{14}CO_2$ measurements made outside of the ICOS framework	15					
5	Atm	nospheric APO measurements	16					
6	Urban flux tower measurements16							
7	Conclusion							
8	Ref	erences	19					

## **Figures**

Figure 1: The location of ground-based sites measuring atmospheric volume mixing ratios of CO <sub>2</sub>
and/or CO. ICOS Class 1 sites are shown in green, Class 2 sites are shown in blue, and non-ICOS
sites are shown in red. Class 1 sites, in green, will also make continuous measurements of APO
and integrated measurements of $\Delta^{14}CO_2$ . Some sites may be outside the domain of the figure.13
Figure 2: Currently operational European ground-based FTIR spectrometers contributing to the
TCCON network14
Figure 3: The location of the currently operating European urban flux sites given in Table 6

## **Tables**

#### C0<sub>2</sub>HUMAN EMISSIONS 2018

height. These sites will all be equipped with integrated radiocarbon measurements as described
in section 4. According to the ICOS Atmospheric station specifications, measurement of the
$O_2:N_2$ ratio, necessary for the calculation of ffCO <sub>2</sub> by APO (see section 5) is only a recommended
parameter
Table 3: A listing of the location of Class 2 ICOS atmospheric sites, based on information from the
ICOS Carbon Portal. These sites will all measure CO <sub>2</sub> continuously, and continuous
measurement of CO is a recommended parameter11
Table 4: European sites measuring CO <sub>2</sub> and/or CO that are not part of the ICOS framework. The
location, species, and frequency (freq.) of the measurements are summarized, where Cont.
means continuous measurements and Wkly means weekly flask measurements
Table 5: The location of all European TCCON sites, along with the year in which measurements
began. All stations measure XCO <sub>2</sub> and XCO, with varying temporal coverage depending on cloud
cover and instrument downtime14
Table 6: A listing of currently operating urban CO <sub>2</sub> flux towers found within Europe, with location,
starting year, and reference literature17

## **1** Executive Summary

This report describes the in-situ atmospheric measurements already being performed in Europe that are relevant for the determination of anthropogenic fluxes. These measurements include atmospheric volume mixing ratios of carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), the ratio of radiocarbon in carbon dioxide ( $\Delta^{14}CO_2$ ), and atmospheric potential oxygen (APO). Routine measurements made from ground-based remote sensing are included in this analysis, following the Copernicus definition of *in situ* to include everything except for satellite measurements. An attempt is made to summarize the location and frequency of these measurements, as well as their uncertainties. Additionally, the currently available urban flux tower measurements of CO<sub>2</sub> fluxes in Europe are described, as these measurements may provide relevant observational constraints on the diurnal, weekly, and annual fluctuations in anthropogenic emissions.

## 2 Introduction

#### 2.1 Background

Efforts are underway to develop a European capacity to monitor anthropogenic  $CO_2$  emissions, and the CHE project is a preparatory step in this process. While it is expected that such a capacity will rely heavily upon satellite measurements of column-integrated  $CO_2$  (and perhaps other tracers), it is clear that *in situ* measurements of  $CO_2$  will continue to play an essential role. These *in situ* measurements of  $CO_2$  provide continuity with measurement records on climate-relevant timescales, as well as providing a high-precision anchor with traceability to the WMO/GAW calibration scale.

However the atmospheric volume mixing ratio measures only the total CO<sub>2</sub>, which is the integral of all surface flux processes; additional tracers may provide information to help separate the signals of the biogenic and anthropogenic flux components from each other. While some of these tracers, such as CO (or NO<sub>2</sub>, which is not considered in this report) may be measurable by space-based remote sensing, other relevant species such as  $\Delta^{14}CO_2$  or APO are not. As such, the use of *in situ* measurements can play a pivotal role in helping to separate the atmospheric signals related to the different flux processes.

An example of such an additional tracer is carbon monoxide, or CO. CO emissions are correlated with anthropogenic  $CO_2$  fluxes as they are both largely the result of combustion processes. While in some parts of the world and in some seasons the CO signals are dominated by wildfires, this is not typically the case in Europe. Using measurements from an aircraft campaign in Asia, Palmer et al. (2006) found error correlations between the two species in excess of 0.7, and significant benefit when simultaneously assimilating both species. Similar results have been shown for the simultaneous assimilation of satellite measurements (Wang et al., 2009) and routine aircraft-based measurements of multiple species (Boschetti et al., 2017).

Assuming that one knows the combustion ratio of  $CO:CO_2$  ( $R_{CO:CO_2}$ ) well, one can use atmospheric measurements of CO to determine the  $CO_2$  signal of fossil fuel emissions in the atmosphere, ffCO<sub>2</sub>, via

$$ffCO_2 = \frac{CO_{obs} - CO_{bg}}{R_{CO:CO_2}}$$
(1)

In practice this is often not the case, and large uncertainties and spatiotemporal variability are associated with the ratio  $R_{CO:CO2}$  (Gamnitzer et al., 2006; Vogel et al., 2010).

 $\Delta^{14}$ CO<sub>2</sub> measurements make use of the fact that the carbon in CO<sub>2</sub> is a mixture of isotopes, i.e. atoms with the same number of protons and electrons, but a different number of neutrons. About 98.9% of naturally occurring carbon is <sup>12</sup>C, while most of the rest (~1.1%) is the stable isotope <sup>13</sup>C. Only minute trace amounts are found as radiocarbon, or <sup>14</sup>C, approximately 0.000000001%. This radiocarbon is radioactive and will eventually decay over time from <sup>14</sup>C to <sup>14</sup>N with a half-life of ~5700 years. Because the carbon being released through the combustion of fossil fuels is much older than this, it no longer contains any <sup>14</sup>C. Thus, by measuring the dilution of the background values of  $\Delta^{14}$ CO<sub>2</sub>, one can directly determine the signal of fossil fuel combustion in the air (ffCO<sub>2</sub>).

