

We thank the reviewers for their thoughtful and detailed comments. We have revised this manuscript based on the comments. Here are the major changes in the revised version:

- The title was changed to: “Discrepancy in assimilated atmospheric CO over East Asia in 2015-2020 by assimilating satellite and surface CO measurements”.
- Table 2 was updated to provide more detailed validations for the assimilation effects.
- Figures 6 and 7 were simplified.
- The surface CO observations and Kalman Filters were re-processed by generating grid "super-observation" instead of grid average. We didn't find a noticeable discrepancy between the applications of super-observations and grid averages. The unit of column CO was converted from molec/cm<sup>2</sup> to column-averaged dry-air mole fractions ( $X_{co}$ ).

Below we respond to the individual comments.

Reviewer #1:

The paper evaluates the effects of satellite and surface measurements on atmospheric carbon monoxide assimilation over East Asia. It is well-written and can provide valuable information for numerical studies on atmospheric carbon monoxide. I recommend the publication of the paper after the following issues have been addressed. The authors showed the underestimation of the modeling on surface carbon monoxide by comparing the a priori simulation with observations in the first two Figures. However, in the latter part of the paper, how much will the data assimilation with different types of carbon monoxide measurements improve the simulations is not evaluated. The authors can add some statistical metrics in the examinations, for example, the correlation coefficient and absolute bias.

**Answer:** Thank you for the comments! Table 2 was revised to provide more detailed validations for the assimilation effects. We find the assimilations led to significant improvement in assimilated CO concentrations:

- Surface CO: the a priori, a posteriori (by assimilating raw MEE CO observations) and MEE CO observations are 397, 631 and 781 ppb over E. China in 2015-2020.
- Column CO ( $X_{co}$ ): the a priori, a posteriori (by assimilating MOPITT CO column data) and MOPITT CO observations are 97.4, 124.1 and 128.6 ppb over E. China in 2015-2020. Here the modeled CO profiles were smoothed with MOPITT averaging kernels.

Reviewer #2:

**General comments:**

This manuscript presents results from data assimilation experiments using surface CO observations and satellite CO retrievals. The assimilation system is based on GEOS-Chem

model and Kalman Filter approach. The CO analysis fields from two data sources are compared and discussed. The authors found the discrepancy between assimilating surface observations and satellite retrievals and concluded that this discrepancy reflects the different vertical sensitivities of satellite and surface observations to CO concentrations in the lower and free troposphere. The comparative study of assimilating surface CO observations and satellite CO retrievals is of interest to inverse modeling and air quality forecasting communities. However, the manuscript in the current shape seems to me incomplete and lacks the scientific depth. The evaluation of both the efficiency of assimilation method adopted and the accuracy of analysis CO fields is not given in the manuscript, making the results and conclusions less convincing. The differences are analyzed and discussed in details, but what they imply for the better use of both surface observations and satellite retrievals and what strategy could be used to improve the assimilation is not provided. More detailed analysis and extra experiments are required to reach robust conclusions which can truly contribute to a better understanding of assimilating CO observations.

**Answer:** Thank you for the comments! The manuscript has been revised based on the comments.

**Specific comments:**

**Question:** Line 1: This title is too general and should be specific to what you found or what you conclude.

**Answer:** Thank you for this suggestion! The title has been changed to: “Discrepancy in assimilated atmospheric CO over East Asia in 2015-2020 by assimilating satellite and surface CO measurements”.

**Question:** Line 21-29: Why use molc/cm<sup>2</sup> as the unit for column CO other than ppb the same as that for surface concentration? Suggest to use the same unit for column and surface CO. Only the inconsistency of column CO is emphasized. Does that mean your study focus on the investigation of column CO?

**Answer:** The unit of CO column has been converted to column-averaged dry-air mole fractions ( $X_{CO}$ ).

As the reviewer suggested, the abstract has been revised to include a discussion about surface level CO: “the adjusted surface CO concentrations are about 631, 806 and 657 ppb by assimilating surface CO measurements, in contrast to 418-427, 627-639 and 500-509 ppb by assimilating MOPITT CO observations over E. China, NCP and YRD, respectively”.

**Question:** Line 74-79: The logic here is not complete. The authors provided descriptions of literatures on assimilating surface CO observations and satellite CO retrievals and pointed out the unsolved problems in the fields. But it is still not clear why the comparative study could help us get closer to solving the exiting problems in assimilating CO observations.

