Responses to Referee's Comments

We are grateful for the referee's valuable and insightful comments. The referee's comments are presented in black, our responses are highlighted in blue, and the revisions made in the manuscript are indicated in red.

Referee #1:

Ha et al. introduce the GEMS glyoxal retrieval algorithm and show comparisons against similar observations made from TROPOMI, and a set of MAX-DOAS instruments in the GEMS field of regard. In general they find reasonable agreement with both, however the TROPOMI comparison revealed a bias in GEMS glyoxal at high NO2 concentrations. The correction required in the spectral-fitting algorithm is not currently implemented in the GEMS algorithm.

The paper is well written and will be a useful reference for other researchers who plan to use the product. I recommend publication after the following comments are addressed.

1. L80: Would the spectral fitting not be stable for the retrieval to be performed at native spatial resolution? Aggregation generally causes problems, the most important probably being increased cloud contamination. In general I would have thought the main reason for aggregation is computational expediency (which is fine). I would just have thought that you would get at least equivalent precision/accuracy by aggregating the retrieved glyoxal at native resolution compared to aggregating at L1.

   The reason for co-adding L1 products before the spectral fitting is to ensure stable spectral fitting rather than to enhance computational expediency. We compared the signal-to-noise ratio (SNR) of glyoxal VCDs retrieved at native resolution and co-added resolution. The SNR of glyoxal VCDs retrieved at native resolution and co-added resolution was 0.18 and 0.33, respectively. Figure R1 compares glyoxal VCDs aggregated at L1 and L2 for one scene. Glyoxal VCDs aggregated at L1 and L2 represent similar spatial distribution. However, glyoxal VCDs aggregated at L2 show extreme values along some tracks and in the region where cloud fraction is high. This implies that the retrieval at native resolution is a bit unstable, therefore we use co-added L1 products at the cost of reducing the spatial resolution.

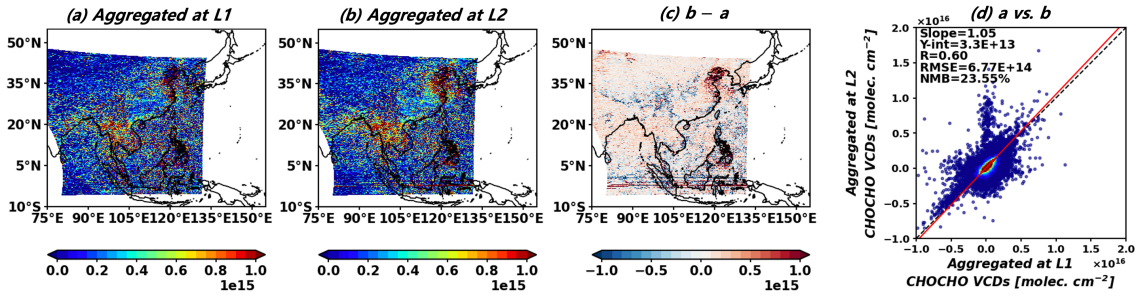

Figure R1. Comparison of the glyoxal VCDs aggregated at L1 and L2 at 03:45 UTC on 11 March 2021. Panel d is the scatter plot comparing glyoxal VCDs aggregated L1 and L2 with

filtering glyoxal VCDs lower than -1×10^{16} molecules cm^{-2} and higher than 2×10^{16} molecules cm^{-2}.

2. L85: I know this will be in basically every NO2, HCHO and CHOCHO retrieval paper, but you probably should add equation 6 from Kwon to make the paper self contained.

I added in the revised manuscript Eq. (6) from Kwon et al. (2019) as Eq. (1) with a brief explanation.

