

FRM4GHG 2.0

Fiducial Reference Measurements for Greenhouse Gases



Semesterly Report Month T0+36

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1 DOCUMENT CHANGE RECORD

Issue	Date	Item	Comment
V1.0	2024-09-11	D4.1.3	Initial version

2 ACCESS LIST

This document is a deliverable “D4.1.3: Semesterly report Month T0+36 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 August 2023 to August 2024 (6 monthly report up to T0 + 30 months and 6 monthly report up to T0 + 36 months).

4 DOCUMENT STRUCTURE

The document gives the progress report for the month T0+36. There were a total of 6 WebEx meetings taking place during the last reporting period. Telecons were held on 7 September 2023, 13 November 2023, 16 January 2024, 12 March 2024, 25 June 2024, and 26 July 2024. All WPs of FRM4GHG-2 make good progress.

5 DELIVERABLES

The deliverables, D1.1.1, D1.1.2, D1.3.1, D2.1.1, D2.1.2, D2.2, D2.3.2, D2.5.1, D2.6.1, D3.1.3 and D4.1.3 are due for submission in the current reporting period. They are all worked on, some of the deliverables have been already submitted to ESA for review while the others are in the pipeline to be finalized and soon submitted to ESA for review.

6 RESULTS

WP 1: Instrumental improvements to low-resolution spectrometers for GHG observations

Develop a more mobile, stand-alone version of the Bruker Vertex70 instrument for deployment in the field (BIRA & Bremen)

Embedding the Vertex spectrometer in a thermally isolated, waterproof enclosure, including a rain sensor and compact solar tracker mounted to the spectrometer, and an automatically controlled cover of the tracker. Enclosure system including rain sensor and compact solar tracker is assembled and currently undergoing testing at our campus in Uccle, Belgium.

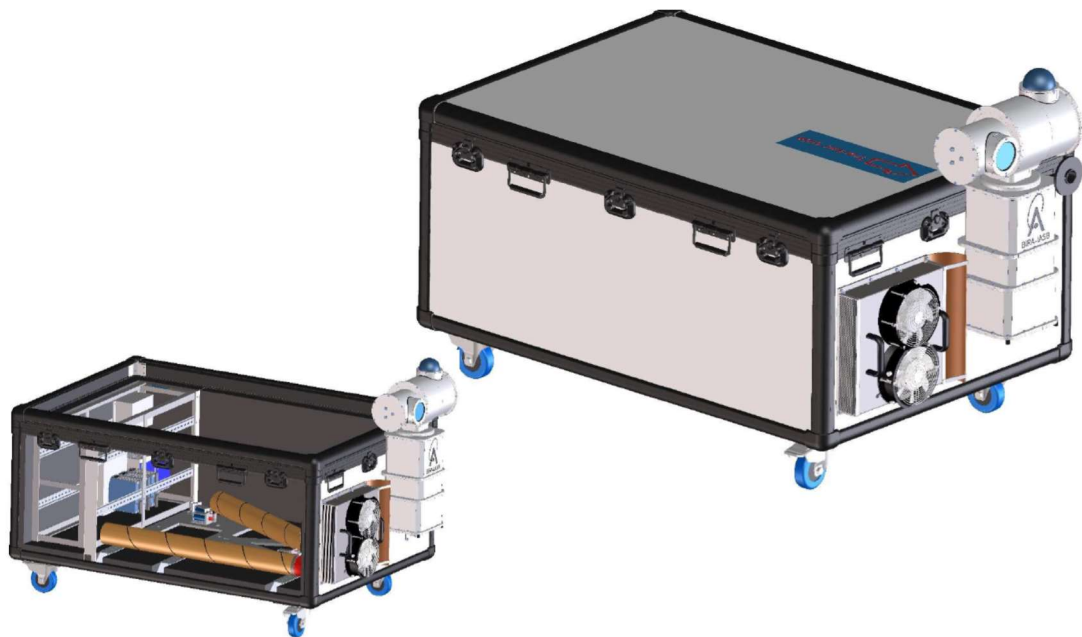


Figure: CAD drawing of the enclosure box and solar tracker



Figure: Enclosure box and meteorological station deployed for testing at the campus of BIRA



Figure: Enclosure box and solar tracker

Develop AirCore for retrieving OCS and N₂O in addition to CO₂/CH₄/CO (FMI & RUG)

Additional AirCore launches were made in April 2024 and in early September 2024 by FMI in collaboration with RUG. The latter flight included a combined launch of two AirCore instruments. This setup allowed to involve two CRDS systems. First system was used to analyse CO₂, CH₄, CO profiles, and the second system to analyse N₂O

and OCS profiles. The preliminary retrievals of both OCS and N₂O have been carried out at FMI.

AirCore retrieval procedure updates (RUG)

To improve the accuracy of the altitude registration of AirCore profiles, AirCore retrievals have been realized using the equilibrium method. A number of sensitivity tests have been performed to determine the most appropriate starting point during the retrieval procedure, which is evaluated against three methods, i.e., Tans' pressure correction method, the spiking technique, and LISA sampling method.

