

Response to Reviewer #1

We thank the reviewer for his/her constructive comments and suggestions to improve the quality and clarity of our manuscript. We have made major and careful modifications to the original manuscript according to all the comments and suggestions from the reviewers. The major modifications include:

- (1) We have changed to using a fixed CO profile as the a priori for all the retrievals, and make changes to all relevant figures. The use of a fixed a priori make it easy to interpret and compare with models and detect anomalies such as wildfires emissions. Specifically, for our purpose of retrieving the diurnal changes of CO columns, the main topic of this study, a fixed a priori is preferred because any significant perturbation to the constant a priori, which does not change diurnally, may indicate information that is retrieved from the observed spectra;
- (2) We removed the 0-1km results and replaced it with the bottom 3-layer, which ranges from the surface to 3km above sea level. In addition, we added North India as the 4th representative region in the analysis, besides North China Plain, Mongolia, and East China Sea.
- (3) We added diurnal cycle comparison of the retrieved CO columns and DOFS, boundary layer height from ERA5 reanalysis, and model simulations of CO columns from CAMS EAC4 reanalysis. All time in the revised manuscript has been changed to Beijing Time (UTC +8).
- (4) A detailed comparison between our retrievals and IASI has been added, including spatial and temporal comparisons. We showed that the DOFS between GIIRS and IASI are comparable, and the vertical sensitivities, as quantified by the AK matrix, are also similar between GIIRS and IASI.
- (5) We added in the Discussions section about the applicability of the algorithm in retrieving CO using GIIRS observation in the winter season, and used December observations as examples.

Item-by-item responses to the specific comments are provided below, in which the reviews' comments are in **blue**, our responses in **black**, and modifications of the original manuscript are indicated by highlight in **yellow** in the revised manuscript.

Review of Zeng et al., 2022

This paper describes CO retrievals from the geostationary hyperspectral infrared sounders GIIRS onboard FY-4B. This is the first publication presenting CO retrievals from a geostationary platform that could be valuable to document the diurnal cycle of this species in the lower troposphere. The paper is correctly written and structured with some interesting information. It is therefore adapted to AMT.

Nevertheless I have some important concerns and questions that have to be adressed before publication. My most important concern is about the diurnal cycle itself which is

the main topic of the paper and included in its title. It should be better documented and compared to other datasets to be validated at minima.

Thank you for your positive comment and constructive suggestions. We have made major changes related to the analysis of the diurnal cycle of CO column retrieval, including (1) We have added Fig. 8 and Fig. 10 to compare the diurnal cycles of the retrieved CO columns, DOFSs from the retrievals, PBLH from ERA5 reanalysis, and model simulations from ECMWF EAC4; (2) In Fig. 13 and Fig. 14, we compare our CO column retrievals with IASI retrievals, especially the day-night contrast of CO columns; (3) In our synthetic experiment as shown in Fig. 4, we separated the results into hours to investigate the accuracy of the retrieval algorithm in reproducing the diurnal change of CO columns; (4) In Section 6.3, the DOFS for the total column and the bottom 3-layer are shown separately to examine how the retrievals are sensitive to the total column and the lower atmosphere. For details, please see our responses to your specific comments below.

CO a priori profiles :

The advantages and inconvenients of a climatological a priori are mentioned in section 7.1 and the use of a single a priori as a way to improve the algorithm to detect anomalies. It should also be mentioned that using such an a priori makes the retrievals more complicated to interpret and to use for model validation.

The use of a 3 hourly profile climatology based on 5 years simulation is done to help to provide the correct diurnal cycle to the retrieval algorithm. But CO is a pollutant with a lifetime much larger than a day. The daily cycle for CO is not as important as for NOx. The authors should provide the plots of the daily variations of CO in the troposphere and lower troposphere in the 3 selected regions from the ECMWF CAMS for instance together with the surface and bottom air temperatures in Fig 2.

