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Comparison of one year of XCH_4 and XCO measurements using a EM27/SUN low resolution FTIR spectrometer to S5P/TROPOMI methane and carbon monoxide columns at Thessaloniki, Greece (revised results)

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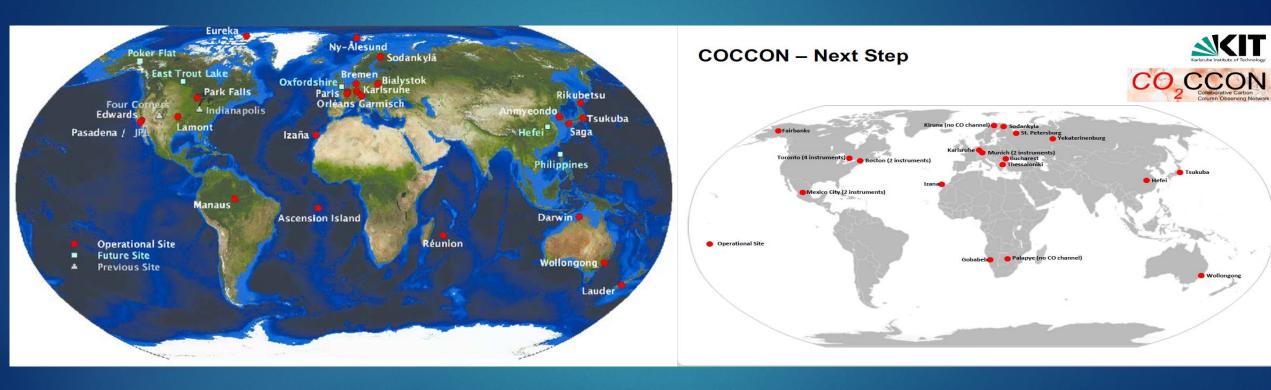
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¹Laboratory of Atmospheric Physics, Department of Physics, Aristotle University of Thessaloniki, Greece ²Institute of Meteorology and Climate Research (KIT-IMK), Karlsruhe Institute of Technology, Karlsruhe, Germany ³SRON Netherlands Institute for Space Research, Utrecht, TheNetherlands The Total Carbon Column Observing Network (TCCON) (Wunch et al., 2011) measures total columns of CO_2 and CH_4 , using Bruker IFS 125HR spectrometers. These high resolution instruments instruments are rather expensive and need large infrastructure to be set up and expert maintenance, which has to be performed on site. (Frey et al, 2019)

The **Bruker EM27/SUN** portable FTIR spectrometer (Gisi et al., 2011; Frey et al., 2015; Hedelius et al., 2016) developed by KIT in collaboration with Bruker Optics[™], is a promising instrument to overcome the above-mentioned shortcomings as it is a mobile, reliable, easy-to-deploy and low-cost supplement to the Bruker IFS 125HR spectrometer used in the TCCON network (Frey et al, 2019)

The **COllaborative Carbon Column Observing Network (COCCON)**, constituted by 60 EM27/SUN spectrometers, with stations around the globe, produces greenhouse gas observations based on common instrumental standards and data analysis procedures, working as an important supplement of TCCON for the quantification of local sinks and sources, to increase the global density of column-averaged greenhouse gas observations

TCCON- COCCON NETWORK



The Bruker EM27/SUN FTIR spectrometer





It consists of a spectrometer body with dimensions of 35x40x27cm and a solar tracker directly mounted on the spectrometer. It weighs approximately 25 kg and its design achieves high stability and is resistant to thermal influences and mechanical disturbances.

The Bruker EM27/SUN FTIR spectrometer

	Spectral window (cm-1)	Spectral resolution (cm-1)	Semi- FOV (mrad)	Detector	Retrieval software	Spectroscopy	A priori profiles & averaging kernels
02	7765-8005	0.5	2.26	InGaAs Spectral range:		HITRAN O ₂ : 2007 (Geoff	TCCON map files
CH_4	5897-6145	0.5	2.36	main channel standard		Toon's ATM line	Column sensitivities
СО	4208-4318			(5000 cm-1 low wavenumber cut off) CO channel extended (4000 cm-1 low wavenumber cut off).	PROFFAST (Sha et al. 2019)	list) H ₂ 0 : 2009 +empirical corrections CH ₄ : 2008 CO: 2012	from retrieval software

