

author comments on the manuscript amt-2018-132-RC1, reviewer 1

We would like to thank reviewer 1 for the constructive comments that aided us to improve our manuscript. In this document we provide our replies to the reviewer's comments. The original comments made by the reviewer are numbered and typeset in italic and bold face font. Following every comment we give our reply. Here line numbers, page numbers and figure numbers refer to the original version of the manuscript, if not stated differently. Additionally, the revised version of the manuscript is added.

General Comments

1. *This brief manuscript reports early findings from CO retrievals by the TROPOMI instrument launched in late 2017. In that sense, the manuscript is similar to a GRL paper (Measuring Carbon Monoxide With TROPOMI: First Results and a Comparison With ECMWF-IFS Analysis Data) recently published by Borsdorff et al. The main novel aspect of this new manuscript is an intercomparison with ground-based CO measurements from nine TCCON stations. The manuscript also presents several short case studies demonstrating CO gradients associated with urbanization and long-range transport. Readers of Atmospheric Measurement Techniques may find this paper interesting, although it contains little technical information. Since the recent GRL paper by Borsdorff et al. already demonstrated that the TROPOMI instrument and SICOR retrieval algorithm yield physically reasonable CO retrievals, this manuscript should really go into the quantitative use of the TROPOMI CO product in some depth. Without such detailed technical information, users will not be able to fully exploit this valuable new dataset. Some of this information is contained in the 2014 AMT paper by Borsdorff et al. (Insights into Tikhonov regularization ...) and the 2016 AMT paper by Landgraf et al. (Carbon monoxide total column retrievals from TROPOMI ...), although the relevance of those theoretical papers to operational TROPOMI retrievals (which are inevitably more complicated to analyze than simulated retrievals) is unclear. Thus, as currently written, this paper lacks important information needed by potential users for quantitative applications.*

Both the submitted manuscript and Borsdorff et al., GRL 2018 discuss the TROPOMI CO data product. Whereas the latter focuses on a first preliminary analysis of the product using a comparison with the ECMWF-IFS CO analysis data. The focus of this study is to quantify the uncertainty of the TROPOMI product by means of a comparison with ground-based reference measurements of the TCCON network. These measurements represent a well-accepted standard for the validation of satellite products, reported in several peer-reviewed papers. Most TCCON sites are located at remote regions and so the use of the NDACC site at Mexico City is another important and innovative element of the paper, demonstrating the validity of the product at regions of hot spot CO emissions. For users of the TROPOMI CO data, we consider such a study as essential, which quantifies the data quality. The technical details of the algorithm are well documented in the literature (Borsdorff et al., 2014 and Landgraf et al., 2016) has not changed since then, so we consider a proper referencing as sufficient. Based on the reviewers comments we see the clear need to discuss instrument specific aspects and the validation approach in more detail, which is described in our specific reply below.

2. *For readers of AMT, the manuscript should specifically address the following questions. What do the operational TROPOMI total column AKs look like, and how do they vary (for example, with respect to solar and satellite zenith angle, CO concentration, etc.)? Do the operational total column AKs depart from unity (i.e., the ideal case) far enough to be significant to users? In other words, is the smoothing error for operational TROPOMI CO column retrievals significant or not? For the operational TROPOMI retrievals, what are typical statistics for smoothing error and retrieval noise (as determined by the retrieval algorithm)? Do these quantities vary, or can they be fairly represented as fixed values?*

adjusted We added the following discussion at p3,113 and included a figure illustrating the vertical sensitivity of the retrieval and its dependence on the atmospheric state and on the observation geometry.

“For the total column of CO, the vertical sensitivity of the retrieval is described by the total column averaging kernel (Borsdorff et al., 2014), which is illustrated in Fig. 1 for TROPOMI data of one particular day, 10th November 2017. It shows the dependence of the averaging kernel on the cloudiness of the scene, where the standard deviation indicates its variation due to different observation and atmospheric parameters, e.g. solar zenith angle, viewing zenith angle and ground reflectivity. For very strict cloud

clearing of the data (with $z < 5$ km and $\tau < 0.01$), the total column averaging kernel is close to 1 for all altitudes with little variation, meaning that the derived column can be interpreted as an estimate of vertically integrated amount of CO. Filtering the data less strict using the clear-sky filter from above ($z < 5$ km and $\tau < 0.5$) results in a slightly reduced sensitivity with a moderate standard deviation and Borsdorff et al. (2017) concluded that those measurements are usually clear-sky equivalent for remote regions without local pollution sources and the induced errors due to the choice of the reference profile to be scaled by the inversion to be on a percentage level (Borsdorff et al., 2014). The presence of clouds changes significantly the vertical sensitivity of the retrieval. Figure 1 shows the column averaging kernel when filtering for optical thick clouds at 5 km altitude. The sensitivity below the cloud is significantly reduced (values lower than 1) due to cloud shielding, and the retrieval estimates a CO total column mainly based on the measurement sensitivity to CO above the cloud (values higher than 1). This can lead to errors $> 30\%$ when the averaging kernel is not used for data interpretation (Borsdorff et al., 2014). However, the TROPOMI CO dataset provides total column averaging kernels for each retrieval and we recommend to use them when ever possible. ”

Minor Revisions and Technical Corrections

1. *Somewhere, the manuscript should clearly define the relationship between total column density of CO, which would have units of molecules per area, and the dry air mixing ratio with units of ppb. Which of these is the actual TROPOMI CO product? Exactly where and how is the conversion between these two quantities made?*

adjusted We added the following sentence on p3,l7:

“Hence, the retrieval result is the total column density of CO [molec/cm²]. To compare it with other measurements we also represent the data product as a dry column mixing ratio XCO [ppb] by dividing the CO total column density by the dry air column density derived from colocated ECMWF pressure fields. ”

2. *p. 1, line 8 - The adjective game-changing should probably be replaced with a more formal term like groundbreaking.*

adjusted We changed the term to “groundbreaking”

3. *p. 2, line 8 - Please include a number for typical background CO concentrations after low background concentration.*

adjusted We added a typical background value of CO at p2,l8:

“With a typical background concentration of ca. 80ppb (in the Northern Hemisphere) ...”

4. *p. 2, line 21 - The SICOR algorithm exploits a CO reference profile which is scaled to produce the retrieved CO total column. Is this reference profile a fixed profile, or does it vary geographically and/or seasonally? What method was used to obtain the reference profile?*

adjusted We added the following sentence with a reference to the source of the reference profile:

“The reference profile of CO that is scaled during the retrieval is taken from simulations of the global chemical transport model TM5 (Krol et al., 2005) and monthly averaged over 3 degree \times 2 degree latitude/longitude grid boxes. ”

5. *p. 3, line 10 - The text is not clear regarding the criteria for classifying clear and cloudy cases, and which cases are discarded entirely. For example, are all cases where cloud optical thickness $\hat{>}$ 0.5 OR cloud height $\hat{>}$ 5000 considered cloudy, or is it really where cloud optical thickness $\hat{>}$ 0.5 AND cloud height $\hat{>}$ 5000? Please clarify.*

adjusted We changed the paragraph p3,l10 from:

“Furthermore, we distinguished between retrievals under clear-sky ($\tau < 0.5$, $z < 5000$ m, over land) and cloudy condition ($\tau > 0.5$, $z < 5000$ m over land and ocean). ”

to

“Furthermore, we distinguished between retrievals under clear-sky ($\tau < 0.5$ and $z < 5$ km, over land) and cloudy condition ($\tau > 0.5$ and $z < 5$ km, over land and ocean). The remaining retrievals are not considered in this study. ”

