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# COMPARISON OF CARBON MONOXIDE DATA PRODUCTS FROM SENTINEL-5PRECURSOR WITH LOW RESOLUTION FTIR SPECTROMETER

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*The TROPOspheric Monitoring Instrument, part of the European Space Agency's (ESA) Sentinel-5 Precursor satellite platform was launched in August 2017. The instruments goal is to globally monitor atmospheric greenhouse gases, trace gases and provide aerosols and cloud information. The carbon monoxide total column density is derived from the backscatter sunlight in the short-wave infrared region. This work presents the comparison of the satellite retrieval of total column CO with a low resolution, 0.5 cm$^{-1}$, FTIR, in peri-urban area near Bucharest, Romania for a period of two months in 2018*

**Keywords** : satellite, FTIR, validation, carbon monoxide.

## 1. Introduction

The anthropogenic activities are the main actions responsible for increasing in GHG (greenhouse gases) concentrations. The amount of GHG and other trace gases into the atmosphere is a result of complex interactions between emissions and processes like chemical reactions, biological transformation and also physical conditions that remove the gases from the atmosphere. The long-term assessment of trace gases and GHG shows a significant increase of concentrations over the past 800,000 years, the increasing rate in the past century exceeding any recorded rate in the previous 20,000 years [1]. The accurate measurements with high spatial and temporal resolution of trace gases and GHG is a stringent necessity nowadays taking into account the continuous changes of anthropogenic activity (industry, land use, agricultural practices etc.) [2] [3] with significant discrepancies between northern and southern hemisphere. The differences in CO concentration between

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northern and southern latitudes during wintertime can reach values of 135 ppb, while in summer, in southern part, concentration is less than half compared with high northern latitudes [1]. Even though the carbon monoxide is atmospheric molecule with a short lifetime, there are high uncertainties associated with its global distribution related to the increase gradients into the atmosphere. CO has not a direct role in a radiative budget because of low absorption of terrestrial infrared radiation, but even so CO is a constituent of $O_3$ cycle and influence the $CH_4$ budget [4].

Currently, the most used technique to monitor column density of CO, $CH_4$, $H_2O$ or other trace gases concentrations are based on Fourier Transform Infrared principle, several types of spectrometers being available. For this study, a low resolution ground-based Fourier Transform Infrared (FTIR) spectrometer measuring direct solar spectra [5] was used to retrieve column density of CO for the purpose to be compared with space satellite CO measurements derived from the new available products of Sentinel-5Precursor observations.

The long term measurements with high regional coverage of carbon monoxide concentration will allow the confirmations of atmospheric patterns of air pollution and also the determination of sources and sinks. Combined measurements and retrievals from ground based instrumentation and from satellite data will assure additional input data and validations tools for the models purposes. In this paper the EM27/SUN spectrometer, developed by Karlsruhe Institute of Technology (KIT) in collaboration with Bruker Optics™, was used to acquire solar spectra and retrieve CO column density, the results being compared with CO retrieval obtained from Sentinel 5-Precursor for a period of two months (August – October 2018) in a peri-urban area in Magurele, Romania (44.348$^o$N, 26.030$^o$E, 93 m above sea level).

Comparable with satellite methods the ground based column measurements are sensitive to local sources, but in the same time can be a suitable method to validate long term satellite measurements, correlated with the meteorological parameters like atmosphere stability or wind directions.

Even the method is not an automatically and have limitation related with solar radiation, it has a high advantage because column measurements is sensitive to long range transport pollution that are not recorded by equipment that measure in situ and are less comparable with satellite observations.

Previous validation were made with a high-resolution FTIR spectrometers as part of the Total Carbon Column Observing Network (TCCON) [7]. Comparison with the high resolution system showed a mean bias of 6 ppb for both clear-sky and cloudy observations and a mean standard deviation of 3.9 and 2.4 ppb [19].

## 2. Instruments and methodology

### 2.1. Low resolution FTIR

For this study a low resolution FTIR was used, that is based on a Michelson Interferometer. The interferometer contains a semi-transparent mirror which is used as a beam splitter that divides the incoming radiation into two partial beams and leads them into the interferometer arms. At the end of the interferometer arms there are cube corner mirror (mirrors that are insensitive to tilt) that reflects back the radiation into the beam splitter. One of the mirrors is static while the other can move so the optical path between the partial beams can vary. The beam splitter thus also acts as a recombiner, resulting in constructive and destructive interference patterns in the recombined beam. The intensity of the recombined radiation due to interference is then recorded by the detector (Fig. *1*).

