



Horizon 2020 Societal challenge 5:  
Climate action, environment, resource  
efficiency and raw materials

# VERIFY

## Observation-based system for monitoring and verification of greenhouse gases

GA number 776810, RIA

<b>Deliverable number (relative in WP)</b>	D3.15
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## Changes with respect to the DoA

The shipment of the spectrometer pair, customs procedures, preparative measures for the very successful St. Petersburg city campaign, and finally realization of posterior shipment of one of the instruments across Russia to Yekaterinburg took longer than expected; nevertheless, 11 days of measurement were acquired at Yekaterinburg. The main delay was the data availability from the Russian partners because they prepared and submitted a publication before data submission to VERIFY.

## Dissemination and uptake

(Who will/could use this deliverable, within the project or outside the project?)

There are 2 different kinds of ground-based measurements:

1. The city campaign measurements, which was a mobile one in the sense that the instruments were re-distributed daily to measurement sites along the circumference of the city in order to have one instrument in the upwind and the other one in the downwind direction. The aim was the estimation of the city emissions.
2. Measurements at two different places (fixed): Yekaterinburg and St. Petersburg.

The information collected here can be used as input by WP3.3.3, W3.4.4 and the scientific community as a whole for validation of Satellite XCO<sub>2</sub>, for instance OCO-2, GOSAT and Sentinel-S5P. The city campaign data are especially useful for assessing the quality of emission inventories.

The satellite XCO<sub>2</sub> retrievals are useful for the inverse modelling community as observational input data set to obtain information on CO<sub>2</sub> sources and sinks. The latest version of this data set (FOCAL v08) has been used as input data set for the generation of the multi-sensor, multi-algorithm merged Level 2 and Level 3 data products as generated for the Copernicus Climate Change Service (C3S). The FOCAL algorithm development for OCO-2 and corresponding data set generation, validation and interpretation is co-funded, e.g., by ESA in the framework of the Climate Change Initiative (CCI). These activities will continue in 2021 and very likely also after the end of the VERIFY project. FOCAL is also one of three algorithms for retrieval of XCO<sub>2</sub> and other parameters used by EUMETSAT for inclusion into the ground system for processing future CO<sub>2</sub>M data.

## Short Summary of results (<250 words)

This report summarizes the execution of and the results achieved by the city emission campaign carried out in St. Petersburg Russia by using a pair of FTIR instruments in 2019.

Before shipment to Russia, the spectrometers were checked and calibrated at KIT, following the requirements of the COCCON (Collaborative Carbon Column Observing Network). After completion of the city campaign, one of the instruments was moved to Yekaterinburg while the other one remained at St. Petersburg, both collecting data at fixed locations. Column-averaged dry air mole fractions of CO<sub>2</sub> (XCO<sub>2</sub>) are reported.

The city emission campaign in St. Petersburg is a case of study in Eastern Europe, aiming at empirically quantifying the CO<sub>2</sub> emissions and the CO / CO<sub>2</sub> emission ratios for the city source by using a pair of ground based FTIR remote sensing instruments operated in the framework of COCCON (type EM27/SUN manufactured by Bruker). Both instruments were set-up in variable daily locations along the up and downwind directions of St. Petersburg's city. These locations were chosen depending on wind forecast and the NO<sub>2</sub> plume orientation as modeled by HYSPLIT one day before field deployment.

A multi-year global data set of column-averaged mole fraction of CO<sub>2</sub>, i.e., XCO<sub>2</sub> has been generated from NASA's OCO-2 mission using an improved version of the FOCAL retrieval algorithm.

#### Evidence of accomplishment (report, manuscript, web-link, other)

As a result of this milestone, two papers are in process: one is in the review process <https://amt.copernicus.org/preprints/amt-2020-87/amt-2020-87.pdf> and the second is planned to be submitted to ACP soon. The lead authorship of the publications has been granted to the Russian partners, as they were not eligible for direct financial support from VERIFY for recognizing their participation.

Moreover, all ground-based XCO<sub>2</sub> results collected in the framework of this activity are submitted with this report.

Version	Date	Description	Author (Organisation)
V0	09/10/20	Creation/Writing	Carlos Alberti (KIT) Michael Buchwitz (UB)
V0.1	20/10/20 - 30/10/20	Writing/Formatting/Delivery	Carlos Alberti(KIT), Michael Buchwitz (UB), Frank Hase (KIT)
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## 1. Glossary

Abbreviation / Acronym	Description/meaning
<b>FTIR</b>	Fourier Transform Infrared (spectrometer or spectrometry)
<b>C3S</b>	Copernicus Climate Change Service
<b>CCI</b>	Climate Change Initiative
<b>CO2M</b>	Anthropogenic CO <sub>2</sub> Monitoring (satellite mission)
<b>COCCON</b>	Collaborative Carbon Column Observing Network
<b>DOAS</b>	Differential Optical Absorption Spectroscopy
<b>EM27/SUN</b>	Commercial designation of the FTIR spectrometer type used
<b>EMME</b>	Emission Monitoring Mobile Experiment
<b>ESA</b>	European Space Agency
<b>EUMETSAT</b>	European Organization for the Exploitation of Meteorological Satellites
<b>FOCAL</b>	Fast atmospheric trace gas retrieval
<b>FTS</b>	Fourier Transform Spectrometer
<b>GHG</b>	GreenHouse Gases
<b>GNSS</b>	Global Navigation Satellite System
<b>HYSPLIT</b>	HYbrid Single Particle Lagrangian Integrated Trajectory Model
<b>JAXA</b>	Japan Aerospace Exploration Agency
<b>NASA</b>	National Aeronautics and Space Administration
<b>OCO-2</b>	Orbiting Carbon Observatory 2 (NASA mission)
<b>TCCON</b>	Total Carbon Column Observing Network
<b>XCH<sub>4</sub></b>	Column-averaged dry air molar fraction of methane
<b>XCO</b>	Column-averaged dry air molar fraction of carbon monoxide
<b>XCO<sub>2</sub></b>	Column-averaged dry air molar fraction of carbon dioxide

## 2. Executive Summary

XCO<sub>2</sub> and XCO measurements in Eastern Europe have been performed using a pair of portable FTIR spectrometers. The activity consisted of two parts: (1) a city campaign devoted to the quantification and characterization of the city emissions of St. Petersburg. For this purpose, the two spectrometers operated in the framework of COCCON (Frey et al., 2019) were arranged daily to appropriate sites upwind and downwind from the city. The observed column differences allow the quantification of the city emissions. After successful completion of this task, one spectrometer was moved to Yekaterinburg and started observations there, while the operation of the other spectrometer continued at St. Petersburg. This campaign activity allowed the observation of concentration gradients on a ~ 2000 km scale. Both campaign activities - St. Petersburg city observations and large baseline observations - were performed to support modelling activities and satellite validation.

A multi-year global data sets of column-averaged mole fraction of CO<sub>2</sub>, i.e., XCO<sub>2</sub> has been generated from NASA's OCO-2 mission using an improved version of the FOCAL retrieval algorithm. The latest version is FOCAL v08 and has been used to generate a 4-year data set covering the period 2015-2018. Using TCCON ground-based XCO<sub>2</sub> retrievals it has been estimated that the single observation random error is approximately 1.5 ppm and the spatial bias or relative accuracy is about 0.64 ppm. These estimates have been derived assuming an error free comparison method and error free TCCON data, i.e., the approximately 0.4 ppm uncertainty of the TCCON data has been neglected. Current, a new FOCAL version is in preparation including the generation of a data set which also covers 2019 and at least the first months of 2020. This new data set will overlap with the new COCCON measurements described in this document and will be used for comparisons of the satellite and COCCON XCO<sub>2</sub> retrievals in Russia. The further development of FOCAL and its use for the generation of global multi-year satellite-derived XCO<sub>2</sub> data sets is a major activity important for Europe not only for VERIFY but also to prepare for processing of future CO2M data (FOCAL has recently been selected by EUMETSAT to be one of three candidate operational retrieval algorithms for CO2M). At present OCO-2 data have been already re-processed several times with improved versions of FOCAL. This is an ongoing activity receiving co-funding also from other projects (CHE, ESA Climate Change Initiative (CCI)). Compared with the latest version of NASA's OCO-2 XCO<sub>2</sub> data product it has been identified that especially the yield of data after final quality filtering needs to be enlarged and to increase the yield is a focus of the ongoing development of FOCAL. FOCAL is designed not only to be accurate but also very fast. This is very important especially for future operational processing of CO2M data as CO2M will provide an order of magnitude more data than OCO-2. Thanks to the VERIFY project the FOCAL algorithm has been significantly improved such that high-quality multi-year global data sets can be generated and to be prepared to generate high-quality data sets also from CO2M in the future using an accurate and fast European retrieval algorithm.

### 3. Introduction

St. Petersburg and Yekaterinburg in Russia had been chosen as test region/cities. The main aim was the evaluation of XCO<sub>2</sub> gradients and the XCO / XCO<sub>2</sub> ratio in a very important region with high emissions and large biosphere fluxes in an eastern European country. To achieve the foreseen objectives two different activities were carried out:

- Mobile City Emission campaign in St. Petersburg: The quantification of the CO<sub>2</sub> emission and of CO / CO<sub>2</sub> emission ratios of the city can empirically be calculated by using the data obtained with two mobile ground based FTIR spectrometers (EM27/SUN, for technical details see: Gisi et al., 2012; Hase et al., 2016); one placed up and the other one downwind of St. Petersburg.
- Separated fixed FTIR measurements at St. Petersburg and Yekaterinburg: After the mobile campaign one of the instruments remained at St. Petersburg and the other one was moved to Yekaterinburg. With the raw data obtained from both instruments the column-averaged dry air mole fractions of CO<sub>2</sub> (termed XCO<sub>2</sub>) have been derived at the two sites separated by about 2000 km distance. These XCO<sub>2</sub> and XCO records and detected gradients can be used for validation of satellite data and to assess model predictions based on different emission inventories.

