### **Responses to Referee's Comments**

We appreciate careful reading and lots of valuable comments. We wrote referee's comments in black, our responses to comments in blue and italics,

5 *and the revised manuscript in red.* 

### Referee #1:

In this paper, Kwon et al. described the HCHO retrieval algorithm to be implemented with the Geostationary Environment Monitoring Spectrometer (GEMS). The authors
discussed the three main steps in the algorithm (namely preprocessing, spectral fitting, and postprocessing), carried out uncertainty analysis, and also compared GEMS results (using OMI radiance data) with existing OMI HCHO results and MAX-DOAS at a few stations. Once launched, the HCHO data from GEMS can potentially be used in studies on regional air quality, and biomass burning in large areas over East and Southeast Asia.

- 15 A paper providing detailed documentation of the retrieval algorithm is certainly of great interest to data users and the satellite remote sensing community. Overall, the paper is well organized, and figures and tables are mostly clear. I would recommend publication in AMT after some clarifications (see below):
- 20 Specific comments: One would assume that there are some similarities and differences between the GEMS and OMHCHO algorithms. Some of these are discussed throughout the text, but it would be useful to have a table or a paragraph summarizing the different setups (and the resulting differences in HCHO) between the two instruments.
- We used the same fitting options with OMHCHO products (González Abad et al., 2015), but auxiliary data such as model data for background corrections and AMF LUT are different. Also, we do not use undersampling correction and latitudinal bias correction for GEMS in default. For clarity, we only described GEMS fitting options in Section 2.2 and added Section 4.1 to explain differences of fitting options between GEMS and OMI as follows:

### 4.1 Retrieval of OMI HCHO

GEMS fitting options described in Table 1 are largely consistent with those of OMHCHO products (González Abad et al., 2015). However, we do not include spectral undersampling (Chance et al., 2005) in the fitting process for GEMS, and reference sectors for a radiance reference are 143-150°E (shaded areas in Fig. 1). For OMI products, spectral undersampling needs to be included, and radiance references are from the Pacific Ocean as described in González Abad et al. (2015). We use simulated HCHO vertical columns for the background correction, which are zonally and monthly averaged over the reference sector (140-160°W, 90°S-90°N) except for Hawaii (154-160°W, 19-22°N).

- 10 reference sector (140-160°W, 90°S-90°N) except for Hawaii (154-160°W, 19-22°N). In addition, we need to correct latitudinal biases for OMI. Previous studies explained that the latitudinal biases result from spectral interferences of BrO and O<sub>3</sub>, whose concentrations are a function of latitude and are high in high latitudes (De Smedt et al., 2008; De Smedt et al., 2015; González Abad et al., 2015). Therefore, the latitudinal biases
- 15 were corrected when a radiance reference was used as the reference spectrum (De Smedt et al., 2008; González Abad et al., 2015; De Smedt et al., 2018). We correct the latitudinal biases, which are slant columns retrieved for a radiance reference and are averaged as a function of latitude, by subtracting the biases from the corrected slant columns in Eq. 11. Figure 6 shows OMI HCHO slant columns from OMHCHO products (Fig. 6a) and the
- 20 GEMS algorithm without and with latitudinal bias corrections (Fig. 6b and 6c). HCHO slant columns without latitudinal bias corrections (Fig. 6b) are retrieved larger in 5°N-25°N than OMHCHO products, but HCHO slant columns with the bias corrections are in better agreement with OMHCHO products. Figure 6d shows the absolute differences between OMI HCHO slant columns with and without latitudinal bias corrections from
- 25 the GEMS algorithm as latitudinal biases. Slant columns with bias corrections increase at latitudes lower than 5°N and higher than 25°N but decrease at latitudes from 5°N-25°N. However, latitudinal biases can be minimized when using a radiance reference as a function of each cross-track position in the south to north direction for GEMS. In default fitting options, therefore, we do not include latitudinal correction and do not analyze uncertainty of latitudinal corrections in Section 3. However, a further investigation for
- 30 uncertainty of latitudinal corrections in Section 3. However, a further investigation fo the latitudinal biases needs to be required after GEMS is launched.

Figure 7 shows an example of retrieved HCHO optical depths and fitting residuals as functions of wavelengths for a pixel in Indonesia (March 23 2005; orbit 3655). The retrieved HCHO slant column is  $3.2 \times 10^{16}$  molecules cm<sup>-2</sup>, which is relatively high due to biomass burning in that region. Average slant column and random uncertainty for all

- 5 pixels on the orbit are  $7.6 \times 10^{15}$  and  $6.9 \times 10^{15}$  molecules cm<sup>-2</sup>, respectively, over the GEMS domain. The large random uncertainty of 100% or larger results from pixels with low concentrations, where averaged slant columns and random uncertainties are  $2.2 \times 10^{15}$  and  $6.2 \times 10^{15}$  molecules cm<sup>-2</sup>.
- 10 It appears that the background correction is a main contributor to the differences between GEMS and OMHCHO. And the two used different versions of GEOS-Chem for background VCDs. Is it possible to compare the model HCHO VCDs from the same model over the GEMS "background" area and OMHCHO "background" area? The easternmost part of GEMS FOR is still relatively close to Asia (and biomass burning and CH4 sources). A comparison may help to determine if the GEMS background is
- "background" enough.

In operation for GEMS, we will use easternmost regions as GEMS reference sectors (143-150°E), which are relatively clean areas for GEMS. Figure S1 shows model
HCHO VCDs in GEMS background area (dashed) and OMI background area (solid) and absolute differences between the two.
We discussed it as follows:

For GEMS, we plan to use simulated HCHO columns over easternmost regions (143-150°E) as GEMS reference sectors, which are shaded areas in Fig. 1. The GEMS reference sectors include part of islands near the equator and Japan but are relatively clean areas in south/north direction over the GEMS domain. In comparisons with background HCHO vertical columns over the Pacific Ocean for OMI (Fig. S1), annual mean of GEMS background columns over 4°S–45°N is 3.3 × 10<sup>15</sup> molecules cm<sup>-2</sup> slightly higher than that

30 of OMI background columns  $(3.2 \times 10^{15} \text{ molecules cm}^2)$ , showing that we can use easternmost regions as background in the GEMS domain. Occasionally, local differences

between GEMS and OMI background columns can be as large as  $3.8 \times 10^{15}$  molecules cm<sup>-2</sup> in the tropical region of the southern hemisphere due to biogenic activity and biomass burning, but the standard deviation of background values in that region is  $5.1 \times 10^{14}$  molecules cm<sup>-2</sup> even lower than that of  $1.2 \times 10^{15}$  molecules cm<sup>-2</sup> in the middle latitude (>30°N), indicating that the influences from biogenic activity and biomass burning can be corrected by model simulations.

5



10 Figure S1. Simulated HCHO vertical column densities in GEMS background area (dashed lines) and OMI background area (solid lines) (top), and absolute differences between the two (bottom).

15 Some symbols used in equations are not defined (immediately before or after the

introduction of the equation), for example i and j in equation 11.

### We explained symbols of i, j, and $VCD_m$ after Eq. 11 as follows:

5 where i and j indicate pixel indices of cross and along tracks, respectively, and VCD<sub>m</sub> denotes a background vertical column density from the model.

Page 7, Line 6: is 300 DU VCD of ozone for the pseudo cross section calculation?

10 The value of 300 DU is a slant column density of ozone for the pseudo cross section calculation and is scd<sub>ref</sub> in Eq. 5.

Page 8, Line 7 and Figure 1: maybe you can define and plot the background areas?

# 15 I defined longitudinal ranges (143-150 °E) for common modes and radiance references. We updated Fig. 1:



Figure 1. GEMS field of regard (red), nominal daily scan (blue), full central scan (magenta),
 full western scan (cyan), and GEMS location (blue star). Shaded areas (143-150°E) are regions for radiance references and common mode.

*We also updated Table 1 and modified some sentences related with reference sectors as follows:* 

- 5 The common mode denotes fitting residuals caused by instrument properties which have not been determined from physical analysis. Accounting for the common mode can reduce fitting residuals and fitting uncertainties without affecting the retrieved slant columns (González Abad et al., 2015). The common mode for GEMS can be calculated by averaging fitting residuals at every cross-track over easternmost swaths (143-150°E)
- 10 shown as shaded areas in Fig. 1, which are relatively clean regions. Table 1 summarizes the detailed information used in the GEMS HCHO retrieval algorithm. We follow fitting options in González Abad et al. (2015). We use measured radiances as the reference spectrum, called a radiance reference, and measured radiances are averaged over the easternmost swaths (143-150°E; shaded areas in Fig. 1) for a day
- 15 as a function of cross-track positions in the south to north direction. Background corrections are required when we use a radiance reference and are discussed in Section 2.2.5. Also, GEMS has cross-track swaths in the south to north directions while instruments such as OMI and TROPOMI have west to east swath. Therefore, latitudinal biases resulting from BrO and O<sub>3</sub> latitude-dependent interferences can be minimized for
- 20 GEMS and are discussed in Section 4.1.

GEMS system attributes		
Spectral range	300–500 nm	
Spectral resolution	< 0.6 nm	
Wavelength sampling	< 0.2 nm	
Signal-to-noise ratio	> 720 at 320 nm	
	> 1500 at 430 nm	
Field - Conservat	$\geq 5000 \text{ (N/S)} \times 5000 \text{ (E/W)} \text{ km}^2$	
Field of regard	(5°S-45°N, 75°E-145°E)	

 Table 1. Summary of GEMS system attributes, parameters for radiance fitting, and parameters for the AMF look-up table.

Spatial resolution (at Secul)	$< 3.5 \times 8 \text{ km}^2$ for aerosol		
Spanar resolution (at Scour)	$< 7 \times 8 \text{ km}^2$ for gas		
Duty cycle	$\sim 8 \text{ times/day}$		
Imaging time	$\leq$ 30 minutes		
Radiance fitting parameters <sup>a</sup>			
Fitting window (calibration window)	328.5–356.5 nm (325.5–358.5 nm)		
De lieure reference	Measured radiances from far east swaths		
Radiance reference	(143-150°E) for a day		
Solar reference spectrum	Chance and Kurucz (2010) <sup>b</sup>		
Absorption cross-sections	HCHO at 300 K (Chance and Orphal, 2011)		
	O3 at 228 K and 295 K (Malicet et al.,		
	1995; Daumont et al., 1992)		
	NO <sub>2</sub> at 220 K (Vandaele et al., 1998) <sup>b</sup>		
	BrO at 228 K (Wilmouth et al., 1999)		
	O4 at 293 K (Thalman and Volkamer,		
	2013) <sup>b</sup>		
Ring effect	Chance and Spurr (1997) <sup>b</sup>		
Common mode	On-line common mode from easternmost		
Common mode	swaths (143-150°E) for a day		
Scaling and baseline polynomials	3 <sup>rd</sup> order		
AMF look-up table parameters			
Longitude (degree) (n=33)	70 to 150 with 2.5 grid		
Latitude (degree) (n=30)	-4 to 54 with 2.0 grid		
Solar Zenith Angle (degree) (n=9)	0, 10, 20, 30, 40, 50, 60, 70, 80		
Viewing Zenith Angle (degree) (n=9)	0, 10, 20, 30, 40, 50, 60, 70, 80		
Relative Azimuth Angle (degree) (n=3)	0, 90, 180		
Cloud Top Pressure (hPa) (n=7)	900, 800, 700, 600, 500, 300, 100		
Surface Albedo (n=7)	0, 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.0		

<sup>a</sup> GEMS fitting parameters follow González Abad et al. (2015). However, undersampling is not included in the fitting parameters for GEMS, and reference sectors for radiance reference and common mode are different.

<sup>b</sup> The datasets are used in QA4ECV retrievals. Please refer to De Smedt et al. (2018) for other datasets and fitting options.

Page 11, Line 15: would you expect that destriping would be necessary in the south/north direction?

5

GEMS has cross tracks (swaths) in the south to north directions. When we use radiance references, we make radiance references as a function of cross-track positions, already including effects of latitudinal dependent ozone absorption. Therefore, we think stripe patterns or biases would not be expected in the south/north direction.

10 We explained latitudinal biases in more detail as follows:

In addition, we need to correct latitudinal biases for OMI. Previous studies explained that the latitudinal biases result from spectral interferences of BrO and  $O_3$ , whose concentrations are a function of latitude and are high in high latitudes (De Smedt et al.,

15 2008; De Smedt et al., 2015; González Abad et al., 2015). Therefore, the latitudinal biases were corrected when a radiance reference was used as the reference spectrum (De Smedt et al., 2008; González Abad et al., 2015; De Smedt et al., 2018). We correct the latitudinal biases, which are slant columns retrieved for a radiance reference and are averaged as a function of latitude, by subtracting the biases from the corrected slant columns in Eq. 11.

20

. . .

However, latitudinal biases can be minimized when using a radiance reference as a function of each cross-track position in the south to north direction for GEMS. In default fitting options, therefore, we do not include latitudinal correction and do not analyze uncertainty of latitudinal corrections in Section 3. However, a further investigation for

25 the latitudinal biases needs to be required after GEMS is launched.

It appears that latitudinal correction is implemented for the GEMS prototype algorithm – can the authors discuss uncertainties associated with this?

30 As we mentioned above, latitudinal corrections are not included for GEMS. For clarity, we remained explanation only related with GEMS in Section 2.2 and explained fitting options for OMI in Section 4.1. Please refer to the first answer. Figure 7: the figure is quite confusing – can the authors provide more detailed description and discussion?

- 5 AMF uncertainties are as functions of parameters in the AMF LUT and are sensitive to measurement conditions. Therefore, figure 7 is too confusing to explain the contributions of parameters so that we deleted Fig. 7. However, we added Table 2 to describe retrieval uncertainties of GEMS HCHO VCDs due to AMF uncertainties. We discussed it as follows:
- 10

Table 2 summarizes estimated retrieval uncertainties of GEMS HCHO VCDs due to AMF uncertainties as functions of surface albedos, cloud top pressures, and cloud fractions. Values are calculated assuming conditions with solar zenith angle of 30°, viewing zenith angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile

- height of 700 hPa. Uncertainties of HCHO VCDs due to AMF uncertainties can be as large as 20% and 24% of HCHO VCDs in clean and polluted areas, respectively. Maximum values occur for conditions with low surface albedo and clouds at high altitudes, and high cloud fractions, but they do not differ much between clean and polluted areas. However, AMF driven HCHO uncertainty with respect to the profile height in polluted areas is higher than that in clean areas, implying that accurate HCHO profile
- information in polluted areas is important for the GEMS HCHO retrieval. We can minimize the a priori HCHO profile uncertainties by using averaging kernels.
- Table 2. Retrieval uncertainties of GEMS HCHO VCD due to AMF uncertainties as functions of surface albedos, cloud top pressures, cloud fractions, and HCHO profile heights for clean and polluted areas. Values are calculated for conditions with solar zenith angle of 30°, viewing zenith angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile height of 700 hPa.

AMF contribution to	Clean	Polluted	
HCHO VCD uncertainty	Clean		
Surface albedo ( $\alpha_s$ )	1-10%	1-12%	
Cloud top pressure $(p_c)$	0-11%	0-11%	
Cloud fraction $(f_c)$	0-19%	0-17%	

HCHO height $(p_h)$	0-11%	0-17%
Total	2-20%	3-24%

Page 16, Line 29-34: can the authors briefly mention what kind of method/strategy/data will be used for aerosol correction, in the case of dust/smoke?

5

*We briefly referred to methods for aerosol correction. We added sentences below the paragraph as follows:* 

We plan to update our AMF LUT as a function of aerosol optical depth, single scattering
albedo, and aerosol height, which will be retrieved in GEMS, to account for the effect of absorbing aerosols. On-line AMF calculation can also be used for aerosol correction with cloud information and model simulation (Lin et al., 2014).

Figure 11 and related discussion: if ozone is an important contributor to the differences
between GEMS and BIRA OMI, maybe the authors can also compare the results from tropics and mid-latitude areas separately? One may expect somewhat better agreement between the two in the tropics? Or maybe the authors can run some test GEMS retrievals using the ozone cross section as used in BIRA retrievals?

20 We found that differences between GEMS and QA4ECV (BIRA) OMI results are caused by polynomial orders. We tested effects of polynomial orders on slant columns. Also, we tested effects of different O<sub>3</sub> absorption cross-sections and non-linear O<sub>3</sub> absorption.

We found that using the 4<sup>th</sup> polynomial order improves both correlation coefficient and
regression slopes although OMI GEMS HCHO slant columns are higher than those in
QA4ECV (Fig. S2). Using different O<sub>3</sub> datasets and considering non-linear effects of
O<sub>3</sub> improve statistics and relative differences between GEMS and QA4ECV products
(Table S1 and S2).

We discussed them in the manuscript as follows:

The discrepancy between the two products could result from the radiance fitting. The OMI QA4ECV products use the DOAS method while the GEMS algorithm uses a nonlinearized fitting method (BOAS) for radiance fitting. We also find that polynomial orders

- 5 accounting for Rayleigh and Mie scatterings are important factors, causing differences between the two products. Retrieved slant columns using the 4<sup>th</sup> polynomial order are in better agreement with the QA4ECV products (Fig. S2). Both correlation coefficient and regression slope are improved although OMI GEMS HCHO values are higher than those of the QA4ECV. We use the 4<sup>th</sup> order polynomial instead of the 5<sup>th</sup> order used in the
- QA4ECV products because slant columns retrieved using the 5<sup>th</sup> order in the GEMS algorithms are much higher than the QA4ECV products.
  Also, different O<sub>3</sub> absorption cross sections (Serdyuchenko et al., 2014) are used in the OMI QA4ECV at different temperatures (220 and 243 K), and a non-linear O<sub>3</sub> absorption effect (Pukīte et al., 2010) is included in the OMI QA4ECV. We examine the O<sub>3</sub> effects
- 15 on retrieved slant columns in GEMS algorithm using O<sub>3</sub> datasets used in QA4ECV and considering a non-linear O<sub>3</sub> absorption effect. Correlation coefficient and regression slopes are slightly improved (Table S1), and relative differences in the four regions defined above are slightly reduced in most seasons and regions (Table S2).

20

Table S1. Spatial correlation coefficients and slopes between OMI GEMS and OMI QA4ECV. Left values are statistics in Fig. 10, and right values are statistics of OMI GEMS using O<sub>3</sub> datasets used in QA4ECV and considering non-linear O<sub>3</sub> absorption effects.

Statistics	(	OMI GEMS vs.	OMI QA4ECV	V
Statistics	Mar.	Jun.	Sep.	Dec.
R	0.76 / 0.75	0.66 / 0.70	0.64 / 0.67	0.52 / 0.54
Slope	0.92 / 1.02	0.76 / 0.82	0.85 / 0.91	0.79 / 0.84

25



considering non-linear O <sub>3</sub> absorption effects.					
Degion	<b>OMIGEMS vs. OMI QA4ECV</b>				
Region	Mar.	Jun.	Sep.	Dec.	
Sumatra/Malaysia (95°-110°E, 0°-7°N)	-0.5% / 3%	-18% / -17%	-6% / -4%	-15% / -13%	
Indochina Peninsula (97°-110°E, 10°-20°N)	-7% / -3%	-20% / -18%	-20% / -15%	-17% / -12%	
China (110°-120°E, 30°-40N)	-21% / -25 %	-25% / -20%	-20% / -14%	-23% / -23%	
Borneo (110°-118°E, 5°S-0°)	-9% / -5%	-13% / -9%	-0.4% / 5%	-18% / -16%	

slant columns in four regions. Left values are relative differences in Table 3 and right values are relative differences of OMI GEMS using O<sub>3</sub> datasets used in QA4ECV and considering non-linear O<sub>3</sub> absorption effects.





Figure S2. The same as Fig.10 but OMI GEMS products are retrieved using the 4<sup>th</sup> order polynomial instead of the 3<sup>rd</sup> order polynomial in default fitting options.

10

### **Responses to Referee's Comments**

We appreciate careful reading and lots of valuable comments. We wrote referee's comments in black, our responses to comments in blue and italics,

5 *and the revised manuscript in red.* 

### Referee #2:

10

This is a useful and timely manuscript on the algorithm for HCHO retrievals with the GEMS geostationary sensor that will observe the atmosphere in the near future over eastern Asia. It is useful because the algorithm is discussed in a step-by-step manner, and a thorough uncertainty assessment is included, and a comparison to independent data is provided. The discussion of the systematic component of the uncertainty is very strong.

It is timely because the launch of GEMS is imminent, and the community would like to

learn how retrievals are different or better than what we know from OMI and TROPOMI.

15 I recommend publication of the paper after the following issues are accounted or considered for.

### Major issues

1. The paper focuses on testing the retrieval algorithm for OMI-type viewing conditions.

- 20 It therefore remains unclear how the GEMS HCHO retrieval approach will account for diurnally varying measurement conditions. Surface reflectivity, HCHO profile shape, clouds will all change throughout the day, and it is unclear how these changes will affect the retrieval and their uncertainties. This is a major hiatus in this paper should be addressed.
- 25 Thanks for your suggestions about our weakness. As you mentioned, HCHO products can be sensitive to diurnally varying parameters such as surface reflectivity, HCHO profile shape, and clouds. Even though these parameters can affect the radiance fitting, we can estimate effects of variations of the parameters on AMF. Uncertainty of AMF as a function of the parameters was discussed in Section 3.2.
- 30 To examine sensitivity of AMF to HCHO profile height, we added Fig. 5d.

In addition, we define a profile height parameter  $(p_h)$  as an altitude below which 75% of HCHO VCDs exist from the surface, to estimate AMF uncertainty with respect to a HCHO profile shape.

5 Figure 5d shows increasing AMF values with an increase in the profile height, resulting from increased HCHO absorptions at high altitudes. The AMF sensitivity to profile heights in clean areas is higher than that in polluted areas because HCHO distributions are more uniform in clean areas than polluted areas.



10

. . .

Figure 5. AMF variations as functions of (a) surface albedo, (b) cloud top pressure (CTP), (c) effective cloud fraction  $(f_c)$ , and (d) profile height over clean (blue) and polluted (red) areas. Conditions of the AMF LUT are given in the figures. For sensitivity to surface albedo, cloud-free conditions are assumed. For sensitivity to cloud fraction, cloud top pressures are 800 hPa (solid line) and 500 hPa (dashed line).

15

We deleted Fig. 7 because it is too confusing to explain the contributions of parameters. Instead, we added Table 2 to describe retrieval uncertainties of GEMS HCHO VCDs due to AMF uncertainties. We discussed it as follows:

Table 2 summarizes estimated retrieval uncertainties of GEMS HCHO VCDs due to AMF uncertainties as functions of surface albedos, cloud top pressures, and cloud fractions. Values are calculated assuming conditions with solar zenith angle of 30°, viewing zenith

- 10 angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile height of 700 hPa. Uncertainties of HCHO VCDs can be as large as 20% and 24% in clean and polluted areas, respectively. Maximum values occur for conditions with low surface albedo and clouds at high altitudes, and high cloud fractions, but they do not differ much between clean and polluted areas. However, AMF driven HCHO uncertainty with
- 15 respect to the profile height in polluted areas is higher than that in clean areas, implying that accurate HCHO profile information in polluted areas is important for the GEMS HCHO retrieval. We can minimize the a priori HCHO profile uncertainties by using averaging kernels.