One limitation of this approach is the error in the background signal of <sup>14</sup>C due to production at nuclear facilities (Graven & Gruber, 2011), which must be taken into account. Typical sensitivities are around -2.6‰/ppm, however this sensitivity is decreasing over time due to cumulative dilution in the atmosphere through the burning of fossil fuel. Graven (2015) shows through simulations that this sensitivity will already drop to -1.6‰/ppm by 2050 given emissions following scenario RCP8.5, and to -0.8‰/ppm by 2100, necessitating considerable increases in the currently available measurement precision to maintain the current sensitivity to fossil fuel emissions via  $\Delta^{14}CO_2$  measurements.

Another method to determine ffCO<sub>2</sub> uses atmospheric potential oxygen (APO). This is defined as:

$$APO = O_2 + \frac{-1.1}{0.2095} (350 - CO_2)$$
 (2)

where  $O_2$  and  $CO_2$  are atmospheric measurements in per meg and ppm units respectively, 1.1 is the global ratio of  $O_2$ : $CO_2$  exchange between the terrestrial biosphere and the atmosphere, which accounts for the partitioning due to photosynthesis and respiration. The mole fraction of  $O_2$  molecules in dry air is 0.2095, and 350 is an arbitrary reference value (Pickers, 2016). Given these definitions, one can define ffCO<sub>2</sub> similarly to Equation 1:

$$ffCO_2 = \frac{APO - APO_{bg}}{R_{APO:CO_2}}$$
(3)

where  $R_{APO:CO2}$  is the APO:CO<sub>2</sub> combustion ratio for fossil fuel emissions, which depends on fuel type (Steinbach et al., 2011). Some uncertainty in the background value (APO<sub>bg</sub>) may be introduced by variability in ocean fluxes, but this is expected to take place on a different time scale.

Finally, one other source of measurements that might provide some constraint on the anthropogenic emissions is eddy covariance flux towers located in urban areas. In addition to potential use for direct measurement of fluxes and comparison with inventories (e.g. Kleingeld et al., 2018), they can provide information about the highly uncertain time factors associated with different emission sectors, which vary on hourly, weekly, and seasonal

scales. This information can provide additional context for the appropriate interpretation of atmospheric mixing ratios as well.

#### 2.2 Scope of this deliverable

#### 2.2.1 Objectives of this deliverables

An overview of the current European *in situ* measurement capacity for both CO and CO<sub>2</sub> is presented in Section 3. Section 4 presents the European sites at which atmospheric  $\Delta^{14}$ CO2 measurements are made, while section 5 presents the same information for APO. For clarification, the Copernicus definition of *in situ*, taken from <u>https://insitu.copernicus.eu/about</u>, is used throughout this report, namely:

ground-based remote sensing measurements, such as that from ground-based, sea-borne or air-borne monitoring systems. This includes, for example, data from sensors placed on the banks of rivers, tall towers, carried on weather balloons or airplanes, pulled through the sea by ships, and drifting in the ocean on floats or buoys. These non-space data are collectively referred to as in situ data (named using the Latin for in position, local or on site).

This rather inclusive definition was chosen in order to include as many non-satellite elements as are likely to be considered in future systems aiming to constrain anthropogenic emissions based on atmospheric measurements.

Finally, the distribution of currently operational urban flux tower measurements in Europe is presented in section 6.

#### 2.2.2 Work performed in this deliverable

The work of this deliverable consisted of the gathering and summarizing of information available from a variety of sources. A variety of sources were used for the collection of this information, including peer-reviewed literature, online databases, and project reports and documents. This literature is cited in the text. In addition to this, discussions with colleagues at my institute helped fill some of the gaps in the available documentation. These included discussions about ICOS data policies with Martin Heimann and Armin Jordan (head of the ICOS Central Analytical Laboratories), up to date information about IAGOS measurements from Christoph Gerbig, and information about radiocarbon measurements in Eastern Europe from Michal Gałkowski.

#### 2.2.3 Deviations and counter measures

The section on the urban flux tower measurements was not originally foreseen in the description of work. This arose following a discussion between partners at the first annual CHE project meeting. The assessment of the utility of these data for an operational system to monitor anthropogenic fluxes is considered exploratory within the CHE project, and is not included in the official deliverables. Nonetheless the decision was made to include the tabulation of these datasets in this deliverable for completeness.

### **3** Atmospheric CO<sub>2</sub> and CO measurements

In this section the two tracers CO<sub>2</sub> and CO are considered together, as these are very often measured simultaneously. Historically this was usually done with flask measurements, whereby discrete air samples are collected in the field in glass flasks, and then shipped to laboratories where they are analyzed for trace gas constituents, perhaps by gas chromatography for tracers such as CO<sub>2</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O, H<sub>2</sub>, and SF<sub>6</sub>. Additional instrumentation such as isotope ratio mass spectrometers allows for the analysis of additional tracers such as stable isotopes of CO<sub>2</sub> ( $\delta^{13}$ C and  $\delta^{18}$ O) or the ratio of O<sub>2</sub> to N<sub>2</sub>. While flasks are still collected at many sites, not least because of the range of tracers that

can be simultaneously measured, there is a practical limit to the temporal resolution that is possible with such an approach, and such flask measurements are rarely collected more frequently than once a week.