Retrieval of XCO<sub>2</sub> from satellites is challenging because of demanding requirements on accuracy. Furthermore, the retrieval algorithm not only has to be very accurate but also very fast due to large amounts of data satellites such as OCO-2 deliver. This will be even more challenging in the future for the planned constellation of Copernicus Anthropogenic CO<sub>2</sub> monitoring (CO<sub>2</sub>M) satellites (CO<sub>2</sub>M) as CO<sub>2</sub>M will deliver more than one order of magnitude more data than OCO-2. University of Bremen (UB) is developing the FOCAL algorithm to meet these requirements. Co-funded by VERIFY and other projects FOCAL has been further developed in recent years and applied to XCO<sub>2</sub> retrieval from OCO-2 and other satellites, including CO<sub>2</sub>M, as FOCAL is one of the candidate algorithms for CO<sub>2</sub>M. Several global multi-year data sets have been generated and made available for VERIFY and other projects. The latest version of FOCAL is version 08 and this version has been used to generate a 4-year data set covering the time period 2015-2018, i.e., the first four years of the OCO-2 mission.

### 4. Mobile City Emission campaign in St. Petersburg:

Relevant information about the campaign and an estimation of the city emissions deduced from the observations can be found in Makarova et al., in press, 2021.

While the actual estimation of a representative emission strength is an involved process and explained in detail in the aforementioned papers, we outline the basic methodology and underlying ideas here: The FTIR spectrometers deliver column-averaged abundances of trace



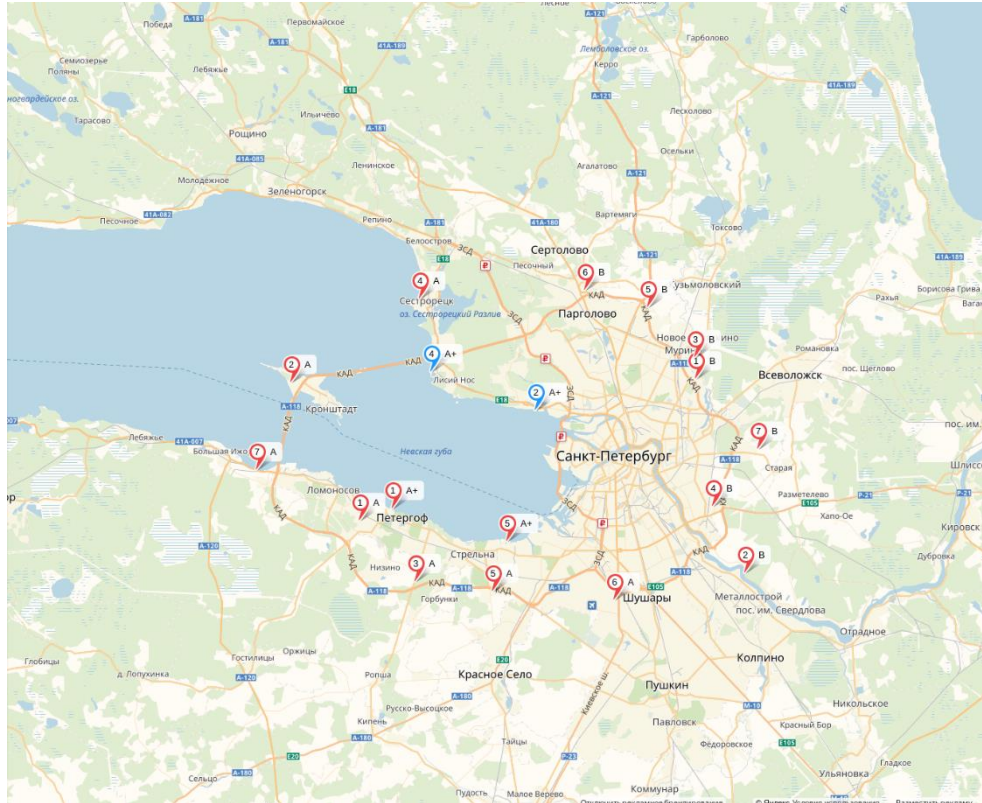
gases with a vertical sensitivity close to unity (or total column amounts), so the altitude of an air parcel enriched in CO<sub>2</sub> due to a local emitter does not matter for the observed enhancement. If a pair of spectrometers is arranged upstream and downstream from a source, then the emitted flux generates a difference in the observed column amounts.

Figure 8 shows the effect nicely for the campaign day April, 25, 2019. As can be seen, on this date the column difference in CO<sub>2</sub> amounts a few ppm. We abstract the emissions of St. Petersburg to form a homogeneous source filling a circular disc of ~20 km diameter, and assume the spectrometers being located diametrically opposed on the circumference of this disc. Then the source strength  $F$  of this circle of diameter  $D$  is connected with the observed column difference  $\Delta col$  by the wind speed  $w$  via  $\Delta col = DF/w$ . The dry air column for a ground pressure of  $10^5 Pa$  amounts to  $2.12 \cdot 10^{29} molec/m^2$ , so the aforementioned differential column signal is  $2.12 \cdot 10^{23} molec/m^2$  for 1 ppm difference between downstream and upstream. The wind speed on the example date was low, in the order of 1 m/s (see table 3). So the released flux of CO<sub>2</sub> molecules is  $10^{19} molec/m^2$  or 23 kt/km<sup>2</sup>yr (per ppm differential column).

In a proper quantification approach, the fact that the source strength varies over the area, changes of wind speed and direction with altitude, and daily and seasonal variations of emissions need to be taken into consideration. For redundancy and explicit sampling of temporal variations of the emissions, the measurements should be repeated under different wind situations, a permanent array of spectrometers would be the optimal choice for emission monitoring.

#### 4.1. Instrument set-up and locations

The basic idea was to select the position of the two instruments one day before good meteorological conditions appear. The desired positions on the circumference of the metropolitan area align with the up and downwind directions as illustrated in Figure 1. For achieving an optimal configuration, the NO<sub>2</sub> plume was modelled using HYSPLIT and then the positions of the FTIR spectrometers were selected accordingly. On the day of the measurements, additional mobile DOAS measurements were carried out by the Russian partners in order to measure the NO<sub>2</sub> total column flux over the city in real time. The last input helped to adjust the location of one or both spectrometers if deviations from the predicted plume orientation are detected.



**Figure 1:** Map with the possible choices for setting up the instruments (red markers).

## 4.2. Results

The results of this campaign have already been published. More details and the complete analysis of the results can be found in the paper by Makarova et al., 2021.

A further paper extending the analysis work on the observational dataset collected in the framework of the St. Petersburg campaign has been submitted recently by Dmitry V. Ionov et al., 2021.

The VERIFY COCCON campaign indicates a higher CO<sub>2</sub> emission flux (by about a factor of two) than reported in the EDGAR database (EDGAR, 2019), using the annual mean per capita emission for Russia reported by EDGAR for 2018, and assuming a population of 5 400 000 for St. Petersburg.

### 4.2.1. Campaign basics

The core instruments of the campaign were two portable ground-based FTIR spectrometers Bruker EM27/SUN (Gisi et al., 2012; Hase et al., 2016), operated in the framework of COCCON (Frey et al., 2019). These spectrometers allow us to obtain column-averaged dry molar fractions of CO<sub>2</sub>, CH<sub>4</sub> and CO and other gases.

The FTIR spectrometers were transported by car to the measurement locations where they were set-up. The geographic coordinates were registered by the GNSS sensor. A detached car battery with an inverter was used as a power supply which ensures about 3 hours of continuous operations. Under cold weather conditions, the instruments were covered by electric heating blankets. The integration time for a single spectrum constitutes about 1 min. Within this period, 10 interferograms are registered and averaged, from these a corresponding spectrum is derived. On each spectrum, a quantitative trace gas analysis is performed. For the processing, the COCCON processing codes have been used, which were developed and tested in the framework of ESA projects (further details on the data processing chain are provided in section 4.2.4).

The tropospheric NO<sub>2</sub> column was derived by with Mobile Zenith DOAS measurements by using an OceanOptics HR4000 spectrometer, which was mounted on board of a car and connected to a portable computer to ensure uninterrupted recording of spectra. Measurements were fully automated while the car was moving. The location of the car was recorded and saved on the computer by the GNSS. The route included the entire city ringway (the highway around St. Petersburg), therefore the main emission sources are inside the route and the position of the megacity plume can be detected with high accuracy. The described approach and the DOAS mobile experiment specific design have been implemented previously and the results have been published by Ionov and Poberovskii (2012, 2015, 2017, 2019).