6. *p. 3, line 16 - The striping issue should be discussed in more detail, with a specific example (including a figure) describing the problem and demonstrating the improvement associated with the method. Does the 5 % value refer to the bias in CO or to the number of retrievals affected by the issue?*

adjusted We added a figure showing a typical striping pattern and changed the paragraph p3,l14 from:

“ The TROPOMI instrument is still in the early phase of the mission and the performance of the CO retrieval is expected 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 NO₂ data product of the Ozone Monitoring Instrument (OMI, Levelt et al. (2006)). This aspect will be the subject of future investigations of the instrument calibration key data. ”

to

“ The TROPOMI instrument is still in the early phase of the mission and the performance of the CO retrieval is expected to improve in the future. For example, single overpasses show stripes of erroneous CO in 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 per orbit. Figure ?? provides an example, where the average the average of the stripe pattern in cross flight direction is -0.03 ppb with a standard deviation of 1.1 ppb. Some stripes can reach values higher than 5 ppb. Hence, the stripe pattern can be removed from the data a posteriori to the retrieval and its removal is indicated accordingly in the remainder of the paper. Boersma et al. (2011) suggested a similar approach to improve the quality of the NO₂ data product of the Ozone Monitoring Instrument (OMI, Levelt et al. (2006)). ”

7. *p. 3, Section 3.1 - This section does not include any discussion or consideration of the effects of the different total column averaging kernels for TROPOMI and TCCON. As a result, the influence of the retrieval vertical sensitivity and smoothing error can not be distinguished from other conceivable types of retrieval bias. Therefore, in the title of Section 3.1 (and elsewhere in the manuscript), I would use the term Intercomparison rather than Validation. In-situ profiles are typically needed for proper validation work since retrieval vertical sensitivity and smoothing error can be explicitly represented.*

adjusted In general the reviewer is right. The TCCON columns are based on remote sensing and are affected by a smoothing error. However, Wunch et al. (2015) showed that the total error including the smoothing error of XCO is below 4%. The smoothing error of TROPOMI clear sky measurements is very similar and so within this uncertainty margin both data sets can be seen as an estimate of the true CO concentration for our study. We add the following sentence p3,l24:

“ Wunch et al. (2015) reported that the total error of the XCO columns measured by TCCON is below 4%. Hence, within this error margin we can assume the TCCON measurements as an estimate of the truth. ”

8. *p. 3, line 29 - The handling of discrepancies between the TCCON station altitude and the assumed altitude (or surface pressure) in the TROPOMI retrievals requires more explanation, preferably with an example. Is it possible to estimate an uncertainty (or an upper limit) for TROPOMI/TCCON intercomparisons due to this effect? How does this uncertainty vary among the different TCCON sites?*

adjusted We quantified the effect of the altitude correction and added the following paragraph at p3, l31:“

“The retrieved CO column of TROPOMI is adapted to the altitude of the station 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. For mountain stations like Zugspitze and Izana, this reduces the TROPOMI CO column on average by 10 and 4 ppb, respectively, improving the agreement between the ground-based and satellite measurements accordingly.”

9. *p. 4, line 17 - The text implies, but does not clearly state, that neither temporal nor spatial averaging is necessary to ... to distinguish the CO enhancement of 20 ppb of the total column from the surrounding background concentrations ... Please clarify. An analysis of TROPOMI retrieval noise (random error) would be an essential part of this discussion.*

adjusted We calculated the retrieval noise error for the point sources shown in this study and add the following paragraph to the manuscript p4, l30:

“ Neither temporal nor spatial averaging is necessary to distinguish the CO enhancements of the total column above the shown point sources. The average noise error of the retrievals from the individual ground pixels shown in Fig. 6 is < 2.3 ppb.”

10. *p. 4, line 18 - The text 20 ppb of the total column should be reworded since ppb is not the unit for total column.*

adjusted We changed the sentence from :

“ ...distinguish the CO enhancement of ≤ 20 ppb of the total column from the surrounding background concentrations in the order of ...”

to

“ ...distinguish typical the CO enhancements of ≤ 20 ppb of the total column dry air mixing ratio from the surrounding background concentrations in the order of ...

”

11. *p. 6, line 25 - retrieval is misspelled Figures 1 and 2 include many data points for days where TROPOMI data were available but TCCON were not, or vice-versa. Such data were not used to calculate TROPOMI/TCCON bias statistics. I would suggest revising the figures somehow to identify which points were actually used to calculate the bias statistics.*

adjusted we recalculated the Fig. 1 and 2. Data without time coincidence for TCCON or TROPOMI is plotted in grey. Furthermore we added the following sentence to the figure caption of Fig.1:

“ Data points without time coincidence between TCCON and TROPOMI are plotted in grey. ”

References

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author comments on the manuscript amt-2018-132-RC1, reviewer 2

We would like to thank reviewer 2 for the constructive comments that aided us to improve our manuscript. In this document we provide our replies to the reviewer's comments. The original comments made by the reviewer are numbered and typeset in italic and bold face font. Following every comment we give our reply. Here line numbers, page numbers and figure numbers refer to the original version of the manuscript, if not stated differently. Additionally, the revised version of the manuscript is added.

Major Comments

1. ***In the abstract the game-changing nature of TROPOMI is emphasized. I would like to see further discussion on this topic perhaps in the Introduction chapter and later in the manuscript to more specifically address this point. E.g. it would be good to include some reference what the heritage instruments measured and why TROPOMI is a game-changer. Need for averaging data is briefly mentioned in Sec 3.2, but I would welcome a bit more discussion on this.***

adjusted We add the following text

p2,15:

“...and a high radiometric accuracy to infer the CO total column over dark vegetation surfaces with an precision < 10 % (Veefkind et al., 2012)”

and p2, 111

“The S5P mission builds upon the heritage of SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography; Bovensmann et al. (1999)), which provided atmospheric CO total column concentrations from the same spectral range (Borsdorff et al., 2017a, Buchwitz et al., 2007, Frankenberg et al., 2005, Gimeno Garcia et al., 2011, Gloudemans et al., 2009)). Measurements of SCIAMACHY in the SWIR have a spatial resolution of about 30 km × 120 km (along-track × across-track) for an integration time of 0.5 s with a global coverage cycle of 3 days. Most importantly, the SCIAMACHY noise error of single CO retrievals can exceed 100 % for a dark scenes. Hence, spatial and temporal averaging of single CO measurements is required (de Laat et al., 2007, Gloudemans et al., 2006), which limits the data interpretation of SCIAMACHY CO data.”

2. ***In the text both terms XCO and CO are used. Just by looking at the notation one might get the impression that XCO denotes daily values, which is perhaps not meant (Section 3.1). Please, clarify what is the TROPOMI data product and whether the validation was based on XCO or CO products.***

adjusted We added the following sentence on p3,17:

“Hence, the retrieval result is the total column density of CO [molec/cm²]. To compare it with other measurements we also represent the data product as a dry column mixing ratio (XCO, [ppb]) by dividing the CO total column density by the dry air column density. ”

3. ***In the validation both cloudy (low clouds) and cloud-free conditions are compared. Please, add discussion how valid FTS cloudy observations are, or clarify if only cloud free FTS observations are considered. Are TCCON and FTS in cloudy cases measuring the same air mass? Cloud optical thickness is used to select clear sky observations where is this information coming from?***

adjusted We added the following sentence on p3,125:

“ Cloud contaminated measurements are rejected and so TCCON measurements refer to clear-sky observations only. Here, TCCON CO columns are provided as column averaged dry air mole fractions XCO (Wunch et al., 2010). ”

4. ***Please add a paragraph on FTS measurements since they are used as reference data here, their accuracy etc. The geographical distribution of the validation sites is limited to 50S-50N. Please, this is also good to be included in the text.***

adjusted We added the following paragraph on p3,l25:

“TCCON is a network of ground-based Fourier Transform Spectrometers to measure total column concentrations of atmospheric trace gases including CO with high accuracy and precision e.g. for satellite validation. The trace gas columns are retrieved from spectrally highly resolved near-infrared radiance measurements recorded in direct-sun geometry (Wunch et al., 2015)”

5. *Soft calibration is done during the validation please, discuss if this is also recommended when operational data is available.*

adjusted The stripe correction mentioned by the reviewer was not applied during the validation. This is an important point that was not mentioned in the manuscript before. We added the following sentence on p3,l14:

“It is important to note that we only applied the de-stripping approach on the Figs. 6 and 8 but not on the TROPOMI CO data used in the later validation. The CO data striping will be the topic of upcoming studies with the first preference to improve the instrument calibration key data.”