With the development of cavity ring-down spectrometry as a common and robust measurement method, CO<sub>2</sub> and CO are now often available simultaneously from the same continuous samplers, such as the Picarro G2401. This instrument is widely used in the research infrastructure ICOS (Integrated Carbon Observing System, https://www.icos-ri.eu/). The Picarro G2301 and G1301 also meet the ICOS specifications for the measurement of CO<sub>2</sub> but are not capable of measuring atmospheric CO simultaneously. Conversely, the Los Gatos instrument LGR 907-0015 and LGR 913-0015 instruments as well as the Picarro G5310 meet the ICOS specifications for the measurement of CO but not CO<sub>2</sub>. The ICOS guidelines for the measurement of CO<sub>2</sub> and CO, adapted from Table 3 of the ICOS Atmospheric Station Specification document v1.3 (November, 2017 https://icosatc.lsce.ipsl.fr/filebrowser/download/69422) can be found in Table 1. Here, continuous measurements are taken to mean hourly or more frequently, such that the diurnal cycle can be well resolved. While the instruments themselves measure much more frequently than that, due to calibration, averaging, and switching between inputs from multiple levels in the case of tall tower measurements, the resultant "continuous" concentration measurements are often at something closer to half-hourly temporal resolution.

Table 1: The measurement performance requirements required by ICOS for the measurement
of CO <sub>2</sub> and CO.

Component	Guaranteed range	Precision (1- σ 1' average raw data)	Precision (1-σ 60' average raw data)	Repeatability (1-σ 10' average raw data)
CO <sub>2</sub>	350-500 ppm	< 50 ppb	< 25 ppb	< 50 ppb
CO	30-1000 ppb	< 1 ppb	< 1 ppb	< 1 ppb

#### 3.1 Surface-based observations

Many of the surface-based observations of  $CO_2$  and CO in Europe have now been incorporated into the ICOS research infrastructure. This framework categorizes two tiers of atmospheric measurement sites: the more thoroughly instrumented Class 1 and Class 2.  $CO_2$  will be measured continuously from all Class 1 and Class 2 sites, and CO will be measured continuously from all Class 1 and most Class 2 sites, depending on the instrumentation used. The 17 Class 1 sites are listed in Table 2, while the 16 European Class 2 ICOS sites are listed in Table 3. All 33 of these ICOS Atmosphere sites will be providing continuous measurements of  $CO_2$ , and almost all will provide continuous measurements of CO as well. Those that do not will still have flasks analyzed for CO, albeit at lower temporal resolution. While not all of these stations are operational at the moment, the additional stations are expected to come online over the next year or two, and are thus considered "current" for the purpose of this forward-looking report. All surface-based *in situ* measurements of  $CO_2$  and CO are shown in Figure 2.1.

Table 2: A listing of the location of Class 1 ICOS atmospheric sites, based on information from the ICOS Carbon Portal. As for Class 2 sites, these will measure CO<sub>2</sub> and CO continuously from each height. These sites will all be equipped with integrated radiocarbon measurements as described in section 4. According to the ICOS Atmospheric Station Specifications, measurement of the O<sub>2</sub>:N<sub>2</sub> ratio, necessary for the calculation of ffCO<sub>2</sub> by APO (see section 5) is only a recommended parameter.

Station name	Code	Country	Latitude	Longitude	Altitude (masl)	Sampling height (magl)
SMEAR II-ICOS Hyytiälä	SMR	Finland	61.85 N	24.30 E	24.30 E 181	
Pallas	PAL	Finland	67.97 N	24.12 E	565	5
Gartow	GAT	Germany	53.07 N	11.44 E	69	30/60/132/ 216
Hohenpeißenberg	HPB	Germany	47.80 N	11.02 E	934	50/93/131
Potenza	POT	Italy	40.60 N	15.72 E	760	
Jungfraujoch	JFJ	Switzerland	46.55 N	7.98 E	3561.5	10
Křešín u Pacova	KRE	Czech Republic	49.57 N	15.08 E	534	250
Lindenberg	LIN	Germany	52.17 N	14.12 E	93	2.5/10/40/99
Karlsruhe	KIT	Germany	53.07 N	11.44 E	110	30/60/ 100/200
Jülich	JUE	Germany	50.93 N	6.23 E	110	12.5/20/ 32.5/52.5/ 82.5/102.5/ 120.0
Observatoire Pérenne	OPE	France	48.55 N	5.50 E	390	10/50/120
Ochsenkopf	OXK	Germany	50.03 N	11.81 E	1015	20/90/163
Saclay	SAC	France	48.72 N	2.14 E	160	5/60/100
Hyltemossa	HTM	Sweden	56.10 N	13.42 E	104	150
Norunda	NOR	Sweden	60.08 N	17.46 E	46	35/70/100
Svartberget	SVB	Sweden	64.24 N	19.43 E	235	150
Zeppelin Observatory	ZPO	Norway	78.92 N	11.84 E	474	

Of course not all surface-based measurements of CO<sub>2</sub> and CO have been incorporated into the research infrastructure of ICOS, not least because countries are required to commit to a national funding framework to support such measurements in order to officially join. Other non-ICOS European *in situ* measurements of CO<sub>2</sub> and CO are collected in Table 4, and included in Figure 2.1. Most of these stations contribute their data to the World Data Centre for Greenhouse Gases (<u>https://ds.data.jma.go.jp/gmd/wdcgg/</u>), operating under the World Meteorological Organization's Global Atmosphere Watch (WMO-GAW) programme.