20	Table 2. Retrieval uncertainties of GEMS HCHO VCD due to AMF uncertainties as functions
	of surface albedos, cloud top pressures, cloud fractions, and HCHO profile heights for clean and
	polluted areas. Values are calculated for conditions with solar zenith angle of 30°, viewing zenith
	angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile height of
	700 hPa.

HCHO VCD uncertainty	Clean	Polluted	
due to AMF uncertainty	Clean		
Surface albedo ( $\alpha_s$ )	1-10%	1-12%	
Cloud top pressure $(p_c)$	0-11%	0-11%	
Cloud fraction $(f_c)$	0-19%	0-17%	
HCHO height $(p_h)$	0-11%	0-17%	
Total	2-20%	3-24%	

25

5

# We additionally discussed how to consider diurnally varying parameters in the GEMS in detail.

Surface albedo, effective cloud fraction, and cloud top pressure are retrieved from GEMS and are used in the AMF calculations. GEMS Level 2 surface properties include

- 5 Lambertian equivalent reflectivity (LER) and the daily bidirectional reflectance distribution function (BRDF) (Lee and Yoo, 2018). GEMS LER products are retrieved as composites of minimum LER values for 15 days every hour with fixed viewing geometry so that geometry dependent LER are yielded. The effective cloud fraction and cloud top pressure (effective cloud pressure) are retrieved from GEMS with the assumption of a
- 10 Lambertian cloud surface (cloud surface albedo = 0.8) (Veefkind et al., 2016). GEMS surface reflectivity products are also used for cloud retrievals. In addition, the radiative cloud fraction  $(f_{rc})$  will be provided from GEMS Level 2 cloud products, and is defined by Eq. 9, where  $I_{cld}$  and  $I_{clr}$  are radiances over cloud and cloud-free surfaces, respectively.

15

. . .

However, the horizontal resolution of  $2^{\circ} \times 2.5^{\circ}$  for HCHO profiles in AMF LUT is much coarser than the GEMS horizontal resolution of  $7 \times 8 \text{ km}^2$  to discern spatial variations by local source emissions. HCHO profiles in AMF LUT are monthly averaged so that hourly variations are not accounted for. In order to resolve these rough conditions, we can use

- 20 HCHO profiles with a finer resolution as a function of time. For example, Kwon et al. (2017) showed that HCHO retrievals using monthly mean hourly AMF values were in better agreement with the model simulations in observation system simulation experiments (OSSE) than those using monthly mean AMF values. Also, air quality forecasting data can be used to consider hourly varying HCHO profiles. Further studies 25 are required to examine the dependency of AMF calculations on spatial resolutions and

30

temporal variations of HCHO profiles and its effect on GEMS retrieval.

2. Even though the instrument is still to be launched, the paper should give more information on the GEMS instrument and how its data will be explored. What is the anticipated signal-to-noise for the HCHO spectral window, or how would it compare to OMI and TROPOMI?

Requirements of the signal-to-noise ratio for GEMS are greater than 720 at 320 nm and 1500 at 430 nm for natural spatial resolutions of  $3.5 \times 8 \text{ km}^2$ . Required signal-to noise-ratios of OMI are 1450 in 335-365 nm, 700 in 365-420 nm, and 2600 in 420-450 nm for spatial resolution of  $13 \times 24 \text{ km}^2$  (OMI L1B ATBD). Signal-to-noise ratios of

5 TROPOMI are 800-1000 in 310-405 nm and 405-500 nm (Veefkind et al., 2012). GEMS signal-to-noise ratios are comparable with those of OMI and TROPOMI. We added sentences and updated Table 1 as follows:

Requirements of signal-to-noise ratio for GEMS are 720 and 1500 at 320 and 430 nm,
respectively, for natural spatial resolutions (3.5 × 8 km<sup>2</sup> over Seoul). However, pixels are co-added in order to increase signal-to-noise ratio, and GEMS will provide spatial resolutions of 7 × 8 km<sup>2</sup> or less over Seoul, South Korea for trace gases.

GEMS system attributes		
Spectral range	300–500 nm	
Spectral resolution	< 0.6 nm	
Wavelength sampling	< 0.2 nm	
Signal to poice ratio	> 720 at 320 nm	
Signal-to-noise ratio	> 1500 at 430 nm	
Field - Conservat	$\geq 5000 \text{ (N/S)} \times 5000 \text{ (E/W)} \text{ km}^2$	
Field of regard	(5°S-45°N, 75°E-145°E)	
Spatial resolution (at Seoul)	$<$ 3.5 $\times$ 8 km <sup>2</sup> for aerosol	
	$< 7 \times 8 \text{ km}^2$ for gas	
Duty cycle	~ 8 times/day	
Imaging time	$\leq$ 30 minutes	
Radiance fitting parameters <sup>a</sup>		
Fitting window (calibration window)	328.5–356.5 nm (325.5–358.5 nm)	
Dediance reference	Measured radiances from far east swaths	
	(143-150°E) for a day	

Table 1. Summary of GEMS system attributes, parameters for radiance fitting, and parameters15156016

Solar reference spectrum	Chance and Kurucz (2010) <sup>b</sup>		
Absorption cross-sections	HCHO at 300 K (Chance and Orphal, 2011)		
	O3 at 228 K and 295 K (Malicet et al.,		
	1995; Daumont et al., 1992)		
	NO <sub>2</sub> at 220 K (Vandaele et al., 1998) <sup>b</sup>		
	BrO at 228 K (Wilmouth et al., 1999)		
	O4 at 293 K (Thalman and Volkamer,		
	2013) <sup>b</sup>		
Ring effect	Chance and Spurr (1997) <sup>b</sup>		
Common mode	On-line common mode from easternmost		
	swaths (143-150°E) for a day		
Scaling and baseline polynomials	3 <sup>rd</sup> order		
AMF look-up table parameters			
Longitude (degree) (n=33)	70 to 150 with 2.5 grid		
Latitude (degree) (n=30)	-4 to 54 with 2.0 grid		
Solar Zenith Angle (degree) (n=9)	0, 10, 20, 30, 40, 50, 60, 70, 80		
Viewing Zenith Angle (degree) (n=9)	0, 10, 20, 30, 40, 50, 60, 70, 80		
Relative Azimuth Angle (degree) (n=3)	0, 90, 180		
Cloud Top Pressure (hPa) (n=7)	900, 800, 700, 600, 500, 300, 100		
Surface Albedo (n=7)	0, 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.0		

<sup>a</sup> GEMS fitting parameters follow González Abad et al. (2015). However, undersampling is not included in the fitting parameters for GEMS, and reference sectors for radiance reference and common mode are different.
 <sup>b</sup> The datasets are used in QA4ECV retrievals. Please refer to De Smedt et al. (2018) for other datasets and fitting options.

5

How will the cloud retrieval from GEMS work? What surface reflectivity data will be used for the cloud and HCHO retrievals? How does the GEMS team address the issue of viewing geometry dependent surface reflectivity? These issues are not discussed, and thus

10

the paper runs the risk of being read as just another OMI HCHO approach, i.e. of little specificity to GEMS.

Thanks for your comments. Effective cloud fraction and cloud top pressure (effective

cloud pressure) from GEMS will be retrieved by using  $O_4$  absorption band with the assumption of Lambertian surface reflectors. Surface reflectance is provided as Lambertian equivalent reflectivity (LER) from GEMS Level 2 surface properties and is used for cloud and HCHO retrievals. GEMS LER products are retrieved as

- 5 composites of minimum LER values for 15 days every hour with fixed viewing geometry so that geometry dependent LER is yielded. We briefly referred to input parameters provided from GEMS Level 2 for AMF calculation and added references as follows:
- 10 Surface albedo, effective cloud fraction, and cloud top pressure are retrieved from GEMS and are used in the AMF calculations. GEMS Level 2 surface properties include Lambertian equivalent reflectivity (LER) and the daily bidirectional reflectance distribution function (BRDF) (Lee and Yoo, 2018). GEMS LER products are retrieved as composites of minimum LER values for 15 days every hour with fixed viewing geometry
- 15 so that geometry dependent LER are yielded. The effective cloud fraction and cloud top pressure (effective cloud pressure) are retrieved from GEMS with the assumption of a Lambertian cloud surface (cloud surface albedo = 0.8) (Veefkind et al., 2016). GEMS surface reflectivity products are also used for cloud retrievals. In addition, the radiative cloud fraction ( $f_{rc}$ ) will be provided from GEMS Level 2 cloud products, and is defined

20 by Eq. 9, where  $I_{cld}$  and  $I_{clr}$  are radiances over cloud and cloud-free surfaces, respectively.

3. The paper would be strengthened if the authors would provide a breakup of the uncertainty budget for typical polluted and clean conditions, e.g. in the form of a table.

25

We summarized the uncertainties of HCHO VCD due to AMF uncertainties for polluted and clean regions in Table 2 and discussed it as follows:

30 Table 2 summarizes estimated retrieval uncertainties of GEMS HCHO VCDs due to AMF uncertainties as functions of surface albedos, cloud top pressures, and cloud fractions. Values are calculated assuming conditions with solar zenith angle of 30°, viewing zenith

angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile height of 700 hPa. Uncertainties of HCHO VCDs due to AMF uncertainties can be as large as 20% and 24% of HCHO VCDs in clean and polluted areas, respectively. Maximum values occur for conditions with low surface albedo and clouds at high

5 altitudes, and high cloud fractions, but they do not differ much between clean and polluted areas. However, AMF driven HCHO uncertainty with respect to the profile height in polluted areas is higher than that in clean areas, implying that accurate HCHO profile information in polluted areas is important for the GEMS HCHO retrieval. We can minimize the a priori HCHO profile uncertainties by using averaging kernels.

10

15

Table 2. Retrieval uncertainties of GEMS HCHO VCD due to AMF uncertainties as functions of surface albedos, cloud top pressures, cloud fractions, and HCHO profile heights for clean and polluted areas. Values are calculated for conditions with solar zenith angle of 30°, viewing zenith angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile height of 700 hPa.

AMF contribution to HCHO VCD uncertainty	Clean	Polluted	
Surface albedo ( $\alpha_s$ )	1-10%	1-12%	
Cloud top pressure $(p_c)$	0-11%	0-11%	
Cloud fraction $(f_c)$	0-19%	0-17%	
HCHO height $(p_h)$	0-11%	0-17%	
Total	2-20%	3-24%	

4. I'm missing a discussion of the GEOS-Chem 2x2.5 a priori profile shapes. These are much coarser than the 7x8 km2 viewing scenes, and this will result in a substantial AMF

20

uncertainty. It is true that this can be accounted for via application of the averaging kernels, or by recomputing the AMFs with high-resolution profiles from a regional CTM or zoom-version of GEOS-Chem. In any case this issue should be discussed in more detail, and also included in the uncertainty budget.

## 25 We defined a profile height parameter $(p_h)$ as an altitude below which 75% of HCHO VCDs exist from the surface to estimate AMF uncertainty with respect to a HCHO

profile shape. We included AMF uncertainty with respect to a profile height. We discussed as follows:

- The AMF uncertainty can be estimated by each parameter in Eq. 16. We examine AMF uncertainties for surface albedo ( $\alpha_s$ ), cloud top pressure ( $p_c$ ), and effective cloud fraction ( $f_c$ ) with a solar zenith angle of 30°, a viewing zenith angle of 30°, and a relative azimuth angle of 0°. In addition, we define a profile height parameter ( $p_h$ ) as an altitude below which 75% of HCHO VCDs exist from the surface, to estimate AMF uncertainty with respect to a HCHO profile shape. The uncertainties of parameters ( $\sigma_{\alpha_s} = 0.02$ ,  $\sigma_{p_c} =$
- 10 50 *hPa*, and  $\sigma_{f_c} = 0.05$ ) are based on De Smedt et al. (2018) and will be replaced to those from GEMS Level 2 products. The uncertainty of profile height ( $\sigma_{p_h}$ ) is defined as a standard deviation of profile heights in AMF LUT, and  $\sigma_{p_h}$  in polluted and clean areas are 84 and 55 hPa, respectively.

15 
$$\sigma_{AMF}^{2} = \left(\frac{\partial AMF}{\partial \alpha_{s}}\sigma_{\alpha_{s}}\right)^{2} + \left(\frac{\partial AMF}{\partial p_{c}}\sigma_{p_{c}}\right)^{2} + \left(\frac{\partial AMF}{\partial f_{c}}\sigma_{f_{c}}\right)^{2} + \left(\frac{\partial AMF}{\partial p_{h}}\sigma_{p_{h}}\right)^{2}$$
(16)

20

. . .

Figure 5d shows increasing AMF values with an increase in the profile height, resulting
from increased HCHO absorptions at high altitudes. The AMF sensitivity to profile heights in clean areas is higher than that in polluted areas because HCHO distributions are more uniform in clean areas than polluted areas.



Figure 5. AMF variations as functions of (a) surface albedo, (b) cloud top pressure (CTP), (c) effective cloud fraction  $(f_c)$ , and (d) profile height over clean (blue) and polluted (red) areas. Conditions of the AMF LUT are given in the figures. For sensitivity to surface albedo, cloud-free conditions are assumed. For sensitivity to cloud fraction, cloud top pressures are 800 hPa (solid line) and 500 hPa (dashed line).

5. It remains very much unclear how the latitude-bias is being determined. The text onpage 11 (lines 9-11) is not clear, and the patterns shown in Figure 5(d) need explanation.

Latitudinal bias is determined by retrieved slant columns for radiance references.

Figure 5d (Figure 6d in the new manuscript) shows latitudinal bias, which is equal to averaged slant columns for radiance references as a function of latitude. We modified sentences as follows:

- 5 In addition, we need to correct latitudinal biases for OMI. Previous studies explained that the latitudinal biases result from spectral interferences of BrO and O<sub>3</sub>, whose concentrations are a function of latitude and are high in high latitudes (De Smedt et al., 2008; De Smedt et al., 2015; González Abad et al., 2015). Therefore, the latitudinal biases were corrected when a radiance reference was used as the reference spectrum (De Smedt
- et al., 2008; González Abad et al., 2015; De Smedt et al., 2018). We correct the latitudinal biases, which are slant columns retrieved for a radiance reference and are averaged as a function of latitude, by subtracting the biases from the corrected slant columns in Eq. 11.
   ...

Figure 6d shows the absolute differences between OMI HCHO slant columns with and

15 without latitudinal bias corrections from the GEMS algorithm as latitudinal biases. Slant columns with bias corrections increase at latitudes lower than 5°N and higher than 25°N but decrease at latitudes from 5°N-25°N.

### Minor comments

20 P2, L12: suggest to remove 'instrument' after SCIAMACHY.*We removed it.* 

P3, L15: suggest to use air quality in the singular*We changed the in the singular.* 

25

30

P3, L16-17: compared to TROPOMI's 7x7 km2 pixels, the 7x8 km2 resolution from GEMS is not that superior, so I suggest to nuance that statement.

### We modified the sentence as follows:

Instruments on-board these geostationary satellites have good spatial resolutions corresponding to those of TROPOMI and high signal-to-noise ratios, ...

P6: eq. (3) and (4) – suggest to use that mathematical e rather than exp which reads as computer code.

We changed "exp" into "e" in other equations (Eq. (5) and (6)) as well as Eq. (3) and (4).

5

Eq. (15) appears wrong. *We corrected Eq. 15 as follows:* 

$$\sigma_{s,j}^2 = RMS^2 \frac{m}{m-n} C_{j,j} C_{j,j},$$
(15)

10

Figure 7: what are the relative uncertainties in the AMF?

We deleted Figure 7 but add Table 2 to describe retrieval uncertainties of GEMS HCHO VCDs due to AMF uncertainties. We discussed Table 2 above.

15

20

Validation: which spatio-temporal selection criteria were used?

For comparison with OMI other products, we used monthly averages weighted by fitting uncertainties and overlapped areas between pixels and grid boxes with a horizontal resolution of  $0.25^{\circ} \times 0.25^{\circ}$ .

- For MAX-DOAS comparison, we also used the weighted monthly averages for OMI in a grid box of 0.25° at the center of site locations, and MAX-DOAS data were arithmetically averaged within OMI overpass time (12:00-15:00 local time). We updated comparisons between MAX-DOAS and OMI products for a year at OHP and Burney in 2005 and red Views herin 2016
- 25 Bremen in 2005 and at Xianghe in 2016.

### We modified paragraphs as follows:

We also compare satellite results with MAX-DOAS ground observations at HauteProvence Observatory (OHP) in France, Bremen in Germany, and Xianghe in China (Table 4). MAX-DOAS data are collected within the OMI overpass time (12:00–15:00

local time) at OHP and Bremen in 2005, and at Xianghe in 2016, respectively. We collect OMI data pixels that are overlapped by a grid box of 0.25° at the center of the site location, and average values of OMI data are weighted by uncertainties and overlapped areas between pixels and grid boxes.

- 5 Comparisons of HCHO VCDs between MAX-DOAS and satellite products are shown in Fig. 11 and Table 4. Averaged MAX-DOAS HCHO VCDs for a year are  $7.6 \times 10^{15}$ ,  $6.7 \times 10^{15}$ , and  $1.6 \times 10^{16}$  molecules cm<sup>-2</sup> at OHP, Bremen, and Xianghe, respectively. HCHO VCDs show a seasonal variation with the maximum concentrations in summer at all sites (Fig. S3). The largest monthly change is shown at Xianghe, likely driven by abundant
- 10 VOC precursors for HCHO productions compared to OHP and Bremen. Averaged HCHO VCDs from OMI GEMS are by 16%, 9%, 25% lower than those from MAX-DOAS at OHP, Bremen, and Xianghe. At Bremen, HCHO VCDs from the GEMS algorithm are in the best agreement with those of MAX-DOAS and show similar monthly variations with MAX-DOAS. OMI GEMS results at Xianghe show a monthly variation
- 15 but at OHP do not show monthly variation despite of a bit increment in summer. In particular, the GEMS algorithm yields lower HCHO VCDs in summer. These lower values may be caused by the a priori HCHO profiles used in AMF calculation. In summer, HCHO is produced and concentrated near the surface, which results in lower AMFs (higher VCDs). S. W. Kim et al. (2018) showed the anti-correlation between AMF values
- 20 and the HCHO mixing ratios at 200 m above ground level. OMHCHO products show similar tendencies as OMI GEMS, but they are much lower than those of OMI GEMS. OMI QA4ECV products are higher than MAX-DOAS at OHP and Bremen but are in the best agreement with MAX-DOAS at Xianghe compared to other satellite products.
- 25 Table 4. Averaged HCHO VCDs (molecules cm<sup>-2</sup>) from MAX-DOAS ground observations and OMI satellite data at OHP in France, Bremen in Germany, and Xianghe in China. For satellites, mean values are weighted by uncertainties and overlapped areas between satellite pixels and 0.25° grid cells for each site. Relative differences between OMI and MAX-DOAS are given in parentheses.

Site <sup>a</sup>	Class <sup>b</sup>	MAX-DOAS <sup>c</sup>	OMHCHO	OMI QA4ECV	OMI GEMS
-------------------	--------------------	-----------------------	--------	---------------	----------

OHP		$7.5 \times 10^{15}$	$5.8 \times 10^{15}$	$1.1 \times 10^{16}$	$6.3 \times 10^{15}$
(449NI 5 59E)	Rural		( 249/)	(500())	
$(44^{-1}N, 5.5^{-1}E)$			(-24%)	(50%)	(-16%)
Bremen	Urban	$6.7 \times 10^{15}$	$5.1  imes 10^{15}$	$9.3  imes 10^{15}$	$6.1 \times 10^{15}$
(53°N, 9°E)			(-23%)	(40%)	(-9%)
Xianghe	Sub-urban	1.6 × 10 <sup>16</sup>	$1.0 \times 10^{16}$	$1.7  imes 10^{16}$	$1.2 \times 10^{16}$
(39°N, 117°E)			(-37%)	(4%)	(-25%)
<sup>a</sup> HCHO VCDs are averaged at OHP and Bremen in 2005 and at Xianghe in 2016.					

<sup>b</sup> Class is assigned in a QA4ECV MAXDOAS website (http://uv-vis.aeronomie.be/groundbased/QA4ECV MAXDOAS)

<sup>c</sup> Fitting windows of 336–359 nm and 324–359 nm are used at OHP and Bremen, and at Xianghe, respectively.





Figure 11. HCHO vertical columns from MAX-DOAS, OMHCHO, OMI QA4ECV, and OMI GEMS at OHP and Bremen in 2005, and at Xianghe in 2016. Orange lines are median values for each dataset, and blue diamonds are mean values. We computed mean values of each satellite product weighted by uncertainties and overlapped areas between satellite pixels and 0.25° grid cells for each site. Boundaries of boxes indicate first and last quantiles of datasets.

10

# Reference

15 OMI L1B ATBD,

https://projects.knmi.nl/omi/documents/data/OMI\_ATBD\_Volume\_1\_V1d1.pdf

Veefkind, J. P., et al. (2012). "TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric composition for climate, air quality

20 and ozone layer applications." <u>Remote Sensing of Environment</u> **120**: 70-83.



Figure S3. Monthly variations of HCHO VCDs from MAX-DOAS (black) and OMI at OHP, Bremen, and Xianghe. Blue indicates OMHCHO, red indicates QA4ECV, and green indicates GEMS.

### **Responses to Referee's Comments**

We appreciate careful reading and lots of valuable comments. We wrote referee's comments in black, our responses to comments in blue and italics,

5 *and the revised manuscript in red.* 

### **Referee #3:**

### **General Description:**

The authors describe the retrieval algorithm of formaldehyde (HCHO) for the future
 GEMS instrument and estimate the likely uncertainties and biases relative to OMI and ground-based MAX-DOAS measurements. The content is appropriate for AMT. Suggested changes, comments and concerns are included below.