**Answer:** Thank you for pointing out this point! The discussion has been revised: “However, comparative analyses to investigate the effects of satellite and surface CO measurements in data assimilation systems are still lacking, which poses a significant

barrier to integrating the information provided by satellite and surface measurements in data assimilation applications”.

**Question:** Line 103-109: How is the quality of MEE CO measurements? The information on the accuracy of those measurements should be provided and discussed.

**Answer:** The manuscript has been revised: “Concentrations were reported by the MEE in units of  $\text{mg}/\text{m}^3$  with a precision of  $0.001\text{mg}/\text{m}^3$ ”. “To ensure the reliability of the data before assimilation, we screened the data on the numerical range and time range. In the first step, we removed data with CO concentrations larger than 6000ppbv ( $\sim 7.5\text{mg}/\text{m}^3$ ), and the selection of this empirical value is relatively close to the  $7\text{mg}/\text{m}^3$  selected by Feng et al. (2020). In the second step, to ensure the rationality of the daily variation of the assimilation results, we eliminated 327 sites with missing data for more than 14 consecutive days, accounting for 19.5% of the total number of sites”.

**Question:** Line 132: The ensemble Kalman Filter and 4D-Var are commonly implemented assimilation techniques to account for the complexity of atmospheric transport and chemistry. I doubt the capability of simple scheme such as sub-optimal Kalman Filter to assimilate CO observations efficiently. The authors should provide evidence on the legitimacy of adopting sub-optimal Kalman Filter approach. BTW, the authors should provide more detailed description on assimilation setup such as initial conditions, DA time window, and how the forecast steps are advanced, etc.

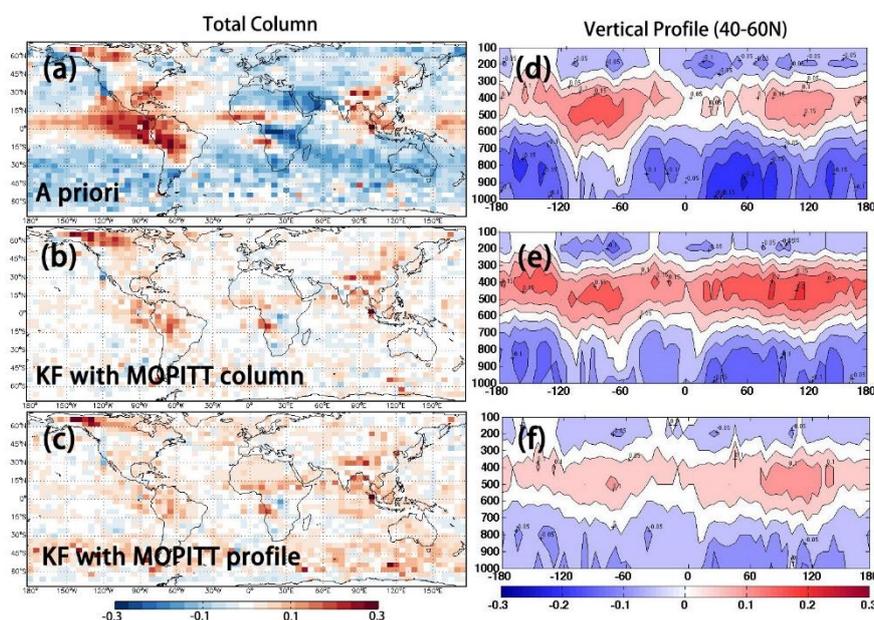
**Answer:** As shown in the revised manuscript: “The sub-optimal Kalman Filter has been applied in previous studies to provide quick optimization for initial and boundary atmospheric CO concentrations (Jiang et al., 2015; Jiang et al., 2017). Han et al. (2022) further provided a comparative analysis between sub-optimal Kalman Filter and a hybrid deep model (DL) to predict surface CO concentrations in China in 2015-2020, and found the good performance of Kalman Filter in respect to independent observations. We note that the optimization effect of sub-optimal Kalman Filter is expected to be weaker than more complicated methods such as Ensemble Kalman Filter, particularly because the latter can optimize CO emissions and concentrations simultaneously (Miyazaki et al., 2017; Feng et al., 2020), for example, Ma et al. (2019) indicated that updated anthropogenic emissions led to improved CO forecast by about 10% during the first 36 hour of forecasts”.