The interferogram, which is the AC part of the recorded interference pattern, depends on the optical path difference (OPD). The recorded spectrum and the interferogram are connected using the Fourier Transformation (FT):

$$S(\tilde{\nu}) = F\big(I(x)\big) = \int_{-\infty}^{\infty} e^{-i\,2\pi\bar{\nu}x} I(x)dx \tag{1}$$

$$I(x) = F^{-1}\big(S(\nu)\big) = \int_{-\infty}^{\infty} e^{i\,2\pi\bar{\nu}x} S(\tilde{\nu})d\tilde{\nu} \tag{2}$$

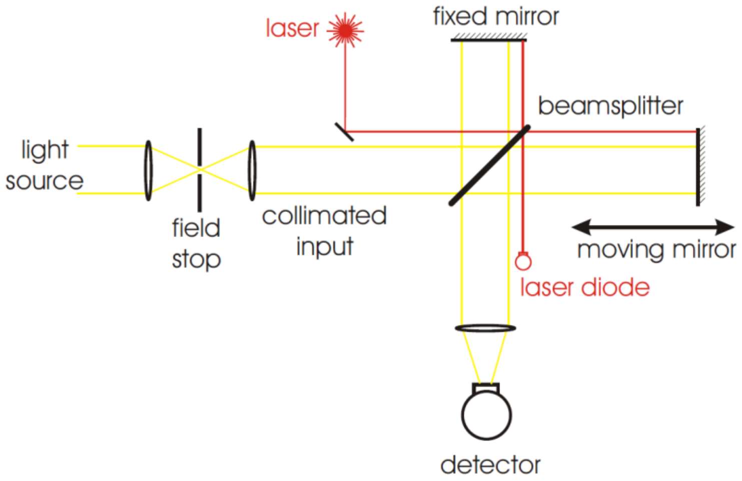

Fig. 1 Schematic drawing of the Michelson Interferometer used by the FTIR [6]. The laser diode is used for the precise determination of the position of the moving mirror

Where $S(\tilde{\nu})$, with $\tilde{\nu} = 1/\lambda$, denotes the spectrum and $I(x)$ represents the interferogram and *x* is the OPD.

For the instrument used in this work, the recorded interferogram is neither infinite nor continuous. Instead, one has $N$ equidistant discrete sampling points with distance $\Delta x$ recorded and equations (1) and (2) become:

$$S(\widetilde{\nu_m}) = \sum_{n=1}^{N} I(x_n) e^{-i\,2\pi\overline{\nu_m}x_n} \tag{3}$$

$$I(x_n) = \frac{1}{N}\sum_{m=1}^{N} S(\widetilde{\nu_m})e^{i\,2\pi\overline{\nu_m}x_n} \tag{4}$$

The maximum path difference of the instrument is realized by movable mirror in the interferometer, and therefore the size of the interferogram is finite and limited to the maximal OPD. The spectral resolution of the instrument is derived from its OPD by using the approximation [6]

$$resolution = \frac{0.9}{OPD} \tag{5}$$

The retroflectors move a geometrical distance of 0.45 cm leading to an OPD of 1.8 cm which corresponds to a spectral resolution of 0.5 $cm^{-1}$. This is considered a low-resolution instrument compared with high resolution instrument from the Total Column Carbon Network (TCCON) [7]. The instrument has an off-axis mirror with a focal length of 1.27 cm for centering the solar beam on the detector and, together with the field stop of 0.6 mm leads to a semi field of view (FOV) of 2.36 mrad which results in an external FOV of about 56% of the apparent solar disc diameter [8]. The measurements are recorded using two detectors, InGaAs diodes with a spectral coverage from 5000 to 11 000 $cm^{-1}$ and the second has a cut-off at 4000 $cm^{-1}$.

### 2.2. Sentinel-5 Precursor

The Sentinel-5 Precursor (S5P) mission is a single-payload satellite in a low Earth orbit that provides daily global information on concentrations of trace gases (ex. $NO_2$, $SO_2$, etc.), greenhouse gases (CO, $CO_2$) and aerosols important for air quality, climate forcing, and the ozone layer. The payload of the mission is the TROPOspheric Monitoring Instrument (TROPOMI) [9] which consists of a spectrometer with spectral band in the ultraviolet, the visible, the near-infrared and the shortwave infrared. The selected wavelength range for TROPOMI allows observation of key atmospheric constituents, including ozone, nitrogen dioxide, carbon monoxide, methane, aerosols and clouds.

