Response to reviewer #1

We really appreciate your constructive comments and suggestions on our manuscript. We have considered every comment carefully, and responded on a point to point and marked every change in red in the revised version.

General comments

This paper presents TROPOMI formaldehyde retrievals over China. The main difference with respect the operational HCHO retrievals distributed by ESA is the use of specific a-priori information during the air mass factor (AMF) calculation to much better the current knowledge of HCHO distributions over China.

Overall, the results presented here are interesting but the paper lacks details essential to understand the value and implications of the new retrieval. The specific comments section suggests may aspects where the manuscript could improve by including further details.

Definitely providing information about TROPOMI level 1 radiances and irradiances is absolutely necessary since they are the base for the retrievals described in section 3.

Responses: Thank you very much for this suggestion. Information about TROPOMI level 1 radiances and irradiances and the S5P cloud operational product are added in Section 2.1. TROPOMI has four spectrometers for medium wave ultraviolet (UV), long wave ultraviolet combined with visual (UVIS), near infrared (NIR), and short wave infrared (SWIR), covering non-overlapping and non-contiguous wavelengths from 270 to 2385 nm which are divided into eight spectral bands. The Band 3 with wavelength from 320 to 405 nm is used for HCHO retrieval. Radiance in Band 3 is measured by UVIS spectrometer. The detector for UVIS spectrometer is a two-dimensional charge-coupled device (CCDs) with one dimension for wavelengths and the other dimension for across track spatial coverage (450 rows). Earth radiance is collected along the dayside of the earth, while solar irradiance measurements are performed near the North Pole every 15 orbits, approximately once a day. In the UVIS channel, the spectral resolution and spectral sampling is about 0.5 nm and 0.2 nm, respectively. Individual ground pixels size of radiance measurement is approximately 3.5 km in the across-track and 7 km (5.5 km since August 2019), with integration time of 1.08 s (0.84 s since August 2019). The Level 1B radiance and solar irradiance are available at the Copernicus Open Access Hub (https://scihub.copernicus.eu/, last access: 22 May 2019).

Changes in manuscript: L66-76, P3-4 in the revised version.

Likewise, for the S5P cloud operational product whose uncertainties will become of paramount importance while assessing the uncertainties of the HCHO retrieval presented here.

Responses: Thank you very much for this suggestion. Information about the S5P cloud operational product are added in Section 2.3.

Changes in manuscript: L100-105, P5 in the revised version.

The description of the retrieval process also lacks many details. To start with, Figure 1 is included but never referenced in the text.

Responses: Thank you very much for this suggestion. Figure 1 is cited at L123, P6 in Section 2.5

Figure 2 shows two HCHO signals superimposed with the spectral fit residual. It will be more informative to include a similar figure showing the contributions from each one of the parameters included in Table 2.

Responses: Thank you very much for this suggestion. We plotted the spectral fitting residuals with considering different cross sections (shown in Figure. A1) on 6 August 2018 over China (orbit 4211). For HCHO polluted case, considering O_3 cross sections at 228K and 295K reduces residual significantly while considering NO_2, BrO and O_4 cross sections affects residual little (Figure. A1 (a)). Moreover, Ring effect and O_3 cross sections affect SCD largely and help reduce random error. For HCHO clean case, considering NO_2, BrO and O_4 cross sections reduces residual largely (Figure. A1 (b)). Moreover, the uncertainties of selection of fitting window, polynomial and TROPOMI slit function are discussed in Section 4.1.

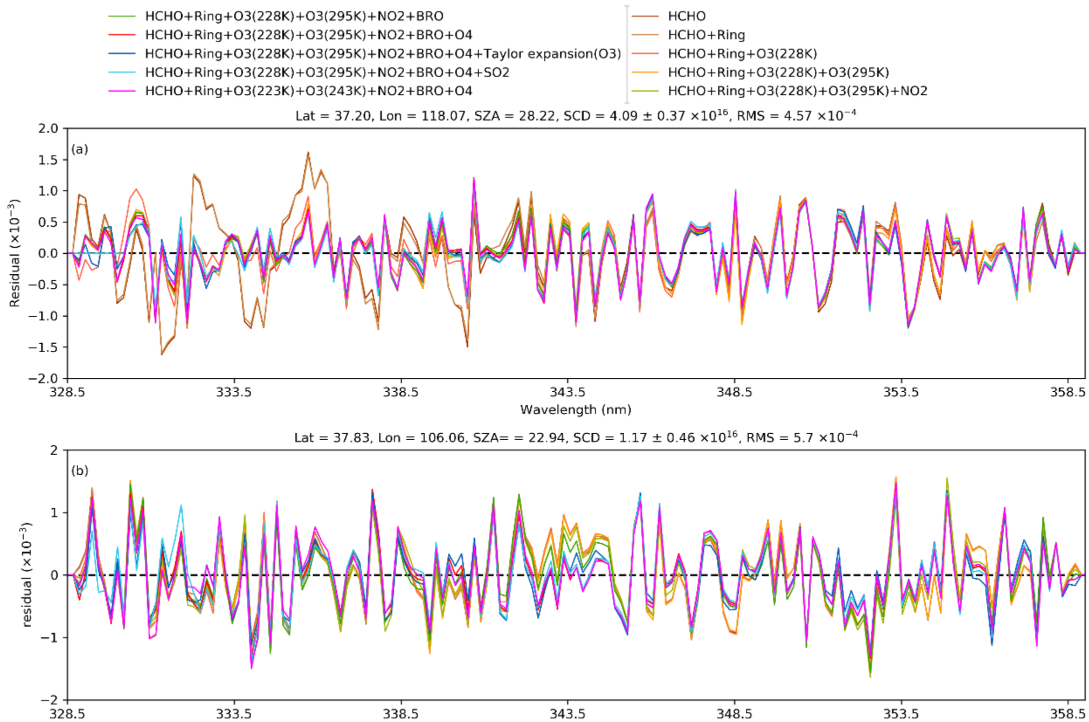

Figure. A1 Fitting residuals with considering different cross sections on 6 August 2018 over China for (a) polluted case and (b) clean case. The legend "HCHO" represents only considering HCHO cross section in spectral fitting and the legend "HCHO+Ring+O3(228K)+ O3(295K)+NO2+BRO+O4+Talor expansion(O3)" represents considering all cross sections listed in Table 1 and taking into account the first order Taylor series expansion for O_3 SCD at two temperatures in spectral fitting.

Changes in manuscript: L183-185, P8 in the revised version and L25-45, P3 and Figure S3 in supplement file.

Table A1. The spectral fitting results with considering different cross sections. The unit of DSCD and random error of DSCD is 10^{16} molec cm^{-2} and the RMS is expressed in 10^{-4}. The fitting results which cross sections are considered in our DSCD retrieval are marked in red.