Track down and solve identified LHR instrumental problems (RAL)

An extensive lab-based characterization of the re-assembled Laser Heterodyne Spectro-Radiometer (LHR) was performed. During this characterization, a noise budget was developed and validated to describe the contribution of different subsystems to the overall instrument noise. Furthermore, the physical origins of unwanted background signals were identified, which identified a need for narrow-pass optical filters to reject out-of-band solar radiation. Such a filter has been installed for our CO₂ channel. In addition, water cooling and insulation was added to improve the thermal stability of the instrument, and the instrument was re-wired to reduce the number of external connections. The instrument measuring configuration was also updated to improve spectral calibration. The instrument was deployed in the RAL dome alongside the Harwell TCCON station in August 2024.

EM27 with fibre optic-coupled solar tracker at Karlsruhe (Wollongong & Karlsruhe)

Summary

- EM27 with fibre-optic-coupled solar tracker developed at UOW
 - Similar to the IRcube system as used at Sodankyla
 - Weather proof tracker + telescope outdoors
 - FTS indoors, weather-safe
 - Automated, operates 24/7, takes spectra only when sunny
- Operated over ~ 1 year at UoW with EM27
- Set up at KIT on 14 Mar 2024 beside COCCON reference system (SN037) on IMK building
 - TCCON station is 1 km north on same KIT site
- To run through summer-autumn 2024
- To 27 May:
 - EM27-FO ~ 15,000 spectra
 - COCCON ~ 4,400 spectra