L85: The modeled radiative transfer equation demonstrates the attenuation of the reference spectrum by gas absorptions based on the Lambert-Beer law (Kwon et al., 2019), and can be expressed as follows:

$$I(\lambda) = \left[\left(aI_0(\lambda) + c_r\sigma_r(\lambda)\right) e^{-\sum_i SCD_i\sigma_i(\lambda)} + c_{cm}\sigma_{cm}(\lambda)\right]P_{sc}(\lambda) + P_{bl}(\lambda) \quad (1)$$

In Eq. (1), a represents an amplification factor applied to the reference spectrum $I_0(\lambda)$; $c_r\sigma_r(\lambda)$ accounts for the contribution of the Ring effect; the term $e^{-\sum_i SCD_i\sigma_i(\lambda)}$ characterizes the attenuation of light by absorbing species i; $c_{cm}\sigma_{cm}(\lambda)$ reflects the influence of the common mode; and $P_{sc}(\lambda)$ and $P_{bl}(\lambda)$ stand for scaling and baseline polynomials, respectively.

3. L90: The spectra region used for the radiance reference may contain significant liquid water absorption, which has caused significant negative biases in retrieved glyoxal, as demonstrated from the previous instruments (GOME-2, OMI) cited in the introduction. If these are used as a radiance reference it may artificially increase glyoxal concentrations over land. Perhaps some of this may be mitigated by the updated Mason and Fry cross section. How does this compare to the previous Pope and Fry liquid water cross section used by the previous studies?

Thanks for the constructive suggestion. Following your recommendation, we conducted a sensitivity test to retrieve glyoxal VCDs using the liquid water absorption cross-sections from Mason et al. (2016) and Pope and Fry (1997). Figure R2 compares glyoxal VCDs using the two liquid water absorption cross-sections. Glyoxal VCDs increase on land and decrease in the ocean when a liquid water cross-section from Mason et al. (2016) is used. Figure R3 shows the retrieved liquid water dSCDs while retrieving glyoxal VCDs in Figure R2. Liquid water dSCDs retrieved with absorption cross-section from Mason et al. (2016) show lower values on land and higher values in the ocean. Lower liquid water dSCD on land resulted in higher glyoxal VCDs using a liquid water absorption cross-section from Mason et al. (2016). The change in glyoxal VCDs is evident in 0–30° N, which is the latitude range where open oceans exist and liquid water concentration is high. Using absorption cross-section from Mason et al. (2016) increased glyoxal VCDs on land and slightly worsened the overestimation bias of GEMS glyoxal VCDs.

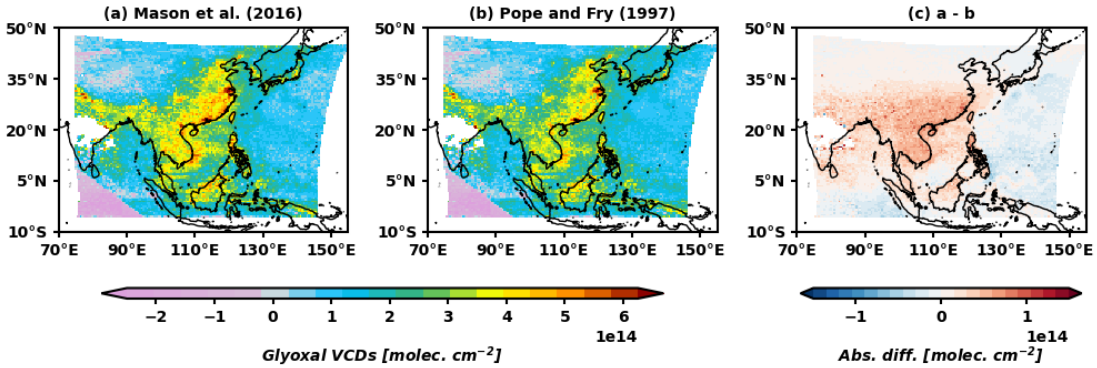

Figure R2. GEMS glyoxal VCDs retrieved using liquid water absorption cross-sections from (a) Mason et al. (2016) and (b) Pope and Fry (1997) in August 2020. Panel c shows the absolute difference between Panel a and Panel b.

