Response to Reviewer #1

We thank the reviewer#1 for the insightful and detailed comments and suggestions, which helped to significantly improve the manuscript. The reviewer's comments are shown in *blue italics* with the author responses in black.

General comments:

Kou et al. conducted a regional atmospheric inversion analysis using GOSAT satellite XCO_2 products to constrain yearly CO_2 net fluxes in China. To quantify China's net ecosystem-atmosphere CO_2 exchange, they utilized the CMAQ-ENKS system. The regional inversion study is crucial in complementing commonly-used global inversion frameworks to accurately diagnose CO_2 NEE over complex environments. The manuscript is well-written and includes an in-depth discussion. I recommend it for publication in ACP pending the authors' response to the following comments.

1. After data screening, the size of GOSAT XCO_2 data may not be sufficient to inversely constrain all yearly 64 ×64km^2 and hourly CO_2 fluxes in China. How did you reconcile the observational limit and the specific spatial-temporal flux state vector? How did you determine the posterior uncertainty associated with the posterior CO_2 flux estimation, and did the posterior uncertainty involve the uncertainty due to the unbalance between the numbers of obs and the specific resolutions applied here? Please explain and clarify these questions in the response and main manuscript.

Thank the reviewer for the comment. (1) The update for CO_2 flux is given by the observation innovation and the correlations between CO_2 concentrations and emissions, while the correlations are naturally provided by the physics- and dynamics-based numerical model. (2) Although there are limited observation numbers, the observations are available of 1 hour. Thus through hourly update along with hourly model advances, the spatially sparse observations can sufficiently constrain the CO_2 flux, which can be demonstrated by the results. (3) Given the EnKF algorithm, the posterior uncertainty is proportional to the prior uncertainty but with a smaller magnitude. Based on hourly update, the posterior uncertainty contains the same flow-dependent information as the prior uncertainty. (4) For both chemistry assimilation and numerical weather prediction, it is commonly that the dimension of observation is much smaller than the dimension of state vector. Thus data assimilation helps to use the limited observations to constrain the state vector.

We have modified the relevant parts in the revised manuscript (Line 292–301); please check if it is clear now.

2. Line 316: How do you justify that the 7-day spin-up is enough to construct the inversion estimation, given that the domain is relatively large? Please clarify this point.

Thank the reviewer for the comment. The 7-day spin-up has been testified by a series of OSSEs (Observing System Simulation Experiments) in Peng et al., 2015, which conducted pseudo-satellite-observation and CMAQ assimilation with the same model domain and horizontal resolution (i.e. 64km× 64km over East Asia). Over the first few days, the assimilated CO_2 diverged from the modeled fields, generally moving closer to the observations, indicating that a spin-up time of about 7 days is required for the assimilation system to respond. In addition, the spin-up time for different assimilation systems implies that the long lifetime of atmospheric CO_2 and limited number of observations need to be taken into account in the regional joint assimilation framework (Tian et al., 2014, Peng et al., 2015, Kou et al., 2017).

We have modified the relevant parts in the revised manuscript (Line 334–337).

Here are the above-mentioned references.

Kou, X. X., Tian, X. J., Zhang, M. G., Peng, Z., & Zhang, X. L. (2017). Accounting for CO_2 variability over East Asia with a regional joint inversion system and its preliminary evaluation. *Journal of Meteorological Research*, 31(5), 834–851. https://doi.org/10.1007/s13351-017-6149-8.

Peng, Z., Zhang, M. G., Kou, X. X., Tian, X. J., & Ma, X. G. (2015). A regional carbon flux data assimilation system and its preliminary evaluation in East Asia. *Atmospheric Chemistry and Physics*, 15, 1087–1104. https://doi.org/10.5194/acp-15-1087-2015.

Tian, X., Xie, Z., Liu, Y., Cai, Z., Fu, Y., Zhang, H., & Feng, L. (2014) A joint data assimilation system (Tan-Tracker) to simultaneously estimate surface CO_2 fluxes and 3-D atmospheric CO_2 concentrations from observations. *Atmospheric Chemistry and Physics*, 14, 13281–13293. https://doi.org/ doi:10.5194/acp-14-13281-2014, 2014.

3. The Results section needs to be modified to be concise. It's better to move some content in Section 3 (Results) to Section 4 (Discussion). Please focus on your estimates for Section 3, modify and condense the previous studies (Lines 363-376) to either Discussion or Introduction sections.

Thank the reviewer for the comment. We have modified the Results section to be concise, and focused on our estimates for Section 3. Line 363–376 has moved to Discussion section.

We have modified the relevant parts in the revised manuscript (Line 381–392, Line 408–411, Line 538–539 and Line 549–561).