Air samples were collected at the locations of both FTIR spectrometers in two air bags: when FTIR measurements started (the first bag) and before completion of FTIR measurements (the second bag). Each bag was sampled for about 40 min. In case of suitable weather and landscape conditions at the location of one of the FTIR spectrometers, sampling bags were lifted by a kite to an altitude of about 100 m. The laboratory analysis of the air samples was performed with the help of gas analysers. Gas analyser Los Gatos Research GGA 24r-EP was used for measuring volume mixing ratio (vmr) of CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>O. Gas analyser Los Gatos Research CO 23r was used for measuring vmr of CO and H<sub>2</sub>O. The concentration of NO and NO<sub>2</sub> (NO<sub>x</sub>) was measured by gas analyser ThermoScientific 42i-TL. For the monitoring of meteorological parameters, two weather stations and the microwave radiometer RPG-HATPRO were used. One portable weather station was operating either at upwind or at the downwind location of FTIR spectrometers. The atmospheric pressure measurements were performed at both up- and downwind locations. The second stationary weather station was operating on the roof of the building (56 m a.s.l.) of the Institute of Physics of St. Petersburg State University (SPbU) located about 25 km west from the city centre. The RPG-HATPRO radiometer was operating also on the roof of this 7<sup>th</sup> building and delivered information on the temperature and humidity vertical profiles together with the information on the cloud liquid water path (Kostsov, 2015; Kostsov et al., 2018).

#### **4.2.2. Field observations, weather conditions and auxiliary data**

The EMME field campaign in 2019 consisted of 11 days of measurements in March-April. For all days of the field campaign, Table 1 presents the information on the location of FTIR spectrometers, FTIR spectrometer identifier, number of bags of air samples, flight of a kite and

air sampling altitude. The last column of Table 1 includes information on the experimental setup (up-and downwind or cross sectional setup) and FTIR spectrometer operator's notes about meteorological phenomena, changes in cloud cover, and local air pollution events observed during FTIR field measurements. Below, we refer to the two Fourier Transform Spectrometers (FTS) as FTS#80 and FTS#84.

Date	Outside the city plume				Inside the city plume				DOAS Mobile	Comment
	Loc	FTS#	AS	Kite	Loc	FTS#	AS	Kite		
21.03	A1	#80	2	No	B7	#84	2	Yes	No	U&d setup, test FTIR field measurements, test flight of the kite without air sampling
27.03	A2	#84	2	No	B2	#80	2	No	Yes	U&d setup, A2 – no clouds, B2 – groups of clouds
01.04	A2	#84	2	No	B2	#80	2	No	Yes	U&d setup, A2 – no clouds, B2 – groups of clouds
03.04	A1	#84	2	No	B3	#80	2	No	Yes	U&d setup, clear sky for both locations
04.04	A5	#84	2	No	B3	#80	2	No	Yes	U&d setup, clear sky for both locations
06.04	B7	#84	2	No	A2	#80	2	No	No	U&d setup, clear sky and burning grass for both locations
16.04	A2	#84	2	No	A5+	#80	2	No	Yes	Cs setup, clear sky for both locations
18.04	B3	#80	2	No	A5,A6+	#84	2	No	Yes	U&d setup, clear sky for both locations
24.04	A2	#84	2	No	B2	#80	2	Yes, 100m	Yes	U&d setup, A2 – clear sky, B2 – light cirrostratus, sun halo
25.04	B3	#80	2	No	A5	#84	2	Yes, 70m	Yes	U&d setup, B3 – smoke plum in the field of view of FTIR spectrometer, A5 – light cirrostratus
30.04	B2	#80	2	No	A2	#84	2	No	Yes	U&d setup, B2 – cirrostratus, A2 – quickly developing altocumulus translucidus

**Table 1:** EMME-2019 observation details: the field experiment setup (up- and downwind “u&d” or cross sectional “cs”), the FTS location (Loc), the FTS identifier (FTS#), the number of bags of air samples (AS), indication of the kite launch and the corresponding air sampling altitude.

During the EMME-2019 two types of field experiment setup were implemented regarding the position of FTIR spectrometers relative to the dominant air flow (wind) direction:

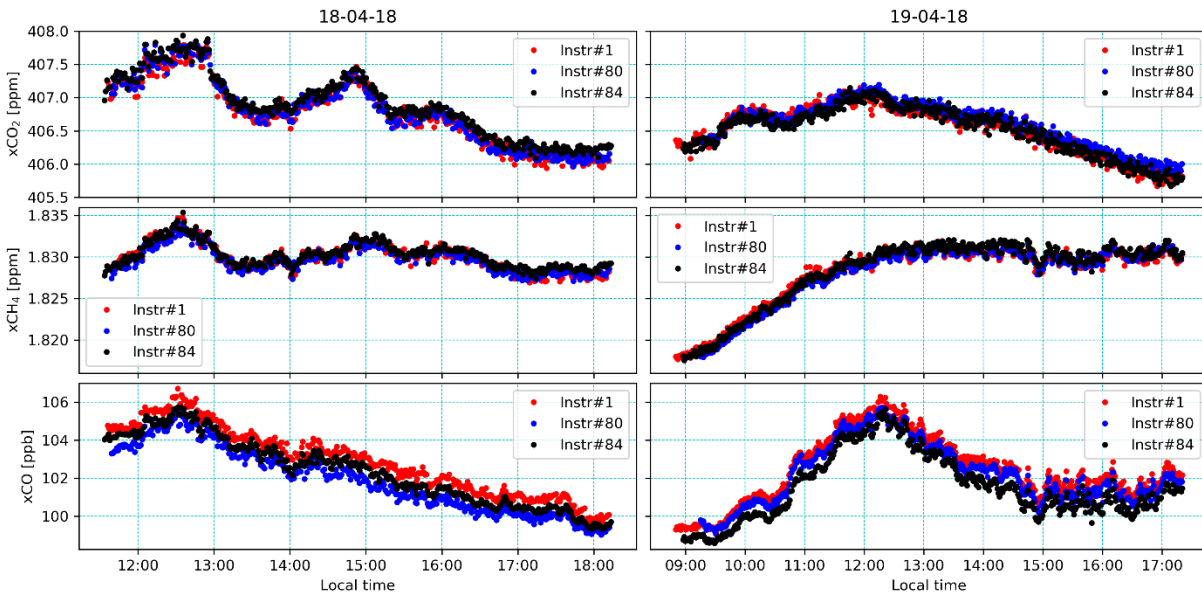
- for most of the days of observations (ten of the eleven), FTIR spectrometers were installed along the wind direction line -in up- and downwind locations on the opposite sides of the city of St. Petersburg (Fig.1, locations #1 and #2);
- for 16 April – the cross sectional setup was implemented. FTIR spectrometers were located on the line which is nearly perpendicular to the dominant wind direction line (not shown in Fig.1).

In order to forecast the spatial distribution of urban air pollution on each day of campaign observations, the HYSPLIT model was used. Following the Russian partner's previous experience of simulating the dispersion of urban contamination from St. Petersburg, the NO<sub>2</sub> content in the lower troposphere was set as a tracer of the polluted air mass distribution (Ionov and Poberovskii, 2019). This numerical modelling was done by means of the dispersion module within the offline version of HYSPLIT. Based on the plume evolution forecasts, the optimal pair of the FTIR spectrometer locations for the upcoming day of measurements was chosen. This approach to planning of the city campaign was implemented during 11 days of EMME-2019, and the necessity to change the location of the FTIR spectrometers occurred only once, on April 18 (see Table 1). For this day, the real-time information on the NO<sub>2</sub> tropospheric column (TrC) acquired along the ring-road by the crew #3 using mobile DOAS observations showed that the actual location of the most polluted city plume area was different from one which had been predicted by the HYSPLIT simulations.

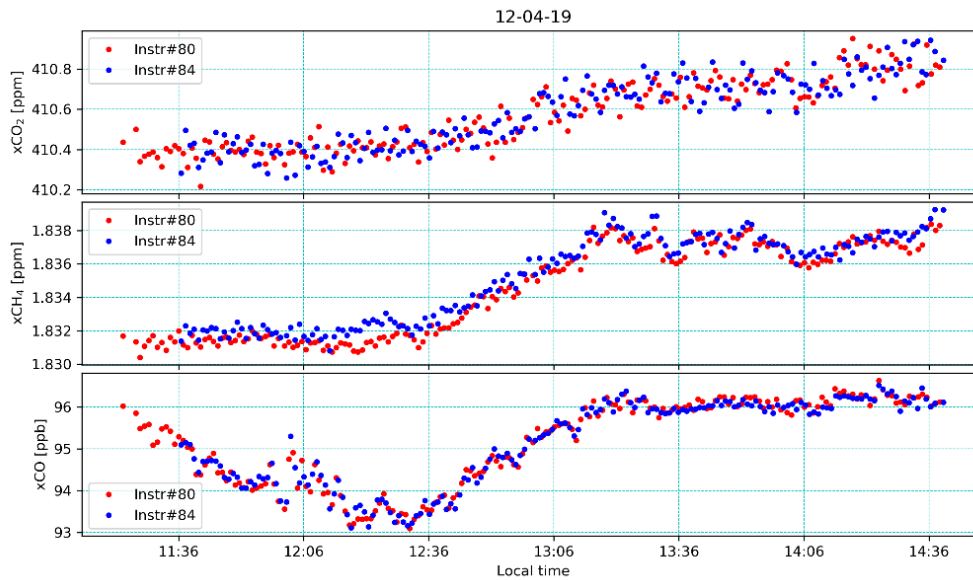
#### **4.2.3. Side-by-side calibration of FTIR spectrometers**

The target quantity of the observations is the small difference between two large values that are measured by different instruments of the same type. Therefore, a careful cross-calibration of the instruments is of primary importance for the considered experiment. Side-by-side calibrations of FTS#80 and FTS#84 were carried out before the instrument was sent to Russia and during the campaign period on the days: 12 April, 26 April, 15 May, and 16 May, 2019. Figures 2 and 3 show trace gas results collected side-by-side with the spectrometers involved in the campaign, indicating the high reliability of the devices.





