Mapping carbon monoxide pollution from space down to city scales with daily global coverage

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Abstract. On 13th October, 2017, the European Space Agency (ESA) successfully launched the Sentinel-5 Precursor satellite with the Tropospheric Monitoring Instrument (TROPOMI) as its single payload. TROPOMI is the first of ESA's atmospheric composition Sentinel missions, which will provide complete long-term records of atmospheric trace gases for the coming 30 years as a contribution to the European Union's Earth Observing programme Copernicus. One of TROPOMI's primary

5   products is atmospheric carbon monoxide (CO). It is observed with daily global coverage and a high spatial resolution of $7x7$ km^2. Due to its moderate atmospheric residence time, its atmospheric abundance provides information on both localized pollution hot spots and the pollutant transport on regional to global scales. In this contribution, we demonstrate the game-changing performance of the TROPOMI CO product, sensing CO enhancements above cities and industrial areas and tracking, with daily coverage, the atmospheric transport of pollution from biomass burning regions. The CO data product is validated

10 with two months of Fourier-transform spectroscopy (FTS) measurements at nine ground-based stations operated by the Total Carbon Column Observing Network (TCCON). We found a good agreement between both data sets with a mean bias of 6 ppb for both clear-sky and cloudy TROPOMI CO retrievals. Together with the corresponding standard deviation of the station-to-station bias of 3.9 ppb for clear-sky and 2.4 ppb for cloudy-sky, it indicates that the CO data product is already well within the mission requirement.

1 Introduction

The Sentinel-5 Precursor (S5P) satellite was successfully launched on 13th October, 2017, from Plesetsk in northern Russia with the Tropospheric Monitoring Instrument (TROPOMI) aboard. The instrument is a grating spectrometer which measures sunlight reflected by the Earth's atmosphere and its surface from the ultraviolet to the shortwave infrared (SWIR) with daily global coverage, a spatial resolution of about 7x7 km^2, and a high radiometric performance (Veefkind et al., 2012). One of the primary targets of the mission is to monitor the atmospheric concentration of CO. This trace gas is emitted to the atmosphere by incomplete combustion e.g. by traffic, industrial production, and biomass burning. Its major sink is the reaction with the OH radical (Spivakovsky et al., 2000). With a low background concentration and an atmospheric residence time from days to months (Holloway et al., 2000), the trace gas is established as a tracer of how pollution is transported, redistributed, and depleted in the atmosphere.

The S5P mission builds upon the heritage of SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography; Gloudemans et al. (2009); Frankenberg et al. (2005); Buchwitz et al. (2007); Gimeno Garcia et al. (2011); Borsdorff et al. (2017a)), which retrieved atmospheric CO total column concentration from the same spectral range. From space, CO is also measured by other satellite instruments with global coverage, e.g. MOPITT (Measurements of Pollution in the Troposphere; Deeter (2003)), AIRS (Atmospheric Infrared Sounder; McMillan (2005)), TES (Tropospheric Emission Spectrometer; Rinsland et al. (2006)), IASI (Infrared Atmospheric Sounding Interferometer; Turquety et al. (2004)). The S5-P mission is the first of a sequence of ESA's atmospheric composition satellites, which also comprises the Sentinel-5 mission, a series of spectrometers with the first launch in the 2021-2023 time frame.

In this study, we use the Shortwave Infrared CO Retrieval Algorithm (SICOR). It is developed by SRON the Netherlands Institute for Space Research, for the operational processing of TROPOMI data (Landgraf et al., 2016a, b) and also serves as algorithm baseline for the data processing of the successor mission Sentinel 5. The algorithm infers the vertical column concentration of CO (the vertically integrated amount of CO above the surface) simultaneously with effective cloud parameters from TROPOMI's 2.3 μm spectra (Vidot et al., 2012; Landgraf et al., 2016b). A first comparison of the TROPOMI CO data product with CO fields from the European Center for Medium-Range Weather Forecast (ECMWF) was performed by Borsdorff et al. (2018). In this study, we deployed SICOR on TROPOMI measurements taken during the early months of the instrument in orbit, and show the capability of the instrument to detect and monitor the air pollution from hot spots like larger cities and industrial regions. Investigating the temporal evolution of CO enhancements over the Atlantic and Indian oceans shows the capability of the instrument to track the atmospheric transport of pollution on a day-to-day basis in agreement with co-located ground-based measurements. Moreover, a validation with collocated ground-based Fourier Transform Spectrometer (FTS) measurements at nine TCCON sites, indicates the TROPOMI CO data quality. The paper is structured as follows: Section 2 describes the data set and methodology and Sec. 3 presents our analysis of the TROPOMI CO data product comprising a first validation with TCCON ground-based measurements, the detection of CO hot spots and the transport of CO pollution over the oceans. Section 4 gives the conclusions of the study and finally the data availability is described in Sec. 5.

2 Data set and methodology

For this study, we used nominal Earth radiance and Solar irradiance measurements of TROPOMI during the commissioning phase of the instrument from 9th November, 2017 to 4th January, 2018. We deployed the SICOR algorithm on the 2.3 μm spectra of TROPOMI and retrieved the total column density of CO simultaneously with interfering trace gases and effective
5   cloud parameters (cloud height and optical thickness) describing the cloud contamination of the ground scene (Landgraf et al., 2016b). The retrieval approach is based on the profile scaling method (Borsdorff et al., 2014) and the implementation and retrieval settings are discussed in detail by Landgraf et al. (2016a).