In this study, column-averaged dry-air mole fractions XCH₄ and XCO are presented for one year of measurements in Thessaloniki, Greece, an urban site $(40.5N^{\circ}, 22.9E, 60 \text{ m a.s.l})$. For a gas G:

$$X_{Gas} = \frac{Column_{Gas}}{Column_{dryair}}$$

Since O_2 's mixing ratio in the atmosphere can be considered stable

$$Column_{dryair} = \frac{Column_{O_2}}{0.2095}$$

the column-averaged dry-air mole fractions (DMF), denoted X_G for gas G, are computed using the retrieved O_2 columns as a measure of the dry air column

$$X_{Gas} = \frac{Column_{Gas}}{Column_{O_2}} \cdot 0.2095$$

Dividing by O_2 improves the precision of the measurement by significantly reducing the effects of instrumental or measurement errors that are common to both the gases (tracker problems leading to mis-pointing and zero-level offsets) or by errors the use of in the surface pressure

EM27/SUN measurements have undergone correction with air mass dependent (ADCF) and air mass independent (AICF) factors in order to bring COCCON data in agreement with TCCON (https://www.imk-asf.kit.edu/)

Species	MW	AICF $a_{\rm gas}$	ADCF $b_{\rm gas}$
XH_2O	(8353.4, 8463.1)	0.8300	0.000
XAIR	(7765.0, 8005.0)	0.9737	-0.007
XCO_2	(6173.0, 6390.0)	0.9862	0.005
XCH_4	(5897.0, 6145.0)	0.9905	-0.014
XCO	(4208.7, 4318.8)	0.9250	0.103
$XCH_4^{(b)}$	(4208.7, 4318.8)	0.9727	-0.017

(b) XCH₄ S5P (Sentinel-5 Precursor)

Xair is defined as

$$X_{\text{air}} = \frac{0.2095}{\text{VC}_{\text{O}_2} \cdot \overline{\mu}} \cdot \left(\frac{P_{\text{S}}}{g} - \text{VC}_{\text{H}_2\text{O}} \cdot \mu_{\text{H}_2\text{O}} \right),$$

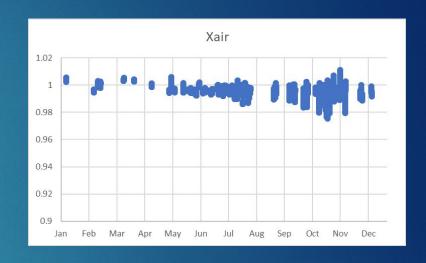
where μ and μ_{H20} are molecular masses of dry air and water vapor g is column averaged gravitational acceleration, Ps is the surface pressure and VCO₂, VCH₂O are the total columns of oxygen and water vapor

For an ideal measurement and retrieval with accurate O_2 and H_2O spectroscopy and surface pressure, Xair would be 1.



- TCCON measurements Xair is typically ~ 0.98 (Wunch et al., 2015).
- EM27/SUN measurements show a factor of ~ 0.97 (Frey et al., 2015; Hase et al., 2015; Klappenbach et al., 2015).

Large deviations (~ 1 %) from these values indicate severe problems, e.g., errors with the surface pressure, pointing errors, timing errors or changes in the optical alignment of the instrument. (Frey et al, 2019)



Xair at Thessaloniki passes TCCON standard quality check (between 0.96 and 1.04) with a mean value of 0.996 and a STD of ~ 0.003

Two methane products of EM27/SUN measurements:

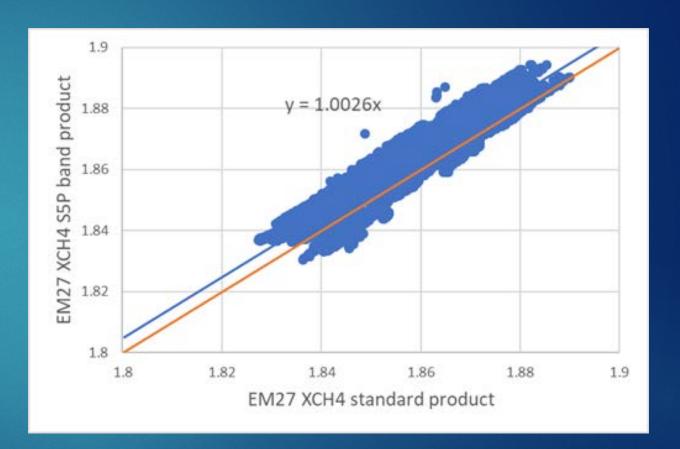
- in the 1.6µm band (standard product)
- in the S5P band (2.6μm) (not official)

are in good agreement, with a mean ratio of 1.0026 (between 0.993 and 1.012 and a std < 0.003)