**Figure 2:** Side by side measurements before the instruments were shipped to Russia. Comparisons between instrument #1(SN37), which is the COCCON reference unit operated at KIT, with instruments FTS #80 and FTS#84.



**Figure 3:** Side by side measurements during the campaign but only with instruments FTS#80 and FTS#84.

From the measurement showed in Figure 2, the corrections factors for XCO<sub>2</sub>, XCO and XCH<sub>4</sub> for both instruments have been calculated as described by Frey, M. et. al., 2019. These results are presented in the table 2:

Instrument	XCO <sub>2</sub> _factor	XCO_factor	XCH <sub>4</sub> _factor
FTS#80	0.99987659	1.00635991	1.00012521
FTS#84	0.99989555	1.00748183	0.99986552

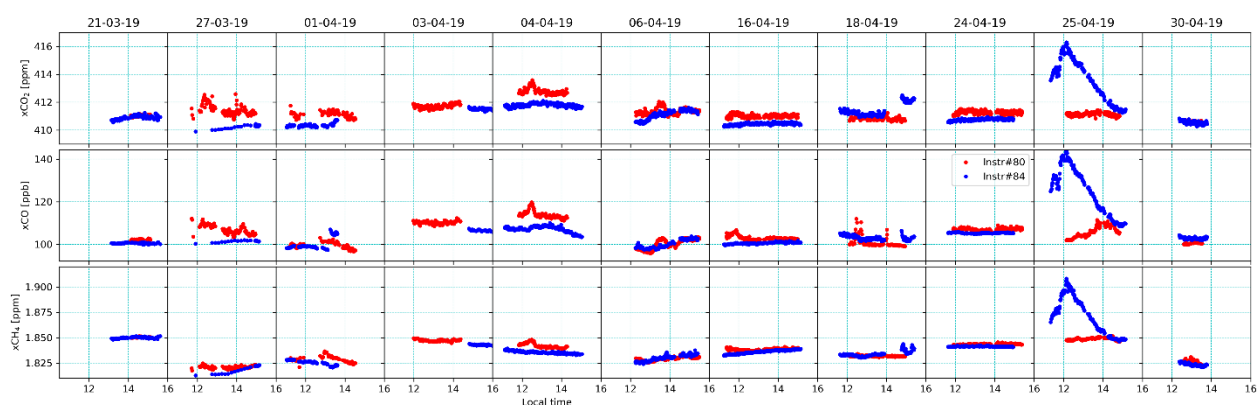
**Table 2:** Instrument-specific calibration factors for instruments FTS#80 and FTS#84.

#### 4.2.4. FTIR and DOAS data processing

The processing of the raw EM27/SUN data was performed using the software tools provided by the COCCON (Frey et al., 2019; <https://www.imk-asf.kit.edu/english/COCCON.php>). The required software is open source and freely available; the development of these tools has been supported by ESA. The interferograms recorded with FTS#80 and FTS#84 were the main input data. In the first processing step, spectra are generated from the recorded DC-coupled interferograms, including a DC correction (Keppel-Aleks et al., 2007) and quality filtering. In the second processing step, column-averaged abundances of the target species are derived from the spectra. The required auxiliary data are the local ground pressure, the temperature profile and the a priori mixing ratio profiles of the gases. For ensuring consistency with the TCCON reference network in this regard, these atmospheric profiles were retrieved from TCCON. As a result, the time series of Xgas and total column (TC) were obtained for CO<sub>2</sub>, CO and CH<sub>4</sub> for each day of measurements at each observational location. For the interpretation of spectral measurements and the derivation of tropospheric NO<sub>2</sub> content, the well-known DOAS method for Azimuth Mobile measurements was used (Platt and Stutz, 2008).

#### 4.2.5. General results

In the following figure, the general variations of the data are showed.



**Figure 4:** General overview of the full campaign results

#### 4.2.5.1. Example of a favourable day of measurements: 25 April

In order to illustrate the interpretation of experimental data and describe the main error sources of final results, we consider one day of the campaign: 25.04.2019. On that day, both FTS locations appeared to be inside the polluted area. This happened due to the specific weather conditions that contribute to the accumulation of air pollutants in the boundary layer: calm night before and light winds of 1 m s<sup>-1</sup> in the daytime (see Table 3 and 4).

Date	T(°C)	RH(%)	WD	WS(m/s)
25 April	20.9	23	WSW	1

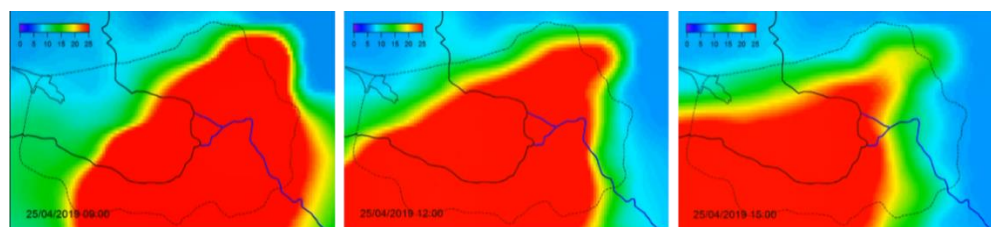
**Table 3:** Basic meteorological data for 25.04.2019: surface air temperature (T), relative humidity (RH), wind speed (WS) and wind direction (WD) at local noon. The meteorological data refers to one of the observational sites in the city of St. Petersburg ([http://rp5.ru/Weather\\_archive\\_in\\_Saint\\_Petersburg](http://rp5.ru/Weather_archive_in_Saint_Petersburg)).

Date	Wind Speed(m/s)			Wind direction (°)		
	Local	GDAS	HYSPLIT	Local	GDAS	HYSPLIT
25 April	1	2	1	69	95	71

**Table 4:** The wind speed and the direction for 25.04.2019 retrieved from different data sources: in situ observations (LOCAL), globally gridded assimilated data (GDAS) and backward trajectory calculations (HYSPLIT).

#### 4.2.5.2. Hysplit forecast for NO<sub>2</sub> plume

In the next figure the Hysplit results are presented for 10:00, 12:00 and 15:00. The orientation of the NO<sub>2</sub>-plume as predicted by Hysplit served for the daily selection of optimal locations of the spectrometers in order to achieve an upwind-downwind configuration around St. Petersburg.

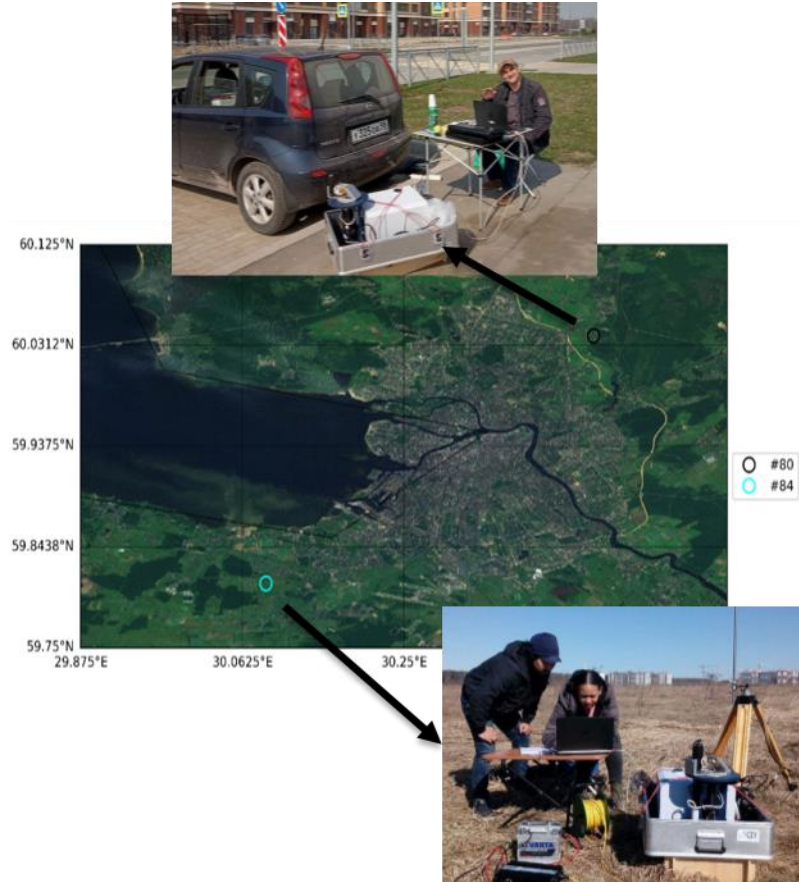


**Figure 5:** Hysplit simulation of NO<sub>2</sub> plume at 10, 12 and 15 pm on 25.04.2019.



#### 4.2.5.3. Instrument locations

After the NO<sub>2</sub> plume was computed, the initial position of the instruments was selected as it can be seen in the next figure.