	Polluted case			Clean case		
	DSCD	Random error of DSCD	RMS	DSCD	Random error of DSCD	RMS
HCHO	4.51	0.43	6.3	0.59	0.42	6.3
HCHO+Ring	4.32	0.44	6.3	0.57	0.44	6.3
HCHO+Ring+O_3(228K)	4.27	0.34	4.9	0.57	0.44	6.3
HCHO+Ring+O_3(228K)+O_3(295K)	3.99	0.35	4.7	0.68	0.46	6.3
HCHO+Ring+O_3(228K)+O_3(295K)+NO_2	3.96	0.34	4.6	0.64	0.45	6.1
HCHO+Ring+O_3(228K)+O_3(295K)+BrO	4.09	0.37	4.6	1.15	0.48	6.0
HCHO+Ring+O_3(228K)+O_3(295K)+BrO+O_4	4.09	0.37	4.6	1.17	0.46	5.7
HCHO+Ring+O_3(228K)+O_3(295K)+BrO+O_4 +Taylor expansion (O_3)	4.38	0.42	4.6	1.10	0.49	5.6
HCHO+Ring+O_3(228K)+O_3(295K)+BrO+O_4 +SO_2	4.40	0.40	4.5	1.73	0.51	5.6
HCHO+Ring+O_3(223K)+O_3(243K)+BrO+O_4	4.12	0.37	4.6	1.19	0.46	5.8

It is very difficult to estimate the quality of this new retrieval without seen the results for one full orbit at least (where issues such as stripes and noise are easier to appreciate). While current figure 3 does this in part, the color scale employed masks may of the possible issues (leaving out negative values) and providing little sensitivity to the region between 0.5 and 1.5 x 1016 molecules cm^{-2} most relevant for HCHO retrievals. It will also be very helpful to have plots showing the WRF-Chem simulations used to extract a priories as well as the GEOS-Chem simulation used for background corrections. Finally and of outmost importance, a discussion of the retrieval uncertainties is completely missing. While section 4 provides some bias estimates, the retrieval section should include a description of the random and systematic uncertainties linked to spectral fit and AMF calculation. Because there is no description of the operational HCHO TROPOMI retrieval in detail, it is impossible to assess the weight and validity of the conclusions derived from the comparison exercise between both products (the one presented here and the operational one). The authors focus on the impact due to the ΔSCD retrieval and the AMF calculation but do not devout any time evaluating the impact of using different earthshine radiances. It is also necessary to include a discussion of the MAX-DOAS errors.

Responses: These comments are also referred in Specific comments. We responded on a point to point in Specific comments.

Figure 5 shows some vertical profiles. What are they? Seasonal averages, daily averages,… Are they representative of similar periods of time and time of day? This results should be place into context with literature recently published (https://www.atmos-meas-tech-discuss.net/amt-2020-30/).

Responses: Thank you very much for this suggestion. The Figure 5 in the old version is Figure 7 in the revised version. Figure

7 shows seasonal averages of vertical HCHO profiles from MAX-DOAS, WRF-Chem and TM5-MP model at CAMS site. In AMF calculations, both WRF-Chem and TM5-MP simulations are interpolated to TROPOMI spatial resolution. Interpolated WRF-Chem and TM5-MP simulations within 20 km of the MAX-DOAS site are spatially averaged to compare with MAX-DOAS profiles. MAX-DOAS profiles are temporally averaged in the period 13:30-14:30 (Local Time) within ±1 h around the TROPOMI overpass time. And the error bars in Figure 7 represent 1σ standard deviation of seasonal variation.

Changes in manuscript: L366-370, P16 in the revised version.

Given the statistical uncertainties and the methodology used, an affirmation such as "These results suggest that our retrieval is better than the operational product both in urban and suburban in China" is probably over confident. First the MAX-DOAS measurements are representative of a small region in China dominated by large urban development. Second, small differences in the SCDs and the use of a higher spatial resolution model in the new retrieval indicates that both perform similarly. If the authors could provide some details about the appearance and differences of box-AMFs between both retrievals it will be possible to get a better idea of the impact of the a priori profiles.

Responses: Thank you very much for this suggestion. Due to limitation of MAX-DOAS data sets, we validate TROPOMI HCHO observations at three MAX-DOAS sites in Beijing including one urban site (CAMS site) and two suburban sites (NC and UCAS sites) (shown in Figure A3). The distances of three MAX-DOAS sites from central Beijing are 8, 27 and 61 km of CAMS, NC, and UCAS sites. The three MAX-DOAS sites are representative of urban and suburban region.

We using a different spectral retrieval technique (BOAS method) of HCHO slant columns. Moreover, the background correction has also been improved. Although DOAS and BOAS HCHO DSCDs show a similar spatial pattern. The spatial distribution of BOAS HCHO SCD is expected to be smooth, less noisy. Besides, operational product using different SCD retrieval methods is compared with MAX-DOAS HCHO measurements. Using the BOAS HCHO SCDs reduced the underestimation in summer and overestimation in winter of the operational product. In summer, using different SCD retrieval methods results a difference of 7.00% (± 1.71%, ± Error) from the TROPOMI operational HCHO VCD. The result shows that using the BOAS HCHO SCDs reduced the underestimation in summer and overestimation in winter of the operational product. Fig. A2 (a) and (c) show daily averaged a priori HCHO profiles from WRF-Chem and TRM5-MP simulations and MAX-DOAS measured profiles. Besides higher horizontal resolution, WRF-Chem simulation also has a higher vertical spatial resolution compared to TM5-MP data set. Although the difference in vertical resolution only shows negligible effect on box AMF under clear sky condition (Fig. A2 (b)), lower vertical resolution profiles would cause significant impact for cloudy cases due to the interpolation of coarse grid (Fig. A2 (d)).

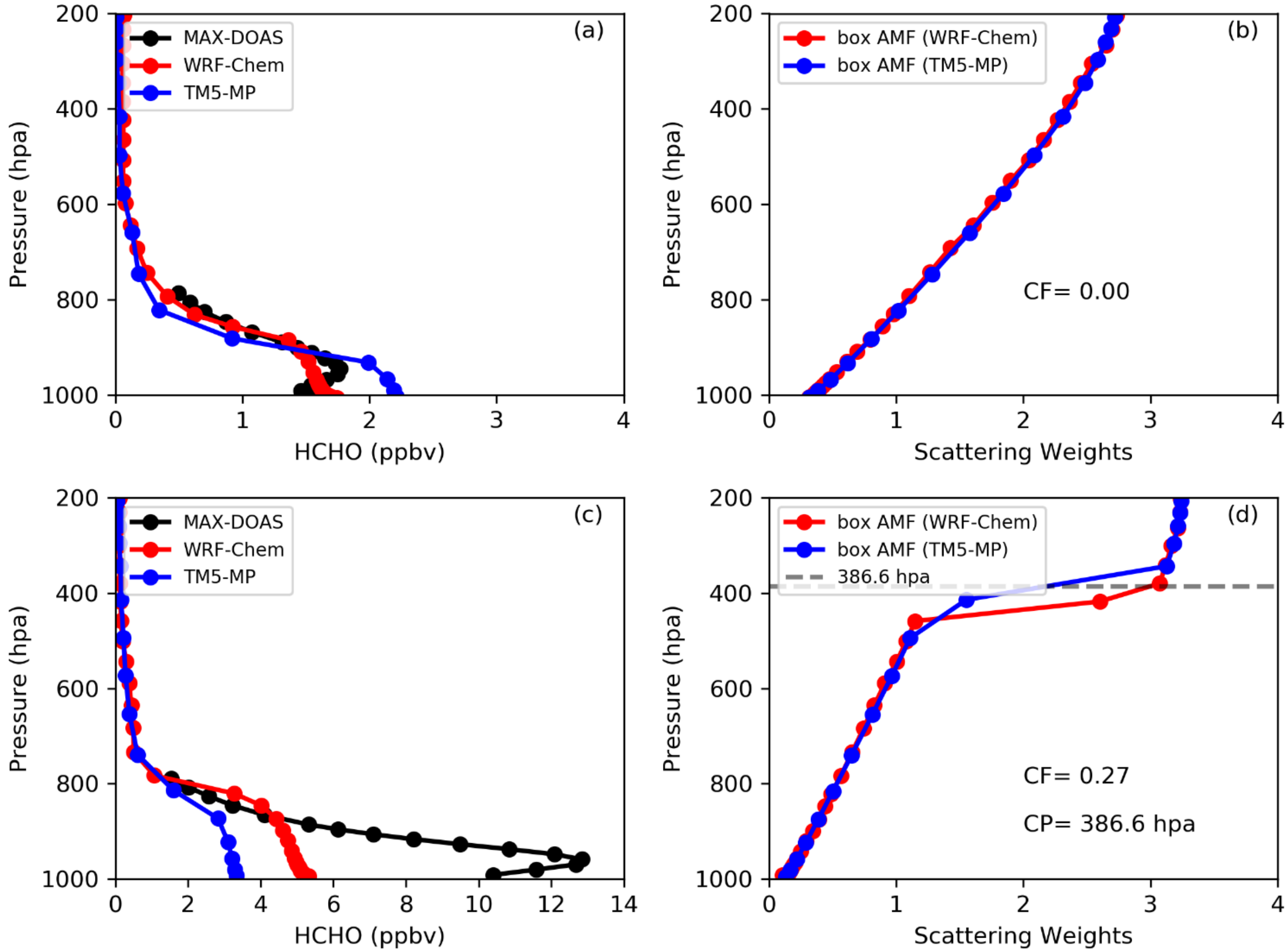

Figure A2. Daily averaged vertical HCHO profiles obtained from MAX-DOAS, WRF-Chem and TM5-MP model in clean case on 03 March 2019 (a) and in polluted case on 26 June 2019. Comparisons of box AMF using WRF-Chem and TM5-MP simulations in clean case with clear sky on 03 March 2019 (b) and in polluted case with cloudy sky on 26 June 2019 (c). The locations of two pixels are within 20 km of the CAMS site.