The TROPOMI instrument is a nadir-viewing hyperspectral imager with four separate spectrometers covering non-overlapping and non-contiguous wavelength bands between the above-mentioned spectral regions and has a ground pixel area of 7 x 7 $km^2$ implying global coverage of the Earth's surface day by day. The instrument uses passive remote sensing techniques by measuring at the top of the atmosphere the solar radiation reflected by and radiated from the Earth. Light from the entire swath is recorded simultaneously and dispersed onto two-dimensional imaging detectors, the position along the swath is projected onto one direction of the detectors, and the spectral information for each position is projected on the other direction. From the spectra obtained, the information on the total column densities and vertical profiles of atmospheric trace gases like $NO_2$, $SO_2$,

$CO_2$, CO, $CH_4$, and O3 is retrieved. In addition, information relating to clouds, surface reflectance and aerosols can be derived.

The retrieval of CO total column density approach is based on the profile scaling method [10] and the implementation and retrieval settings are according to [11]. The reference profile of CO that is used for in the processing chain is taken from the chemical transport model TM5 [12], which is averaged over $3^o$ x $2^o$ latitude and longitude. Final result represents the total column density of CO measured in molec x $cm^{-2}$. In order to compare satellite retrievals with ground based measurements, the dry air column mixing ratio is represented XCO, by dividing the CO total column density by the dry air column density derived from co-located European Center for Medium-Range Weather Forecast (ECMWF) pressure fields [13].

## 3. Results and discussions

### 3.1. Column mixing ratios of carbon monoxide from ground based and satellite retrievals

To retrieve atmospheric trace gas abundances, the recorded spectra are analyzed with a software program which applies a model of the radiative transfer in the Earth's atmosphere to generate an atmospheric spectrum from a set of relevant input parameters (forward model). These contain atmospheric variables such as temperature, pressure and various volume mixing ratios (VMRs), auxiliary data such as the solar zenith angle (SZA) and the instrumental line shape (ILS) parameters and spectral information of the relevant gases, most often provided by line lists such as HITRAN 2008 [14].

First, the recorded interferograms are Fourier transformed using the Norton-Beer medium apodization function [8]. The apodization reduces the sidelobes around the spectral lines and a DC-correction is performed. Also is applied a quality filter over the interferograms which discards the ones that have an intensity fluctuation above 10% and intensities below 10% of the maximum modulation amplitude. Typical recorded spectra are shown in Fig. *2*.

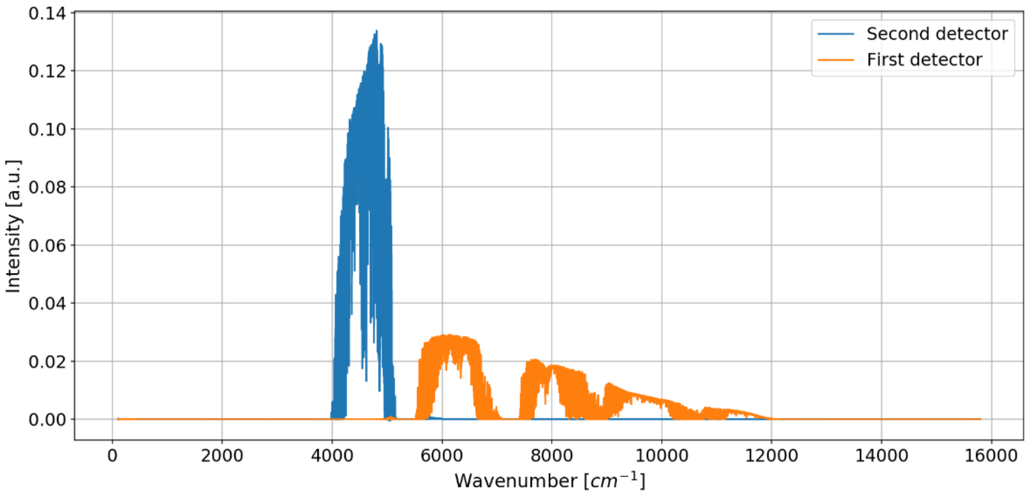

Fig. 2 Typical spectra recorded with FTIR (29.08.2018 07:14 UTC)

A second part of the analyzing software calls the forward model repetitively and reconstructs the profile shape of the initial one with the measured spectrum within spectral window 4210 – 4320 cm$^{-1}$. Example of the fitted spectra for 29.08.2018, 07:14h UTC is shown in Fig. 3. The method involves varying a selected set of variables to a priori trace-gas profiles. As source of the input profiles, the WAACM ver.6 climatology [15] is used, which is part of the PROFFIT software used successfully by TCCON [7] and tested by Buchwitz et.al. [16].