### **General Comments:**

- 15 It's not clear what's unique about the retrieval to GEMS. Seems more like a recapitulation of the OMI retrieval description paper of González Abad et al. (2015). A way to address this would be to assess the implication of the unique temporal component of GEMS (i.e. observations throughout the day) on uncertainties in the retrieval.
- 20 Thanks for suggestions. We analyzed expected random uncertainty for GEMS by using simulated radiances, which are convoluted with GEMS bandpass functions at 330 nm and include noises based on signal-to-noise ratio for co-added pixels with spatial resolutions of 7 × 8 km<sup>2</sup>. We updated related paragraphs as follows:
- 25 We analyze expected uncertainties for the GEMS algorithm by using simulated radiances from Kwon et al. (2017) and OMI Level 1B data. In order to estimate the expected random uncertainty for GEMS (Section 3.1.1), we use simulated radiances, which are convoluted with GEMS bandpass functions at 330 nm as a function of cross-track positions in the south to north direction. Simulated radiances include noises based on the expected signal-
- 30 to-noise ratio for co-added pixels with spatial resolutions of  $7 \times 8 \text{ km}^2$ . We use absorption cross-sections of Ring effect, O<sub>3</sub>, NO<sub>2</sub>, HCHO, and additionally SO<sub>2</sub> (Hermans et al.,

2009; Vandaele et al., 2009) in radiance fitting because  $O_3$ ,  $NO_2$ , and HCHO, and  $SO_2$  were considered in radiance calculation (Kwon et al. 2017).

For other uncertainty analyses, we use OMI Level 1B data with OMI slit function data (Dirksen et al., 2006) in order to examine algorithm sensitivities to individual parameters.

5 Fitting options such as absorption cross-section data and the fitting window are summarized in Table 1. It will be necessary to conduct an additional uncertainty analysis for GEMS HCHO retrievals after GEMS is launched.

# 10

. . .

Random uncertainties from the GEMS algorithm are estimated using simulated radiances. RMS of fitting residuals and random uncertainty for the GEMS domain range from  $2.9 \times 10^{-4}$  to  $2.1 \times 10^{-3}$  and  $2.1 \times 10^{15}$  to  $1.6 \times 10^{16}$  molecules cm<sup>-2</sup>, respectively, which are comparable with those (RMS:  $4 \times 10^{-4}$  to  $2.0 \times 10^{-3}$ ; random uncertainty:  $3.3 \times 10^{15}$  to 1.8

- 15  $\times 10^{16}$  molecules cm<sup>-2</sup>) obtained from the GEMS algorithm using OMI Level 1B data. GEMS measures target species every hour in daytime so that changes of solar location for a day can affect the accuracy of radiance fitting. An averaged fitting RMS value and a random uncertainty are  $6.9 \times 10^{-4}$  and  $5.0 \times 10^{15}$  molecules cm<sup>-2</sup> for conditions with both solar and viewing zenith angles less than 70, which happen at 8:00–18:00 and 9:00–
- 20 16:00 local time of Seoul in summer and winter, respectively. However, the fitting RMS value and the random uncertainty increase to  $1.1 \times 10^{-3}$  and  $8.2 \times 10^{15}$  molecules cm<sup>-2</sup>, respectively, when solar and viewing zenith angles are higher than 70.
- 25 To clarify, we remained descriptions related with GEMS in the Section 2. Descriptions related with OMI to validate the GEMS algorithm were moved to new Section 4.1.

### We described a radiance reference for GEMS in Section 2.2.3 as follows:

30 Table 1 summarizes the detailed information used in the GEMS HCHO retrieval algorithm. We follow fitting options in González Abad et al. (2015). We use measured

radiances as the reference spectrum, called a radiance reference, and measured radiances are averaged over the easternmost swaths (143-150°E; shaded areas in Fig. 1) for a day as a function of cross-track positions in the south to north direction. Background corrections are required when we use a radiance reference and are discussed in Section

5

2.2.5. Also, GEMS has cross-track swaths in the south to north directions while instruments such as OMI and TROPOMI have west to east swath. Therefore, latitudinal biases resulting from BrO and  $O_3$  latitude-dependent interferences can be minimized for GEMS and are discussed in Section 4.1.

### 10

# We described GEMS surface reflectivity and cloud information used for AMF calculation in Section 2.2.4.

Surface albedo, effective cloud fraction, and cloud top pressure are retrieved from GEMS and are used in the AMF calculations. GEMS Level 2 surface properties include

- 15 Lambertian equivalent reflectivity (LER) and the daily bidirectional reflectance distribution function (BRDF) (Lee and Yoo, 2018). GEMS LER products are retrieved as composites of minimum LER values for 15 days every hour with fixed viewing geometry so that geometry dependent LER are yielded. The effective cloud fraction and cloud top pressure (effective cloud pressure) are retrieved from GEMS with the assumption of a
- 20 Lambertian cloud surface (cloud surface albedo = 0.8) (Veefkind et al., 2016). GEMS surface reflectivity products are also used for cloud retrievals. In addition, the radiative cloud fraction  $(f_{rc})$  will be provided from GEMS Level 2 cloud products, and is defined by Eq. 9, where  $I_{cld}$  and  $I_{clr}$  are radiances over cloud and cloud-free surfaces, respectively.
- 25

# Also, we wrote a plan to consider temporal variations of a priori HCHO profiles as follows:

30 However, the horizontal resolution of  $2^{\circ} \times 2.5^{\circ}$  for HCHO profiles in AMF LUT is much coarser than the GEMS horizontal resolution of  $7 \times 8 \text{ km}^2$  to discern spatial variations by

local source emissions. HCHO profiles in AMF LUT are monthly averaged so that hourly variations are not accounted for. In order to resolve these rough conditions, we can use HCHO profiles with a finer resolution as a function of time. For example, Kwon et al. (2017) showed that HCHO retrievals using monthly mean hourly AMF values were in

5 better agreement with the model simulations in observation system simulation experiments (OSSE) than those using monthly mean AMF values. Also, air quality forecasting data can be used to consider hourly varying HCHO profiles. Further studies are required to examine the dependency of AMF calculations on spatial resolutions and temporal variations of HCHO profiles and its effect on GEMS retrieval.

10

Throughout, use the standard symbol  $\otimes$  for convolution. This will help clarify terms in equations that are confusing, as brackets are used to denote dependence, but also operators, e.g.  $f \otimes g(\lambda)$  to replace  $(f * g)(\lambda)$  in Equation (2) is clearer. Please correct these issues throughout.

15

Thanks for suggestion. We replaced the symbol \* to the symbol  $\otimes$  in Eq. (1)-(5) as follows:

$$I_R(\lambda) = I_0^h \otimes g(\lambda + \Delta \lambda) P_{sc}(\lambda) + P_{bl}(\lambda), \tag{1}$$

20

$$f \otimes g(\lambda) = \int_{-\infty}^{\infty} f(\Lambda)g(\lambda - \Lambda)d\Lambda$$
<sup>(2)</sup>

attenuated radiance in radiance fitting =  $I_0^h \otimes g(\lambda) e^{-\tau^h \otimes g(\lambda)}$ , (3)

attenuated radiance in reality = 
$$(I_0^h(\lambda)e^{-\tau^h(\lambda)})\otimes g(\lambda).$$
 (4)

$$\sigma_{ps}(\lambda) = \frac{1}{scd_{ref}} \ln\left(\frac{I_0^h \otimes g(\lambda)}{\left(I_0^h(\lambda)e^{-scd_{ref}\sigma^h(\lambda)}\right) \otimes g(\lambda)}\right),\tag{5}$$

25

Inconsistent use of wavelength dependence in equations. For example, why do  $I_R$  and  $I_0^h$  not depend on wavelength in Equations (1)-(4), but do in Equation (5)?

We changed those equations above, and we also modified variables related with Eq. (6)
30 as a function of wavelength.

$$I(\lambda) = \left[ \left( a I_0(\lambda) + c_r \sigma_r(\lambda) \right) e^{-\sum_i SCD_i \sigma_i(\lambda)} + c_{cm} \sigma_{cm}(\lambda) \right] P_{sc}(\lambda) + P_{bl}(\lambda), \quad (6)$$

Many sub-sections in Section 2.2. are the same as in González Abad et al. (2015). Why not just refer the reader to that paper and only state aspects specific to GEMS and that are

5 different between the two approaches?

As we answered to first comments, we remained descriptions related to GEMS. We also added new sub-section 4.1 to describe fitting options in the GEMS algorithm for OMI HCHO retrievals.

10

### 4.1 Retrieval of OMI HCHO

GEMS fitting options described in Table 1 are largely consistent with those of OMHCHO products (González Abad et al., 2015). However, we do not include spectral undersampling (Chance et al., 2005) in the fitting process for GEMS, and reference

- 15 sectors for a radiance reference are 143-150°E (shaded areas in Fig. 1). For OMI products, spectral undersampling needs to be included, and radiance references are from the Pacific Ocean as described in González Abad et al. (2015). We use simulated HCHO vertical columns for the background correction, which are zonally and monthly averaged over the reference sector (140-160°W, 90°S-90°N) except for Hawaii (154-160°W, 19-22°N).
- 20 In addition, we need to correct latitudinal biases for OMI. Previous studies explained that the latitudinal biases result from spectral interferences of BrO and O<sub>3</sub>, whose concentrations are a function of latitude and are high in high latitudes (De Smedt et al., 2008; De Smedt et al., 2015; González Abad et al., 2015). Therefore, the latitudinal biases were corrected when a radiance reference was used as the reference spectrum (De Smedt
- et al., 2008; González Abad et al., 2015; De Smedt et al., 2018). We correct the latitudinal biases, which are slant columns retrieved for a radiance reference and are averaged as a function of latitude, by subtracting the biases from the corrected slant columns in Eq. 11. Figure 6 shows OMI HCHO slant columns from OMHCHO products (Fig. 6a) and the GEMS algorithm without and with latitudinal bias corrections (Fig. 6b and 6c). HCHO
- 30 slant columns without latitudinal bias corrections (Fig. 6b) are retrieved larger in 5°N-25°N than OMHCHO products, but HCHO slant columns with the bias corrections are in

better agreement with OMHCHO products. Figure 6d shows the absolute differences between OMI HCHO slant columns with and without latitudinal bias corrections from the GEMS algorithm as latitudinal biases. Slant columns with bias corrections increase at latitudes lower than 5°N and higher than 25°N but decrease at latitudes from 5°N-25°N.

- 5 However, latitudinal biases can be minimized when using a radiance reference as a function of each cross-track position in the south to north direction for GEMS. In default fitting options, therefore, we do not include latitudinal correction and do not analyze uncertainty of latitudinal corrections in Section 3. However, a further investigation for the latitudinal biases needs to be required after GEMS is launched.
- 10 Figure 7 shows an example of retrieved HCHO optical depths and fitting residuals as functions of wavelengths for a pixel in Indonesia (March 23 2005; orbit 3655). The retrieved HCHO slant column is  $3.2 \times 10^{16}$  molecules cm<sup>-2</sup>, which is relatively high due to biomass burning in that region. Average slant column and random uncertainty for all pixels on the orbit are  $7.6 \times 10^{15}$  and  $6.9 \times 10^{15}$  molecules cm<sup>-2</sup>, respectively, over the
- 15 GEMS domain. The large random uncertainty of 100% or larger results from pixels with low concentrations, where averaged slant columns and random uncertainties are  $2.2 \times 10^{15}$  and  $6.2 \times 10^{15}$  molecules cm<sup>-2</sup>.
- 20 It's not clear why Section 2.2.5 is relevant, as it describes bias corrections specific to OMI. Is it anticipated that the same bias corrections will be needed for GEMS? If this section is relevant, the readers could just be referred to González Abad et al. (2015) and this section be kept brief.
- 25 Thanks for your comments. For GEMS, background corrections are only used when we use a radiance reference. To clarify, therefore, we explained background corrections for GEMS in Section 2.2.5, and corrections and discussions for OMI were moved to Section 4.1. We modified paragraphs in Section 2.2.5 as follows:
- 30

An alternative method to avoid the above-mentioned biases in the fitting procedure is to

use measured radiances over a clean background region (referred to as radiance references) as the reference spectrum in radiance fitting. As measured radiance includes instrument noise and attenuation by interfering gases in the background atmosphere, the interfering effects can be minimized in radiance fitting, which results in negligible cross-track biases.

- 5 For GEMS, we plan to use simulated HCHO columns over easternmost regions (143-150°E) as GEMS reference sectors, which are shaded areas in Fig. 1. The GEMS reference sectors include part of islands near the equator and Japan but are relatively clean areas in south/north direction over the GEMS domain. In comparisons with background HCHO vertical columns over the Pacific Ocean for OMI (Fig. S1), annual mean of GEMS
- background columns over 4°S-45°N is 3.3 × 10<sup>15</sup> molecules cm<sup>-2</sup> slightly higher than that of OMI background columns (3.2 × 10<sup>15</sup> molecules cm<sup>-2</sup>), showing that we can use easternmost regions as background in the GEMS domain. Occasionally, local differences between GEMS and OMI background columns can be as large as 3.8 × 10<sup>15</sup> molecules cm<sup>-2</sup> in the tropical region of the southern hemisphere due to biogenic activity and biomass burning, but the standard deviation of background values in that region is 5.1 ×
- $10^{14}$  molecules cm<sup>-2</sup> even lower than that of  $1.2 \times 10^{15}$  molecules cm<sup>-2</sup> in the middle latitude (>30°N), indicating that the influences from biogenic activity and biomass burning can be corrected by model simulations.
- The retrieved slant columns using a radiance reference are differential slant columns 20  $(\Delta SCD = SCD - SCD_0)$  and do not include background HCHO columns  $(SCD_0)$  that are mainly from the oxidation of methane. To account for the background columns, we use HCHO vertical columns simulated in 2014 from a chemical transport model, GEOS-Chem (Bey et al., 2001) with a spatial resolution of  $2^\circ \times 2.5^\circ$ . Simulated HCHO vertical columns are zonally and monthly averaged over the reference sectors and are interpolated
- 25 to 720 latitudinal grid points with a resolution of  $0.25^{\circ}$  from 90°S to 90°N. In order to account for dependency of measured radiances on geometric angles, we then convert simulated background vertical columns into slant columns by applying AMF values over the reference sector (*AMF*<sub>0</sub>), which are calculated with cloud information and geometric angles on the reference sectors. Corrected GEMS HCHO slant columns are
- 30 formulated as the sum of the retrieved differential slant columns and the simulated background slant columns as shown in Eq. 11,

$$\Omega_s(i,j) = SCD_{corr}(i,j) = \Delta SCD(i,j) + AMF_0(lat)VCD_m(lat),$$
(11)

where i and j indicate pixel indices of cross and along tracks, respectively, and VCD<sub>m</sub>
denotes a background vertical column density from the model. We finally apply AMF values from the LUT to the corrected slant columns to obtain GEMS HCHO vertical column densities.

It's also not clear why data quality flags are provided for a future product. This would only be important for the user when the data is ready for release.

The data quality flag is provided for basic information of data quality in radiance fitting, and we followed the flag definition from González Abad et al. (2015). We are planning to provide flags including much information such as geometry angles, clouds, surface information.

15

20

Section 3 appears to just be testing uncertainties inherent in fitting parameters and retrieval terms that would be an issue for all space-based instruments measuring HCHO, rather than being specific to GEMS. Is there anything unique to GEMS (instrument configuration, viewing domain, repeat time etc.) that would increase or decrease sensitivity to these uncertainties relative to other instruments?

Uncertainty related to GEMS instrument is considered in random uncertainty. Random uncertainty, called fitting uncertainty, is calculated from fitting residuals caused by

- 25 instrument noise, radiance measurement uncertainty from dark current and stray lights, and polarization. We estimated expected random uncertainty by using simulated radiances with GEMS bandpass functions and signal-to-noise ratios. We discussed it in the first answer.
- 30 *GEMS* does not include a polarization scrambler while OMI and TROPOMI include a polarization scrambler. In the operation, polarization correction will be conducted when L1B data are produced. The correction could minimize polarization, but it would
not be perfect. The effects could increase random uncertainty. We need to have a process to minimize polarization. We discussed it in Section 5 as follows:

5 We currently use a broad fitting window (328.5–356.0 nm). However, we may need to use a different fitting window to reduce interference from polarization effects because GEMS does not include a polarization scrambler. A polarization correction is planned to minimize its interference during GEMS Level 1B production, but we need to examine the retrieval sensitivity to polarization.

10

## **Specific Comments:**

P2, Line 18: the spatial resolution of TROPOMI is finer than 7 x 7 km2 for HCHO (De Smedt et al., 2018).

15

Veefkind et al. (2012) showed the spatial resolution of TROPOMI UVIS band 3 (310-405 nm) is  $7 \times 7 \text{ km}^2$ . However, we found TROPOMI HCHO products are provided with  $7 \times 3.5 \text{ km}^2$ . We corrected it from  $7 \times 7 \text{ km}^2$  to  $7 \times 3.5 \text{ km}^2$ .

# 20

Equation (1): Why are *Psc* and *Pbl* not dependent on wavelength?

*P<sub>sc</sub>* and *P<sub>bl</sub>* are functions of wavelength. Therefore, we changed it as follows:

25 
$$I_R(\lambda) = I_0^h \otimes g(\lambda + \Delta \lambda) P_{sc}(\lambda) + P_{bl}(\lambda),$$
 (1)

P3, Line 22: Can aerosol optical properties be retrieved across this wavelength and for this type of instrument? Do the authors mean AOD and aerosol index (AI)?

30

We meant AOD and SSA. We clarified it as follows:

Geostationary Environment Monitoring Spectrometer (GEMS) will be launched by South Korea, and it will measure radiances ranging from 300 to 500 nm every hour with fine spatial resolutions of  $3.5 \times 8 \text{ km}^2$  for aerosols or  $7 \times 8 \text{ km}^2$  for gases over Seoul in South Korea to monitor column concentrations of air pollutants including O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, and

5 HCHO, and aerosol optical properties (aerosol optical depth and single scattering albedo).

Table 1: add references for these parameters as footnotes to point to consistency with existing retrievals.

10 We marked 'a' and 'b' on datasets used in OMHCHO and QA4ECV, respectively, and explanation was written in footnotes as follows:

<b>Radiance fitting parameters</b> <sup>a</sup>			
Fitting window (calibration window)	328.5–356.5 nm (325.5–358.5 nm)		
De liener reference	Measured radiances from far east swaths		
Radiance reference	(143-150°E) for a day		
Solar reference spectrum	Chance and Kurucz (2010) <sup>b</sup>		
Absorption cross-sections	HCHO at 300 K (Chance and Orphal, 2011)		
	O3 at 228 K and 295 K (Malicet et al.,		
	1995; Daumont et al., 1992)		
	NO <sub>2</sub> at 220 K (Vandaele et al., 1998) <sup>b</sup>		
	BrO at 228 K (Wilmouth et al., 1999)		
	O4 at 293 K (Thalman and Volkamer,		
	2013) <sup>b</sup>		
Ring effect	Chance and Spurr (1997) <sup>b</sup>		
Common mode	On-line common mode from easternmost		
Common mode	swaths (143-150°E) for a day		
Scaling and baseline polynomials	3 <sup>rd</sup> order		

<sup>a</sup> GEMS fitting parameters follow González Abad et al. (2015). However, undersampling is not included in the fitting parameters for GEMS, and reference sectors for radiance reference and common mode are different.

<sup>b</sup> The datasets are used in QA4ECV retrievals. Please refer to De Smedt et al. (2018) for other datasets and fitting options.

P8, Lines 14-17: What about clouds (Millet et al., 2006)?

## We corrected the sentence as follows:

5

AMF uncertainties contribute to retrieval uncertainties by multiple factors including cloud, HCHO vertical distribution, aerosol vertical distribution, and aerosol optical properties (Millet et al., 2006; Chimot et al., 2016; Kwon et al., 2017; Hewson et al., 2015).

10

P18, Lines 8-14: Comment too on the implications of more observations over the same scene per day on uncertainty compared to OMI.

Thank your comments. We wanted to show an example of OMI HCHO results retrieved
15 from the GEMS algorithm. Therefore, we showed HCHO optical depths and fitting residuals and explained averaged HCHO slant column density and random uncertainty. In addition, we explained slant columns and random uncertainties in pixels with low concentrations as follows:

- 20 Averaged slant column and random uncertainty for all pixels on the orbit are  $7.6 \times 10^{15}$ and  $6.9 \times 10^{15}$  molecules cm<sup>-2</sup>, respectively, over the GEMS domain. The large random uncertainty of 100% or larger results from pixels with low concentrations, where averaged slant columns and random uncertainties are  $2.2 \times 10^{15}$  and  $6.2 \times 10^{15}$  molecules cm<sup>-2</sup>.
- 25

P18, Lines 26-28: Provide an appropriate reference for this statement.

## We added a reference as follow:

Zhong, L., Louie, P. K. K., Zheng, J., Yuan, Z., Yue, D., Ho, J. W. K., and Lau, A. K. H.:
Science–policy interplay: Air quality management in the Pearl River Delta region and Hong Kong, Atmospheric Environment, 76, 3-10,

# https://doi.org/10.1016/j.atmosenv.2013.03.012, 2013

Referencing: some references are missing the doi number (e.g., González Abad et al., 2015).

