

FRM4GHG 2.0

Fiducial Reference Measurements for Greenhouse Gases



Semesterly Report Month T0+24

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1 DOCUMENT CHANGE RECORD

Issue	Date	Item	Comment
V1.0	2023-12-10	-	Initial version
V1.1	2024-01-14	-	Updated version following comments from ESA

2 ACCESS LIST

This document is a deliverable “D4.1.3: Semesterly report Month T0+24 created for the project FRM4GHG 2.0 and will be submitted to ESA. The document can be downloaded from the project webpage <http://frm4ghg.aeronomie.be>.

3 PURPOSE

The document describes the progress of the WPs over the time period January 2023 to December 2023 (two 6 monthly reports up to T0 + 24 months).

4 DOCUMENT STRUCTURE

The document gives the progress report for the month T0+24. There were a total of 8 WebEx meeting taking place during the last reporting period. Telecons were held on 18 January 2023, 26 January 2023, 9 February 2023, 21 April 2023, 25 May 2023, 13 July 2023, 7 September 2023 and 13 November 2023. All WPs of FRM4GHG-2 make good progress. However, only details of the progress reports presented during the current reporting period outlaid per work package in the following section.

5 DELIVERABLES

The two deliverables, D2.4.1 and D3.1.2 have been also submitted and accepted during the current reporting period.

6 RESULTS

WP1: Instrumental improvements to low-resolution spectrometers for GHG observations:

The RAL group presented several changes and updates, which have been performed with the Laser Heterodyne Spectroradiometer (LHR).

RAL scientists have worked on identifying the problems and finding a solution for them. The list of problems (described below as P) and the solutions (described below as S) are given as a summary in this report.

P1) Problem with the background heterodyne signal caused by the wide bandwidth solar radiation

S1) Addition of narrow-band optical filters

P2) Problem with unstable amplitude modulation because of unstable chopper wheel.

S2) The chopper wheel was replaced with tuning fork modulator.

P3) Laser instability caused by light propagating back into the laser.

S3) Addition of an isolator (polariser & $\lambda/4$)

P4) Small image and wasted information caused due to single lens optics and monochrome.

S4) Solved by using two lens optics and hyperspectral camera.

P5) High instrument temperature caused by heat coming from internal electronics and low ventilation in the system.

S5) Added a water cooled base plate.

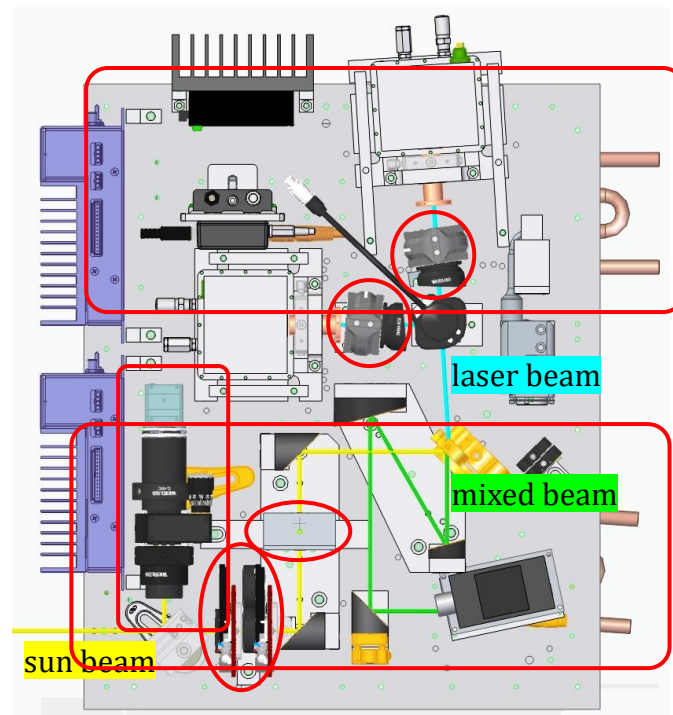
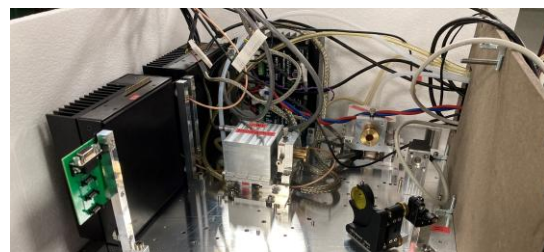
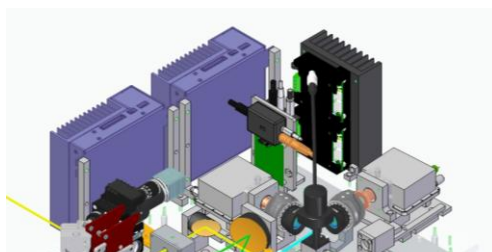


Figure: LHR system and the marked circles show the area of problems and solutions as discussed above.



The rebuilding of the optics is underway. Once this is ready then the next step is to perform atmospheric measurements against TCCON station.

University of Wollongong performed thermo-electrically-cooled PV-MCT (photo-voltaic Mercury Cadmium Telluride) detector tests for solar Fourier transform spectrometer (FTS) measurements. The results of the initial assessment have been presented at one of the WebEx meetings.

