FRM4GHG
Fiducial Reference Measurements for Greenhouse Gases

Deliverable D24 for CCN2
FRM4GHG Outcomes and Conclusions for XCO_2, XCH_4, XCO and HCHO measurements from low-resolution instruments

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1 DOCUMENT CHANGE RECORD

Issue	Date	Item	Comment
V0.0	2020-06-16	-	Initial version
V1.0	2020-07-16	-	Added comments from all partners
V1.1	2020-07-24	-	Implemented comments from ESA
V1.2	2020-08-06	-	Implemented comments from all partners

2 ACCESS LIST

This document is a deliverable "D24: FRM4GHG Outcomes and Conclusions for XCO_2, XCH_4, XCO and HCHO measurements from low-resolution instruments" created for the project FRM4GHG CCN2 and will be submitted to ESA. The document can be downloaded from the project webpage http://frm4ghg.aeronomie.be.

The measurement data obtained during the project are also available from the project Webpage, as well as from EVDC

3 PURPOSE

The aim of our project was to perform an intercomparison of measured total column concentrations of CO_2, CH_4 and CO (referred to hereafter as GHG) using several different spectroscopic instruments at collocated TCCON sites. Measurements were performed for the first two years at Sodankylä/Finland and for the IRcube for the third year in Wollongong and Darwin in Australia.

The measurements were supported by an in-situ AirCore balloon system which measures the vertical concentration profile of the trace gases, and which can be calibrated relative to the WMO standards.

We brought four different spectrometers, working with different resolution and different techniques (Michelson interferometer, and a laser heterodyne spectro-radiometer). The AirCore system and data analysis, sampling the atmospheric concentration profile up to 30 km, was supported by the Dutch (University of Groningen) subcontractor. The intercomparison between all instruments was done in a first step in a quasi-blind approach, i.e. each partner carries out the measurements and retrieval independently for his instrument; then another partner carries out the intercomparison between the results. Following this first exercise, AirCore data were included as a-priori information in the retrieval process. The whole retrieval was repeated to study the impact of the vertical concentration profile on the results and intercomparison. Observed differences between the data were discussed on a regular basis with the partners and the measurement specifics were modified if necessary. Measurements and analysis was then continued to get more statistically significant results, and study the seasonal impact (e.g. by humidity and solar zenith angle) on the results. Formaldehyde (HCHO) was measured by the Vertex70 with a liquid nitrogen cooled InSb detector and compared to HCHO measured at both high and low spectral resolution with the TCCON Bruker IFS 125HR instrument.

Numerous stakeholders could benefit from our campaign. On one side, the satellite community needs to know which instruments are suitable for future validation campaigns. Since the global coverage of TCCON is very limited, more validation sites in areas with limited and difficult access are required. This includes for example sites with high albedo in the Himalaya or in desert regions of the Earth, where the large TCCON instruments are difficult to be installed. Here small, low-resolution instruments will be the only possibility to perform validation campaigns. These instruments could serve as a possible extension and complement of the TCCON network. Satellites of interest are: GOSAT-1 and -2, OCO-2 and -3, all future Copernicus Sentinel instruments measuring GHG, as well as the Chinese Tansat mission, and possibly the Merlin and Microcarb missions. Furthermore, we used the acquired data within the S5P validation efforts through the AOs in which the team is involved. Furthermore, the results of this project are of importance for the modelling community. Inversion models, to retrieve the sources and sinks on a regional or global scale, require observational data with a good global coverage, precision and consistency as input for the models. The demonstrated consistency between the GHG measurements from the low-resolution spectrometers and the standard TCCON instruments proves that these low-resolution spectrometers are suitable for complementing the TCCON network in a more cost-effective and flexible way.

The exchange and cooperation with existing networks and initiatives as well as the feedback from the satellite teams constitute an important objective to ensure that the outcome is also taken into account by all stakeholders. This implies continuous interfacing with TCCON and NDACC, specifically the NDACC Infrared Working Group (IRWG), and with the GHG modelling communities. All project activities followed the overarching principles of QA4EO.

This document provides the outcome and conclusions of the measurement campaigns performed with the low-resolution instruments during the years 2017 – 2019.

4 DOCUMENT STRUCTURE

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5 RESULTS

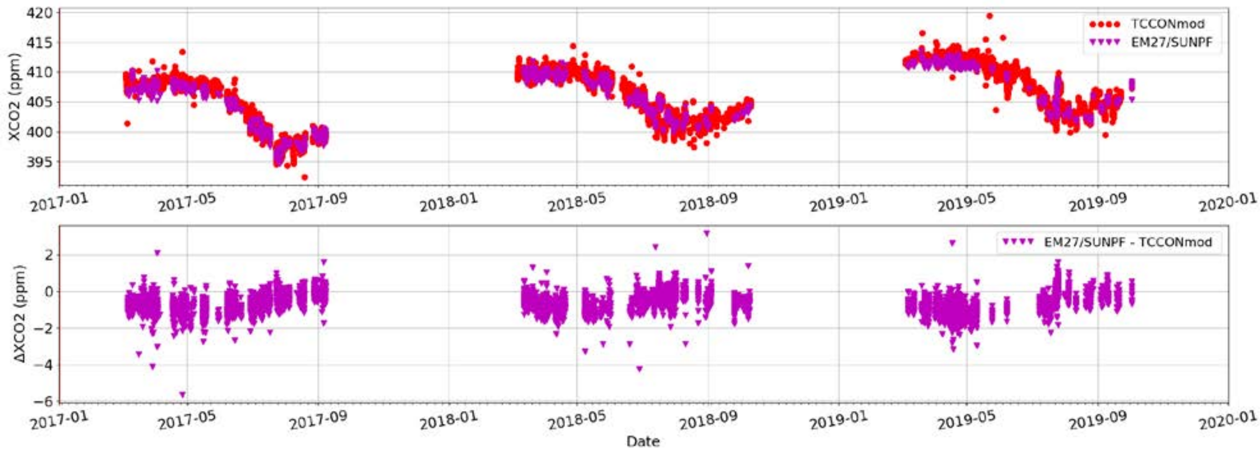
This section summarises briefly the results of the three years of the FRM4GHG campaign performed during 2017 – 2019. An extensive discussion of the results is available in Sha et al., AMTD, 2019.

5.1 EM27/SUN

Measurements during 2017 – 2019 were performed at Sodankylä site.
A new shelter was installed on 20 September 2018 to the EM27/SUN. Since then the EM27/SUN was left outside even when no measurements were performed (e.g., night time). The EM27/SUN was packed in the container during the winter of 2018/19.

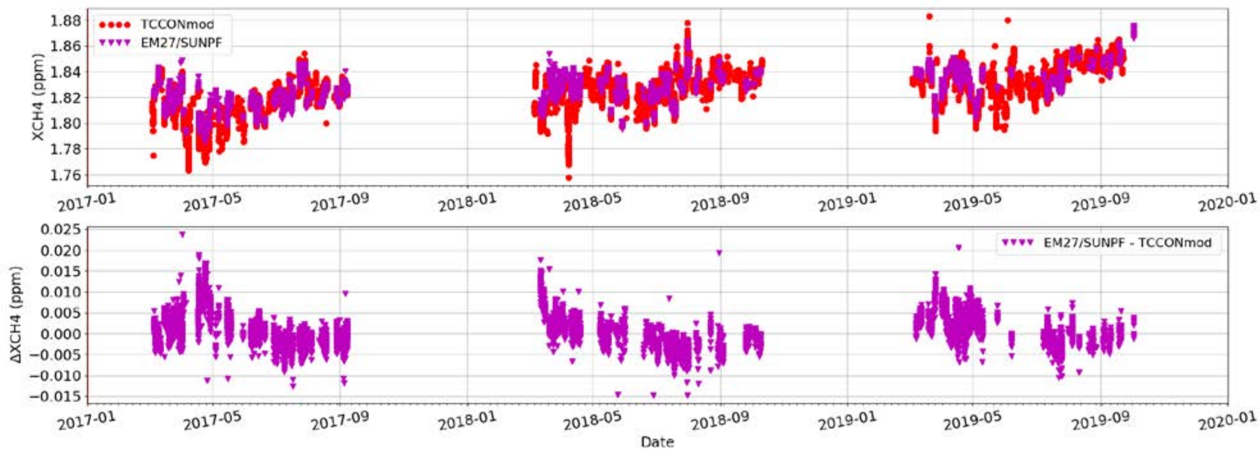
XCO2:

Seasonality seen in the bias (~ 1 ppm) ➔ significant ~ the accuracy requirement (0.25%) of TCCON.
The seasonality of the bias for other sites will depend on the variability of the profile shape during the year and their difference to the true profile.