6. *Related to figures 4 and 6: Is here both cloudy data and cloud free data? Is there difference in the interpretation of the pixels depending on whether they are cloud free or cloudy? Please, add some discussion on this.*

adjusted In Figure 4 and 6, we do not distinguish between clear-sky and cloudy data to obtain a good data coverage. For clarification, we added the following sentence at p4, l29

“Figure 6 shows predominately clear-sky observations but also includes retrievals from cloud contaminated scenes, which in case of optical thick and high clouds reduces the sensitivity to boundary layer CO pollution at emission hot spots (Borsdorff et al., 2017b).”

7. *Conclusions: I would appreciate discussion on what exactly has been validated and to elaborate more what type of validation is needed in the future (in terms of spatial and temporal coverage, atmospheric and observational conditions).*

adjusted We already state in the conclusion (p6,l10-14) that the TROPOMI CO dataset (clear-sky, cloudy) was validated with TCCON and NDACC measurements. Furthermore, we reported the biases with the validation measurements. We add the following sentence at p6,l13:

“ For this study, only a limited amount of TCCON data was available with confined spatial and temporal coverage. The Sentinel 5 Precursor as an operational mission requires a continuous monitoring of the CO data quality, which will be performed as part of the operational validation activities. In this context, future work will consider the validation of the TROPOMI CO data for longer time scales including additional TCCON and NDACC stations to improve the significance of the product validation.”

Minor comments

1. *Fig 1 & Fig 2. Please, indicate if this refers to cloud free data / cloudy data or both. If both, would it be possible to indicate this somehow.*

adjusted The daily means in the plots include both clear-sky and cloudy satellite data. We indicated this in the caption of Fig.1 by adding the sentence:

“ Clear-sky and cloudy satellite data are used to calculate the daily means. ”

2. *P 1, L 6: Sentence starting Due to you could re-formulate this to make the message more clear.*

adjusted We change the sentence p1,l6 from:

“ 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. ”
to

“ The moderate atmospheric resistance time and the low background concentration leads to localized pollution hot spots of CO and allows to track the atmospheric transport of pollution on regional to global scales.
”

3. *P 1, L 13: Please, clarify station-to-station bias?*

adjusted We changed the sentence at p1,l13 from:

“ 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.”

to

“ We found a good agreement between both data sets with a mean bias of 6 ppb (average of individual station biases) for both clear-sky and cloudy TROPOMI CO retrievals. Together with the corresponding standard deviation of the individual station biases of 3.8 ppb for clear-sky and 4.0 ppb for cloudy-sky, it indicates that the CO data product is already well within the mission requirement.

”

4. *P2,L 13. led-;lead?*

corrected

5. *Figure 5, lower panel, indications on how many observations correspond to varying radius values would be very nice to have.*

adjusted We add the following two sentences in the caption of Fig. 5:

“ For the smallest radius (15 km) we found 20 cloudy and 160 clear-sky collocations. However, for the widest radius (100 km) 92 cloudy and 4425 clear-sky collocations are found. ”

6. *Fig 7 I think this is a zoom of figure 2. Is this needed?*

adjusted The reviewer is right we removed Fig. 7 from the manuscript.

References

- Borsdorff, T., aan de Brugh, J., Hu, H., Nédélec, P., Aben, I., and Landgraf, J.: Carbon monoxide column retrieval for clear-sky and cloudy atmospheres: a full-mission data set from SCIAMACHY 2.3 μm reflectance measurements, *Atmospheric Measurement Techniques*, 10, 1769–1782, <https://doi.org/10.5194/amt-10-1769-2017>, URL <https://www.atmos-meas-tech.net/10/1769/2017/>, 2017a.
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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 products is atmospheric carbon monoxide (CO). It is observed with daily global coverage and a high spatial resolution of 7x7 km². ~~Due to its moderate atmospheric residence time, its atmospheric abundance provides information on both~~ The moderate atmospheric resistance time and the low background concentration leads to localized pollution hot spots ~~and the pollutant transport of CO and allows to track the atmospheric transport of pollution~~ on regional to global scales. In this contribution, we demonstrate the ~~game-changing groundbreaking~~ 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 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 (average of individual station biases) for both clear-sky and cloudy TROPOMI CO retrievals. Together with the corresponding standard deviation of the ~~station-to-station bias of 3.9~~ individual station biases of 3.8 ppb for clear-sky and ~~2.4~~ 4.0 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 $7 \times 7 \text{ km}^2$, and a high radiometric ~~performance~~ accuracy to infer the CO total column over dark vegetation surfaces with an precision $< 10 \%$ (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~~ typical background concentration of ca. 80 ppb (in the Northern Hemisphere) 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. (2010)~~ which ~~retrieved~~ provided atmospheric CO total column ~~concentration~~ concentrations from the same spectral range ~~-(Gloudemans et al., 2000)~~ Measurements of SCIAMACHY in the SWIR have a spatial resolution of about $30 \text{ km} \times 120 \text{ km}$ (along-track \times across-track) for an integration time of 0.5 s with a global coverage cycle of 3 days. Most importantly, the SCIAMACHY noise error of single CO retrievals can exceed 100 % for dark scenes. Hence, spatial and temporal averaging of single CO measurements is required (de Laat et al., 2007; Gloudemans et al., 2006), which limits the data interpretation of SCIAMACHY CO data.

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 $2.3 \mu\text{m}$ 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~~ Based on this, our study deploys 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 $2.3 \mu\text{m}$ spectra of TROPOMI and retrieved the total column density of CO simultaneously with interfering trace gases and effective cloud parameters (cloud height (z) and optical thickness (τ)) 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). The reference profile of CO that is scaled during the retrieval is taken from simulations of the global chemical transport model TM5 (Krol et al., 2005) and monthly averaged over $3 \text{ degree} \times 2 \text{ degree}$ latitude/longitude grid boxes. Hence, the retrieval result is the total column density of CO [molec/cm^2]. To compare it with other measurements we also represent the data product as a dry column mixing ratio XCO [ppb] by dividing the CO total column density by the dry air column density derived from colocated ECMWF pressure fields.

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 that is still under investigation. Furthermore, we distinguished between retrievals under clear-sky (~~cloud-optical-thickness $< \tau < 0.5$, cloud-height $< 5000 \text{ m}$ and $z < 5 \text{ km}$~~ , over land) and cloudy condition (~~cloud-optical-thickness $> \tau > 0.5$, cloud-height $< 5000 \text{ m}$ and $z < 5 \text{ km}$~~ , over land and ocean). The remaining retrievals are not considered in this study. 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.