Changes in manuscript: L126-128, P6 and L370-374, P16-17 in the revised version.

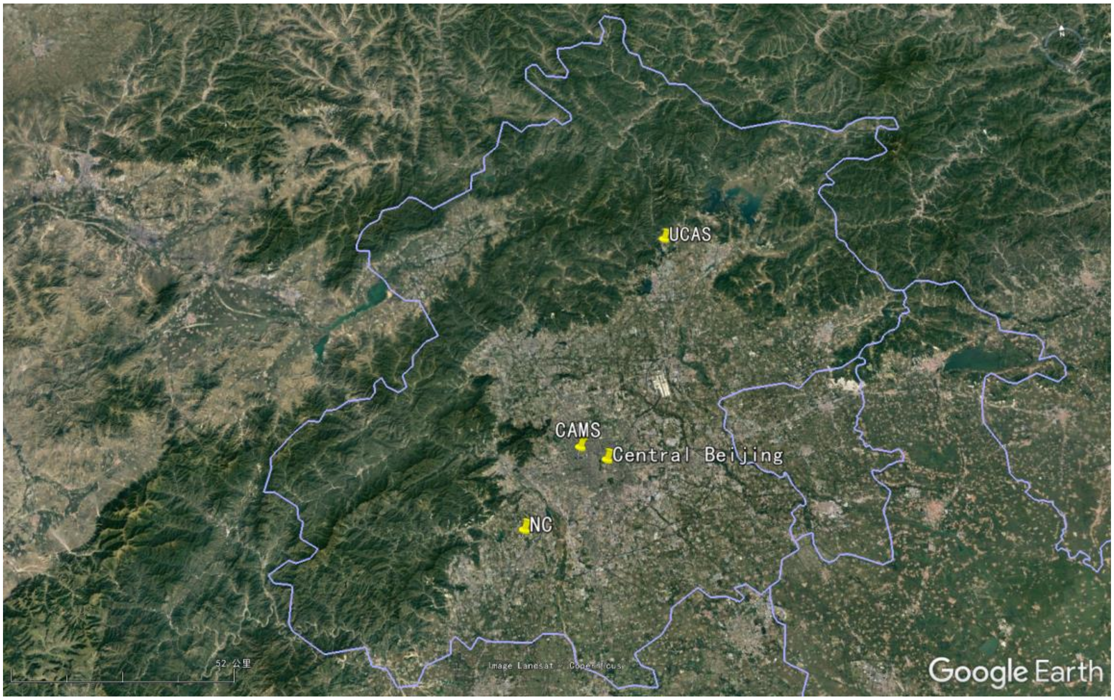

Figure A3. Locations of three MAX-DOAS sites in Beijing

The organization of the paper is clear (despite missing important aspects). However, while the use of English in the paper is good enough to be understandable it could benefit from further proofreading and grammar correction. Some language specific suggestions provided in the technical corrections section represent few of the possible improvements. While this reviewer will love to provide detailed language corrections it is time consuming and out of the scope of the reviewer duties.

For the reasons mentioned above the paper needs several major revisions and needs to expand to include essential details. The results shown are promising but in its current form, it will be difficult for anyone outside the authors to use the presented retrievals for a scientific study.

Specific comments

Abstract

A reference to the paper describing the operational TROPOMI HCHO retrieval by De Smedt et al., should be added here.

Recently Vigouroux et al., have published an excellent operational TROPOMI HCHO retrieval validation paper (https://www.atmos-meas-tech-discuss.net/amt-2020-30/). To cite in the introduction will provide a nice platform to place the results from this study into context later on.

Responses: Thank you very much for this suggestion. We have cited De Smedt et al. and Vigouroux et al. in the introduction.

Changes in manuscript: L50, P1 and L54-57, P3 in the revised version.

Data sets

The retrieval of slant column densities requires the use of calibrated TROPOMI radiances. This is maybe, the most important dataset that necessary to obtain the results presented in this work. Its description and how the authors obtained it should be a section in data sets.

Responses: Thank you very much for this suggestion. We have added the introduction of TROPOMI radiance in Sect. 2.1.

Changes in manuscript: L66-76, P3-4 in the revised version.

1. The original spatial resolution of TROPOPMI observations at NADIR was 3.5 km x 7 km but it improved to 3.5 km x 5.5 km in August 2019.

Responses: Thank you very much for this suggestion. We have modified it.

Changes in manuscript: L74-75, P4 in the revised version.

2. Fitting parameters considered in the spectral fitting of the operational product?

Responses: Thank you very much for this suggestion. The fitting parameters considered in the spectral fitting of the operational product are added in Table 1.

Changes in manuscript: L81-82, P4 and Table 1 in the revised version.

3. How is the reference sector correction applied in the operational product?

Responses: Thank you very much for this suggestion. The reference sector correction in the operational product is described in Sec. 2.2.

Changes in manuscript: L90-99, P4-5 in the revised version.

4. Procedures used to generate earthshine radiance over the remote Pacific in the operational HCHO retrieval.

Responses: Thank you very much for this suggestion. Daily detector row averaged radiance over the equatorial Pacific (latitude from 5°S to 5°N and longitude from 180°W to 140°W) is used as reference spectra. Due to residual HCHO signals in reference, the differential SCD (DSCD) is retrieved in spectra fitting.

Changes in manuscript: L85-87, P4 in the revised version.

5. Besides a-priori profiles and observation geometry, information about clouds, aerosols and surface reflectance play important roles in AMFs calculations. How is the operational product accounting for them?

Responses: Thank you very much for this suggestion. Information about data sets used in AMF calculation was added in Table 1.

Changes in manuscript: L222, P10 and Table 1 in the revised version.

6. Does the model simulation include information about pyrogenic sources?

Responses: Thank you very much for this suggestion. The open burning emission is obtained from the Fire INventory from NCAR (FINN) model (Wiedinmyer et al., 2010).

Changes in manuscript: L118-119, P5 in the revised version.

7. What was the spin off period of the simulations?

Responses: Thank you very much for this suggestion. Maybe the referee has a doubt about the spin-up period of the WRF-Chem simulations. WRF-Chem simulation is carried out from July 2019 to July 2019 with five days spun up prior to the simulation.

Changes in manuscript: L119-120, P5 in the revised version.

8. It will be interesting to have a reference for the MEIC inventory.

Responses: Thank you very much for this suggestion. The paper of (Li et al., 2017) was cited in the revised version.

Changes in manuscript: L116, P5 in the revised version.

9. What are the uncertainties associated with TROPOMI HCHO retrievals and MAX-DOAS observations. Table 1 could be expanded with information about the estimated uncertainties for each site as well as the dates and amount of available data.

Responses: Thank you very much for this suggestion. Error analysis of TROPOMI HCHO retrieval is added in Section 4. Available days and averaged relative error of MAX-DOAS measurements within ±1 h around the TROPOMI overpass time are added in Table 2.

Changes in manuscript: L241-292, P11-13 and Table 2 in the revised version.