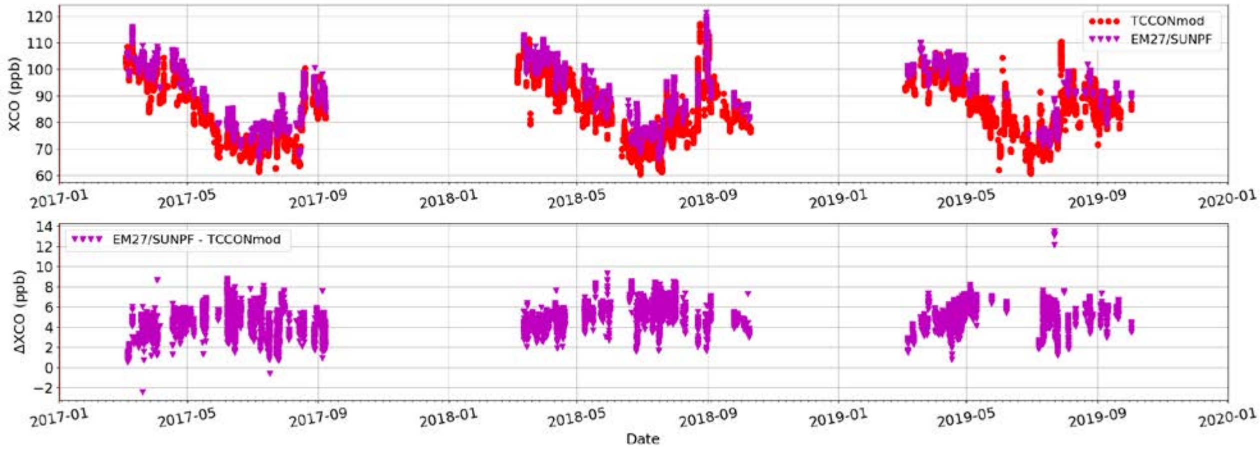
XCH4:

Seasonality seen in the bias. About 10 ppb during spring polar vortex conditions – not so usual for standard TCCON site, ~ 3 – 5 ppb bias during the summer – Autumn period ➔ significant ~ the accuracy requirement (0.2%) of TCCON.



XCO:

Seasonality seen in the bias. About 2 – 4 ppb bias ➔ significant ~ the accuracy requirement (2%) of TCCON.



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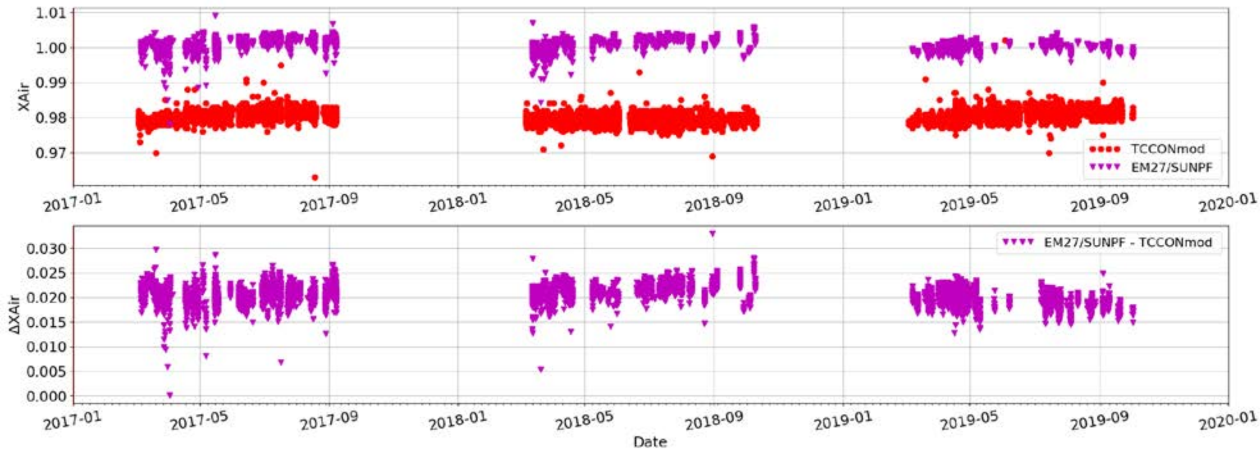
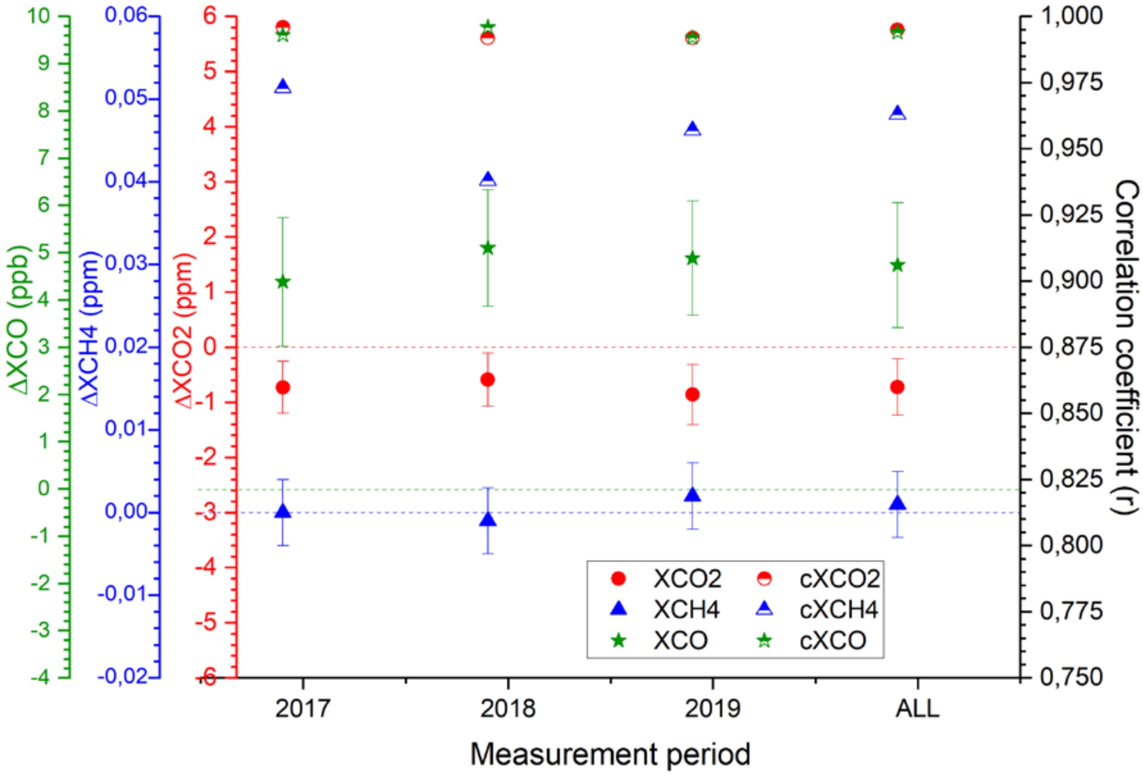


Statistics:

Species	Duration	XCO2 / ppm	XCH4 / ppm	XCO / ppb
Bias (mean standard ± deviation) and correlation coefficient (r)				
EM27/SUN vs TCCON (Sodankylä)	2017	-0.727±0.474 (0.996)	0.000±0.004 (0.973)	4.384±1.361 (0.993)
EM27/SUN vs TCCON (Sodankylä)	2018	-0.587±0.485 (0.992)	-0.001±0.004 (0.938)	5.101±1.234 (0.996)
EM27/SUN vs TCCON (Sodankylä)	2019	-0.859±0.548 (0.992)	0.002±0.004 (0.957)	4.886±1.210 (0.992)
EM27/SUN vs TCCON (Sodankylä)	2017 – 2019	-0.722±0.510 (0.995)	0.001±0.004 (0.963)	4.738±1.321 (0.994)



Overall, stability looks very good even over several years.