PROFFIT is a nonlinear fitting algorithm which analyses solar absorption spectra based on inversion methods and is utilized for a full profile retrieval using inverse covariance. On the selected spectral windows, it is applied a constrained profile retrieval. The a priori volume mixing ratio profile is obtained from the Whole Atmospheric Chemistry Climate Model (WAACM). For intra-day variability, pressure and temperature profiles from NCEP based on one profile at 12UTC for the given location are considered.

Final product is represented by the vertical column density [mol cm$^{-2}$] of the interested gas which is the integrated value of the fitted CO vertical profile.

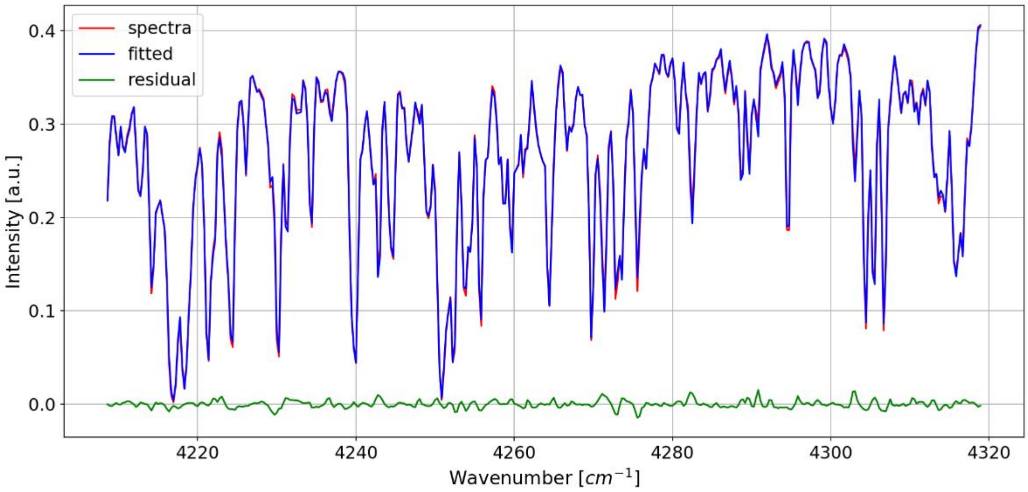

Fig. 3 Example of fitted spectra for CO column density (29.08.2018, 07:14h, UTC)

The vertical column density represents the number of molecules above the surface while column mixing ratio, $X_\mu$ of molecule $\mu$ is related to the total column according to following formula:

$$N_\mu = f_\mu N_{air}^{wet} \tag{6}$$

Where $N_{air}^{wet} = \sum\left(N_\mu + N_{H_2O}\right) = N_{air} + N_{H_2O}$ is the vertical column density of wet air. It is necessary to focus on dry column mixing ratio rather than the wet mixing ratios because dry mixing ratio is a better tracer, not being subject to strongly varying $H_2O$ variations. The mole fraction and the dry air mole fraction are associated according to:

$$\sum_\mu f_\mu + f_{H_2O} = 1 \tag{7}$$

$$\sum_\mu d_\mu = 1 \tag{8}$$

$$f_\mu = d_\mu\left(1 - X_{H_2O}\right) \tag{9}$$

Relation $\bar{m} = m_{air}\left(1 - X_{H_2O}\right) + f_{H_2O} m_{H_2O}$ accounts for this by correcting the mass for dry air mixing ratio with the fraction of the $H_2O$ mixing ratio, as $H_2O$ contributes less to the surface pressure $p$ on a per molecule basis than air. Rewriting equation (6) [13] we obtain the following equation:

$$X_\mu = N_\mu \left(\frac{p}{m_{air} g} - N_{H_2O}\frac{m_{H_2O}}{m_{air}}\right)^{-1} \tag{10}$$

Where $p$ is the surface pressure, $m_{air} \approx 28.96\ g\ mol^{-1}$ is the molar mass of air, $m_{H2O} = 18.02\ g\ mol^{-1}$ is the molar mass of $H_2O$ and $g$ is the gravitational constant.