HCHO SCD retrieval: wavelength calibration

One has to assume that S represents the preflight instrument slit function. It will be very beneficial to discuss the behavior of the preflight slit function. Are they available to the public? How stable the instrument slit function has been after launch?

Responses: Thank you very much for this suggestion. The Full-Width at Half-Maximum (FWHM) and asymmetric factor of the instrument slit function are obtained by fitting the daily irradiance to the high resolution solar spectra using the Gauss-Newton Nonlinear Least Squares (NLLS) method assuming the asymmetric Gaussian shape of slit function. The time series of the fitted TROPOMI slit function parameter from August 2018 to July 2019 in Fig. A4 shows that the TROPOMI slit function is stable after lunch. Therefore, the preflight TROPOMI slit function is used in wavelength calibration procedure. The preflight slit function is obtained from the TROPOMI Calibration Key Data (CKD) (available at http://www.tropomi.eu/data-products/isrf-dataset, last access: 22 May 2019) which is derived from TROPOMI calibration measurements performed in March 2015 at CSL in Liege. Comparing the spectral fit residual of using different versions of preflight slit function in the spectral fitting, we found that using version v3.0.0 results in lowest residual (Fig. S3). The preflight instrument slit function version v3.0.0 is used in our retrieval, while the operational product uses version v1.0.0 preflight slit function.

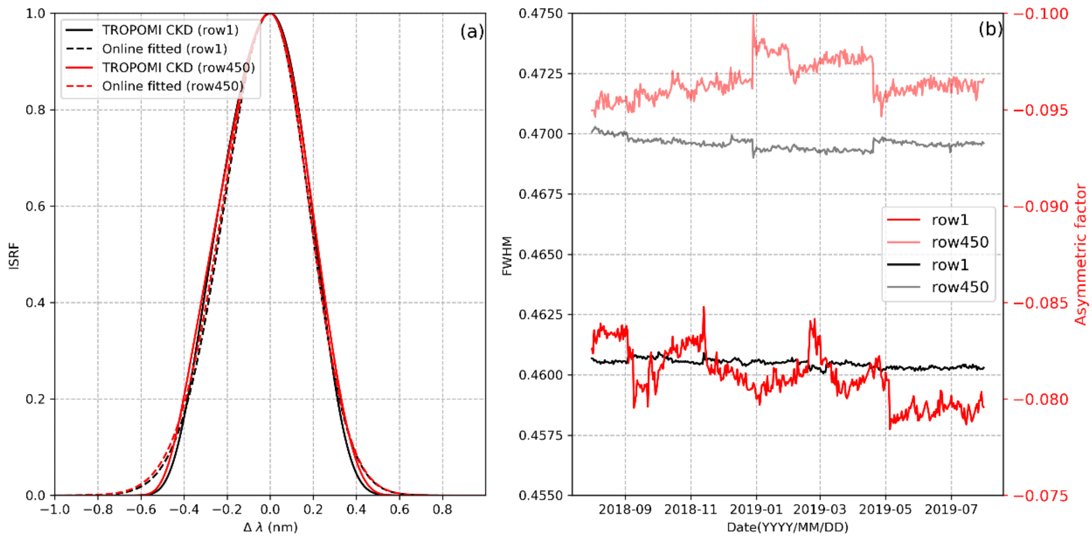

Figure A4. (a) The slit function from TROPOMI CKD at 340nm and online fitted for row 1 and row 450 on 01 August 2018. (b) Time series of FWHM and asymmetric factor of online fitted slit function from August 2018 to July 2019.

Changes in manuscript: L159-169, P7-8 in the revised version.

HCHO SCD retrieval: Radiance fitting

As mentioned above, there is lack of detail in the description of the methodology employed. Please explain the following questions:

(1) Methodology employed to calculate the daily average earthshine radiance over the Pacific.

Responses: Thank you very much for this suggestion. Radiances measured 1 day before the processing day over the Pacific with latitudes ranging from 30°S to 30°N and longitude ranging from 180°W to 140°W are averaged and used as reference in the spectral fit.

Changes in manuscript: L179-180, P8 in the revised version.

(2) Any I0 corrected cross sections in the spectral fit.

Responses: Thank you very much for this suggestion. O_3 cross sections at two temperatures are I_0 corrected in the spectral fitting.

Changes in manuscript: Table 1 in the revised version.

(3) What is the impact of not including SO_2 when if SO_2 optical thickness becomes relevant.

Responses: Thank you very much for this suggestion. We plotted the spectral fitting residual with (red lines) and without (wathet lines) considering SO_2 cross section for HCHO polluted (a) and clean (b) cases in SO_2 polluted region (Fig. A1). Considering SO_2 cross section has no contribution on reducing residual and increases residual at some wavelengths on the contrary.

(4) What is the impact of not including water vapor?

Responses: Thank you very much for this suggestion. Laboratory measurements of water vapor absorption lines only extend to 25,470 cm^{-1} (393 nm) (Dupre et al., 2005). (Lampel et al., 2016) indicated that water vapor can potentially have an impact on the spectral retrievals of tropospheric HCHO while the absorption at 335 nm could not be unambiguously identified in measurements so far.

(5) Which method is employed to estimate Raman spectra?

Responses: Thank you very much for this suggestion. The high resolution solar spectrum (Chance and Kurucz, 2010) is convolved with the rotational Raman spectra of O_2 and N_2 to produce the Raman spectrum (Chance and Spurr, 1997). The paper (Chance and Spurr, 1997) is cited in the revised version.

Changes in manuscript: L181, P8 and Table 1 in the revised version.

(5) How was the fitting window selected? HCHO retrievals show big dependencies with fitting windows.

Responses: Thank you very much for this suggestion. We use the same fitting window with operational product in DSCD retrieval. The uncertainty of fitting window selection is discussed in Section 4.1.

(6) New O3 cross sections have become available in recent years (for example Serdyuchenko et al., 2014 (https://www.atmos-meas-tech.net/7/609/2014/)). (7) Have the authors taken into account the effect of ozone in the fitted slant columns as described by Pukite et al., 2008 (https://www.atmos-meas-tech.net/3/631/2010/amt-3-631-2010.pdf).

Responses: Thank you very much for this suggestion. In our retrieval, we didn't take into account the first order Taylor series expansion for O_3 SCD. The uncertainty without considering Taylor expression for O_3 SCD is discussed in Section 4.1. We did the sensitivity tests about effect from difference of O_3 cross sections and Taylor series approach for HCHO polluted and clean cases. The spectral fitting residuals are plotted in Figure. A1 and fitting SCDs, random errors and RMS are presented in Table A1. For HCHO polluted case, using newly published O_3 cross sections changes SCD by 0.7% with random error and RMS unchanged. For HCHO clean case, using newly published O_3 cross sections changes SCD by 1.7% with random error unchanged and RMS increasing 0.1. The effect of using Taylor series of O_3 cross section in the spectral retrieval is evaluated through sensitivity analysis (Table 3). After reference sector correction, systematic difference regarding to Taylor series of O_3 cross section is estimated to be 3.49%. The uncertainties from O_3 cross section and without using Taylor series approach are discussed in Section 4.1.

AMF calculation

Details missing in the AMF calculation include: (1) Terrain height and surface pressure corrections.

Responses: Thank you very much for this suggestion. The surface pressure and terrain height we used are obtained from the operational HCHO product which are corrected.

(2) Origin of cloud information.

Responses: Thank you very much for this suggestion. Cloud parameters used in AMF calculation in our retrieval are from the operational TROPOMI cloud product. Cloud fraction is retrieved using the Optical Cloud Recognition Algorithm (OCRA) and cloud top height (pressure) and optical thickness (albedo) are retrieved using Neural Networks (ROCINN) algorithm using the "Clouds-as-Reflecting-Boundaries" (CRB) model, treating clouds as simple Lambertian surfaces (Loyola et al., 2018). The operational TROPOMI cloud product is also available at the Copernicus Open Access Hub (https://scihub.copernicus.eu/, last access: 22 May 2019).

