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Climate action, environment, resource
efficiency and raw materials

VERIFY

Observation-based system for monitoring and verification of greenhouse gases

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Dissemination and uptake (Who will/could use this deliverable, within the project or outside the project?)
Modelers performing CH ₄ inversions using TROPOMI data. Within the project, LCSE will use this deliverable for high resolution inversions over Europe.
Short Summary of results (<250 words)
VUA has characterized S5P TROPOMI retrieved column averaged mixing ratio of methane (XCH ₄) for use in inversions. A key part of this characterization is to use the measurements in an inversion using the TM5 atmospheric transport model and 4DVAR data assimilation. In addition to an inversion using the TROPOMI operational product, an inversion with a bias correction applied to the measurements has been performed. The TROPOMI XCH ₄ measurements were also compared to independent TCCON measurements.
Evidence of accomplishment (report, manuscript, web-link, other)
This report and an accompanying data file with the results. The data will be made available on the VERIFY portal after a period of one year to allow VUA to publish the associated results.

Version	Date	Description	Author (Organisation)
V0.1	15/03/2021	Creation of document	J. van Peet (VUA)
V1	17/03/2021	Formating/Delivery on the portal	Aur�lie Paquirissamy and Philippe Peylin (CEA)

1. Glossary	5
2. Executive Summary	6
3. Introduction	7
4. Main section	8
4.1. TM5 4DVAR inversion system	8
4.1.1. Sources	8
4.1.2. Initial concentration	8
4.2. Surface measurements	9
4.3. TROPOMI measurements	10
4.4. Bias correction	11
4.5. Results	12
4.6. Discussion	15
5. Conclusions	16
6. References	17

1. Glossary

Abbreviation / Acronym	Description/meaning
4DVAR	4D variational data assimilation
AK	Averaging Kernel
AOT	Aerosol Optical Thickness
CAMS	Copernicus Atmosphere Monitoring Service
CH ₄	methane
GOSAT	Greenhouse gases Observing SATellite
NIR	Near InfraRed
NOAA	National Oceanic and Atmospheric Administration
S5P	Copernicus Sentinel-5 Precursor. The satellite platform carrying TROPOMI.
SWIR	ShortWave InfraRed
TCCON	Total Carbon Column Observing Network
TM5	global chemistry transport model used for the CH ₄ inversions
TROPOMI	TROPOspheric Monitoring Instrument, the satellite instrument on board the Copernicus Sentinel-5 Precursor satellite.
XCH ₄	column averaged mixing ratio of methane

2. Executive Summary

VUA has characterized S5P TROPOMI retrieved column averaged mixing ratio of methane (XCH₄) for use in inversions. A key part of this characterization is to use the measurements in an inversion using the TM5 atmospheric transport model and 4DVAR data assimilation. In addition to an inversion using the TROPOMI operational product, an inversion with a bias correction applied to the measurements has been performed. The bias correction was derived by comparing the TROPOMI measurements to the results of an inversion using surface measurements only. The relative differences between the TROPOMI measurements and the surface-only inversion are within 2%. The TROPOMI XCH₄ measurements have also been compared to independent TCCON observations. The mean difference between co-located TCCON and TROPOMI XCH₄ for all available TCCON sites is between -0.4 and 0.8 ppbv. Within the uncertainty of the total column retrieval there is no systematic relation between the TCCON – TROPOMI difference and surface albedo or solar zenith angle, although the possibility remains of regional varying biases that cancel out in the bias corrected global mean.

3. Introduction

The overarching goals of WP4 are to deliver estimates of CH₄ and N₂O surface to atmosphere fluxes, including anthropogenic as well as natural sources, and to build this capacity into a pre-operational system, the Community Inversion Framework. Therefore, it is important to improve the understanding of the processes driving surface-atmosphere fluxes of CH₄ and N₂O, and reduce the uncertainties in their budgets and trends at national, regional and continental scales.

The current deliverable will explore the potential of TROPOMI XCH₄ measurements to improve flux estimates from atmospheric inversions. More specifically, this deliverable will characterise S5P TROPOMI retrieved column CH₄ mole fractions (XCH₄) for use in inversions. TROPOMI is the first instrument to use the 2.3 µm absorption band and will, therefore, require specific data selection criteria (e.g. for the presence of partial cloud cover), uncertainty characterization, as well as evaluation and correction of systematic errors.