**Figure 6:** Locations of the instrument on 25.04.2020

#### 4.2.5.4. Mobile Zenith DOAS results

In order to check the accurateness of the NO<sub>2</sub> plume forecast, one loop of Mobile Zenith DOAS measurements was carried out as shown in figure 7:

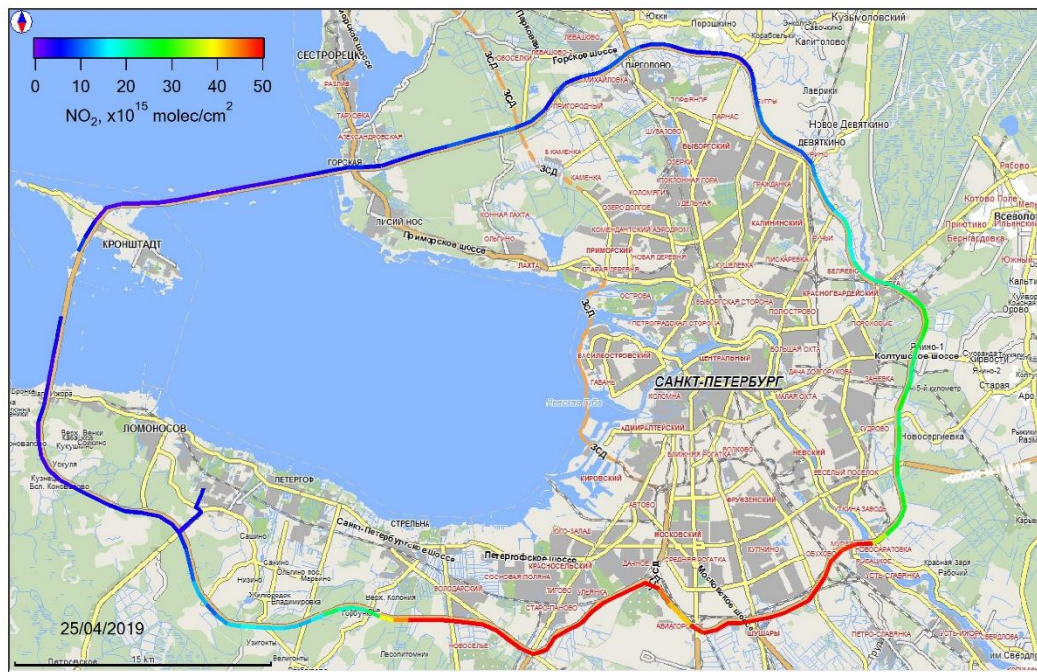
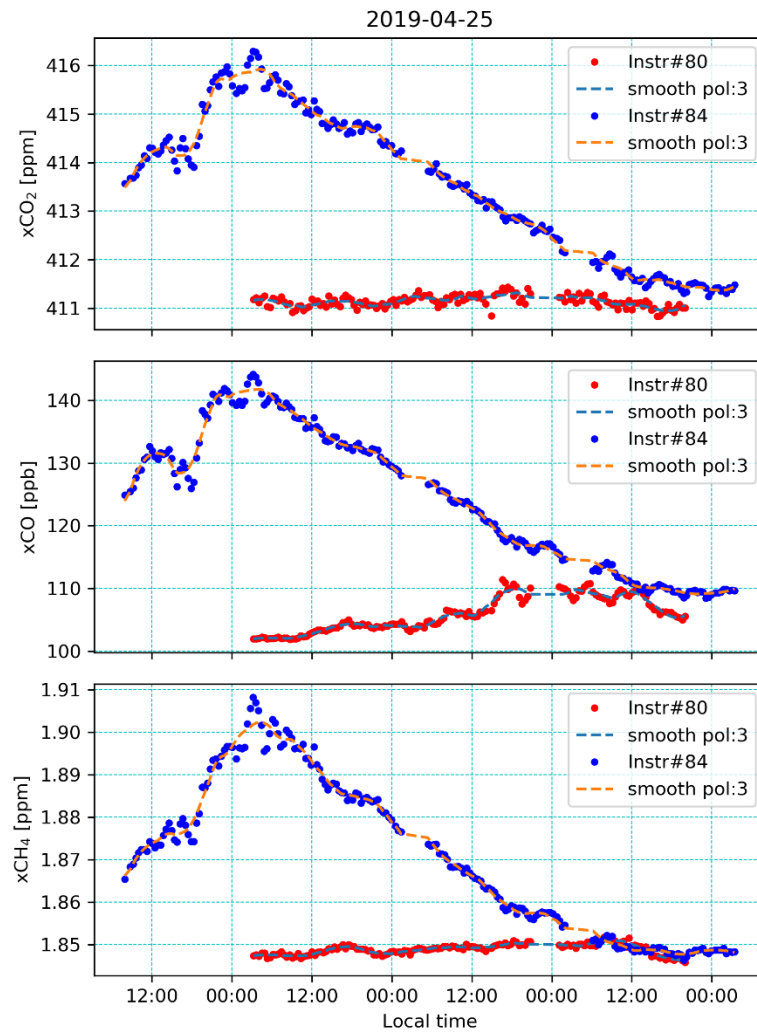


Figure 7: Mobile Zenith DOAS results of one loop over the city ring for 25.04.2020.

#### 4.2.5.5. FTIR Results

The final results of the FTIR measurements for a sample day can be seen in figure 8 below. Strong enhancements in XCO<sub>2</sub>, XCO, and XCH<sub>4</sub> are observed by the spectrometer

downwind from St. Petersburg, while the spectrometer located upwind records background concentrations



**Figure 8:** Results for XCO<sub>2</sub>, XCO and XCH<sub>4</sub> on 25.04.2020

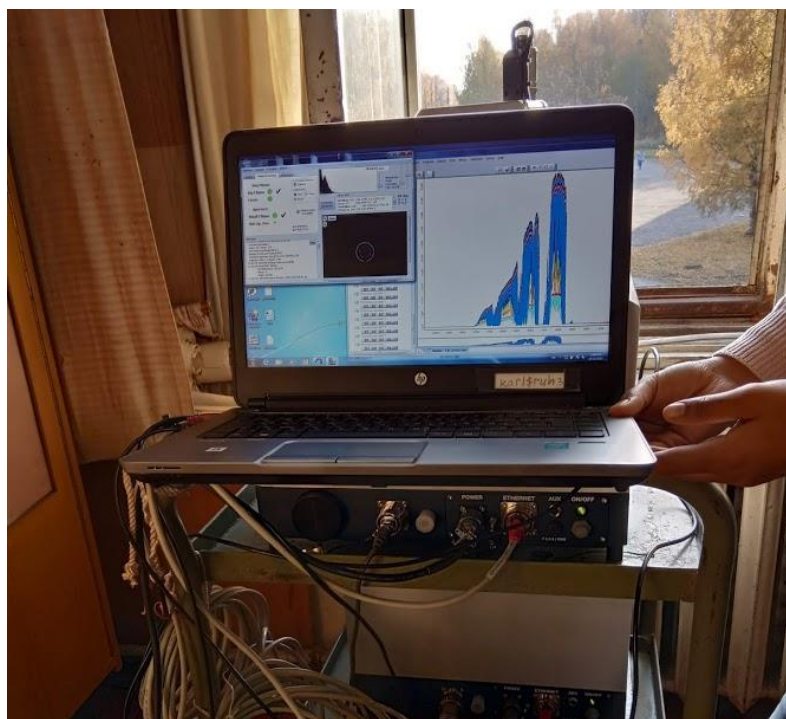
## 5. Distributed measurements at St. Petersburg and Yekaterinburg:

### 5.1. St. Petersburg

For the long-term long-baseline campaign, the instrument FTS#80 remained at Peterhof station of St. Petersburg State University, while the other spectrometer FTS#84 was moved to Yekaterinburg. Peterhof is a suburb of St. Petersburg located circa 35 km southwest from Saint Petersburg's city center. The instrument in St. Petersburg was operated by the Russian partners lead by Dr. Maria Makarova at the Faculty of Physics at the St. Petersburg State University, while the spectrometer in Yekaterinburg was operated by the Ural State University.

#### 5.1.1. Instrument location

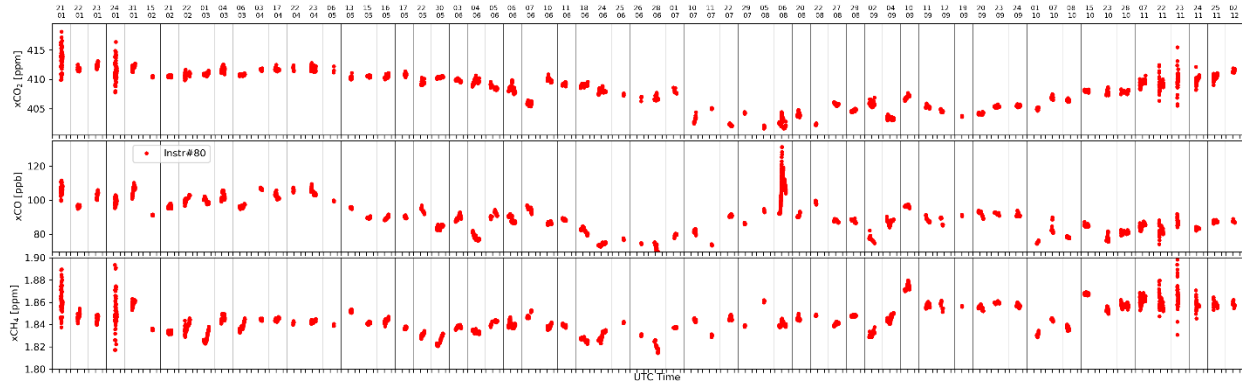
The instrument was set-up every almost or completely sunny day (out from the city campaign period) on the 4<sup>th</sup> floor of the FTIR remote sensing group.