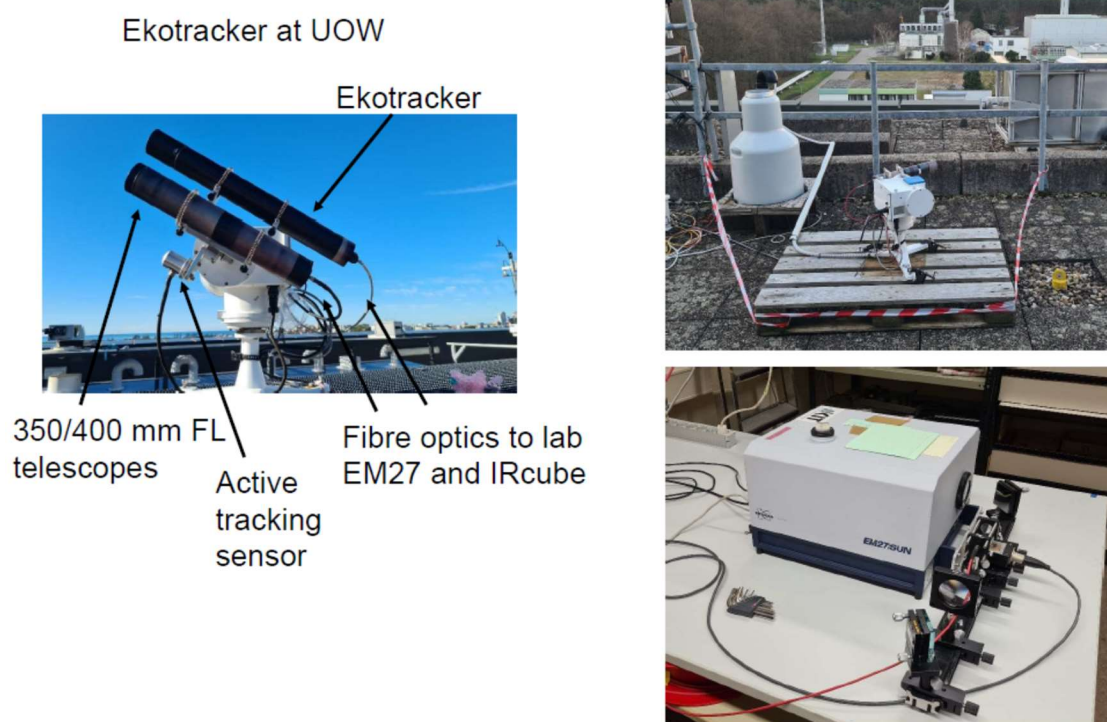


Figure: EM27-FO in pictures

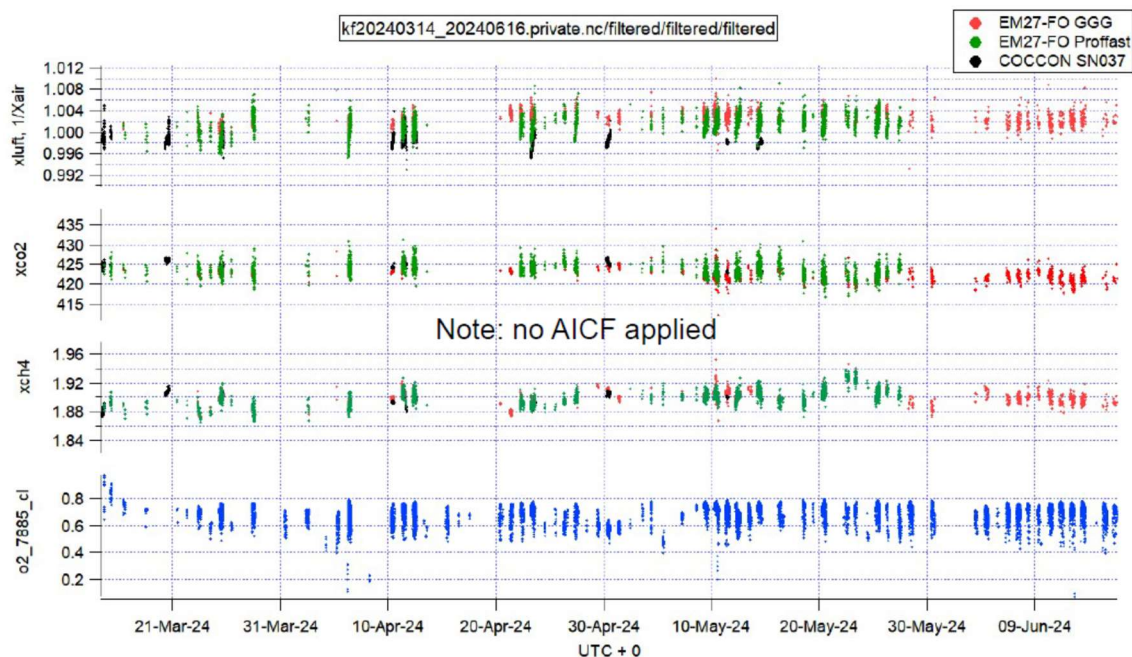


Figure: Timeseries of the first data (1) collected during the Karlsruhe side-by-side measurements with EM27-FO processed with GGG and PROFFAST and reference EM27SUN (SN037) processed with PROFFAST. Note that no Airmass Independent Correction Factor (AICF) has been applied to the data shown here.

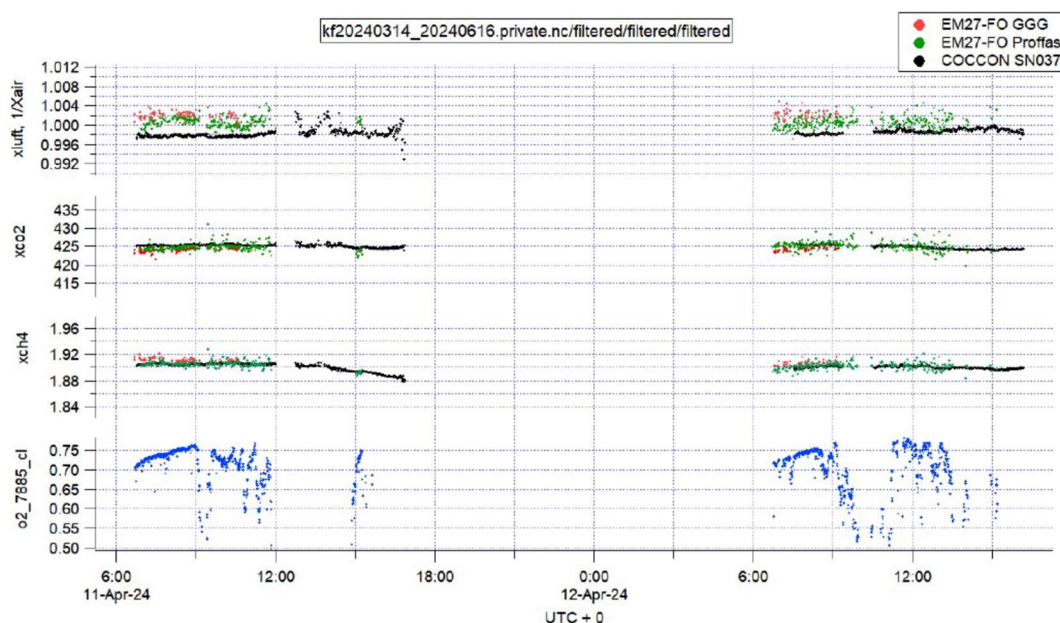


Figure: First data (2) same as above but plotted for two individual days

Final comments

- 2-3 systems also trialling in Toronto
Debra Wunch, Niki Jacobs from University of Toronto are working on this effort
- More detail and further comparisons in time has been presented at the FTIR annual meeting in Boulder 2024
- Plans to locate the IRcube + FO at Cape Grim GAW station, late 2024

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

Update on COCCON processing (Karlsruhe)

The COCCON data processing chain has three steps: (1) PREPROCESS: Generates spectra from raw interferograms (2) PCXS: Generates daily lookup tables informing about x-sections of gases (3) INVERS: Trace gas retrievals for all spectra measured during a day. The code has been updated to the operational version 2.4: (a) PREPROCESS – support of variable observer coordinate (b) PCXS – faster generation of xsc-lookup table. Furthermore the update of the current beta version includes: (c) nonlinearity correction for the EM27/SUN (d) residual solar Doppler scale diagnostics