A key part of this characterization is to use TROPOMI measurements in an inversion using the TM5 atmospheric transport model and 4DVAR data assimilation. In addition to an inversion using the TROPOMI operational product, an inversion with a bias correction applied to the measurements has been performed. The TROPOMI XCH₄ measurements have also been compared to independent TCCON observations.

4. Main section

4.1. TM5 4DVAR inversion system

The flux inversions within the current project are based on the TM5-4DVAR inversion system (P. Bergamaschi et al., 2010, 2013; Peter Bergamaschi et al., 2009; Meirink et al., 2008). It is also used within the Copernicus Atmosphere Monitoring Service project (CAMS¹) to derive methane fluxes on the global scale (Segers et al., 2021). In this section, we briefly summarize the main characteristics of the TM5 4DVAR inversion system (Bergamaschi et al., 2010, 2013; Bergamaschi et al., 2009; Meirink et al., 2008). The current implementation is based on the version used in CAMS to derive methane fluxes on the global scale (Segers et al., 2021).

In the current implementation, the model runs on a horizontal grid of 6° longitude × 4° latitude. In the vertical direction, the grid consists of 25 layers, which are a subset of the 137 layers used in the ERA-5 meteorology fields used to drive the model. The M1QN3 algorithm is used to minimise the cost function, which is an iterative process. The iteration process is stopped after a maximum of 40 iteration, or sooner if convergence is reached. The model runs from January 1st, 2018 till July 1st, 2019, but the results are only analysed from May 1st, 2018 till May 1st, 2019, to allow for four months spin-up and two months spin-down.

4.1.1. Sources

The inversion system optimizes sources in four source categories: wetlands, rice fields, biomass burning and other. Anthropogenic emissions fall into the “other” source category. These sources have been chosen such that their spatial and temporal characteristics enable the inversion system to distinguish their effect on the CH₄ concentration. For a detailed description of the emission inventories that are used to derive the a priori estimates, see Segers et al. (2021). The emissions that are used for the inversions in this deliverable are the same as those used in version v19r1 of the CAMS reanalysis².

4.1.2. Initial concentration

The inversion system requires an a priori estimate of the initial concentration. In the version used in the CAMS project, the initial concentration is part of the state vector and is therefore updated during the inversion. However, to facilitate the interpretation of our results and avoid a trade-off between updating emissions and initial concentrations, the optimization of the initial condition was disabled. This requires an accurate estimate of the initial concentrations, for which we use a daily averaged concentration field from the CAMS reanalysis (v18r1), which was optimized using surface measurements. A global map of column average mixing ratios, derived from this initial concentration field, is shown in Figure 1.

¹ <https://atmosphere.copernicus.eu/>

² <https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-greenhouse-gas-inversion?tab=overview>

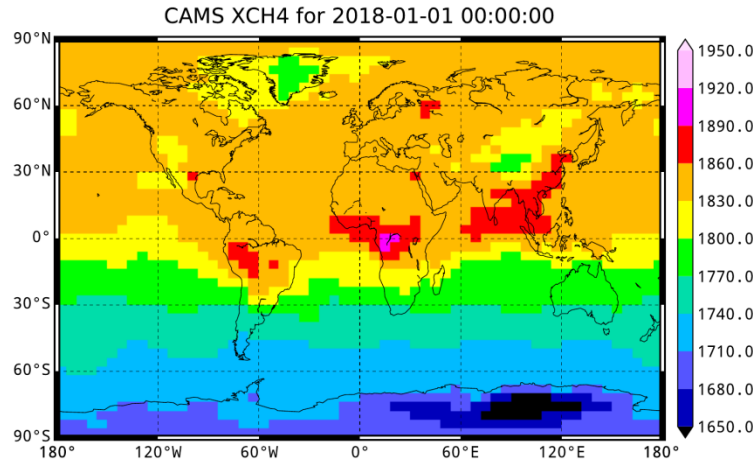


Figure 1: Initial methane concentration field used for the inversions in the VERIFY project.