**Figure 9:** Instrument set-up at Peterhof. A huge window allowed measurements from ~ 10:00 am to ~15:30 every day.

### 5.1.2. Results

Sixty-six days of measurements were successfully carried out. In figure 10, the results can be seen.



**Figure 10:** Time series for XCO<sub>2</sub>, XCO and XCH<sub>4</sub> obtained during the long-term period at Peterhof.

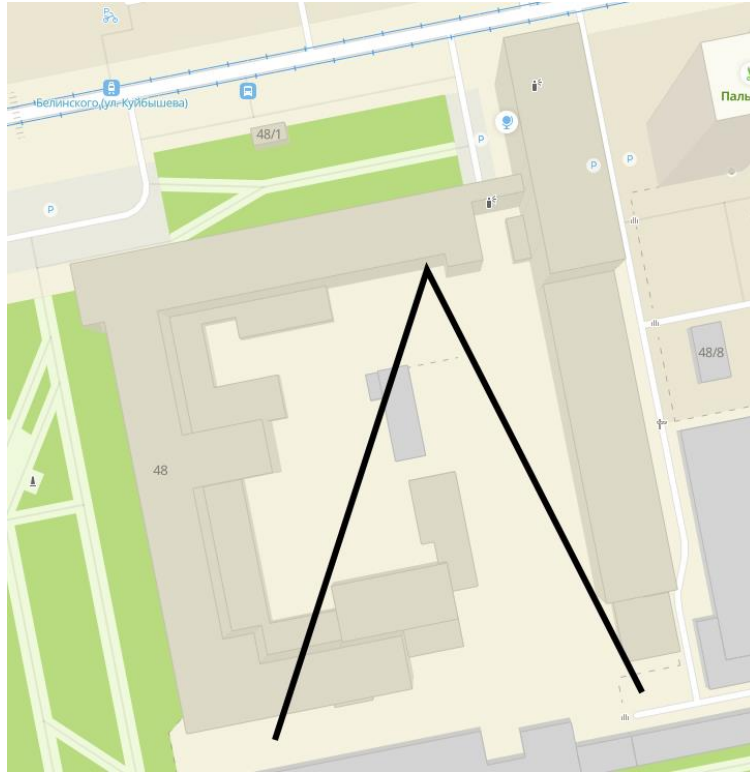
## 5.2. Yekaterinburg

After the city campaign was carried out, the instrument FTS#84 was transported to Yekaterinburg. This process was not as fast as expected mainly due to the preparations required for ensuring a safe inland transport in Russia. Nevertheless, the instrument was finally set-up on October 2019 and kept measuring until the very last day before to be shipped back to KIT in Germany. Dr. Konstantin Gribov, leading researcher at the Climate and Environmental Physics Laboratory INSMA of the Ural Federal University (UrFU) operated the instrument.

### 5.2.1. Instrument location and set-up

The instrument was set-up in an internal yard of Ural Federal University building (56.828N, 60.619E); therefore, good weather and the building structure, which blocked the sunlight, were a limitation. Sometimes high trucks passing in the yard blocked the field of view of the instrument. The spectrometer stood on the windowsill of the basement, so it was located exactly at ground level ~ 260 m. Under good weather conditions, measurements were carried out in the time approximately between 11:00 and 14:30 of local time.

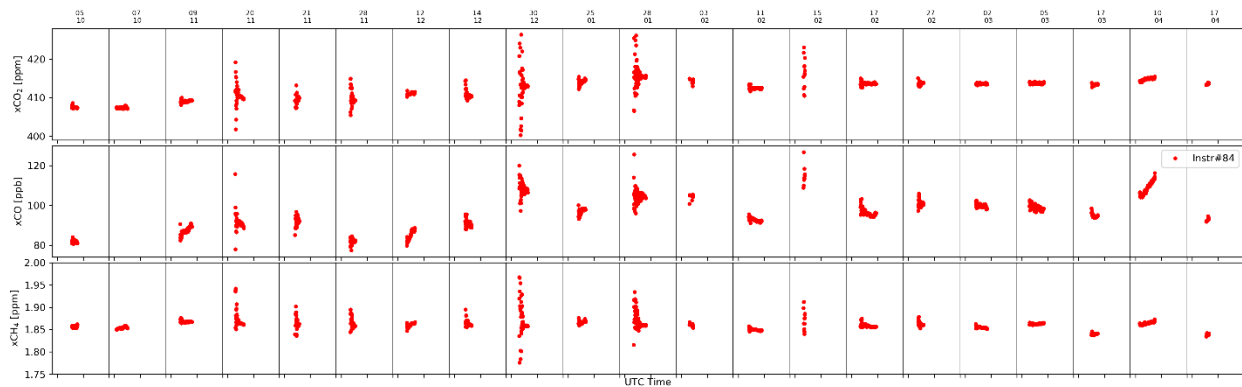




**Figure 11:** Map diagram of a building on which an approximate viewing angle is indicated.

### 5.2.2. Results

Twenty-two successfully day of measurements were successfully carried out on this site. The next plot shows the time series for the main GHG's retrieved.



**Figure 12:** Times series for XCO<sub>2</sub>, XCO and XCH<sub>4</sub> retrieved at Yekaterinburg.

## 6. Data availability

The final results for the city campaign and the long term run in Peterhof and Yekaterinburg are available in one single \*.dat file internally at the VERIFY website. The header of that document is self-explaining and contains all the necessary information for the users. Appendix A provides an overview over the available data, listing the locations of spectrometers, dates, start and end time of instrument operation during each day, and the total number of observations collected during each day.

The long-term stability of instrumental characteristics and the measures taken to ensure network consistency within COCCON has been described by Frey et al., 2019. The performance of a COCCON including the data processing chain has been demonstrated in the framework of ESA's FRM4GHG project. The campaign results have been reported by Sha et al., 2020.

Because FOCAL data products for 2019 and later are still in preparation, we cannot demonstrate a direct use of the COCCON observations for validation of the FOCAL XCO<sub>2</sub> data product in this document (as the COCCON campaign was conducted in 2019). As example of application of boreal COCCON data, we refer the reader to the recent study by Tu et al., 2020, using COCCON data collected in Northern Scandinavia for investigating the skills of CAMS model data and S5P measurements.

## 7. Satellite XCO<sub>2</sub> retrievals

The Fast atmospheric trace gas retrieval (FOCAL) algorithm (Reuter et al., 2017a, 2017b) has been further improved and used to generate multi-year data sets of XCO<sub>2</sub> retrieved from NASA's OCO-2 mission.

The latest version of FOCAL is version 08. The data set generated with FOCAL v08 covers the time period 2015 – 2018. This version of FOCAL and the corresponding data set will be described in this section including validation.

Previous data product versions as generated using VERIFY funding have been made available during and for the VERIFY project.

The improvement of FOCAL is an ongoing activity and will not stop when the VERIFY project is finished. New versions will be used to generate improved data sets. For example, an extended data set generated with v09 covering (at least) the time period 2015 – 2019 is currently in preparation. This new data set overlaps with the COCCON XCO<sub>2</sub> retrievals described in this document and will be used for comparisons of the satellite data with the COCCON retrievals.

Since the start of the VERIFY project a number of FOCAL related activities have been carried out in parallel. For example, FOCAL is also applied to simulated CO<sub>2</sub>M data as FOCAL is one of the candidate algorithms for CO<sub>2</sub>M. CO<sub>2</sub>M ("CO<sub>2</sub> Sentinels") is a planned constellation of EU Copernicus Anthropogenic CO<sub>2</sub> monitoring satellites to be launched from 2025 onwards. Furthermore, the Japanese space agency JAXA has established a contract with University of Bremen to apply FOCAL also to GOSAT and GOSAT-2. The further development of FOCAL and its application to different satellites and/or for different projects is currently co-funded by several institutions and related projects in addition to VERIFY. These co-funding sources are the EU (CHE project), ESA (CO<sub>2</sub>M, OCO-2), EUMETSAT (CO<sub>2</sub>M) and JAXA (GOSAT, GOSAT-2).

Concerning ESA projects, FOCAL is not only used for CO<sub>2</sub>M related assessments but ESA also co-funds the further development and application of FOCAL w.r.t. XCO<sub>2</sub> retrieval from OCO-2 as part of the ESA Climate Change Initiative (CCI).

The FOCAL v08 data product is available from the FOCAL website hosted by University of Bremen (<http://www.iup.uni-bremen.de/~mreuter/focal.php>) but also from the ESA CCI open Data Portal (<https://climate.esa.int/en/odp/#/dashboard>). Data access and additional information on this product is also available via the CEDA Archive, see <https://catalogue.ceda.ac.uk/uuid/b06213c3f3934a689f89ab22aa50e471> and <https://catalogue.ceda.ac.uk/uuid/b06213c3f3934a689f89ab22aa50e471?jump=related-docs-anchor>.



