A win-win success story: the HCHO and stratospheric NO2 TROPOMI/S5P validation using the FTIR ground-based network

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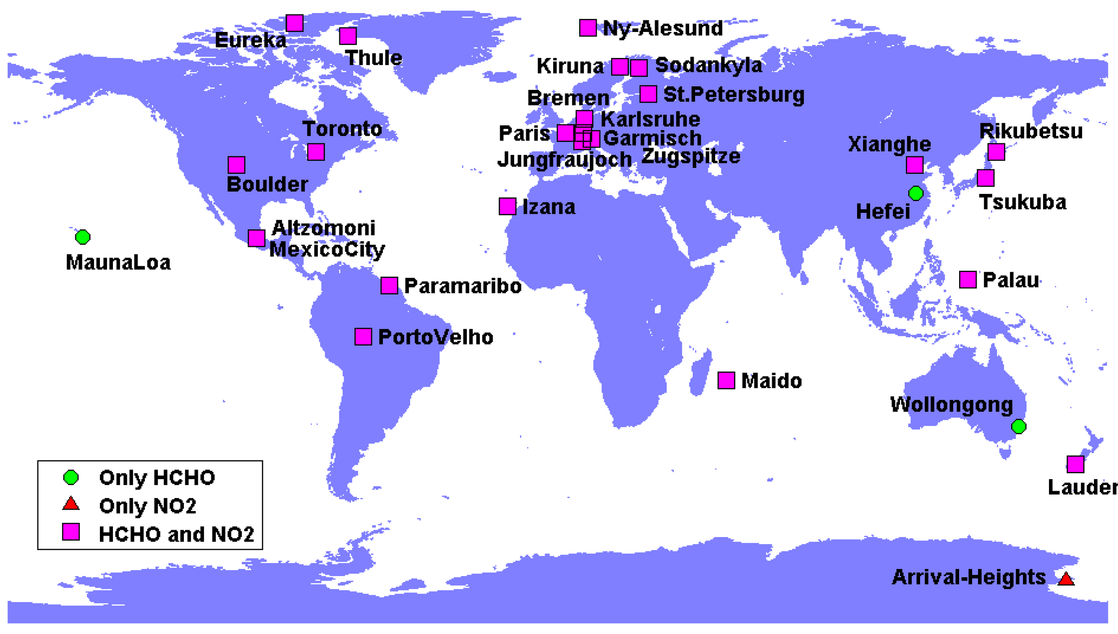
As part of the S5P Validation Team, we present the results of the **NIDFORVal project** (AO ID 28607: S5P NItrogen Dioxide and FORmaldehyde VALidation using NDACC and complementary FTIR and UV-Vis DOAS ground-based remote sensing data), **focusing on the FTIR (Fourier Transform Infrared)** part.

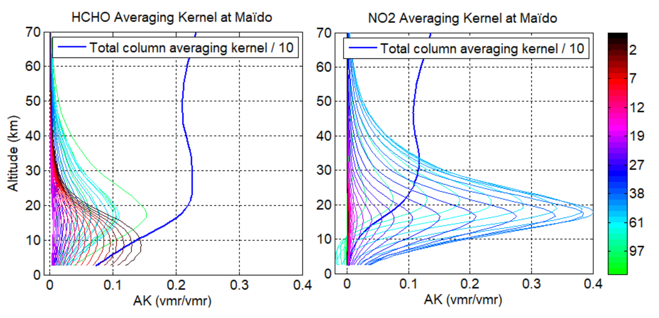
Why a "win-win" success ?

1) This project is a huge success for the FTIR network:

- Before the S5P launch, an extensive number of ground-based FTIR stations were already providing excellent data sets for the satellite validation of CO and CH4, through the TCCON (Total Carbon Column Observing Network) and NDACC (Network for the Detection of Atmospheric Composition Change) networks. **The situation was much less advanced for formaldehyde (HCHO) and nitrogen dioxide (NO2), these two gases being not official TCCON nor NDACC target species. Only a very few FTIR stations were producing HCHO and/or NO2 data**, and using different retrieval settings, which complicates the interpretation of satellite validation.
- Within this project, and thanks to the very good collaboration within the InfraRed Working group (https://www2.acom.ucar.edu/irwg) of NDACC, **we have obtained a unique harmonized HCHO data set (Vigouroux et al., 2018) currently at 28 FTIR stations, providing a wide range of observation conditions and ensuring a consistent validation among the sites.**
- Thanks to this work, **HCHO has now been declared as an official NDACC target species.**
- In addition to the S5P validation presented here, **our FTIR HCHO data set are used for many other applications** (Souri et al., 2022; Hyeong-Ahn Kwon, in preparation; Stavrakou et al., in preparation;…).
- Based on this success, **we have more recently optimized the NO2 retrieval settings and applied them to 25 FTIR stations, building a harmonized reference data set for satellite validation** (Vigouroux et al., in preparation).