5

## **DOI numbers are added as follows:**

González Abad et al. (2015): 10.5194/amt-8-19-2015 Barkley et al. (2013): 118, 6849-6868, 10.1002/jgrd.50552, 2013

- Bey et al. (2001): 10.1029/2001JD000807
  Cantrell et al. (1990): 10.1021/j100373a008
  Chance et al. (1997): 10.1364/AO.36.005224
  Chance et al. (2000): 10.1029/2000GL011857
  Daumont et al. (1992): 10.1007/BF00053756
- De Smedt et al. (2008): 10.5194/acp-8-4947-2008
  Hewson et al. (2013): 10.5194/amt-6-371-2013
  Malicet et al. (1995): 10.1007/BF00696758
  Marais et al. (2012): 10.5194/acp-12-6219-2012
  Palmer et al. (2001): 10.1029/2000JD900772
- 20 Spurr (2006): 10.1016/j.jqsrt.2006.05.005 Zhu et al. (2014): 10.1088/1748-9326/9/11/114004

### **References:**

González Abad et al., Atmos. Meas. Tech., 8, 19-32, 2015, doi:10.5194/amt-8-19-2015.
De Smedt et al., Atmos. Meas. Tech., 11, 2395–2426, 2018, doi:10.5194/amt-11-2395-2018. Millet et al., J. Geophys. Res., doi:10.1029/2005JD006853, 2006

# Description of a formaldehyde retrieval algorithm for the Geostationary Environment Monitoring Spectrometer (GEMS)

5 Hyeong-Ahn Kwon<sup>1</sup>, Rokjin J. Park<sup>1</sup>, Gonzalo González Abad<sup>2</sup>, Kelly Chance<sup>2</sup>, Thomas P. Kurosu<sup>3</sup>, Jhoon Kim<sup>4</sup>, Isabelle De Smedt<sup>5</sup>, Michel Van Roozendael<sup>5</sup>, Enno Peters<sup>6,\*</sup>, and John Burrows<sup>6</sup>

<sup>3</sup>Earth Science, Jet Propulsion Laboratory, Pasadena, California, USA

<sup>4</sup>Department of Atmospheric Sciences, Yonsei University, Seoul, Republic of Korea

- <sup>5</sup>Royal Belgian Institute for Space Aeronomy (BIRA-IASB), Brussels, Belgium
   <sup>6</sup>Institute of Environmental Physics, University of Bremen, Bremen, Germany
   \*now at: DLR Institute for Protection of Maritime Infrastructures, German Aerospace Center, Bremerhaven, Germany
- 20 Correspondence to: Rokjin J. Park (rjpark@snu.ac.kr)

**Abstract.** We describe a formaldehyde (HCHO) retrieval algorithm for the Geostationary Environment Monitoring Spectrometer (GEMS) that will be launched by the Korean Ministry of Environment in 2019. The algorithm comprises three steps: pre-processes, radiance fitting,

- 25 and post-processes. The pre-processes include a wavelength calibration, and interpolation and convolution of absorption cross-sections; radiance fitting is conducted using a non-linear fitting method referred to as basic optical absorption spectroscopy (BOAS); and post-processes include air mass factor calculations and bias corrections. In this study, several sensitivity tests are conducted to examine the retrieval uncertainties using the GEMS HCHO algorithm. We
- evaluate the algorithm with the OMI Level 1B irradiance/radiance data by comparing our retrieved HCHO column densities with OMI HCHO products of the Smithsonian Astrophysical Observatory (OMHCHO) and of the Quality Assurance for Essential Climate Variables project (OMI QA4ECV). Results show that OMI HCHO slant columns retrieved using the GEMS algorithm are in good agreement with OMHCHO, with correlation coefficients of 0.77–0.91
   and regression slopes of 0.94–1.04 for March, June, September, and December 2005. Spatial

-	Deleted:	for stripe patterns, background HCHO, and latitudinal
	hiases	

Deleted: protection of maritime infrastructures

Deleted: Belgian Institute	
Deleted: Space Aeronomy	
Deleted: BIRA	

Deleted: 92

 <sup>&</sup>lt;sup>1</sup>School of Earth and Environmental Science, Seoul National University, Seoul, Republic of
 Korea

<sup>&</sup>lt;sup>2</sup>Atomic and Molecular Physics Division, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, USA

distributions of HCHO slant columns from the GEMS algorithm are consistent with the OMI QA4ECV products, but relatively poorer correlation coefficients of 0.52 to 0.76 are found compared to those against the OMHCHO products. Also, we compare the satellite results with ground-based MAX-DOAS observations. OMI GEMS HCHO vertical columns are by 9–25% lower than those of MAX-DOAS at Haute-Provence Observatory (OHP) in France, Bremen in Germany, and Xianghe in China. We find that the OMI GEMS retrievals have less bias than the OMHCHO and OMI QA4ECV products at OHP and Bremen in comparison with MAX-DOAS.

#### **1** Introduction

5

- 10 Formaldehyde (HCHO) is mainly produced by the oxidation of non-methane volatile organic compounds (NMVOCs), and it has been observed from space since the GOME instrument on ERS-2 satellite first began conducting column measurements in 1995 (Chance et al., 2000). The subsequent instrument, SCIAMACHY, on ENVISAT, collected continuous HCHO column data for 2002–2012 (Wittrock et al., 2006), and GOME-2A and -2B instruments
- 15 have been conducting measurements from 2007 to the present day (De Smedt et al., 2012; De Smedt et al., 2015). The OMI instrument was launched in 2004 and has provided global HCHO vertical column data with a higher spatial resolution of 13 × 24 km<sup>2</sup> at the nadir than that of former instruments. Furthermore, the TROPOMI equivalent to OMI has been offering consecutive data with an even finer spatial resolution of 7 × <u>3.5 km<sup>2</sup> in UVIS bands at the nadir</u> since 2017. There are thus more than 20 years of HCHO column data available from these various instruments, which enable analyses to be conducted on the global changes of HCHO columns throughout this time period.

All of the low-orbiting sun-synchronous satellite measurements of HCHO columns have played an important role in filling gaps for regions where limited (or no) in-situ measurements

of HCHO have been made, and these measurements have been used to constrain top-down estimates of biogenic and anthropogenic emissions of NMVOCs (Marais et al., 2012; Barkley et al., 2013; Stavrakou et al., 2014; Zhu et al., 2014). Together with satellite glyoxal measurements, HCHO satellite measurements have been used to distinguish dominant VOC sources (e.g., biogenic vs. anthropogenic) (DiGangi et al., 2012; Vrekoussis et al., 2010). In addition, the ratio of HCHO to nitrogen dioxide (NO<sub>2</sub>) columns has been used to determine NOx-limited or VOC-limited ozone production regimes (Martin et al., 2004; Duncan et al.,

Deleted: 7

Deleted: instrument

#### Deleted: mentioned above

Deleted: BIRA	
Deleted: have	
<b>Deleted:</b> 0.8–30	
Deleted: BIRA	
Deleted: for	

2010; Choi et al., 2012). Continuous HCHO column measurements from sun-synchronous satellites are thus invaluable for evaluating and monitoring NMVOC emission trends over long-term periods.

However, as sun-synchronous satellites have measurement frequencies of once or twice a
day, they provide limited explorations of diurnal cycles and transboundary transport of air pollutants. Moreover, their coarse spatial resolutions make discerning local source emissions difficult. With the aim of overcoming the issues, Zhu et al. (2014) used the oversampling method and temporally averaged out pixels of OMI HCHO vertical columns over high-resolution grids of ~2 km, and Kim et al. (2016) developed a downscaling method for OMI
NO<sub>2</sub> measurements by adopting the spatial distribution information from a regional air quality model. However, both these methods have inherent limitations: the former method involves a trade-off between spatial and temporal information and the latter method includes the uncertainties of emission distributions.

To tackle the limitations inherent in low-orbiting satellites measurements, environmental geostationary satellites will be launched in 2019 (or later) by South Korea and the United States, and in 2021 by European Union, to monitor air <u>quality</u> over East Asia, North America, and Europe, respectively. Instruments <u>onboard</u> these geostationary satellites have <u>spatial</u> resolutions <u>corresponding well with those of <u>TROPOMI</u> and <u>high</u> signal-to-noise ratios, and they will conduct column measurements of air pollutants every hour during the daytime. The</u>

- 20 Geostationary Environment Monitoring Spectrometer (GEMS) will be launched by South Korea, and it will measure radiances ranging from 300 to 500 nm every hour with fine spatial resolutions of 3.5 × 8 km<sup>2</sup> for aerosols or 7 × 8 km<sup>2</sup> for gases over Seoul in South Korea to monitor column concentrations of air pollutants including O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, and HCHO, and aerosol optical properties, (aerosol optical depth and single scattering albedo). The measurements from GEMS will then be used in applications such as data assimilation of air
- quality forecasts and top-down constraints of air pollutant emissions.

This paper describes a GEMS retrieval algorithm for HCHO. It also presents an uncertainty analysis and an evaluation of the algorithm, which involves comparing OMI GEMS HCHO results with OMI HCHO products from the Smithsonian Astrophysical Observatory

OMHCHO) and those from the <u>Quality Assurance for Essential Climate Variables (QA4ECV)</u> project. In addition, OMI HCHO results are compared with those of MAX-DOAS ground observations. In Section 2, we describe the GEMS instrument and provide the theoretical basis for HCHO retrievals. In Section 3, sensitivity tests are conducted to examine the retrieval Deleted: observations periods

Deleted: transports

Deleted: qualities	)
Deleted: on-board	
Deleted: superior	
Deleted: to	
Deleted: sun-synchronous satellites	
Deleted: higher	
Deleted: a	
Deleted: resolution	

Deleted:

Deleted: Belgian Institute for Space Aeronomy (OMI BIRA).

30

uncertainties; and in Section 4, we discuss an evaluation of HCHO results retrieved from the GEMS algorithm.

#### 2 GEMS HCHO algorithm

#### 2.1 GEMS instrument

5

GEMS is a scanning ultraviolet-visible spectrometer which will be launched by the Korean Ministry of Environment in 2019 <u>onboard</u> a geostationary satellite (GEO-KOMPSAT 2B), which also carries a Geostationary Ocean Color Imager-2 (GOCI-2). GEMS will be located at ~128.2°E near the equator and will cover East and Southeast Asia (5°S-45°N, 75°-145°E). The instrument will conduct hourly measurements during the day (8 times) over the

10 whole domain. It will <u>measure</u> one swath from south to north and then turn a scan mirror from east to west using an imaging time of 30 minutes and a transmission time of 30 minutes to enable GOCI-2 measurements for a 30 min period.

Solar backscattered radiances will be measured in the 300–500 nm wavelength range with a spectral resolution of 0.6 nm and a wavelength interval of 0.2 nm. <u>Signal-to-noise ratio</u>

requirements for GEMS are 720 and 1500 at 320 and 430 nm, respectively, for natural spatial resolutions (3.5 × 8 km<sup>2</sup> over Seoul). However, pixels are co-added in order to increase signal-to-noise ratio, and GEMS will provide spatial resolutions of 7 × 8 km<sup>2</sup> or less over Seoul, South Korea, for trace gases. The field of regards (FOR) and information about GEMS are shown in Fig. 1 and Table 1, respectively, Detailed information about the GEMS instrument and algorithms for species other than HCHO are found elsewhere (Kim, J. et al., 2018; Kim, M. et al., 2018; Go et al., 2019).

#### 2.2 HCHO algorithm description

25

Figure 2 is a flow chart of the HCHO retrieval algorithm for GEMS. The retrieval procedure consists of three steps: pre-processes, radiance fitting, and post-processes. The pre-processes begin with a wavelength calibration of Level 1B data (irradiance and radiance), and interpolation and convolution of absorption cross-sections at calibrated wavelength grid points. Radiance fitting is then conducted to derive the HCHO slant columns using a non-linear least square method. Finally, the post-processes include an air mass factor (AMF) calculation that employs a look-up table to convert HCHO slant columns to vertical columns, an assignment

 Deleted: on-board
 Deleted: scan
 Deleted: GEMS will provide finer
 Deleted. CEMS will provide line
 <b>Deleted:</b> compared to those of previous low-orbiting satellites (for
example, OMI has a spatial resolution of $13 \times 24$ km <sup>2</sup> at nadir).
 Deleted: ; detailed
 Deleted: 2018:
 Pereteu, 2010a

of data quality flags for each pixel, the removal of a possible stripe pattern along each crosstrack position, and corrections for background values. Each retrieval step is described in more detail in the following sections.

#### 2.2.1 Wavelength calibration and GEMS bandpass function

5 Wavelength grid points of measured irradiances and radiances in a charge-coupled device (CCD) sensor are often shifted or squeezed, and such systematic biases due to wavelength shifts or squeezes need to be corrected when producing Level 1B data. However, as precise wavelength alignments between irradiances/radiances and absorption cross-sections are required to achieve accurate radiance fitting, it is also necessary to conduct wavelength 10 calibration during retrieval.

In wavelength calibration, the solar reference spectrum (Chance and Kurucz, 2010) is firstly convolved with a GEMS bandpass function and is then interpolated to the wavelength grids of the measured spectrum. A convolved solar reference spectrum with wavelength shift parameters and polynomial parameters (Eq. 1) is then fitted to the measured irradiances and radiances in a broader fitting window (325.5–358.5 nm) than that of the radiance fitting for

15

 $I_{R_{0}}(\lambda) = I_{0}^{h} \otimes g(\lambda + \Delta \lambda) P_{so}(\lambda) + P_{bl}(\lambda),$ (1)

HCHO retrievals as follows:

20

where  $I_R(\lambda)$  is the modeled irradiance and radiance,  $I_0^h(\lambda)$  is the solar reference spectrum with a high spectral resolution of 0.01 nm wavelength interval,  $\Delta\lambda$  is the wavelength shift,  $g(\lambda)$  is a bandpass function, and  $P_{sc}(\lambda)$  and  $P_{bl}(\lambda)$  are scaling and baseline polynomials, respectively. The symbol  $\otimes$  denotes the convolution procedure, as shown in Eq. 2.

$$\frac{f \otimes g(\lambda)}{(2)} = \int_{-\infty}^{\infty} f(\Lambda)g(\lambda - \Lambda)d\Lambda$$

30

25

We use the GEMS bandpass functions for the convolution in wavelength calibration to ensure consistency with the spectral resolutions of measured irradiances and radiances. GEMS bandpass functions are provided at seven center wavelengths ranging from -1.8 nm to 1.8 nm Deleted: making

Deleted: and latitudinal biases

Deleted: asterisk

Deleted:

**Deleted:** =  $(I_0^h *$ **Deleted:** )(

Deleted:  $+P_{bl}, \rightarrow$ 

(Deleted:  $(f * g)(\lambda)$ 

at center wavelengths, with wavelength sampling intervals of 0.06 nm (Fig. 3). The right panel in Fig. 3 shows bandpass functions averaged for spatial indices at 330 and 365 nm, and it also shows the relative differences between bandpass functions at 365 nm and 330 nm. The relative differences are smaller near the wavelength center, but they increase over each wing of the function. For GEMS HCHO retrieval, we will conduct calibration for every spatial pixel of the sensor using the bandpass functions at 330 nm. However, as bandpass functions are not linear with wavelengths, it will be necessary to estimate uncertainties for the wavelength dependency

of the bandpass functions after GEMS is launched.

#### 2.2.2 Convolution and reference spectra sampling

10

15

20

25

5

Table 1 shows a summary of absorption cross-section datasets used in GEMS HCHO retrieval. In the retrieval algorithm, absorption cross-section data with fine spectral resolutions (for example, HCHO absorption cross-section data with a spectral resolution of 0.011 nm) are first convoluted with the bandpass functions described in Section 2.2.1, and they are then interpolated to the calibrated wavelength grids of measured radiances. Finally, radiance fitting accounts for attenuation of a reference spectrum (measured irradiance or radiance) by gas

absorption using Eq. 3 with convoluted cross-section data as follows:

attenuated radiance in radiance fitting = $I_0^h \otimes q_1(\lambda) e^{-\tau^h \otimes g_1(\lambda)}$ ,		D	eleted: = (l <sub>0</sub> <sup>h</sup> *
(A)	di	(D	eleted: ) $(\lambda) \exp(-(\tau^h * g)(\lambda)),$
(3)			

where  $\tau^h$  is the optical depth of interfering gases with fine spectral resolutions.

However, radiative transfer in the atmosphere occurs in a slightly different manner. Solar irradiance is firstly reduced by the absorption of interfering gases, and radiances are subsequently measured on discrete wavelength grids of an instrument with predetermined spectral resolutions, as shown in Eq. 4,

attenuated radiance in reality = $(I_0^n(\lambda)e^{-r}(\lambda))\otimes q(\lambda)$ .	* g)( <sup>A</sup> )
(4)	

Deleted: wavelength

Deleted:

Deleted: where the asterisks denote convolution (Eq. 2) and

Therefore, the difference between measured and calculated radiances on discrete grids may provide biases in radiance fitting when sensors have limited spectral resolutions, which is referred to as the solar I<sub>0</sub> effect (Aliwell et al., 2002; Chan Miller et al., 2014).

We calculate pseudo absorption cross-sections to account for the differences in gas absorptions between reality and radiance fitting. We assume that the absorption process in 5 radiance fitting is the same with that in reality, and the pseudo absorption cross-sections are computed using the following equation (Aliwell et al., 2002; Chan Miller et al., 2014),

$$\sigma_{ps}(\lambda) = \frac{1}{scd_{ref}} \ln\left(\frac{l_0^h \otimes g(\lambda)}{\left(l_0^h(\lambda)e^{-scd_{ref}\sigma^h(\lambda)}\right) \otimes g(\lambda)}\right),$$
(5)

where  $\sigma_{ps}(\lambda)$  is a pseudo absorption cross-section,  $scd_{ref}$  is a reference slant column density, and  $\sigma^h(\lambda)$  is an absorption cross-section with a fine spectral resolution.

Although the corrected absorption cross-section can be applied to all the species, we only 15 apply the correction to the O3 absorption cross-section, which is the most important interfering species in the fitting window of HCHO retrievals, and we use an O3 reference slant column density of 300 DU for the correction.

#### 2.2.3 Radiance fitting

10

Three different methods have been used with sun-synchronous satellite measurements in 20 previous HCHO retrievals: differential optical absorption spectroscopy (DOAS) method (De Smedt et al., 2008; De Smedt et al., 2012), a non-linearized fitting method, which is known as basic optical absorption spectroscopy (BOAS) (Chance et al., 2000; González Abad et al., 2015; González Abad et al., 2016), and principal component analysis (Li et al., 2015). Zhu et al. (2016) conducted an inter-comparison of HCHO vertical column densities retrieved using the three retrieval methods for four instruments such as OMI, GOME-2A, GOME-2B, and 25 OMPS; they found that the different retrieval results were consistent in terms of temporal and spatial variations of HCHO columns in the southeastern United States.

To yield HCHO slant columns in this study, we use the BOAS method, which is based on a non-linearized form of the Lambert-Beer law, as shown in Eq. 6 (González Abad et al., 2015). 30

One advantage of the BOAS method is that it uses unprocessed radiance data, and it is thus

**Deleted:**  $\ln \left( \frac{l_0^h(\lambda) * g(\lambda)}{[l_0^h(\lambda) \exp(-scd_{ref}\sigma^h(\lambda))] * g(\lambda)} \right)$ 

more intuitive than the widely used DOAS method, which uses a linearized form by taking the logarithm of radiance to irradiance and high-pass filtering the result. The modeled radiative equation is given as follows:

5 
$$I(\lambda) = \left[ \left( aI_0(\lambda) + c_r \sigma_r(\lambda) \right) e^{-\sum_i SCD_i \sigma_i(\lambda)} + c_{cm} \sigma_{cm}(\lambda) \right] P_{sc}(\lambda) + P_{bl}(\lambda)_s$$
(6)

-

10

15

where *a* is an amplitude factor;  $I_0(\lambda)$  is a reference spectrum (solar irradiance or radiance reference);  $c_r \sigma_r(\lambda)$  is the contribution of the Ring effect;  $e^{-\sum_i SCD_i \sigma_i(\lambda)}$  is the contributions of all gas absorptions;  $SCD_i$  and  $\sigma_i(\lambda)$  are slant column densities and absorption cross-sections for species *i*, respectively;  $c_{cm}\sigma_{cm}(\lambda)$  is the contribution of the common mode; and  $P_{sc}(\lambda)$  and  $P_{bl}(\lambda)$  are scaling and baseline polynomials, respectively, considering low frequency variations due to Rayleigh and Mie scattering. Furthermore, the modeled spectrum is fitted to measured radiances using a non-linear least square method (Wedin and Lindström, 1987) to yield HCHO slant columns.

The common mode denotes fitting residuals caused by instrument properties which have not been determined from physical analysis. Accounting for the common mode can reduce fitting residuals and fitting uncertainties without affecting the retrieved slant columns (González Abad et al., 2015). The common mode for GEMS can be calculated by averaging

20 fitting residuals at every cross-track over <u>casternmost swaths (143-150°E) shown as shaded</u> areas in Fig. 1, which are relatively clean regions.

<u>Table 1 summarizes the detailed information used in the GEMS HCHO retrieval</u> <u>algorithm.</u> Fitting options in González Abad et al. (2015) are followed. We use measured radiances as the reference spectrum, called a radiance reference, and measured radiances are

25 averaged over the easternmost swaths (143-150°E; shaded areas in Fig. 1) for a day as a function of cross-track positions in the south to north direction. Background corrections are required when we use a radiance reference and are discussed in Section 2.2.5. Also, GEMS has cross-track swaths in the south to north direction while instruments such as OMI and TROPOMI have west to east swath. Therefore, latitudinal biases resulting from BrO and O<sub>3</sub>

30 latitude-dependent interferences can be minimized for GEMS and are discussed in Section 4.1.

Deleted:  $[(al_0(\lambda) + c_r\sigma_r(\lambda)) \exp(-\sum_l SCD_l\sigma_l) + c_{cm}\sigma_{cm}]$ Solution Solution Moved down [1]: Table 1 summarizes the detailed information used in the GEMS HCHO retrieval algorithm. m, Deleted: ].we ce ns ng ed Deleted: far east swaths al Moved (insertion) [1]

Deleted:

#### 2.2.4 Air mass factor

HCHO slant column densities  $(\Omega_s)$  from the radiance fitting are then converted to vertical columns  $(\Omega_v)$  with an AMF (Eq. 7), which is a correction factor for the light slant path to the vertical path. Previous studies have shown that AMF uncertainty is one of the crucial factors causing retrieval uncertainties (De Smedt et al., 2012 and 2018), and AMF uncertainties contribute to retrieval uncertainties by multiple factors including cloud top pressure, cloud fraction, HCHO vertical distribution, aerosol vertical distribution, and aerosol optical properties, (Millet et al., 2006; Chimot et al., 2016; Kwon et al., 2017; Hewson et al., 2015).

10 
$$\Omega_{\nu} = \frac{\Omega_s}{AMF}$$
(7)

15

5

of the target species (Eq. 8), which represent radiative sensitivity to the optical depth of the absorber and a partial column density profile normalized by total vertical column density, respectively, at each layer l (l = 1, 2, ..., n) (Palmer et al., 2001). Scattering weights are dependent on the solar zenith angle  $(\theta_s)$ , viewing zenith angle  $(\theta_v)$ , relative azimuth angle  $(\theta_r)$ , surface albedo ( $\alpha_s$ ), cloud top pressure ( $p_{cld}$ ), and effective cloud fraction ( $f_c$ ).

An AMF can be decoupled with a scattering weight  $(w_l)$  and a vertical shape factor  $(S_l)$ 

#### $AMF = \sum_{l=1}^{n} w_{l}(lat, lon, month, \theta_{s}, \theta_{v}, \theta_{r}, \alpha_{s}, p_{cld}, f_{c})S_{l}(lat, lon, month)$ 20 (8)

Surface albedo, effective cloud fraction, and cloud top pressure are retrieved from GEMS and used in the AMF calculations. GEMS Level 2 surface properties include Lambertian 25 equivalent reflectivity (LER) and the daily bidirectional reflectance distribution function (BRDF) (Lee and Yoo, 2018). GEMS LER products are retrieved as composites of minimum LER values for 15 days every hour with fixed viewing geometry so that geometry dependent LER are yielded. The effective cloud fraction and cloud top pressure (effective cloud pressure) are retrieved from GEMS with the assumption of a Lambertian cloud surface (cloud surface

30 albedo = 0.8) (Veefkind et al., 2016). GEMS surface reflectivity products are also used for <u>cloud retrievals</u>. In addition, the radiative cloud fraction  $(f_{rc})$  will be provided from GEMS

Deleted: mainly

Deleted: over polluted areas because AMFs can be influenced Deleted: ( Deleted: ) (

Deleted: can be

Moved (insertion) [2] Deleted: ).