There is an increasing interest in TCCON for spectra from 2000 – 4000 cm^{-1} below the InGaAs cutoff. Measurements in this region is of special interest to co-retrieve OCS and C₂H₆. This is currently achieved by performing InSb detector at limited number of sites. The measurements are performed in dual configuration (InGaAs/InSb). InSb has high sensitivity but requires liquid nitrogen for cooling. As a result it might be difficult to implement such a system for remote sites where liquid nitrogen might not be easily available. Furthermore, InSb detector saturates with full solar signal, and therefore optical bandpass filters are needed.

The Judson J19TE series TE-cooled PV-MCT detector covers a similar frequency range to the InSb and requires no liquid nitrogen. However, it is less sensitive but still accepts higher signal levels without saturation.

The configuration for the detector is given below:

Used in the UoW in situ FT analyser (“ooofti”) since ~ 2007

Bruker IRCube FTS at 0.5 cm^{-1} resolution, 1900 – 5000 cm^{-1}

- Global source, CaF₂ beamsplitter
- 26 m multipass White cell
- high signal throughput

Here we assess this detector in a modified IRCube used for solar FTS measurements at UoW

Rooftop tracker

NIWA design, open path (no fibre optic)

IRcube in lab, modified to emulate EM27 optics (no camtracker)

- Direct parallel solar beam into the IRCube interferometer
- 127 mm FL OAP focussing onto detector, 0.6 mm aperture
- CaF₂ beamsplitter
- TE-MCT in similar detector package to InGaAs

	TE PV-MCT	InSb
Model (1mm circular)	J19TE3	J10D
Operating temperature	-65°C / -45°C	77K
Cooling	3 stage TE	LN2 dewar
Peak wavenumber	2200 cm ⁻¹	2100 cm ⁻¹
Peak responsivity	1.6 A/W	3.0 A/W
Detectivity D*	2 – 5 E10	1 E11

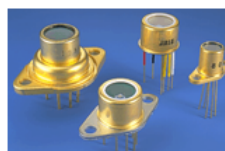
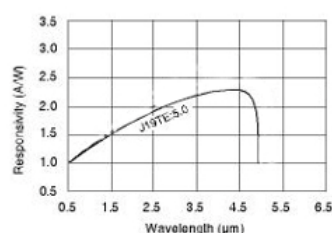


Figure: TE cooled PV-MCT vs InSb detector (Teledyne – Judson Technologies)

Parallel measurements using TCCON IFS125 HR + IRCube have been performed using NIWA solar tracker with about 100 mm parallel beam. The beam was split 50:50 to HR and IRCube FTSs using a pickoff mirror and not a beamsplitter. The TCCON HR has a configuration of using InGaAs + InSb dual detector with spectra recorded at 0.5 (TCCON) and 0.004 (NDACC) cm⁻¹ resolution. The IRCube has a CaF2 beamsplitter and TE-MCT detector configuration.

Survey spectra

— IRCube, CaF2, TE_MCT
— IRCube, Quartz, InGaAs
— HR125_CaF2, InGaAs

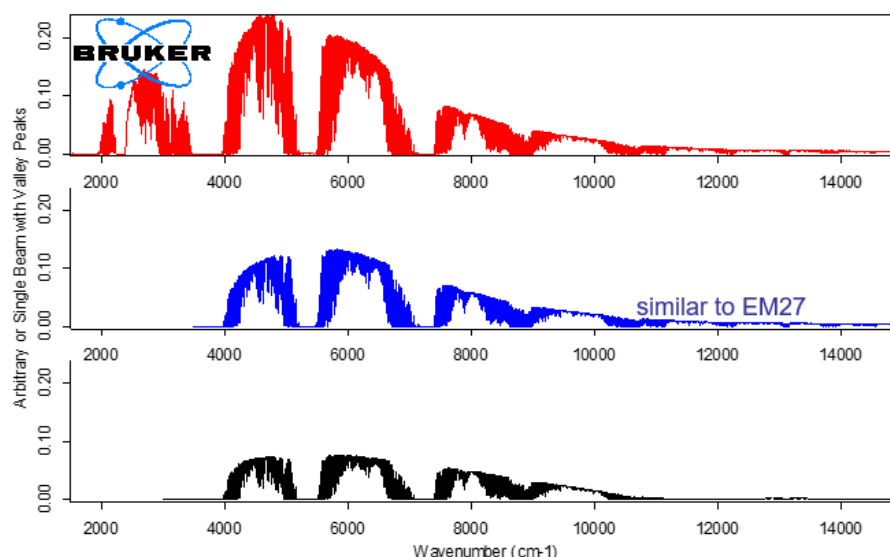
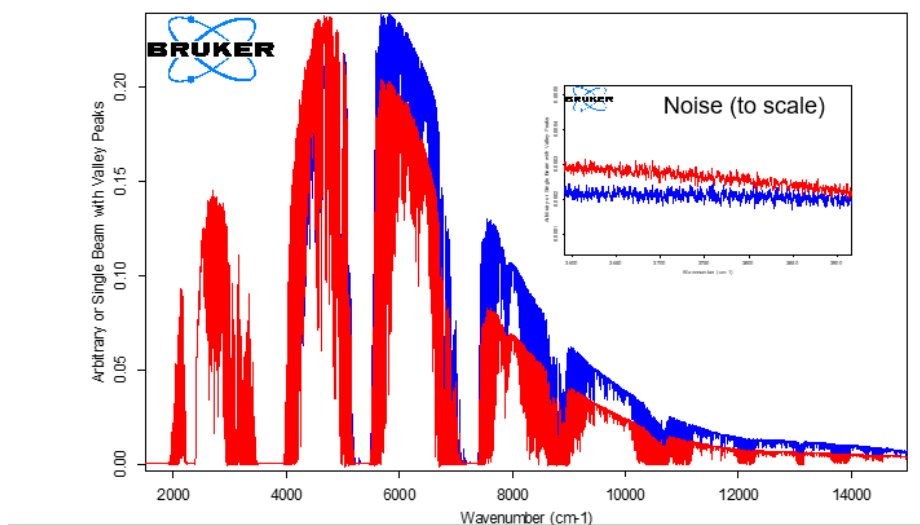