Figure 4 shows that the same low biases for high concentrations in the a priori are partially kept in the retrieval for North China Plain and Mongolia. The disappearance of these biases when the AvKs are applied to the « true » profiles clearly indicates that these biases are linked to the a priori and the lack of sensitivity of the sensor/retrieval to the polluted BL. As stated by the authors, this problem could be related to the too tight a priori covariance matrix used with the climatological profiles but it is not sure. It would be very interesting to provide some results from a simple test using a single a priori profile and its more loose a priori covariance to verify this assumption. In that case the signal to noise ratio for the retrieval which has been tuned according to the a priori covariance matrices (section 6.1) should be lowered which could lead to a destabilisation of the retrieval and possible oscillations in the profiles.

We thank the reviewer for the very constructive suggestions. We have made substantial revisions according to your suggestions:

- (1) In the revised manuscript, we changed the a priori from time-varying profiles to a fixed CO profile derived from model simulations. The reason, as you mentioned, is that a time-varying a priori makes the retrieval results more complicated to interpret and to use for model validation. Also, the small errors with the time-varying a priori profiles also make it difficult to detect anomalies from unexpected events such as wildfire emissions. For our purpose of retrieving the diurnal changes of CO columns, the main topic of this study, a fixed a priori is preferred because any significant perturbation to the constant a priori, which does not change diurnally, may indicate information that is retrieved from the observed spectra. The updated a priori CO profile and its covariance matrix are shown in Fig. (2). As expected, the DOFSs are higher for the retrievals using fixed a priori (with a larger variability) compared to the time-varying profiles. Compared to IASI, we found our results show a very good agreement for the urban source regions as well as natural wildfire emissions, as shown in Fig. 13 and Fig. 14 in the revised manuscript;
- (2) The averaged daily cycle of CO columns and the ground-level CO concentration are shown in Figure 10(c) and the supplementary Fig. S6. They are used to compare with the diurnal change of the retrieved CO columns, and DOFS from the retrieval and the boundary layer height as shown in Fig. 8 and Fig. 10, respectively.
- (3) Using the fixed a priori CO profile, we have repeated the simulation experiment and updated Fig. 4. We found that in the daytime when DOFS is large the retrieval results have a good agreement with the assumed “truth”. This suggests that a single a priori profile with a loose a priori covariance used in this study (Fig. 2) improves the retrieval results compared to previous results using the time-varying profiles but with a tight a priori covariance.

Related statements have been added to the revised manuscript in Section 3.2, Section 5, and Section 6.3.

What diurnal cycle ?

The problem to document the CO diurnal cycle with the GIRS retrievals come from the fact that it could be linked to :

- the real CO cycle that is the objective
- the variability of the BL layer with probably a better detection of pollution in the afternoon when the BL is higher where the sensor is more sensitive
- the variability of the DOFS

In order to disentangle these different sources of diurnal CO cycles

- the diurnal cycles of CO total columns over the 3 selected zones should be provided clearly the same way as the DOFS in Fig 8. The plot of the DOFS for the 0-1 km could be

removed as the retrieval for this layer provides no relevant information (see next comment) and as its diurnal variability is mostly similar to the total column.

- the diurnal cycle of the BL height could also be documented from ECMWF ERA5 data for instance.

- the diurnal cycle of CO from other sources such as local pollution networks in China/ Beijing area, ECMWF CAMS used for the a priori (see above) and some references to relevant publications should be provided to check whether or not GIIIRS retrievals are sensitive to BL pollution diurnal variability.