- FTIR **solar absorption remote sensing** measurements.
- **Retrieval codes** are based on **Optimal Estimation** (**Rodgers, 2000**).



- The sensitivity is mainly in the **troposphere for HCHO**, and in the **stratosphere for NO2**.

FTIR uncertainty	HCHO	NO2
Random (depending mostly on clean vs polluted sites)	Median $2.6x10^{14}$ (1 to $11x10^{14}$) molec/cm^2	Median 3.3E14 (1.3 to 7.7E14) molec/cm^2
Systematic	Median 14%	Median 10%.

2) We celebrate here the S5P mission success:

- The **high quality of the HCHO S5P data** have been demonstrated within this project (Vigouroux et al., 2020, for the FTIR part, De Smedt et al., 2021 for the UV-Vis DOAS part). This validation work was the first extensive study to demonstrate the **good accuracy and precision of the TROPOMI HCHO data, which were both well below the pre-launch requirements. The TROPOMI HCHO uncertainty budget has also been validated through these comparisons.**
- We present **here an update of the HCHO S5P validation using the FTIR network**, including more than 4 years of data, and different versions of the S5P products.
- The NIDFORVal project (with G. Pinardi as co-PI for the UV-Vis part) has also contributed to demonstrate the **high quality of the NO2 tropospheric, stratospheric and total column S5P** data sets, using the MAX-DOAS, **Zenith-Scattered-Light ZSL**, and direct sun ground-based data respectively (Verhoelst, et al. 2021).
- The **FTIR stratospheric NO2 data set can complement the ZSL observations.** Indeed the latter are made during sunset and sunrise which imposes the use of a photochemical box model to adjust the observations to the time of the TROPOMI overpasses, while the FTIR measurements are made during the whole day**, allowing direct comparison between measurements that are collocated in time.** Conclusions about the accuracy and the precision of the S5P stratospheric NO2 product are drawn and compared to the ones obtained using ZSL data set (Verhoelst et al., 2021). Furthermore, **diurnal cycle comparisons can be made using the FTIR data**, which is not the case for the ZSL network.

Validation Methodology

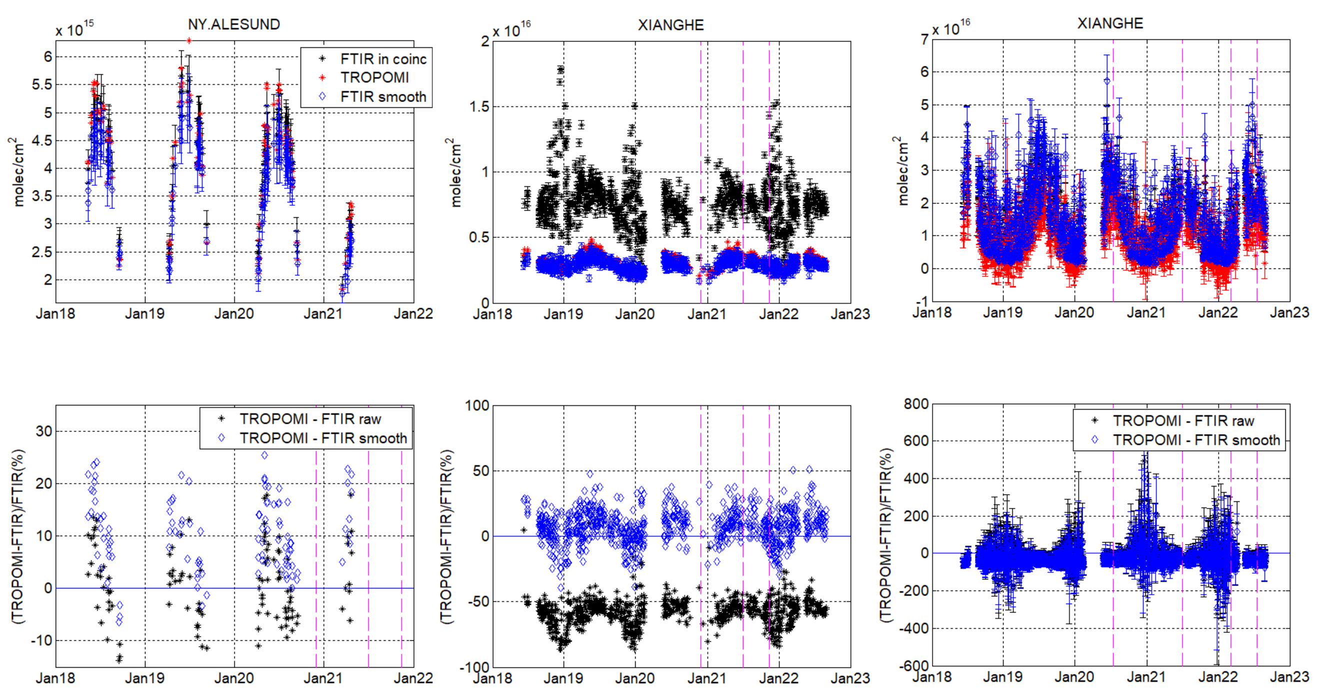
1) Collocation is **not** above the FTIR site: we calculate the **position along the line-of-sight at the altitude where the FTIR averaging kernels shows the maximum of sensitivity: free troposphere** and **~30-35km** for HCHO and NO2, respectively.