For the data analysis, we performed *a posteriori* quality filtering of the TROPOMI data. To this end, we used retrievals with a solar zenith angle < 80 degrees and discarded the two most westward ground pixels of the swath due to a performance issue
10   that is still under investigation. Furthermore, we distinguished between retrievals under clear-sky (cloud optical thickness < 0.5, cloud height < 5000 m, over land) and cloudy condition (cloud optical thickness > 0.5, cloud height < 5000 m over land and ocean). Clear-sky observations over ocean, which is a dark surface in the SWIR, cannot be used for data interpretation because of a too low signal.

The TROPOMI instrument is still in the early phase of the mission and the performance of the CO retrieval is expected
15   to improve in the future. For example, single overpasses show stripes of erroneous CO values < 5% in the flight direction, probably due to calibration issues of TROPOMI. Considering high-frequency variations of CO measurements across flight direction per orbit, we infer the stripe pattern by median filtering of the detected features in flight direction. Subsequently, the stripe pattern is removed from the data. Boersma et al. (2011) suggested a similar approach to improve the quality of the NO$_2$ data product of the Ozone Monitoring Instrument (OMI, Levelt et al. (2006)). This aspect will be the subject of future
20   investigations of the instrument calibration key data.

3 Results

3.1 Validation with TCCON ground-based measurements

The quality of the TROPOMI CO data product needs to be validated with independent reference observations both for clear-sky and cloudy TROPOMI measurements. To this end, we performed a first validation with CO observations at nine ground-based
25   FTS stations operated by the TCCON network (see Table 1). We selected sites in both the northern and southern hemisphere at low and high elevation on the continents and islands (Hase et al., 2015; Sussmann and Rettinger, 2014; Wennberg et al., 2016, 2015; Blumenstock et al., 2014; Feist et al., 2014; De Mazière et al., 2014; Sherlock et al., 2014). For the comparison, we used TROPOMI observations co-located with the TCCON sites by selecting all TROPOMI retrievals from the same day within a radius of 50 km around each station. The retrieved CO column of TROPOMI is adapted to the altitude of the station
30   by either cutting off the scaled mixing ratios profile at the station altitude or extending it assuming a constant elongation of the mixing ratio to lower altitude. Finally, we calculated daily averages of the dry air mixing ratios (XCO) using the adapted TROPOMI retrievals and the TCCON measurements shown in Fig. 1 and Fig. 2. Data gaps in the TROPOMI time series are

partly caused by discarding observations with high clouds (>5000 m) but also due to observation time reserved for in-orbit instrument characterization during the instrument commissioning phase.

Figure 3 depicts the corresponding bias for each TCCON station for clear-sky and cloudy-sky conditions and the combination of both, as well as the standard deviation and the number of coincident daily mean values of TROPOMI and TCCON.

5   With the limited data available at the time of writing, we found good agreement with a small mean bias of TROPOMI CO versus TCCON of 6.0 ppb for clear-sky, 6.2 ppb for cloudy-sky TROPOMI retrievals and 5.8 ppb for the combination of both with a station-to-station deviation of 3.9 ppb for clear-sky, 2.4 ppb for cloudy-sky, and 2.9 ppb for the combination case. Furthermore, the mean standard deviation of the bias is 3.9 ppb for clear-sky, 2.4 ppb for cloud-sky, and 2.9 ppb for the combination. The good agreement between clear-sky and cloudy-sky retrieval underlines the validity of the data retrieval for
10 cloudy scenes, a key aspect of the SICOR algorithm to achieve the data coverage of the TROPOMI CO product.

3.2 Detection of CO hot spots

Today's work and life style supports urbanization and the rise of metropolitan areas all over the world with populations exceeding more than 10 million people. Intense traffic and industrial activities in those regions led to high levels of air pollution affecting human health. For example, at rush hour in Mexico City the CO concentration has reached values as high as 9300 ppb
15 (Stephens et al., 2008). Sensing air pollution from space has the potential to globally monitor trends and variations of atmospheric pollutants affecting human health. The detection of air pollution above cities, urban and industrial areas with satellites comes with the challenge of low CO sensitivity of measurements. Until now, data needs to be temporally and spatially averaged to distinguish the CO enhancement of $\leq$ 20 ppb of the total column from the surrounding background concentrations in the order of 100 ppb (Pommier et al., 2013; Clerbaux et al., 2008; Borsdorff et al., 2017b).

20 In this respect, the CO measurements by TROPOMI represent a breakthrough. The advanced radiometric performance combined with the high spatial resolution and the daily global coverage of TROPOMI allows the sensing of CO enhancements above polluted areas with only single satellite overpasses, given the perspective of day-to-day monitoring. For example, Fig. 4a shows enhanced CO values over the industrial area near to Venice as well as pollution above Turin, Milan, and Rome. Figure 4b depicts an orbit overpass over Saudi Arabia and Egypt and shows distinct pollution patterns over Mecca, Jeddah and Cairo.
25 Furthermore, enhanced CO values along the Nile indicate air pollution in this densely populated region. Figure 4c clearly shows the enhanced CO values above Tehran, in agreement with the urban area of the city. Also smaller cities in the region like Qom, Isfahan, and Mashhad can be distinguished from the background CO level. Finally, Fig. 4d shows strong CO enhancements above Mexico City, Guadalajara, Torreón, and Monterrey. Data gaps in the figures are caused by the filtering of measurements under clear-sky conditions over the oceans and measurements contaminated by high altitude clouds.
30 The daily global coverage of TROPOMI and so the temporal evolution of air pollution on city scales opens up new possibilities to monitor the effect of emission regulations but also requires estimates of the absolute uncertainty of the TROPOMI CO product. Figure 5a shows that the TROPOMI CO concentrations are in good agreement with ground-based measurements of a Fourier Transform Spectrometer (FTS) in Mexico City (Stremme et al., 2009, 2013; Plaza-Medina et al., 2017) when selecting clear-sky satellite observations of the same day, which are spatially co-located in a radius of 15 km around the ground

site. Figure 5b indicates that this data screening is essential for the detection of pollution on city scales. Choosing a wider co-location radius for the satellite data leads to a significant bias with the FTS measurements, which demonstrates the importance of TROPOMI's spatial resolution for this type of application keeping representation errors between ground-based and satellite observations to a minimum. It is important to realize that the comparison with measurements at TCCON sites, as discussed

5 in the previous subsection, are mostly not affected by localized CO emissions and so allow for a looser spatial collocation criterion with a collocation radius of 50 km.