The FOCAL v08 data product has also been used as one of the input data sets for the generation of the multi-sensor, multi-algorithm Level 2 and Level 3 XCO<sub>2</sub> products (Reuter et al., 2020) as generated in the framework of the EU Copernicus Climate Change Service (C3S, <https://climate.copernicus.eu/>). These merged data products are available via the Copernicus Climate Data Store (CDS, <https://cds.climate.copernicus.eu/#!/home>).

Detailed information on the FOCAL OCO-2 XCO<sub>2</sub> v08 data product is available from several documents:

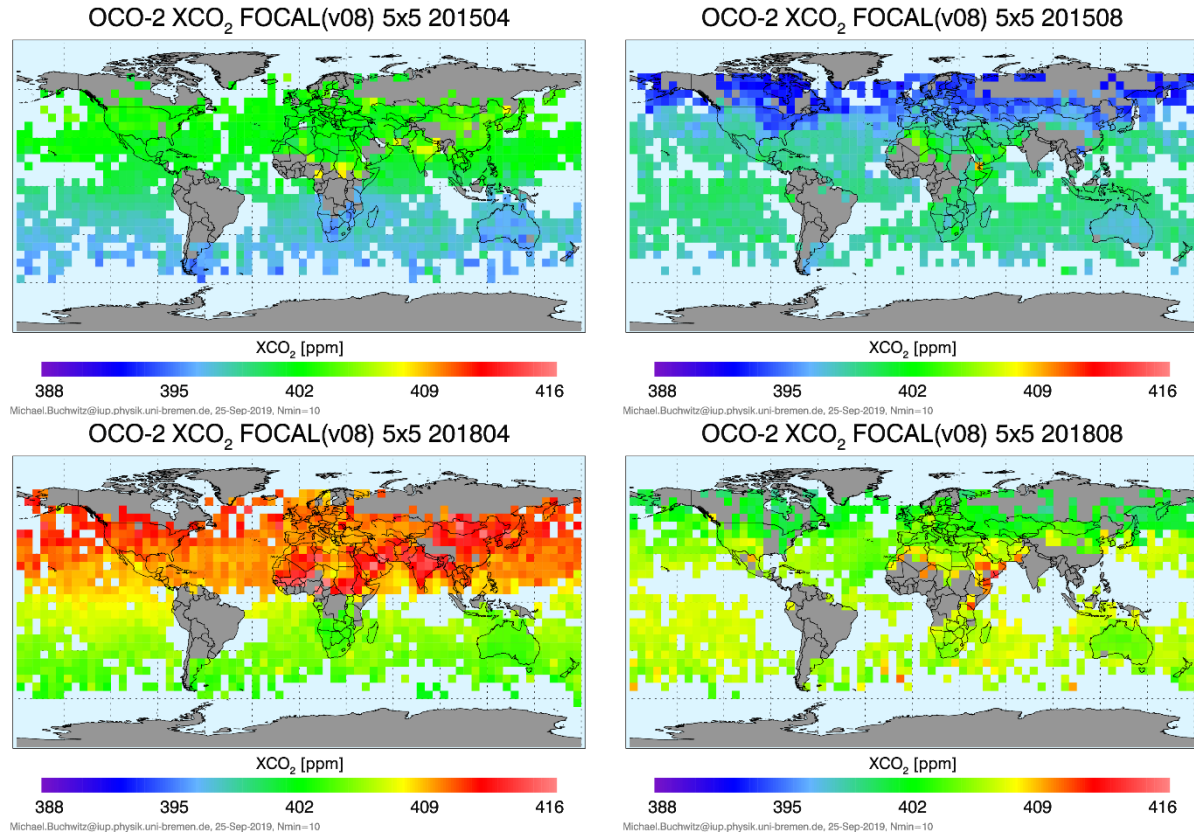
- Algorithm description (ATBD): [http://www.iup.uni-bremen.de/~mreuter/atbd\\_co2\\_oc2\\_foca\\_v1.pdf](http://www.iup.uni-bremen.de/~mreuter/atbd_co2_oc2_foca_v1.pdf)
- Product User Guide (PUG): [http://www.iup.uni-bremen.de/~mreuter/PUGv2\\_GHG-CCI\\_CO2\\_OC2\\_FOCA\\_v08.pdf](http://www.iup.uni-bremen.de/~mreuter/PUGv2_GHG-CCI_CO2_OC2_FOCA_v08.pdf)
- Validation and error analysis: [http://www.iup.uni-bremen.de/~mreuter/E3UBv1\\_GHG-CCI\\_CO2\\_OC2\\_FOCA\\_v08.pdf](http://www.iup.uni-bremen.de/~mreuter/E3UBv1_GHG-CCI_CO2_OC2_FOCA_v08.pdf)

Therefore, we only provide a short overview in this document.

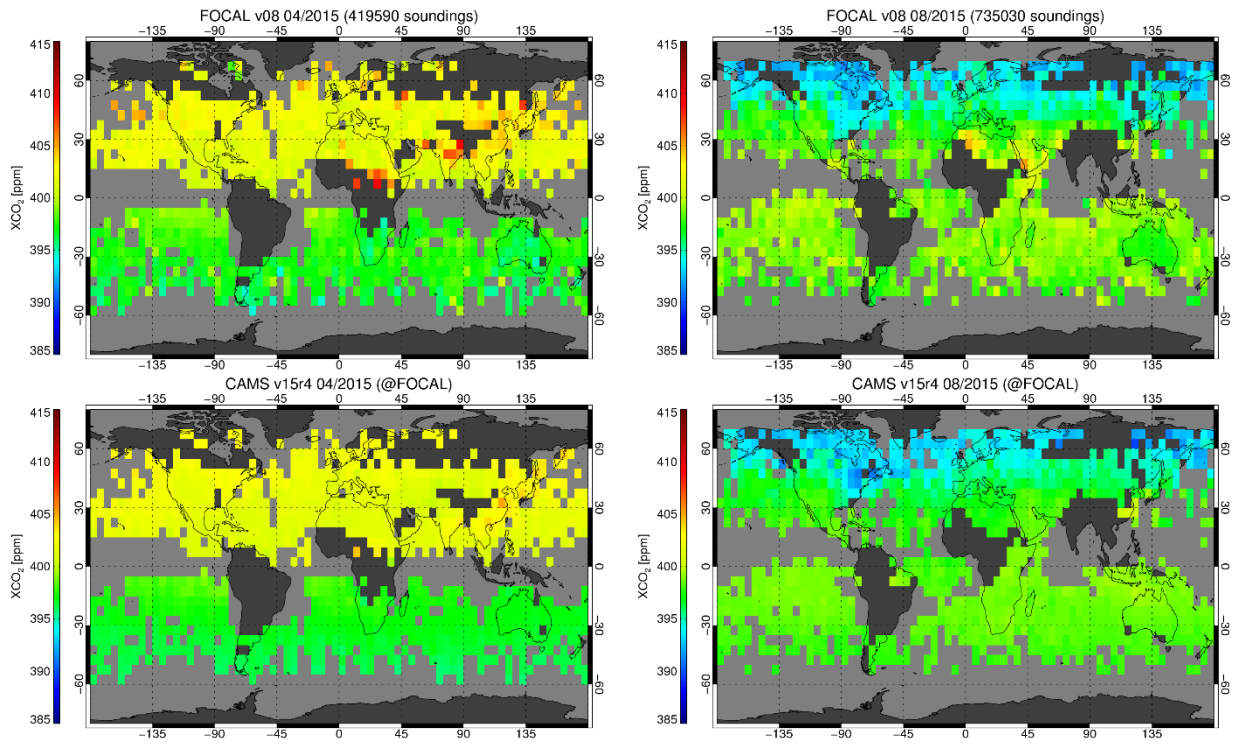
Figure 14 shows how the FOCAL OCO-2 XCO<sub>2</sub> v08 data product “looks like”. Shown are monthly averages at 5°x5° resolution for April and August 2015 and 2018. Clearly visible is the overall increase with time (about 2-3 ppm/year, depending on year), the north-south gradients and seasonal variations with typically high values in April over the northern hemisphere and lower values in August primarily due to uptake of atmospheric CO<sub>2</sub> during the terrestrial biosphere growing season.

A comparison with CAMS XCO<sub>2</sub> is shown in Figure 15 for April and August 2015. Note that the CAMS model data set has been sampled as the satellite observations before gridding to minimize sampling effects.