The bias values are very close to each other and the small differences seen from year-to-year is due to the data representative issue.

Annual cycle in comparison to TCCON is likely due to difference map – a prior vs true atmospheric state which generates differences in Xgas because sensitivities of TCCON and COCCON differ.

5.2 VERTEX70

Measurements during 2017 – 2019 were performed at Sodankylä site.
Vertical lines indicate instrument modifications.

XCO2:

Seasonality seen in the bias for 2019 (~ 1 ppm) ➔ significant ~ the accuracy requirement (0.25%) of TCCON.
The seasonality of the bias for other sites will depend on the variability of the profile shape during the year and their difference to the true profile.

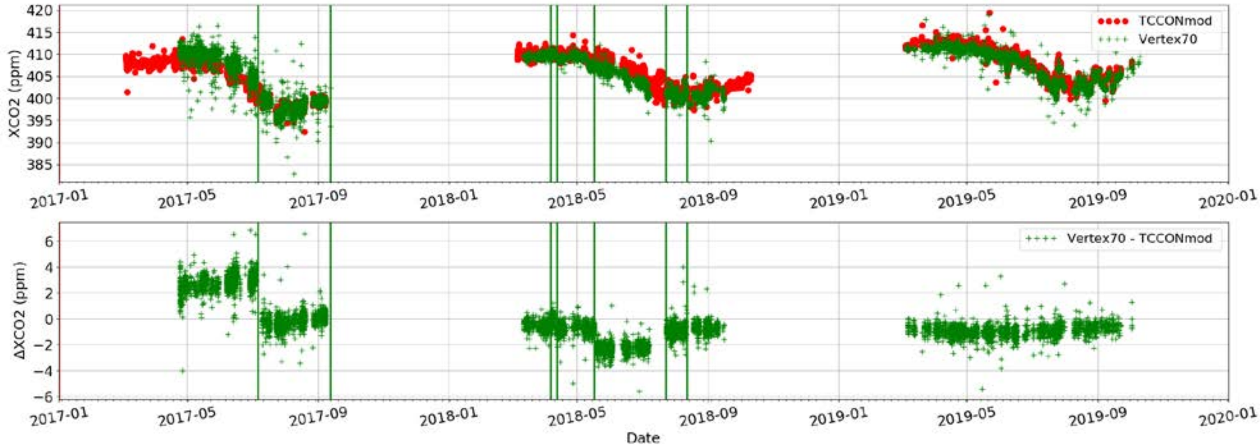


Figure 1: Vertex70 XCO2 for all measurements between 2017 and 2019.

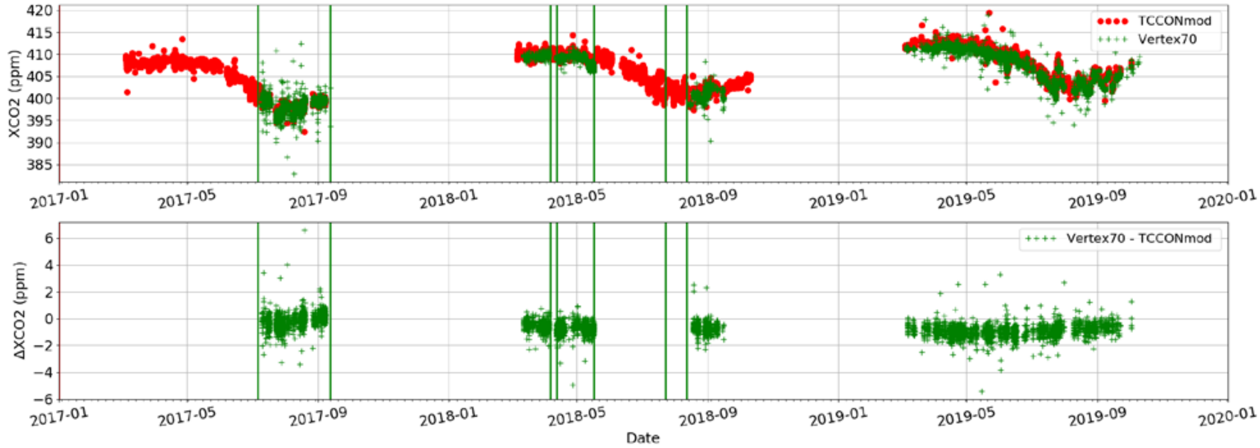
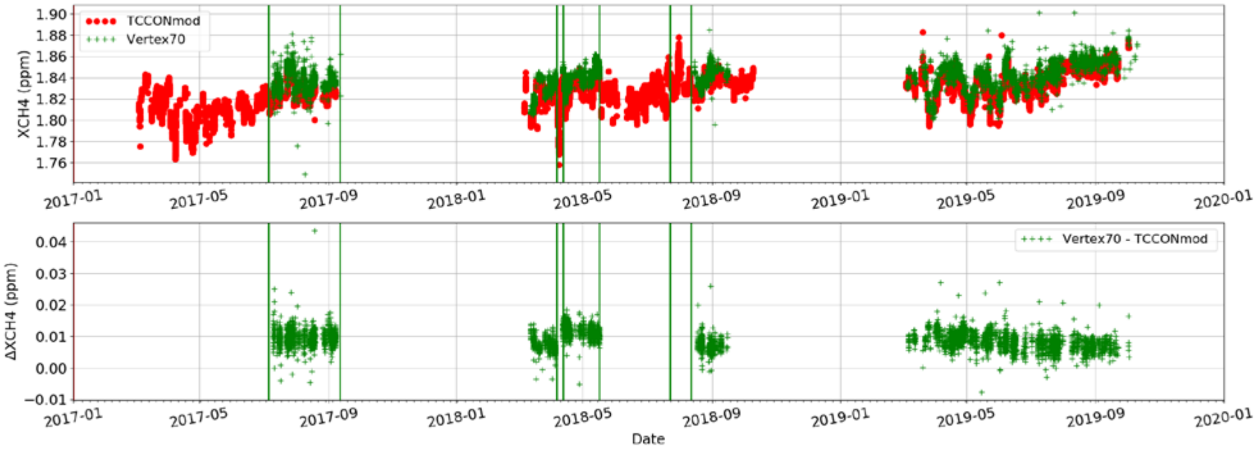


Figure 2: Vertex70 XCO2 for good measurements between 2017 and 2019.

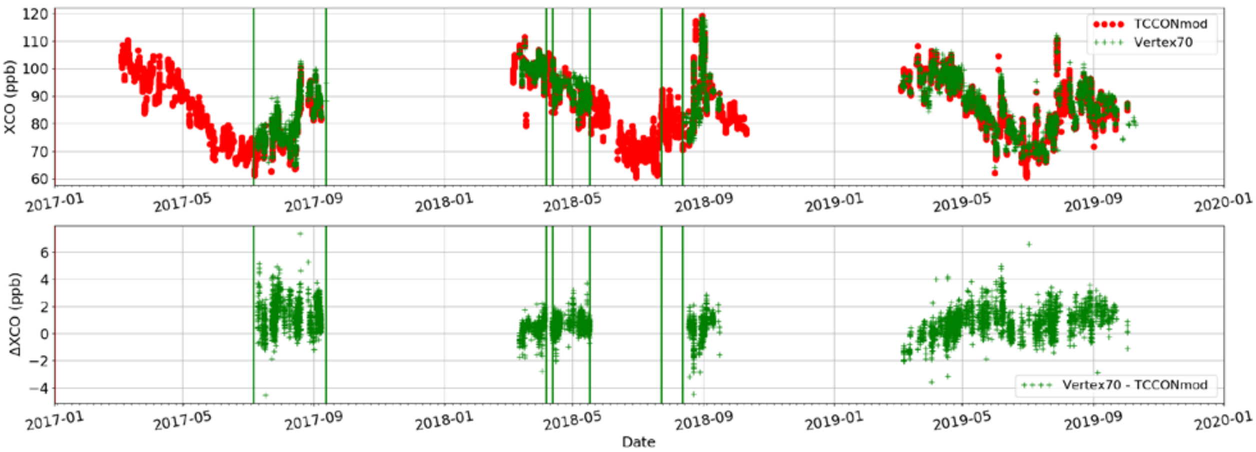
XCH4:

Seasonality in the bias is not so obvious – related to resolution (0.2 cm-1 rather than 0.5 cm-1).