Furthermore, selecting TROPOMI CO observations for cloudy conditions leads to a 5 % bias independent from the selected radius. Due to light shielding by clouds, the satellite measurements become insensitive to the lower atmosphere where most of the pollution is located (Borsdorff et al., 2014) and so the TROPOMI CO product estimates the CO column from the less

10 polluted air above the cloud. This apparent disadvantage of cloudy observations turns into an advantage when analyzing the vertical distribution of trace gases (Borsdorff et al., 2017b). By observing the same pollution event for clear-sky conditions and for varying cloud height, it reveals the vertical extension of the city pollution into the atmosphere.

3.3 Monitoring pollution transport

For several years, measurements of CO have been used to trace the transport of polluted air masses within the atmosphere,

15 mostly with the focus on long-range transport. The atmospheric residence time of CO varies from days to months (Holloway et al., 2000) and so it is well suited to capture advection of atmospheric pollution. For example, Gloudemans et al. (2006) studied the transport of CO emission by biomass burning in the southern hemisphere using SCIAMACHY observations and Yurganov et al. (2004, 2005) analyzed the anomaly in the CO burden of the northern hemisphere caused by biomass burning with ground-based and satellite measurements.

20 The TROPOMI CO data set will advance this research field by providing the global distribution of the atmospheric CO concentration on a daily basis with high spatial resolution. It enables us to study the day-to-day variation of CO on global, regional and local scales. As an example, Fig. 6a-c present TROPOMI CO for three subsequent days depicting the southward transport of enhanced CO concentrations over the Atlantic Ocean originating from fires in north Africa. On 13th December, 2017 Ascension Island is surrounded by air with low CO concentrations (80 ppb) but already a few days later, on 17th December, the

25 first enhanced CO values reach the island. Finally on December 25th, Ascension Island is exposed to strong CO polluted air with values up to 116 ppb. Figure 7a shows that this finding is in agreement with regular ground-based FTS measurements of a Total Carbon Column Observing Network (TCCON) station on the island (Feist et al., 2014). With the help of TROPOMI measurements the localized ground-site measurements can be put into a regional context. Another example is given in Fig. 6d-f. On November 10th, biomass burning in Africa and Madagascar caused an extended plume of enhanced CO concentrations

30 reaching the island of Réunion. The atmospheric situation stayed stable until November 12th (90-103 ppb), but changed to low CO concentration on the 16th of November due to different meteorology (70 ppb). Figure 7b shows an excellent agreement with TCCON measurements at the island (De Mazière et al., 2014) and so illustrates the extra information provided by the satellite product in addition to the ground-based measurement.

4   Conclusions

The CO observations of TROPOMI represent a major step forward for the monitoring of air pollution from space. In this study we investigated the quality of the CO data product applying the operational retrieval software to the first two months of data of the S5-P mission in space. Because only a small fraction of all TROPOMI observations are expected to be cloud free (Krijger

5 et al., 2005), it is essential to account for clouds in the retrieval. Building on previous work (Gloudemans et al., 2009; Buchwitz et al., 2006; Vidot et al., 2012), an important feature of the TROPOMI CO retrieval is to infer cloud parameters and trace gas columns at the same time to achieve a good yield from the data processing. Thereby, we account for the vertical sensitivity of cloudy measurements and enhance the data yield both for observations over land and ocean (Borsdorff et al., 2017a, 2016). On average 8% of all measurements are clear-sky, 22 % cloudy-sky observations over land, and 51% cloud-sky over the oceans.

10 A first validation of the TROPOMI CO data product with collocated TCCON observations at nine selected measurement sites showed good agreement with a mean bias of about 6 ppb for both clear-sky and cloudy observations and a mean standard deviation of 3.9 ppb and 2.4 ppb, respectively, demonstrating a good repeatability of the observations. Here, the station-to-station bias is 3.9 ppb and 2.4 ppb for both types of measurements. Additionally, a comparison with ground-based FTS at Mexico shows that the CO pollution hot spot can be observed with high accuracy. Due to this good data quality in combination

15 with the spatial resolution of 7x7 km^2, TROPOMI can capture the variability of CO due to atmospheric transport of pollution on a day-to-day basis, demonstrated for daily overpasses at Ascension Island and Reunion in agreement with TCCON observations at these sites. Based on these preliminary results, we conclude that the TROPOMI CO product already fulfills the mission requirements on precision (< 10 %) and accuracy (< 15 %) (Veefkind et al., 2012), assuming a background concentration of 100 ppb.

20 ## 5   Data availability

The TROPOMI CO data set of this study is available for download at ftp://ftp.sron.nl/pub/pub/DataProducts/TROPOMI_CO/. The underlying data of the figures presented in this publication can be found at ftp://ftp.sron.nl/open-access-data/. TCCON data are available from the TCCON Data Archive, hosted by CaltechDATA, California Institute of Technology, CA (US), https://tccondata.org/.

