

Authors' response to comments from Anonymous Referee #3

General comments:

The Korean GEMS satellite is the first of a series of geostationary satellite instruments providing hourly observations of key air pollution species, including NO₂. These data are of large interest for air quality studies. As the current operational GEMS tropospheric NO₂ product still has some deficiencies, there is a need for improvements, and this manuscript is aiming at improving on that.

The approach taken in this study is to use the reprocessed PAL version of the TROPOMI NO₂ product to determine a pixel specific slant column offset of the GEMS data at TROPOMI overpass time, and to apply it to all GEMS measurements. The stratospheric correction is based on TM5 stratospheric VC data, again from the TROPOMI product, together with the diurnal variation taken from a GEOS-Chem run. Cloud correction and AMFs are computed using an updated version of the POMINO retrieval framework. The algorithm is applied to three months of data and the resulting columns compared to TROPOMI data, MAX-DOAS observations and in-situ surface measurements.

The manuscript is clearly written, covers a topic of interest to the AMT readership and reports on a relevant study. However, I have some concerns that the authors need to address before the manuscript can be accepted for publication.

We thank the Referee #3 for taking time to review our paper and for providing constructive suggestions and comments for improvement. We updated our POMINO-GEMS algorithm by replacing nested GEOS-Chem v9-02 derived stratospheric NO₂ VCDs with NASA GEOS-CF v1 derived stratospheric NO₂ VCDs, and reprocessed all retrievals. Updated validation results show great improvement in NO₂ diurnal patterns between POMINO-GEMS and ground-based MAX-DOAS measurements. We also use mobile-car MAX-DOAS measurements in the Three Rivers' Source region on the Tibetan Plateau to validate POMINO-GEMS retrievals, and good agreement is also shown in terms of NO₂ diurnal variation. Responses to the major and specific comments are provided below.

Major comments:

My main criticism about the paper is that the approach taken (correction of GEMS SCD data using TROPOMI retrievals) is a temporary solution at best. Clearly, problems in the GEMS SCD retrievals need to be solved in the spectral fit and not using an ad-hoc correction linking it to data from another satellite instrument. also, the assumption that all the problems in GEMS data can be described by a slant column offset determined at the time of TROPOMI overpass is probably not correct, as solar zenith angle and relative azimuth angle change over the day. Therefore, the most important measurement quantity of GEMS, the diurnal variation of NO₂, could be affected by the applied method.

It is also important to realize, that GEMS and TROPOMI data taken at the same time of the day do not have the same scattering geometry, and thus not the same AMF. The slant columns can therefore be different, even after geometric correction. These problems of the current approach need to be discussed in the manuscript.

Thank you very much for your comments.

(1) We agree that current approach to correct GEMS total NO₂ SCDs is a temporary solution at best, and the systematic problems in the official GEMS SCD retrieval should be solved by improving spectral fitting. As we are planning to learn and perform DOAS method to directly retrieve NO₂ SCDs, NO₂ SCDs calculated from this temporary solution can be a valuable reference to evaluate our product in the future. We have added more discussion on this limitation.

In Line 255-257, we added:

“We also note that our simple correction is a temporary solution before the aforementioned systematic problems in the official GEMS SCD retrieval are solved by improving spectral fitting.”

In Line 757-765, we added:

“To address the systematic overestimation and stripes problems in the original GEMS data, we correct GEMS total NO₂ SCDs by using TROPOMI data as a temporary solution. For example, we implement a simple geometric correction to combine GEMS and TROPOMI total NO₂ SCDs, but their differences in scattering geometry are only partly accounted for. Thus this correction works well in most regions, but may introduce SCD uncertainties up to 0.9×10^{15} molec. cm⁻² (20% – 30%) at the edge of the northwestern GEMS FOV. Currently, the Environmental Satellite Center of South Korea is updating the NO₂ SCD data to v2.0. We will update our POMINO-GEMS algorithm accordingly, once the updated official NO₂ product becomes available to provide necessary inputs for our research product.”