XCO:

Seasonality seen in the bias. About 2 – 4 ppb bias ➔ significant ~ the accuracy requirement (2%) of TCCON.



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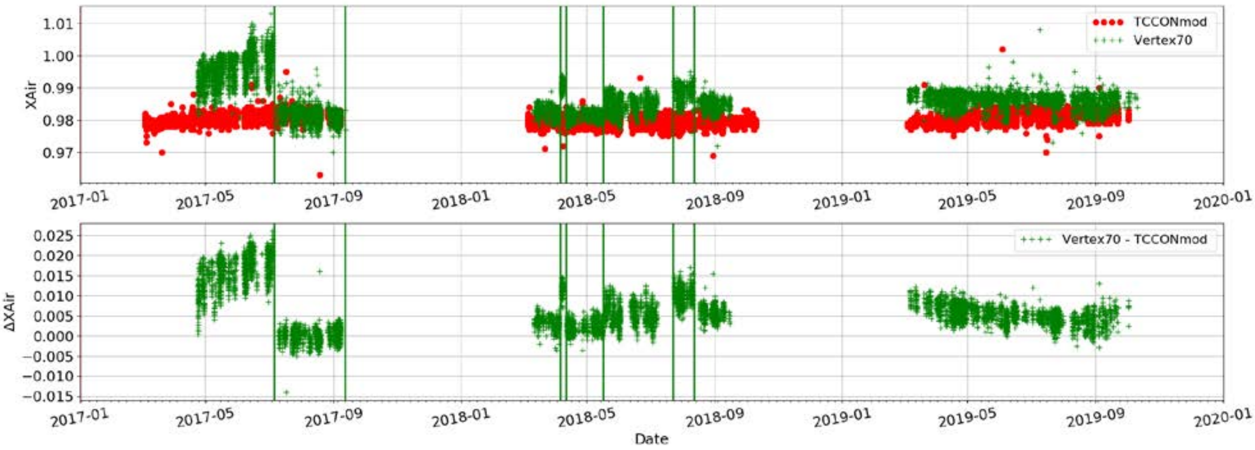
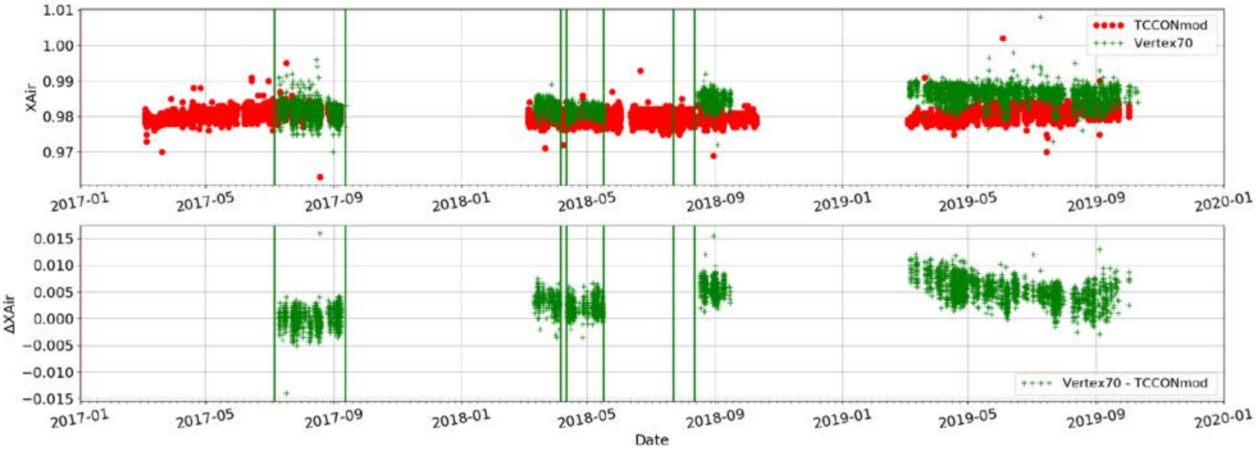
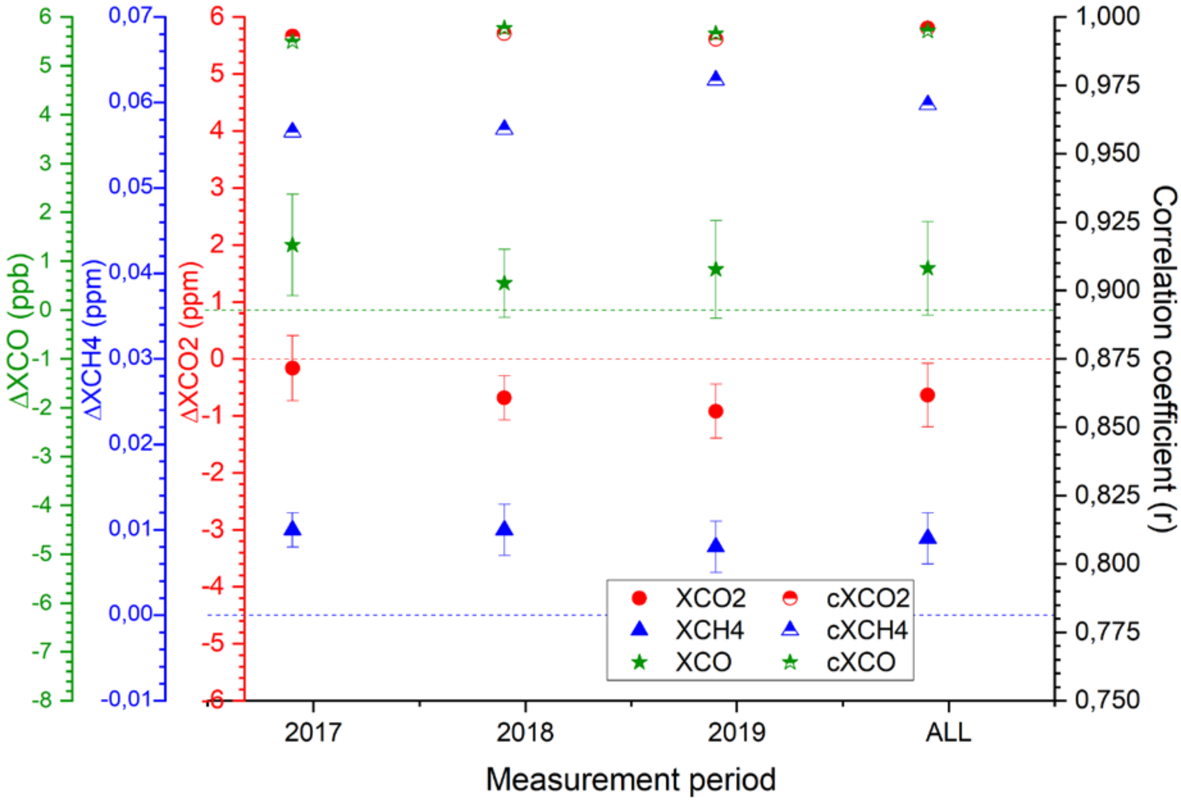




Statistics:

Species	**Duration**	**XCO2 / ppm**	**XCH4 / ppm**	**XCO / ppb**
Bias (mean standard ± deviation) and correlation coefficient (r)				
VERTEX70 vs TCCON (Sodankylä)	2017	-0.160±0.574 (0.933)	0.010±0.002 (0.958)	1.335±1.035 (0.991)
VERTEX70 vs TCCON (Sodankylä)	2018	-0.681±0.389 (0.994)	0.010±0.003 (0.959)	0.548±0.699 (0.996)
VERTEX70 vs TCCON (Sodankylä)	2019	-0.915±0.472 (0.992)	0.008±0.003 (0.977)	0.834±1.002 (0.994)
VERTEX70 vs TCCON (Sodankylä)	2017 – 2019	-0.636±0.558 (0.996)	0.009±0.003 (0.968)	0.856±0.961 (0.995)



The values for 2019 is the most representative for full year since no instrument modification done. The bias values for 2017 and 2018 have data representative issue due to the instrument modifications. However, the bias change from the year-to-year are within the seasonal variability seen in the comparison.