## Table 3: A listing of the location of Class 2 ICOS atmospheric sites, based on information from the ICOS Carbon Portal. These sites will all measure CO<sub>2</sub> continuously, and continuous measurement of CO is a recommended parameter.

Station name	Code	Country	Latitude	Longitude	Altitude (masl)	Sampling height (magl)
Birkenes Observatory	BIR	Norway	58.39 N	8.25 E	190	40
Cabauw	CES	Netherlands	51.97 N	4.93 E	0	20/60/120/200
Monte Cimone	CMN	Italy	44.19 N	10.70 E	2165	12.7
Helgoland	HEL	Germany	54.18 N	7.89 E	4	10
Ispra	IPR	Italy	45.81 N	8.64 E	223	65
Lampedusa	LMP	Italy	35.52 N	12.63 E	45	10
Lutjewad	LUT	Netherlands	53.40 N	6.35 E	1	60
Plateau Rosa	PRS	Italy	45.93 N	7.70 E	3480	-
Puy de Dôme	PUY	France	45.77 N	2.97 E	1465	10
Staion Nord	SNO	Denmark	81.36 N	16.39 W	24	80
Steinkimmen	STK	Germany	53.04 N	8.46 E	29	
Torfhaus	ТОН	Germany	51.80 N	10.53 E	801	279s
Trainou	TRN	France	47.96 N	2.11 E	131	50/100/180
ICOS Utö- Baltic Sea	UTO	Finland	59.78 N	21.37 E	8	60
Weybourne	WAO	UK	52.95 N	1.12 E	15	10

## Table 4: European sites measuring CO<sub>2</sub> and/or CO that are not part of the ICOS framework. The location, species, and frequency (freq.) of the measurements are summarized, where Cont. means continuous measurements and Wkly means weekly flask measurements.

Station name	Country	Lat.	Lon.	Alt. (masl)	Sampling height (magl)	CO <sub>2</sub>	CO	freq.
Białystok	Poland	53.23 N	23.03 E	183	5/30/90/ 180/300	Y	Y	Cont.
Capo Granitola	Italy	37.67 N	12.65 E	5		Y	Y	Cont.
Giordan Lighthouse	Malta	36.07 N	14.22 E	160		Y	Y	Cont.
Hegyhatsal	Hungary	46.95 N	16.65 E	248	10/48/82/ 115	Y		Cont.
Hegyhatsal	Hungary	46.95 N	16.65 E	248	115	Y	Y	Wkly
Heimaey	Iceland	63.40 N	20.28 W	100		Y	Y	Wkly
Izaña	Spain	28.30 N	16.50 W	2367		Y	Y	Cont.
Kasprowy Wierch	Poland	49.23 N	19.98 E	1987	2	Y	Y	Cont.
Lamezia Terme	Italy	38.88 N	16.23 E	6		Y	Y	Cont.
Lecce - ISAC	Italy	40.34 N	18.12 E	36		Y	Y	Cont.
Mace Head	Ireland	53.33 N	9.90 W	25		Y	Y	Cont.
Puszcza Borecka	Poland	54.15 N	22.07 E	157		Y		Cont.
Sonnblick	Austria	47.05 N	12.95 E	3106		Y	Y	Cont.
Summit	Denmark	72.58 N	38.48 W	3238		Y	Y	Wkly
Terceira Island	Portugal	38.77 N	27.37 W	40		Y	Y	Wkly
Teriberka	Russia	69.20 N	35.10 E	40		Y		Wkly
Zugspitze	Germany	47.42 N	10.98 E	2656		Y	Y	Cont.



#### Ground-based in situ measurement sites

Figure 1: The location of ground-based sites measuring atmospheric volume mixing ratios of CO₂ and/or CO. ICOS Class 1 sites are shown in green, Class 2 sites are shown in blue, and non-ICOS sites are shown in red. Class 1 sites, in green, will also make continuous measurements of APO and integrated measurements of △<sup>14</sup>CO₂. Some sites may be outside the domain of the figure.

#### 3.2 Ground-based remote sensing

Ground-based remote sensing relies upon a similar measurement technique as used to make passive near-infrared satellite measurements, only looking directly at the sun as a source of radiation rather than at sunlight reflected and scattered from the earth's surface. This reduces the path-length-dependent errors associated with aerosol and cloud contamination, and leads to a higher-precision measurement that can be used for direct calibration and/or validation of satellite column-integrated measurements. It was with this goal in mind that the Total Carbon Column Observing Network (TCCON, Wunch et al. (2011)) was established, currently numbering more than 20 ground-based FTIR (Fourier Transform infrared) spectrometers worldwide. In addition to providing invaluable validation measurements for existing satellites such as GOSAT and OCO-2, the total column measurements (referred to as Xgas) can be used directly for carbon cycle studies and source-sink attribution. Measurements of XCO<sub>2</sub> and XCO are retrieved from TCCON spectra, with 1- $\sigma$  measurement precision for clear skies and solar zenith angles less than 82° estimated to be <0.25%. There are currently 10 operational TCCON sites in Europe, as listed in

#### C0<sub>2</sub>HUMAN EMISSIONS 2018

Table 5. The locations of the TCCON stations are shown in Figure 2.