4.2. Surface measurements

For inversions with surface measurements only, we use the atmospheric methane dry air mole fractions from the National Oceanic and Atmospheric Administration (NOAA) Global Monitoring Laboratory (GML) carbon cycle cooperative global air sampling network (Dlugokencky et al., 2020). The locations of the surface measurements are shown in Figure 2. The NOAA surface measurements are released ahead of the official release for the CAMS reanalysis. Here, the same observations have been used as in version v19r1 of the CAMS reanalysis.

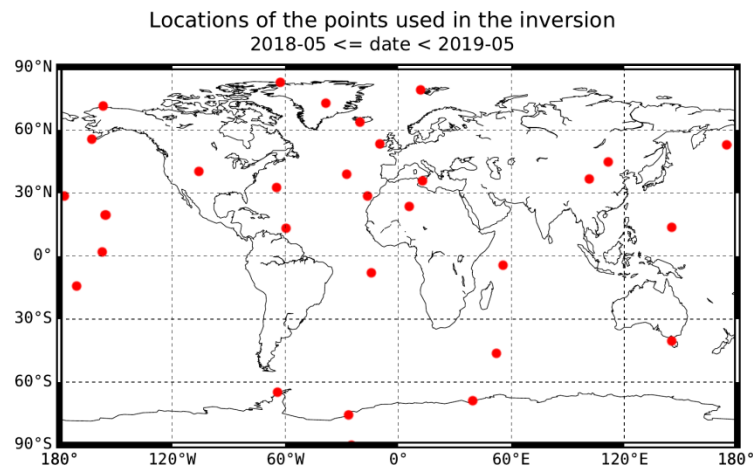


Figure 2: Locations of the surface measurements from the NOAA network.

The inversion results have also been validated with independent measurements from the Total Carbon Column Observing Network (TCCON, 2017). The TCCON measurements are binned into one hour time intervals, which is the same interval as the inversion system uses to save TCCON-

comparison relevant model output at the TCCON locations. The model output is compared to the TCCON measurements taking the averaging kernel (AK) into account. When comparing TROPOMI and TCCON measurements, only measurements that are closer than 300 km and fall in the same time interval are considered. Both TROPOMI and TCCON retrievals have associated AKs, which are taken into account following Rodgers et al. (2003).

4.3. TROPOMI measurements

In VERIFY T4.3.2, the inversion system described in section 4.1 has been updated to make optimal use of TROPOMI XCH₄ measurements. Within VERIFY, the operational TROPOMI XCH₄ product has been used, which is available online³. The product version varies over the model run time interval from 1.1.0 on January 1st, 2018 to 1.2.0 on July 1st, 2019. More information on the differences between the version numbers can be found online⁴. The operational product contains an XCH₄ dataset that is bias corrected with respect to GOSAT measurements (Hasekamp et al., 2019), which is used to characterise the TROPOMI XCH₄ retrievals. A scientific TROPOMI XCH₄ product has been recently released (Lorente et al., 2021), which is not used here since it was not available for the full time period when running the inversions.

Due to the high spatial resolution of TROPOMI, it was too expensive to run the inversions with the original measurements. Therefore, the measurements per orbit have been gridded into superobservations at the same horizontal resolution as is used in the inversion system (6° longitude × 4° latitude). Only measurements with a quality descriptor flag of 1 (i.e. cloud-free pixels) have been used. The gridded value is the weighted average of all observations that fall into the gridcell, with weights being equal to the uncertainty values of the original measurements. The uncertainty of the superobservations is quantified by the standard deviation of the TROPOMI XCH₄ measurements that fall into the same gridcell, with a minimum of two times the weighted average of the retrieval uncertainties of the measurements.