For the ground-based Fourier transform spectrometer it is assumed a dry mole fraction of $d_{O2} = 0.2095$, this is equivalent to the dry air column mixing ratio. Rationing the interested gas, $\mu$, and $O_2$ equations eliminates the denominator in Equation (9) to make $X_\mu$ independent of surface pressure and gravity, leading to [7]:

$$XCO = \frac{N_{CO}}{N_{O_2}} \times 0.2095 \tag{11}$$

All retrieved columns share systematic errors as they are measured and calculated by same instrument. Normalizing them in this way reduces some of the systematic errors.

Because the consistency of retrieved $O_2$ and measured surface pressure is sensitive to the stability of the system, one can compare the surface pressure derived from the column densities of oxygen and water vapor with a co-located pressure sensor. This represents a check-up of data quality and long-term stability of the instrument. The following relationship is used to derive the surface pressure from the FTIR using Equation (10) :

$$\mathrm{p} = \left(\frac{N_{O_2}}{0.2095} \cdot m_{air} + N_{H_2O} \cdot m_{H_2O}\right) \cdot g \cdot \exp\left(-\frac{\Delta h}{h_s}\right) \tag{12}$$

Where $\Delta h$ is the height difference between the FTIR and the pressure sensor used and $h_s$ is the scaling height.

The systematic factor found for the analyzed period of time was 1.066 with a variability of 0.002 (Fig. *4*). This low value of the variability it is an indicator of the systems stability. The systematic factor origin can mainly be attributed to oxygen line intensity errors [17].

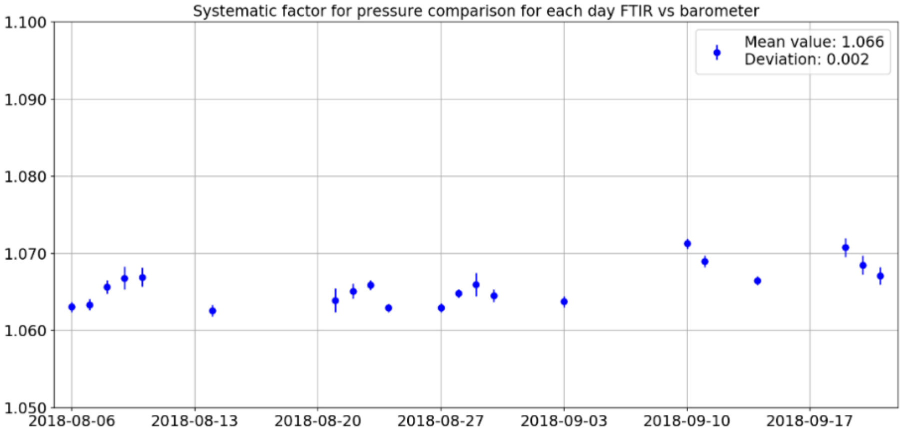

Fig. 4 Systematic factor for pressure comparison between FTIR retrievals and barometric data

To ensure data quality of a low-resolution instrument and to be able to be comparable with the high resolution systems it was performed a 5 days campaign of co-located measurements between the high and low resolution systems. From this, a calibration factor was derived for each of the products for the low-resolution instrument (not shown). The relative bias found is considered afterwards in the post-processing procedure.

The error budget of each data point retrieved with the FTIR is based on the assumption of different factors that can influence the systems performance. This includes the laser sampling errors, zero level offsets, instrument line shape deviation, smoothing error, changes in the a priori profiles of pressure, temperature or carbon monoxide, in situ calibration and random noise. Together with the empirical factor determined by direct comparison with the high resolution FTIR, the error budget was 7.53% for column densities of carbon monoxide.

Regarding the processing of satellite data from the TROPOMI in the Shortwave Infrared (SWIR) region, the algorithm used is a further development of the retrieval method described by Vidot et.al. [18] and includes a significant improvement with respect to the description of atmospheric scattering. The algorithm requires several input fields such as:

- The measured Earth radiance and solar irradiance spectra including noise estimate, solar and viewing geometry and geo-location information;
- Temperature, water-vapor and pressure profiles and geo-potential height at the geo-location, for TROPOMI these profiles are provided by the ECMWF;
- An estimate of the $CH_4$ field using a chemistry transport model, here Transport Model 5 (TM5) is used [12];
- And estimate of the CO column from a chemistry transport model, here TM5 is used.