Figure: Survey spectra from 2 October 2023.

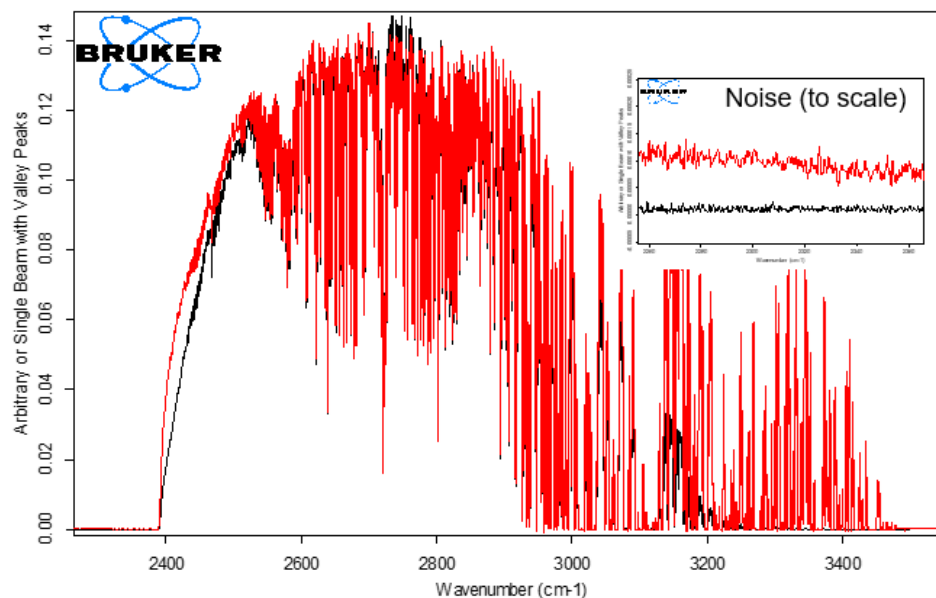
TE-MCT vs InGaAs

— IRcube, CaF2, TE_MCT
— IRcube, Quartz, InGaAs



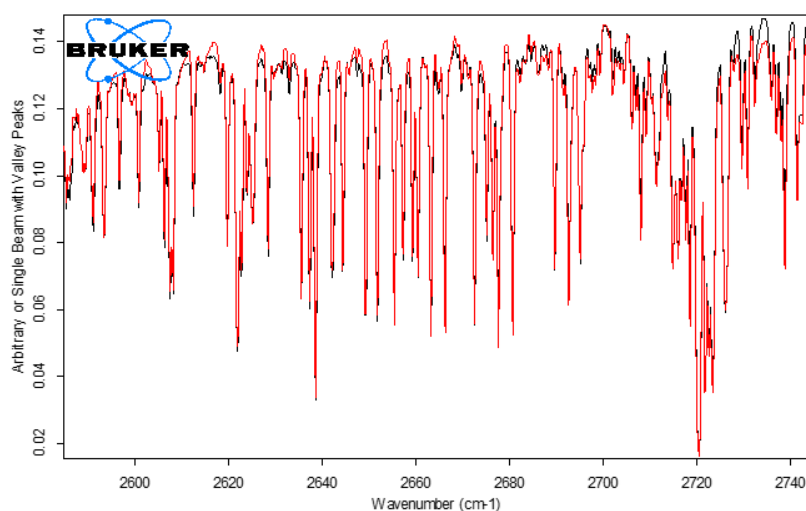
TE-MCT vs InSb Filter 3

— IRcube, CaF2, TE_MCT
— HR125, CaF2, InSb F3



TE-MCT vs InSb Filter 3

— IRcube, CaF₂, TE_MCT
— HR125, CaF₂, InSb F3



<u>FTS</u>	<u>BS</u>	<u>Detector</u>	<u>SNR*</u>
IRcube	CaF ₂	TE-MCT	15000
IRcube/EM27	Quartz	InGaAs	17000
HR125	CaF ₂	InGaAs	18000
HR125	CaF ₂	InSb	46000

* Peak / rms noise, preliminary only
(only 1/2 day of measurements)

Statistics showing the SNR comparison for the test setup.