Site	Country	Latitude	Longitude	Start year
Białystok	Poland	53.23 N	23.03 E	2009
Bremen	Germany	53.10 N	8.85 E	2004
Garmisch	Germany	47.48 N	11.06 E	2007
Izaña	Spain	28.30 N	16.50 W	2007
Karlsruhe	Germany	49.10 N	8.44 E	2009
Ny-Ålesund	Norway	78.90 N	11.90 E	2002
Orléans	France	47.97 N	2.11 E	2009
Paris	France	48.49 N	2.36 E	2014
Sodankylä	Finland	67.37 N	26.63 E	2009
Zugspitze	Germany	47.42 N	10.98 E	2012

## Table 5: The location of all European TCCON sites, along with the year in which measurements began. All stations measure XCO<sub>2</sub> and XCO, with varying temporal coverage depending on cloud cover and instrument downtime.

TCCON measurements are made in the near infrared (NIR) part of the spectrum. Similar measurements can be made with the same or similar instruments in the mid-infrared as well, and these measurements are part of the Network for the Detection of Atmospheric Composition Change (NDACC). These spectra can be used to retrieve a number of gaseous atmospheric components, including CO, but not XCO<sub>2</sub>. Because of this, and because the mid-infrared weighting function tends to peak in the upper troposphere, making it harder to use for source-sink attribution, these measurements are not considered further in this report



## Figure 2: Currently operational European ground-based FTIR spectrometers contributing to the TCCON network.

#### 3.3 Operational aircraft-based measurements

Through the IAGOS research infrastructure, commercial airliners are equipped with *in situ* instruments in order to measure atmospheric composition (Petzold et al., 2015). All 10 IAGOS-Core aircraft are equipped with the so- called Package 1 instrument, which continuously measures carbon monoxide (CO) by infrared absorption using the gas filter correlation technique. This results in a time resolution of 30 s, a precision of  $\pm 5\%$ , and an accuracy of  $\pm 5$  ppb. These aircraft belong to different airlines (including Lufthansa, Air France, Iberia, China Airlines, and Hawaiian Airlines), and as such achieve a well-distributed global coverage, with the largest concentration of profiles (during take-off and landing) over European airports in addition to high-altitude cruise data. There are plans to expand the IAGOS fleet to 20 aircraft, 5 of which will be equipped with Package 2d (Filges et al., 2015), which will additionally measure CO<sub>2</sub> continuously using cavity ring-down spectroscopy, but these measurements have not yet begun.

Because this is a distributed network with the exact location of the measurements being determined by the operational use of the airliners in question, it is difficult to clearly define the exact sampling. Each aircraft makes approximately two long-haul flights per day, resulting in four profiles per day. A continually updated map of all IAGOS flights to date can be found at <u>http://iagos.sedoo.fr/</u>.

### **4** Atmospheric $\Delta^{14}$ CO<sub>2</sub> measurements

Measurements of  $\Delta^{14}CO_2$  in the air are unfortunately rather expensive and difficult to carry out, which limits their availability. One of the leading laboratories in this technique is Heidelberg Radiocarbon Laboratory, which has recently been converted to the Central Radiocarbon Laboratory (CRL) of the ICOS Research Infrastructure. As such, this laboratory will be responsible for most of the radiocarbon measurements made in Europe going forward.

#### **4.1** $\triangle^{14}$ CO<sub>2</sub> measurements made within ICOS

The ICOS-CRL will continue the already established measurement technique of precision proportional gas counting, with a measurement precision of 2% or better, but the measurement capacity is limited to approximately 500 samples per. This is being extended through the use of Accelerator Mass Spectrometry (AMS) radiocarbon analysis, which is carried out at the CEZA (Curt-Engelhorn-Zentrum Archäometrie gGmbH) laboratory in Mannheim. The ICOS-CRL will extract the CO<sub>2</sub> from flask samples and graphitize it for subsequent analysis by CEZA. It is expected that up to 1500 samples per year might be analyzed in this way, with a target long-term compatibility between the two methods of 1‰.

While the flask measurements of  $\Delta^{14}CO_2$  provide instantaneous snap- shots of ffCO<sub>2</sub> in a given air mass, the decision was made within ICOS to rely primarily on high-volume  $\Delta^{14}CO_2$  samplers, which integrate two weeks' worth of sampling into one measurement, allowing the long-term large-scale changes in the European atmospheric ffCO<sub>2</sub> burden to be tracked. According to the ICOS Atmospheric Station Specifications, ICOS will collect integrated radiocarbon measurements from the highest sampling height of all Class 1 stations. The locations of these stations are summarized in Table 2, and plotted in Figure 1. As mentioned in Section 3, while not all of these stations are operational at the moment, the additional stations are expected to come online in the next years.

#### **4.2** European $\Delta^{14}$ CO<sub>2</sub> measurements made outside of the ICOS framework

The University of Heidelberg operated up to nine stations measuring  $\Delta^{14}CO_2$  in the Heidelberg sampling network over many years (Levin et al., 2010), as well as the station Schauinsland in Germany. Of these ten stations, four of which were in Europe (Mace Head, Izaña, Jungfraujoch, and Schauinsland), only Jungfraujoch and Schauinsland have been

operated beyond 2010 (Hammer & Levin, 2017; Graven et al., 2017). Jungfraujoch has since been granted Class 1 status within ICOS (see Table 2). There is also a long-term record directly in Heidelberg, collected on the roof the university, which will continue.

Some  $\Delta^{14}$ CO<sub>2</sub> sites have also been historically operated in Central/Eastern Europe, partly for the monitoring of radiocarbon production from nuclear power plants, as outlined in Wang (2015) and Svetlik et al. (2010), which discuss the stations Prague-Bulovka and Košetice in the Czech Republic, Bratislava in Slovakia, and Dunaföldvár in Hungary. Unfortunately there is no record of these measurements being continued beyond 2007. In contrast to this, there are two ongoing radiocarbon records in Poland, namely from Kasprowy Wierch and Kraków (Pazdur et al., 2013). The coordinates of Kasprowy Wierch are given in Table 4, while the Kraków measurements are collected at 50.07 N, 19.92 E, 220 masl.