The gridded measurements and model data are compared according to the notes in the product user manual (Apituley et al., 2017), taking the averaging kernel (AK) into account. During this comparison, the model CH₄ profile has to be interpolated to the same vertical grid as was used in the retrieval. Model layers for which the pressure is larger than the retrieval surface pressure are ignored. If the model surface pressure is smaller than the retrieval surface pressure, the pressure difference between both surface pressures is subtracted from the model surface pressure. The model pressure levels between that value and the retrieval surface pressure are then redistributed proportionally over the new interval, but the CH₄ concentrations are not changed. After this step, the surface pressures of the interpolated model profile and retrieval are equal. The modelled CH₄ profile is redistributed over the retrieval layers using the fractions by which the model layers overlap with the retrieval layers, based on the pressure of the layer boundaries.

3 <https://scihub.copernicus.eu/>

4 <http://www.tropomi.eu/data-products/methane>

4.4. Bias correction

The TROPOMI measurements have been compared with the results of an inversion using NOAA surface measurements only. Vertical interpolation and AK have been applied according to the procedure described in section 4.3 to calculate the model's XCH₄ as it would be observed by TROPOMI. Latitudinal and seasonal differences between this dataset and the TROPOMI data are similar as found using GOSAT satellite data, and are explained by inaccuracies in the representation of the stratospheric age-of-air in the TM5 transport model. To avoid that this error is compensated by emission adjustments in the inversion, it has been treated as a bias that has been subtracted from the TROPOMI data prior to their use in the inversion. The procedure is the same as described in Monteil et al. (2013) and is used also in the CAMS reanalysis (Segers et al, 2021). The bias is calculated as the relative difference $100.0 \cdot (\text{tm5_xch4} - \text{tropomi_xch4}) / \text{tropomi_xch4}$, where tm5_xch4 and tropomi_xch4 are the column averaged mixing ratios from the surface only inversion and TROPOMI respectively. The results are binned according to time (monthly bins) and latitude. The size of the latitude bins is 4°, which corresponds to the latitudinal grid size of the inversion system (section 4.1) and the superobservations (section 4.3). The mean differences per month can be used as a correction factor on the latitude data before gridding and assimilation. The monthly mean biases as a function of latitude are shown in Figure 3. From this point onward, “bias corrected TROPOMI measurements” will refer to TROPOMI measurement with this time-latitude bias correction applied, in addition to the retrieval bias correction that is provided by the L2 data.

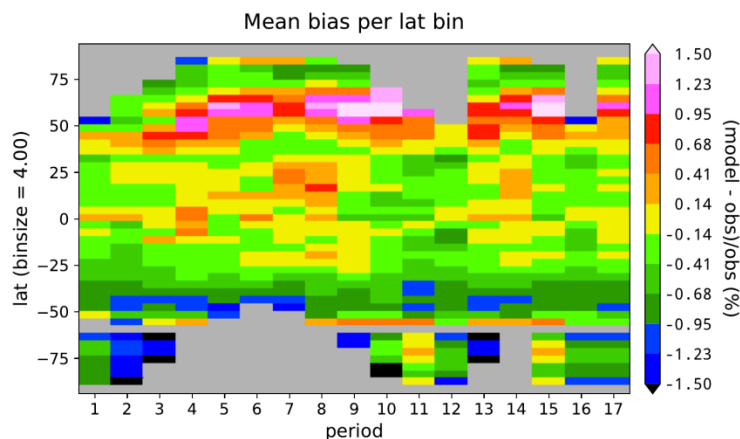


Figure 3: Mean bias for TROPOMI. Period 1-12 is January-December; 13-16 are the seasons: MAM-DJF; and 17 is the yearly mean.

The monthly mean biases as a function of latitude are stored in an hdf-5 file, which is also part of the current deliverable. The attributes and datasets in the hdf-5 file are described in tables Table 1, Table 2, and Table 3.

Name	Type	Note
h5_version	64 bit integer array (shape = 3)	Hdf5 library version (major, minor, and bugfix nr.)
file_version	scalar string	
tm5_run_id	scalar string	
start_date	scalar string	start date of the interval for which the bias correction is derived
end_date	scalar string	end date of the interval for which the bias correction is derived
merge_type	scalar string	None means no horizontal gridding, i.e. the original measurements have been used to derive the bias correction

Table 1: Global attributes in the bias correction hdf5 file.