Potential applications of TE-cooled PV-MCT detectors:

- Cover 2000-11000 cm⁻¹ in a single spectrum
- HR125 – high resolution, TCCON and NDACC
 - Replace InSb when LN₂ is logistically difficult
 - Replace or duplicate InGaAs
- EM27 and IRcube
 - Extend to 2000 cm⁻¹
 - Requires CaF₂ beamsplitter
 - Replace InGaAs completely?
- Caveats / next steps
 - Full assessment of SNR and non-linearity
 - Not tested at high resolution – 0.5 cm⁻¹ only
 - Heat sink for TE cooling in HR125 under vacuum

WP2: Improvements to retrieval algorithms for GHG and additional species from low-resolution and TCCON spectrometers

FMI continues to perform TCCON and EM27/SUN type of measurements and performed several AirCore launches. They have updated the software to run the HR instrument and installed new sensors to operate the HR solar tracker cover. A new enclosure for the EM27/SUN has been built.

FMI hosted a balloon campaign from July 24 until August 11, 2023 in Sodankylä. In total we performed 12 flights with various instruments. This provided an opportunity to test new samplers and sensors and to intercompare results. The focus of the campaign was to measure CO₂, CH₄, N₂O, COS, O₂, O₃, water vapor isotopologues, stratospheric circulation tracers and Ozone-Depleting Substances. Instrumentation included several AirCores, LISA sampler by the Groningen University and sub-samplers to analyze stratospheric gases.

The AirCore profiles were compared to the GGG prior and the differences improved with the new GGG 2020 prior as compared to the GGG 2014 prior.

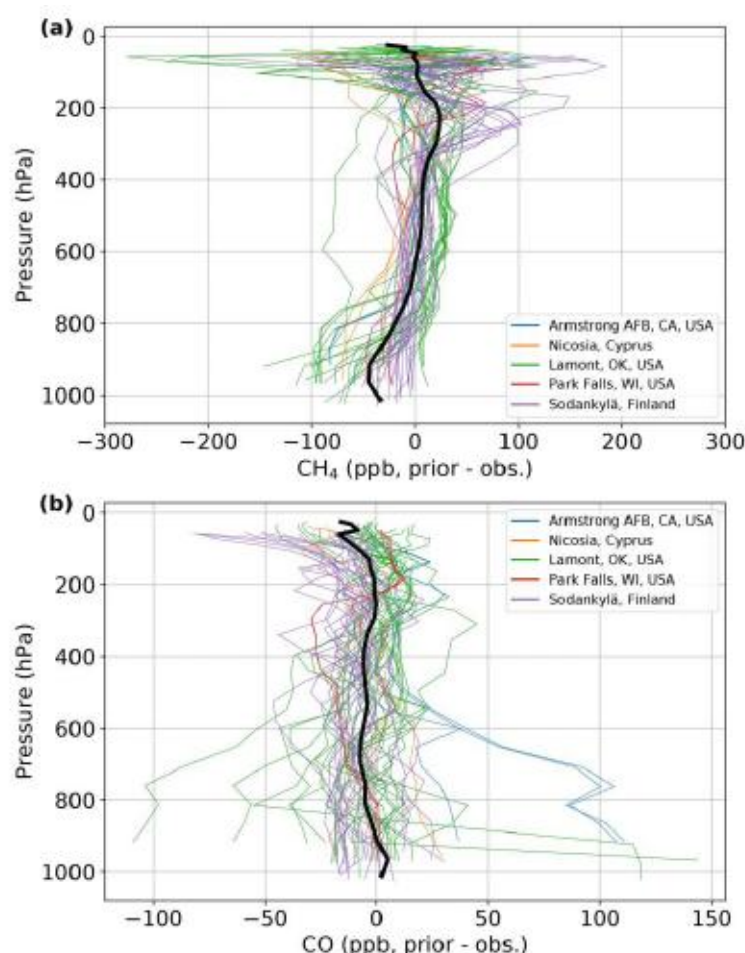


Figure: Difference plots for GGG2020 priors versus (a) CH₄ and (b) CO AirCore data. The thinner, colored lines represent differences for individual profiles, and the thick black line indicates the mean difference across all profiles shown. The individual differences are colored by their TCCON site (Laughner et al., 2023).

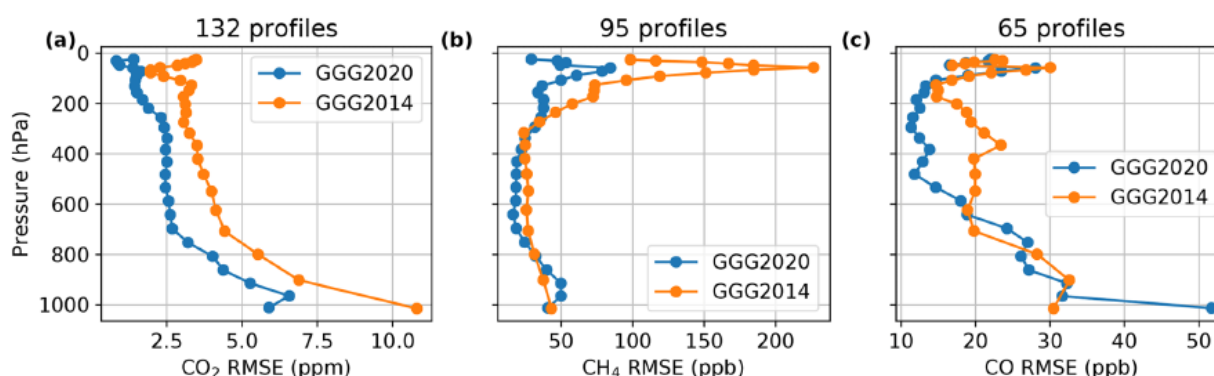


Figure: Root mean square error (RMSE) of (a) CO₂, (b) CH₄, and (c) CO priors versus combined AirCore and aircraft observations (Laughner et al., 2023).