9

<u>Level 2 cloud products</u>, and is defined by Eq. 9, where  $I_{cld}$  and  $I_{clr}$  are radiances over cloud and cloud-free surfaces, respectively.

$$f_{rc} = \frac{f_c I_{cld}}{(1 - f_c) I_{clr} + f_c I_{cld}}$$
(9)

5

10

In order to consider the presence of clouds in the AMF calculations, scattering weights in the partial cloudy scenes are linearly interpolated as a function of radiative cloud fractions with scattering weights for clear sky ( $w_{l,nc}$ ) and fully covered cloudy sky ( $w_{l,wc}$ ) (Eq. 10) (Martin et al., 2002; González Abad et al., 2015). The latter ( $w_{l,wc}$ ) is calculated as a function of cloud top pressures using the assumption of Lambertian cloud surface, with a cloud surface albedo of 0.8.

Deleted: We also	
Deleted: :	
Deleted: w <sub>l,yc</sub> )	
Deleted: w <sub>l,yc</sub>	
Deleted: in	
Deleted: (see Table 1)	

Deleted: are

20

For AMF calculations, we compile an AMF look-up table (LUT) at 340 nm as a function of the variables described in Eqs. 8–10 and Table 1. González Abad et al. (2015) showed that the wavelength dependence of scattering weights in the HCHO fitting window is small. Therefore, we ignore the wavelength dependence and use AMF values for one wavelength. The AMF LUT is calculated using VLIDORT v2.6 (Spurr, 2006) with a priori data including temperature, pressure, and gas profiles (O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, and HCHO), which were simulated from a 3-D chemical transport model (GEOS-Chem v9-01-02; Bey et al., 2001) driven by Modern-Era Retrospective Analysis for Research and Applications (MERRA) with 47 vertical levels and a  $2^{\circ} \times 2.5^{\circ}$  horizontal resolution, for 2014.

25

30

However, the horizontal resolution of  $2^{\circ} \times 2.5^{\circ}$  for HCHO profiles in AMF LUT is much coarser than the GEMS horizontal resolution of  $7 \times 8 \text{ km}^2$  to discern spatial variations by local source emissions. HCHO profiles in AMF LUT are monthly averaged so that hourly variations are not taken into account. In order to resolve these coarser conditions, we can use HCHO profiles with a finer resolution as a function of time. For example, Kwon et al. (2017) showed that HCHO retrievals using monthly mean hourly AMF values were in better agreement with

the model simulations in observation system simulation experiments (OSSE) than those using

monthly mean AMF values. Additionally, air quality forecasting data can be used to consider hourly varying HCHO profiles. Further studies are required to examine the dependency of AMF calculations on spatial resolutions and temporal variations of HCHO profiles and its effect on GEMS retrieval.

5

10

Figure 4 shows examples of scattering weights and vertical profile shapes from the AMF LUT in June with conditions of a solar zenith angle of  $30^{\circ}$ , a viewing zenith angle of  $0^{\circ}$ , a relative azimuth angle of  $90^{\circ}$ , a surface albedo of 0.1, a cloud top pressure of 800 hPa, and an effective cloud fraction of 0.3 over clean and polluted grids. Clean grids are classified as having a HCHO column density less than  $3.0 \times 10^{15}$  molecules cm<sup>-2</sup> and a surface pressure higher than 990 hPa, and polluted grids have a HCHO column higher than  $1.0 \times 10^{16}$  molecules cm<sup>-2</sup> and a surface pressure higher than 990 hPa. In Fig. 4, although scattering weights are not significantly changed, the normalized vertical profile (a vertical shape factor) over a polluted area is larger near the surface compared to a clean area, which results in AMF values of 1.55 and 1.28 over the clean and polluted areas, respectively.

#### 15 2.2.5 Post processes

Post processes include systematic bias corrections and a statistic data quality flag calculation for each pixel. Systematic bias corrections include cross-track bias correction and background HCHO column correction. Cross-track biases <u>can appear in two-dimensional CCD sensors such as GEMS</u> as functions of each cross-track position when a solar irradiance is used
as a reference spectrum <u>(Chan Miller et al., 2014; Nowlan et al., 2016)</u>. The cross-track biases for OMI are constant at different latitudes; therefore, the biases <u>are shown as stripes in the along-track direction</u>. The cross-track biases are estimated by a polynomial fit through medians of HCHO slant columns for each cross-track position in clean sectors, which are the <u>casternmost</u> swaths for GEMS. The biases are removed from all data measured on the same day for each cross-track position.

An alternative method, to avoid the above-mentioned biases in the fitting procedure is to use measured radiances over a clean background region (referred to as radiance references) as the reference spectrum in radiance fitting. As measured radiance includes instrument noise and

30 attenuation by interfering gases in the background atmosphere, the interfering effects can be minimized in radiance fitting, which results in negligible cross-track biases. Deleted:

Deleted: appear

Deleted: and

Deleted: show

Deleted: Pacific Ocean for OMI and

Deleted: that can be used

Deleted: In this case, the

150°E) as GEMS reference sectors, shown as shaded areas in Fig. 1. The GEMS reference sectors include part of islands near the equator and Japan but are relatively clean areas in south/north direction over the GEMS domain. In comparison with background HCHO vertical
columns over the Pacific Ocean for OMI (Fig. S1), the annual mean of GEMS background columns over 4°S-45°N is 3.3 × 10<sup>15</sup> molecules cm<sup>-2</sup> slightly higher than that of OMI background columns (3.2 × 10<sup>15</sup> molecules cm<sup>-2</sup>), showing that we can use the easternmost regions as background in the GEMS domain. Occasionally, local differences between GEMS and OMI background columns can be as large as 3.8 × 10<sup>15</sup> molecules cm<sup>-2</sup> in the tropical

For GEMS, we plan to use simulated HCHO columns over the easternmost regions (143-

10 region of the southern hemisphere due to biogenic activity and biomass burning, but the standard deviation of background values in that region is  $5.1 \times 10^{14}$  molecules cm<sup>-2</sup>, which is even lower than that of  $1.2 \times 10^{15}$  molecules cm<sup>-2</sup> in the middle latitude (>30°N), indicating that the influences from biogenic activity and biomass burning can be corrected by model simulations.

15

The retrieved slant columns using a radiance reference are differential slant columns  $(\Delta SCD = SCD - SCD_0)$  and do not include background HCHO columns  $(SCD_0)$  that are mainly from the oxidation of methane. To account for the background columns, we use HCHO vertical columns simulated in 2014 from a chemical transport model, GEOS-Chem (Bey et al., 2001) with a spatial resolution of  $2^\circ \times 2.5^\circ$ . Simulated HCHO vertical columns are averaged

20 <u>zonally and monthly</u> over the reference sectors and are interpolated to 720 latitudinal grid points with a resolution of 0.25° from 90°S to 90°N.

In order to account for dependency of measured radiances on geometric angles, we then convert simulated background vertical columns into slant columns by applying AMF values over the reference sector ( $AMF_0$ ), which are calculated with cloud information and geometric angles on the <u>reference sectors</u>. Corrected <u>GEMS</u> HCHO slant columns are formulated as the sum of the retrieved differential slant columns and the simulated background slant columns as shown in Eq. 11,

$$\Omega_{s}(i,j) = SCD_{corr}(i,j) = \Delta SCD(i,j) + AMF_{0}(lat)VCD_{m}(lat),$$

Deleted: related to

Deleted: )

(11)

-(	Deleted: For OMI products, simulated				
(	Deleted: zonally				
Deleted: clean background regions for					
<ul> <li>Deleted: sector (140-160°W, 90°S-90°N), except for Hawaii (1: 160°W, 19-22°N),</li> </ul>					
Deleted: ,					
<b>Deleted:</b> . For GEMS, we plan to use simulated HCHO co over easternmost regions as GEMS reference sectors					
Y	Deleted: To derive				
Y(	Deleted: from the model,				
(	<b>Deleted:</b> same orbit. Pixels for $AMF_0$ are selected with cloud fractions of less than 0.4, data quality flags of 0 (good pixels), and retrieved slant columns of $\pm 1.0 \times 10^{16}$ molecules cm <sup>-2</sup> .				

30

25

where i and j indicate pixel indices of cross and along tracks, respectively, and VCD<sub>m</sub> denotes a background vertical column density from the model. We finally apply AMF values from the LUT to the corrected slant columns to obtain GEMS HCHO vertical column densities.

After the correction of systematic biases and conversion to vertical column densities with 5 AMFs, all pixels are flagged with vertical columns and fitting uncertainties.(González Abad et al., 2015). We assign a data quality flag of 0 for good pixels, where retrieved vertical columns plus two-times fitting uncertainties are positive. The pixels in which retrieved vertical columns are negative within two-times fitting uncertainties, but positive within three-times fitting uncertainties, are assigned with a data quality flag of 1, which represents suspected quality pixels. Pixels with negative vertical columns within three-times fitting uncertainties are designated as bad quality pixels and given a data quality flag of 2, and missing values are flagged by -1. It is of note that these conditions are generous and that tighter conditions for good data may be required for analysis.

10

#### **3** Uncertainty analysis

15 We use error and uncertainty terminology from the Guide to the Expression of Uncertainties in Measurements (GUM) (https://www.bipm.org/utils/common/documents/jcgm /JCGM 100 2008 E.pdf). Following GUM, "error" means the difference between a measurement result and a true value, and "uncertainty" means the dispersion of measurement values, such as a standard deviation and a full width at the half maximum. Because there is a 20 lack of true values for HCHO vertical columns, we consider that the word "uncertainty" is

more appropriate for use in our analysis. Uncertainties in the retrieval steps mentioned in Section 2.2 are assumed to be uncorrelated, because the steps are independently performed. Total uncertainty in HCHO vertical column density (VCD) using a radiance reference can be formulated as follows

25 (Boersma et al., 2004; De Smedt et al., 2008),

transformed using Eqs. 7 and 11 into Eq. 13,

$$\sigma_{v}^{2} = \left(\frac{\partial\Omega_{v}}{\partial\Delta SCD}\sigma_{s}\right)^{2} + \left(\frac{\partial\Omega_{v}}{\partialAMF}\sigma_{AMF}\right)^{2} + \left(\frac{\partial\Omega_{v}}{\partial VCD_{m}}\sigma_{m}\right)^{2} + \left(\frac{\partial\Omega_{v}}{\partialAMF_{0}}\sigma_{AMF_{0}}\right)^{2}, \tag{12}$$

where  $\sigma$  is the uncertainty in each part,  $\Omega_v$  is the vertical column density, and subscripts v,

s, and m represent vertical, slant, and model, respectively. The total uncertainty equation is

30

Deleted: However, we may also need to correct latitudinal biases resulting from  $O_3$  interference dependent on latitudes. Previous studies conducted latitudinal bias corrections when using a radiance reference as the reference spectrum (González Abad et al., 2015; De Smedt et al., 2018). Latitudinal biases result from stratospheric O<sub>3</sub> distributions dependent on latitude; however, they can be corrected by subtracting slant columns retrieved for a radiance reference from the slant columns analyzed. Figure 5 shows OMI HCHO slant columns from OMHCHO products

(Fig. 5a) and the GEMS algorithm without and with latitudinal bias corrections (Fig. 5b and 5c).

Moved down [3]: HCHO slant columns without latitudinal bias corrections (Fig. 5b)

Deleted: 5b) are retrieved larger in 5°N-25°N than OMHCHO products, but HCHO slant columns with the bias corrections are better agreement with OMHCHO products. Figure 5d shows the ns with the bias corrections are in absolute differences between OMI HCHO slant columns with and without latitudinal bias corrections from the GEMS algorithm. Slant as with bias corrections increase at latitudes lower than 5°N colu and higher than 25°N but decrease at latitudes from 5°N-25°N. However, GEMS has south to north swaths, while sun-synchronous satellites have west to east swaths; therefore, latitudinal biases can be minimized when using a radiance reference for GEMS. However, the latitudinal biases need to be further investigated after GEMS is launched. Deleted:

Deleted: where

	Deleted: 4
(	Deleted: As
(	Deleted: exact

$$\sigma_{\nu}^{2} = \frac{1}{AMF^{2}} \left[ \sigma_{s}^{2} + \frac{(\Delta SCD + AMF_{0}VCD_{m})^{2}}{AMF^{2}} \sigma_{AMF}^{2} + AMF_{0}^{2} \sigma_{m}^{2} + VCD_{m}^{2} \sigma_{AMF_{0}}^{2} \right].$$
(13)

We analyze expected uncertainties for the GEMS algorithm by using simulated radiances
from Kwon et al. (2017) and OMI Level 1B data. In order to estimate the expected random uncertainty for GEMS (Section 3.1.1), we use simulated radiances which are convoluted with GEMS bandpass functions at 330 nm as a function of cross-track positions in the south to north direction. Simulated radiances include noises based on the expected signal-to-noise ratio for co-added pixels with spatial resolutions of 7 × 8 km<sup>2</sup>. We use absorption cross-sections of Ring
effect, O<sub>3</sub>, NO<sub>2</sub>, HCHO, and additionally SO<sub>2</sub> (Hermans et al., 2009; Vandaele et al., 2009) in radiance fitting because O<sub>3</sub>, NO<sub>2</sub>, and HCHO, and SO<sub>2</sub> were considered in the radiance calculation (Kwon et al. 2017).

For other uncertainty analyses, we use OMI Level 1B data with OMI slit function data (Dirksen et al., 2006) in order to examine algorithm sensitivities to different parameters. Fitting

15 options such as absorption cross-section data and the fitting window are summarized in Table 1. It will be necessary to conduct an additional uncertainty analysis for GEMS HCHO retrievals after GEMS is launched.

#### 3.1 Uncertainties in slant columns

#### 3.1.1 Random uncertainty

20

Uncertainties in slant columns result from random uncertainties ( $\sigma_{rand}$ ) and systematic uncertainties ( $\sigma_{sys}$ ) (Eq. 14) (De Smedt et al., 2018),

$$\sigma_s^2 = \sigma_{rand}^2 + \sigma_{sys}^2. \tag{14}$$

25 Random uncertainties are fitting uncertainties when yielding slant columns, and they mainly result from instrument <u>noises</u>. We can reduce random uncertainties by using measured radiances over clean areas as reference spectra instead of irradiances, as the use of measured radiances can minimize instrument <u>noises</u> and interference of O<sub>3</sub> and BrO in the stratosphere. In addition, averaging the resulting slant columns for individual pixels can reduce random

14

(	Deleted: this
(	Deleted: retrieving HCHO VCDs with
(	Deleted: level
(	Deleted: For the
~(	Deleted: analysis, we use

Deleted: , and use OMI slit function data (Dirksen et al., 2006). However, when GEMS is launched, it Deleted: another

Deleted: noise

Deleted: noise

uncertainties, but this is achieved at the expense of a loss of temporal and spatial resolution (De Smedt et al., 2018).

Random uncertainties can be calculated from root mean square (RMS) values of fitting residuals, degrees of freedom (m - n), and diagonal <u>components</u> of a covariance matrix  $(C_{j,j})$  for fitting parameters,

$$\sigma_{s,j}^2 = RMS^2 \frac{m}{m-n} C_{j,j} C_{j,j},\tag{15}$$

where m and n are the number of spectral grids and fitting parameters, respectively, and j denotes specific species in fitting parameters.

Random uncertainties from the GEMS algorithm are estimated using simulated radiances. RMS of fitting residuals and random uncertainty for the GEMS domain range from  $2.9 \times 10^{-4}$  to  $2.1 \times 10^{-3}$  and  $2.1 \times 10^{15}$  to  $1.6 \times 10^{16}$  molecules cm<sup>-2</sup>, respectively, which are comparable with those (RMS:  $4 \times 10^{-4}$  to  $2.0 \times 10^{-3}$ ; random uncertainty:  $3.3 \times 10^{15}$  to  $1.8 \times 10^{16}$  molecules cm<sup>-2</sup>) obtained from the GEMS algorithm using OMI Level 1B data. GEMS measures target

- 15 cm<sup>-2</sup>) obtained from the GEMS algorithm using OMI Level 1B data. GEMS measures target species every hour in daytime, and changes of solar location during the day can affect the accuracy of radiance fitting. An averaged fitting RMS value and a random uncertainty are 6.9 × 10<sup>-4</sup> and 5.0 × 10<sup>15</sup> molecules cm<sup>-2</sup> for conditions with both solar and viewing zenith angles less than 70, which happen at 8:00–18:00 and 9:00–16:00 local time of Seoul in summer and
- 20 winter, respectively. However, the fitting RMS value and the random uncertainty increase to  $1.1 \times 10^{-3}$  and  $8.2 \times 10^{15}$  molecules cm<sup>-2</sup>, respectively, when solar and viewing zenith angles are higher than 70.

#### 3.1.2 Systematic uncertainty

5

10

Systematic uncertainties result from uncertainties of wavelength calibration, the bandpass
function for convolution, and absorption cross-sections. We estimate systematic uncertainties
from sensitivity tests to parameters, using OMI Level 1B data. First, systematic uncertainties
associated with absorption cross-sections are estimated using alternative absorption crosssections: we compare resulting slant columns to the baseline <u>calculation</u> with conditions in
Table 1 for a one-month period (March 2005). In the analysis, we define an uncertainty as a
standard deviation of differences between the sensitivity and baseline <u>calculations</u>. Absorption

Deleted: component	
RMS <sup>2</sup>	
Deleted: $\frac{mn}{m-n}$	

**Deleted:** To examine random uncertainties from the GEMS algorithm for HCHO, we select pixels with the following conditions: a main data quality flag of 0, an effective cloud fraction of less than 0.3, and a solar zenith angle of less than 60° in the GEMS field of view (5°S-45°N, 75°-145°E) for four months (March, June, September, and December) in 2005. Random uncertainties vary between  $3.3 \times 10^{15}$  and  $1.8 \times 10^{16}$  molecules cm<sup>2</sup>, corresponding to pixels with fitting RMS from  $4.4 \times 10^4$  to  $2.0 \times 10^3$ , and accounting for 97% of all pixels. In comparison with OMHCHO retrievals, González Abad et al. (2015) showed fitting uncertainties ranging from  $6 \times 10^{15}$  molecules cm<sup>2</sup> to 100% or larger of slant columns and RMS values from  $0.4 \times 10^3$  to  $2.0 \times 10^3$ , which are similar to those from the GEMS algorithm.<sup>4</sup>

Deleted:	
Deleted: run	
Deleted: run	
Deleted: run	

cross sections are convoluted and interpolated using the same spectral resolution and wavelength sampling to enable comparisons between datasets.

To test the retrieval sensitivity to HCHO absorption cross-sections, we use HCHO absorption cross-section datasets from Cantrell et al. (1990) instead of those of Chance and  $O_{1}$  is the retrieval to the section of the section o

Deleted: )

Deleted: run

Deleted: run

Deleted: run

Deleted: run

Orphal (2011), which provide a rescaling of the datasets in Cantrell et al. (1990) to those of Meller and Moortgat (2000). Absorption cross-sections of Cantrell et al. (1990) are ~10% lower than those of Chance and Orphal (2011), and the differences are directly linked to slant column retrieval (Pinardi et al., 2013). Therefore, slant columns using Cantrell et al. (1990) are by a factor of 1.1 higher than those of the baseline <u>calculation</u>. The slant column changes are similar to values from previous studies (Pinardi et al., 2013; De Smedt et al., 2018).

10

5

We conduct a sensitivity test to O<sub>3</sub> absorption cross-sections at 223 K and 293 K from Chehade et al. (2013). Uncertainties of these datasets at both temperatures are ~4% in the fitting window of the GEMS algorithm. Compared to the baseline <u>calculation</u>, use of the O<sub>3</sub> datasets of Chehade et al. (2013) at 223 K and 293 K changes the slant columns by ~20% and ~8% on average, respectively, which provides uncertainties in slant columns of  $1.4 \times 10^{15}$  and  $0.57 \times 10^{15}$  molecules cm<sup>-2</sup>. These uncertainties are larger than those of 13% and 5% from Pinardi et al. (2013) and De Smedt et al. (2018), respectively. It thus appears that the GEMS HCHO retrieval algorithm is the most sensitive to O<sub>3</sub> absorption, especially at low temperatures in the stratosphere, due to strong absorption in the ultraviolet.

20

15

A sensitivity test is conducted against the BrO dataset of Fleischmann et al. (2004), which is by ~9% lower than the baseline BrO dataset in the GEMS HCHO fitting window, and results in ~4% slant column changes compared to the baseline <u>calculation</u> (with an uncertainty of 0.28 × 10<sup>15</sup> molecules cm<sup>-2</sup>).

We then examine slant column uncertainties for O<sub>4</sub> and NO<sub>2</sub> absorption cross-sections.
We use alternative O<sub>4</sub> absorption cross-sections from <u>http://spectrolab.aeronomie.be/o2.htm</u> that have differences of 28% compared to the dataset used in the baseline <u>calculation</u>. Due to the large uncertainties of the data compared to other absorption cross-section data, the resulting slant column changes are significant, ~24%, with an uncertainty of 1.6 × 10<sup>15</sup> molecules cm<sup>-2</sup>. This uncertainty could thus be decreased by reducing the uncertainties of the O<sub>4</sub> datasets. Also, O<sub>4</sub> should be included in the large fitting interval for HCHO because it has strong peaks near

344.0 and 361 nm (De Smedt et al., 2015; Thalman and Volkamer, 2013). The NO<sub>2</sub> datasets

Deleted: run

Deleted: run

Deleted:

from Burrows et al. (1998) are 2% larger than those in the baseline <u>calculation</u>; switching to them leads to ~5% slant column changes with uncertainty of  $0.37 \times 10^{15}$  molecules cm<sup>-2</sup>.

We also estimate the systematic uncertainties of slant columns for wavelength calibration and solar  $I_0$  effects by using an alternative solar irradiance reference. As described in Section 2.2.1 and 2.2.2, a solar irradiance reference spectrum was used in the wavelength calibration

5 2.2.1 and 2.2.2, a solar irradiance reference spectrum was used in the wavelength calibration and calculation of pseudo absorption cross-sections related with solar I<sub>0</sub> effect. An alternative reference spectrum from Kitt Peak National Observatory (Kurucz et al., 1984) is almost identical to that of the baseline <u>calculation</u>, but the resulting slant column changes are up to ~14% with uncertainty of 0.92 × 10<sup>15</sup> molecules cm<sup>-2</sup>. We thus consider that the solar I<sub>0</sub> effect
 10 associated with the strongest interfering gas (O<sub>3</sub>) is very sensitive to the reference spectrum.