Name	Type	Note
lat_bin_lbound	64 bit float array (shape = 47)	lower boundaries of the latitude bins used
lat_bin_ubound	64 bit float array (shape = 47)	upper boundaries of the latitude bins used
periods	String array (len = 9, shape = 17)	names of the periods used (i.e. time bins)

Table 2: dimensions and bin boundaries in the bias correction hdf5 file.

Name	Type	Note
lat_count	64 bit int array	number of collocations
lat_mean	64 bit float array	mean bias
lat_sdev	64 bit float array	standard deviation of the bias

Table 3: datasets characterising the bias in each time-latitude bin, each array shape = 17 × 47

4.5. Results

In total, three inversions have been performed using the setup described in the previous sections: an inversion using NOAA flask measurements only, an inversion using TROPOMI measurements only, and an inversion using the bias correction described in section 4.4 applied to the TROPOMI measurements. Figure 4 shows the differences between TM5 and the measurements that are used in the inversion in the top row, and the differences between TM5 and TCCON in the bottom row.

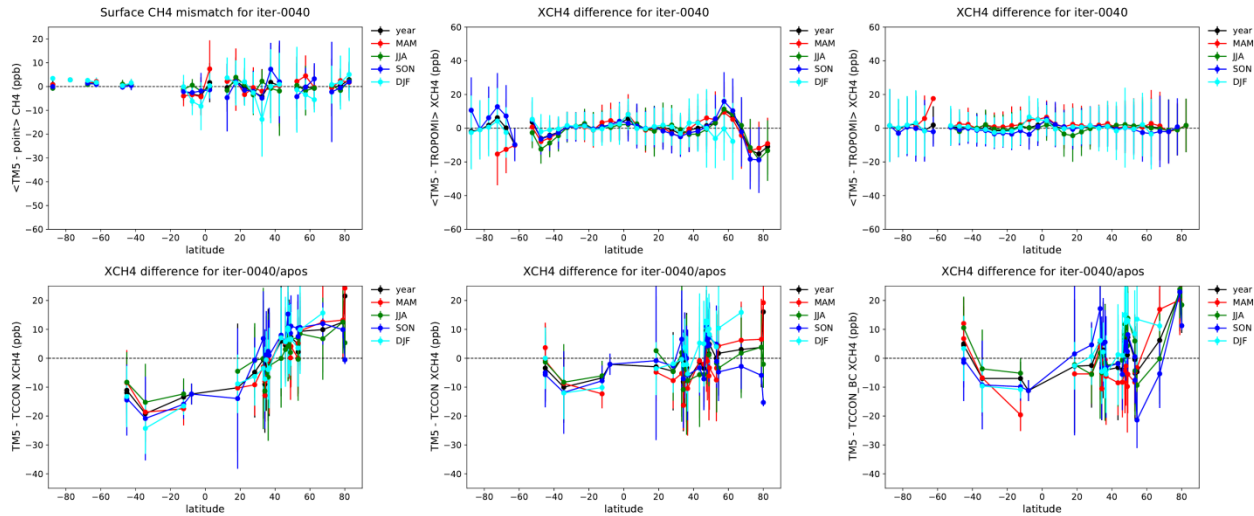


Figure 4: Differences between TM5 and the measurements that are used in the inversion (top row) and TCCON (bottom row). From left to right: inversion with surface measurements only, with TROPOMI measurements only and with the bias corrected TROPOMI measurements only. Note that the top left plot gives the difference in surface CH₄ mixing ratio, while the top-middle and top-right plots give the difference in XCH₄.

At high latitudes, the average difference between TM5 and TROPOMI XCH₄ can reach up to 20 ppbv (top-middle), but applying the bias correction from section 4.4 fixes that (top-right). There is a North-South gradient visible in the comparison between TM5 and TCCON (bottom row), which is somewhat smaller for the inversion with TROPOMI measurements only compared to the other two inversions. Note that in the bottom-right plot, the bias correction has been applied as well, for a correct comparison between TCCON measurements and the inversion results.

As an example of the emission updates, Figure 5 shows the total prior emissions for September 2018 in the top left, and the differences between the prior and posterior emissions for the three inversions in the other panels.

