Response to the comments of Reviewer #1

We would like to thank the anonymous referee for his/her comprehensive review and valuable comments. These comments helped improve and clarify the submitted manuscript. In response, we have made changes according to the referee's suggestions. Below we reply to each comment point by point, showing the reviewers' comments in black and our responses in blue. Changes to the original manuscript are highlighted in **bold blue**. Note that the line numbers in the response are updated based on the revised manuscript, which we provide with our response.

General comments

Zhu et al. investigated the terrestrial GPP estimation using COS within a two-leaf modelling framework. COS flux data were used to calibrate the ecosystem model parameters and to optimize GPP simulations among different ecosystems within the Monte Carlo-based methodology base on the coupling of COS modeling and the BEPS model. The approach is with novelty, and brings new method and knowledge to the field of carbon cycle and also improves the estimation of GPP. In general, the work presented in the manuscript is interesting and worthy of publication. However, there are a few issues the authors should address before publication. The figures, tables and citations are not carefully maintained. The storyline is sometimes hard to follow.

Response: Thank you for your valuable feedback. We acknowledge the oversight in maintaining the figures, tables, and citations, as well as the shortcomings in ensuring the coherence of the storyline. In response, we have revised the inappropriate figures, tables, and citations in the original manuscript, and rewritted relevant sentences to enhance the coherence and readability of the article.

Specific comments

COS fluxes measurements are used to assimilate and improve the BEPS model and GPP estimate. COS itself is also a trace gas in the atmosphere, and the authors are suggested to summarize the measurements of COS mole fractions and relevant modelling studies. The manuscript is related to another manuscript, Zhu et al, (2023 under review). Authors are advised to cite the previous one and discuss relevance to the current manuscript, e.g. the model approach. The discussion part is suggested to include a discussion of advantage and disadvantage of the model work.

Response: Thanks for the valuable comments. As you mentioned, this manuscript is related to another manuscript, i.e., Zhu et al. (2023). Indeed, both of these works are implemented within the two-leaf model framework. The difference lies in the fact that in the other manuscript, our main objective is to introduce our newly developed adjoint-based assimilation system (Nanjing University Carbon Assimilation System, NUCAS v1.0), demonstrate the robustness of the assimilation system, and investigate the constraints of the tracer-gas, e.g. carbonyl sulfide (COS) on water, energy and carbon related parameters and processes. While COS assimilation has proven effective in constraining COS-related model parameters and optimizing GPP, there remains a gap of systematic understanding of the interaction, identifiability of the optimized model parameters from different processes as well as the ability of COS in reducing model prediction uncertainty of GPP.

In this manuscript, we address the shortcomings of the adjoint-based sensitivity analysis, which is based on the Bayesian approach, by employing the Monte Carlo-based parameter calibration method, and conducted a global sensitivity analysis to provide general results over the entire parameter space. Notably, we also analyzed COS-related parameter interaction, identifiability, as well as the constraint ability of COS on GPP uncertainty.

As for the measurements and modelling studies of COS mole fractions, they are also integral parts of research in the COS field, closely related and crucial to this study. Consequently, we fully agree with your point that it is necessary to summarize and discuss the measurements and modelling studies of COS mole fractions.

In view of this, we have made modifications to three aspects of the manuscript: (1) Clarifying the advantages of the two-leaf model, and explaining why we did not conduct comparative analyses between the two-leaf model and other models (i.e., the big-leaf model). (2) Increasing the citation of another study; discussing the advantages and disadvantages of the Monte Carlo-based parameter optimization approach. (3) Adding a summary and discussion of the observed and simulated studies related to COS mole fraction. Details are as follows:

(1) In the introduction, we have added a paragraph to introduce the rationality of the two-leaf model and the necessity of applying two-leaf model in LSM, as shown below: “**Due to the dissimilar illumination conditions, there are the significant variability of leaf photosynthesis between sunlit and shaded leaves (Chen et al., 1999; Pignon et al., 2017; Wang et al., 2018; Bao et al., 2022). It is now clearly recognized that big-leaf models are conceptually flawed and practically inaccurate and sunlit-shaded leaf stratification is necessary to make accurate canopy-level photosynthesis estimation (Chen et al., 2012; Luo et al., 2018). Consequently, in the process-based LSM that simulates COS plant uptake and photosynthesis in a coupled manner (Ball et al., 1987; Berry et al., 2013), the application of the two-leaf model shows promise for providing precise simulation of plant COS uptake.**” (line 71-76)