The AirCore data were also compared to the model data from FMI (InvSURF). The results show good agreement.

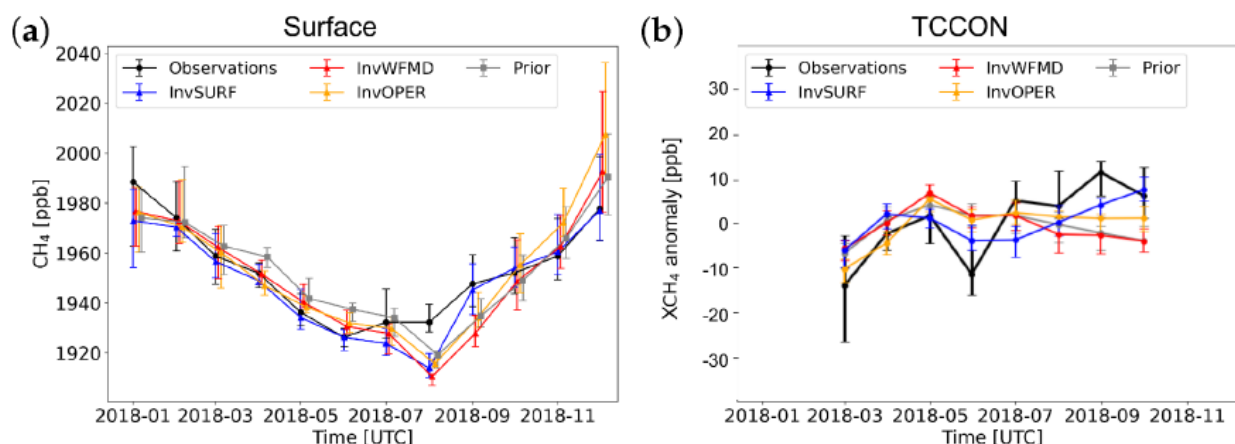


Figure: A) Monthly median and first and third quantiles at surface station against the assimilated data in InvSURF (b) quantiles of XCH₄ anomaly against TCCON retrievals where annual means are subtracted. (Tsuruta et al., 2023).

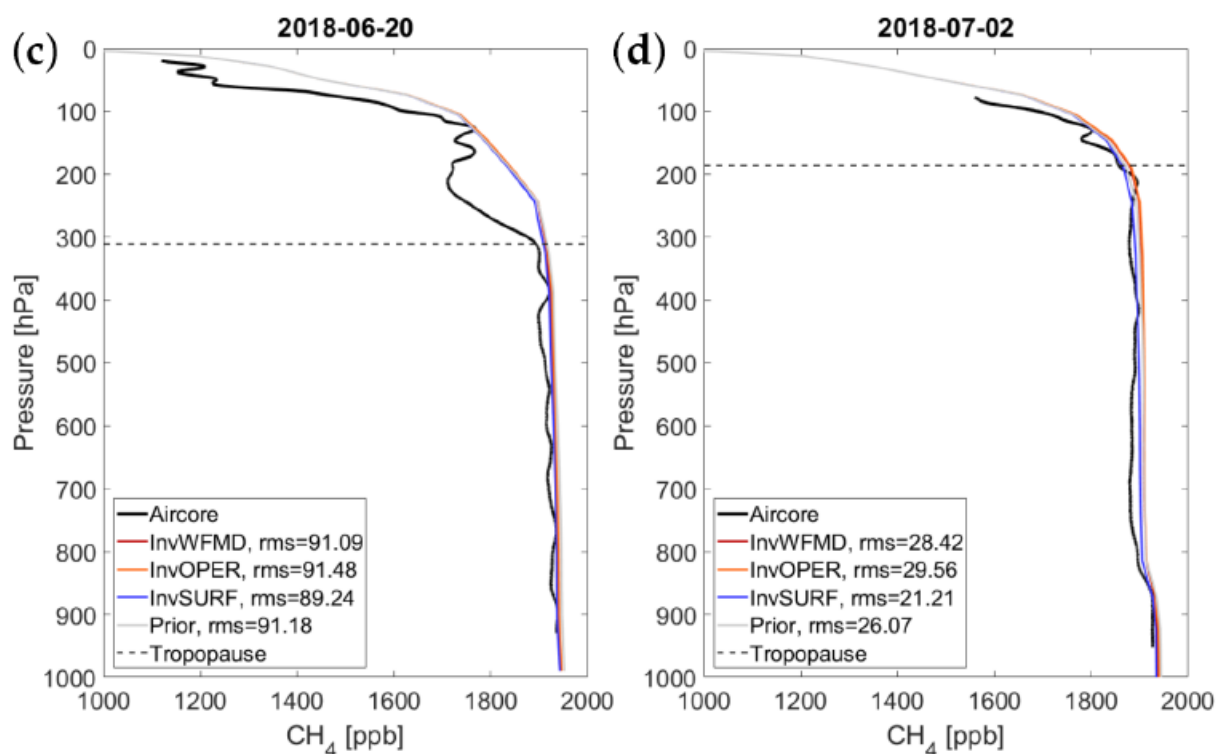


Figure: CH₄ profile against AirCore measurements (Tsuruta et al., 2023).