## 5 Atmospheric APO measurements

High precision measurements of APO, or rather the ratio of  $O_2:N_2$ , are part of the Class 1 measurements of ICOS, and are suggested to be added to Class 2 sites as possible. The ICOS sites contain most of the few sites from which there is a historical record of APO in Europe, including Ochsenkopf, Jungfraujoch, Puy de Dôme, and Lutjewad (van der Laan-Luijkx et al., 2010) as well as Weybourne (Pickers, 2016). The only extant exceptions are Mace Head and Białystok, which are currently not part of ICOS. Indeed, the ICOS sites, as given in Tables 2 and 3 and summarized in Figure 1, promise to greatly extend the availability and applicability of the APO method of assessing anthropogenic fluxes.

### 6 Urban flux tower measurements

While the annual emissions on an annual scale are known with relatively high certainty based on national fuel use statistics, there are larger uncertainties associated with the spatial and temporal disaggregation of these national totals (Denier van der Gon et al., 2017). Here atmospheric measurements may be able to provide some constraint, if they are able to characterize the concentration gradients on smaller scales. The temporal disaggregation may further be constrained through the use of eddy flux tower measurements conducted in urban environments.

While these flux tower measurements are representative of fluxes over a rather small area of the city, careful footprint analysis can help estimate the contribution of each emission sector to the measured fluxes. In total, 17 flux towers were found that are currently measuring CO2 fluxes in urban environments in Europe, located in 13 different cities. (Multiple stations are operating in Basel, Helsinki, and London.) Further flux towers exist that are focussing on energy fluxes, but that is not the focus of this report. The location of these stations, the year they began collecting measurements, and reference literature where available are found in Table 6. The distribution of these urban flux sites can be found in Figure 3.

Most of these stations were found through their listing online in the Urban Flux Network (<u>http://ibis.geog.ubc.ca/urbanflux/</u>), with reference literature found through a combination of citations mentioned in Grimmond & Christen (2012), institute websites, and further literature searches. Comparison of the Urban Flux Network sites with those listed on the European Flux Database Cluster at http://www.europe-fluxdata.eu with the conditions of IGBP land cover type "URB" and flux type CO<sub>2</sub> yielded three additional sites in Italy: Giuliano, Osservatorio Valerio, and San Marcellino. All three are comparatively recent sites, beginning data collection in 2015, 2014, and 2014, respectively. As no peer-reviewed literature references are available yet to describe these comparatively young sites, the reader is referred to the PI contact information in the listing on the European Flux Database Cluster website.

## Table 6: A listing of currently operating urban CO2 flux towers found within Europe, with location, starting year, and reference literature.

Name	Country	Latitude	Longitude	Start yr.	Reference
Arnhem	Netherlands	51.98 N	5.92 E	2012	Kleingeld et al. (2018)
Basel (Aeschenplatz, Ba09)	Switzerland	47.55 N	7.60 E	2009	Lietzke et al. (2015)
Basel (Klingelbergstrasse, Ba04)	Switzerland	47.56 N	7.58 E	2004	Schmutz et al. (2016)
Berlin (Charlottenburg, Cfb)	Germany	52.51 N	13.33 E	2014	ucahs.org
Dublin (Marrowbone Lane, Db09)	Ireland	53.34 N	6.29 W	2009	Keogh (2015)
Florence (Ximeniano, Fl05)	Italy	43.78 N	11.25 E	2005	Matese et al. (2009)
Giugliano (IT-GIn)	Italy	40.95 N	14.12 E	2015	www.europe- fluxdata.eu
Helsinki (Kumpala, He05)	Finland	60.20 N	24.96 E	2005	Vesala et al. (2008)
Helsinki (Hotel Torni, He10h)	Finland	60.17 N	24.94 E	2010	Nordbo et al. (2013)
Łódź (Lipowa, Lo06)	Poland	51.76 N	19.45 E	2006	Pawlak et al. (2013)
London (BT Tower, Ld06)	UK	51.52 N	0.14 W	2006	Helfter et al. (2011)
London (KSK, Ld08)	UK	51.50 N	0.12 W	2008	Kotthaus & Grimmond (2012)
London (KSS, Ld09)	UK	51.50 N	0.12 W	2009	Kotthaus & Grimmond (2012)
Nantes (Na06)	France	47.25 N	1.53 W	2005	Ruban et al. (2010)
Osservatorio Valerio (It-OVr)	Italy	43.91 N	12.90 E	2014	www.europe- fluxdata.eu
San Marcellino (IT- SMc)	Italy	40.85 N	14.26 E	2014	www.europe- fluxdata.eu
Swindon (Sw11)	UK	51.58 N	1.80 W	2011	Ward et al., (2013)





## 7 Conclusion

This report provides a description of the currently available European measurement capacity for atmospheric mixing ratios of  $CO_2$ , as well as of tracers that might be relevant for the separation of anthropogenic and biogenic fluxes in an inverse modelling system. These additional tracers include carbon monoxide,  $\Delta^{14}CO_2$ , and atmospheric potential oxygen, calculated from measured  $O_2:N_2$  ratios. All operational non-satellite measurements were considered, in keeping with the Copernicus definition of *in situ*.

This information, providing the location and frequency with which these measurements are being made (or will be made in the near future, in the case of some not yet operational ICOS sites) will provide a baseline for the inverse modeling activities within WP4 in CHE.