For the total column of CO, the vertical sensitivity of the retrieval is described by the total column averaging kernel (Borsdorff et al., 2014), which is illustrated in Fig. 1 for TROPOMI data of one particular day, 10th November 2017. It shows the dependence of the averaging kernel on the cloudiness of the scene, where the standard deviation indicates its variation due to different observation and atmospheric parameters, e.g. solar zenith angle, viewing zenith angle and ground reflectivity. For very strict cloud clearing of the data (with $z < 5 \text{ km}$ and $\tau < 0.01$), the total column averaging kernel is close to 1 for all altitudes with little variation, meaning that the derived column can be interpreted as an estimate of vertically integrated amount of CO. Filtering the data less strict using the clear-sky filter from above ($z < 5 \text{ km}$ and $\tau < 0.5$) results in a slightly reduced sensitivity with a moderate standard deviation and Borsdorff et al. (2017a) concluded that those measurements are usually clear-sky equivalent for remote regions without local pollution sources and the induced errors due to the choice of the reference profile to be scaled by the inversion to be on a percentage level (Borsdorff et al., 2014). The presence of clouds

changes significantly the vertical sensitivity of the retrieval. Figure 1 shows the column averaging kernel when filtering for optical thick clouds at 5 km altitude. The sensitivity below the cloud is significantly reduced (values lower than 1) due to cloud shielding, and the retrieval estimates a CO total column mainly based on the measurement sensitivity to CO above the cloud (values higher than 1). This can lead to errors $> 30\%$ when the averaging kernel is not used for data interpretation (Borsdorff et al., 2014). However, the TROPOMI CO dataset provides total column averaging kernels for each retrieval and we recommend to use them when ever possible.

The TROPOMI instrument is still in the early phase of the mission and the performance of the CO retrieval is expected to improve in the future. For example, single overpasses show stripes of erroneous CO values $< 5\%$ in the in 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, per orbit, Figure 2 provides an example, where the average the average of the stripe pattern is in cross flight direction is -0.03 ppb with a standard deviation of 1.1 ppb. Some stripes can reach values higher than 5 ppb. Hence, the stripe pattern can be removed from the data a posteriori to the retrieval and its removal is indicated accordingly in the remainder of the paper. 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 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 FTS stations operated by the TCCON network (see Table 1) which are located preferably at remote areas.

TCCON is a network of ground-based Fourier Transform Spectrometers to measure total column concentrations of atmospheric trace gases including CO with high accuracy and precision e.g. for satellite validation. The trace gas columns are retrieved from spectrally highly resolved near-infrared radiance measurements recorded in direct-sun geometry (Wunch et al., 2015). Cloud contaminated measurements are rejected and so TCCON measurements refer to clear-sky observations only. Here, TCCON CO columns are provided as column averaged dry air mole fractions XCO (Wunch et al., 2010).

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). Wunch et al. (2015) reported that the total error of the XCO columns measured by TCCON is below 4% . Hence, within this error margin we can assume the TCCON measurements as an estimate of the truth.

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 by either cutting off the scaled mixing ratios ratio profile at the station altitude or extending it assuming a constant elongation of the mixing ratio to lower altitude. For mountain stations like Zugspitze and Izana, this reduces the

TROPOMI CO column on average by 10 and 4 ppb, respectively, improving the agreement between the ground-based and satellite measurements accordingly. Finally, we calculated daily averages of the ~~dry air mixing ratios (XCO)~~ XCO values using the adapted TROPOMI retrievals and the TCCON measurements shown in Fig. 3 and Fig. 4. Data gaps in the TROPOMI time series are partly caused by discarding observations with high clouds (~~>5000-m~~5 km) but also due to observation time reserved for in-orbit instrument characterization during the instrument commissioning phase.

Figure 5 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.

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~~ standard deviation of the individual station biases of 3.8 ppb for clear-sky, ~~2.4~~ 4.0 ppb for cloudy-sky, and ~~2.9~~ 3.4 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 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~~ lead 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 (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~~ typical the CO enhancements of ≤ 20 ppb of the total column dry air mixing ratio from the surrounding background concentrations in the order of 100 ppb (Pommier et al., 2013; Clerbaux et al., 2008; Borsdorff et al., 2017b).

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. 6a shows enhanced CO values over the industrial area near to Venice as well as pollution above Turin, Milan, and Rome. Figure 6b depicts an orbit overpass over Saudi Arabia and Egypt and shows distinct pollution patterns over Mecca, Jeddah and Cairo. Furthermore, enhanced CO values along the Nile indicate air pollution in this densely populated region. Figure 6c 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. 6d 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. Figure 6 shows

predominately clear-sky observations but also includes retrievals from cloud contaminated scenes, which in case of optical thick and high clouds reduces the sensitivity to boundary layer CO pollution at emission hot spots (Borsdorff et al., 2017b). Neither temporal nor spatial averaging is necessary to distinguish the CO enhancements of the total column above the shown point sources. The average noise error of the retrievals from the individual ground pixels shown in Fig. 6 is < 2.3 ppb.

5 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 7a 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
10 site. Figure 7b indicates that this data screening is essential for the detection of pollution on city scales. Choosing a wider collocation 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 in the previous subsection, are mostly not affected by localized CO emissions and so allow for a looser spatial collocation
15 criterion with a collocation radius of 50 km.

Furthermore, selecting TROPOMI CO observations for cloudy conditions leads to a ~~5 %~~ 25 % (5 ± 7 molec/cm²) 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 polluted air above the cloud. This apparent disadvantage of cloudy observations turns into an
20 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, mostly with the focus on long-range transport. The atmospheric residence time of CO varies from days to months (Holloway
25 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.

30 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. 8a-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 De-

cember, the 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 [??a-4 \(panel 3\)](#) 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. 8d-f. On November 10th, biomass burning in Africa and Madagascar caused an extended plume of enhanced CO concentrations 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 [??b-4 \(panel 4\)](#) 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.

10 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 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.

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~~ [standard deviation of the station biases is 3.8 ppb and 4.0](#) 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~~ [For this study, only a limited amount of TCCON data was available with confined spatial and temporal coverage. The Sentinel 5 Precursor as an operational mission requires a continuous monitoring of the CO data quality, which will be performed as part of the operational validation activities. In this context, future work will consider the validation of the TROPOMI CO data for longer time scales including additional TCCON and NDACC stations to improve the significance of the product validation.](#)

[Due to the](#) good data quality in combination with the spatial resolution of $7 \times 7 \text{ km}^2$, 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.

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),

5 <https://tccondata.org/>. [The TROPOMI CO data is available via the Copernicus Open Access Hub https://s5phub.copernicus.eu.](https://s5phub.copernicus.eu/)

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10 O. Wennberg, and Geoffrey C. Toon performed FTS measurements and retrievals for the various stations. Jochen Landgraf supervised the study. All authors discussed the results and commented on the manuscript.