Changes in manuscript: L100-105, P5 in the revised version.

(3) Descriptions of the nodes of the box AMF look up table.

Responses: Thank you very much for this suggestion. The grid points of parameters in creating the LUT are same with De Smedt et al (2018).

Changes in manuscript: L208, P9 in the revised version.

(4) VLIDORT set up.

Responses: Thank you very much for this suggestion. The paper (Spurr, 2008) is cited introducing VLIDORT 2.6

Changes in manuscript: L207, P9 in the revised version.

(4) Impact of aerosols.

Responses: Thank you very much for this suggestion. Aerosol extinction profiles are also retrieved by the MAX-DOAS. Aerosol optical properties, such as single-scattering albedo (SSA) and Ångström exponent obtained from the Aerosol Robotic Network (AERONET) station in Beijing (https://aeronet.gsfc.nasa.gov/, last access: 22 May 2019) are used as input parameters for the MAX-DOAS aerosol profile retrieval. As the aerosol profiles from MAX-DOAS are retrieved at 360nm, we further converted the profile to 340nm using Ångström exponent obtained from the AERONET measurements. To estimate aerosol effect on TROPOMI HCHO retrieval, we calculate the AMFs using MAX-DOAS measured aerosol extinction profiles using VLIDORT (version 2.6) (Spurr, 2008). The AMFs are applied on the operational product and our retrieval. The TROPOMI HCHO VCDs with and without considering aerosols are compared to MAX-DOAS HCHO VCDs. The comparison results are shown in Figure 10. The results show that considering aerosol in the AMF calculations does not improve the agreement with ground based measurements. Considering aerosol effect in TROPOMI retrieval reduces HCHO VCDs by 11.46% (± 1.48%) for the operational product and 17.61% (± 1.92%) for our retrieval in winter. The reduction over urban site is more significant than suburban sites, mainly due to higher aerosol load. Operational product using both HCHO and aerosol extinction profiles from MAX-DOAS shows underestimation of 8.36 % (± 4.63 %). Our retrieval using MAX-DOAS HCHO and aerosol extinction profiles for AMF calculation underestimates HCHO VCD by 18.53% (± 4.04 %).

Changes in manuscript: L452-466, P20 in the revised version.

(5) Error analysis.

Responses: Thank you very much for this suggestion. Error analysis in TROPOMI HCHO retrieval is discussed in Section 4.2.

Changes in manuscript: L241-292, P11-13 in the revised version.

Reference sector correction of SCDs

Add description of the GEOS-Chem configuration employed in the reference sector correction. Which longitudes define remote Pacific? Is the correction applied only to -30 degrees to 30 degrees? That region is used to calculate the earthshine radiance. Any contributions outside -30 to 30 degrees will not be correcting for the residual HCHO column but for biases of different origin.

Responses: Thank you very much for this suggestion. Radiances measured 1 day before the processing day over the Pacific with latitudes ranging from 30°S to 30°N and longitude ranging from 180°W to 140°W are averaged and used as reference in the spectral fit. The first step of reference sector correction is retrieving the DSCDs using average earthshine radiance reference, calculating the corresponding AMFs and storing them as a separate database. The HCHO VCD over remote Pacific Ocean is simulated by GEOS-Chem assuming HCHO over this region is mainly from the oxidation of CH_4. The simulated HCHO SCD is calculated by multiplying the VCD (VCD_G) taken from GEOS-Chem with the corresponding AMF (M_0). Assuming HCHO in the reference sector correction is well simulated by GEOS-Chem, the difference between the simulated and retrieved DSCD (DSCD_0) is recognized as the SCD bias caused by residual HCHO signal in reference spectrum. TROPOMI measurements over the Pacific (latitude from 90°S to 90°N and longitude from 160°W to 140°W) are first binned according to their latitude to 500 bins with a resolution of 0.36°. The median value of each bin is then used for the calculation of the SCD background correction (González Abad et al., 2015; González Abad et al., 2016). Assuming the SCD correction is constant in the longitude direction, the SCD correction at 500 gridded latitude points from 90°S to 90°N are linearly interpolated to the latitude of each pixel over China. The interpolated SCD correction is then applied on the retrieved DSCDs to calculate SCDs. Therefore, DSCDs outside -30 to 30 degrees are also corrected. We have clarified in the revised version.

Changes in manuscript: L179-180, P8 and L231-240, P11 in the revised version.

Comparison of operational and improved HCHO product

What are the coincidence criteria to match TROPOMI and MAX-DOAS measurements? To use daily averages in the case of MAX-DOAS seems inappropriate considering the diurnal variations of HCHO columns.

Responses: MAX-DOAS measurements are temporally averaged within ± 1 h around the TROPOMI overpass time, while TROPOMI pixels within 20 km of the MAX-DOAS site are spatially averaged for comparison. TROPOMI pixels in our retrieval and operational product are both filtered for intensity-weighted cloud fraction smaller than 0.3, root mean square of spectral fit residual (RMS) smaller than 10^{-3}, AMF larger than 0.1 and SZA smaller than 70°, quality assurance value (QA value) larger than 0.55 and successful SCD retrieval. The histograms showing the distributions of RMS of spectral fit of the operational product and our retrieval on 06 August 2018 over China are shown in Figure. S5. About 15% and 17% of measurements in the operational product and our retrieval show RMS larger than 10^{-3}.

Changes in manuscript: L307-313, P14 in the revised version.

Comparisons of SCD retrievals

This discussion applies to SCDs or ΔSCDs? The result of the spectral fit is in both cases the differential slant column but the title of the section indicates the comparison of slant columns. In that case, differences are not only due to the spectral fit but also to the reference sector correction. What is the impact of using different earthshine radiances?

Responses: Thank you very much for this suggestion. In section 5.1.1, we compare DSCDs and SCDs after applying reference sector correction between operational product and the new retrieval. The biases between DSCDs and RMS in operational product and our retrieval are mainly related to the difference of retrieval method, retrieval settings and selection of reference. To investigate the impact of selection of earthshine radiance reference, we retrieved HCHO DSCDs using daily detector row averaged radiance over the equatorial Pacific (latitude from 5°S to 5°N and longitude from 180°W to 120°W) as reference using BOAS method. Comparisons of HCHO DSCDs and HCHO SCDs after applying reference sector correction using different earthshine radiance reference are presented in Figure. A5. The retrieved DSCDs using different earthshine radiance reference correlated well (R=0.99, Slope=0.99) and with a difference of 30% ($0.11 \pm 0.08 \times 10^{16}$ molec cm^{-2}). The bias is compensated (1.9%) by the reference sector correction (Figure. A5 (b)).