COCCON processing: PREPROCESS

Envisaged procedure: (1) PREPROCESS needs to accept variable observer coordinates (2) A daily map file for the ship's noon coordinate is requested (3) The pressure data measured on board is used by INVERS (no change here) Extension of PREPROCESS code (done); Extensions on the wrapper means: generate PREPROCESS input (use ship track) request map file for noon coordinate

Relevant sections from standard preprocess.inp:

- T/F: fixed or variable observer coordinates
- specify site name
- specify time offset of OPUS time versus UT (hours+decimals)
- if fixed observer coordinates: specify fixed observer coords (lat_deg,long_deg (East pos),alt_km)

COCCON processing: PCXS

PREPROCESS and INVERS are pretty efficient, the currently distributed version of

- PCXS requires ~ 7 min (single core, Intel i7-3770 CPU @ 3.40GHz) for generating the required lookup table.
- For (re)processing of a large number of measurement days, the computational effort for PCXS becomes noticeable, a reduction of computational effort is desirable.
- Current scheme:
 - Each line contribution (Voigt + extensions (LM, SDV)) is added on each spectral grid point of a fine global spectral grid ($\pm 25 \text{ cm}^{-1}$).
- New scheme:
 - Each line contribution is split in two parts. The first part contains the narrow section around the line center peak (sampled on the fine global spectral grid. The remaining second low-res part is sampled on a coarser spectral grid ($\Delta \nu_{coarse} = 4 \Delta \nu_{fine}$). After collecting all line contributions, the coarse grid contribution is interpolated and added on the fine grid.

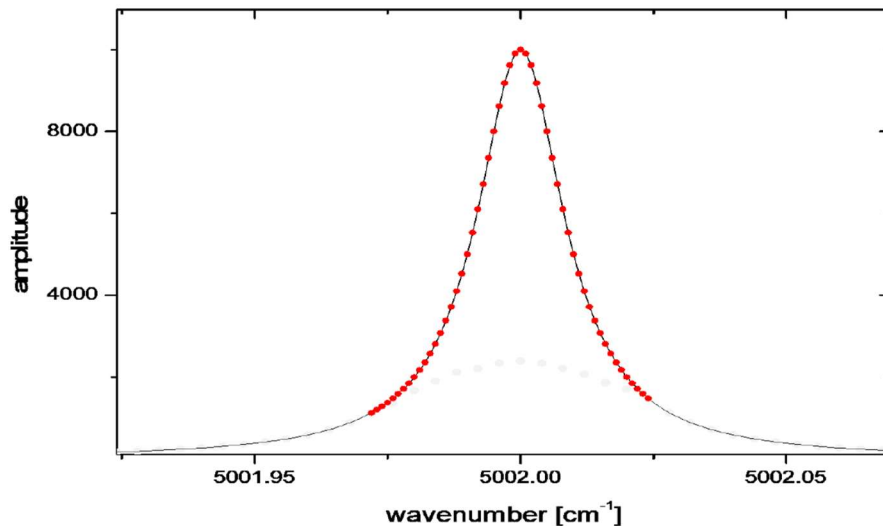


Figure: Graphical outline of the procedure (a smooth polynomial replacement for completion of the coarse grid contribution is used. This needs to be subtracted from the fine grid contribution):

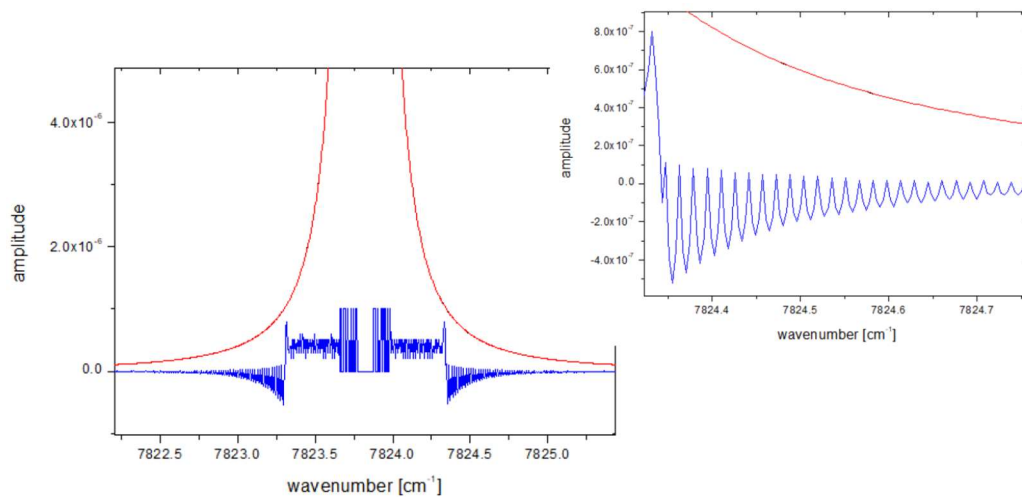


Figure: Effects on modelled line shapes (difference x 1000)

Effects on XGAS:

XH₂O ~7e-5

XCO₂ ~8e-5

XCH₄ ~1e-5

XCO ~2e-5

Speed gain for creating one table: 7 min -> 1 min 40 s.