4. How does the performance of the posterior CO_2 flux estimates for the ocean area of the domain compare? Does GOSAT have the same algorithm to handle XCO_2 over land and ocean? Did you use the same QA/QC to determine your assimilated XCO_2 data for land and ocean?

Thank the reviewer for the comment. In this study, GOSAT XCO_2 retrievals were from NASA's ACOS_L2_Lite_FP.9r (data available at https://oco2.gesdisc.eosdis.nasa.gov/data/GOSAT_TANSO_Level2/), this version of processing supports both nadir and glint soundings. In the case of soundings over water, a check was made to ensure the observation was made in glint mode in ACOS retrievals.

In the present study, the GOSAT XCO_2 were introduced in the EnKS-based assimilation framework to constrain China's biosphere sink. The CMAQ-simulated CO_2 concentrations profiles were mapped into the GOSAT satellite retrieval levels and then vertically integrated based on the satellite averaging kernel according to the following equation:

$$XCO_2^f = XCO_2^p + \sum_{k=1}^{N_{lev}} \left\{ \left[\left(y_k^f - y_k^p \right) \boldsymbol{A}_k \right] \boldsymbol{h}_k (1-\boldsymbol{w})^{-1} \right\} \quad \text{(S1)}$$

Then state variables can be updated by applying the EnKS constrained by GOSAT retrievals over land and ocean in the analysis step (Equation S2).

$$\boldsymbol{x}^a = \boldsymbol{x}^f + K(y - H(\boldsymbol{x}^f)) \quad \text{(S2)}$$

Details of the Equation S1 and S2 are provided in Section 2.2.2.

Before being applied in assimilation, we use the same data screening strategy to handle XCO_2 over land and ocean. The retrievals for the glint soundings over oceans have relatively larger uncertainty, and thus many data over oceans are excluded in our inversions in terms of data screening strategy (Figure 2). On the other hand, our present study focus on top-down estimation of China's biosphere sink, fully investigation of posterior CO_2 flux estimates for the ocean area is outside the scope of this work and is therefore not discussed any further here. In the future, we'll further study China's ocean carbon source and sinks based on satellite retrievals and regional CTM assimilation in depth.

We have modified the relevant parts in the revised manuscript (Line 274–278, Line 292–301, and Line 673–676).

5. Line 344-345: "This discrepancy of the seasonal scale..." This sentence is not clear to me. Did you mean the mixing between biospheric and fossil-fuel sources or the differences between them?

Thank the reviewer for the comment. Table 1 indicates that the point-by-point uncertainty is larger in summer and lower in spring and autumn. The difference in seasonal performance could be partly due to the uncertainties in the spatial and temporal variations of the biosphere flux estimation and fossil-fuel inventories.

We have modified the relevant parts in the revised manuscript (Line 361–364); please check if it is clear now.

6. Line 341: "(1.99 and 2.41..)" lacks a unit.

Thank the reviewer for the comment. The unit for MAE and RMSE is ppm. We have added unit (i.e. ppm) in the revised manuscript (Line 360).

7. Was the system designed to prevent unrealistic and non-physical negative flux estimates due to Gaussian assumption/perturbation? Please clarify this point.

Thank the reviewer for the comment. In anthropogenic emission assimilation, negative flux estimates are unrealistic and non-physical, so they are eliminated. This might result in the Gaussian assumption not being satisfied. However, in carbon data assimilation, negative flux refers to the uptake of atmospheric

CO_2 by photosynthesis exceeds CO_2 released by respiration, especially in the growing season. Negative flux in carbon assimilation is realistic and reasonable, which are not excluded. In this way, Gaussian assumption is satisfied in JDAS carbon assimilation.

We have modified the relevant parts in the revised manuscript (Line 219–221).

8. Section 2.3, the last two paragraphs (Lines 284-302) have redundant information in terms of the XCO_2 data QA/QC (3-step screening strategy). Please modify them to be more concise.

Thank the reviewer for the comment. The assimilated and non-assimilated GOSAT XCO_2 observations are selected by different process of sifting (Table R1). These two sets of observations both used XCO_2 with "outcome_flag = 1" and precluded absolute biases between the observation and simulations greater than 5 ppm. Nevertheless, the main difference lies in step 2. The XCO_2 with the minimum "xco₂_uncert" in the same model grid point at the same hour were used to assimilate, and other XCO_2 were used to validate.

Table R1 GOSAT XCO_2 for assimilation and validation

	XCO_2 for assimilation	XCO_2 for validation
Step 1	Select XCO_2 with "outcome_flag = 1",	Select XCO_2 with "outcome_flag = 1",
Step 2	Select XCO_2 with the minimum "xco_2_uncert" in the same model grid point at the same hour	Select XCO_2 except for values minimum "xco_2_uncert", in order to filter out all of the assimilated XCO_2
Step 3	Preclude record with absolute biases between the observation and simulations greater than 5 ppm	Preclude record with absolute biases between the observation and simulations greater than 5 ppm

We have modified the relevant parts in the revised manuscript (Line 303−320).