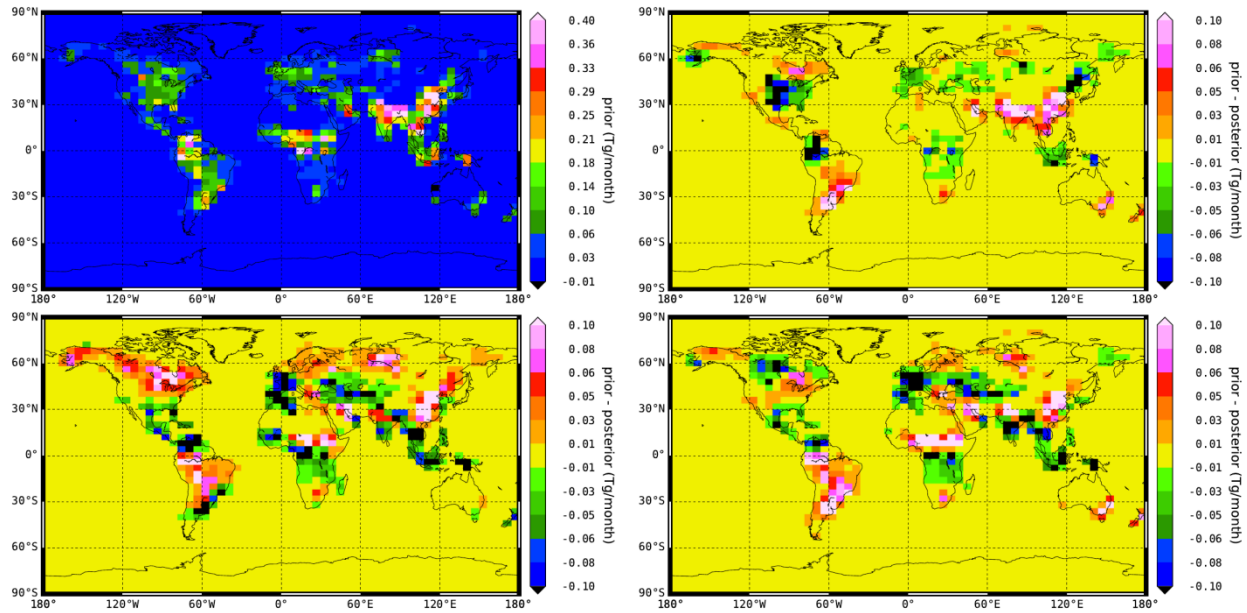


Figure 5: Inversion-derived CH₄ emission adjustments. Top left: prior emissions. The other panels show the difference between prior and posterior emission. Top right: inversion with surface measurements only, bottom left: inversion with TROPOMI measurements only, bottom right: inversion with bias correction applied to TROPOMI measurements.

The TROPOMI XCH₄ measurements have also been compared to independent TCCON observations. Since both TROPOMI and TCCON retrievals have associated averaging kernels (AKs), these should be taken into account as well (Rodgers, 2003). The profiles from the TM5 inversion using surface measurements only have been used as a common a priori. Figure 6 shows the XCH₄ ratio of TCCON/TROPOMI, plotted against various parameters. The black dots show the ratio of the a priori corrected measurements, the red dots show the ratio of the TM5 profile as observed by TCCON and TROPOMI respectively (with the a priori profiles used in the retrievals). The latter is an indication of the impact of the AK differences between TCCON and TROPOMI on the column averaged mixing ratio.

The mean difference for all available TCCON sites between co-located TCCON and not bias corrected TROPOMI XCH₄ is 0.9 ± 17.8 ppbv. When using the TM5 inversion as a common a priori, the mean increases to 2.1 ± 18.2 ppbv. If the bias correction from section 4.4 is applied to the TROPOMI data, the mean difference is -0.4 ± 18.7 when using the retrieved measurements directly and 0.8 ± 18.8 when using TM5 as a common a priori. Lorente et al. (2021) report a mean difference of -3.4 ± 5.6 ppbv, based on a selection of the TCCON stations over a two year period. It should be noted that they report the mean and standard deviation of the station biases instead of the mean and standard deviation of all measurements combined reported above.