- S5P pixels are then selected within:
  - **20 km** of this position for **HCHO (about 35 pixels)**
  - **50 km** of this position for **NO2 (about 150-200 pixels).**
- Time coincidence is **±3h (HCHO)** and **±1h (NO2)** around the **satellite overpass time.**

2) Compared pairs:

- The FTIR a priori profile is substituted with the TROPOMI one to **take into account the different TROPOMI and FTIR a priori profiles** (Rodgers and Connor, 2003).
- The corrected profile is **smoothed with the TROPOMI averaging kernel AK** (Rodgers and Connor, 2003). In this process, since the TROPOMI AK are zero below the tropopause for stratospheric NO_2, the tropospheric part of the FTIR profile is removed, and only stratospheric columns from both products are indeed compared. For HCHO, the similar AK of both instruments give a smaller impact of this smoothing operation.
- Both individual manipulated FTIR columns and S5P manipulated pixel columns are then **averaged**.

NO2 **HCHO**



Effect of removing the tropospheric part by using the TROPOMI stratospheric NO2 AK: about 10% at clean site (Ny-Alesund); much larger for polluted sites (e.g. Xianghe > 50%).

The vertical magenta bars are showing when **new S5P processor versions are released.**

3) Metrics for validation :

- The **bias** at a single station is estimated by the median relative difference:

  BIAS=Median[(TROPOMI-FTIR)/FTIR]

  To be compared to systematic error budget / S5P requirements.

- The **dispersion** at a single station is estimated by the scaled median absolute deviation of the differences TROPOMI-FTIR:

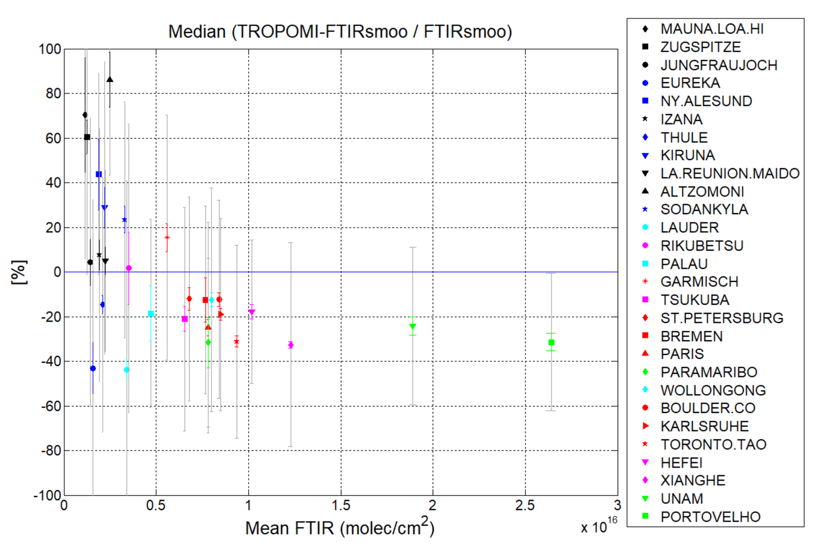
  MAD=1.4826*Median[ABS(DIFF-Median(DIFF))]