Additionally, this report compiles metadata on the currently operating urban flux towers within Europe. Following discussions at the first annual CHE meeting, these data may be examined in an exploratory way to assess their utility for determining time factors for different sectors in emission inventories, currently a significant source of uncertainty.

## 8 References

Boschetti, F., Thouret, V., Maenhout, G. J., Totsche, K. U., Marshall, J. & Gerbig, C. (2017) Multi-species inversion and IAGOS airborne data for a better constraint of continental scale fluxes. Atmospheric Chemistry and Physics Discussions 2017:1–37.

Denier van der Gon, H. A. C., Kuenen, J. J. P., Janssens-Maenhout, G., Do'ring, U., Jonkers, S. & Visschedijk, A. (2017) TNO CAMS high resolution European emission inventory 2000–2014 for anthropogenic CO<sub>2</sub> and future years following two different pathways. Earth System Science Data Discussions 2017:1–30.

Filges, A., Gerbig, C., Chen, H., Franke, H., Klaus, C. & Jordan, A. (2015) The IAGOS-core greenhouse gas package: a measurement system for continuous airborne observations of CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O and CO. Tellus B: Chemical and Physical Meteorology 67(1):27989.

Gamnitzer, U., Karstens, U., Kromer, B., Neubert, R. E. M., Meijer, H. A. J., Schroeder, H. & Levin, I. (2006) Carbon monoxide: A quantitative tracer for fossil fuel CO<sub>2</sub>? Journal of Geophysical Research: Atmospheres 111(D22).

Graven, H., Allison, C. E., Etheridge, D. M., Hammer, S., Keeling, R. F., Levin, I., Meijer, H. A. J., Rubino, M., Tans, P. P., Trudinger, C. M., Vaughn, B. H. & White, J. W. C. (2017) Compiled records of carbon isotopes in atmospheric CO<sub>2</sub> for historical simulations in CMIP6. Geoscientific Model Development 10(12):4405–4417.

Graven, H. D. (2015) Impact of fossil fuel emissions on atmospheric radio- carbon and various applications of radiocarbon over this century. Proceedings of the National Academy of Sciences 112(31):9542–9545.

Graven, H. D. & Gruber, N. (2011) Continental-scale enrichment of atmospheric  ${}^{14}CO_2$  from the nuclear power industry: potential impact on the estimation of fossil fuel-derived CO<sub>2</sub>. Atmospheric Chemistry and Physics 11(23):12339–12349.

Grimmond, C. & Christen, A. (2012) Flux measurements in urban ecosystems. FluxLetter 5:1–8.

Hammer, S. & Levin, I. (2017) Monthly mean atmospheric  $\delta^{14}CO_2$  at Jungfraujoch and Schauinsland from 1986 to 2016. https://doi.org/10.11588/data/10100.

Helfter, C., Famulari, D., Phillips, G. J., Barlow, J. F., Wood, C. R., Grimmond, C. S. B. & Nemitz, E. (2011) Controls of carbon dioxide concentrations and fluxes above central London. Atmospheric Chemistry and Physics 11(5):1913–1928.

Keogh, S. (2015) An assessment of radiative, energy and carbon dioxide fluxes over three urban locations in Dublin. Ph.D. thesis, Maynooth University, National University of Ireland, Irish Climate Analysis and Re- search Units, Department of Geography.

Kleingeld, E., van Hove, B., Elbers, J. & Jacobs, C. (2018) Carbon dioxide fluxes in the city centre of Arnhem, A middle-sized Dutch city. Urban Climate 24:994 – 1010.

Kotthaus, S. & Grimmond, C. (2012) Identification of micro-scale anthropogenic  $CO_2$ , heat and moisture sources - processing eddy covariance fluxes for a dense urban environment. Atmospheric Environment 57:301–316.

Levin, I., Naegler, T., Kromer, B., Diehl, M., Francey, R., Gomez-Pelaez, A., Steele, P., Wagenbach, D., Weller, R. & Worthy, D. (2010) Observations and modelling of the global distribution and long-term trend of atmospheric <sup>14</sup>CO<sub>2</sub>. Tellus B: Chemical and Physical Meteorology 62(1):26–46.

Lietzke, B., Vogt, R., Feigenwinter, C. & Parlow, E. (2015) On the controlling factors for the variability of carbon dioxide flux in a heterogeneous urban environment. International Journal of Climatology 35(13):3921–3941.

Matese, A., Gioli, B., Vaccari, F. P., Zaldei, A. & Miglietta, F. (2009) Carbon dioxide emissions of the city center of Firenze, Italy: Measurement, evaluation, and source partitioning. Journal of Applied Meteorology and Climatology 48(9):1940–1947.

Nordbo, A., Järvi, L., Haapanala, S., Moilanen, J. & Vesala, T. (2013) Intra-city variation in urban morphology and turbulence structure in Helsinki, Finland. Boundary-Layer Meteorology 146(469).

Palmer, P. I., Suntharalingam, P., Jones, D. B. A., Jacob, D. J., Streets, D. G., Fu, Q., Vay, S. A. & Sachse, G. W. (2006) Using CO<sub>2</sub>:CO correlations to improve inverse analyses of carbon fluxes. Journal of Geophysical Re- search: Atmospheres 111(D12).

Pawlak, W., Fortuniak, K. & Siedlecki, M. (2013) Carbon dioxide flux in the centre of Łódz, Poland – Analysis of a 2-year eddy covariance measurement data set. International Journal of Climatology 31(2):232-243.