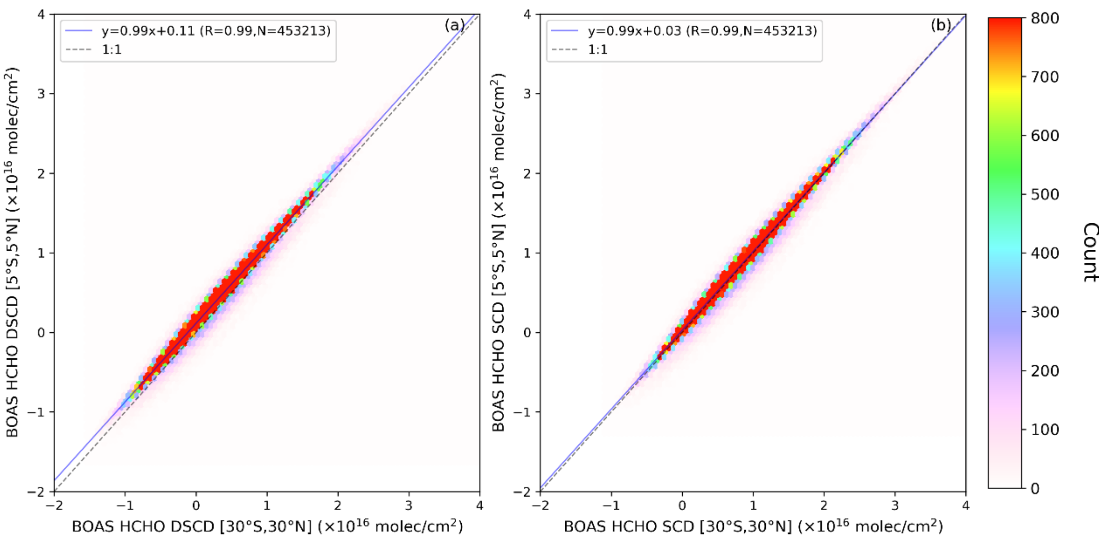

Figure A5. Pixel to pixel comparisons of BOAS HCHO DSCDs (a) and BOAS HCHO SCDs (b) using different earthshine radiance reference on 06 August 2018 in the region between 73° E and 130° E, and 18° N and 54° N. The labels with [30°S, 30°N] represent that average of radiances of the equatorial Pacific (latitude from 30°S to 30°N and longitude from 180°W to 140°W) is used as reference spectra. The labels with [5°S, 5°N] represent that average of radiances of the equatorial Pacific (latitude from 5°S to 5°N and longitude from 180°W to 120°W) is used as reference spectra.

Changes in manuscript: L339-345, P15 in the revised version and Figure. S6 in the supplement file.

What are the reasons to filter out retrievals with RMS higher than 10^{-3}. What is the percentage of retrievals with RMS above the threshold? What is the definition of outliers? Showing a histogram of the distributions of both datasets will help to understand the outlier definition.

Responses: Thank you very much for this suggestion. The histograms of the distributions of RMS in operational product retrieval and our retrieval are shown in Figure. A6. About 15% and 17% of measurements in the operational product and our retrieval show RMS larger than 10^{-3}.

Changes in manuscript: L312-313, P14 in the revised version and Figure. S5 in the supplement file.

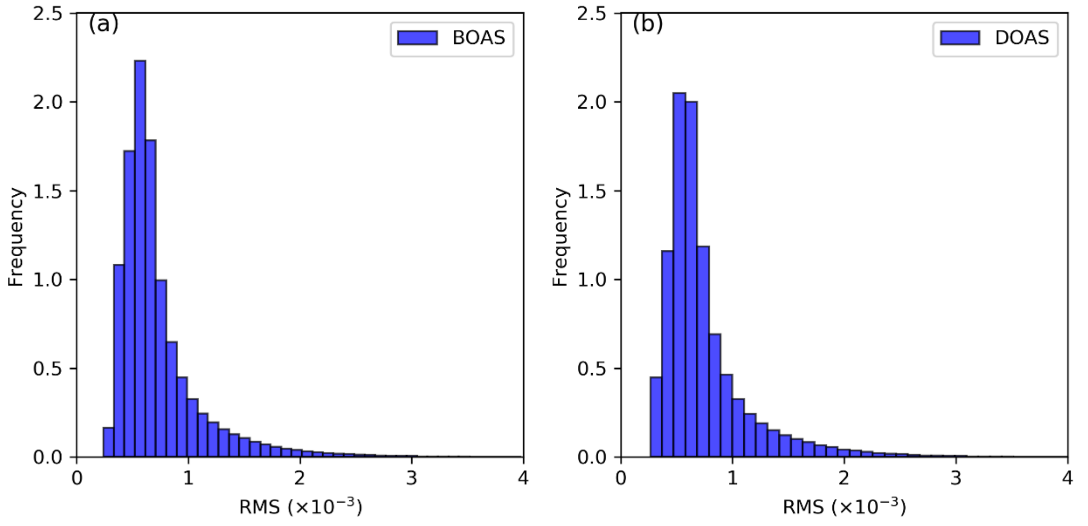

Figure A6. The histograms of the distributions of RMS in operational product retrieval (a) and our retrieval (b).

The authors make a distinction between non-corrected and corrected SCDs. It is not clear in the text what corrected implies. One has to assume we are talking about reference sector corrected and non-corrected SCDs. If that is the case, one could argue that part of the differences observed between SCDs in the non-corrected case are due to the different selection (or calculation) of earthshine radiance reference. To make a real comparison of the performance of booth fitting algorithms they should be using consistent earthshine radiances. The results seem to indicate that part of the biases are due to using different earthshine radiances since the correlation for the non-corrected and corrected SCDs columns is similar for both products but the bias between them is significantly reduced after applying the correction.

Responses: Thank you very much for this suggestion. Due to residual HCHO signals in reference, the differential SCD (DSCD) is retrieved in spectra fitting. Therefore, reference sector correction has to be applied on the retrieved DSCDs to calculate SCDs. The reference sector correction is described on Sec. 3.3. We have cleared it in the revised version. To investigate the impact of selection of earthshine radiance reference, we retrieved HCHO DSCDs using daily detector row averaged radiance over the equatorial Pacific (latitude from 5°S to 5°N and longitude from 180°W to 120°W) as reference using BOAS method. Comparisons of HCHO DSCDs and HCHO SCDs after applying reference sector correction using different earthshine radiance reference are presented in Figure. A5. The retrieved DSCDs using different earthshine radiance reference correlated well (R=0.99, Slope=0.99) and with a difference of 30% ($0.11 \pm 0.08 \times 10^{16}$ molec cm^{-2}). The bias is compensated (1.9%) by the reference sector correction (Figure. A5 (b)). To investigate the impact from retrieval method, BOAS HCHO DSCDs using same retrieval settings with operational DSCD retrieval are compared with DOAS HCHO DSCDs (Figure. A7). Using same retrieval settings, difference between DOAS HCHO DSCDs and BOAS HCHO DSCDs (27.33%) is significantly reduced and the remaining difference is due to retrieval algorithm. Besides, smaller difference (4.41%) in HCHO SCDs indicates that reference sector correction reduces the effect from retrieval method (Figure. A7 (b)).

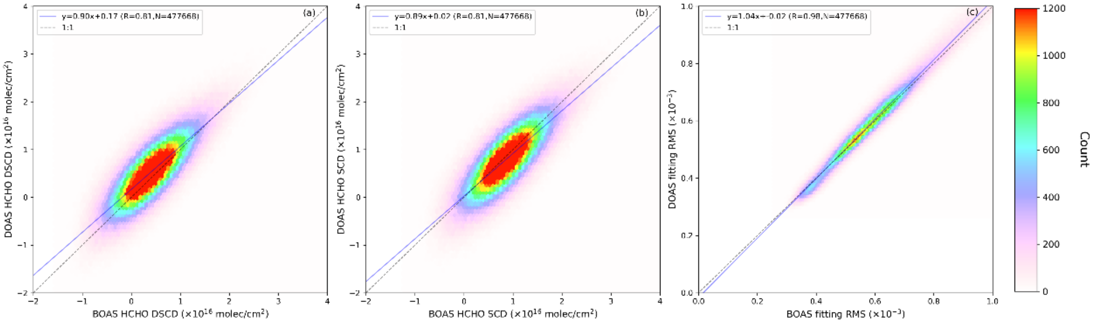

Figure A7. (a) Pixel to pixel comparisons of DOAS and BOAS HCHO DSCDs, (b) DOAS and BOAS HCHO SCDs and (c) DOAS and BOAS fitting RMS on 06 August 2018 in the region between 73° E and 130° E, and 18° N and 54° N. Same retrieval settings are used in DOAS and BOAS retrieval.

Changes in manuscript: L340-349, P15-16 in the revised version and Figure. S7 in the supplement file.

Comparisons of retrievals after AMF calculations

One question that raises this section and always permeates the use of high-resolution models is how much information is folded back from the model in the retrieval. To better understand this question it will be very useful to know more about the WRF-Chem simulations and how they were used. The operational product employs daily forecast interpolated in time and space. This procedure should be added to the methodology. How do the box-AMFs of the different retrievals compare?