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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

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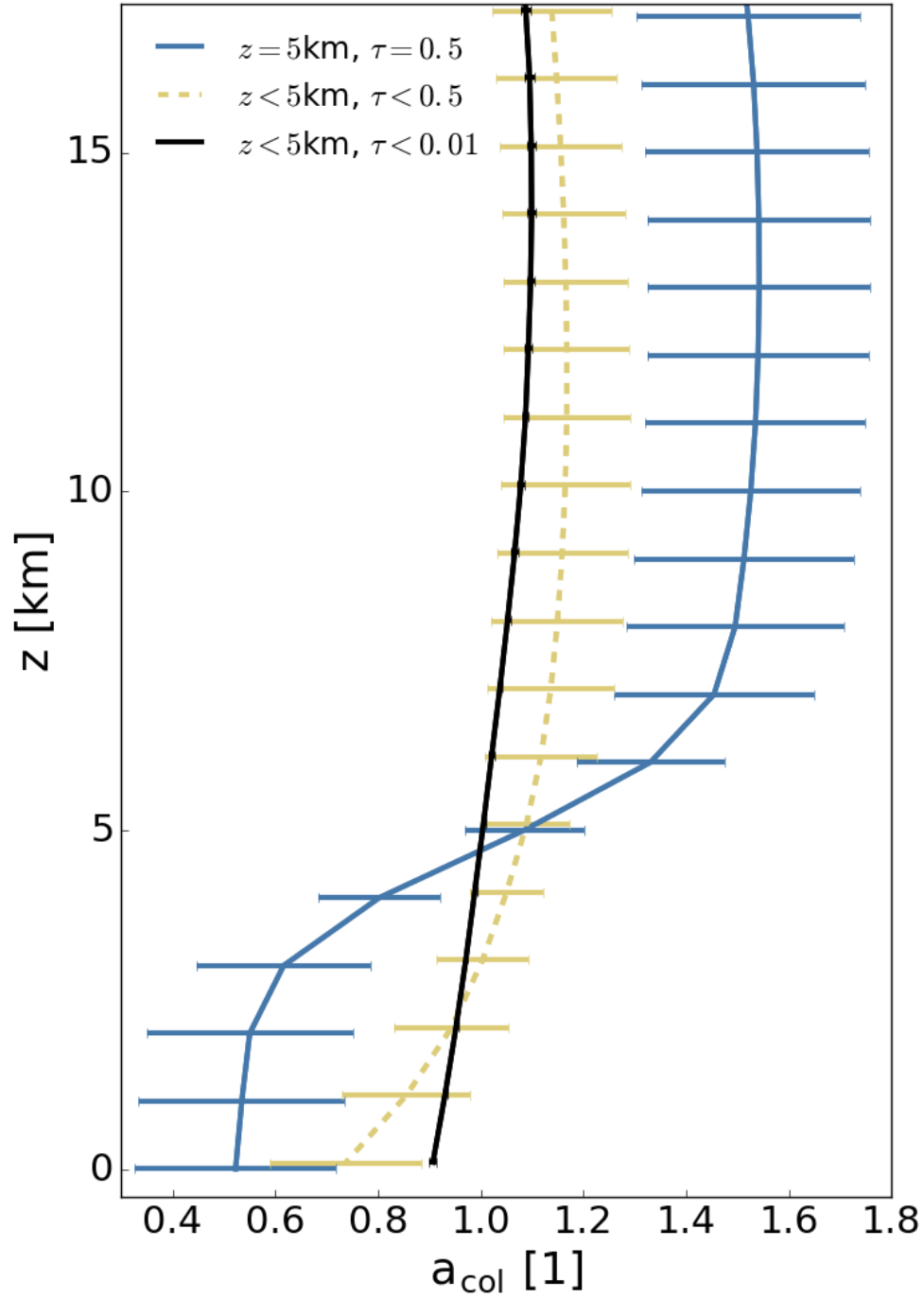


Figure 1. TROPOMI CO total column averaging kernels for November the 10th, 2017. The global average is shown for three different categories of cloudiness (black) strict cloud clearing, (yellow) clear-sky equivalent, and (blue) high optical thick clouds. The standard deviation is indicated as error bars.

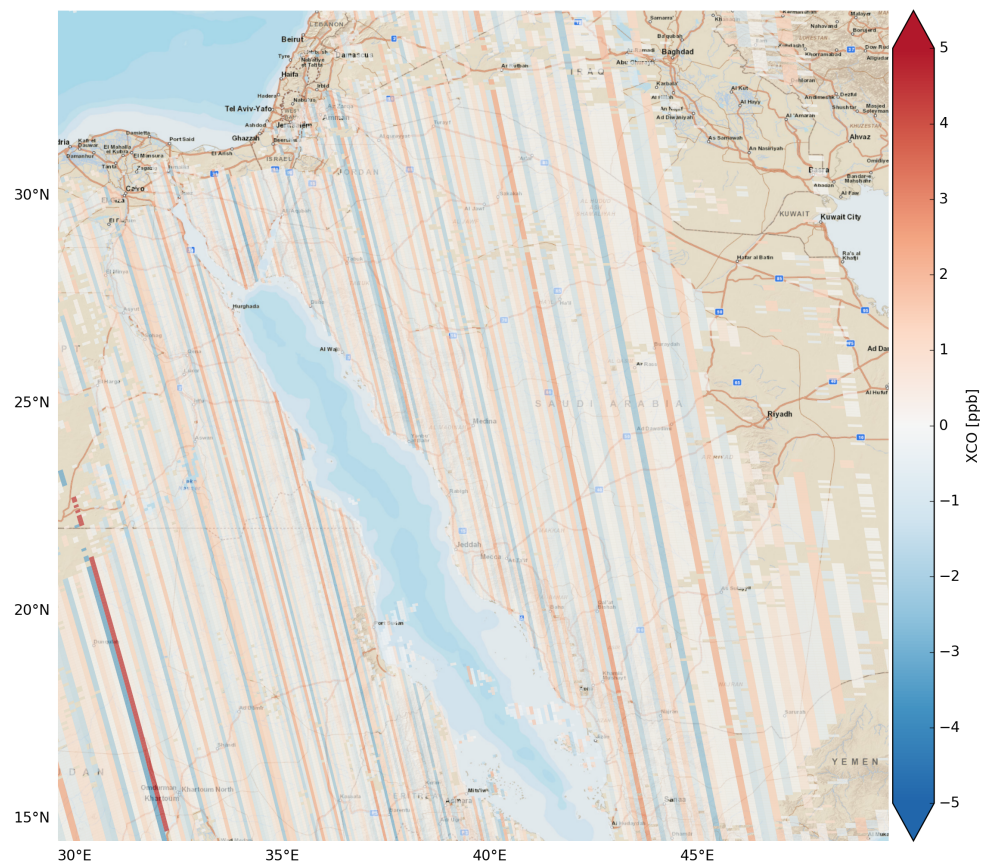


Figure 2. Stripe pattern derived by median filtering from a TROPOMI CO orbit above Saudi Arabia and Egypt on 12th November 2017.