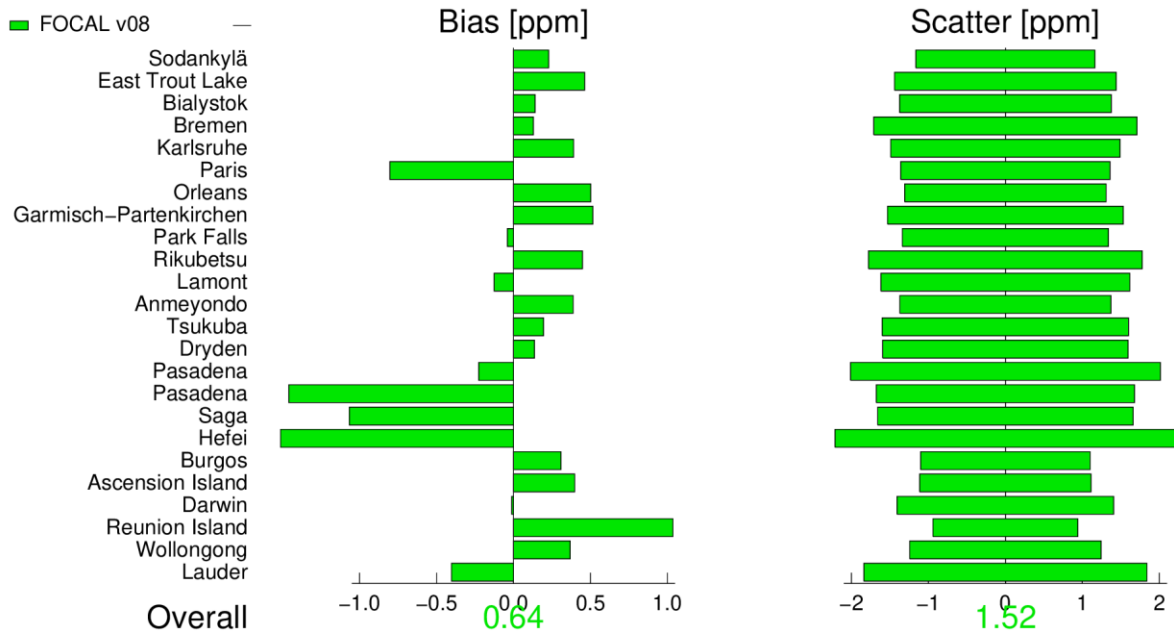
An overview about the validation results using TCCON is shown in Figure 16. As can be seen, the bias (mean difference) and the scatter (standard deviation of the difference) depends on TCCON sites. The standard deviation of biases, which is a measure of spatial biases, is 0.64 ppm and the mean value of the scatter, which is a measure of the single observation precision or random errors, is 1.52 ppm. Note that this includes representation errors and assumes error free TCCON retrievals, i.e., neglects the TCCON uncertainty of approx. 0.4 pm (1-sigma).



**Figure 13:** Monthly maps of the FOCALv08 OCO-2 XCO<sub>2</sub> data product. Top: April (left) and August (right) 2015. Bottom: April (left) and August (right) 2018. To generate these maps, the Level 2 (individual soundings) data product has been gridded (averaged) to monthly time and 5x5o spatial resolution.



**Figure 14:** Monthly mean XCO<sub>2</sub> at 5° x 5°. Top: FOCAL v08. Bottom: CAMSv15r4 sampled as FOCAL. Left: April 2015. Right: August 2015.



**Figure 15:** Validation statistics bias (left) and scatter (right) per TCCON site with more than 250 co-locations for FOCALv08 (bias corrected). The summarizing values (“overall”) represent the standard deviation of the site biases and the average scatter relative to TCCON. The sites are ordered from north (top) to south (bottom).

Because FOCAL data products for 2019 and later are still in preparation, we cannot demonstrate a direct use of the COCCON observations for validation of the FOCAL XCO<sub>2</sub> data product in this document (as the COCCON campaign was conducted in 2019). In order to substantiate the assumption that the FOCAL data are reliable and suitable for further use by model / inventory studies, we have included a comparison with the TCCON.

## 8. Conclusions

For the first time, St. Petersburg CO<sub>2</sub> city emissions were quantified in collaboration with Russian partners using a pair of portable FTIR spectrometers of the COCCON network. Subsequent to the city campaign, one spectrometer was moved to Yekaterinburg and the measurements continued for the detection of XCO<sub>2</sub> gradients on synoptic scales across western Russia. By accomplishing both tasks, highly accurate and precise measurements of column-averaged greenhouse gas abundances have been demonstrated in regions still lacking infrastructure for the quantification of GHG emissions. This has been made possible by use of a novel type of FTIR spectrometer, which is sufficiently stable to maintain its instrumental characteristics even after transport events and in harsh environmental conditions and by merging these favorable instrumental features with the stringent COCCON requirements of pre-deployment performance and calibration checks and use of a standardized processing scheme. The successful completion of the Russian campaign tasks therefore suggests that COCCON is a highly useful component of future observational strategies for quantification of greenhouse gas emissions.

The FOCAL retrieval algorithm has been improved and used to generate global XCO<sub>2</sub> data sets from OCO-2 for VERIFY. The latest version is FOCAL v08 and has been used to generate a 4-year data set covering the period 2015-2018. Using TCCON ground-based XCO<sub>2</sub> retrievals it has been estimated that the single observation random error is approximately 1.5 ppm and the spatial bias or relative accuracy is about 0.64 ppm. These estimates have been derived assuming an error free comparison method and error free TCCON data, i.e., the approximately 0.4 ppm uncertainty of the TCCON data has been neglected. The further development of FOCAL and its use to generate global multi-year XCO<sub>2</sub> data sets from OCO-2 is a major activity and significantly benefits from co-funding from other projects (CHE, ESA GHG-CCI+). FOCAL has been selected by EUMETSAT to be one of three candidate algorithms to retrieve XCO<sub>2</sub> from CO2M. The FOCAL development and corresponding data set generation (from OCO-2, GOSAT, GOSAT-2 and in the future from CO2M) will continue in 2021 and even after the end of the VERIFY project.

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## 10. Appendix A: measurement days and locations of COCCON campaign

Place Identifier	Latitude	Longitude	Instrument-SN	Date	start time	end time	# of spectra
St.Petersburg_EMME	59.88	29.83	FTS#80	21.03.19	14:07:31	15:07:35	39
	59.85	30.54		27.03.19	11:42:36	14:57:33	111
	59.85	30.54		01.04.19	11:14:16	14:31:30	86
	60.04	30.47		03.04.19	11:58:07	14:22:10	133
	60.04	30.47		04.04.19	11:49:50	14:16:31	137
	60.01	29.69		06.04.19	12:16:09	15:29:35	176
	59.86	30.11		16.04.19	11:21:53	15:00:16	200
	60.04	30.47		18.04.19	12:07:18	14:56:30	155
	59.85	30.54		24.04.19	11:52:34	15:23:11	182
	60.04	30.47		25.04.19	12:08:03	14:49:59	145
	59.85	30.54		30.04.19	12:35:54	13:31:54	53
	59.95	30.59	FTS#84	21.03.19	13:08:30	15:36:53	128
	60.01	29.69		27.03.19	11:54:42	15:09:04	28
	60.01	29.69		01.04.19	11:02:14	13:37:19	115
	59.88	29.83		03.04.19	14:47:40	16:05:00	72
	59.81	30.09		04.04.19	11:06:49	15:01:56	225
	59.95	30.59		06.04.19	12:14:54	15:25:10	168
	60.01	29.69		16.04.19	11:14:24	15:08:40	224
	59.72	30.23		18.04.19	11:38:23	15:24:41	168
	60.01	29.69		24.04.19	11:37:27	14:56:12	190
	59.81	30.09		25.04.19	11:19:49	15:08:35	201
	60.01	29.69		30.04.19	12:22:24	13:46:45	82
Peterhof (SPbU)	59.88	29.83	FTS#80	21.01.19	10:54:51	12:16:36	71
				22.01.19	10:03:00	11:39:13	42
				23.01.19	10:46:50	11:57:04	67
				24.01.19	10:52:51	12:17:34	67
				31.01.19	10:22:33	12:29:10	123
				15.02.19	11:42:19	12:24:35	42
				21.02.19	10:13:17	12:40:59	135



				22.02.19	09:15:46	12:58:07	152
				01.03.19	09:51:07	13:38:21	179
				04.03.19	09:31:54	12:06:47	117
				06.03.19	09:39:16	13:34:35	204
				03.04.19	11:47:40	13:05:00	72
				17.04.19	09:16:04	12:11:13	123
				22.04.19	09:19:54	09:51:14	25
				23.04.19	09:07:36	12:35:02	130
				06.05.19	12:27:32	12:38:58	12
				13.05.19	11:35:07	12:33:51	56
				15.05.19	10:41:12	12:59:14	107
				16.05.19	10:07:26	12:59:57	120
				17.05.19	11:24:15	12:53:59	78
				22.05.19	10:20:47	12:51:00	102
				30.05.19	08:47:36	13:16:38	175
				03.06.19	09:18:50	13:18:45	218
				04.06.19	08:23:45	13:19:19	257
				05.06.19	08:27:35	13:19:33	137
				06.06.19	08:28:49	13:19:32	238
				07.06.19	08:47:36	12:06:49	107
				10.06.19	10:24:44	13:21:19	165
				11.06.19	08:12:50	10:50:21	122
				18.06.19	08:16:32	13:34:58	292
				24.06.19	08:11:55	13:22:38	243
				25.06.19	13:08:39	13:24:05	16
				26.06.19	13:02:09	13:14:32	9
				28.06.19	09:32:30	12:13:25	68
				01.07.19	10:11:54	12:37:16	32
				10.07.19	11:57:12	13:24:55	81
				11.07.19	11:37:14	12:02:59	20
				22.07.19	11:25:22	13:22:02	113
				29.07.19	09:21:04	09:47:54	23
				05.08.19	10:22:15	10:47:10	15
				06.08.19	08:18:00	13:14:06	272
				20.08.19	08:24:53	10:25:43	98
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				02.09.19	08:10:10	12:04:52	45
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				24.09.19	10:14:00	12:51:52	91
				01.10.19	11:53:16	13:10:41	56
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				08.10.19	07:21:51	09:23:43	91
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				22.11.19	08:58:17	12:00:25	90
				23.11.19	08:59:11	09:52:55	53
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