Thank you for your thoughtful and very constructive suggestions. We have made the following revisions according to your suggestions:

- (1) In Section 6.3, we added what you have just mentioned to explain what may drive the diurnal changes of the CO column retrievals: **“The diurnal changes of CO from FY-4B/GIIRS retrievals may be linked to (a) the real CO variabilities, the objective of this study, driven by emissions and transport, and (b) the change in the detectivity by the instrument as reflected in the DOFS from the retrieval algorithm, which can be affected by the change in TC and CO concentration.”**
- (2) As an attempt to disentangle these different factors, in Fig. 8, we show the diurnal changes of the DOFS for both the total column and the bottom 3-layer (from the surface to 3km a.s.l.), the CO columns from retrievals, the boundary layer height (BLH) from ECMWF reanalysis data, and the ECMWF CAMS EAC4 CO simulations over the 4 selected regions. The 0-1km figures have been removed in the revised manuscript. The comparison results suggest that a direct interpretation of the authentic diurnal column variabilities from the retrieved CO columns is challenging given the entangled effects of the real CO changes and the variable detectivity. A better solution is to use mode assimilation (e.g., EAC4) that takes into account the retrieved CO profile and the vertical sensitivity in order to disentangle the different contributions. Please see the details in Section 6.3 in the revised manuscript.
- (3) Unfortunately, the most recent data from local measurement network are not available for the public yet. Instead, we use model simulations from CAMS EAC4 that has assimilated satellite observations (IASI and MOPPIT) for the comparison, as shown by the CO column in Fig. 9(c) and the ground-level CO concentration in Fig. S6. From the model simulations, which has assimilated satellite observations of IASI and MOPPIT, we see the change in total columns in all selected regions are very small (less than 2% on average) which can be primarily attributed to the diurnal change in BLH. In the supplementary Fig. S6, the model simulated ground-level CO concentrations show a much larger variation compared to the CO columns. However, model simulations from EAC4 have large grids ($0.75^\circ \times 0.75^\circ$) and low temporal resolution (3-hour) which are not sufficient to resolve the local CO changes that are usually measured over the urban centrals. To the first order, the change in the ground-level CO concentration averaged

over the selected region is primarily driven by the BLH change, and the traffic emission peaks are not discernable from the time series.

- (4) Moreover, we collected information about the CO diurnal cycle in China and India from published papers. Please see our response to your next comment.

I have some doubts about the diurnal variability displayed in fig 10 and 11 in the NCP : the maxima are detected between 16 and 22 UTC that is between midnight and 6AM Beijing time (If I understood correctly). So it does not correspond to the time of day (i) with the highest activity where we expect the largest emissions and concentrations (this should be highlighted by surface /CAMS data as proposed above) (ii) with the highest BL which is in the afternoon (iii) with the largest DOFS which is the beginning of the afternoon (see Fig 8 and Fig 10 b). The authors have to provide some explanations about this peculiar diurnal cycle.

A typical diurnal change of ground-level CO, like what you described, is directly affected by the diurnal emission pattern from traffic besides BLH change. In urban regions, ground level CO concentrations from in-situ ground-based observations show a distinctive double-peak diurnal cycle corresponding to the morning and evening traffic rush hours. In the morning, the increase in traffic emissions results in the morning peak; As BLH gets larger, air dilution takes place and the concentration drops; In the evening, traffic emissions increases while the BLH quickly decreases which results in an evening peak. These diurnal patterns have been observed across many cities in Asia (e.g., Ran et al., 2009; Chen et al., 2020; Meng et al., 2009; Verma et al., 2017).

However, the diurnal changes of CO columns and the surface layer concentration are not expected to be the same. As shown in Stremme et al. (2009), which retrieved diurnal CO column changes in the Mexico City using ground-based solar and lunar infrared spectroscopy, found that the diurnal changes in the total CO column and the surface level concentration can be very different. The total CO column within the city presents large variations with contributions from urban CO emissions at the surface and the transport of cleaner or more polluted air masses into the study area.