9. The posterior uncertainty is not presented in the study. It would be expected that this information is displayed for Figures 3, 4, and 6. Echoing my previous comment, please add or clarify the posterior uncertainty considerations/treatment in the study.

Thank the reviewer for the comment. Similar to CarbonTracker which uses transport model as a forward operator in an ensemble fixed-lag Kalman smoother, JDAS is also extended to incorporate the EnKS feature. The EnKS allows for a sequential processing of the measurements in time and is used to assimilate the concentrations and update the fluxes. Thus, EnKS that can take into future observations into account is used to assimilate the concentrations and update the fluxes. The smoothing window of EnKS (i.e. denoted as assimilation window hereafter) was set to 24 h in this study. In an assimilation cycle, the fluxes for the 24-h smoothing window have been designed to be optimized hour by hour successively.

In the joint assimilation framework, besides the application of CMAQ to generate ensemble CO_2 concentrations, a flux forecast model was also designed to represents flux variations on account of fluxes acting as model forcing. Consequently, after completing the "forecast step", Kalman gain matrix K is obtained by minimizing the analysis error covariance with evolved forecast error covariance over time. Then, the associated analyzed state variables, $\boldsymbol{x}^a = \left[\boldsymbol{C}^a, \boldsymbol{E}^a\right]^T$, can be updated by applying the EnKS constrained by GOSAT retrievals in the "analysis step". Furthermore, the distribution of ensemble spread of CO_2 flux in January 2016 is provided in Figure R1. It shows that the values of the ensemble spread ranges from 0.2 to 0.8 in most areas, which are consistent with our previous studies (Peng et al., 2015 in Figure 11c and Peng et al., 2023).

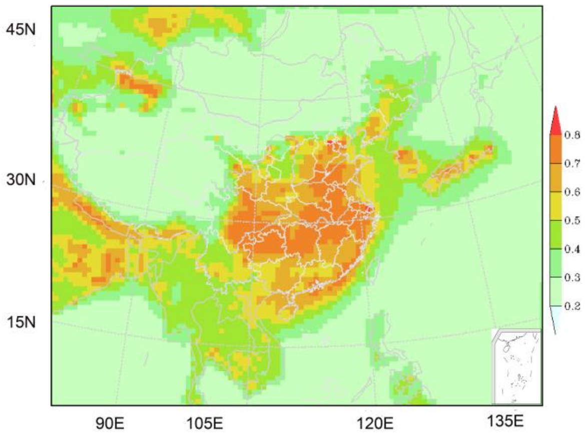

Figure R1. The ensemble spread of $\boldsymbol{\lambda}_{i,t}^{a}$ at model level 1 in January 2016, when β=80.

Here are the above-mentioned references.

Peng, Z., Zhang, M. G., Kou, X. X., Tian, X. J., & Ma, X. G. (2015). A regional carbon flux data assimilation system and its preliminary evaluation in East Asia. *Atmospheric Chemistry and Physics*, 15, 1087–1104. https://doi.org/10.5194/acp-15-1087-2015.

Peng, Z., Kou, X. X., Zhang, M. G., Lei, L. L., Miao, S. G., Wang, H. M., Jiang, F., Han, X., and Fang, S. X. (2023). CO_2 flux inversion with a regional joint data assimilation system based on CMAQ, EnKS, and surface observations. *Journal of Geophysical Research-Atmosphere*, 128, e2022JD037154. https://doi. org/10.1029/2022JD037154

We have modified the relevant parts in the revised manuscripts (Line 271−278), and Figure 1 is further added and discussed.

10. Analysis increments are not clear to me. How did you calculate them? Did you use FC minus CTRL? The adjustments between FC and CTRL are very small over the ocean.

Thank the reviewer for the comment. (1) The analysis-minus-background, AMB, (i.e., $\boldsymbol{x}^a - \boldsymbol{x}^{\mathrm{b}}$) is denoted as "analysis increments". Fig. 2 focus on the discussion of flux analysis increments (i.e., $\boldsymbol{E}^a - \boldsymbol{E}^{\mathrm{b}}$) to certify that GOSAT XCO_2 is effectively absorbed in JDAS. (2) FC and CTRL experiments were further designed to assess the quality of the inversion results. One set of experiments was forced by the optimized *a posteriori* fluxes (denoted as FC), and the other was forced by the prescribed *a priori* fluxes as a control experiment (denoted as CTRL). This traditional approach was adopted as a compromise to assess whether the *a posteriori* fluxes would enable improvements in the fit to observed CO_2 concentrations, including non-assimilated GOSAT as well as surface observations from 14 sites. (3) The retrievals for the glint soundings over oceans have relatively larger uncertainty, and thus many data over oceans are excluded in our inversions in terms of data screening strategy (Figure 2). In consequence, the adjustments between FC and CTRL are very small over the ocean.