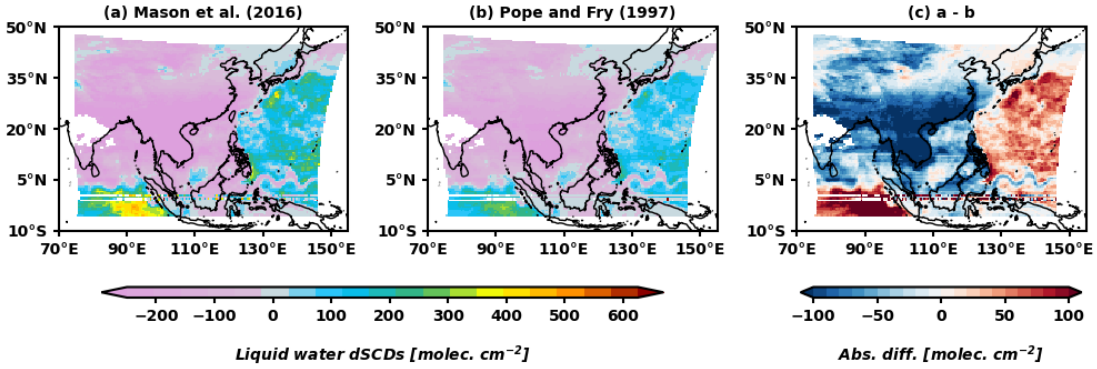

Figure R3. Liquid water dSCDs fitted (a) using the same setting used to retrieve GEMS glyoxal V2.0 and (b) using Pope and Fry (1997) liquid water absorption cross-section in August 2020.

L91: This reference sector contains open oceans where interference of liquid water absorption occurs, which could lead to a high bias of glyoxal VCDs in land. However, we inevitably selected this region as a reference sector since it generally shows low concentrations of glyoxal and other pollutants.

4. L97: The retrieval optimization is mentioned in the intro and conclusion, but not really presented in the text. I think it would be worth adding a figure showing the fit window optimization, and provide more details of the analysis.

We included in the revised manuscript a description of the sensitivity test for the fitting window selection as follows:

L99: We conduct sensitivity tests of fitting window selection to minimize fitting RMS and column uncertainty of the retrieved glyoxal averaged over the entire domain by varying lower and upper wavelengths with 0.5 nm increments. Figure 1 shows the results of our sensitivity tests and our optimal fitting window of 433.0–461.5 nm for glyoxal retrieval. The fitting window of 433.0–461.5 nm was selected considering its low fitting RMS and column uncertainty. However, we find that the differential slant column densities (dSCDs) over the reference sector (120–150° E) retrieved with this fitting window have a positive value, which could result in a high systematic bias, which we discuss below.

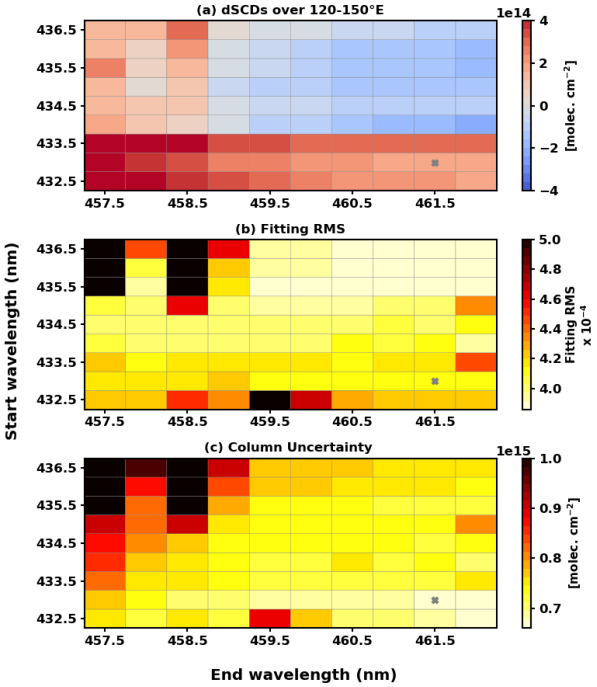

Figure 1. Metrics used to select an optimal fitting window. Panel a shows dSCDs averaged over 120–150°E, and panels b and c show fitting RMS and column uncertainty averaged over the entire domain. Values are calculated from the retrieval at 04:45 UTC on 17 March 2021.