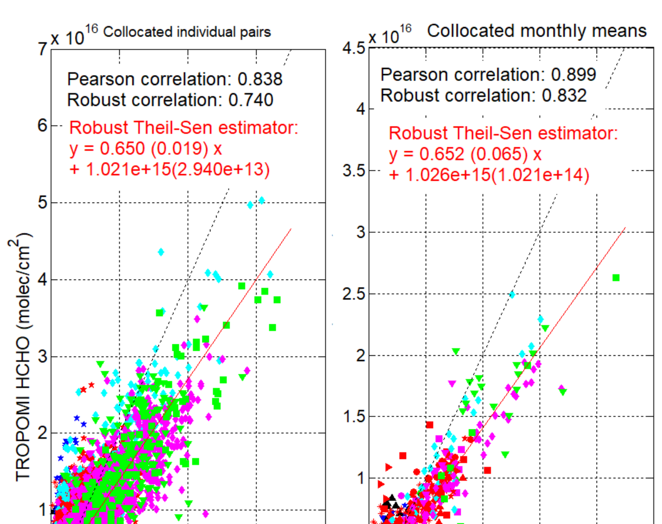
  The scaling factor of 1.4826 ensures that for a normal distribution, the MAD = 1sigma standard deviation.

  To be compared to random error budget / S5P requirements.

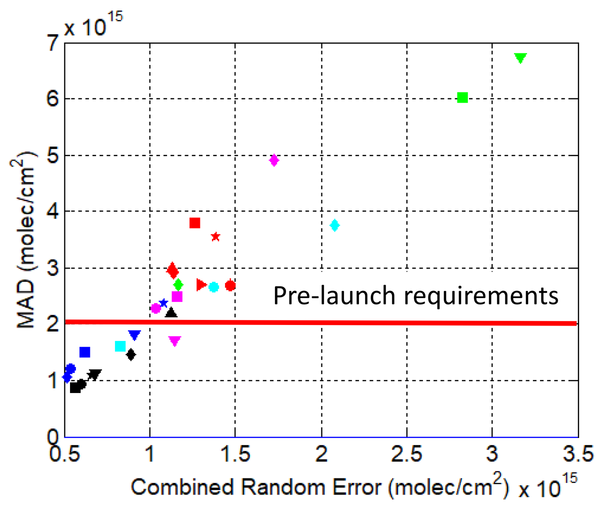
HCHO validation results

- Results presented here are updated from Vigouroux et al., ACP, 2020. Check this reference for more details.
- Our results are also available and updated in the quarterly validation reports here: https://mpc-vdaf.tropomi.eu/