RUG performed AirCore launches with the new AirCore at Sodankylä. Because of the new AirCores the CO enhancements seen in the previously is not observed any more (see figure below).

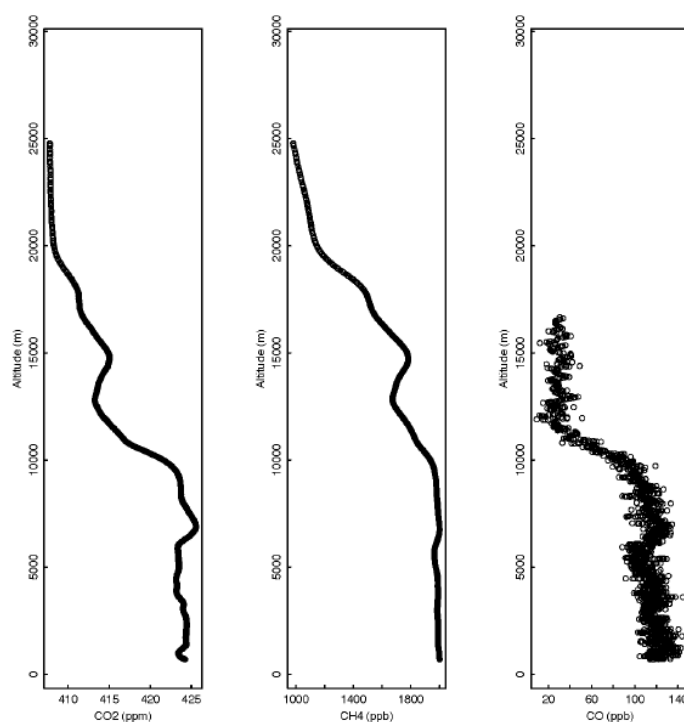
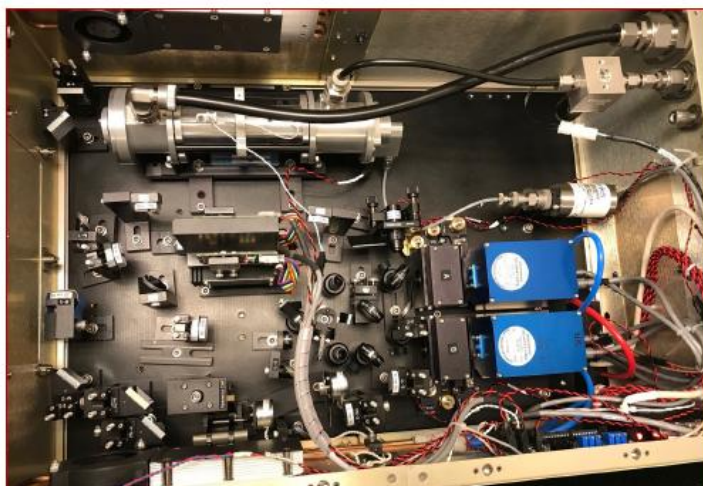


Figure: AirCore flight on April 27, 2023 using a new AirCore.

New dual AirCore and LISA launches were performed at Sodankylä between 31 July and 14 August 2023. The key changes performed to the system are electronics updates, addition of differential pressure sensor, AirCore coating performed in the US. Furthermore, a LISA bags have been chosen, which were not affecting COS and an ozone scrubber is in testing in the laboratory (see picture below).

- Dual laser QCLS: OCS/N₂O/CO₂/CH₄/CO
- Sub-sampling of air samples

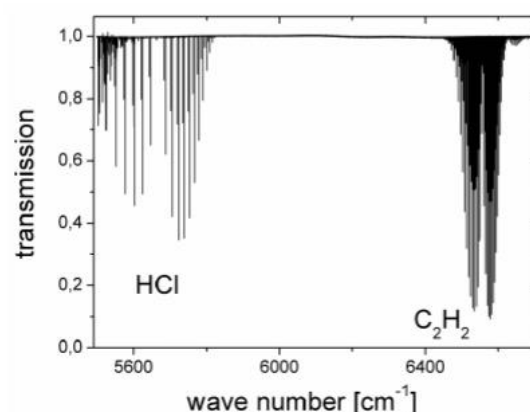
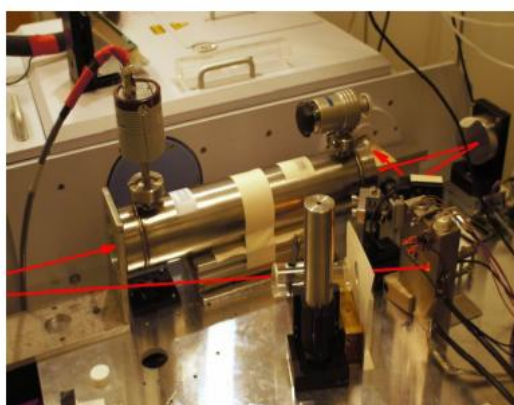


WP3: Improvements to TCCON and COCCON network operations, consistency and data delivery

KIT performed sample test of new HCl cell parameters and found some very interesting results and conclusions.