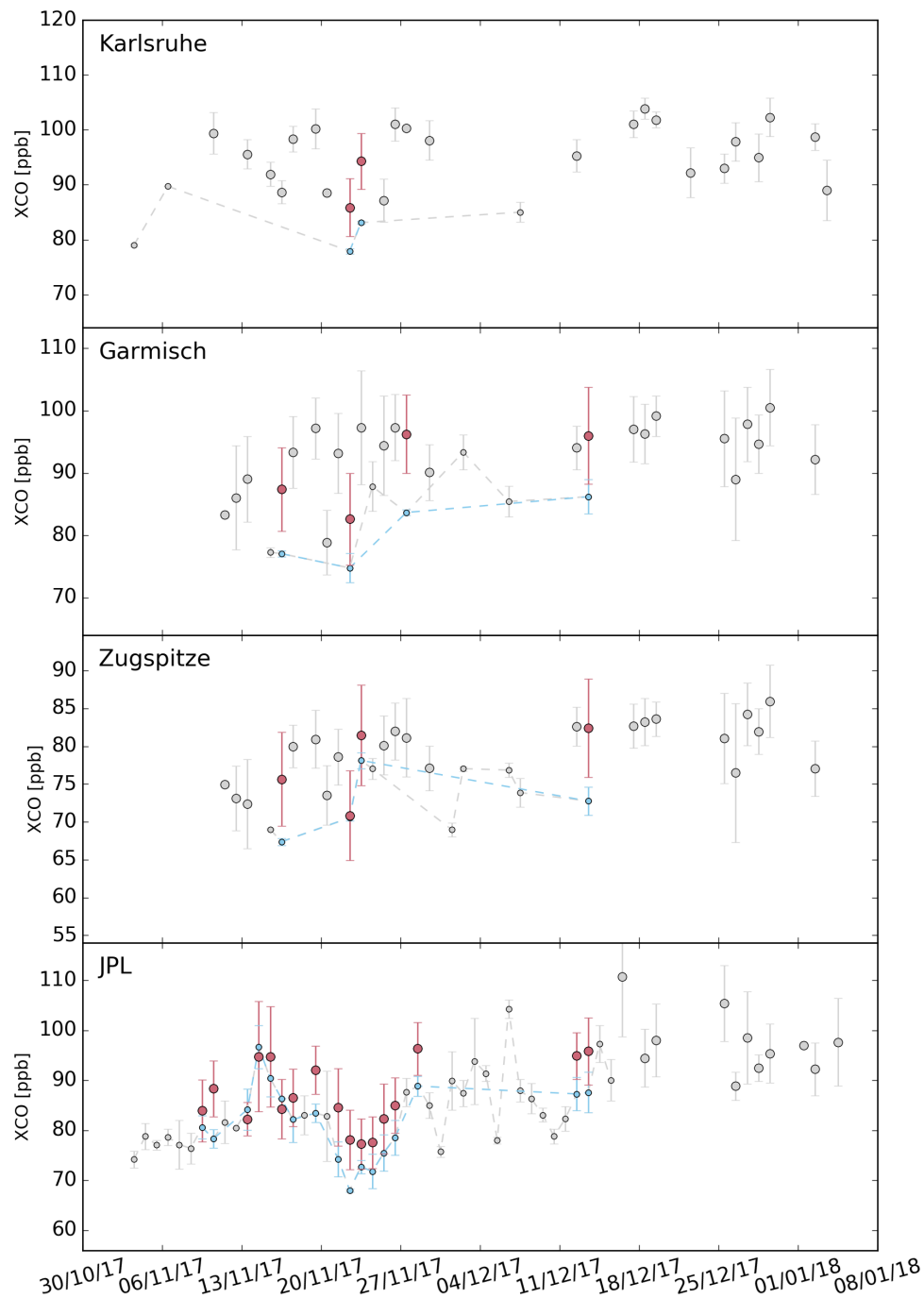


Figure 3. Daily means of dry air column mixing ratios (XCO) measured by TROPOMI (pink) and various TCCON stations (blue). A co-location radius of 50 km is used. The standard deviation of individual retrievals within a day is shown as an error bar. Data points without time coincidence between TCCON and TROPOMI are plotted in grey. No de-striping was applied on the TROPOMI data.

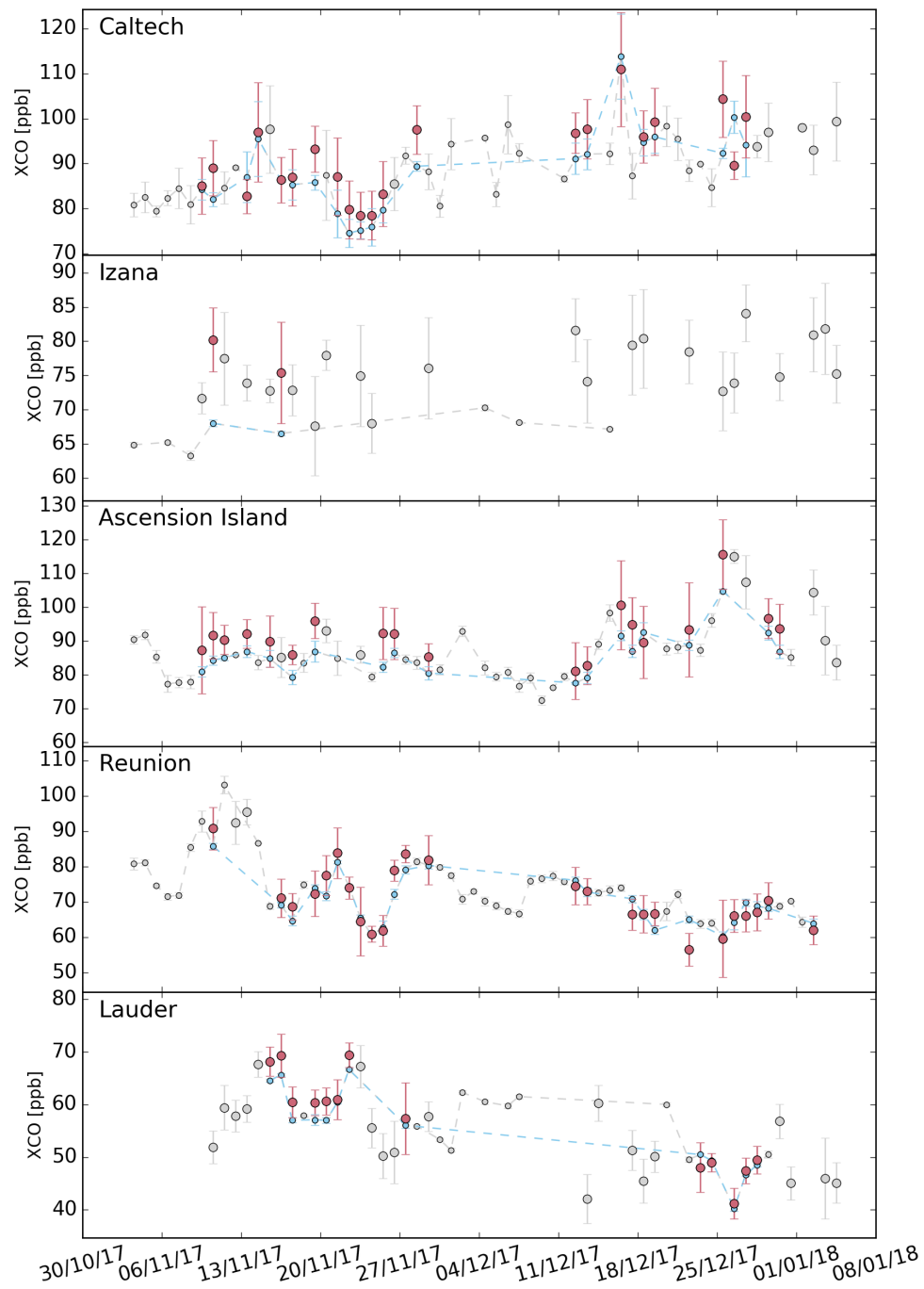


Figure 4. As Fig. 3 but with different TCCON stations.