We plan to include these updates in the next PROFFAST release (2.4):

COCCON processing: Nonlinearity correction

Nonlinearity correction scheme implemented (for operational use). Two-step procedure:

Step (1):

- Perform fit of characteristic curve (using dedicated PREPROCESS option: based on a set (~12) of selected measurements), result: set of parameters
- This task needs to be done for all pre / gain configurations used on spectrometer SNXXX

Step (2):

- Add table of parameters for all required pre / gain configurations for selected spectrometer SNXXX
- Redo the preprocessing of all raw ifgs recorded with SNXXX (switch on NLC corr option)
- Redo the trace gas analysis

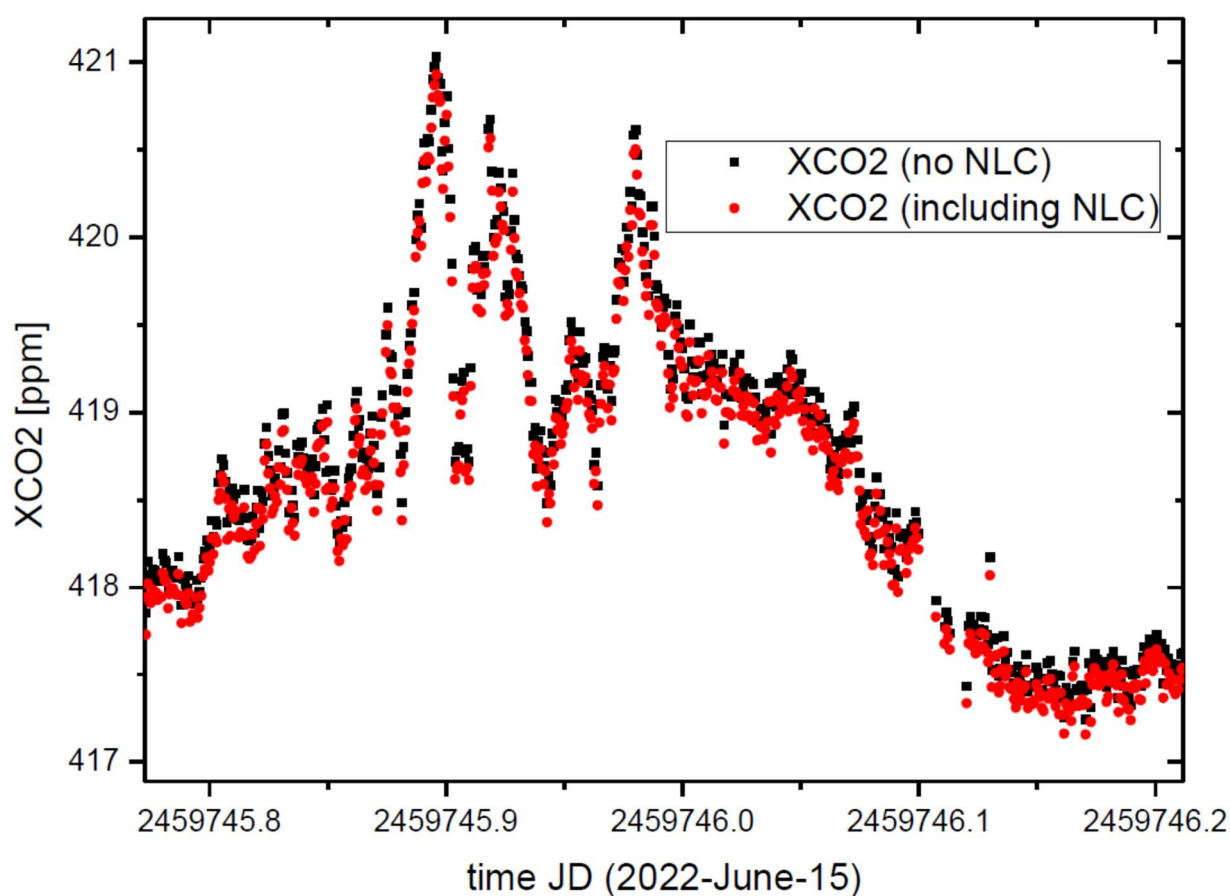


Figure: Results for CO₂, retrieved with and without Non Linearity Correction

SN37 (2022-June-15)	Rel change NLC / no NLC
XCO ₂	-2.4e-4 (-0.1 ppm)
XCH ₄	-2.4e-4 (-0.5 ppb)
SN37 (2022-Mar-25)	Rel change NLC / no NLC
XCO ₂	+8.5e-5 (+0.03 ppm)
XCH ₄	+1.1e-4 (+0.2 ppb)

Preliminary conclusions:

- Moderate effects, but not negligible
- Corrections vary through year, during a day nearly constant (expect spurious annual cycle)
- Volunteers for code testing / evaluation of corrections required (AEMET/COCCON, Spain?)
- Application would impose quite some work (characterisation of each unit required)!