(2) The assumption of the correction of GEMS SCD data using TROPOMI retrievals is not clearly presented before, so we have added more discussion about the assumption in our geometric correction.

In line 237-241, we added:

“In Eq. (2), we implement a simple geometric correction (concerning SZAs and VZAs) for AMFs instead of using the actual AMFs; the latter could account for the differences in relative azimuth angles and other factors. Specific derivation of this assumption is given in Section 1 of the Supplement Information (SI). The correction is assumed to be acceptable with an extra uncertainty introduced to the total NO₂ SCDs, as will be further discussed in Section 3.5.”

In Line 680-683, we added:

“Given the assumption we made in adjusting GEMS total SCDs to match TROPOMI values, we tentatively estimate the error in our corrected total SCD data to be $0.5 - 0.7 \times 10^{15}$ molec. cm⁻² (10% in a relative sense) for most regions and 0.9×10^{15} molec. cm⁻² (20% – 30%) at the edge of the northwestern GEMS FOV.”

(3) It's true that TROPOMI-guided correction for GEMS total NO₂ SCDs could affect the diurnal variations of NO₂ from GEMS observations, so we have added additional comparisons and discussion about the diurnal variations of uncorrected GEMS NO₂ VCDs. The comparison results show that no significant influence on the diurnal variation of POMINO-GEMS tropospheric NO₂VCDs is brought in through TROPOMI-based correction for total NO₂ SCDs.

In Line 569-577, we added:

“As we use TROPOMI total NO₂ SCDs to correct those of GEMS, this may influence the NO₂ diurnal variation of original GEMS observations. Thus we also compare MAX-DOAS data with re-calculated POMINO-GEMS tropospheric NO₂ VCDs without correction in total SCDs (red dashed lines in Figure 9). Compared to our default POMINO-GEMS data (with correction), excluding the correction leads to lower diurnal correlation coefficients at Xuzhou, Hefei, Fudan University, Nanhui and Dianshan Lake, but higher correlation coefficients at Xianghe, Chongming and Fukue. Excluding the correction increases the NMB at three sites but decreases the NMB at five sites. We conclude that at these eight sites (in the eastern areas), no significant influence on the diurnal variation of POMINO-GEMS tropospheric NO₂VCDs is brought in through TROPOMI-based correction for total NO₂ SCDs.”

In Line 586-588, we added:

“In contrast, POMINO-GEMS without total SCD correction exhibits much poorer correlation with mobile-car MAX-DOAS data, due to the erroneous increase in the afternoon.”

In Line 639-640, we added:

“Note that the consistency between POMINO-GEMS and MEE data does not depend on the total SCD correction (Table S4).”

My second concern is about the comparison of GEMS and TROPOMI data shown in the manuscript. As GEMS slant columns are forced to agree with TROPOMI data, this comparison makes little sense and only shows that no technical mistake was made. The only comparisons providing additional information are those to external data.

Thank you for your comment and suggestion. The discrepancies between POMINO-GEMS and POMINO-TROPOMI v1.2.2 tropospheric NO₂ VCDs are caused by differences in both tropospheric NO₂ SCDs and AMFs. We have added detailed discussion in Section 3 of the SI. Besides, we have also added the comparison results between POMINO-GEMS and independent OMNO2 v4 and GOME-2 GDP 4.8 tropospheric NO₂ VCDs.

In Line 496-506, we added:

“Figure 7d-f and g-i show the comparison results of POMINO-GEMS tropospheric NO₂ VCDs with OMNO2 v4 on a 0.25° × 0.25° grid and GOME-2 GDP 4.8 on a 0.5° × 0.5° grid averaged over JJA 2021, respectively. POMINO-GEMS NO₂ VCDs exhibit good spatial consistency with the two independent products ($R = 0.87$ and 0.83), although with slightly lower values than OMNO2 v4 (by 16.8%) and GOME-2 GDP 4.8 (by 1.5%). These VCD differences are expected, considering the differences in the retrieval algorithm. For example, the POMINO-GEMS algorithm implements explicit aerosol corrections in the radiative transfer calculation, while OMNO2 v4 and GOME-2 GDP 4.8 treat aerosols as “effective clouds”. POMINO-GEMS accounts for the anisotropy of surface reflectance by adopting MODIS BRDF coefficients, whereas OMNO2 v4 and GOME-2 GDP 4.8 use geometry-dependent and regular LER, respectively. The horizontal resolution of a priori NO₂ profiles in POMINO-GEMS is 25 km (and interpolated to 2.5 km), 1° × 1.25° in OMNO2 v4 and 1.875° × 1.875° in GOME-2 GDP 4.8.”

My third point is, that the uncertainty discussion is very superficial and in my opinion not correct. The

SC uncertainty should be driven by shot noise and therefore be described as an absolute, not a relative uncertainty. The overall uncertainty of $0.2E15$ molec/cm² derived for the tropospheric SCDs appears very low, but it is anyway not clear if this is the uncertainty for an individual GEMS measurement, a monthly average, or the three monthly average discussed here. this discussion needs to be improved.

Thank you very much for your comment. We have re-written Section 3.5 to improve the discussion about the error estimates. All uncertainties discussed here are for the summertime retrieval. For the uncertainty of NO₂ slant columns, we have discussed it both using an absolute value and in a relative sense. The relative uncertainty of NO₂ SCDs is used for following estimation of relative uncertainty of tropospheric NO₂ VCDs. We agree that the overall uncertainty for tropospheric NO₂ SCDs is underestimated, and updated analysis has been added in the revised manuscript.

In the data availability section, it is stated that the data is available through <http://www.pku-atmos-acm.org/acmProduct.php/>. This does not appear to be the case and data could therefore not be checked for this review.

Thank you very much for your comment. At first, we were processing the retrieval data beginning in December 2020 and hadn't upload them online. Since we updated our retrieval algorithm by using NASA GEOS-CF v1 product, we now are reprocessing all the data and will upload them for public use as soon as possible.

We have changed the data availability statement to “The POMINO-GEMS NO₂ data will be freely available soon at the ACM group product website (<http://www.pku-atmos-acm.org/acmProduct.php/>).”

Minor comments:

Line 188: Isn't the current GEMS NO₂ product provided at 3.5×8 km²?

Yes, the current GEMS NO₂ product is provided at 3.5×8 km², but the spatial resolutions of other trace gases are different. Therefore we decide to quote the statement in Kim et al. (2020) to generally describe the spatial resolution of GEMS products.

Line 128: How does the known GEMS uncertainty in irradiances affect the reflectances and thereby cloud retrievals?

The uncertainties in the measured radiances at the top of atmosphere and extraterrestrial solar irradiances can directly affect the cloud fraction retrieval, and also be propagated to the uncertainties of DOAS-fitted continuum reflectances and O₂-O₂ SCDs used for the inversion of cloud-top pressure.

Currently we don't exactly know the uncertainty in radiances measured by GEMS instrument. In our POMINO-GEMS algorithm, we re-retrieve cloud parameters in order to assure the consistency of ancillary parameters used for cloud and NO₂ retrieval, such as aerosol optical parameters and surface reflectance.

Line 262: this ad hoc factor needs to be mentioned again when later comparing the retrievals with the in-situ observations.

Thank you for your suggestion. We have added more discussion about the limitation of the ad hoc factor in section 3.4 in the revise manuscript.

In line 606-608, we added:

“These differences reflect errors in POMINO-GEMS NO₂ VCDs, in the conversion from tropospheric VCDs to surface concentrations, and in MEE data (due to potential contamination by nitric acid and organic nitrates (Liu et al., 2018)).”