The retrieval is performed in two steps, first as part of the SWIR pre-processing module, the vertically integrated amount of the methane is retrieved from a dedicated fit window of the SWIR band between 2315 nm and 2324 nm using a non-scattering radiative transfer model. If the measurements are strongly affected by clouds and/or aerosols they are associated with a lower quality flag. The criteria is based on the comparison of the retrieved $CH_4$ with the a priori $CH_4$ information. If their difference exceeds a certain threshold then the retrievals are considered affected by clouds, therefore rejected. In the second step, the methane absorption features are used to infer information on atmospheric scattering by clouds and aerosols, which passed the cloud filter, together with the atmospheric CO and $H_2O$ abundances, surface albedo and a spectral calibration of the reflectance spectrum. The scattering layer has a triangular height distribution of fixed geometrical thickness, and its optical depth and height are parameters to be retrieved.

Finally, the retrieved level 2 product consists of a CO column estimate including a column averaging kernel and a random error estimate [11]. The dry-air mixing ratio of carbon monoxide is derived using the co-located ECMWF pressure field according to Equation (10).

### 3.2. Data selection ground based and satellite retrieval

Two months of data were selected for the direct comparison between ground based retrieval of column mixing ratios of CO and satellite measurements from TROPOMI between August and October 2018. The ground based instrument is located in a peri-urban area near Bucharest, Romania (44.35N, 26.03E, 93m asl).

All measurements were taken under clear sky conditions with the solar zenith angle below 80 deg, using the same instrumental parameters.

Fig. 5 includes the time series of CO column mixing ratios retrieved with the FTIR (shade of blue means the whole time series of the day while the dark blue means the selected points for comparison with the satellite retrievals), the red point represents the mean value of the selected period of time with its associated variability. The yellow points represent the satellite retrievals over a selected area around the ground based instrument and the green point represents its mean value with its associated precision. The error bars of the carbon monoxide mixing ratios retrieved from FTIR represents the error budget described in Sect. 3.1. The selected period of time from ground-based measurements have a difference of 1 hour before and after the time of the satellite overpass.

In this example (Fig. 5), the average column mixing ratios for the satellite retrieval was 104.97 ppb with a spatial variability of 3.29 ppb while the ground based instrument shows a column density of 116.95 ppb with a temporal variability of 1.06 ppb and the difference in the mean values shows an overestimation of ground based measurements of 11.41 ± 1.14%.

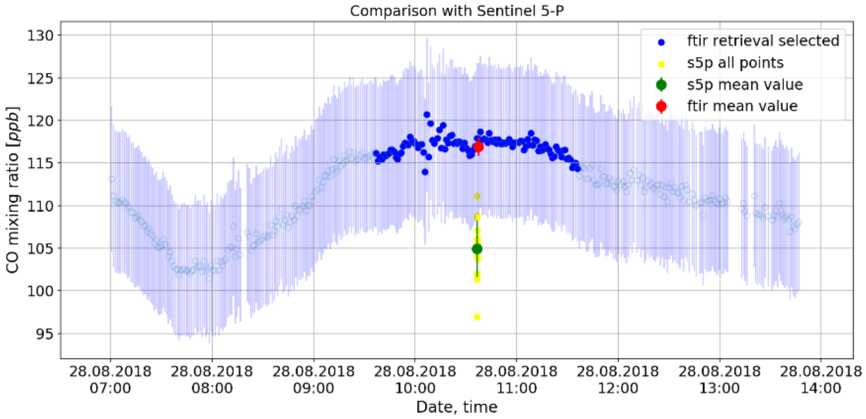

Fig. 5 Time series of CO mixing ratio. Data selection criteria (time)

Satellite data were selected by considering all the retrieval points around a 30 km radius around the ground-based location for each time of the overpass. The area is selected so that, the influence of the local sources of CO to be minimized. Also the time period selected from the ground based measurements are chosen so that the local sources to be minimized. Fig. 6 shows the TROPOMI overpass over

Romanian map (semi-transparent color) and the selected data from satellite retrieval (opaque color).