The total systematic uncertainty of slant columns for the parameters discussed is 38% of the slant column densities on average for one month. This uncertainty is larger than that of De Smedt et al. (2018), which is 20% of the slant column densities. However, there are remaining slant column uncertainties resulting from uncertainties relating to other parameters, such as polynomial orders in Eq. 6, instrument bandpass functions, and temperature dependency of cross-sections. Therefore, we estimate the systematic uncertainty as being 38% of the slant

#### 3.2 Uncertainty in AMF

The AMF uncertainty can be estimated by each parameter in Eq. 16. We examine AMF
uncertainties for surface albedo (α<sub>s</sub>), cloud top pressure (p<sub>c</sub>), and effective cloud fraction (f<sub>c</sub>) with a solar zenith angle of 30°, a viewing zenith angle of 30°, and a relative azimuth angle of 0°. In addition, we define a profile height parameter (p<sub>h</sub>) as an altitude below which 75% of HCHO VCDs exist from the surface to estimate AMF uncertainty with respect to a HCHO profile shape. The uncertainties of parameters (σ<sub>asy</sub> = 0.02, σ<sub>pcy</sub> = 50 hPa, and σ<sub>fcy</sub> = 0.05)
are based on De Smedt et al. (2018) and will be replaced to those from GEMS Level 2 products. The uncertainty of profile height (σ<sub>ph</sub>) is defined as a standard deviation of profile heights in AMF LUT, and σ<sub>ph</sub> in polluted and clean areas is 84 and 55 hPa, respectively.

columns, prior to conducting uncertainty analyses on other parameters.

$$\sigma_{AMF}^2 = \left(\frac{\partial AMF}{\partial \alpha_s}\sigma_{\alpha_s}\right)^2 + \left(\frac{\partial AMF}{\partial p_c}\sigma_{p_c}\right)^2 + \left(\frac{\partial AMF}{\partial f_c}\sigma_{f_c}\right)^2 + \left(\frac{\partial AMF}{\partial p_h}\sigma_{p_h}\right)^2 \tag{16}$$

-(	Deleted: Derivatives
-(	Deleted: AMF to parameters (slope) are calculated
(	Deleted: AMF LUT, while the
(	Deleted: ,
~(	Deleted: ,
Y	Deleted: )
4	Deleted: ),

#### 30

15

Figure 5a shows AMF sensitivities to surface albedos with clear sky conditions ( $f_c = 0$ ). AMF values increase linearly with an increase in surface <u>albedos</u> and are slightly higher for <u>clean regions</u> than <u>polluted regions</u> in which HCHO is <u>concentrated</u> near the surface.

- Clouds below or within HCHO layers increase photon path lengths due to multiple 5 scattering, while clouds at altitudes above HCHO layers shield photons from reaching the 5 surface. Therefore, the AMF values decrease with a decrease in cloud top pressures (with an increase in cloud heights) due to fewer photons reaching to the surface (Fig. 5b). AMFs rapidly change with increasing cloud top pressures near the surface, implying a high AMF sensitivity to the changes in photon path lengths with respect to multiple scattering.
- 10 Cloud fractions also play an important role in determining AMF values and their effects are shown in Fig. <u>5c</u>. For a cloud with its top pressure at 800 hPa, AMF values increase with an increase in the cloud fraction, which implies that more photons arrive at the near surface <u>due to</u> multiple scattering by clouds with increasing cloud fractions. However, for clouds with its top pressure at high altitudes (500 hPa), AMF values decrease with an increase in cloud fractions due to the shielding effect of clouds.
  - Figure 5d shows increasing AMF values with an increase in the profile height, resulting from increased HCHO absorptions at high altitudes. The AMF sensitivity to profile heights in clean areas is higher than that in polluted areas because HCHO distributions are more uniform in clean areas than polluted areas.
- 20 Table 2 summarizes estimated retrieval uncertainties of GEMS HCHO VCDs due to AMF uncertainties as functions of surface albedos, cloud top pressures, and cloud fractions. Values are calculated assuming conditions with solar zenith angle of 30°, viewing zenith angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile height of 700 hPa. Uncertainties of HCHO VCDs due to AMF uncertainties can be as large as 20% and 24% of
- 25 HCHO VCDs in clean and polluted areas, respectively. Maximum values occur for conditions with low surface albedo, clouds at high altitudes, and high cloud fractions, but they do not differ significantly between clean and polluted areas. However, AMF driven HCHO uncertainty with respect to the profile height in polluted areas is higher than that in clean areas, implying that accurate HCHO profile information in polluted areas is important for the GEMS
- 30 <u>HCHO retrieval. We can minimize the a priori HCHO profile uncertainties by using averaging kernels.</u>

Aerosol vertical distributions and aerosol optical properties are also important sources of AMF uncertainty (Chimot et al., 2016; Kwon et al., 2017; Hewson et al., 2013). Non-absorbing

Deleted: 6a
Deleted: from AMF LUT
Deleted: albedo,
Deleted: sensitivities of AMF to surface albedo
Deleted: low surface albedos of less
Deleted: 0.2. The
Deleted: mixing ratio
Deleted: higher
<b>Deleted:</b> in polluted areas compared to clean areas, which relates to lower AMF values in polluted areas with low surface albedos.
Deleted: high
Deleted: near
Deleted: near Deleted: 6b
Deleted: near Deleted: 6b Deleted: are more sensitive to
Deleted: near Deleted: 6b Deleted: are more sensitive to Deleted: pressure
Deleted: near Deleted: 6b Deleted: are more sensitive to Deleted: pressure Deleted: ; this implies
Deleted: near Deleted: 6b Deleted: are more sensitive to Deleted: pressure Deleted: ; this implies Deleted: is more sensitive
Deleted: near Deleted: 6b Deleted: are more sensitive to Deleted: pressure Deleted: ; this implies Deleted: is more sensitive Deleted: than blocking by clouds near the surface
Deleted: near Deleted: near Deleted: 6b Deleted: are more sensitive to Deleted: pressure Deleted: ; this implies Deleted: is more sensitive Deleted: than blocking by clouds near the surface Deleted: roles
Deleted: near Deleted: near Deleted: 6b Deleted: are more sensitive to Deleted: pressure Deleted: ; this implies Deleted: is more sensitive Deleted: than blocking by clouds near the surface Deleted: roles Deleted: 6c
Deleted: near Deleted: near Deleted: 6b Deleted: are more sensitive to Deleted: pressure Deleted: ; this implies Deleted: is more sensitive Deleted: than blocking by clouds near the surface Deleted: roles Deleted: foc Deleted: clouds
Deleted: near         Deleted: 6b         Deleted: are more sensitive to         Deleted: pressure         Deleted: is implies         Deleted: is more sensitive         Deleted: than blocking by clouds near the surface         Deleted: roles         Deleted: clouds         Deleted: the

**Deleted:** Figure 7 shows AMF uncertainty as functions of each parameter. AMF uncertainties with respect to surface albedo are estimated using a surface albedo ( $\sigma_{\alpha_s}$ ) uncertainty of 0.02 (Kleipool et al., 2008) and are uniform compared to those with respect to cloud properties. In cloud free conditions ( $f_c = 0$  in Fig. 7c), uncertainties of surface albedo almost account for total AMF uncertainty. For cloudy conditions, we use the uncertainties of cloud top pressure ( $\sigma_{p_c} = 50$  hPa) and cloud fraction ( $\sigma_{f_c} = 0.05$ 

#### (Moved up [2]: ) (Veefkind et al., 2016).

Deleted: In conditions where the surface albedo is less than 0.6 and the cloud top pressure is higher than 550 hPa, AMF uncertainties for cloud top pressures account for most of the total AMF uncertainties (Fig. 7a and 7b). In the opposite conditions, the uncertainties (Fig. 7a and 7b). In the opposite conditions, the uncertainties. In addition, AMF uncertainties for cloud top pressures account for almost all the AMF uncertainties result in 4% to 16% of HCHO vertical column uncertainties from Eqs. 13 and 16.¶ HCHO profile shapes, aerosol vertical distributions,

**Deleted:** AMF uncertainties from HCHO profiles are estimated to range from 0.27–0.44; they are thus the largest uncertainties of all other parameters in Eq. 16. The AMF uncertainty in HCHO profiles leads to a 15%–63% uncertainty in HCHO vertical columns. However, we can minimize the a priori HCHO profile uncertainties by using averaging kernels.<sup>¶</sup>

aerosols <u>play a similar role as that of clouds in radiative transfer and are implicitly considered</u> when cloud information is used. However, absorbing aerosols, such as mineral dust and black carbon, counteract the effects of non-absorbing aerosols and clouds. In the GEMS domain, dust storms and biomass burning occur seasonally, and we may therefore need to consider <u>the effect</u> of absorbing aerosols on the retrieval. We plan to update our AMF LUT as a function of aerosol optical depth, single scattering albedo, and aerosol height – all of which will be retrieved by <u>GEMS – to account for the effect of absorbing aerosols</u>. On-line AMF calculation can also be used for aerosol correction with cloud information and model simulation (Lin et al., 2014).

#### 3.3 Uncertainty in background correction

10

5

The uncertainties of model results for background concentrations ( $\sigma_m$ ) and AMFs over reference sectors ( $\sigma_{AMF_0}$ ) are important errors for background corrections ( $\sigma_{bg}$ ) in Eq. 17, which represents the 3<sup>rd</sup> and 4<sup>th</sup> terms on the right-hand side of Eq. 13,

$$\sigma_{bg}^2 = \frac{1}{AMF_0^2} \left[ AMF_0^2 \sigma_m^2 + VCD_m^2 \sigma_{AMF_0}^2 \right].$$
(17)

15

25

We estimate model uncertainties by <u>using standard deviations of model results as a</u> <u>function of latitude. Standard deviations range</u> from  $2.3 \times 10^{14}$  to  $1.4 \times 10^{15}$  molecules cm<sup>-2</sup>. Uncertainties related to AMF in the reference sectors are identical to those discussed in Section 3.2.

#### 20 4 Results and validation

In this section, we use OMI Level 1B data to validate our retrieval algorithm, Resulting HCHO products are compared with OMI products of other institutes, for one month of each season (March, June, September, and December) in 2005 to provide seasonal variation in the GEMS domain. We also compared OMI products with ground-based MAX-DOAS at two sites in Europe for 2005 and one site in China for 2016.

### **4.1 Retrieval of OMI HCHO**

GEMS fitting options described in Table 1 are largely consistent with those of OMHCHO products (González Abad et al., 2015). However, we do not include spectral undersampling

# Deleted: have Deleted: roles to

between aerosols and clouds

Deleted:	,
Deleted:	non-absorbing aerosols
Dolotodu	

Deleted: contribute to uncertainties in

#### Deleted: the differences in

**Deleted:** between the GEMS algorithm and the OMHCHO products, and find that model HCHO vertical columns used in the GEMS algorithm are lower than those used in OMHCHO, with absolute differences ranging...

## Deleted: -1

**Deleted:** to  $6.1 \times 10^{14}$ . In Section 4, we discuss the impacts of these differences on retrieved HCHO columns

Deleted: Validation

Deleted: is validated by a comparison

**Deleted:** . First, we yield HCHO column densities using the GEMS HCHO algorithm with OMI Level 1B data in the fitting options described in Table 1, and also include spectral undersampling in the fitting process (Chance et al., 2005). We conduct our calculations

(Chance et al., 2005) in the fitting process for GEMS, and reference sectors for a radiance reference are 143-150°E (shaded areas in Fig. 1). For OMI products, spectral undersampling needs to be included, and radiance references are from the Pacific Ocean as described in González Abad et al. (2015). We use simulated HCHO vertical columns for the background

5 correction, which are zonally and monthly averaged over the reference sector (140-160°W, 90°S-90°N) except for Hawaii (154-160°W, 19-22°N).

In addition, we need to correct latitudinal biases for OMI. Previous studies explained that the latitudinal biases result from spectral interferences of BrO and O<sub>3</sub>, whose concentrations are a function of latitude and are high in high latitudes (De Smedt et al., 2008; De Smedt et al.,

- 10 2015; González Abad et al., 2015). Therefore, the latitudinal biases were corrected when a radiance reference was used as the reference spectrum (De Smedt et al., 2008; González Abad et al., 2015; De Smedt et al., 2018). We correct the latitudinal biases, which are slant columns retrieved for a radiance reference and are averaged as a function of latitude, by subtracting the biases from the corrected slant columns in Eq. 11.
- 15 Figure 6 shows OMI HCHO slant columns from OMHCHO products (Fig. 6a) and the GEMS algorithm without and with latitudinal bias corrections (Fig. 6b and 6c), HCHO slant columns without latitudinal bias corrections (Fig. 6b) are retrieved larger in 5°N-25°N than OMHCHO products, but HCHO slant columns with the bias corrections are in better agreement with OMHCHO products. Figure 6d shows the absolute differences between OMI HCHO slant
- 20 columns with and without latitudinal bias corrections from the GEMS algorithm as latitudinal biases. Slant columns with bias corrections increase at latitudes lower than 5°N and higher than 25°N but decrease at latitudes from 5°N-25°N.

<u>However</u>, latitudinal biases can be minimized when using a radiance reference as a function of each cross-track position in the south to north direction for GEMS. In default fitting

25 options, therefore, we do not include latitudinal correction and do not analyze uncertainty of latitudinal corrections in Section 3. However, a further investigation for the latitudinal biases needs to be required after GEMS is launched.

Figure 7 shows an example of retrieved HCHO optical depths and fitting residuals as functions of wavelengths for a pixel in Indonesia (March 23 2005; orbit 3655). The retrieved HCHO slant column is  $3.2 \times 10^{16}$  molecules cm<sup>-2</sup>, which is relatively high due to biomass burning in that region. Averaged slant column and random uncertainty for all pixels on the orbit

30

(Moved (insertion) [3]

Deleted: Figure 8

Deleted: Average

are  $7.6 \times 10^{15}$  and  $6.9 \times 10^{15}$  molecules cm<sup>-2</sup>, respectively, over the GEMS domain. The large

random uncertainty of 100% or larger results from pixels with low concentrations, where averaged slant columns and random uncertainties are  $2.2 \times 10^{15}$  and  $6.2 \times 10^{15}$  molecules cm<sup>-2</sup>

### **4.2 Comparison with other OMI products**

5

10

25

Figure <u>8</u> compares monthly mean slant columns retrieved using the GEMS algorithm and those of OMHCHO products (González Abad et al., 2015). We select pixels with (1) vertical columns ranging from  $-5.0 \times 10^{15}$  to  $10 \times 10^{16}$  molecules cm<sup>-2</sup>, (2) a main data quality flag of 0, (3) an effective cloud fraction of less than 0.3, and (4) a solar zenith angle of less than 60. Monthly mean slant column densities are weighted by uncertainties and overlapped areas between pixels and grid boxes with a horizontal resolution of  $0.25^{\circ} \times 0.25^{\circ}$ .

We find similar spatial patterns of HCHO slant columns in both products; this shows that high HCHO columns occur over Indonesia and the Indochina Peninsula in March and over Indonesia in September, owing to biomass burning and biogenic activities. In summer, HCHO enhancements over China are caused by biomass burning and the oxidation of biogenic and anthropogenic VOCs due to photochemical reactions. In addition, high HCHO slant column

anthropogenic VOCs due to photochemical reactions. In addition, high HCHO slant column densities occur over the Pearl River Delta, where anthropogenic VOCs are emitted from petrochemical industries, cargo ports, paint production, and many other activities. (Zhong et al., 2013). The scatter-plot comparisons with OMHCHO products show that GEMS HCHO slant columns are in good agreement with OMHCHO products, with correlation coefficients of 0.77–
 0.91 and regression slopes of 0.94–1.04. The relative differences between GEMS slant columns

and those of OMHCHO products are -3% to 0.1% on average over the domain.

However, some discrepancies are found despite overall good agreement between GEMS and OMHCHO products, and these are mainly related to the background correction from the different model results. Figure <u>9</u> shows the simulated HCHO vertical columns used for background corrections in OMHCHO (solid lines) and the GEMS algorithm (dashed lines), respectively. <u>Similar</u> latitudinal variations are shown with a peak in the tropics and gradual decreases in high latitudes, which reflects the photochemical production of HCHO; however, the magnitudes slightly differ. The model results used in GEMS are smaller than those used in the OMHCHO products (especially within  $\pm 20^{\circ}$  latitudes) by  $-1.3 \times 10^{15}$  molecules cm<sup>-2</sup> near

21

Deleted: results from random uncertainties
Deleted: in

Deleted: 9

Deleted: (1)

Deleted:

Deleted: 92

Deleted: 10

Deleted: )

Deleted: Values show similar

**Deleted:** Latitudinal biases are also corrected using the method discussed in Section 2.2.5.

<sup>30 6°</sup>N in September. Both results were obtained from the same 3-D global chemical transport model (GEOS-Chem), but different assimilated meteorological products were employed.

HCHO vertical columns used in the OMHCHO products were from GEOS-Chem with GEOS-4 meteorological data (Millet et al., 2006), which have lower cloud optical depths near the equator compared to GEOS-3 and GEOS-5. Therefore, low cloud optical depths in the tropics resulted in faster methane oxidation and the production of more HCHO in relation to high hydroxyl radical concentrations (<u>http://wiki.seas.harvard.edu/geos-</u>

5 hydroxyl radical concentrations (<u>http://wiki.seas.harvard.edu/geos-chem/index.php/GMAO\_GEOS-4</u>). González Abad et al. (2016) also showed that the OMHCHO products are larger than OMI BIRA-IASB products near the equator, such as in Southeastern Asia, Tropical Africa, and the Amazon Basin.

We select four regions (Sumatra/Malaysia, the Indochina Peninsula, China, and Borneo),
where HCHO is abundant from biomass burning and biogenic and anthropogenic sources.
Table <u>3</u> provides the relative differences between OMI GEMS HCHO slant columns and
OMHCHO slant columns in these four regions. GEMS HCHO slant columns are 1% to 13%
lower than those of OMHCHO in Sumatra/Malaysia and Borneo, because the differences in
simulated HCHO column densities for background corrections are relatively large near the
equator compared to the mid-latitudes, as previously mentioned. In the Indochina Peninsula

and China, however, the GEMS HCHO slant columns are 6% to 25% higher than the OMHCHO slant columns, although the simulated HCHO concentrations used in the GEMS algorithm for background corrections are lower than those used in OMHCHO.

We also compare OMI GEMS HCHO slant columns with OMI QA4ECV products (OMI
 QA4ECV) (De Smedt et al., 2018). The QA4ECV project was proposed to provide reliable satellite and ground-based measurements of climate and air quality variables with detailed uncertainty information (http://www.qa4ecv.eu). Figure 10 shows that the spatial distributions of the GEMS HCHO slant columns are consistent with those of the OMI QA4ECV products, but relatively poorer correlation coefficients of 0.52 to 0.76 are found compared to those with

25 <u>the OMHCHO products. The relative differences between GEMS and QA4ECV slant columns</u> range from -11% to -22% on average over the GEMS domain.

Magnitudes of relative differences between OMI GEMS and OMI QA4ECV slant columns vary regionally and seasonally (Table 3). The differences are relatively small near the equator (Sumatra/Malaysia and Borneo) compared to those in subtropics and mid-latitudes

30 (Indochina Peninsula and China) because biomass burning often occurs and biogenic sources are more abundant near the equator. The HCHO differences near the equator are lower in spring and fall than those in summer and winter due to biomass burning. The effects of biomass burning on HCHO slant columns are also found over Indochina Peninsular in spring. As a result,

Deleted: 2

Deleted:	from BIRA-IASB
Deleted:	BIRA) in Fig. 11
Deleted:	The
Deleted:	BIRA
Deleted:	and provide

Deleted: This

Deleted: of

Deleted: BIRA

magnitudes of relative differences in regions and seasons with high HCHO concentrations are small, implying that HCHO can be well-retrieved because of the abundant HCHO concentrations, regardless of the fitting method used.

<u>The discrepancy between the two products could result from the radiance fitting. The OMI</u>
 <u>QA4ECV</u> products use the DOAS method while the GEMS algorithm <u>uses</u> a non-linearized fitting method (BOAS) for radiance fitting. <u>We also find that polynomial orders accounting for</u> Rayleigh and Mie scatterings are important factors, causing differences between the two products. Retrieved slant columns using the 4<sup>th</sup> polynomial order are in better agreement with the QA4ECV products (Fig. S2). Both correlation coefficient and regression slope are

10 improved although OMI GEMS HCHO values are higher than those of the QA4ECV. We use the 4<sup>th</sup> order polynomial instead of the 5<sup>th</sup> order used in the QA4ECV products because slant columns retrieved using the 5<sup>th</sup> order in the GEMS algorithms are much higher than the QA4ECV products.

Also, different O<sub>3</sub> absorption cross sections (Serdyuchenko et al., 2014) are used in <u>the</u>
 OMI<u>QA4ECV</u> at different temperatures (220 and 243 K), and a non-linear O<sub>3</sub> absorption effect
 (Pukīte et al., 2010) is included in the OMI QA4ECV. We examine the O<sub>3</sub> effects on retrieved slant columns in GEMS algorithm using O<sub>3</sub> datasets used in QA4ECV and considering a non-linear O<sub>3</sub> absorption effect. Correlation coefficient and regression slopes are slightly improved (Table S1), and relative differences in the four regions defined above are slightly reduced in

20 most seasons and regions (Table S2).

25

30

#### **4.3 Comparison with ground- based MAX-DOAS**

We also compare satellite results with MAX-DOAS ground observations at Haute-Provence Observatory (OHP) in France, Bremen in Germany, and Xianghe in China (Table <u>4</u>). MAX-DOAS data are <u>collected</u> within the OMI overpass time (12:00–15:00 local time) at OHP and Bremen in 2005, and at Xianghe in 2016, respectively. We collect OMI data pixels that are overlapped by a grid box of 0.25° at the center of the site location, and average values of OMI data are weighted by <u>uncertainties and</u> overlapped areas between pixels and grid boxes.

Comparisons of HCHO VCDs between MAX-DOAS and satellite products are shown in Fig. <u>11</u> and Table <u>4</u>. <u>Averaged MAX-DOAS HCHO VCDs for a year are 7.6 × 10<sup>15</sup>, 6.7 × 10<sup>15</sup>, and 1.6 × 10<sup>16</sup> molecules cm<sup>-2</sup> at OHP, Bremen, and Xianghe, respectively. <u>HCHO VCDs</u></u>

show a seasonal variation with the maximum concentrations in summer at all sites (Fig. S3).

## Deleted: BIRA

Deleted: use

**Deleted:** OMI BIRA uses a two-step method: radiance fitting is conducted in a broad fitting window (328.5–359.0 nm) to remove interferences from BrO and O<sub>4</sub>, and HCHO slant columns are then derived in a narrow fitting window (328.5–346.0 nm) with fitted BrO slant columns...