5.3 IRCUBE

IRcube measurements in 2017 and 2018 are performed in Sodankylä.
17 Jan 2019 – 23 Aug 2019 IRcube measurements are performed in Wollongong (WE)
12 Sep 2019 – 31 Dec 2019 IRcube measurements performed in Darwin (DB)

XCO2:

Seasonality seen in the bias for 2017 SO (~ 1 ppm) ➔ significant ~ the accuracy requirement (0.25%) of TCCON.
The seasonality of the bias is not seen for 2019 WE & DB for other sites as the variability of the profile shape during the year is closer to the true profile.

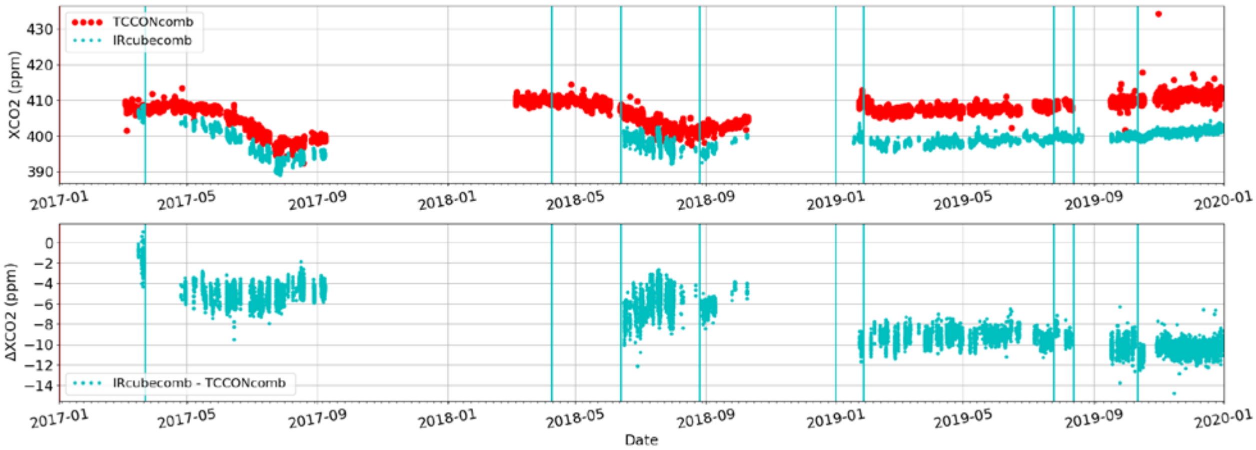


Figure 3: IRCUBE XCO2 for all measurements between 2017 and 2019.

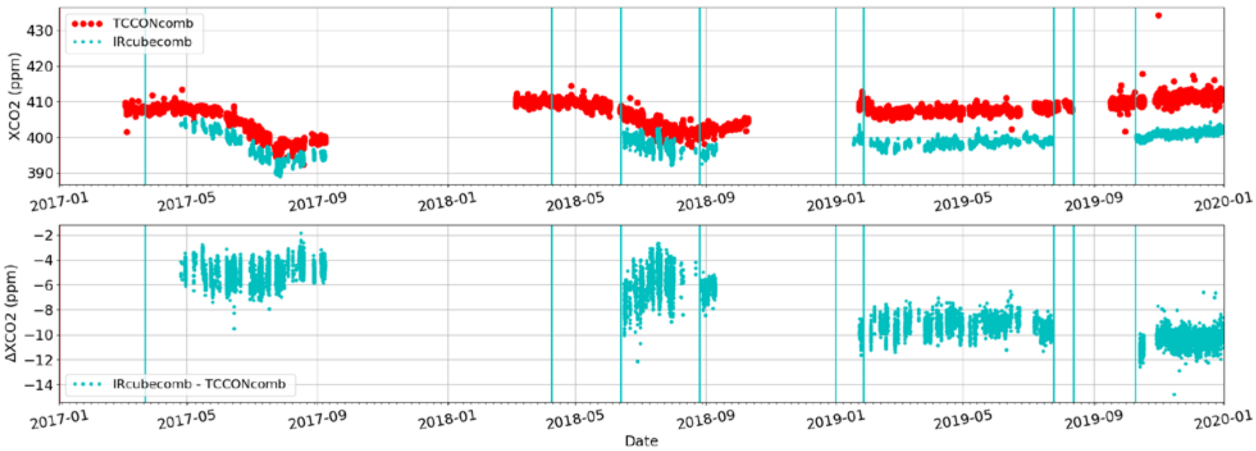
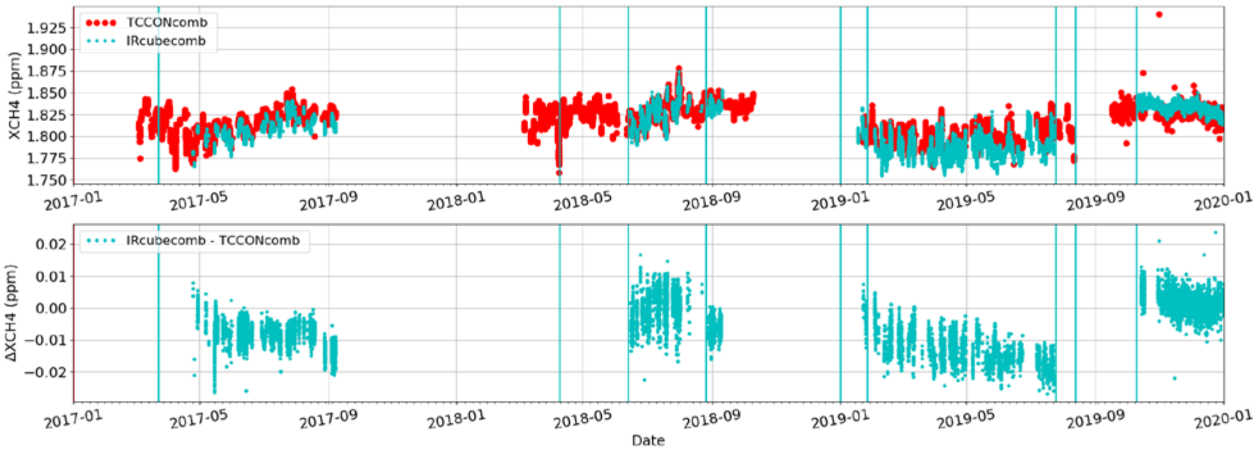


Figure 4: IRCUBE XCO2 for good measurements between 2017 and 2019.

XCH4:

Decreasing bias as a function of time needs further investigation.