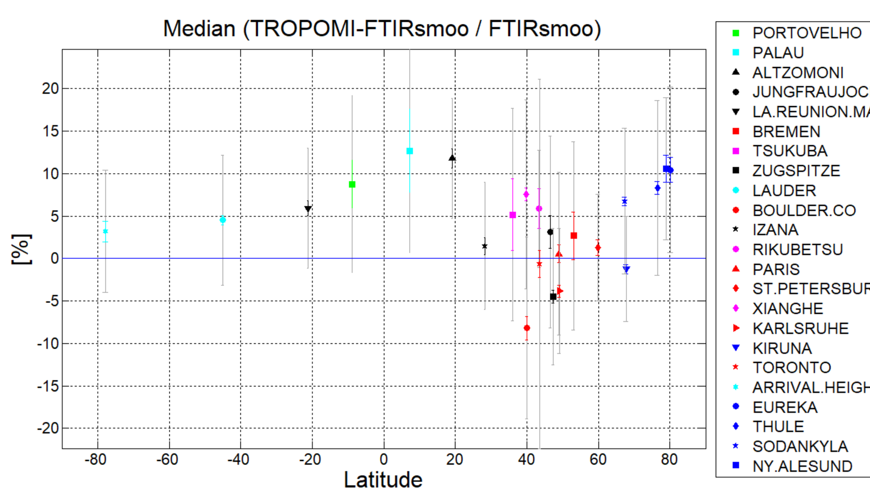




- The **biases are within pre-launch requirements**: accuracy < 80%.
- The S5P HCHO bias depends on the HCHO-levels: **clean sites show positive biases and polluted sites negative biases.**
- Also well described in the scatter plots: S5P HCHO has a **constant bias (offset=1.02x10[15] molec/cm2), and a proportional bias (slope=0.65).**
- This robust finding could have been obtained **thanks to the diversity of the FTIR network (clean/polluted) and its harmonization (no bias dependence due to the FTIR station).**
- The **S5P HCHO precision is within the pre-lunch requirements:** this can be proven **looking at the MAD at clean sites** (see plot): < $2x10^{15}$ molec/cm^2 for ~36 pixels (=$12x10^{15}$ molec/cm^2 for one pixel).
- We found an underestimation with a factor of ~ 2 of the S5P random uncertainty (Vigouroux et al., 2020).

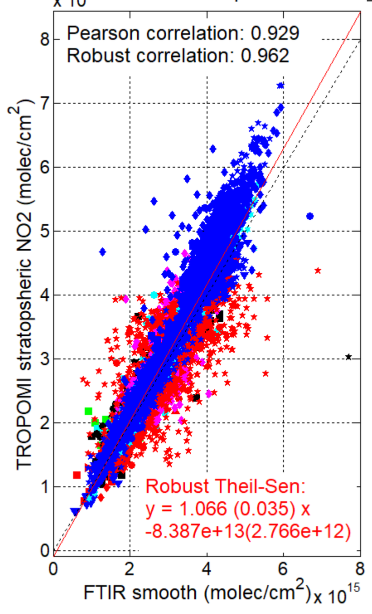


NO2 validation results

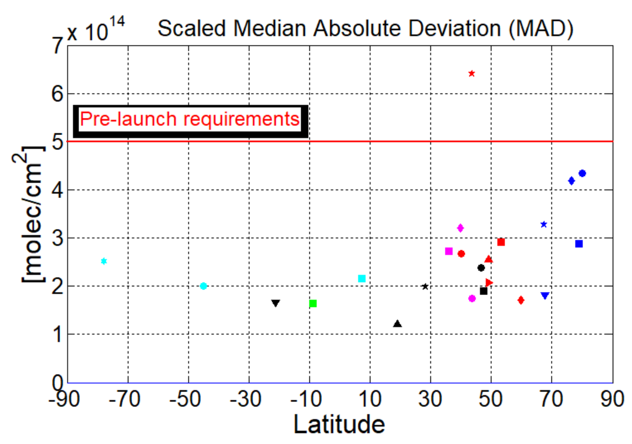


- **Station-to-station dispersion=5.4%**, very similar to DOAS network dispersion, showing **overall good consistency of the FTIR network.**
- The **biases are within the S5P mission requirements (accuracy <10%)**, except for the 2 highest latitude and tropical stations, where they are +10-13%.
- **Median of the individual bias: +4.5%. While the validation with ZSL DOAS data gives a negative bias (-6%)** (Verhoelst et al., 2021):
  - We performed **FTIR – DOAS comparisons** at stations with both techniques (see Table), using a photochemical box model to correct for the diurnal cycle effect. **Results are coherent with a median bias of 10% between the two ground-based measurements,** and **consistent with the FTIR systematic uncertainty.**

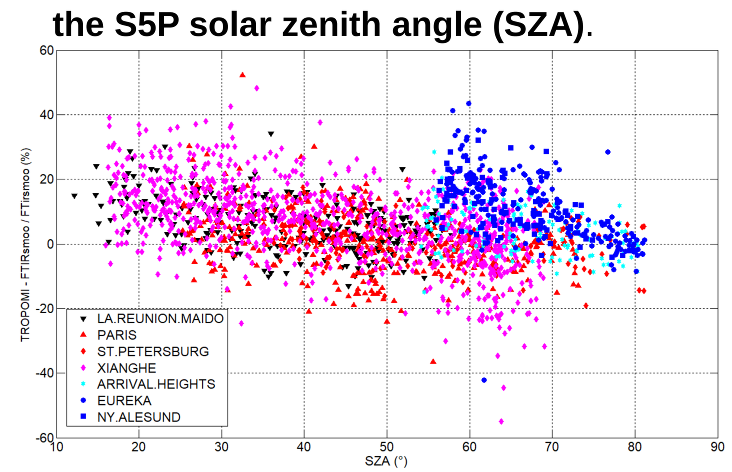
shortlabel	count	(DOAS-FTIR)/FTIR median NMB [%]	std-from-mad [%]
EUREKA	19	−0.17	4.74
SODANKYLA	69	11.29	17.88
BREMEN	26	15.96	14.06
IZANA	125	5.64	15.17
REUNION.MAIDO	327	13.11	12.99
median	69	11.29	14.06