Differences can be attributed to

- inconsistent spectroscopic linelists between bands
- different vertical sensitivity of the retrievals
- spectral interference of water vapor on the S5P band being stronger

A new linelist to be implemented into PROFFAST will be available soon, for an improved XCH₄ S5P band product (Framk Hase, personal



EM27/SUN comparison to TROPOMI

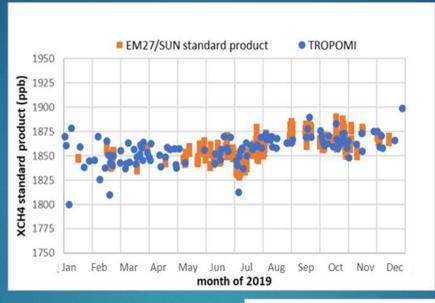
Methane	Carbon monoxide			
<i>TROPOMI</i> : XCH ₄ OFFL product, no bias-corrected, qa=100	<i>TROPOMI</i> : XCO OFFL product, qa=100			
EM27/SUN: XCH ₄ standard (1.6μm) XCH ₄ S5P using S5P band (2.3μm) Mission data requirements for XCH ₄ : 1.5% ±1%	<i>EM27/SUN</i> : XCO Mission data requirements for XCO: 15% ±10%			
100km & 1h co-location criteria ¹	50km & 1h co-location criteria ¹			
1 as used in TROPOMI Validation Reports				

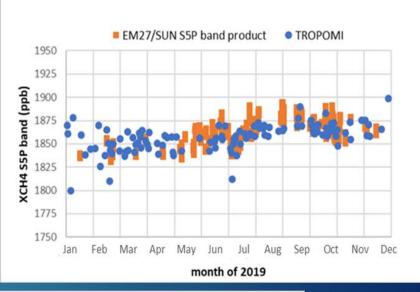
EM27/SUN vs TROPOMI full time series

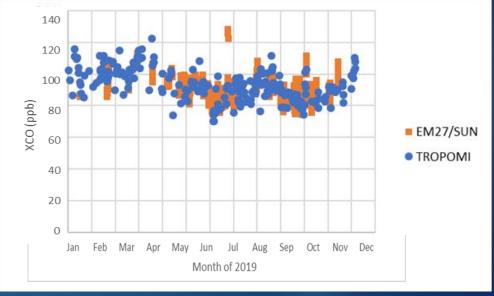
Examining the full time series with no spatial/time criteria, TROPOMI seems to show

- higher CO values compared to ground based measurements
- lower XCH₄ values than the standard ground based XCH₄ and almost similar to the S5P band XCH₄

Seasonal variations captured by EM27 are reproduced by TROPOMI



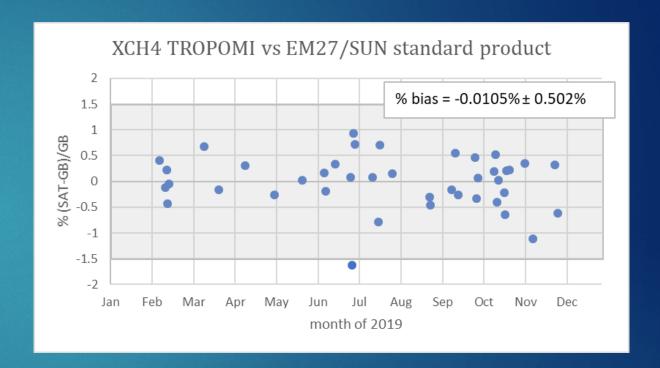




EM27/SUN vs TROPOMI (co-located measurements)

Methane (XCH₄ EM27/SUN standard product)





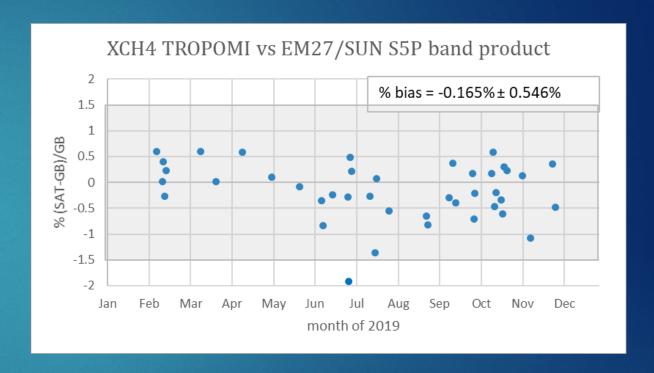
% bias = $-0.0105\% \pm 0.502\%$, well within mission requirements (1.5% ± 1.0%)