In Line 631-639, we added:

“The discrepancies between POMINO-GEMS and MEE surface NO₂ concentrations at different hours are likely caused by the assumed constant correction factor of 2 to account for the vertical gradient of NO₂ from the height of ground instrument to the center of the first model layer (Section 2.2). In the morning when the PBL is low, most NO₂ molecules are near the ground and the vertical gradient of NO₂ over polluted regions is the largest in the daytime, so the factor of 2 may lead to underestimation of derived surface NO₂ concentrations. In contrast, in the afternoon, the PBL mixing is much stronger and the vertical gradient of NO₂ is much smaller, thus the factor of 2 may lead to overestimated surface NO₂ concentrations.”

Line 277: please provide a bit more information on this – how many data points were excluded? What exactly were the criteria?

Thank you for your suggestion. We have added more information about the Grubbs statistical test, and have also shown the comparison between the original data and those after excluding outliers in Figure S7.

In line 357-361, we added:

“The Grubbs statistical test, which is used to detect outliers in a univariate data set assumed to exhibit normal distribution (Grubbs, 1950), is performed to exclude outliers in both MAX-DOAS and satellite data before comparison. Only one data pair from Fudan University site is identified as an outlier and removed (Figure S7), and we get 1348 matched hourly data pairs in total.”

Figure 4: What are the regions shown in grey in the figure? Are these negative values of missing data?

The regions in grey mean there are no GEMS observations or valid retrievals in June-July-August 2021. They are either because of the spatial limitation of GEMS FOV, or because the pixels are excluded due to the quality control criteria. We have added the note in the caption of all corresponding figures.

Figure 6 / Line 323: I do not find the discussion of the observed increase in NO₂ convincing. The observed changes are large and have clear patterns, and I suspect they are retrieval artefacts.

Thank you very much for your comment. Previous studies have discovered the NO_x emissions from natural sources such as soil and lakes. After using more reasonable stratospheric NO₂ information from GEOS-CF v1 product, the increase in NO₂ over this region is still evident, so we believe it is hardly a

retrieval artefact. We will further explore this issue in the future.

Figure 7: As discussed above, the only surprise with this figure is that the agreement is not even better.

Thank you for your comment. We have added detailed analysis for the differences between POMINO-GEMS and POMINO-TROPOMI v1.2.2 tropospheric NO₂ VCDs in Section 3 of the SI.

Besides, we have also compared POMINO-GEMS tropospheric NO₂ VCDs with those of external OMNO2 v4 and GOME-2 GDP 4.8 products. Comparison results have been shown in the reply to the second major comment.

Line 433: Maybe mention that the main difference between column and surface concentrations is that the column is insensitive to boundary layer height changes.

Done.

Line 443: Please provide information on for what the uncertainty calculations are made – individual measurements or averages?

Our uncertainty analysis is for the general summertime retrieval.

Line 448: why should SCD have a relative uncertainty?

Thank you for your comment. According to previous studies, the SCD uncertainty can both be described using an absolute value and in a relative sense. Here, we use the relative uncertainty of NO₂ SCDs to estimate the relative uncertainty of tropospheric NO₂ VCDs. We have updated our discussion about the SCD uncertainty in the revised manuscript.

In Line 676-683, we added:

“As described in Section 2, we calculate hourly total SCDs based on the original GEMS SCD data and daily TROPOMI-guided corrections. According to the GEMS ATBD of NO₂ retrieval algorithm, the SCD errors from the DOAS method are < 5.65% at high-NO₂ conditions (NO₂ VCD > 1 × 10¹⁵ molec. cm⁻²) (Lee et al., 2020). The NO₂ SCD errors of TROPOMI are reported to be 0.5 – 0.6 × 10¹⁵ molec. cm⁻² (10% in a relative sense) (Van Geffen et al., 2022). Given the assumption we made in adjusting GEMS total SCDs to match TROPOMI values, we tentatively estimate the error in our corrected total SCD data to be 0.5 – 0.7 × 10¹⁵ molec. cm⁻² (10% in a relative sense) for most regions and 0.9 × 10¹⁵ molec. cm⁻² (20% – 30%) at the edge of the northwestern GEMS FOV.”

References:

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