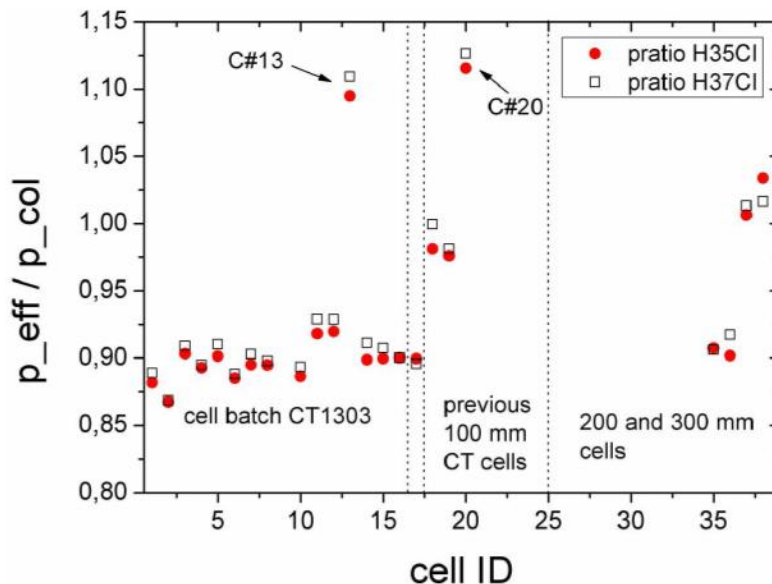
HCl cell measurements are performed at the TCCON sites using lamp and/or solar measurements to get information about the instrumental line shape (ILS). The method requires cells of known content (ideally pure HCl). However, the practical problems of this realization were initially underestimated (Hase et al., 2013).

The applied method of testing was to measure HCl + reference C₂H₂ cell in the same beam. See setup below.



The key finding of the investigation was that the fabricated cells do not contain pure HCl but rather a HCl-air mixtures of variable composition.

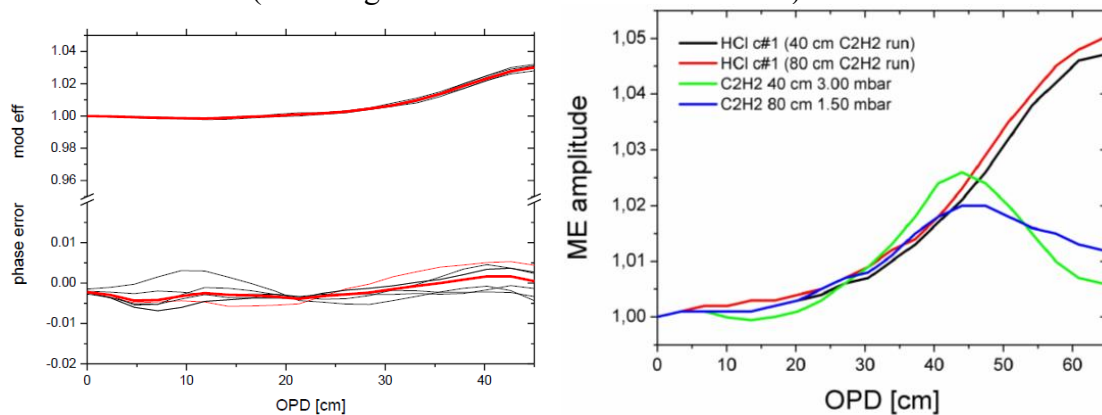
As a solution specific “effective pressure parameters” were determined for each cell. The retrieved modulation efficiency amplitudes can change by 4% at 45 cm OPD if the HCl pressure is in error by 0.5 mbar in the ILS retrieval setup (see below figure).

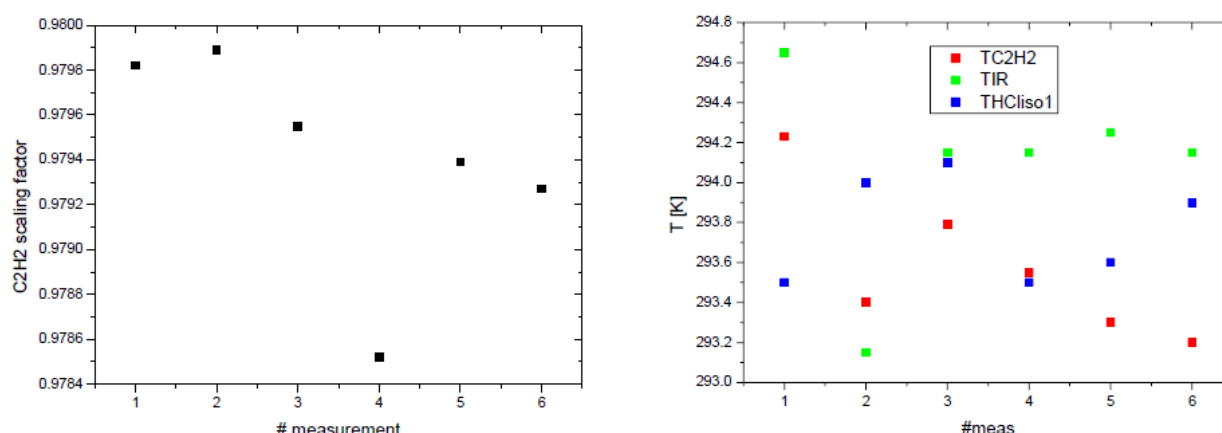


The assumption was that the origin of the problem might be due to the too long waiting period for some of the cells before being molted off and some leaks in teflon valve.