Results in agreement with those of the Validation Reports of the Copernicus Sentinel-5 Precursor Operational Data Products that show TROPOMI 's underestimation of XCH_4 vs NDACC and TCCON data with an overall agreement of -0.68% (negative bias) for not bias-corrected TROPOMI XCH_4 and a standard deviation of the relative bias is on an average 0.6%

EM27/SUN vs TROPOMI (co-located measurements)

Methane (XCH₄ EM27/SUN standard product)



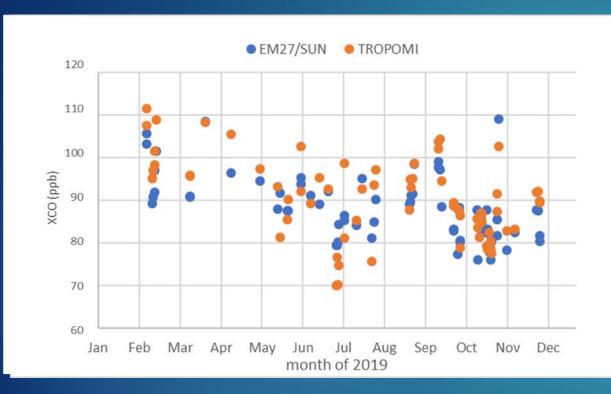


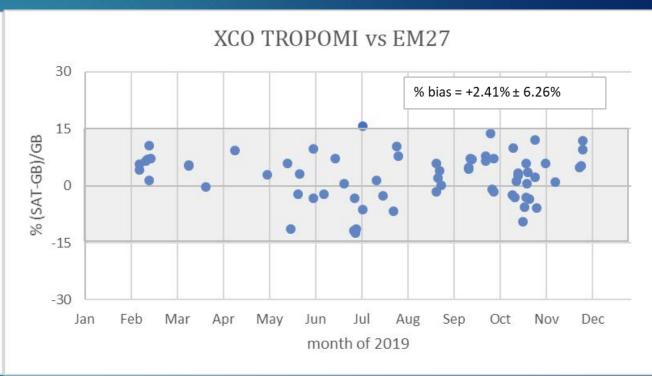
% bias = $-0.165\% \pm 0.546\%$, well within mission requirements (1.5% ± 1.0%)

Results in agreement with those of the Validation Reports of the Copernicus Sentinel-5 Precursor Operational Data Products that show TROPOMI 's underestimation of XCH_4 vs NDACC and TCCON data with an overall agreement of -0.68% (negative bias) for not bias-corrected TROPOMI XCH_4 and a standard deviation of the relative bias is on an average 0.6%

EM27/SUN vs TROPOMI (co-located measurements)

Carbon monoxide (XCO)





XCO % bias = $+2.41\% \pm 6.26\%$, well within mission requirements (15% \pm 10%)

Results in agreement with those of the Validation Reports of the Copernicus Sentinel-5 Precursor Operational Data Products that show TROPOMI 's overestimation of XCO vs NDACC and TCCON data with an overall agreement of 7-10% (positive bias) and a standard deviation of the relative bias is on an average 5%

EM27/SUN vs TROPOMI comparison for other distances

Distance (km)	XCH _{4standard} (1h, qa=1) %bias	XCO (1h, qa=1) % bias
30	-0.0946	+4.416
50	+0.2088	+2.411
100	-0.0105	+0.736

- As the distance from Thessaloniki overpass increases, the bias decreases demonstrating a lower value of TROPOMI
 average value for this area, probably caused by fewer CO sources in the surrounding area of Thessaloniki, which is a
 high polluted urban site.
- For methane, it is not clear whether conclusions can be drawn mainly due to a very low number of common measurement days between EM27/SUN and TROPOMI. A detailed recording of CO and CH₄ sources in the area of Thessaloniki and within 100km, could provide more insight

Summary

- The very first year of measurements of XCH₄ and XCO using a Bruker EM27/SUN low resolution spectrometer provided by Karlsruhe Institute of Technology, are presented for Thessaloniki, an urban site in Northern Greece.
 Our measurements show similar levels of XCH4 and XCO for similar sites
- Comparison to TROPOMI measurements show that seasonal variations are captured. Mission requirements are met for both the bias level and the standard deviation:

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\Rightarrow XCH<sub>4</sub> (1 hour, 100km) : % bias = -0.0105% ± 0.502% (within 1.5% ± 1.0%) \Rightarrow XCO (1 hour, 50km) : % bias = +2.41% ± 6.26% (within 15% ± 10%)
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(these are revised results compared to the positive TROPOMI bias for methane stated in the abstract)

- Data set will be re evaluated after incorporating improved parameters such as ME and PE for ILS function and new linelists for secondary XCH₄ products
- Important to establish a dense data set of ground based measurements in order to
 to confirm the comparison to TROPOMI in the future
 provide more common days for comparison to the satellite (especially for methane)
 explore variability of the measurements

References

Frey, M., Hase, F., Blumenstock, T., Groß, J., Kiel, M., Mengistu Tsidu, G., Schäfer, K., Sha, M. K., and Orphal, J.: Calibration and instrumental line shape characterization of a set of portable FTIR spectrometers for detecting greenhouse gas emissions, Atmos. Meas. Tech., 8, 3047–3057, https://doi.org/10.5194/amt-8-3047-2015, 2015

Frey, M., Mahesh K. Sha, Hase, F., Kiel, M., Blumenstock, T., Harig, R., Surawicz G., Deutscher, N.M., Shiomi, K., Franklin, J.E., Bösch, H., Chen, J., Grutter, M., Ohyama, H., Sun, Y., Butz, A., Mengistu Tsidu, G., Ene, D., Wunch, D., Cao, Z., Garcia, O., Ramonet, M., Vogel F., and Orphal, J., Building the Collaborative Carbon Column Observing Network (COCCON): long-term stability and ensemble performance of the EM27/SUN Fourier transform spectrometer, Atmos. Meas. Tech., 12, 1513–1530, 2019, https://doi.org/10.5194/amt-12-1513-2019

Gisi, M., Hase, F., Dohe, S., Blumenstock, T., Simon, A., and Keens, A.: XCO₂-measurements with a tabletop FTS using solar absorption spectroscopy, Atmos. Meas. Tech., 5, 2969–2980, https://doi.org/10.5194/amt-5-2969-2012, 2012.

Hasekamp, O., Lorente, A., Hu, H., Butz, A., Aan de Brugh, J., Landgraf, J.,: ATBD for Sentinel-5 Precursor Methane Retrieval, SRON-S5P-LEV2-RP-001

Hedelius, J. K., Parker, H., Wunch, D., Roehl, C. M., Viatte, C., Newman, S., Toon, G. C., Podolske, J. R., Hillyard, P. W., Iraci, L. T., Dubey, M. K., and Wennberg, P. O.: Intercomparability of XCO₂ and XCH₄ from the United States TCCON sites, Atmos. Meas. Tech., 10, 1481–1493, https://doi.org/10.5194/amt10-1481-2017, 2017

Tu, Qiansi, Observation of atmospheric greenhouse gas abundances on regional scales in boreal areas using portable FTIR Spectrometers, Dissertation

Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W. T., Sherlock, V., and Wennberg, P. O.: **The Total Carbon Column Observing Network**, Philos. T. R. Soc. A, 369, 2087–2112, https://doi.org/10.1098/rsta.2010.0240, 2011

Lambert, J.-C., S. Compernolle, K.-U. Eichmann, M. de Graaf, D. Hubert, A. Keppens, Q. Kleipool, B. Langerock, M.K. Sha, T. Verhoelst, T. Wagner, C. Ahn, A. Argyrouli, D. Balis, K.L. Chan, I. De Smedt, H. Eskes, A.M. Fjæraa, K. Garane, J.F. Gleason, F. Goutail, J. Granville, P. Hedelt, K.-P. Heue, G. Jaross, ML. Koukouli, J. Landgraf, R. Lutz, S. Nanda, S. Niemejer, A. Pazmiño, G. Pinardi, J.-P. Pommereau, A. Richter, N. Rozemeijer, M. Sneep, D. Stein Zweers, N. Theys, G. Tilstra, O. Torres, P. Valks, C. Vigouroux, P. Wang, and M. Weber., Quarterly Validation Report of the Copernicus Sentinel-5 Precursor Operational Data Products #06: April 2018 – February 2020, S5P MPC Routine Operations Consolidated Validation Report series, Issue #06, Version 06.0.1, 154 pp., March 2020.

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