The diurnal cycle of the retrieved CO columns from FY-4B/GIIRS, shown in Fig. 10(a), presents impacts by the diurnal change of DOFS. Noted that the a priori CO columns averaged for the year of 2021 are 0.038, 0.028, 0.039, 0.036 mole/m², respectively, for North China Plain, Mongolia, East China Sea, and North India. Since the DOFS for the nighttime is low, especially for the lower atmosphere, the nighttime column retrievals generally tend to close to the a priori columns, resulting in a bow shape. In the daytime, the CO column retrievals are well constrained by the observations. Since the summer time CO is generally lower than the yearly mean (Chen et al., 2020) based on CO's seasonal cycle, we see the column retrievals averaged in July are generally lower than the a priori column value which is derived from annual mean of model simulations. These results suggest that a direct interpretation of the authentic diurnal column variabilities from the retrieved CO columns is challenging given the entangled effects of the real CO changes and the variable detectivity. A better solution is to use mode assimilation (e.g.,

EAC4) that takes into account the retrieved CO profile and the vertical sensitivity in order to disentangle the different contributions.

We have added the above statements in the revised manuscript, please see Section 6.3.

Why 0-1 km layer ?

The 0-1 km could be interesting to document BL pollution but it is characterised by a very low DFS mostly below 0.1 to 0.15 (Fig 3 and 9) and below 0.125 on Fig 8 which means that there is almost no information about this layer in the retrieval whatever the thermal contrast. DOFS is even negative (what does that means?) in Fig 8 and 9 showing that this layer is absolutely not a good choice.

The sentence line 400 « The DOFS can be as large as 0.3 providing a strong constraint on the bottom 0-1 km » is a flagrant overstatement (just a couple of points at 0.3 in Fig 9!!!) and should be removed or changed. Even a DOFS of 0.3 would have meant that the information for this layer is low.

In Figure 7 that displays the AvKs we see that the AvKs peak at 800 or 700 hPa in the best cases.

I therefore do not see the relevance to display results about the 0-1 km layer in the different figures. As the DOFS for the total columns are roughly between 0.8 and 1.2, the authors should separate the atmosphere/troposphere in the two layers in which the information is equally provided and display results for the lowermost of those layers.

Thanks for your thoughtful suggestions. We have removed the 0-1km results and replaced with results of the bottom 3-layer, which ranges from the ground to 3km above the sea level. As shown in Fig. 8, in the daytime, the bottom 3-layer has a DOFS that is about half of the total column. This separation of the upper and lower atmosphere also corresponds to the peak sensitivity in Fig. 7 as you mentioned. Related changes have been made in Fig. 3, Fig. 8, Fig. 9, Fig. 10 and Fig. 11. We have also removed the statement that overstate the DOFS.

Comparisons with IASI :

The comparison with IASI data is made to partly validate the diurnal variations but some important information is missing :

- as there are only two overpasses of IASI daily at 9:30 LST AM and PM, the authors have to detail how they average the GIIRS data temporaly which is not clear at all.

- the correlation coefficients and rmse are given in the Fig 12 but a table with those figures and other basic statistics such as mean biases +/- rmsd should be added.

- the comparison methodology to smooth IASI with GIIRS AvKs is assuming that IASI has a much better vertical resolution than GIIRS which is not the case (IASI has probably a DOFS of 1.5). In that sense it is worth to display IASI AvKs to compare with GIIRS (as in Fig 7) and to provide IASI's DOFS. It would probably be better to avoid to apply equation 11 assuming that both sensors have similar vertical sensitivity or to use the more (too) complicated methodology detailed in Rodgers and Connor, JGR (2003).

- IASI CO "diurnal cycle" is mostly related to its decreased sensitivity at night. So the agreement with GIIRS for day and night described as good by the authors is just indicating that GIIRS has the same decreased sensitivity at night. There should be some statements about this issue.