#### Deleted: BIRA

Deleted: . Differences caused by

**Deleted:** absorption datasets is the largest term in the

Deleted: (as mentioned in Section 3.1.2); therefore, relatively lower slant columns in the GEMS algorithm compared to OMI BIRA may result from the different

# Deleted: employed

Deleted: The regional and seasonal magnitudes of relative differences between OMI GEMS and OMI BIRA slant columns are larger over the Indochina Peninsula and China and in summer and winter, respectively, compared to those over Sumatra/Malaysia and Borneo, and compared to in spring and fall (Table 2). Biomass burning often occurs in spring and fall within Sumatra/Malaysia and Borneo, and during the spring in the Indochina Peninsula, which results in strong HCHO enhancements (see Fig. 11). That implies that HCHO can be well-retrieved because of the abundant HCHO concentrations, regardless of the fitting method used.<sup>¶</sup>

···(	Deleted: QA4ECV
••(	Deleted: 3
(	Deleted: daily averaged
(	Deleted: and are collected
···(	Deleted: in March, June, and September 2005, at
`(	Deleted: June and September
Y	Deleted: May
Å	Deleted: 12
4	Deleted: 3.
-(	Deleted: averaged
•••(	Deleted: each period
)	Deleted: 9
Y	Deleted: .2
Y	Deleted: 8

The largest monthly change is shown at Xianghe, likely driven by abundant VOC precursors for HCHO production compared to OHP and Bremen.

Averaged HCHO VCDs from OMI GEMS are by 16%, 9%, 25% lower than those from MAX-DOAS, at OHP, Bremen, and Xianghe. At Bremen, HCHO VCDs from the GEMS
algorithm are in the best agreement with those of MAX-DOAS and show similar monthly variations with MAX-DOAS. OMI GEMS results at Xianghe show a monthly variation but, at OHP, do not show monthly variation despite of a bit increment in summer. In particular, the GEMS algorithm yields lower HCHO VCDs in summer. These lower values may be caused by the a priori HCHO profiles used in AMF calculation. In summer, HCHO is produced and

10 concentrated near the surface, which results in lower AMFs (higher VCDs). <u>S. W. Kim et al.</u> (2018) showed the anti-correlation between AMF values and the HCHO mixing ratios at 200 m above ground level. <u>OMHCHO products show similar tendencies as OMI GEMS, but they are much lower than those of OMI GEMS. OMI QA4ECV products are higher than MAX-DOAS at OHP and Bremen but are in the best agreement with MAX-DOAS at Xianghe compared to other satellite products.</u>

#### **5** Conclusions and discussions

20

25

30

We have developed a GEMS HCHO algorithm based on a non-linearized fitting method and described the algorithm in detail. The GEMS HCHO algorithm consists of three steps: preprocesses, radiance fitting, and post-processes. Pre-processes include wavelength calibration, and interpolation and convolution of absorption cross-sections. In the radiance fitting, HCHO slant column densities are retrieved by minimizing the difference between calculated radiances with initial guesses of absorbing gases and measured radiances in HCHO fitting windows. Finally, AMF values are calculated from an AMF LUT, and bias corrections are conducted if necessary.

We estimated the uncertainties of slant columns, AMF, and background corrections, <u>using</u> <u>simulated radiances and OMI Level 1B data</u>. The random <u>uncertainties</u> of slant columns are <u>estimated using simulated radiances and are comparable</u> with <u>those of OMI GEMS</u> products. <u>The</u> systematic uncertainty is 38% of the slant columns, which is higher than that of De Smedt et al. (2018). However, the systematic uncertainty can be reduced by using up-to-date absorption cross-sections. AMF uncertainty <u>amounts</u> to <u>by 20%</u> and <u>24%</u> of the HCHO vertical columns, in clean and polluted areas, respectively, and mainly results from uncertainties

#### Deleted: results Deleted: %, 0.8%, and 30 Deleted: , respectively,

**Deleted:** by 45% and 27% in June at OHP and Bremen, respectively, compared to those from MAX-DOAS

Deleted: 2018b

Deleted: OMHCHO products show similar tendencies as OMI GEMS, but they are much lower than those of OMI GEMS. OMI BIRA products are higher by 19%–66% than ground observations at all three sites. Of the three satellite products, the OMI GEMS products show the smallest relative differences at OHP and Bremen against MAX-DOAS, where HCHO VCDs are relatively low. However, compared to the other two sites, there are smaller differences for OMI BIRA products in Xianghe where there are high HCHO VCDs.<sup>§</sup>

**Deleted:** for stripe patterns, background concentrations, and latitudinal biases  $\dots$ 

Deleted:	
Deleted: uncertainty	
Deleted: corresponds	
Deleted: OMHCHO	
Deleted: , but the	

-	Deleted: with respect
(	Deleted: surface albedo, cloud top pressure,
(	Deleted: cloud fraction contributes to 4–16
~(	Deleted: . AMF uncertainty with respect to

associated with HCHO profile heights and cloud information (cloud top pressure and cloud fractions).

# **Deleted:** height leads to 15%–63% uncertainty in the HCHO vertical columns, but this can be reduced by using averaging kernels.

Deleted: results

Deleted: 92	
Deleted: were found despite good agreement	
Deleted: GEMS and OMHCHO	
Deleted: , and these are caused by the use	
Deleted: different model results for	
Deleted: Although both	
Deleted: are	
Deleted: products are employed.	
Deleted: are	

Deleted: are	
Deleted: BIRA	
Deleted: they have	
Deleted: the	
Deleted: BIRA	
Deleted: consider	
Deleted: BIRA	
Deleted: may be	
Deleted: employed	
Deleted: compare	

(	<b>Deleted:</b> are 25%, 0.8
(	Deleted: 30
(	Deleted: are
(	Deleted: among
X	Deleted: three
(	Deleted: as GEMS handpass functions depend on wavelength

#### Deleted:

#### Deleted: narrow

**Deleted:** (De Smedt et al., 2018). Also, latitudinal biases for a radiance reference can also be minimized in GEMS employing its south to north swaths, as we plan to use easternmost swaths (except for islands such as Japan and Indonesia) for a radiance reference. Therefore, we will further investigate latitudinal biases after GEMS is launched...

OMI HCHO <u>columns</u> from the GEMS algorithm were compared to the OMHCHO products with consistent fitting conditions applied. OMI GEMS slant columns show good

- agreement with OMHCHO products, with correlation coefficients of 0.77–0.91 and regression slopes of 0.94–1.04. However, some differences between two products were found because of background corrections. Both products used the model results simulated by GEOS-Chem, but driven with different assimilated meteorological datasets. The simulated HCHO vertical columns used in OMHCHO products were from GEOS-Chem with GEOS-4 meteorological data, which have lower cloud optical depths near the equator compared to GEOS-3 and GEOS-5. Low cloud optical depths in the tropics result in faster methane oxidation and greater HCHO production caused by high hydroxyl radical concentrations. Therefore, OMHCHO slant columns are 1% to 13% higher than OMI GEMS slant columns in Sumatra/Malaysia and
- 15

20

Borneo near the equator.

The spatial distributions of GEMS HCHO slant columns <u>were</u> consistent with OMI <u>QA4ECV</u> products, but relatively poorer correlation coefficients of 0.52 to 0.76 are found compared to <u>those with</u> OMHCHO products. Relative differences between GEMS and <u>QA4ECV</u> slant columns range from -11% to -22% on average over the GEMS domain. We found that the discrepancy between the GEMS and <u>QA4ECV</u> products were mainly caused by the different radiance fitting methods, polynomial orders in the fitting window, the different O<sub>3</sub>

absorption datasets, and consideration of the non-linear O3 absorption effect.

We also <u>compared</u> satellite results with MAX-DOAS ground observations at OHP in France, Bremen in Germany, and Xianghe in China. HCHO VCDs from the GEMS algorithm <u>were by 16%, 9</u>%, and <u>25</u>% lower than <u>those of MAX-DOAS</u>, but the <u>GEMS</u> discrepancies at

25 OHP and Bremen were the smallest compared to the other satellites against the in-situ data. After GEMS is launched, several options need to be tested. As described in Section 2.2.1,

it will be necessary to estimate uncertainties resulting from the wavelength dependence of bandpass functions. We may also need to conduct <u>additional</u> sensitivity tests to optimize the fitting window, and fitting options. We currently use a broad fitting window (328.5–356.0 nm).

30 However, we may need to use a <u>different</u> fitting window to reduce interference from polarization effects because GEMS does not include a polarization scrambler, <u>A polarization</u> <u>correction is planned to minimize its interference during GEMS Level 1B production, but we</u>

need to examine the retrieval sensitivity to polarization. Additionally, an update to the optimized AMF LUT with finer spatial and temporal resolutions is required.

#### Acknowledgements

This subject is supported by Korea Ministry of Environment (MOE) as "Public Technology 5 Program based on Environmental Policy (2017000160001)". Part of the research was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with NASA.

#### References

15

- 10 Aliwell, S. R., Van Roozendael, M., Johnston, P. V., Richter, A., Wagner, T., Arlander, D. W., Burrows, J. P., Fish, D. J., Jones, R. L., Tørnkvist, K. K., Lambert, J. C., Pfeilsticker, K., and Pundt, I.: Analysis for BrO in zenith-sky spectra: An intercomparison exercise for analysis improvement, Journal of Geophysical Research: Atmospheres, 107, ACH 10-11-ACH 10-20, 10.1029/2001JD000329, 2002.
  - Barkley, M. P., De Smedt, I., Van Roozendael, M., Kurosu, T. P., Chance, K., Arneth, A., Hagberg, D., Guenther, A., Paulot, F., and Marais, E.: Top-down isoprene emissions over tropical South America inferred from SCIAMACHY and OMI formaldehyde columns, Journal of Geophysical Research: Atmospheres, <u>J18</u>, 6849-6868, 10.1002/jgrd.50552, 2013.
     Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., Li, Q., Liu, H. Y., Mickley, L. J., and Schultz,
  - M. G.: Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, Journal of Geophysical Research Atmospheres, 106, 23073-23095, <u>10.1029/2001JD000807</u>, 2001.
     Boersma, K. F., Eskes, H. J., and Brinksma, E. J.: Error analysis for tropospheric NO2 retrieval from space, Journal of
- 20 Geophysical Research: Atmospheres, 109, n/a-n/a, 10.1029/2003JD003962, 2004. Burrows, J. P., Dehn, A., Deters, B., Himmelmann, S., Richter, A., Voigt, S., and Orphal, J.: ATMOSPHERIC REMOTE-SENSING REFERENCE DATA FROM GOME: PART 1. TEMPERATURE-DEPENDENT ABSORPTION CROSS-
  - SECTIONS OF NO2 IN THE 231–794 nm RANGE, Journal of Quantitative Spectroscopy and Radiative Transfer, 60, 1025-1031, https://doi.org/10.1016/S0022-4073(97)00197-0, 1998. 25 Cantrell, C. A., Davidson, J. A., McDaniel, A. H., Shetter, R. E., and Calvert, J. G.: Temperature-dependent formaldehyde
  - cross sections in the near-ultraviolet spectral region, Journal of Physical Chemistry, 94, 3902-3908, 10.1021/j100373a008, 1990. Chan Miller, C., Gonzalez Abad, G., Wang, H., Liu, X., Kurosu, T., Jacob, D. J., and Chance, K.: Glyoxal retrieval from the
- 30 Ozone Monitoring Instrument, Atmos. Meas. Tech., 7, 3891-3907, 10.5194/amt-7-3891-2014, 2014. Chance, K., and Spurr, R. J. D.: Ring effect studies: Rayleigh scattering, including molecular parameters for rotational Raman
- scattering, and the Fraunhofer spectrum, Applied Optics, 36, 5224-5230, 10.1364/AO.36.005224, 1997. Chance, K., Palmer, P. I., Spurr, R. J. D., Martin, R. V., Kurosu, T. P., and Jacob, D. J.: Satellite observations of formaldehyde over North America from GOME, Geophysical Research Letters, 27, 3461-3464, 10.1029/2000GL011857, 2000.
- 35 Chance, K., Kurosu, T. P., and Sioris, C. E.: Undersampling correction for array detector-based satellite spectrometers, Applied Optics, 44, 1296-1304, 10.1364/AO.44.001296, 2005.
  - Chance, K., and Kurucz, R. L .: An improved high-resolution solar reference spectrum for earth's atmosphere measurements in the ultraviolet, visible, and near infrared, Journal of Quantitative Spectroscopy and Radiative Transfer, 111, 1289-1295, https://doi.org/10.1016/j.jqsrt.2010.01.036, 2010.
- 40 Chance, K., and Orphal, J.: Revised ultraviolet absorption cross sections of H2CO for the HITRAN database, Journal of Quantitative Spectroscopy and Radiative Transfer, 112, 1509-1510, http://dx.doi.org/10.1016/j.jqsrt.2011.02.002, 2011. Chehade, W., Gür, B., Spietz, P., Gorshelev, V., Serdyuchenko, A., Burrows, J. P., and Weber, M.: Temperature dependent ozone absorption cross section spectra measured with the GOME-2 FM3 spectrometer and first application in satellite retrievals, Atmos. Meas. Tech., 6, 1623-1632, 10.5194/amt-6-1623-2013, 2013.
- 45 Chimot, J., Vlemmix, T., Veefkind, J. P., de Haan, J. F., and Levelt, P. F.: Impact of aerosols on the OMI tropospheric NO2 retrievals over industrialized regions: how accurate is the aerosol correction of cloud-free scenes via a simple cloud model?, Atmos. Meas. Tech., 9, 359-382, 10.5194/amt-9-359-2016, 2016.
- Choi, Y., Kim, H., Tong, D., and Lee, P.: Summertime weekly cycles of observed and modeled NO<sub>x</sub> and O<sub>3</sub> concentrations as a function of satellite-derived ozone production sensitivity and land use types over the Continental United States, Atmos. Chem. Phys., 12, 6291-6307, 10.5194/acp-12-6291-2012, 2012. 50

26

Deleted:

Deleted: 2013

Daumont, D., Brion, J., Charbonnier, J., and Malicet, J.: Ozone UV spectroscopy I: Absorption cross-sections at room temperature, Journal of atmospheric chemistry, 15, 145-155, 10.1007/BF00053756, 1992

Smedt, I., Müller, J.-F., Stavrakou, T., van der A, R., Eskes, H., and Van Roozendael, M.: Twelve years of global De observations of formaldehyde in the troposphere using GOME and SCIAMACHY sensors, Atmospheric Chemistry & Physics, 8, 4947-4963, <u>10.5194/acp-8-4947-2008</u>, 2008. Smedt, I., Van Roozendael, M., Stavrakou, T., Müller, J. F., Lerot, C., Theys, N., Valks, P., Hao, N., and van der A, R.:

5

- De Improved retrieval of global tropospheric formaldehyde columns from GOME-2/MetOp-A addressing noise reduction and instrumental degradation issues, Atmos. Meas. Tech., 5, 2933-2949, 10.5194/amt-5-2933-2012, 2012. De Smedt, I., Stavrakou, T., Hendrick, F., Danckaert, T., Vlemmix, T., Pinardi, G., Theys, N., Lerot, C., Gielen, C., Vigouroux,
- 10 C., Hermans, C., Fayt, C., Veefkind, P., Müller, J. F., and Van Roozendael, M.: Diurnal, seasonal and long-term variations of global formaldehyde columns inferred from combined OMI and GOME-2 observations, Atmos. Chem. Phys., 15, 12519-12545, 10.5194/acp-15-12519-2015, 2015.
- De Smedt, I., Theys, N., Yu, H., Danckaert, T., Lerot, C., Compernolle, S., Van Roozendael, M., Richter, A., Hilboll, A., Peters, E., Pedergnana, M., Loyola, D., Beirle, S., Wagner, T., Eskes, H., van Geffen, J., Boersma, K. F., and Veefkind, P.: Algorithm theoretical baseline for formaldehyde retrievals from S5P TROPOMI and from the QA4ECV project, Atmos. 15 Meas. Tech., 11, 2395-2426, 10.5194/amt-11-2395-2018, 2018.
- DiGangi, J. P., Henry, S. B., Kammrath, A., Boyle, E. S., Kaser, L., Schnitzhofer, R., Graus, M., Turnipseed, A., Park, J. H., Weber, R. J., Hornbrook, R. S., Cantrell, C. A., Maudlin Iii, R. L., Kim, S., Nakashima, Y., Wolfe, G. M., Kajii, Y., Apel, E. C., Goldstein, A. H., Guenther, A., Karl, T., Hansel, A., and Keutsch, F. N.: Observations of glyoxal and formaldehyde 20 as metrics for the anthropogenic impact on rural photochemistry, Atmospheric Chemistry and Physics, 12, 9529-9543, 10.5194/acp-12-9529-2012, 2012.
  - Dirksen, R., Dobber, M., Voors, R., and Levelt, P.: Prelaunch characterization of the Ozone Monitoring Instrument transfer
- function in the spectral domain, Applied Optics, 45, 3972-3981, 10.1364/AO.45.003972, 2006.
  Duncan, B. N., Yoshida, Y., Olson, J. R., Sillman, S., Martin, R. V., Lamsal, L., Hu, Y., Pickering, K. E., Retscher, C., Allen, D. J., and Crawford, J. H.: Application of OMI observations to a space-based indicator of NOx and VOC controls on 25 surface ozone formation, Atmospheric Environment, 44, 2213-2223, http://dx.doi.org/10.1016/j.atmosenv.2010.03.010, 2010.
- Fleischmann, O. C., Hartmann, M., Burrows, J. P., and Orphal, J.: New ultraviolet absorption cross-sections of BrO at atmospheric temperatures measured by time-windowing Fourier transform spectroscopy, Journal of Photochemistry and 30 Photobiology A: Chemistry, 168, 117-132, https://doi.org/10.1016/j.jphotochem.2004.03.026, 2004.
- Go, S., Kim, J., Mok, J., Irie, H., Yoon, J. M., Torres, O., Krotkov, N., Labow, G., Kim, M., Koo, J.-H., Choi, M., and Lim, H.: Column Effective Imaginary part of refractive index derived from UV-MFRSR and SKYNET in Seoul, and implications for retrieving UV Aerosol Optical Properties from GEMS measurem ents, Remote Sensing of Enviro mited 2019
- 35 González Abad, G., Liu, X., Chance, K., Wang, H., Kurosu, T., and Suleiman, R.: Updated Smithsonian Astrophysical Observatory Ozone Monitoring Instrument (SAO OMI) formaldehyde retrieval, Atmospheric Measurement Techniques, 8, 19-32, 10.5194/amt-8-19-2015, 2015
- González Abad, G., Vasilkov, A., Seftor, C., Liu, X., and Chance, K.: Smithsonian Astrophysical Observatory Ozone Mapping and Profiler Suite (SAO OMPS) formaldehyde retrieval, Atmos. Meas. Tech., 9, 2797-2812, 10.5194/amt-9-2797-2016, 40 2016
- Hermans, C., Vandaele, A. C., and Fally, S.: Fourier transform measurements of SO2 absorption cross sections:: I. Temperature dependence in the 24 000-29 000 cm-1 (345-420 nm) region, Journal of Quantitative Spectroscopy and Radiative Transfer, 110, 756-765, http://dx.doi.org/10.1016/j.jcgsrt.2009.01.031, 2009.
- Hewson, W., Bösch, H., Barkley, M., and De Smedt, I.: Characterisation of GOME-2 formaldehyde retrieval sensitivity, 45
  - Atmospheric Measurement Techniques, 6, 371-386, <u>10.5194/amt-6-371-2013</u>, 2013.
    Hewson, W., Barkley, M. P., Gonzalez Abad, G., Bösch, H., Kurosu, T., Spurr, R., and Tilstra, L. G.: Development and characterisation of a state-of-the-art GOME-2 formaldehyde air-mass factor algorithm, Atmos. Meas. Tech., 8, 4055-4074, 10.5194/amt-8-4055-2015, 2015.
- Kim, H. C., Lee, P., Judd, L., Pan, L., and Lefer, B.: OMI NO2 column densities over North American urban cities: the effect 50 of satellite footprint resolution, Geosci. Model Dev., 9, 1111-1123, 10.5194/gmd-9-1111-2016, 2016
- Kim, J., Ukkyo Jeong, Myoung-Hwan Ahn, Jae H. Kim, Rokjin J. Park, Hanlim Lee, Chul Han Song, Yong-Sang Choi, Kwon-Ho Lee, Jung-Moon Yoo, Myeong-Jae Jeong, Seon Ki Park, Kwang-Mog Lee, Chang-Keun Song, Sang-Woo Kim, Young-Joon Kim, Si-Wan Kim, Xiong Liu, Kelly Chance, Christopher Chan Miller, Jay Al-Saadi, Ben Veihelmann, Pawan K. Bhartia, Omar Torres, Gonzalo González Abad, David P. Haffner, Dai Ho Ko, Seung Hoon Lee, Jung-Hun
- 55 Woo, Mijin Kim, Sujung Go, Heesung Chong, Sang Seo Park, Dennis Nicks, Won Jun Choi, Kyung-Jung Moon, Ara Cho, Jong-Min Yoon, Sang-Kyun Kim, Hyunkee Hong, Kyunghwa Lee, Hana Lee, Seoyoung Lee, Myungje Choi, Pepijin Veefkind, Pieternel Levelt, David Edwards, Mina Kang, Mijin Eo, Juseon Bak, Kanghyun Baek, Hyeong-Ahn Kwon, Jiwon Yang, Kyung Man Han, Boram Kim, Hee-Woo Shin, Haklim Choi, Ebony Lee, Jihyo Chong, Yesol Cha, Hitoshi Irie, Sachiko Hayashida, Yasko Kasai, Yugo Kanaya, C. L., Jintai Lin, James H. Crawford, Gregory R. Carmichael,
- 60 Michael J. Newchurch, Barry L. Lefer, Robert J. Swap, Alexis K H Lau, Thomas P. Kurosu, Glen Jaross, Berit Ahlers, and Dobber, M.: New Era of Air Quality Monitoring from Space: Geostationary Environment Monitoring Spectrometer (GEMS) Bulletin of the American Meteorological Society, submitted, <u>2018</u>.
- Kim, M., Kim, J., Torres, O., Ahn, C., Kim, W., Jeong, U., Go, S., Liu, X., Moon, J. K., and Kim, D.-R.: Optimal Estimation Based Algorithm to Retrieve Aerosol Optical Properties for GEMS Measurements over Asia, Remote Sensing, 10, 65 10.3390/rs10020162, 2018.