Responses: Thank you very much for this suggestion. Information about more WRF-Chem is added in the Sect. 2.4. A priori HCHO profile for TROPOMI AMF calculations are calculated by interpolating WRF-Chem simulation spatio-temporally to the measurement time and location. This information is added in Sect. 3.2. The information that the operational product employs daily forecast interpolated in time and space is also added in Sect. 2.2.

Besides higher horizontal resolution, WRF-Chem simulation also has a higher vertical spatial resolution compared to TM5-MP data set. Although the difference in vertical resolution only shows negligible effect on box AMF under clear sky condition (Fig. 6 (b)), lower vertical resolution profiles would cause significant impact for cloudy cases due to the interpolation of coarse grid (Fig. 6 (d)).

Changes in manuscript: L89, P4, L200-201, P9 and L371-374, P17 in the revised version.

Comparison between HCHO VCDs observed by MAX-DOAS and TROPOMI

Blue lines in figure 7 right panels, more than speaking of the retrieval, speak about the difference between the a priori profiles for both models and the retrieved MAX-DOAS profiles. Slopes and correlation coefficients are similar (within error estimates) for both retrievals when using model a priori. It is easier to imagine how the comparison of the operational retrieval with MAX-DOAS observations will also suffer a dramatic improvement if MAX-DOAS a priori were to be used in the AMF calculations.

Responses: Thank you very much for this suggestion. We have recalculated HCHO VCD for the operational product by using MAX-DOAS HCHO profiles as a priori profiles. The Pearson correlation coefficient (R) between the recalculated operational product and MAX-DOAS HCHO VCD decreases by 0.02 to 0.79. The slope of the regression line increases by 0.19 to 0.84 with offset reduces by 0.24×10^{16} molec cm^{-2} to 0.15×10^{16} molec cm^{-2}. Although the recalculated TROPOMI operational product shows improved correlation with MAX-DOAS observations, our retrieval using MAX-DOAS measurements as a priori profile still shows a better agreement with MAX-DOAS HCHO VCDs. It also indicates that the BOAS spectral retrieval of SCDs agree better to the ground based observations over China.

Changes in manuscript: L406-412, P18 in the revised version.

Technical corrections

Line 18 (grammatical suggestion (GS)): Since what is improved is the HCHO retrieval a more correct grammatical structure will be "We present an improved retrieval of formaldehyde (HCHO) over China from the TROPO…"

Responses: Thank you very much for this suggestion. Changed.

Line 23 (GS): remove "the" from "agreement with the ground based…" since it is possible there are more than MAXDOAS measurement than those used in this study.

Responses: Thank you very much for this suggestion. Changed.

Line 25 (GS): add "s" to "profile" "…higher resolution a priori profiles".

Responses: Thank you very much for this suggestion. Changed.

Line 26: The percentage of what is reported by 61.11% and 0.15%? With the current text it is impossible to know if it refers to the change in the mean VCD, or the percentage of the bias correction attributed to each step. Please specify.

Responses: Thank you very much for this suggestion. We added the error bar in Table 3 and find the conclusion that changing SCD retrieval method only shows a tiny effect of 0.15% is less rigorous. The sentence is changed into "The improvements are mainly related to the AMF calculation with more precise a priori profiles in winter. Using more precise a priori profiles in general reduces HCHO VCDs by 52.37 % ($\pm$ 27.09 %) in winter.

Changes in manuscript: L24-25, P1 in the revised version.

Line 29 (GS): Change "indicating" to "indicate".

Responses: Thank you very much for this suggestion. Changed.

Line 45 (GS): Add "to" after compared "Compared to its predecessor"

Responses: Thank you very much for this suggestion. Changed.

Line 53 (GS): Add "s" to "profile" and "a" after "from". Which regional model is employed?

Responses: Thank you very much for this suggestion. The AMF calculation is improved by using higher resolution a priori profiles from the regional Weather Research and Forecasting model (WRF-Chem). Changed. A priori HCHO profile for TROPOMI AMF calculations are calculated by interpolating WRF-Chem simulation spatio-temporally to the measurement time and location.

Line 64 (GS): Change “is” to “are” “.., which are devided in …”.

Responses: Thank you very much for this suggestion. Changed.

Line 91 (GS): Add “the” in “… is located in the Chinese…”

Responses: Thank you very much for this suggestion. Changed.

Line 105: The first step of the methodology explained in this paper is the calculation of ΔSCD, not SCD.

Responses: Thank you very much for this suggestion. Changed.

Line 111: There can be other causes for wavelength miss-alignment can be Doppler shift, non-uniform illumination of the slit due to presence of clouds or other high reflectance surface in the pixel.

Responses: Thank you very much for this suggestion. Added.

Changes in manuscript: L147-148, P7 in the revised version.

Line 115: What is the benefit of having two closure polynomials during wavelength calibration considering that it is done using TOA irradiances and there for are not affected by the presence of clouds or aerosols that may introduce low-frequency structures?

Responses: Thank you very much for this suggestion. We compared fitting residuals with and without considering the third baseline and scaling polynomials during wavelength calibration. Considering the third scaling and baseline polynomials decreases the residual largely.

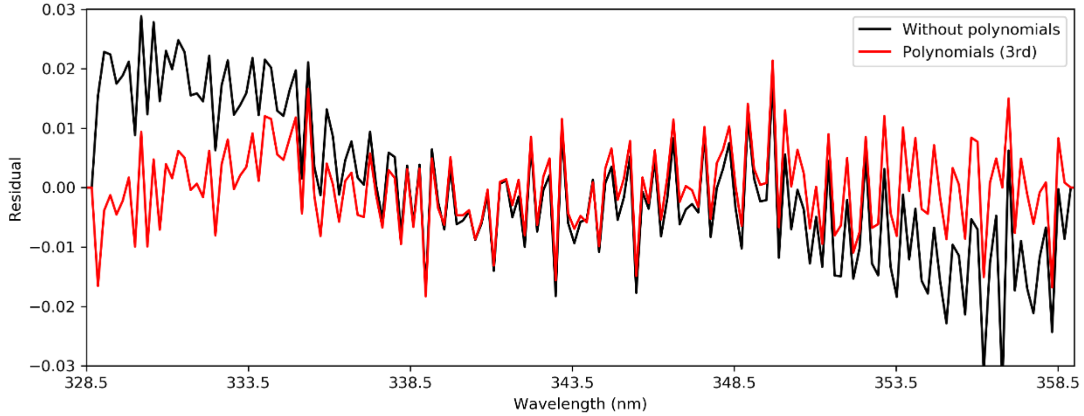

Figure A8. Comparison of fitting residuals with and without considering the third baseline and scaling polynomials during wavelength calibration.

Changes in manuscript: L68-171, P8 in the revised version.

Line 142 (GS): Add “presence of” to “… atmosphere (presence of clouds, vertical HCHO distribution…”

Responses: Thank you very much for this suggestion. Changed.

Line 143: What is a comprehensive radiative transfer model?

Responses: Thank you very much for this suggestion. The sentence seems superfluous because the following content has a detailed introduction about AMF calculation. The sentence is deleted.

Line 150 (GS): Add “on” as “The box AMF depends on wavelength, …”

Responses: Thank you very much for this suggestion. Changed.

Line 173 (GS): Add “be” to “… is known to be caused by …”

Responses: Thank you very much for this suggestion. The sentence is changed into “Using earthshine radiance over remote Pacific Ocean as reference significantly reduces the influence from unresolved spectral structures which could significantly improve the spectral retrieval of weak absorber, i.e., HCHO.”

Line 173: Could the authors provide a reference for the known cause of stripping?

Responses: Thank you very much for this suggestion. The sentence is changed into “Using earthshine radiance over remote Pacific Ocean as reference significantly reduces the influence from unresolved spectral structures which could significantly improve the spectral retrieval of weak absorber, i.e., HCHO.”