COCCON processing: solar Doppler scale

- Diagnosis of residual solar Doppler scale after convergence is reached
- Performed for each existing job (variable quality), new output variable "jobXY_sol"
- Good pointing stability found for EM27/SUN FTS standard camtracker

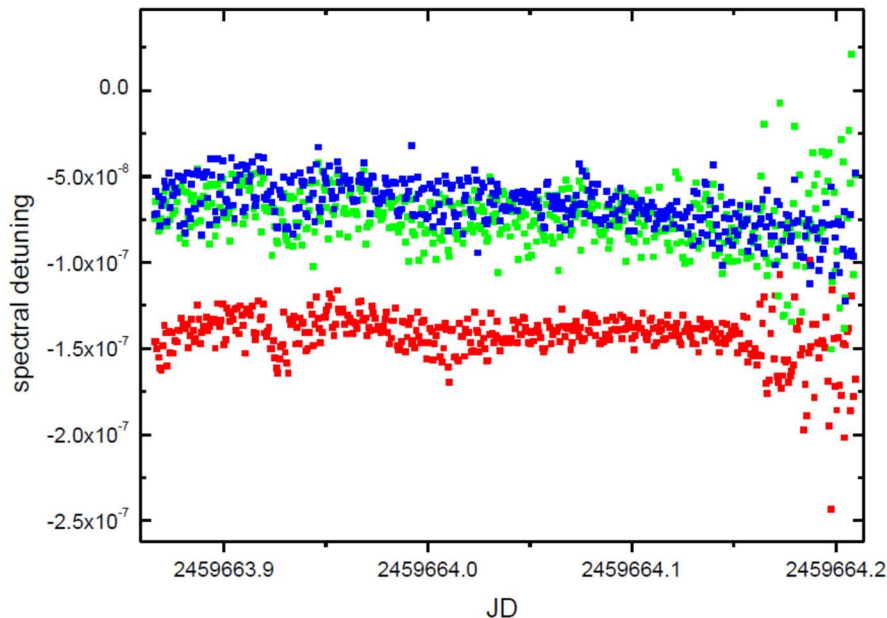


Figure: The Figure shows results from three different spectral fitting regions. Note: solar equatorial rotation speed ~ 2 km/s, therefore: $1e-7$ spectral detuning $\sim 0.2'$ offset (equatorial component).

Summary

Implemented in PROFFAST operational ver 2.4

- New PREPROCESS supports data analysis for shipborne measurements
- Calculation of daily x-section tables is accelerated

Implemented in internal beta version of PROFFAST:

- Nonlinearity correction scheme
- Residual solar Doppler analysis

Uptake of low resolution spectrometer in the COCCON schema (Karlsruhe & Wollongong)

KIT supported the installation, operation, data analysis & comparisons of a fiber-coupled EM27/SUN, which has been installed by UoW in March (activity lead: D. Griffith, C. Alberti).

KIT has implemented the required extensions in the PROFFAST code in order to serve the other portable spectrometers operated in the framework of FRM4GHGII (Vertex70, Invenio, IRcube). Because these spectrometers deliver single-sided interferograms, phase correction is a critical issue, so a robust phase unwrapping scheme and the option of using an analytical phase model have been developed and

implemented in the preprocessing tool of PROFFAST (activity lead: F. Hase). A publication describing the new scheme has been submitted to AMT.

The previous observations collected with KIT's EM27/SUN FTS operated in Sodankyla have been reprocessed using the latest version of PROFFAST (D. Dubravica). Moreover, the forward model of PROFFAST has been further refined (e.g. SDV line shape support, more accurate handling of airmass dependency, acceleration of cross-section calculation, ...). The calibration of Xgas results has been reviewed and readjusted for the latest PROFFAST version in order to achieve agreement with TCCON GGG2020 (Laughner et al., (2024)) data (Benedikt Herkommer, Jasmin Vestner). In the framework of this work we discovered that the slope of XCH₄ versus XH₂O has changed substantially between GGG2014 and GGG2020. In order to mimic the results of GGG2020, a further empirical degree of freedom - an explicit a-posteriori correction of XGAS vs XH₂O slope - has been implemented in PROFFAST and the required tuning parameters were adjusted.

Improvements to retrieval algorithm for LHR based on Optimal Estimation Method, OEM (RAL)

The LHR data level1 processor (L1P) has been updated and improved. Overall, the structure of the L1P has been streamlined and made robust enough to process both historical data and the updated instrument data. Relative spectral calibration has been improved by fitting a sinusoidal function to each calibration trace to provide a global fit to the etalon response. For absolute spectral calibration, a global fit to the atmospheric spectral traces has been applied. This fit includes the atmospheric absorption features (assuming a Voigt line shape) and a polynomial instrumental baseline. Together, these changes allow more robust spectral calibration with well-defined errors and quality parameters. This also allows the spectral baseline to be more reliably estimated (including variability) for input into the level 2 processor. The removal of non-spectral background contributions to the heterodyne signal was updated following the lessons learned from the experimental noise model. The quality control of the spectral data has been simplified to reflect a smaller number of more relevant inputs, including the intensity of the associated solar image and the pointing stability and QC metrics from the spectral calibration and background correction modules.