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Disclaimer. The presented work has been performed in the frame of the Sentinel-5 Precursor Validation Team (S5PVT) or Level 1/Level 2 Product Working Group activities. Results are based on preliminary (not fully calibrated/validated) Sentinel-5 Precursor data that will still change.

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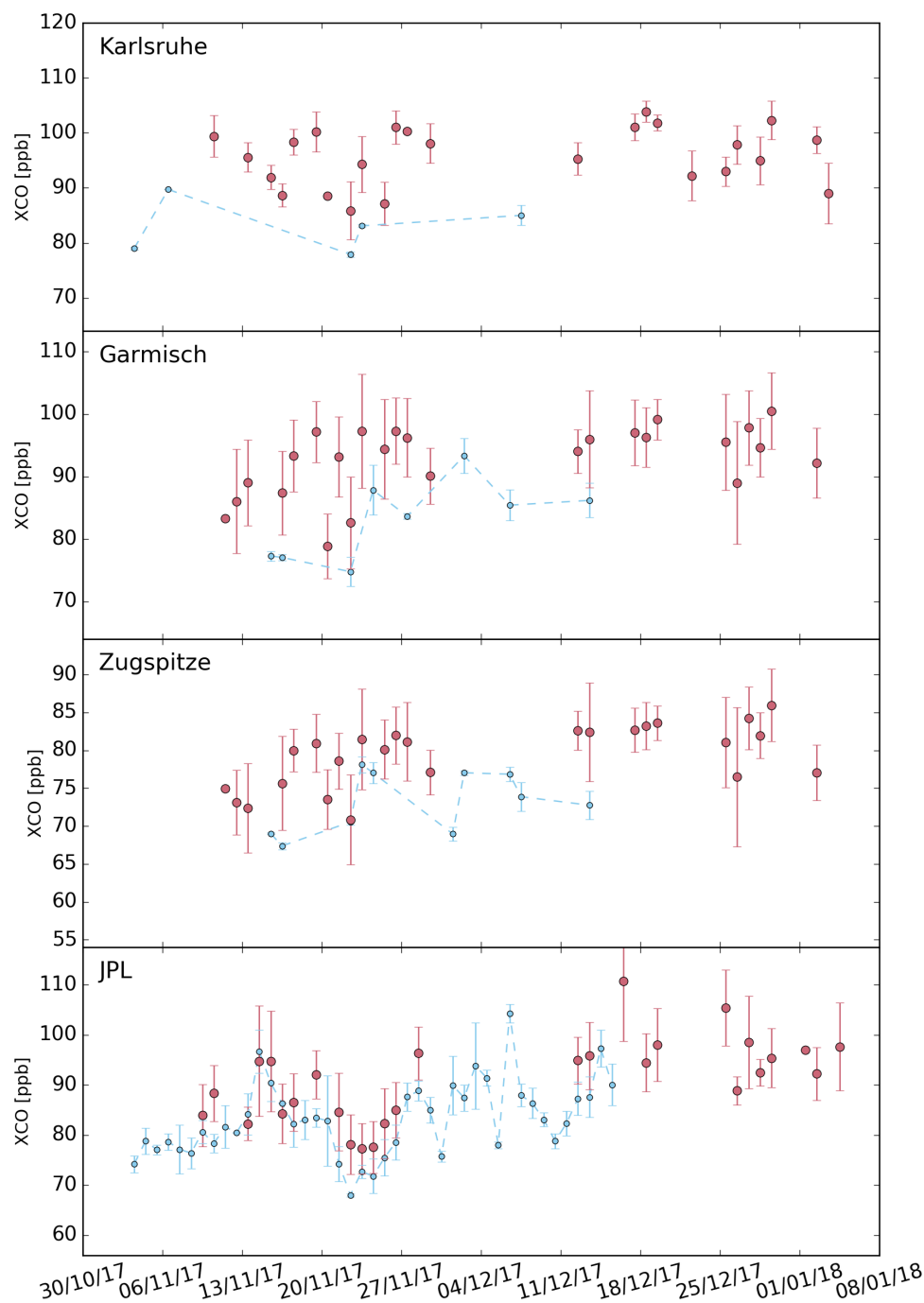
Figure 1. Daily means of dry air column mixing ratios (XCO) measured by TROPOMI (pink) and various TCCON stations (blue). A co-location radius of 50 km is used. The standard deviation of individual retrievals within a day is shown as an error bar.