A new batch of HCl cells has been fabricated meanwhile. The problem with the shipping and distribution of the cell remains requiring additional round trip of new cell batch using ATA carnet.

As the origin of problem with the first batch is believed to be known, it was decided to skip the calibration. But a subset of cells has been provided to KIT by Univ of Bremen for sample testing (#15, #16, #18, #20). The same measurements have been performed as in the previous calibration exercise (but using MPD = 45 cm instead of 65 cm). The results are shown below:

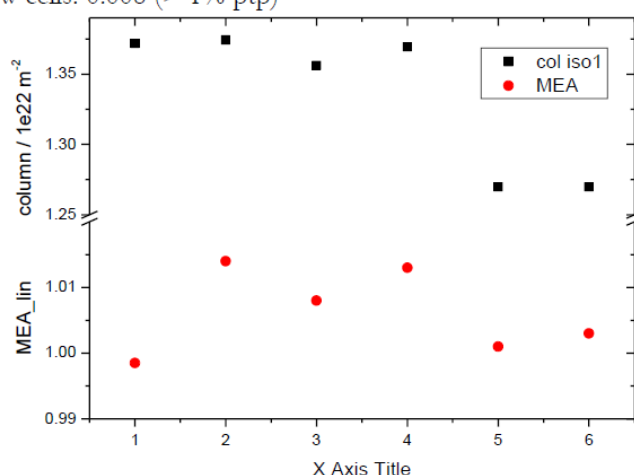




Results show that the Temperature has as standard deviation of about 0.5 K and C2H2 column shows a standard deviation of 0.05% with max-min of 0.1%.

Results for HCl cells: retrieved linear MEA on top of C2H2 ILS
(effective pressure calculated assuming pure HCl: $p_{\text{eff}} = 0.875 * p_{\text{col}}$)
(#15: 1, #16: 2, #18: 3, #20: 4, #1-1: 5, #1-2: 6)

Stdev MEA new cells: 0.008 (> 1% ptp)



The conclusion of the work was that the preliminary results need to be rechecked. Cell contents do not appear identical indicating still variable purity. The sub-sample test was OK for current +/- 4% MEA requirements. Station-to-station biases due to differing cell characteristics might be expected on about 1% level.

Projection towards future enhanced requirements (MicroCarb, CO2M) suggests that bypassing the C2H2 calibration was a bad idea. Characterization of distributed cells will be considerably more work (and the results probably more prone to inconsistencies...).

KIT provided a report on travel standard tasks in the last months:

The travel standard completed successful deployment to Izana. It has been checked at KIT upon its arrival from Izana. The transport from Izana back to Germany has been extremely rough, the g-sensor detected up to about 30 g. It seems that the CO detector suffered due to this, the Xgas calibration changed. Because the colleagues in Paris urged to receive the spectrometer due to weather and other side constraints, KIT nevertheless decided to send

immediately the spectrometer on to the TCCON Paris site (as the main channel calibration seems ok).

A suggestion would be to better have a pair of spectrometers available for realizing the TS activity, this would give more time for the calibration at KIT and more flexibility to choose the other spectrometer if the one coming back from deployment has suffered (so the shelter would then be sent with the other spectrometer). Therefore, KIT will reserve a further spectrometer for the TS activity. The additional spectrometer will not be available immediately, as KIT needs to do careful side-by-side measurements with the existing TS spectrometer to avoid biases between TSA and TSB results.

In addition, the work on the HCHO retrieval from low-resolution spectrometers (**D2.4.1**), demonstrating the strategy for retrieving HCHO from low-resolution spectrometers was successfully finished. The Report on the PROFFAST update and data-stream verification (**D3.1.2**) where PROFFAST includes the option to ingest ECMWF P/T and CAMS a priori profile data (verified) was also finished. Both reports can be found in the deliverables section on the project website in the outreach section.

7 OUTLOOK

During the next 12 months the partners will work on

- the instrumental improvement of the Vertex70
- the experimental instrumental improvement of the IRCube.
- an update of the Linefit program,
- an update of the PROFFAST retrieval,
- the Vertex and IRCube data uptake in the COCCON schema,
- the LHR data retrieval
- an enhanced retrieval product from the TCCON spectra,
- an update on the AirCore retrieval procedure,
- a reprocessed L1 and L2 dataset,
- a comparison exercise on the data retrieval

A status report will be given by the semesterly report after 6 months.

8 APPLICABLE DOCUMENTS

Statement of Work: Fiducial Reference Measurements for Ground-Based IR Greenhouse Gas Observations (FRM4GHG 2.0)

Prepared by: EOP-GMQ, Reference: ESA-EOPG-EOPGMQ-SOW-21

9 REFERENCE DOCUMENTS

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The Total Carbon Column Observing Network's GGG2020 Data Version, Earth Syst. Sci. Data Discuss. [preprint], <https://doi.org/10.5194/essd-2023-331>, in review, 2023.

Tsuruta, A.; Kivimäki, E.; Lindqvist, H.; Karppinen, T.; Backman, L.; Hakkarainen, J.; Schneising, O.; Buchwitz, M.; Lan, X.; Kivi, R.; et al. CH₄ Fluxes Derived from Assimilation of TROPOMI XCH₄ in CarbonTracker Europe-CH₄: Evaluation of Seasonality and Spatial Distribution in the Northern High Latitudes. Remote Sens. 2023, 15, 1620.
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