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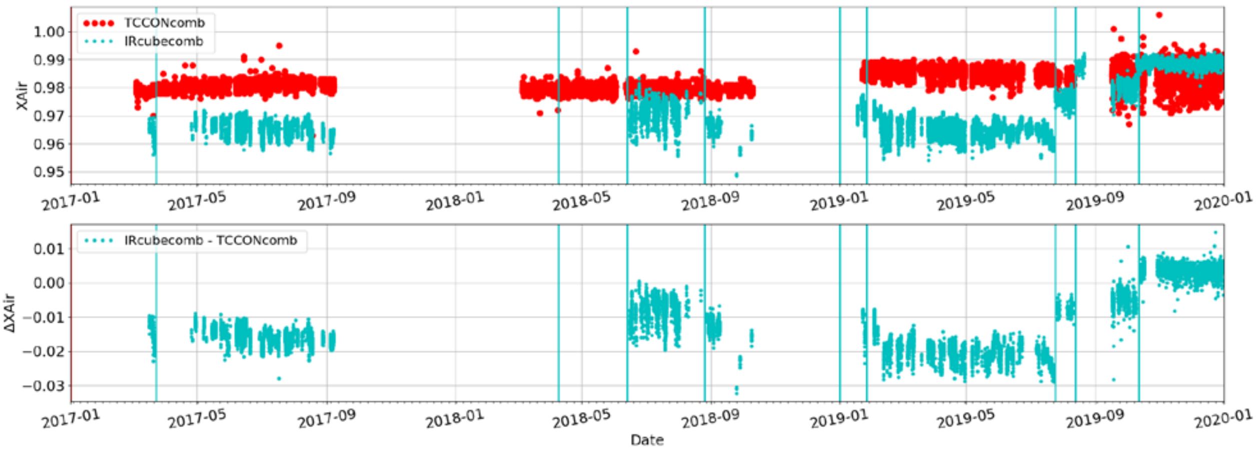
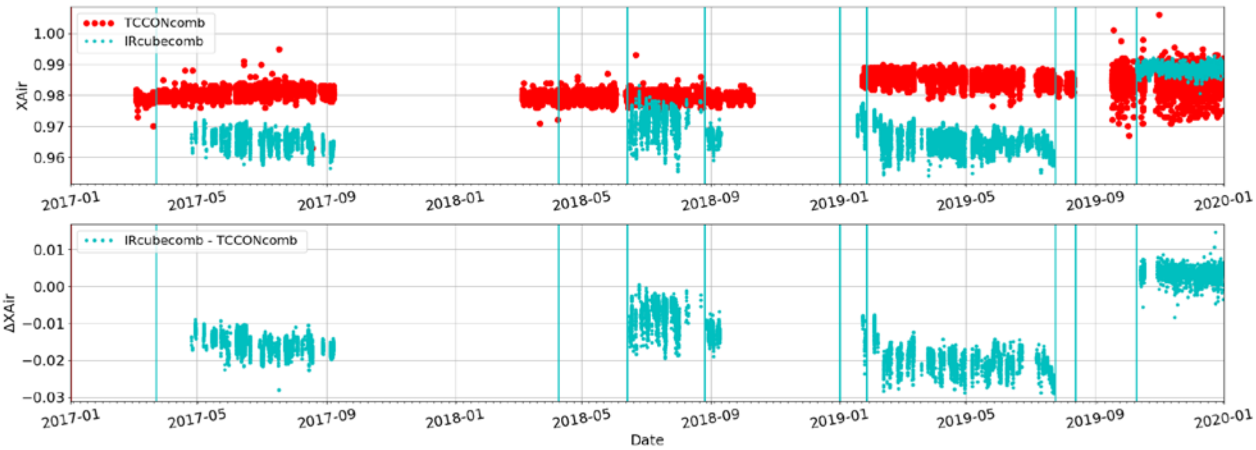
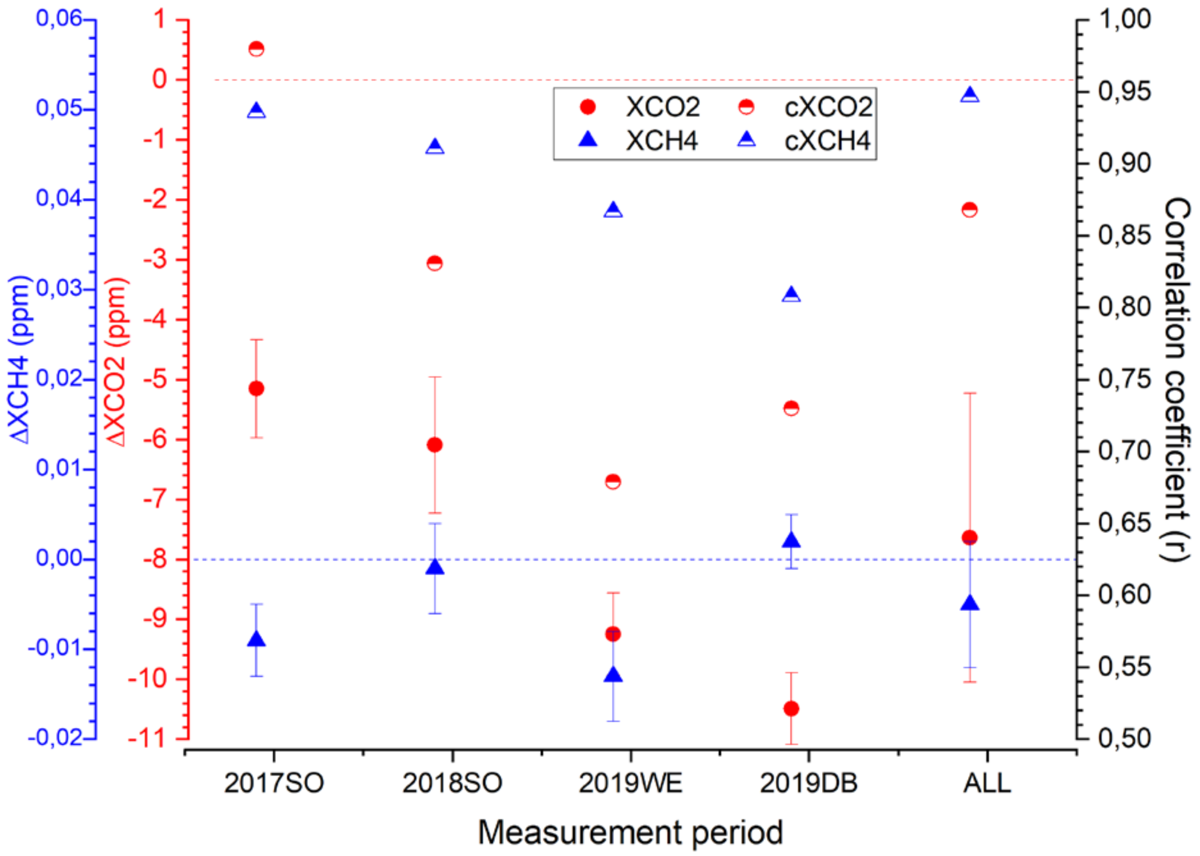




Statistics:

Species	**Duration**	**XCO2 / ppm**	**XCH4 / ppm**
Bias (mean standard ± deviation) and correlation coefficient (r)			
IRcube vs TCCON (Sodankylä)	2017	-5.15±0.819 (0.98)	-0.009±0.004 (0.936)
IRcube vs TCCON (Sodankylä)	2018	-6.09±1.133 (0.831)	-0.001±0.005 (0.911)
IRcube vs TCCON (Wollongong)	2019	-9.243±0.683 (0.679)	-0.013±0.005 (0.867)
IRcube vs TCCON (Darwin)	2019	-10.487±0.597 (0.73)	0.002±0.003 (0.808)
IRcube vs TCCON (all)	2017 – 2019	-7.634±2.409 (0.868)	-0.005±0.007 (0.947)



The bias values are strongly dependent on the instrument modifications.
The precision is best for Darwin 2019.