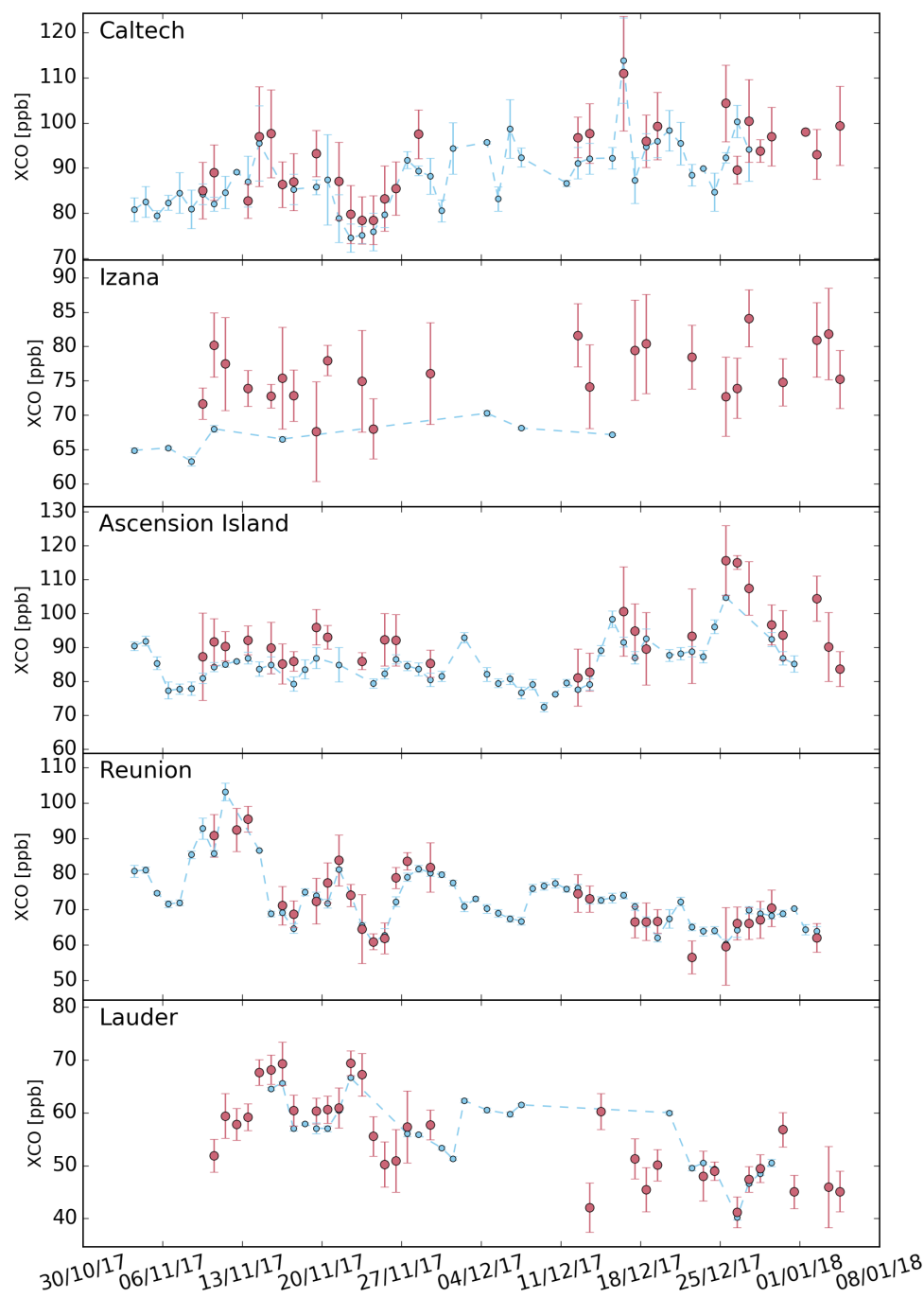
Figure 2. As Fig. 1 but with different TCCON stations.
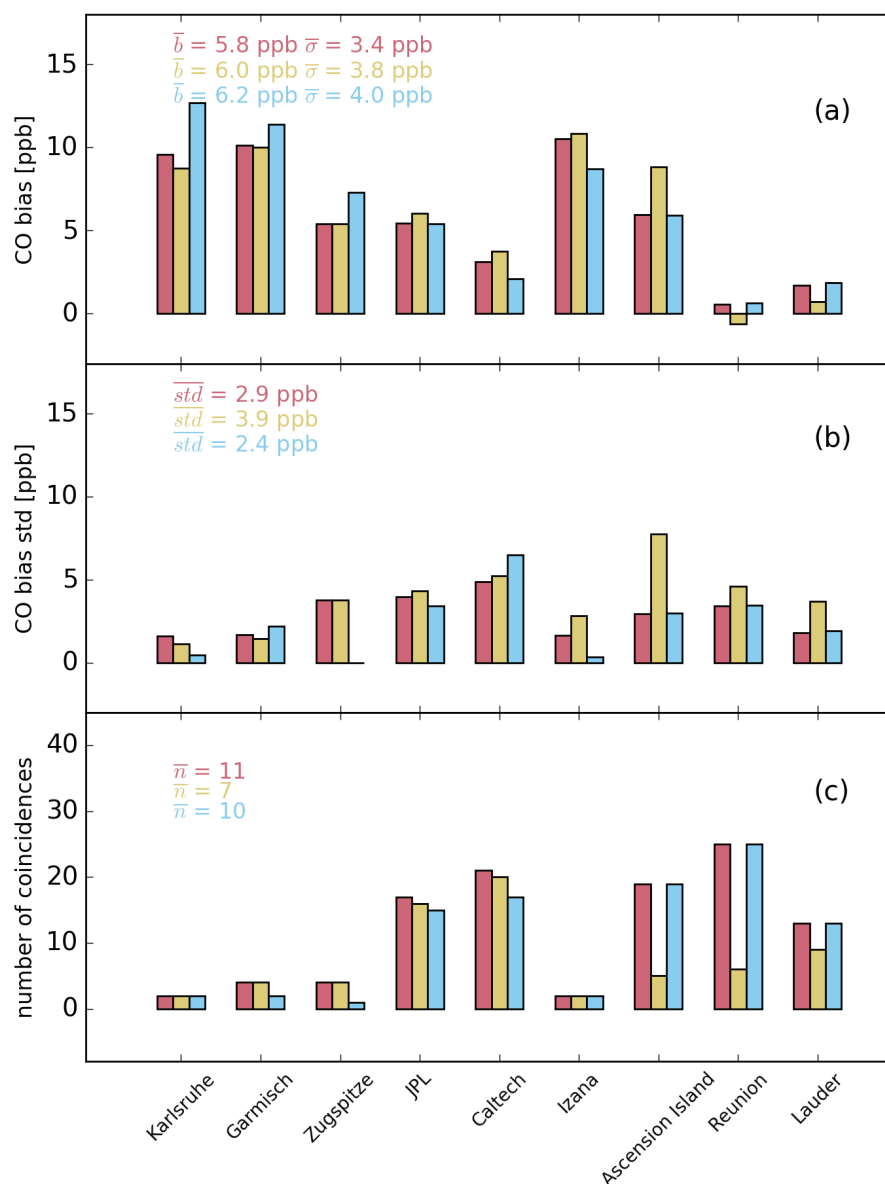
Figure 3. Mean bias (TROPOMI - FTS) between co-located daily mean XCO values (see Fig. 1, 2) of TROPOMI and TCCON (a), the standard deviation of the bias (b), and the number of coincident daily mean pairs (c). $\bar{b}$ is the global mean bias (average of all station biases) and $\bar{\sigma}$ its station-to-station . $\overline{std}$ is the average of all standard deviations and $\bar{n}$ the average number of coincident pairs. TROPOMI retrievals under clear-sky (yellow), cloudy-sky (blue) and the combination of both (pink) are distinguished.