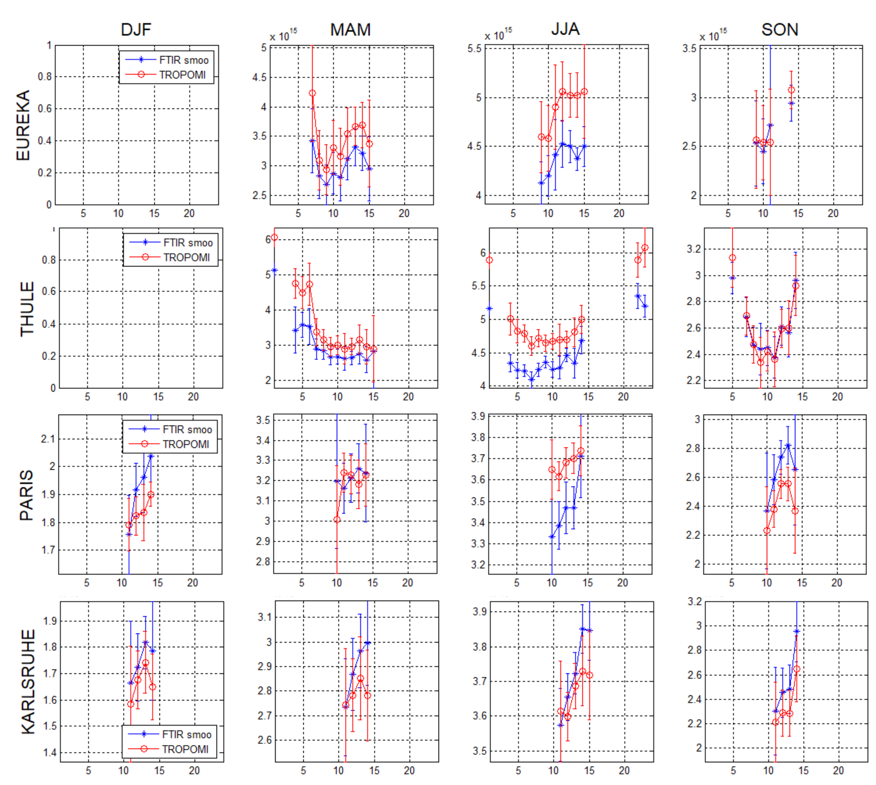


- **Correlation** is very good.
- Offset is very small= $-0.8x10^{14}$ molec/cm^2 (~1%)
- **MAD=2.9 x10[14] molec/cm[2]. This is within the S5P requirements of 5.0 x10[14] molec/cm[2].** (but requirements are given for single pixel). **And similar to S5P comparison with ZSL DOAS.**



- The **S5P and FTIR NO2 diurnal cycles look in good agreement at all sites.** They can differ from one site to another similar one (e.g. Thule vs Eureka…): to explore when more years of data will be available.
- At some sites, we observe a **dependence of the bias to the S5P solar zenith angle (SZA).**





Summary and outlook

- **Success for the FTIR network**: **HCHO and NO2 data sets built during the project !**
  - FTIR HCHO data set published (Vigouroux et al., 2018), used now in other studies, and **HCHO became an official NDACC species**.
  - All metrics using FTIR NO2 are **as good as when using ZSL DOAS network** (Verhoelst et al., 2021), with the additional advantage to provide comparisons of the diurnal cycle.
- **Success for TROPOMI HCHO and stratospheric NO2 products: they reach the pre-launch requirements** of maximum 80% and 10% bias, respectively, and the $2x10^{15}$ molec/cm^2 and 5.0 $x10^{14}$ molec/cm^2 precision, respectively.
- **On-going & future HCHO and NO2 S5P validation:** ESA reports, new algorithm versions, long-term changes,….. **Understand/improve** the extreme values in NO2 biases: due to TROPOMI or to FTIR ? Same question for some observed SZA dependence of the bias. **Publication of NO2 results.**