Thank you for your great suggestions. We have made changes related to the comparison with IASI:

- (1) Besides the comparison of daily time series in July, we have added Fig. 13 and selected July 07 as an example to compare the spatial distribution between GIIRS and IASI, which shows a good agreement between the two datasets.
- (2) We have added more details about how the comparison was made. For the spatial comparison, we added **“For the spatial comparison, we use the daytime and nighttime observations on July 07, 2022 as examples. The observation hours of IASI (Supplementary Fig. S7) over the North China Plain are about 9.7h BJT for the daytime and 21h BJT for the nighttime, which correspond to the GIIRS measurement cycles of 10-11h BJT and 20-21h BJT, respectively, for the daytime and nighttime. Because the GIIRS and IASI have different footprints and grid configurations, it is not straightforward to make point-by-point comparisons. We re-grid the retrieval data into 0.5°×0.5° grids and then make comparisons of the collocated grid data points.”** For the time series comparison, we added **“For comparing the time series of GIIRS and IASI, we focus on the four representative regions (North China Plain, Mongolia, East China Sea, and North India) and compare the regionally averaged CO total columns between GIIRS and IASI. The data processing is similar to the spatial comparison. First, the retrieval data are re-gridded into 0.5°×0.5° grids and the comparisons are made using the collocated grid data points to avoid bias related to uneven distribution of CO data points within the selected region. Then, according to the averaged observation hour of IASI in a specific region, the temporally closest measurement cycle of GIIRS retrievals is used. Finally, the GIIRS CO retrievals are also adjusted to the IASI a priori following Equation (6). The comparison results of daily averaged CO columns are shown in Fig. 14. The statistics of the comparisons are shown in the Supplementary Table S1.”**
- (3) Supplementary Table 1 is added to provide statistics of the comparison, including correlation coefficient, RMSE, mean bias and the error standard deviation.

- (4) Since we have used a fixed CO profile as the a priori, we have re-evaluated the comparison using a priori adjustment and AK smoothing. Our results show that GIIRS retrievals with a fixed CO profile as the a priori have sensitivity (reflected by the AK diagonal) that is similar to IASI (Appendix Fig. A2). We therefore only apply the a priori adjustment to the GIIRS data using the IASI a priori, but not the AK smoothing assuming that the two sensors have similar vertical sensitivity as you mentioned.
- (5) We have added the statement to Section 6.4: **“Noted that in the nighttime, both instruments have weak sensitivity to the bottom atmosphere, the agreement may just indicate that GIIRS and IASI have the same decreased sensitivity at nighttime.”**

For details, please see Section 6.4 in the revised manuscript.

Detailed comments :

Section 3 and 4 :

Some generalities about radiative transfer and retrieval methodology and basic known equations could be removed. Equations 3 to 8 have been largely documented such as in Rodgers (2000) and it is unnecessary to repeat this here.

We have removed Equations 3, 5, 6 and 7, but keep Equations 4 and 8 as they are important to the description of the observation characteristics and the settings of the retrieval algorithms.

Time : the time is given in UTC but it is not a correct choice to interpret diurnal cycles around China. Beijing local solar time would be much better. Furthermore the time is often given without the precision that it is UTC.

We have changed the time used in the paper to Beijing Time, which is UTC+8.0.

Figure 7 : precise the time system chosen.

We have changed it to Beijing Time (UTC+8)

Figure 8 : we suppose it is UTC !

We have changed it to Beijing Time (UTC+8)

Figure 10 : please provide hour in LST because UTC is not adapted to the geographical zone.

We have changed it to Beijing Time (UTC+8)

Fig 12 : Dayth => Day

Changed.

Line 59 : The authors mention Kobayashi et al. (1999) as one of the first attempt to document CO from space with the japanese IMG ADEOS. Nevertheless, this paper do not present CO retrievals from IMG. The only retrievals of CO from this first spaceborne IR FTS have been published later by Barret et al. (2005).

Thanks. We have replaced “Kobayashi et al. (1999)” by “Barret et al. (2005)”, and Kobayashi et al. (1999) is move to be referenced for IMG.

refs:

C.D. Rodgers and B.J. Connor, Intercomparison of remote sounding instruments, JGR, 2003.

B. Barret, et al., Global carbon monoxide vertical distributions from spaceborne high-resolution FTIR nadir measurements, ACP, 2005.

We have added the second reference. The first one was already cited in the original manuscript.