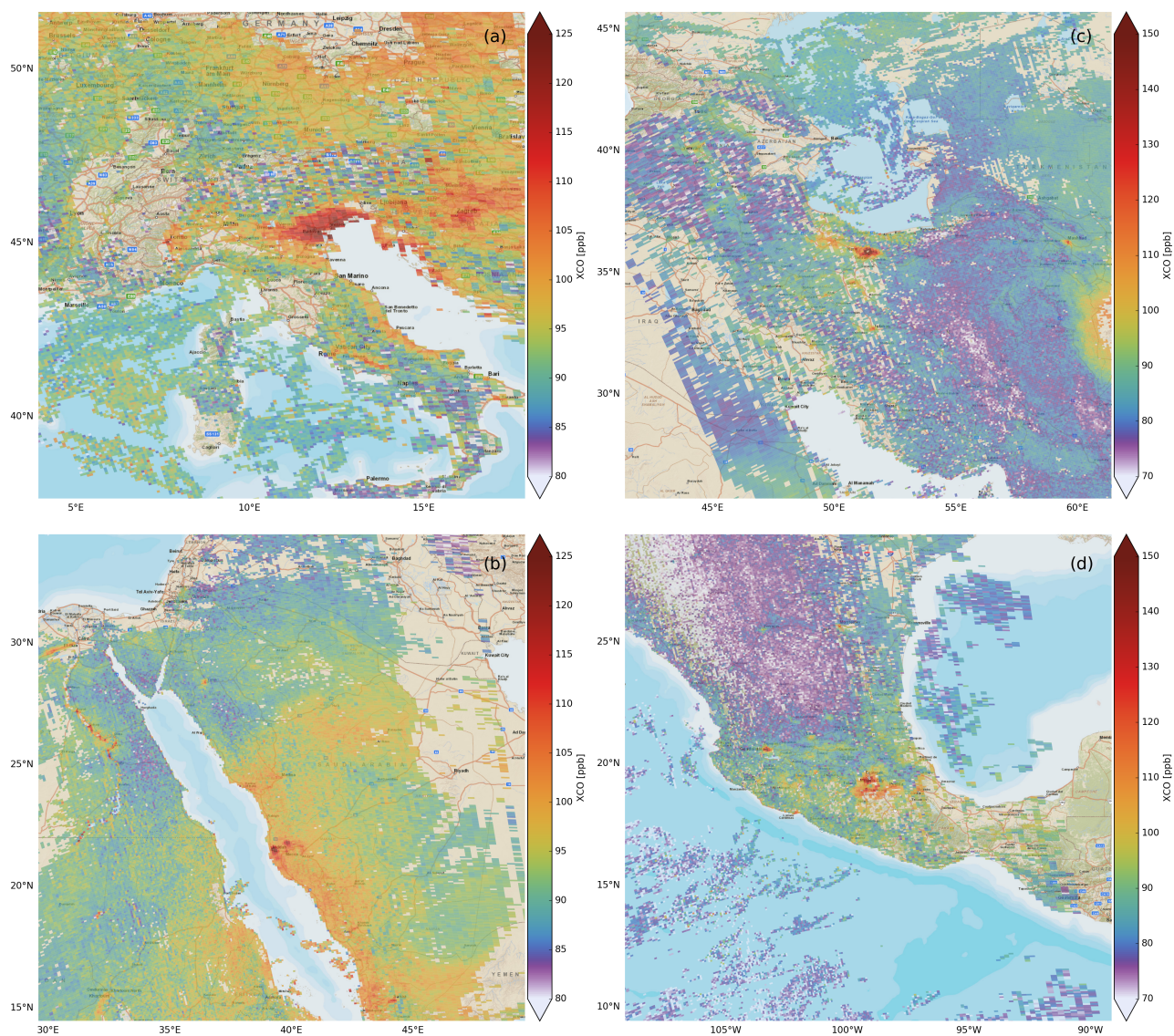
Figure 4. Total column mixing ratio (XCO) for individual TROPOMI ground pixels for (a) Italy on 25th December, (b) Saudi Arabia and Egypt on 12th November 2017, (c) Iran on 17th November 2017, and (d) Mexico on 25th November 2017.
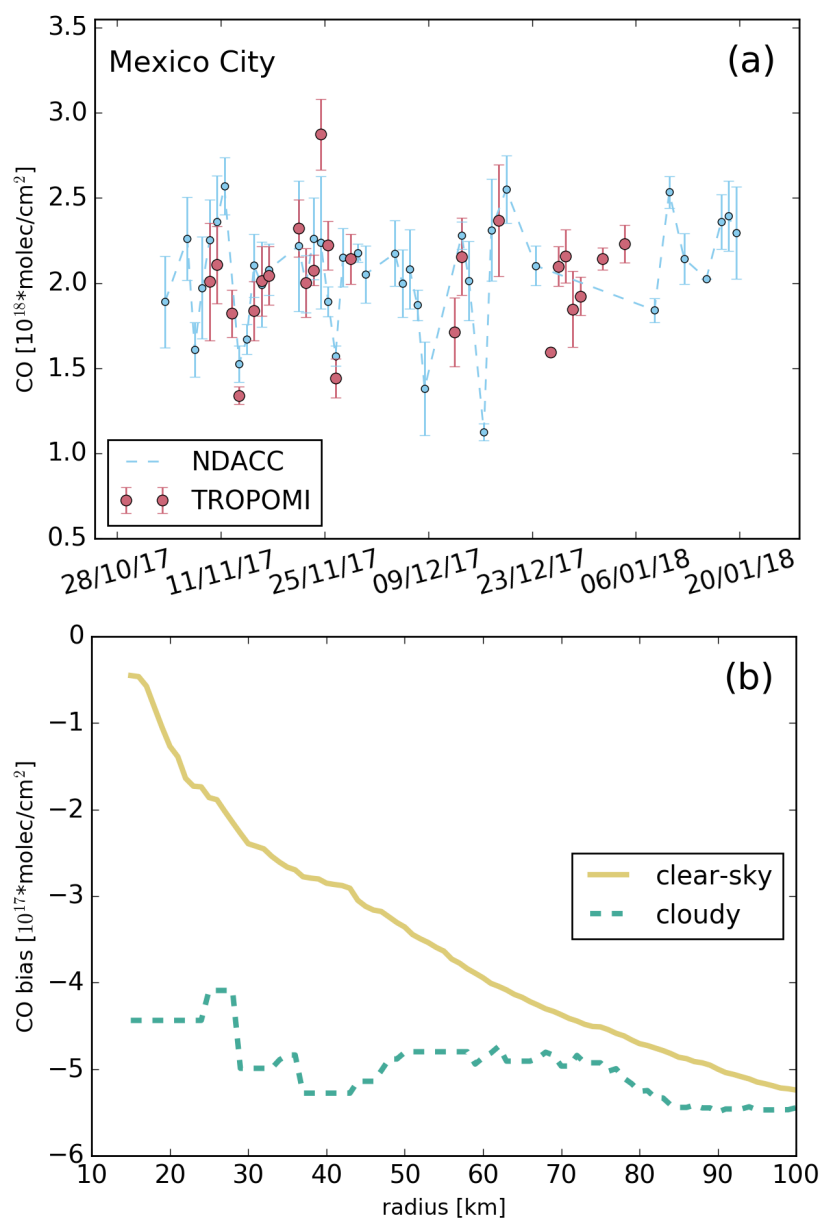
Figure 5. (a) Mexico City daily mean CO columns from TROPOMI (pink) and FTS (blue) with standard deviation of the individual retrievals (errors). TROPOMI observations are filtered for clear-sky within 15 km around the ground site. The TROPOMI columns are altitude corrected to the station elevation. (b) Bias of the CO columns (TROPOMI - FTS) as