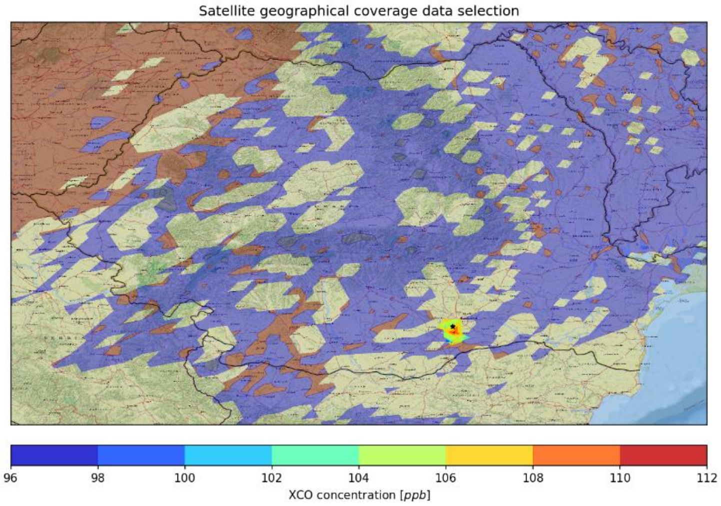

Fig. 6 Romanian map with satellite retrievals and the area around the ground based location for 28.08.2018

Fig. 6 shows higher concentrations on the south side of the city. These can be explained by the main sources of carbon monoxide, which are traffic and industrial activities.

A number of 20 days and 31 overpasses of the satellite are analyzed in the period of time described above. Fig. *7* shows the scatter plot between satellite mean values and ground based FTIR instrument mean values for each satellite overpass.

For the period analyzed, the mixing ratios retrieved from ground-based instrument were between 81.38 ppb and 126.54 ppb as for the satellite retrievals, the column mixing ratios ranged from 83.19 ppb to 115.51 ppb.

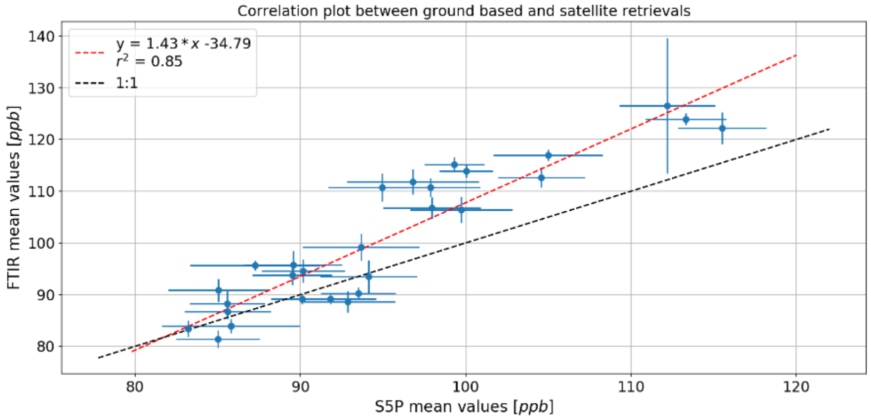

Fig. 7 Scatter plot between satellite overpasses and ground based measurements

The overall comparison between ground based and satellite retrieval show an average overestimation of ground based of 5.52 ± 0.64% with ranges from -4.7% to 14.2% and the 'r squared' value of 0.85. According to the satellite accuracy requirements the bias should not exceed 15% in the long-term analysis [19].

## 4. Conclusions

Total column carbon monoxide densities were derived from a low resolution (0.5 $cm^{-1}$) FTIR in a peri-urban area near Bucharest, Romania for a period of two months in 2018 and compared with satellite retrieval from Sentinel-5 Precursor.

To have a good data comparison for both methods satellite and ground based techniques several criteria were taking into account to reduce the input of punctual spots:

-time period of FTIR CO retrieval was selected based on average time of ± 1h around sentinel over passing hour in order to diminish the influence of possible local sources like a traffic or local fires, wind directions.

-data collection from S5P, to have an appropriate representativeness, was made taking into account the administrative Romanian map. This was a necessary step considering the Bucharest city proximity. The covered area considered for the satellite data was based on a radius of 30 Km around the ground-based location in order to reduce the uncertainty associated with punctual spots like roads or city influences

The difference between ground-based data measurements and satellite data retrieval are in range of 5.52 ± 0.64% that represent a good agreement according to satellite validation requirements and considering the fact that this work presented a low resolution ground based instrument. The overestimation can be explained by line of sight of the ground-based system and local sources such as traffic.

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