Line 175: AMFs are not retrieved; they are calculated and are independent of a remote Pacific earthshine radiance.

Responses: Thank you very much for this suggestion. Changed. The first step of reference sector correction is retrieving the DSCDs using average earthshine radiance reference, calculating the corresponding AMFs and storing them as a separate database.

Line 204 & 209 (GS): Change “outliners” to “outliers”.

Responses: Thank you very much for this suggestion. Changed.

Line 216 & 217: What is the error associated to the average SCDs? Standard deviations?

Responses: Thank you very much for this suggestion. Standard deviations are shown in the brackets associated to the average DSCDs. Changed.

Line 222: How is the 32.32% lower calculated? A difference of 0.02 x 10^{16} molecules cm^{-2} looks rather small.

Responses: Thank you very much for this suggestion. The sentence is changed to “Averaged SCD taken from the operational product on 06 August 2018 over China ($0.85 \pm 0.69\times10^{16}$ molec cm^{-2}) is on average 4.49 % lower than our retrieval ($0.89 \pm 0.61\times10^{16}$ molec cm^{-2}).”

Line 223-226 (GS): Please rewrite sentence.

Responses: Thank you very much for this suggestion. The sentences are changed into "We use SCDs in our retrieval and AMFs in operational product to calculated the updated HCHO VCD. The NMB of the two data sets are shown in Table 5. The bias between the updated HCHO VCD and operational HCHO VCD is caused by difference in SCD retrieval. The updated and operational product is also compared to the MAX-DOAS observations."

Line 228: How does BOAS compute mean random errors? Does this calculation use the operation product random error definition? Without describing both is impossible to interpret BOAS random errors lower than DOAS by 22%.

Responses: Thank you very much for this suggestion. The random uncertainty in DSCDs retrieval is described in Sect. 4.1.

Figure 1: What is figure 1 trying to illustrate. It is not mention in the text.

Responses: Thank you very much for this suggestion. Figure 1 shows the location and HCHO concentration of three MAX-DOAS sites. It is cited in L123, P6 in Section 2.5.

Table 1: What is the definition of viewing azimuth angle? Probably clock wise with respect to North.

Responses: Thank you very much for this suggestion. The original Table 1 is Table 2 in the revised version. Viewing azimuth angle of the north is taken as zero degree. It is added in the caption of Table 2.

Table 2: What is the methodology employed to calculate the Raman spectra? What is the definition of Pacific (remote)? Will it be possible to provide a reference for the pre-flight instrument slit function?

Responses: Thank you very much for this suggestion. The original Table 2 is Table 1 in the revised version. The high resolution solar spectrum (Chance and Kurucz, 2010) is convolved with the rotational Raman spectra of O_2 and N_2 to produce the Raman spectrum (Chance and Spurr, 1997). The paper (Chance and Spurr, 1997) is cited in the revised version.

Radiances measured 1 day before the processing day over the Pacific with latitudes ranging from 30°S to 30°N and longitude ranging from 180°W to 140°W are averaged and used as reference in the spectral fit.

The preflight TROPOMI slit function is obtained from the TROPOMI Calibration Key Data (CKD) (available at http://www.tropomi.eu/data-products/isrf-dataset) which is derived from TROPOMI calibration measurements performed in March 2015 at CSL in Liege.

The information is added in the revised version.

Changes in manuscript: Table 2, L163-165, P7 and L179-180, P8, in the revised version.

Figure 2: What the figure shows is the ΔSCD not SCD. Please correct. The size of the residuals in plot (b) seem to be large considering an RMS of 2.78×10^{-4}.

Responses: Thank you very much for this suggestion. Figure 2 shows the spectral retrieval of HCHO DSCDs and Figure 2 is updated. The values are updated.

Figure 3: A different color map, extending to negative values, will provide a better picture of the BOAS retrieval performance since there is a significant number of pixels with negative columns. Which orbits contribute to these plots?

Responses: Thank you very much for this suggestion. The color bar range in Figure 4 is changed into from -2 x 10^{16} to 2 x 10^{16}. And the Figure 3 is updated. The orbits from 04210 to 04213 contribute to these plots.

Figure 4: Does plot (a) show ΔSCDs and plot (b) SCDs? Please clarify. Does this regression consider an independent variable? Probably is better to consider the errors of both variables in the linear fit.

Responses: Thank you very much for this suggestion. Figure 5 (a) shows the DSCDs comparison and (b) shows SCDs comparison. It is clarified in the revised version. We don't consider an independent variables in the linear fit. The linear fit in our study is more direct to show the difference between the two data sets.

Table 3: It is very difficult to interpret. How can be the NMBs between satellite and MAX-DOAS be 0% for the operational product? The caption needs to be re-written to provide a proper description of what the table is showing.

Responses: Thank you very much for this suggestion. $NMB_{S1,S2}$ is NMBs between TROPOMI HCHO VCDs with different retrieval settings, and $NMB_{S1,M}$ is NMBs between TROPOMI and MAX-DOAS observations. The caption is changed into "NMBs between TROPOMI HCHO VCDs with different retrieval settings ($NMB_{S1,S2}$) and NMBs between TROPOMI and MAX-DOAS observations ($NMB_{S1,M}$). TROPOMI HCHO VCDs are calculated with four different settings, (1) operational retrieval setting (2) replacing DOAS SCDs using BOAS SCDs in the operational product, (3) changing the a priori profiles from TM5 to regional WRF-Chem simulations in the operational product and (4) both (2) and (3) changes in the operational product. The error bars ($\pm$ 2 SE) are also presented. All values are in %."

Figure 5: What is the methodology used to calculate the vertical profiles? Time averaging, filtering of MAX-DOAS observations… Co-registration of models and models with in-situ measurements.

Responses: Thank you very much for this suggestion. In AMF calculations, both WRF-Chem and TM5-MP simulations are interpolated to TROPOMI spatial resolution. Interpolated WRF-Chem and TM5-MP simulations within 20 km of the MAX-DOAS site are spatially averaged to compare with MAX-DOAS profiles. MAX-DOAS profiles are temporally averaged in the period 13:30-14:30 (Local Time) within ±1 h around the TROPOMI overpass time.

Changes in manuscript: L366-370, P16 in the revised version.

Figure 6: Using a divergent color map scale in plot © will help to appreciate the positive and negative differences. What is the effect of clouds and aerosols? Some of the big differences resemble cloud structures. What is the correlation between both AMF calculations?

Responses: Thank you very much for this suggestion. The original Figure 6 is Figure 7 in the revised version. The color bar of Figure. 7(c) is updated to appreciate the positive and negative differences easily. We added the spatial distribution of cloud fraction (d) of orbit 04211 on 06 August 2018 in Figure. 7(d). The results show a similar spatial pattern. However, WRF-Chem AMF is mostly higher than the one calculated with TM5-MP HCHO profiles. The big differences of two AMFs are mainly occurred in pixels with large cloud fraction.

Changes in manuscript: L383-384, P17 in the revised version.

Figure 8: Do these plots show improve HCHO data? It seems to show gridded data. What is the spatial sampling of the grid? Which methodology was used to calculate the gridded fields?

Responses: Thank you very much for this suggestion. Figure 8 shows the averaged HCHO VCDs in four seasons. Before averaging, TROPOMI HCHO VCDs in our retrieval are gridded into same grids (0.1°×0.1°). The grid methodology follows the method in (Zhu et al., 2017). The weights are calculated using relative error (Re) and intensity-weighted cloud fraction (cf):

$$\overline{N_V}(i) = \frac{\sum_{p=1}^{p=n(i)} \frac{N_v(p)}{\left(Re(p) * (1 + 3 * cf(p))\right)^2}}{\sum_{p=1}^{p=n(i)} \frac{1}{\left(Re(p) * (1 + 3 * cf(p))\right)^2}}$$

Where $\overline{N_V}(i)$ is gidded HCHO VCDs of grid cell. n(i) is number of satellite pixels falling in the grid cell i.

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