Within the uncertainty of the total column retrieval there is no systematic relation between the TCCON – TROPOMI difference and surface albedo or solar zenith angle, although the possibility remains of regional varying biases that cancel out in the bias corrected global mean.

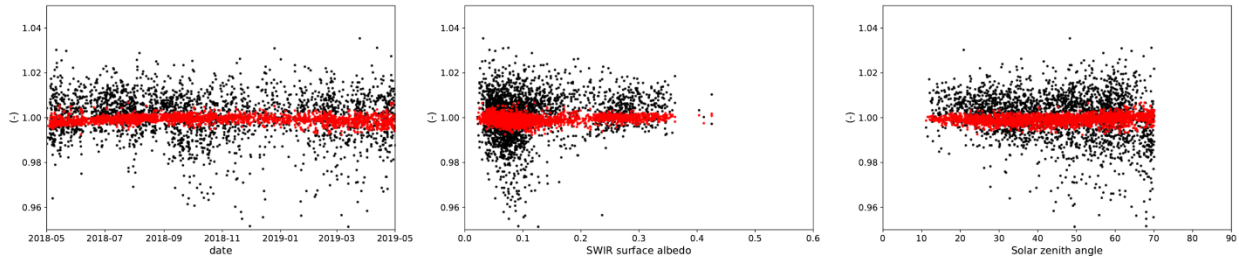


Figure 6: TCCON XCH₄ / TROPOMI XCH₄ as a function of time, surface albedo in the SWIR channel and solar zenith angle. The black dots show the ratio of the a priori corrected measurements, the red dots show the ratio of the TM5 profile as observed by TCCON and TROPOMI respectively (with the a priori profiles used in the retrievals).

4.6. Discussion

In section 4.4, we compared TROPOMI XCH₄ measurements against the results from an inversion using surface measurements only. The derived bias correction was used to correct the TROPOMI measurements in a subsequent inversion. However, we could also have applied the bias correction to the model CH₄ concentration, and the result would be the same. In other words, are the TROPOMI measurements biased against the model, or is the model biased against TROPOMI?

There are inaccuracies in the stratospheric age-of-air in the TM5 transport model, which explain some of the differences found between TROPOMI and the inversion using surface measurements only. A comparison with TROPOMI and TCCON measurements showed a dependence of the ratio between the two instruments that depends on the surface albedo in the SWIR channel (Lorente et al., 2021). This might be an important effect at high latitudes, such as the increase and decrease of the TM5 – TROPOMI difference that is visible poleward of 50°N in the top-middle panel in Figure 4. The bias at high latitudes might therefore be mainly an instrumental effect, but at lower latitudes the attribution of the bias to model or observations will be less clear.

The bottom two panels of Figure 5 show significant differences between a priori and a posteriori emissions around the Sahara when using TROPOMI measurements with respect to the inversion using surface measurements only. TROPOMI retrievals are sensitive to high aerosol optical thickness (AOT), which can be caused by desert dust. Since there are few CH₄ sources in the Sahara, the model will then try to update the CH₄ concentration over the Sahara by changing the sources close to the Sahara. For this deliverable, we used TROPOMI retrievals with a quality factor of 1, which are cloud free pixels with a maximum AOT in the NIR channel of 0.3. The sensitivity of TROPOMI retrievals to AOT could be further characterised by lowering the AOT limit and to observe how the inversion reacts in and around regions such as the Sahara.

5. Conclusions

In this deliverable, we have characterized TROPOMI XCH₄ measurements for use in atmospheric inversions. A key part of this characterization were the 4DVAR inversions using TROPOMI measurements and the TM5 atmospheric transport model. The bias between TROPOMI measurements and the inversion using surface measurements only were within 2%, and are made available as an hdf-5 file accompanying this deliverable. The bias is influenced by issues in both the TROPOMI instrument and the TM5 transport model. Therefore, care should be taken when applying the biases to different models. Modellers might even derive their own bias correction, and use the one provided here as a reference.

The TROPOMI XCH₄ measurements were also compared directly to independent TCCON data, resulting in a mean difference of 0.8 ± 18.8 . Within the uncertainty of the total column retrieval there is no systematic relation between the TCCON – TROPOMI difference and surface albedo or solar zenith angle.

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