Pazdur, A., Kuc, T., Pawełczyk, S., Piotrowska, N., Sensuła, B. & Rozanski, K. (2013) Carbon isotope composition of atmospheric carbon dioxide in southern Poland: Imprint of anthropogenic  $CO_2$  emissions in regional biosphere. Radiocarbon, Vol 55, Nr 2–3, 2013, p 848–864.

Petzold, A., Thouret, V., Gerbig, C., Zahn, A., Brenninkmeijer, C. A. M., Gallagher, M., Hermann, M., Pontaud, M., Ziereis, H., Boulanger, D., Marshall, J., Ndlec, P., Smit, H. G. J., Friess, U., Flaud, J.-M., Wahner, A., Cammas, J.-P., Volz-Thomas, A. & TEAM, I. (2015) Global-scale atmosphere monitoring by in-service aircraft current achievements and future prospects of the European research infrastructure IAGOS. Tellus B: Chemical and Physical Meteorology 67(1):28452.

Pickers, P. (2016) New applications of continuous atmospheric  $O_2$  measurements: meridional transects across the Atlantic Ocean, and improved quantification of fossil fuelderived  $CO_2$ . Ph.D. thesis, University of East Anglia, School of Environmental Sciences.

Ruban, V., Rodriguez, F., Rosant, J. M., Lamprea Maldonado, K., Mosini, M. L., Lebouc, L., Pichon, P., Letellier, L., Rouaud, J. M., Martinet, L., Dormal, G., Panhalleux, V., Demare, D., Andrieu, H., Bagga, I., Piquet, T. & Mestayer, P. (2010) Le secteur atelier pluridisciplinaire (SAP), un observatoire de l'environnement urbain : Présentation des premiers résultats du suivi hydrologique et microclimatique du bassin du Pin Sec (Nantes). Bulletin des Laboratoires des Ponts et Chaussées (277):pp 5–18.

Schmutz, M., Vogt, R., Feigenwinter, C. & Parlow, E. (2016) Ten years of eddy covariance measurements in Basel, Switzerland: Seasonal and interannual variabilities of urban CO<sub>2</sub> mole fraction and flux. Journal of Geophysical Research: Atmospheres 121(14):8649–8667.

Steinbach, J., Gerbig, C., Rödenbeck, C., Karstens, U., Minejima, C. & Mukai, H. (2011) The CO<sub>2</sub> release and Oxygen uptake from Fossil Fuel Emission Estimate (COFFEE) dataset: effects from varying oxidative ratios. Atmospheric Chemistry and Physics 11(14):6855–6870.

Svetlik, I., Povinec, P. P., Molnár, M., Vána, M. and Sivo, A.: Radiocarbon in the air of Central Europe: Long-term investigations, Radiocarbon, 52(2), 823–834, 2010.

van der Laan-Luijkx, I. T., Karstens, U., Steinbach, J., Gerbig, C., Sirignano, C., Neubert, R. E. M., van der Laan, S. & Meijer, H. A. J. (2010)  $CO_2$ ,  $\delta O_2/N_2$  and APO: observations from the Lutjewad, Mace Head and F3 platform flask sampling network. Atmospheric Chemistry and Physics 10(21):10691–10704.

Vesala, T., Järvi, L., Lauiainen, S., Sogachev, A., Rannik, Ü., Mammarella, I., Keronen, P., Rinne, J., Riikonen, A. & Nikinmaa, E. (2008) Surface atmosphere interactions over complex urban terrain in Helsinki, Finland. Tellus B 60(2):188–199.

Vogel, F. R., Hammer, S., Steinhof, A., Bernd & Levin, K. I. (2010) Implication of weekly and diurnal <sup>14</sup>C calibration on hourly estimates of CO-based fossil fuel CO<sub>2</sub> at a moderately polluted site in southwestern Germany. Tellus B 62(5):512–520.

Wang, H., Jacob, D. J., Kopacz, M., Jones, D. B. A., Suntharalingam, P., Fisher, J. A., Nassar, R., Pawson, S. & Nielsen, J. E. (2009) Error correlation between CO<sub>2</sub> and CO as constraint for CO<sub>2</sub> flux inversions using satellite data. Atmospheric Chemistry and Physics 9(19):7313–7323.

Wang, Y. (2016) The potential of observations of radiocarbon in atmospheric  $CO_2$  for the atmospheric inversion of fossil fuel  $CO_2$  emission at regional scale. PhD thesis. Earth Sciences, Université Paris-Saclay.

Ward, H. C., Evans, J. G. & Grimmond, C. S. B. (2013) Multi-season eddy covariance observations of energy, water and carbon fluxes over a suburban area in Swindon, UK. Atmospheric Chemistry and Physics 13(9):4645–4666.

Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W. T., Sherlock, V. & Wennberg, P. O. (2011) The Total Carbon Column Observing Network. Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences 369(1943):2087–2112.

## **Document History**

Version	Author(s)	Date	Changes
0.1	Julia Marshall (MPI- BGC	01/06/2018	Initial Version
0.2	Julia Marshall (MPI- BGC)	12/06/2018	Version for internal review
1.0	Julia Marshall (MPI- BGC)	29/06/2018	Final Version

## **Internal Review History**

Internal Reviewers	Date	Comments
Michal Gałkowski (MPI-BGC)	26/06/2018	Approved with comments
Ignacio Pisso (NILU)	27/06/2018	Approved with comments

## **Estimated Effort Contribution per Partner**

Partner	Effort
MPG	1 PM
Total	1

This publication reflects the views only of the author, and the Commission cannot be held responsible for any use which may be made of the information contained therein.