Deleted: 2018a

- Kim, S. W., Natraj, V., Lee, S., Kwon, H. A., Park, R., de Gouw, J., Frost, G., Kim, J., Stutz, J., Trainer, M., Tsai, C., and Warneke, C.: Impact of high-resolution a priori profiles on satellite-based formaldehyde retrievals, Atmos. Chem. Phys., 18, 7639-7655, 10.5194/acp-18-7639-2018, <u>2018</u>.
- Kleipool, Q. L., Dobber, M. R., de Haan, J. F., and Levelt, P. F.: Earth surface reflectance climatology from 3 years of OMI data, 113, doi:10.1029/2008JD010290, 2008.
   Kurucz, R. L., Furenlid, I., Brault, J., and Testerman, L.: Solar flux atlas from 296 to 1300nm. National Solar Observatory,
  - Sunspot, New Mexico, 1984.
    Kwon, H. A., Park, R. J., Jeong, J. I., Lee, S., González Abad, G., Kurosu, T. P., Palmer, P. I., and Chance, K.: Sensitivity of formaldehyde (HCHO) column measurements from a geostationary satellite to temporal variation of the air mass factor
- 10 in East Asia, Atmos. Chem. Phys., 17, 4673-4686, 10.5194/acp-17-4673-2017, 2017. Lee, K., and Yoo, J.-M.: Determination of directional surface reflectance from geostationary satellite observations, SPIE Asia-Pacific Remote Sensing, 10777-52, 2018.
- Li, C., Joiner, J., Krotkov, N. A., and Dunlap, L.: A new method for global retrievals of HCHO total columns from the Suomi National Polar-orbiting Partnership Ozone Mapping and Profiler Suite, Geophysical Research Letters, 42, 2515-2522, 10.1002/2015GL063204, 2015.
  - Lin, J. T., Martin, R. V., Boersma, K. F., Sneep, M., Stammes, P., Spurr, R., Wang, P., Van Roozendael, M., Clémer, K., and Irie, H.: Retrieving tropospheric nitrogen dioxide from the Ozone Monitoring Instrument: effects of aerosols, surface reflectance anisotropy, and vertical profile of nitrogen dioxide, Atmos. Chem. Phys., 14, 1441-1461, 10.5194/acp-14-1441-2014, 2014.
- 20 Malicet, J., Daumont, D., Charbonnier, J., Parisse, C., Chakir, A., and Brion, J.: Ozone UV spectroscopy. II. Absorption crosssections and temperature dependence, Journal of atmospheric chemistry, 21, 263-273, <u>10.1007/BF00696758</u>, 1995. Marais, E. A., Jacob, D. J., Kurosu, T. P., Chance, K., Murphy, J. G., Reeves, C., Mills, G., Casadio, S., Millet, D. B., and
- Maras, E. A., Jacos, D. J., Rubski, T. T., Chance, R., Hulpiry, J. G., Reves, C., Mins, G., Casado, S., Minet, D. B., and Barkley, M. P.: Isoprene emissions in Africa inferred from OMI observations of formaldehyde columns, Atmospheric Chemistry and Physics, 12, 6219-6235, <u>10.5194/acp-12-6219-2012</u>, 2012.
- 25 Martin, R. V., Chance, K., Jacob, D. J., Kurosu, T. P., Spurr, R. J. D., Bucsela, E., Gleason, J. F., Palmer, P. I., Bey, I., Fiore, A. M., Li, Q., Yantosca, R. M., and Koelemeijer, R. B. A.: An improved retrieval of tropospheric nitrogen dioxide from GOME, Journal of Geophysical Research: Atmospheres, 107, ACH 9-1-ACH 9-21, 10.1029/2001JD001027, 2002. Martin, R. V., Fiore, A. M., and Van Donkelaar, A.: Space-based diagnosis of surface ozone sensitivity to anthropogenic
- emissions, Geophysical Research Letters, 31, n/a-n/a, 10.1029/2004GL019416, 2004. 30 Meller, R., and Moortgat, G. K.: Temperature dependence of the absorption cross sections of formaldehyde between 223 and
- 323 K in the wavelength range 225–375 nm, Journal of Geophysical Research: Atmospheres, 105, 7089-7101, 10.1029/1999JD901074, 2000.
  - Millet, D. B., Jacob, D. J., Turquety, S., Hudman, R. C., Wu, S., Fried, A., Walega, J., Heikes, B. G., Blake, D. R., Singh, H. B., Anderson, B. E., and Clarke, A. D.: Formaldehyde distribution over North America: Implications for satellite retrievals of formaldehyde columns and isoprene emission, Journal of Geophysical Research: Atmospheres, 111, doi:10.1029/2005JD006853.2006.

35

- Nowlan, C. R., Liu, X., Leitch, J. W., Chance, K., González Abad, G., Liu, C., Zoogman, P., Cole, J., Delker, T., Good, W., Murcray, F., Ruppert, L., Soo, D., Follette-Cook, M. B., Janz, S. J., Kowalewski, M. G., Loughner, C. P., Pickering, K. E., Herman, J. R., Beaver, M. R., Long, R. W., Szykman, J. J., Judd, L. M., Kelley, P., Luke, W. T., Ren, X., and Al-Saadi, J. A.: Nitrogen dioxide observations from the Geostationary Trace gas and Aerosol Sensor Optimization (GeoTASO) airborne instrument: Retrieval algorithm and measurements during DISCOVER-AQ Texas 2013, Atmos. Meas. Tech., 9, 2647-2668, 10.5194/antt-9-2647-2016, 2016.
- Palmer, P. I., Jacob, D. J., Fiore, A. M., and Martin, R. V.: Air mass factor formulation for spectroscopic measurements from satellites: Application to formaldehyde retrievals from the Global Ozone Monitoring Experiment, Journal of geophysical research, 106, 14,539-514,550, <u>10.1029/2000JD900772</u>, 2001.
  - Pinardi, G., Van Roozendael, M., Abuhassan, N., Adams, C., Cede, A., Clémer, K., Fayt, C., Frieß, U., Gil, M., Herman, J., Hermans, C., Hendrick, F., Irie, H., Merlaud, A., Navarro Comas, M., Peters, E., Piters, A. J. M., Puentedura, O., Richter, A., Schönhardt, A., Shaiganfar, R., Spinei, E., Strong, K., Takashima, H., Vrekoussis, M., Wagner, T., Wittrock, F., and Yilmaz, S.: MAX-DOAS formaldehyde slant column measurements during CINDI: intercomparison and analysis improvement, Atmos. Meas. Tech., 6, 167-185, 10.5194/amt-6-167-2013, 2013.
  - Pukīte, J., Kühl, S., Deutschmann, T., Platt, U., and Wagner, T.: Extending differential optical absorption spectroscopy for limb measurements in the UV, Atmos. Meas. Tech., 3, 631-653, 10.5194/amt-3-631-2010, 2010.
- Serdyuchenko, A., Gorshelev, V., Weber, M., Chehade, W., and Burrows, J. P.: High spectral resolution ozone absorption crosssections – Part 2: Temperature dependence, Atmos. Meas. Tech., 7, 625-636, 10.5194/amt-7-625-2014, 2014.
- 55 Spurr, R. J.: VLIDORT: A linearized pseudo-spherical vector discrete ordinate radiative transfer code for forward model and retrieval studies in multilayer multiple scattering media, Journal of Quantitative Spectroscopy and Radiative Transfer, 102, 316-342, 10.1016/j.jqsrt.2006.05.005, 2006.
- Stavrakou, T., Müller, J. F., Bauwens, M., De Smedt, I., Van Roozendael, M., Guenther, A., Wild, M., and Xia, X.: Isoprene emissions over Asia 1979-2012: impact of climate and land-use changes, Atmos. Chem. Phys., 14, 4587-4605, 10.5194/acp-14-4587-2014, 2014.
  - Thalman, R., and Volkamer, R.: Temperature dependent absorption cross-sections of O2–O2 collision pairs between 340 and 630 nm and at atmospherically relevant pressure, Physical Chemistry Chemical Physics, 15, 15371-15381, 10.1039/C3CP50968K, 2013.
- Vandaele, A. C., Hermans, C., Simon, P. C., Carleer, M., Colin, R., Fally, S., Mérienne, M. F., Jenouvrier, A., and Coquart, B.:
   Measurements of the NO2 absorption cross-section from 42 000 cm<sup>-1</sup> to 10 000 cm<sup>-1</sup> (238–1000 nm) at 220 K and 294

Deleted: 2018h

K, Journal of Quantitative Spectroscopy and Radiative Transfer, 59, 171-184, http://dx.doi.org/10.1016/S0022-4073(97)00168-4, 1998.

- 40/3(9/)00108-4, 1996.
   Vandaele, A. C., Hermans, C., and Fally, S.: Fourier transform measurements of SO2 absorption cross sections: II.: Temperature dependence in the 29 000-44 000 cm-1 (227–345 nm) region, Journal of Quantitative Spectroscopy and Radiative Transfer, 110, 2115-2126, http://dx.doi.org/10.1016/j.jqsrt.2009.05.006, 2009.
   Veefkind, J. P., de Haan, J. F., Sneep, M., and Levelt, P. F.: Improvements to the OMI 02–02 operational cloud algorithm and the transfer of the Comparison o
- comparisons with ground-based radar-lidar observations, Atmos. Meas. Tech., 9, 6035-6049, 10.5194/amt-9-6035-2016, 2016.
- Vrekoussis, M., Wittrock, F., Richter, A., and Burrows, J. P.: GOME-2 observations of oxygenated VOCs: what can we learn 10 from the ratio glyoxal to formaldehyde on a global scale?, Atmos. Chem. Phys., 10, 10145-10160, 10.5194/acp-10-10145-2010, 2010.
  - Wedin, P. Å., and Lindström, P.: Methods and Software for Nonlinear Least Squares Problems, Umeå University. Institute of Information Processing, 1987
- Wilmouth, D. M., Hanisco, T. F., Donahue, N. M., and Anderson, J. G.: Fourier Transform Ultraviolet Spectroscopy of the A 2II3/2 ← X 2II3/2 Transition of BrO, The Journal of Physical Chemistry A, 103, 8935-8945, 10.1021/jp9916510, 1999. 15 Wittrock, F., Richter, A., Oetjen, H., Burrows, J. P., Kanakidou, M., Myriokefalitakis, S., Volkamer, R., Beirle, S., Platt, U., and Wagner, T.: Simultaneous global observations of glyoxal and formaldehyde from space, Geophysical Research Letters, 33, n/a-n/a, 10.1029/2006GL026310, 2006.
- Zhong, L., Louie, P. K. K., Zheng, J., Yuan, Z., Yue, D., Ho, J. W. K., and Lau, A. K. H.: Science-policy interplay: Air quality 20 management in the Pearl River Delta region and Hong Kong, Atmospheric Environment, 76, https://doi.org/10.1016/j.atmosenv.2013.03.012, 2013. 3-10.
- Zhu, L., Jacob, D. J., Mickley, L. J., Marais, E. A., Cohan, D. S., Yoshida, Y., Ducan, B. N., González Abad, G., and Chance, K. V: Anthropogenic emissions of highly reactive volatile organic compounds in eastern Texas inferred from oversampling of satellite (OMI) measurements of HCHO columns, Environmental Research Letters, 9, 114004, 10.1088/1748-9326/9/11/114004, 2014. 25
- Zhu, L., Jacob, D. J., Kim, P. S., Fisher, J. A., Yu, K., Travis, K. R., Mickley, L. J., Yantosca, R. M., Sulprizio, M. P., De Smedt, I., González Abad, G., Chance, K., Li, C., Ferrare, R., Fried, A., Hair, J. W., Hanisco, T. F., Richter, D., Jo Scarino, A., Walega, J., Weibring, P., and Wolfe, G. M.: Observing atmospheric formaldehyde (HCHO) from space: validation and intercomparison of six retrievals from four satellites (OMI, GOME2A, GOME2B, OMPS) with SEAC4RS aircraft 30 observations over the southeast US, Atmos. Chem. Phys., 16, 13477-13490, 10.5194/acp-16-13477-2016, 2016.

45

35

40

5

Table 1. Summary of GEMS system attributes, parameters for radiance fitting, and parameters for the AMF look-up table.

GEMS system attributes		
Spectral range	300–500 nm	
Spectral resolution	<u>&lt;</u> 0.6 nm	
Wavelength sampling	<u>≤</u> 0.2 nm	
	<u>&gt; 720</u> at <u>320 nm</u>	Deleted: < 56 km <sup>2</sup>
Signal-to-noise ratio	<u>≥ 1500 at 430 nm</u>	Deleted: Seoul
	$\geq$ 5000 (N/S) × 5000 (E/W) km <sup>2</sup>	Deleted: Spatial resolution
Field of regard	(5°S-45°N, 75°E-145°E)	Deleted: Observation coverage
Spatial resolution (at Socul)	$\leq$ 3.5 × 8 km <sup>2</sup> for aerosol	
<u>Spatial resolution (at Seour)</u>	$\leq$ 7 $\times$ 8 km <sup>2</sup> for gas	
Duty cycle	<u>∼</u> 8 times/day	Deleted: ≥
Imaging time	$\leq$ 30 minutes	
Radiance fitting parameters <sup>a</sup>		Deleted: parameters
Fitting window (calibration window)	328.5–356.5 nm (325.5–358.5 nm)	
	Measured radiances from far east swaths	
Radiance reference	<u>(143-150°E) for a day</u>	
Solar reference spectrum	Chance and Kurucz (2010) <sup>b</sup>	
Absorption cross-sections	HCHO at 300 K (Chance and Orphal, 2011)	
	O3 at 228 K and 295 K (Malicet et al., 1995;	
	Daumont et al., 1992)	
	NO <sub>2</sub> at 220 K (Vandaele et al., 1998) <sup>b</sup>	
	BrO at 228 K (Wilmouth et al., 1999)	
	O4 at 293 K (Thalman and Volkamer, 2013) <sup>b</sup>	Deleted: 294
Ring effect	Chance and Spurr (1997) <sup>b</sup>	
Common modo	On-line common mode from <u>casternmost</u>	Deleted: far east
Common mode	swaths (143-150°E) for a day	
Scaling and baseline polynomials	3 <sup>rd</sup> order	
AMF look-up table parameters		
Longitude (degree) (n=33)	70 to 150 with 2.5 grid	
Latitude (degree) (n=30)	-4 to 54 with 2.0 grid	
	30	

Solar Zenith Angle (degree) (n=9)	0, 10, 20, 30, 40, 50, 60, 70, 80 0, 10, 20, 30, 40, 50, 60, 70, 80					
Viewing Zenith Angle (degree) (n=9)						
Relative Azimuth Angle (degree) (n=3)	0, 90, 180					
Cloud Top Pressure (hPa) (n=7)	900, 800, 700, 600, 500, 300, 100					
Surface Albedo (n=7)	0, 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.0					
<sup>a</sup> GEMS fitting parameters follow González Abad et	al. (2015). However, undersampling is not included in the fitting					
parameters for GEMS, and reference sectors for radiance reference and common mode are different.						
<sup>2</sup> Datasets used in QA4EUV retrievals. Please refer to De Smedt et al. (2018) for details on other datasets and fitting option						
	31					
	Solar Zenith Angle (degree) (n=9) Viewing Zenith Angle (degree) (n=9) Relative Azimuth Angle (degree) (n=3) Cloud Top Pressure (hPa) (n=7) Surface Albedo (n=7) <sup>a</sup> GEMS fitting parameters follow González Abad et parameters for GEMS, and reference sectors for radiance <sup>b</sup> Datasets used in QA4ECV retrievals. Please refer to Da					

Table 2, <u>Retrieval uncertainties of GEMS HCHO VCD due to AMF uncertainties as functions of surface albedos, cloud top pressures, cloud fractions, and HCHO profile heights for clean and polluted areas. Values are calculated for conditions with solar zenith angle of 30°, viewing zenith angle of 30°, relative azimuth angle of 0°, cloud fractions less than 0.3, and a profile height of 700 hPa.</u>

**Deleted:** . Relative differences between OMI GEMS HCHO slant columns and OMHCHO and OMI BIRA slant columns in four regions.¶

HCHO VCD uncertainty				(	Deleted: Region			
due to AMF uncertainty	<u>Clean area</u>	Polluted area		(	Deleted: GEMS vs. OMHCHO			
			_	$\sim$	Deleted: GEMS vs. OMI BIRA			
Surface albedo ( $\alpha_s$ )	<u>1-10%</u>	<u>1-12%</u>						
<u>Cloud top pressure <math>(p_c)</math></u>	<u>0-11%</u>	<u>0-11%</u>						
Cloud fraction (f <sub>c</sub> )	<u>0-19%</u>	<u>0-17%</u>						
HCHO height (p <sub>h</sub> )	<u>0-11%</u>	<u>0-17%</u>						
Total	<u>2-20%</u>	<u>3-24%</u>						
Dogion	GEMS vs. OMHCHO				GEMS vs. OMI QA4ECV			
--	-----------------	------	------	------	---------------------	------	------	------
Kegion	Mar.	Jun.	Sep.	Dec.	Mar.	Jun.	Sep.	Dec.
Sumatra/Malaysia (95°-110°E, 0°-7°N)	-7%	-12%	-8%	-4%	-0.5%	-18%	-6%	-15%
Indochina Peninsula (97°-110°E, 10°-20°N)	8%	11%	9%	25%	-7%	-20%	-20%	-17%
China (110°-120°E, 30°- <mark>40°N</mark> )	12%	6%	10%	6%	-21%	-25%	-20%	-23%
Borneo (110°-118°E, 5°S-0°)	-7%	-13%	-7%	-1%	-9%	-13%	0.4%	-18%

Deleted: 40N

## Table 3. Relative differences between OMI GEMS HCHO slant columns and OMHCHO and OMI QA4ECV slant columns in four regions.

Table 4. Averaged HCHO VCDs (molecules cm<sup>-2</sup>) from MAX-DOAS ground observations and OMI satellite data at OHP in France, Bremen in Germany, and Xianghe in China. For satellites, mean values are weighted by <u>uncertainties and</u> overlapped <u>areas</u> between satellite pixels and <u>0.25° grid cells</u> <u>for each site</u>. Relative differences between OMI and MAX-DOAS are given in parentheses.

,Site <sup>a</sup>	Class <sup>b</sup>	MAX- <mark>DOAS</mark> °	ОМНСНО	OMI QA4ECV	OMI
					GEMS
OHP			<b>5</b> 0 1015	1 1 1016	<b>CO</b> 1015
(44°N,	Rural	$7.5 \times 10^{15}$	$5.8 \times 10^{15}$	$1 1 \times 10^{10}$	$6.3 \times 10^{15}$
5 59E)		V	(- <u>24</u> %)	( <u>50</u> %)	(- <u>16</u> %)
3.3°E)					
Bremen	<b>T</b> T 1	<u>6.7 × 10<sup>15</sup></u>	$5.1 \times 10^{15}$	$9.3 \times 10^{15}$	$6.1 \times 10^{15}$
(53°N, 9°E)	Urban		(- <u>23</u> %)	<u>(40</u> %)	(- <u>9</u> %)
Xianghe					
(200)	C11	1 ( 1016	$1.0 \times 10^{16}$	$1.7 \times 10^{16}$	$1.2 \times 10^{16}$
(39 <sup>-</sup> N,	Sub-urban	$1.0 \times 10^{10}$	(-37%)	(4%)	(-25%)
117°E)			( <u>•                                    </u>	₩ <u></u> , s)	

5 # HCHO VCDs are averaged at OHP and Bremen in 2005, and at Xianghe in 2016. Class is assigned in a QA4ECV MAXDOAS website (http://uv-vis.aeronomie.be/groundbased/QA4ECV\_MAXDOAS) <sup>e</sup>\_Fitting windows of 336–359 nm and 324–359 nm are used at OHP and Bremen, and at Xianghe, respectively.

10

20

15

Deleted: area		
Deleted: of 0.25° at locations		
Deleted: Site*		
Deleted: Class		
Deleted: DOAS**		
Deleted: BIRA		
Deleted: 4		
Deleted: 3		
Deleted: 5.9		

Deleted:

Deleted: Class	2
Deleted: DOAS**	2
Deleted: BIRA	$\sum$
Deleted: 4	2
Deleted: 3	$\mathcal{D}$
Deleted: 5.9	2
Deleted: 9	2
Deleted: 32	)
Deleted: 61	$\mathcal{D}$
Deleted: 25	2
Deleted: 6.0	2
Deleted: 8.8	)
Deleted: 7.2	)
Deleted: .2	)
Deleted: 17	)
Deleted: 21	$\mathbb{D}$
Deleted: 0.8	$\mathcal{D}$
Deleted: 1	)
Deleted: 2.2	2
Deleted: 3	$\sum$
Deleted: 8	$\sum$
Deleted: 41	$\sum$
Deleted: 19	$\sum$
Deleted: 30	2
Deleted: *	2
Deleted: in March, June, September 2005; at	2
Deleted: June and September	2
Deleted: ;	2
Deleted: May	)
Deleted: **	



Figure 1. GEMS field of regard (<u>red), nominal daily scan (blue), full central scan (magenta), full</u> <u>western scan (cyan),</u> and GEMS location (blue star). <u>Shaded areas (143-150°E) are regions for</u> <u>radiance references and common mode.</u>





Figure 2. Flow chart for GEMS HCHO algorithm.



Figure 3. Full width at half maximum (FWHM) of GEMS bandpass as a function of spatial index <u>of GEMS detection rows</u> (left) and averaged bandpass functions <u>for spatial indices</u> at 330 and 365 nm and relative differences (right).

Deleted: to
Deleted: index



Figure 4. Scattering weights (left) and normalized profiles (right) from the AMF LUT over clean (blue) and polluted (red) grids. Dashed and dotted lines in the left figure indicate scattering weights <u>with no</u> <u>cloud (nc)</u> and with cloud<u>(wc)</u>, respectively. The solid line in <u>the</u> left figure indicates scattering weights <u>for a partial cloudy scene</u> calculated <u>using</u> Eq. 10.



5





Deleted: and

... [1]

5

10

AMF LUT are given in the figures. For sensitivity to surface albedo, cloud-free conditions are assumed.

For sensitivity to cloud fraction, cloud top pressures are 800 hPa (solid line) and 500 hPa (dashed line).





Figure 7. Fitted HCHO optical depth (top) and fitting residuals (bottom) on a pixel (March 23, 2005; orbit 3655) with main data quality flag of 0 and effective cloud fraction less than 0.3. In the top panel, the black solid line indicates optical depth, and the red solid line indicates HCHO optical depth plus fitting residuals.



Figure 8. Monthly mean slant <u>column densities (SCDs)</u> from GEMS algorithm (1<sup>st</sup> row) and OMHCHO products (2<sup>nd</sup> row) for four months in 2005, and scatter plots between GEMS and OMHCHO. Statistics are given in the figures.

Deleted: columns



Figure 9. Simulated HCHO vertical column densities used in OMHCHO (solid lines) and GEMS algorithm (dashed lines) for background correction (top), and absolute differences of model results between GEMS algorithm and OMHCHO.







Figure 11\_HCHO vertical columns from MAX-DOAS, OMHCHO, OMI\_QA4ECV, and OMI GEMS at OHP and <u>Bremen</u> in 2005, and at Xianghe in 2016, Orange lines are median values for each dataset, and blue diamonds are mean values. We computed mean values of each satellite product weighted by <u>uncertainties and</u> overlapped <u>areas</u> between satellite pixels and 0.25° grid cells for each site. Boundaries of boxes indicate first and last quantiles of datasets.

 Moved (insertion) [5]

 Deleted: BIRA

 Deleted: (March, June,

 Deleted: September

 Deleted: ), Bremen (June, September in 2005),

 Deleted: (May

 Deleted: ).

 Deleted: diamond markers

 Deleted: For satellites,

 Deleted: area

 Deleted: of 0.25° at locations.

Page 39: [1] Deleted

•

Author

5/16/19 2:00:00 PM