“The assimilations were started on July 1 2014 by assimilating MOPITT or MEE CO observations to produce optimized initial conditions on Jan 1 2015. The modeled CO concentrations are compared with observations and updated hourly, based on Eq. 2, and then forwarded to Eq. 1 for the model simulations in the next time step, i.e., the assimilation window is one hour”.

**Question:** Line 157-157: Why profile and column data were used to produce two types of boundary conditions? As stated in the later paragraph, the differences between two approaches are small, why not just stick to results from just one approach to do the analysis?

**Answer:** The attached figure shows the relative differences between a priori simulation and Kalman Filter assimilation in respect to MOPITT CO observations from our previous analysis. The sub-optimal Kalman Filter by assimilating MOPITT column data (panel b) and MOPITT profile data (panel c) led to similar improvements on CO

columns. However, the adjustments on CO vertical profiles are very different (panels e-f), i.e., similar biases in CO columns cannot ensure similar biases in vertical profiles. Consequently, it is helpful to produce different CO boundary concentrations for different datasets.



**Question:** Line 170-173: Since the boundary conditions were produced by assimilating MOPITT CO observations using a global assimilation system, theoretically, assimilating the same MOPITT data over a regional domain are supposed to produce more details on CO field spatially and temporally and there should not be relatively large difference for regional averaged column CO. So where is this marked enhancement coming from?

**Answer:** The a priori CO concentrations were produced with original boundary condition without optimization. The caption of Table 1 was revised to clarify this point: “The E. Asian CO boundary conditions are provided by global a priori simulations, except Kalman Filters by assimilating MOPITT CO while the boundary conditions are provided by global assimilations of MOPITT CO”.

**Question:** Line 194-195: If the boundary conditions used in assimilating MEE data the same as those used in assimilating MOPITT data? If not, the authors may have to account for how much differences produced by using different boundary conditions.

**Answer:** The different boundary conditions may have influenced the assimilation. As shown in the revised manuscript, we find it may not affect our conclusion: “as shown in Fig. 4a, the modeled CO columns from the boundary conditions are biased low by about 40%, which was not removed when assimilating MEE surface CO. While the influence from boundary conditions on surface CO concentrations over E. China could be limited, it is expected to have a noticeable influence on free tropospheric CO over E. China. It further confirms the overestimated enhancements on free tropospheric CO by assimilating MEE CO measurements, because potential negative biases due to the usage of a priori boundary conditions have been completely covered”.

**Question:** The MEE observations were averaged over a  $0.5^{\circ} \times 0.625^{\circ}$  model grid. So the representative information specified for those stations might be smoothed out to a certain extent. Thus the specified representative error might not correspond with the model grid.

**Answer:** Thank you for pointing out this issue! The assimilations and all Figures/Tables have been reproduced by generating grid super-observations and corresponding observation errors. We didn't find a noticeable discrepancy between the applications of super-observations and grid averages.

**Question:** Line 197-199: Data assimilation is expected to improve the agreement between observations and model simulations. The independent observations should be taken to evaluate the assimilation results.

**Answer:** The efficiency of our Kalman Filter method by assimilating MEE surface CO measurements has been evaluated in respect to independent observations by Han et al. (2022). As shown in the revised manuscript: "Han et al. (2022) further provided a comparative analysis between sub-optimal Kalman Filter and a hybrid deep learning model to predict surface CO concentrations in China in 2015-2020, and found the good performance of Kalman Filter in respect to independent observations".

**Question:** Line 229-236: Does the modeled CO concentration refer to CO concentrations analysis? In that case, the ratio of 1.5 is still relatively large, suggesting the observations are not efficiently assimilated. There are fewer observation stations in less polluted area. Consequently, the adjustment of prior CO concentration should be small, giving rise to large ratios up to 6 over less polluted area. Overall, those ratios just indicate that the prior CO concentrations are severely biased and an efficient correction scheme should be devised before doing the assimilation.

Line 245-246: It is not appropriate to use the ratios from fig.1 d to remove systematic background biases since the assimilation results are used to computer those ratios. BTW, why the observations are scaled other than prior CO field? Suggest to identify the possible systematic biases through the evaluation of simulations results from forward model run against observations.

**Answer:** These ratios are calculated based on a priori simulation. The description has been updated in the revision: "Fig. 1c-d exhibit the model a priori simulation and observed surface CO, as well as the ratios between observed and model a priori surface CO in 2019". We are sorry for the confusion.

**Question:** Line 252-253: Is the PBL height the dominant factor regulating the variability of meteorological conditions? Please explain.