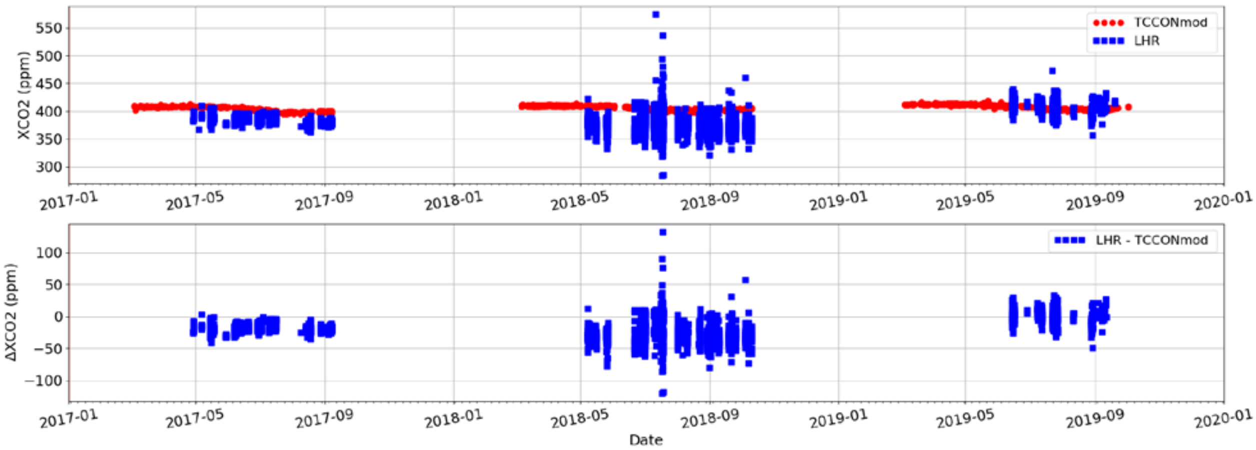
5.4 LHR

In 2017, an existing bench-top laboratory demonstrator LHR was quickly completely re-engineered into a prototype transportable instrument to allow field deployment and remote operation as required for the FRM4GHG project. This was the first deployment of its kind and the instrument was configured to measure CO_2 and H_2O. The results from 2017 showed that the signal to noise ratio (SNR) and the precision was within expectation. However, large diurnal biases with no obvious correlation were found to severely impede the accuracy.

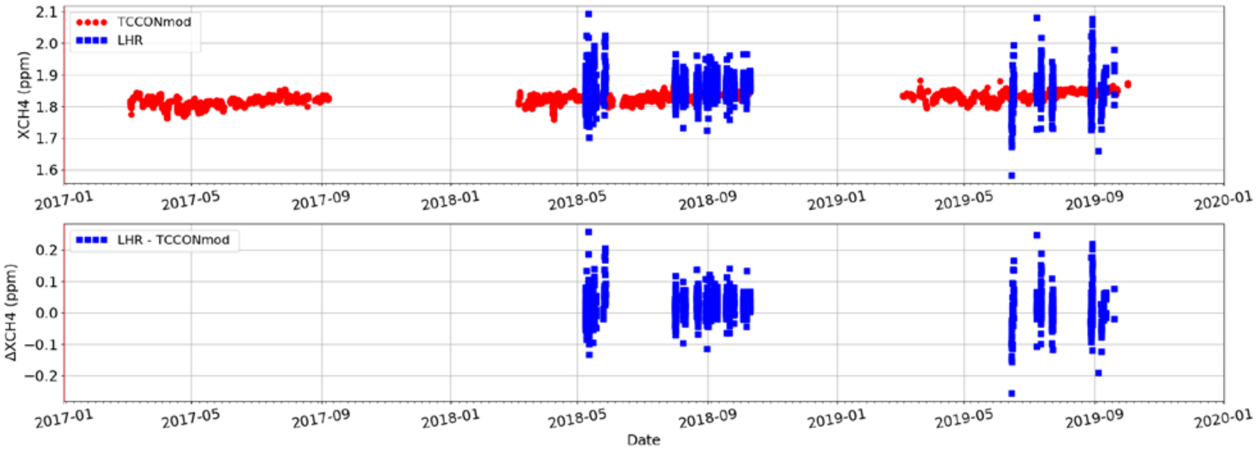
In 2018, the instrument was returned to RAL for modifications. A new channel for CH_4 measurements was added, and consequently the input LWIR filter was widened to capture CH_4 lines at 7.8 µm and CO_2 lines at 9.7 µm. The instrument hence configured was returned to Sodankylä for the 2018 campaign. Analysis of the data obtained revealed that the SNR was severely decreased compared to 2017, and drifts were more significant. Severe misalignments were diagnosed, and a team was send to Sodankylä to address these.

In 2019, with the system realigned and after the installation of a new detector, the SNR was back to its expected values. Analysis of the dataset showed that ~70% of the data were affected by a field of view (FoV) issue (the LHR shared the Vertex70 Sun tracker) producing high level of excess noise in the signals. In 2019, a strong focus was placed on the physical understanding of the instrument biases. Unwanted thermal modulation of the detector and signal correlation to the instrument temperature were understood and corrective measures implemented into the analysis algorithm. As a results, the data were shown to be far closer on average to the TCCON reference. However, the scatter due to measurement noise was very high owing to the FoV issue previously mentioned.

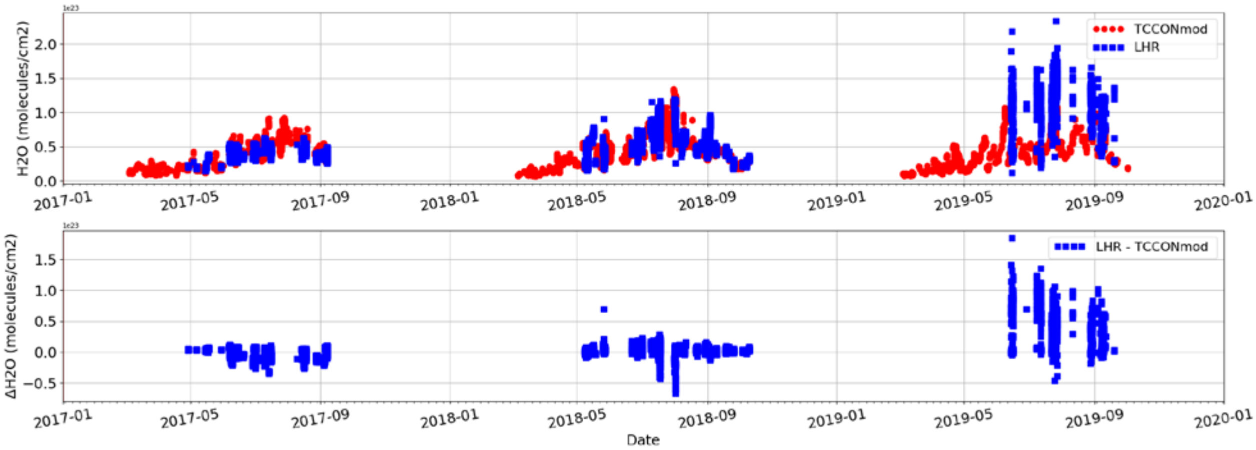
XCO2:



XCH4:

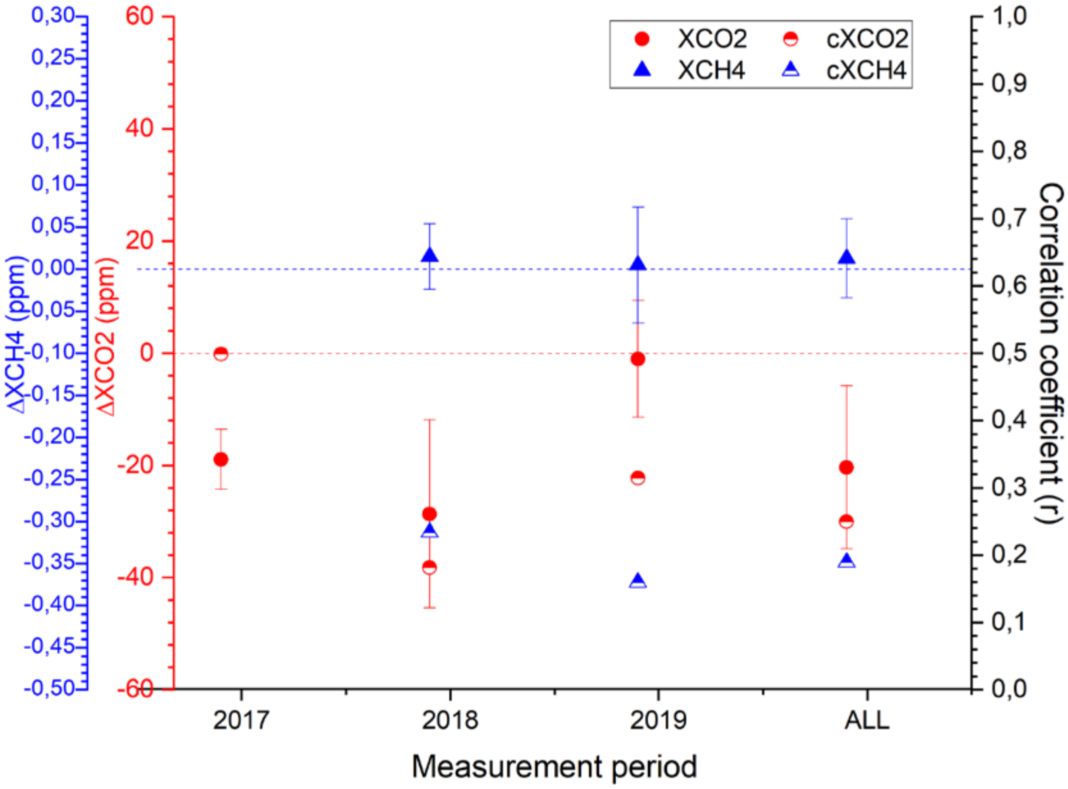


H2O:



Statistics:

Species	Duration	XCO2 / ppm	XCH4 / ppm
Bias (mean standard ± deviation) and correlation coefficient (r)			
LHR vs TCCON (Sodankylä)	2017	-18.891±5.342 (0.499)	-
LHR vs TCCON (Sodankylä)	2018	-28.616±16.752 (0.182)	0.015±0.039 (0.234)
LHR vs TCCON (Sodankylä)	2019	-0.979±10.424 (0.315)	0.005±0.069 (0.160)
LHR vs TCCON (Sodankylä)	2017 – 2019	-20.304±14.518 (0.250)	0.013±0.047 (0.190)



The bias and the sd of the difference are about a factor of 10 greater than the EM27/SUN's. The bias values vary from year to year.

6 CONCLUSION AND OUTLOOK

Except for small interruptions, all instruments were running throughout the measurement periods, which means between March and September when the sun is above the horizon at a sufficiently high solar elevation angle. Our studies not only yielded a detailed comparison of the different instruments, but intermediate comparisons allowed to improve the instrumental set-ups.

The test instruments under investigation are three Fourier transform spectrometers (FTS): a Bruker EM27/SUN, a Bruker IRcube and a Bruker Vertex70; as well as a Laser Heterodyne spectro-Radiometer (LHR) developed by the UK Rutherford Appleton Laboratory. All four remote sensing instruments performed measurements simultaneously next to the reference TCCON instrument, a Bruker IFS 125HR. The remote sensing measurements have been complemented by regular AirCore launches performed from the same site.

The comparisons with the HR125LR data set (low-resolution measurements at the resolution of EM27/SUN performed with the IFS 125HR) gave useful analysis of the resolution dependent effects on the target gas retrieval. The intercomparison results show that the LHR data have a large scatter and biases with a strong diurnal variation relative to the TCCON and other FTS instruments. The LHR is a new instrument under development and these biases are being currently investigated and addressed. The campaign helped to characterize and identify the instrumental biases and possibly retrieval biases for the FTS, which are currently under investigation. The EM27/SUN, HR125LR and Vertex70 showed relatively small biases w.r.t the reference TCCON for the three years of measurements. The bias of the IRcube changed between the measurement sites and the cause is under investigation. Further improvements of the instrument are ongoing. The EM27/SUN, the IRcube, the modified Vertex70 and the HR125LR provided stable and precise measurements of the target gases comparable to the TCCON during the campaign. The data from 2018 and 2019 campaigns were used for the validation of Sentinel-5 Precursor methane and carbon monoxide and OCO-2 carbon dioxide products. The precision of the low-resolution instruments being comparable to the TCCON is therefore well suited as fiducial reference measurements for satellite validation. The bias seen between the TCCON and the low-resolution instrument is also observed in the validation results between S5P vs TCCON and S5P vs low-resolution instruments. The instrument dependent calibration factors for the low-resolution instruments to scale the retrieved Xgas values to the WMO in-situ standards by indirectly scaling to the TCCON standards is currently done based on two sites (Karlsruhe and Sodankylä) for the COCCON EM27/SUN. This will be further improved based on the side-by-side measurements performed at several other TCCON sites with the traveling EM27/SUN. The work for the traveling standard has been proposed for the next phase of the FRM4GHG project. Once these measurements are available, the scaling factors determined from the global measurements can be used for the individual instruments which will then also be used for the measurements performed during the FRM4GHG campaign. The first results of the formaldehyde (HCHO) retrieved with the Vertex70 showed promising results in comparison to the high and low-resolution HCHO measurements performed with the Bruker IFS 125HR. In the next step further improvements of the retrieval strategy is needed to improve the quality of the HCHO product from the low-resolution instruments (Vertex70 and HR125LR data). This work is proposed to be done in the follow-up FRM4GHG project. The HCHO retrieved from the Vertex70 measurements with the optimised settings will be of great value for its use as reference data set for satellite validation.

The EM27/SUN, the IRcube and the Vertex70 portable low-resolution FTS instruments are suitable to be used for campaign deployment or long-term measurements from any site and offer the ability to complement the TCCON and expand the global coverage of ground-based reference measurements of the target gases (CO_2, CH_4 and CO).

Our project resulted in important new findings, but indicated also that further new studies are required. This includes the development of a more portable stand-alone version of the Vertex70 spectrometer, possibly with a thermoelectric cooled MCT-detector to cover the mid infrared spectral region for the detection of other trace gases without the need for liquid nitrogen. Improve stability of IRcube alignment by developing a more robust alignment method using fringes rather than by simple maximisation of the signal. Testing a fibre optic feed for the EM27/SUN, which will help in understanding if the biases seen in the IRcube measurements are due to the use of fibre optic feed to bring the solar light to the interferometer. Furthermore, a detailed comparison between the two codes used, GFIT and PROFFAST, is necessary to understand and minimize the residual differences found between the data from the various instruments using two retrieval methods. This implies also a further development of the retrieval codes and spectroscopic data, for example by including the approximative Rosenkranz line-mixing and speed dependent Voigt schemes in PROFFAST or an improved spectral data set of H_2O. The further development of GHG profiling (profile retrieval from NIR spectra) is also important to improve the consistency between NDACC and TCCON for CO, N_2O, CH_4, CO_2. Further investigation of the use of different pT-input files, from ECMWF or NCEP by GFIT and PROFFAST, is expected to enable a reduction of the differences between both codes. The demonstration of an EM27/SUN travel standard visiting sites operated by European TCCON groups would allow a direct calibration bridge between the different TCCON sites. In addition, it would be desirable to include the most stable instrument beside the EM27/SUN, the Vertex70, into

COCCON, maybe as an extended COCCON.

7 APPLICABLE DOCUMENTS

Statement of Work: Fiducial Reference Measurements for Ground-Based FTIR Greenhouse Gas Observations (FRM4GHG)
Prepared by: T. Fehr/B. Bojkov (EOP-GMQ), Reference: ESA-EOPG-MOM-SOW-0007

8 REFERENCE DOCUMENTS

FRM4GHG deliverables from the Phase 1, phase 2 and phase 3 made available via the project website http://frm4ghg.aeronomie.be/index.php/outreach/deliverables

Sha, M. K., De Mazière, M., Notholt, J., Blumenstock, T., Chen, H., Dehn, A., Griffith, D. W. T., Hase, F., Heikkinen, P., Hermans, C., Hoffmann, A., Huebner, M., Jones, N., Kivi, R., Langerock, B., Petri, C., Scolas, F., Tu, Q., and Weidmann, D.: Intercomparison of low and high resolution infrared spectrometers for ground-based solar remote sensing measurements of total column concentrations of CO2, CH4 and CO, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2019-371, in review, 2019.