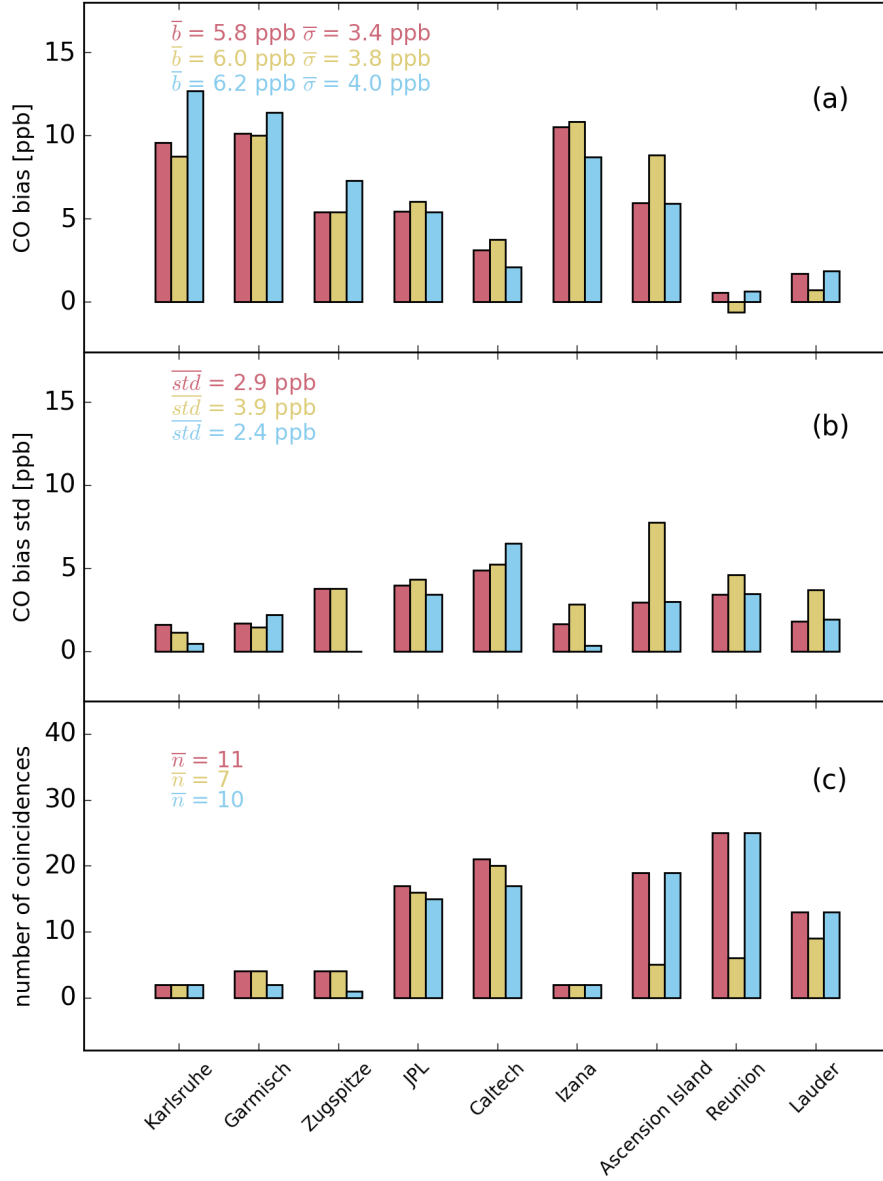


Figure 5. Mean bias (TROPOMI - FTS) between co-located daily mean XCO values (see Fig. 3, 4) of TROPOMI and TCCON (a), the standard deviation of the bias (b), and the number of coincident daily mean pairs (c). \bar{b} is the global mean bias (average of all station biases) and $\bar{\sigma}$ its [station-to-station standard deviation](#). \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. [No de-stripping was applied on the TROPOMI data.](#)

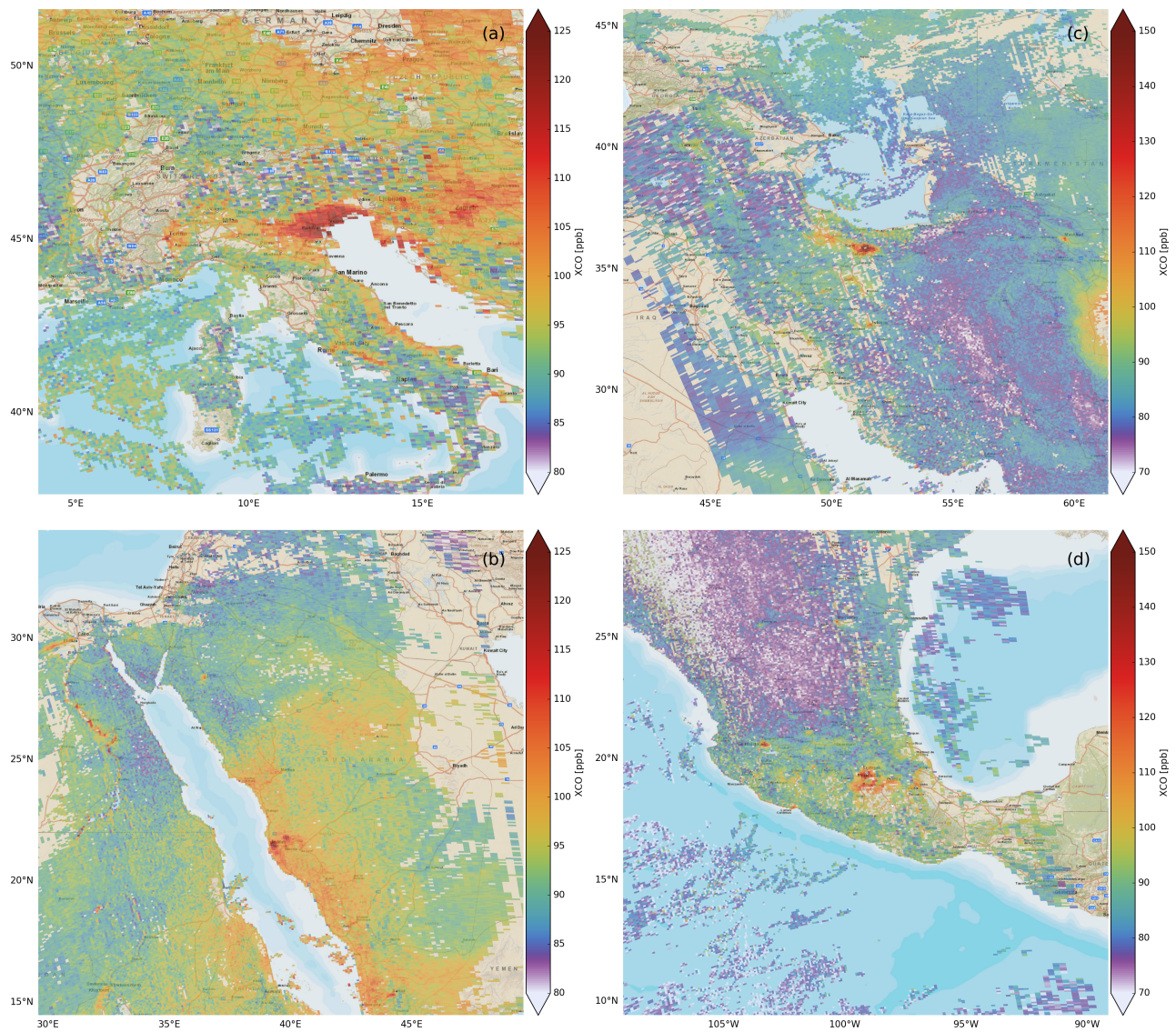


Figure 6. Total column mixing ratio (XCO) for individual TROPOMI ground pixels for (a) Italy on 25th December, (b) Saudi Arabia and Egypt on 12th November 2017, (c) Iran on 17th November 2017, and (d) Mexico on 25th November 2017. De-stripping was applied on the TROPOMI data.