We have modified the relevant parts in the revised manuscripts (Line 366−368).

11. Figure 1: The colors for the analysis increment plots are too light. Please modify the color bar range to have a better display of the contrast between sink and source.

Thank the reviewer for the comment. We have modified the color bar range (Fig. R2) to have a better display of the contrast between increases and decreases in the revised manuscript (Fig. 2); please check if it is clear now.

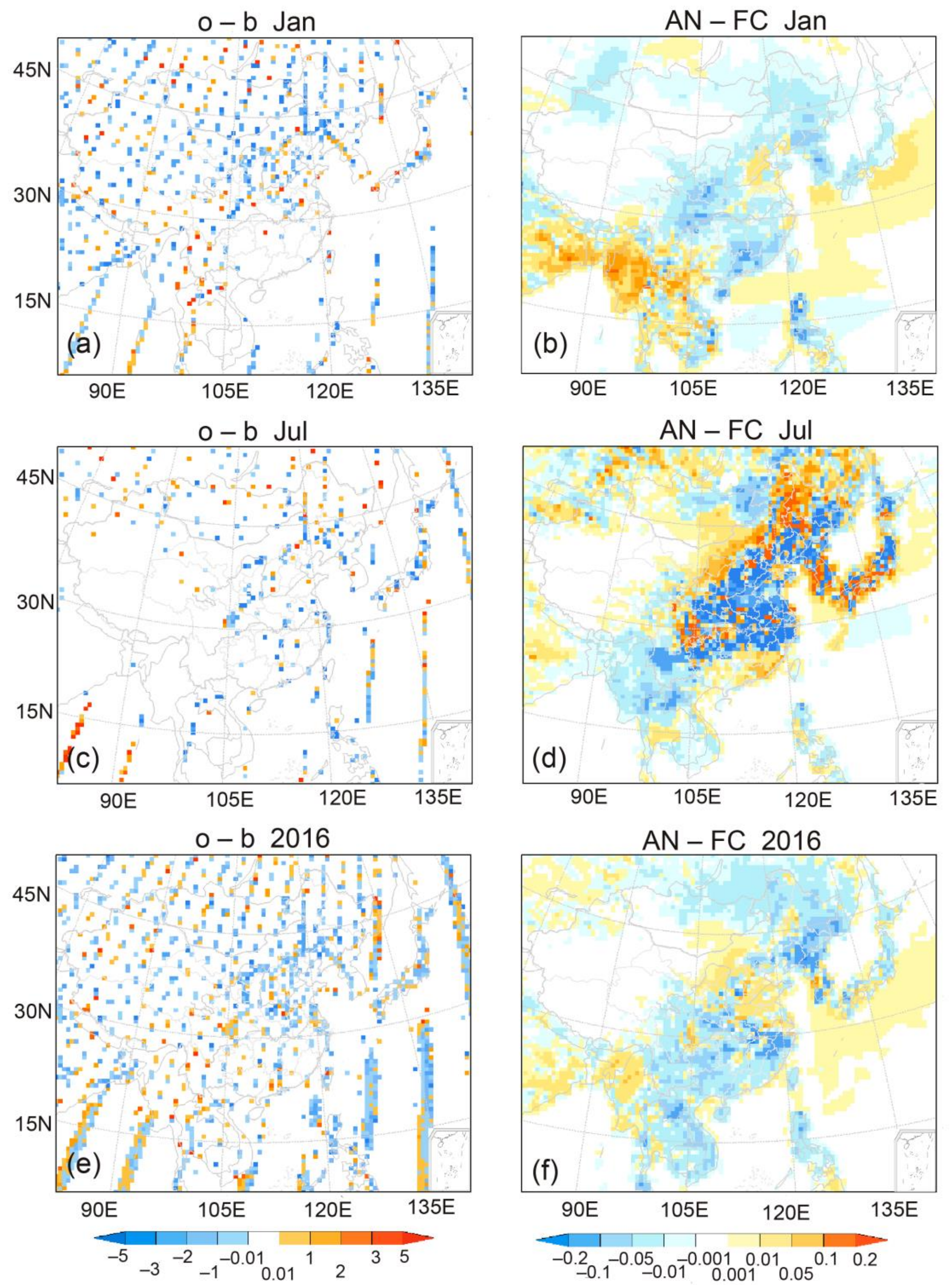

Figure R2. Observation increments (XCO_2; unit: ppm) and analysis increments (biosphere flux; unit: μmole m^{-2} s^{-1}) in (a, b) January, (c, d) July, and (e, f) the whole year of 2016.