**Answer:** Thank you for pointing out this issue! Multiple linear regression (MLR) has been widely used to disentangle the anthropogenic and meteorological contributions to surface O<sub>3</sub> and PM<sub>2.5</sub> variabilities. We could consider a similar analysis to analyze the effects of meteorological variabilities on observed and modeled surface CO concentrations in our future work, which may provide better normalization factors with interannual variabilities.

The discussion has been revised: "More efforts are needed in the future to evaluate the possible influence from meteorological condition changes on the inconsistency

between observations and simulations”. It was also noted in the revised conclusion: “More efforts to analyze the effects of meteorological variabilities on observed and modeled surface CO concentrations is helpful for better assimilation of surface CO observations, and more accurate evaluation for atmospheric CO changes”.

**Question:** Line 255-261: Does the assimilation using normalized surface CO performs better than that using raw surface observations? The authors should give more discussions so that readers could judge the validity of the normalization approach.

**Answer:** Both assimilations using normalized surface CO and raw surface CO have their advantages and disadvantages. It has been clarified in the revised conclusion section: “While the normalized CO measurements in this work are supposed to provide a better representation of atmospheric CO in the free troposphere, Kalman Filter by assimilating raw CO measurements is closer to real urban CO concentrations at surface level”. We are sorry for the confusion.

**Question:** Line 267: The description and discussion of the evolution or trend of CO over E. Asia is relevant only if the authors can show their assimilation results are solid enough. At the current stage, I suggest the authors also to check trends of surface observations and MOPITT CO retrievals and compare with assimilated trends.

**Answer:** Table 2 was revised to provide more detailed validations for the assimilation effects. We find the assimilations led to significant improvement in CO concentrations:

- Surface CO: the a priori, a posteriori (by assimilating raw MEE CO observations) and MEE CO observations are 397, 631 and 781 ppb over E. China in 2015-2020.
- Column CO ( $X_{co}$ ): the a priori, a posteriori (by assimilating MOPITT CO column data) and MOPITT CO observations are 97.4, 124.1 and 128.6 ppb over E. China in 2015-2020. Here the modeled CO profiles were smoothed with MOPITT averaging kernels.

However, we find the improvement in CO trends is weaker: the a priori, a posteriori and MOPITT observed decreasing trends are 0.2, 0.47 and 1.14 ppb/y over E. China in 2015-2020. It has been noted in the revised manuscript: “It should be noted that the decreasing trends in the MOPITT-based assimilations are more affected by the a priori simulations and are thus, weaker than those of MOPITT observations, as exhibited by the neutral changes over central China in Fig. 5c-d”. The subtitle of Section 3.5 was changed from “Evolution of atmospheric CO over E. Asia in 2015-2020” to “Assimilated atmospheric CO over E. Asia in 2015-2020”.

#### **Technical corrections:**

**Question:** Line 71: Replace “sufficient” to “sufficiently”.

**Answer:** Changed.

**Question:** Line 77-81: Please check the consistency of this sentence.

**Answer:** This sentence has been revised.

**Question:** Line 97-98: “radiance” can not measure “radiation”. Please correct.

**Answer:** Changed from “radiance” to “channel”.

**Question:** Line 105: “criteria pollutants”? Please check.

**Answer:** Changed from “criteria” to “critical”.

**Question:** Line 107: “reference state” or “reference temperature”? Please check.

**Answer:** Changed from “reference state” to “reference temperature”.

**Question:** Line 162: In figure 2, suggest to refine the color scheme of lines to make the different lines more distinguishable. The suggestion also applies to lines in other figures.

**Answer:** Thank you for this suggestion! The color lines in all figures were reproduced.

**Question:** Line 165: In Table 1, suggest to use ppb for the unit of column CO.

**Answer:** The unit has been changed.

**Question:** How is the “MEE CO(normalized)+MOPITT column” experiment conducted? It is not explained and discussed in the manuscript.

**Answer:** It is performed by assimilating both MOPITT CO and normalized surface CO measurements simultaneously. The caption of Table 1 has been revised.

**Question:** BTW, the results from last two rows are the same, please check.

**Answer:** Thank you for pointing out this issue! Changed.

**Question:** Line 193: “impacts” of what? Please check.

**Answer:** Changed.

**Question:** Line 220-221: Please check the grammar and rearrange this sentence.

**Answer:** Changed.

**Question:** Line 243: Replace “account for” to “to account for” or use a more appropriate phrase.

**Answer:** Changed.