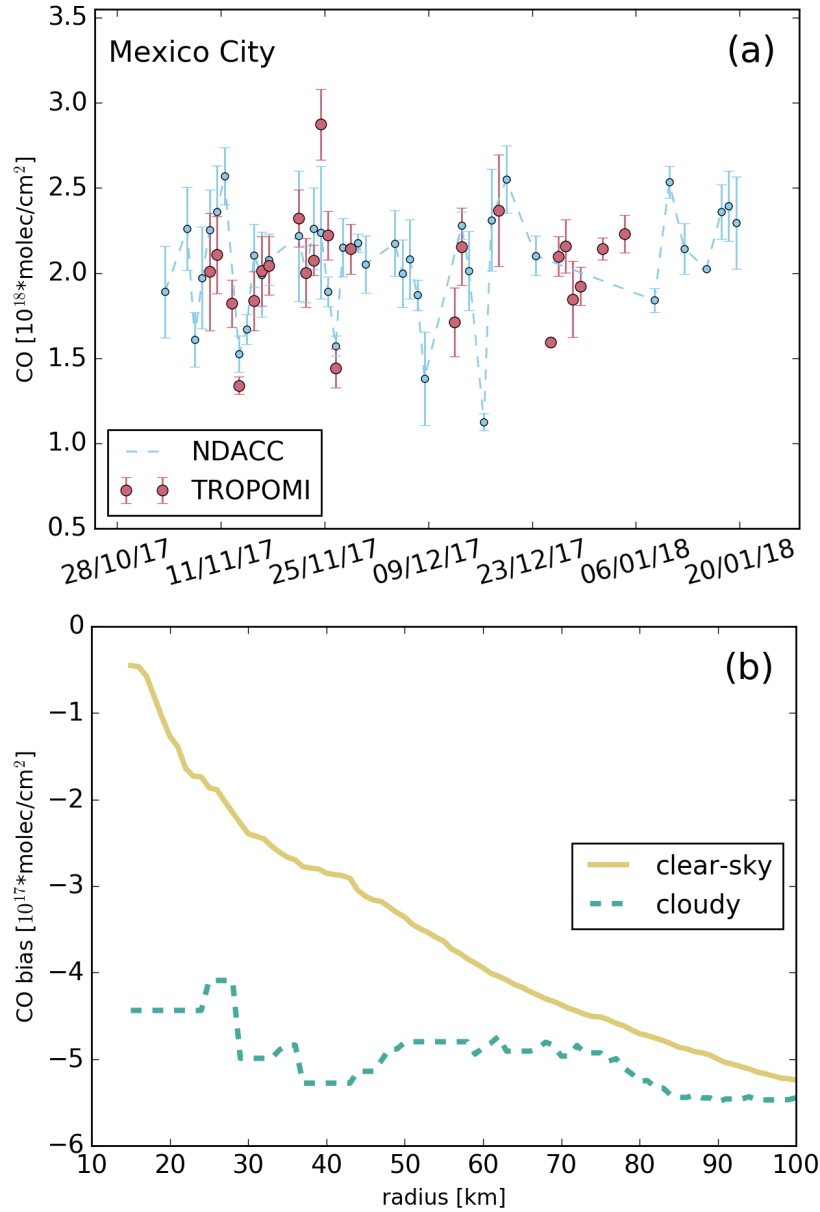


Figure 7. (a) Mexico City daily mean CO columns from TROPOMI (pink) and FTS (blue) with standard deviation of the individual retrievals (errors). TROPOMI observations are filtered for clear-sky within 15 km around the ground site. The TROPOMI columns are altitude corrected to the station elevation. (b) 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. For the smallest radius (15 km) we found 20 cloudy and 160 clear-sky collocations. However, for the widest radius (100 km) 92 cloudy and 4425 clear-sky collocations are found. No de-stripping was applied on the TROPOMI data.

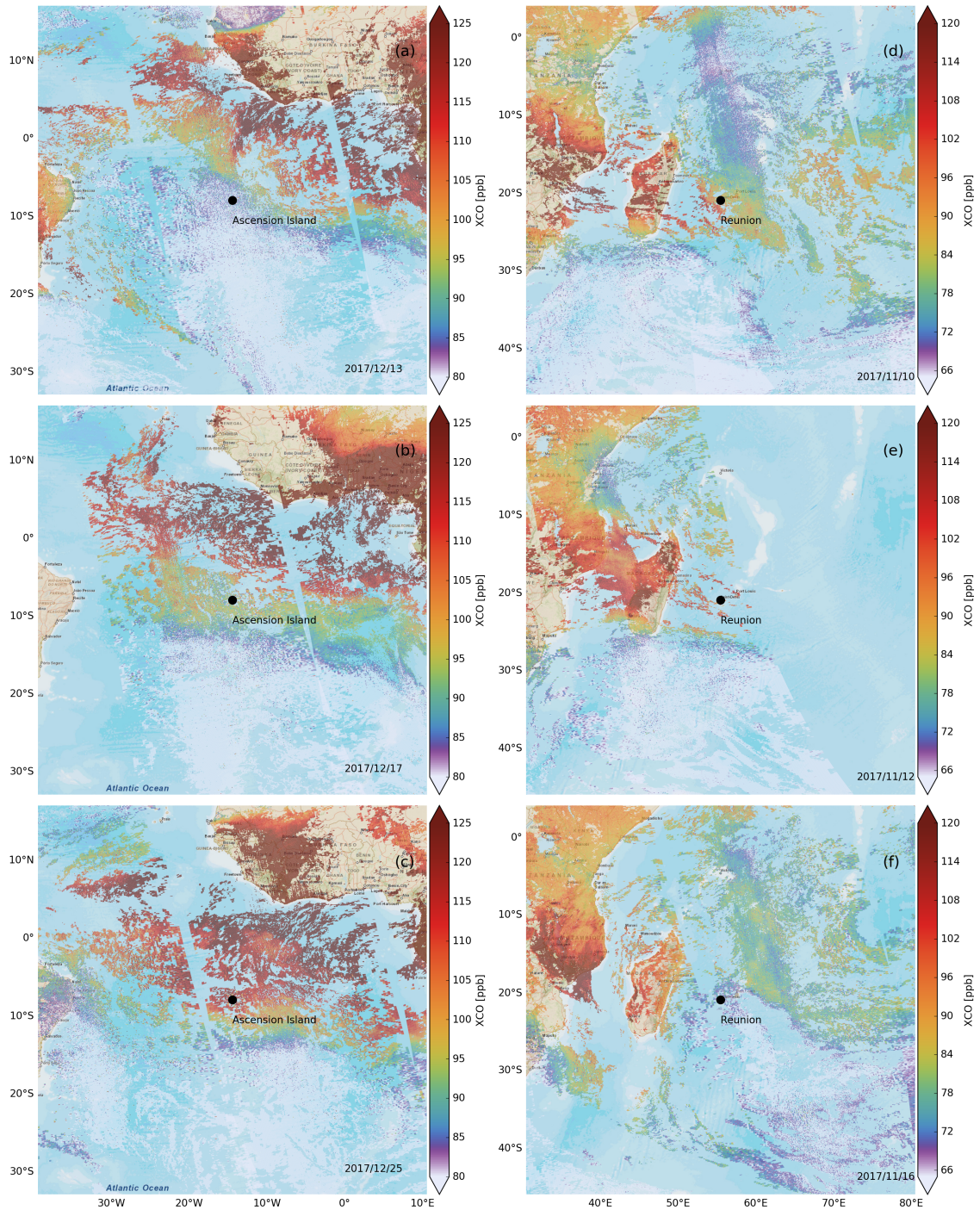


Figure 8. Total column mixing ratio (XCO) for individual TROPOMI ground pixels near Ascension Island on (a) 13th December, 2017, (b) 17th December, 2017, and (c) 25th December, 2017 and near Réunion on (d) the 10th November, 2017 (e) 12th November, 2017, and (f) 17th November, 2017. [De-stripping was applied on the TROPOMI data.](#)

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	
Réunion	−20.90	55.49	0.09	TCCON
Lauder	−45.04	169.68	0.37	TCCON