A novel level 2 processor, based on the RFM (Reference Forward Model) and the MORSE (Multispectral Orbital Retrieval using Sequential Estimation) retrieval developed for MIPAS, IASI, and CubeMAP missions is still being adapted to be tested on the Ground-based LHR. The first tests on actual data is planned to start on the 30th of September.

Investigate feasibility of retrieving OCS from the low-resolution MIR spectra using SFIT4 (Bremen)

Instrument:

- Portable Pendulum based FTIR Vertex-70. Spectral resolution = 0.4cm^{-1} to 0.16cm^{-1} .
- The number of spectra produced and house kept in the database are 60000 from automated measurement during 2019 campaign.

The retrieval procedure:

- The retrieval is performed using the latest SFIT4 v1.0.21.
- Tikhonov regularization is used for both OCS and procedure O3 profile gases.
- The WACCM/IRWG profile version is v7.
- interpolated from NCEP up to an altitude of 34km and WACCM/IRWG up to an altitude of 120km
- Spectroscopy used is HITRAN 2020. Also some tests are currently being performed using ATM19
- The isotopes considered are the following: H_2^{18}O and C^{18}O_2
- Other interfering gases considered: CO_2 , H_2O and CO
- Line mixing gas considered for CO_2 with the Boon et al (2012) LM scheme applied
- The spectra are processed for SFIT4 retrieval (conversion to bnr and creation of spectral database and profiles).

Retrieval Windows:

- The chosen retrieval is performed using multiple windows simultaneously:
- [2047.85, 2048.24], [2049.77, 2050.18], [2051.18, 2051.46] and [2054.33, 2054.67] cm^{-1} .