5. L117: Is the OMI LER product the most appropriate surface reflectance database for GEMS? Given the different instrument viewing geometries, the equivalent GEMS LER may be significantly different due to BRDF effects, and the OMI LER database spatial resolution is coarse compared to the GEMS pixel size. Are there plans to update this in the future?

It is desirable to use GEMS surface reflectance since it considers BRDF effects and has finer spatial and temporal resolution than OMI LER product. However, GEMS background surface reflectance (BSR) V2.0 has issues such as a distinct bias for different wavelengths compared to TROPOMI directionally-dependent LER (DLER), as well as discontinuity between land and ocean. Figure R4 shows GEMS and OMI surface reflectance and the glyoxal VCDs retrieved with each reflectance. GEMS BSR V2.0 displays much lower values over ocean areas than on land, leading to higher glyoxal VCDs in the ocean than on land. However, advancements have been made in GEMS BSR V3.0, showing more consistent values with OMI LER and TROPOMI DLER. Therefore, we plan to update GEMS glyoxal retrieval to v3.0 using GEMS BSR V3.0 product in the future.

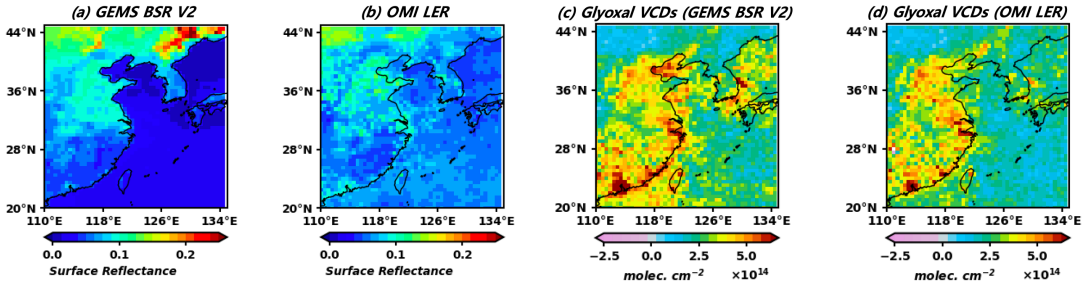

Figure R4. Surface reflectance values of GEMS BSR V2.0 (a) and OMI LER (b) and glyoxal VCDs retrieved using GEMS BSR V2.0 (c) and OMI LER (d) in March 2022.

6. L174: Is this paper describing “GEMS glyoxal V2.0”? It probably should be mentioned explicitly somewhere earlier, as it is helpful for users of the product.

I clarified the version information at the beginning of Sect. 2.

L71: The algorithm descriptions in Sect. 2 and the evaluation results in Sect. 3 and Sect. 4 are based on GEMS glyoxal V2.0, which has been the operational product since 2023.

7. L194: Could some of the influence from the polluted background on the reference be eliminated by expanding the time averaging window of the reference radiance, and screening regions that are typically impacted by pollutant outflow? The generally higher retrieved columns in Fig. 7(a) are also what I would expect from the liquid water interference discussed earlier, as the larger reference sector is incorporating more of the open ocean water scenes to the east.