function of the co-location radius around Mexico City. Here, TROPOMI retrievals under clear-sky conditions (yellow) and optically thick clouds above 4000 m (green) are considered.
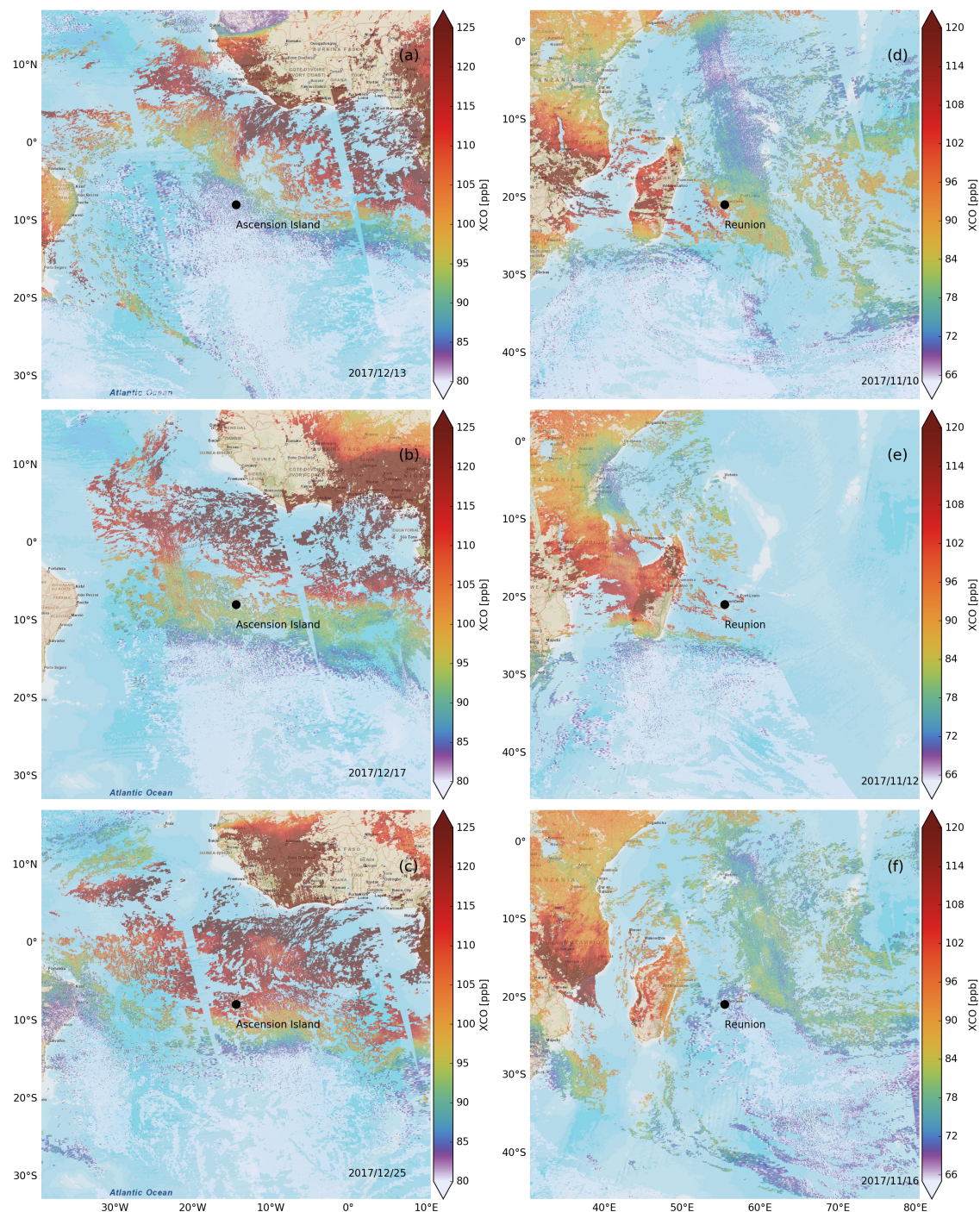
Figure 6. Total column mixing ratio (XCO) for individual TROPOMI ground pixels near Ascension Island on (a) 13th December, 2017, (b) 17th December, 2017, and (c) 25th December, 2017 and near Réunion on (d) the 10th November, 2017 (e) 12th November,2017, and (f) 17th November, 2017.
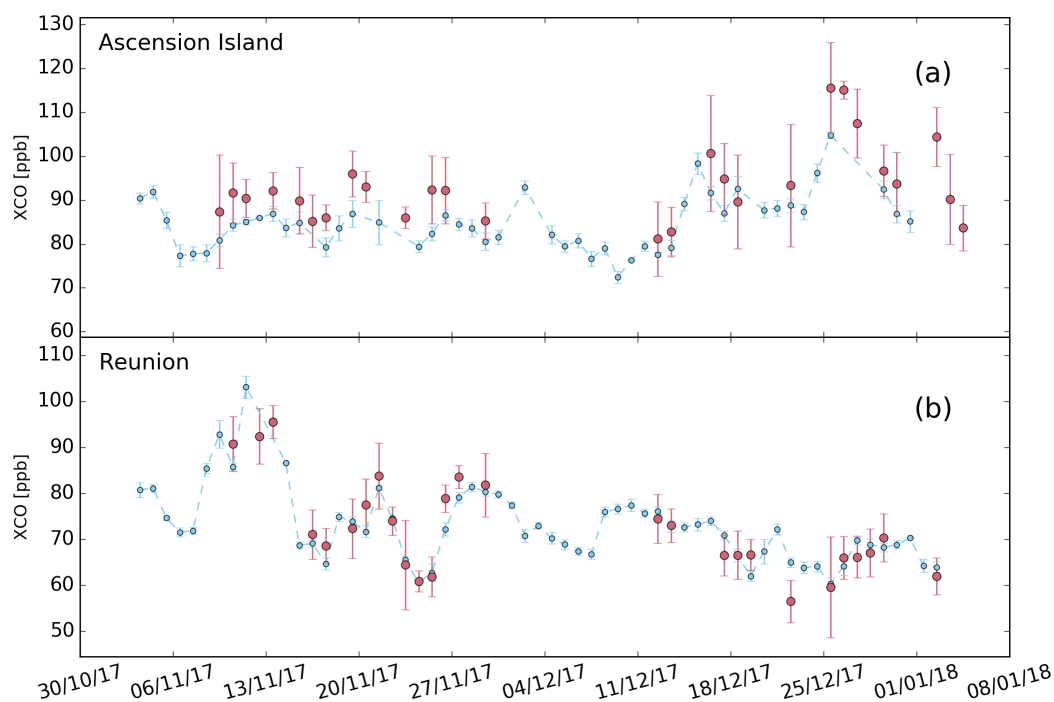
Figure 7. Daily means of dry air column mixing ratios (XCO) measured by TROPOMI (pink) and TCCON stations (blue) for (a) Ascension Island and (b) Réunion. A co-location radius of 50 km is used. The standard deviation of individual retrievals within a day is shown as error bar.

Table 1. Ground-based FTS stations used for validation. The latitude and longitude values are given in degrees, the surface elevation in km.

Name	Latitude	Longitude	Altitude	Type
Karlsruhe	49.10	8.44	0.11	TCCON
Garmisch	47.48	11.06	0.75	TCCON
Zugspitze	47.42	10.98	2.96	TCCON
JPL	34.20	−118.18	0.39	TCCON
Caltech	34.14	−118.13	0.24	TCCON
Izaña	28.31	−16.50	2.37	TCCON
Mexico City	19.33	−99.18	2.26	Bruker Vertex 80
Ascension Island	−7.92	−14.33	0.03	TCCON
Réunion	−20.90	55.49	0.09	TCCON
Lauder	−45.04	169.68	0.37	TCCON