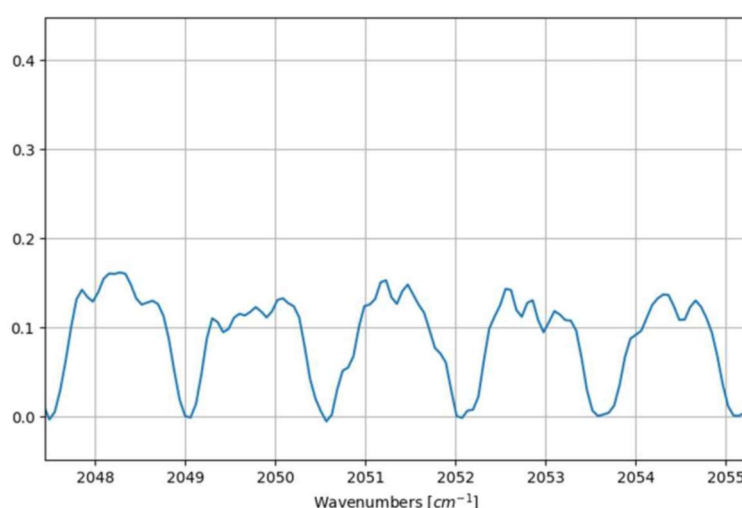


Figure: Spectral lines of OCS

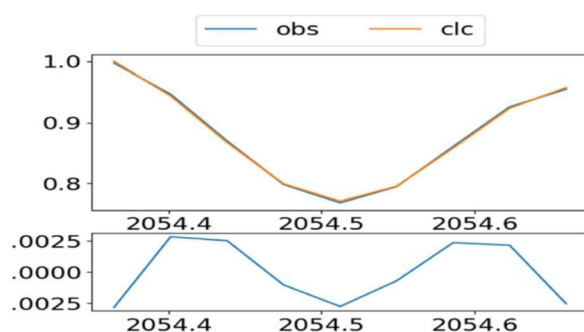


Figure: Example of a fitted OCS window

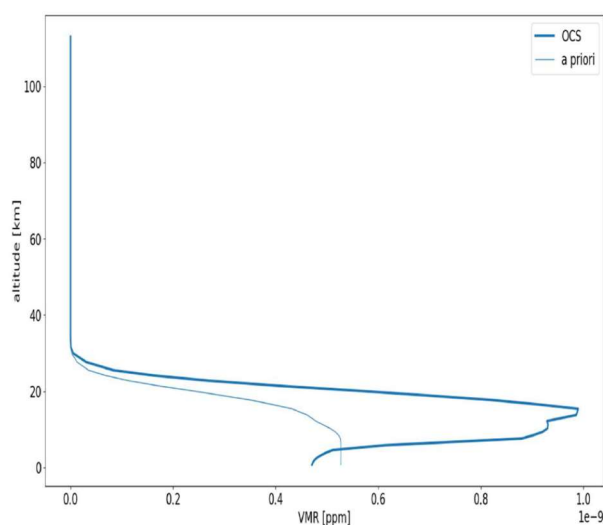


Figure: A-priori profile and resulting VMR profile for OCS

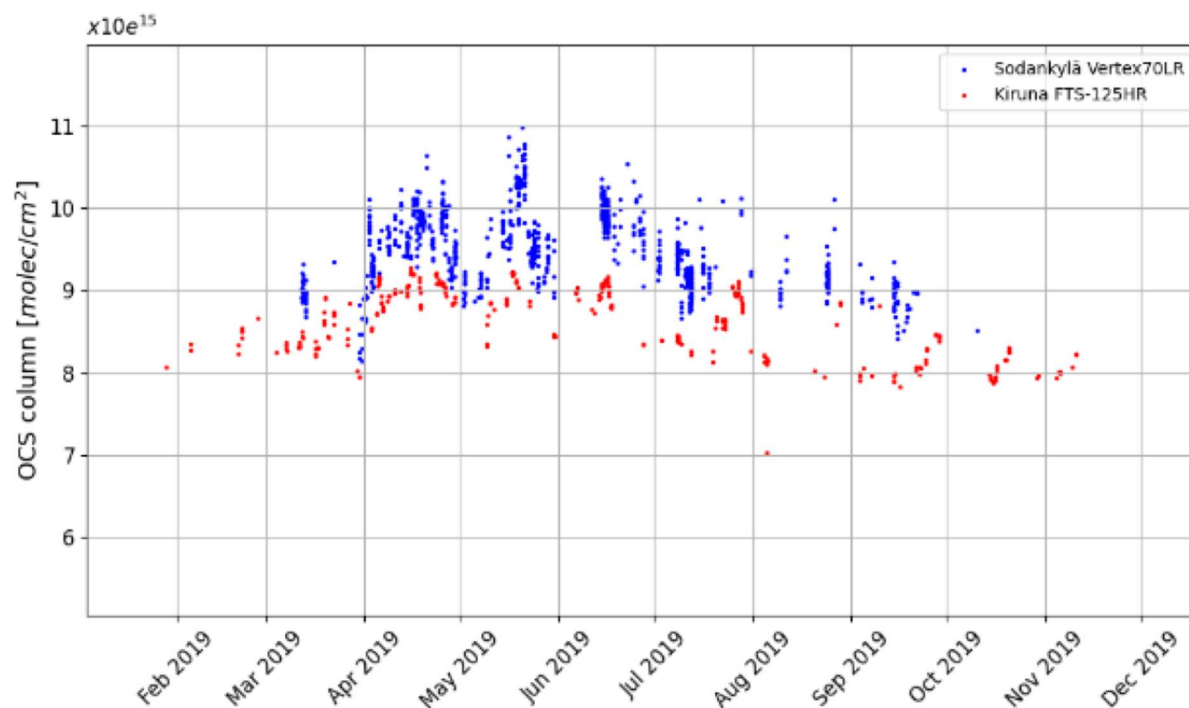


Figure: Time series of OCS measured in Kiruna with the FTS-125HR and Sodankyla with the Vertex70LR

The results are presented in a scientific paper on the topic of Fiducial Reference Measurement for Greenhouse Gases by Sha et al. (2024)

7 OUTLOOK

During the next 12 months the partners will work on

- The further development of GHG profiling using NIR and MIR spectra, for CO₂, CH₄, CO and N₂O
- Continue to develop AirCore for retrieving OCS and N₂O
- Update of the FRM4GHG dataset based on the latest version of PROFFAST and GGG

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

Laughner, J. L., Toon, G. C., Mendonca, J., Petri, C., Roche, S., Wunch, D., Blavier, J.-F., Griffith, D. W. T., Heikkinen, P., Keeling, R. F., Kiel, M., Kivi, R., Roehl, C. M., Stephens, B. B., Baier, B. C., Chen, H., Choi, Y., Deutscher, N. M., DiGangi, J. P., Gross, J., Herkommer, B., Jeseck, P., Laemmle, T., Lan, X., McGee, E., McKain, K., Miller, J., Morino, I., Notholt, J., Ohyama, H., Pollard, D. F., Rettinger, M., Riris, H., Rousogonous, C., Sha, M. K., Shiomi, K., Strong, K., Sussmann, R., Té, Y., Velasco, V. A., Wofsy, S. C., Zhou, M., and Wennberg, P. O.: The Total Carbon Column Observing Network's GGG2020 data version, *Earth Syst. Sci. Data*, 16, 2197–2260, <https://doi.org/10.5194/essd-16-2197-2024>, 2024.

Sha, M.K.; De Mazière, M.; Notholt, J.; Blumenstock, T.; Bogaert, P.; Cardoen, P.; Chen, H.; Desmet, F.; García, O.; Griffith, D.W.T.; et al. Fiducial Reference Measurement for Greenhouse Gases (FRM4GHG). *Remote Sens.* 2024, 16, 3525. <https://doi.org/10.3390/rs16183525>