Following your suggestion, we conducted a sensitivity test to retrieve glyoxal VCDs using radiance references by excluding pixels possibly affected by pollution outflow. We used GEOS-Chem results to determine regions that are typically impacted by pollutant outflow. Figure R5 (a) shows regions in the reference sector that we exclude in radiance reference calculation. Panel b shows the absolute difference between glyoxal retrieved with updated radiance references and GEMS glyoxal V2.0. The updated glyoxal VCDs are, in average, 8.8% higher than GEMS glyoxal V2.0 VCDs. We also tested the sensitivity of our retrieval to expanding time averaging of our radiance reference calculation from three to five days. Panel c shows the absolute difference between glyoxal retrieved with five days mean radiance reference and GEMS glyoxal V2.0, which uses three days mean radiance reference. Glyoxal VCDs retrieved with five days mean radiance show uniformly lower values (NMB of -7.1%). Glyoxal VCDs shown in panel d illustrate the effect when the radiance reference is both screened and averaged for five days. The difference is positive north to 30°N and negative in other regions, and the normalized mean bias compared with GEMS V2.0 is 1.0%. Through the sensitivity tests, we confirmed that the current radiance reference that we use is impacted by pollution and this can be partly resolved by excluding regions that show high simulated concentration. Therefore, we plan to find the optimal criteria for screening polluted regions in radiance reference and apply them to GEMS glyoxal V3.0.
Using clean radiance references by excluding polluted regions and incorporating more open ocean as a reference region could contribute to increasing glyoxal VCDs. Two factors, however, results in increases in glyoxal VCDs in different latitude bands such that the first mostly affected regions north to 30°N, while the latter is more important south to 30°N.

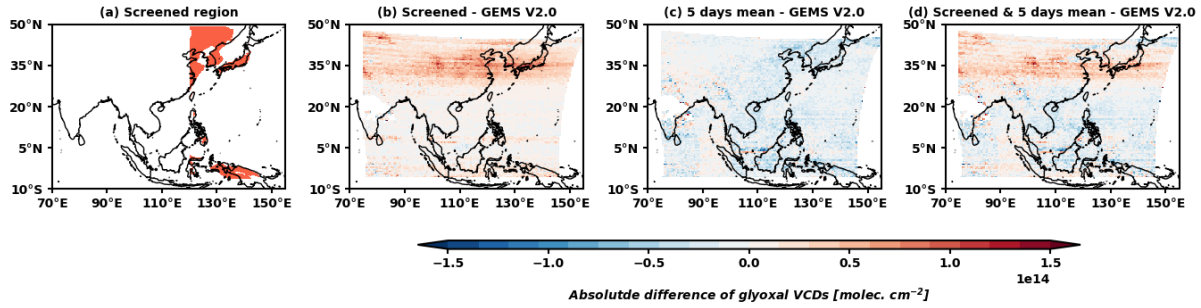

Figure R5. Regions in the reference sector that represent column-averaged mixing ratio of glyoxal higher than 1.2 ppt are indicated in red in panel a. The absolute difference between glyoxal VCDs and GEMS glyoxal V2.0 VCDs retrieved using (b) screened radiance reference, (c) five days averaged radiance, and (d) screened and five days average radiance in August 2020.

8. L222: It may be helpful to mention the magnitude of MAX-DOAS instrument-to-instrument biases to aid the interpretation of Fig. 8. Does CERES have some sort of side-by-side intercomparison?

There were no studies that quantified the magnitude of instrument-to-instrument biases in MAX-DOAS glyoxal retrieval as far as I found. However, Lerot et al. (2021) compared TROPOMI glyoxal columns with MAX-DOAS glyoxal columns from different institutions. We included the comparison result of MAX-DOAS and TROPOMI glyoxal VCDs and a notice about the difference in instruments as follows:

L215: Each institution uses different instruments and retrieval algorithms (fitting intervals, absorption cross-sections, etc.), leading to the possibility that instruments from each institution contain distinct systematic biases. Acknowledging this limitation, Lerot et al. (2021) compared TROPOMI glyoxal VCDs with MAX-DOAS glyoxal VCDs from different institutions. Compared with TROPOMI glyoxal VCDs, a mean difference of MAX-DOAS glyoxal VCDs from BIRA-IASB is -0.8×10^{14} molecules cm^{-2}, within the range of mean differences of MAX-DOAS glyoxal VCDs from CERES (-3.5×10^{14}–0.1×10^{14}). It is uncertain to confirm if there are significant systematic biases between the instruments. Rather, inconsistent biases across the stations could result from the different profiles and aerosol concentrations.

References

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