In the new added section (**Section 4.5 Caveats and implication**), we have clarified the reason why we did not conduct comparative analyses between the two-leaf model and other models (i.e., the big-leaf model), as shown below: “**Compared to big leaf model, two-leaf model has been demonstrated to better describe the canopy radiation distribution, GPP, and stomatal conductance (Luo et al., 2018). In this study, we take the advantage of two-leaf model, to simulate COS fluxes from plant and soil based on the BEPS model within the two-leaf framework. Ecosystem-scale COS flux data were used to calibrate the model parameters belong to BEPS and to optimize GPP simulations among diverse ecosystems within the Monte Carlo-based methodology. Our results demonstrate that COS not only improves the accuracy of GPP simulations but also reduces GPP simulation uncertainty. However, due to the lack of in-situ COS concentration and flux data, as well as BEPS model driving data (e.g. meteorological data, LAI data and clumping index data), However, due to the lack of in-situ COS concentration and flux data, as well as BEPS model driving data (e.g., meteorological data, LAI data, and clumping index data), we are still facing challenges in evaluating the performance of the two-leaf model compared to**

other models in COS simulation. The increasing availability of observational data holds promise for addressing this issue." (line 538-546)

(2) We have added an introduction to another manuscript in the introduction section: "Currently, several studies have been endeavored to refine the model parameters of LSMs through assimilating the COS data, and thereby optimized the modeling of water-carbon fluxes (Chen et al., 2023; Abadie et al., 2023; **Zhu et al., 2023**)" (line 63)

This manuscript shares similarities with another manuscript (Zhu et al., 2023) in that both involve optimizing GPP using COS. In the other manuscript, we constructed an adjoint-based Nanjing University Carbon Assimilation System (NUCAS) and thus assimilated COS within NUCAS. In contrast, we utilized a Monte Carlo-based method to assimilate COS in this manuscript. Here, we took the advantage the Monte Carlo-based parameter optimization method, analyzed the global sensitivity, identifiability as well as interactions of COS-related parameters. Furthermore, the model prediction uncertainty for COS and GPP were evaluated. Thus, we added a discussion regarding this in the new added Section (**Section 4.5 Caveats and implication**), as shown below: "**Taking advantage of the Monte Carlo-based parameter optimization approach, we analyzed the global sensitivity, identifiability as well as interactions of COS-related parameters in this study. Furthermore, we quantified the uncertainty in simulated COS and GPP, thereby revealing the capacity of COS to constrain the uncertainty in GPP simulations. However, the Monte Carlo-based parameter optimization approach subject to controversy (Sambridge and Mosegaard, 2002) due to the numerous subjective decisions involved in its implementation, such as the selection of parameter range, sample size and performance metric, etc. Further research is needed to investigate the impact of these factors on the parameter optimization results related to COS and the assessment of model prediction uncertainty.**" (line 569-575)

(3) With reference the observed and simulated studies related to COS mole fraction, a summary of the trace gas COS in the atmosphere is included in the introduction: "**Carbonyl sulfide (COS) is the most abundant sulfur-containing trace gas in the atmosphere with a lifetime of about 2 years (Montzka et al., 2007; Karu et al., 2023). The tropospheric atmospheric mole fraction of COS is approximately 500 parts per trillion (ppt), exhibiting a typical seasonal amplitude of ~ 100–200 ppt (Montzka et al., 2007; Ma et al., 2021; Hu et al., 2021; Remaud et al., 2022; Remaud et al., 2023; Ma et al., 2023).**" (line 37-40)

A discussion about the modeling of COS mole fraction as well as the COS vegetation sink were included in the new added Section (**Section 4.5 Caveats and implication**), as shown below: "**The spatial and temporal variation in atmospheric COS concentrations has a considerable influence on the COS plant uptake (Ma et al., 2021; Kooijmans et al., 2021) due to the linear relationship between the two (Stimler et al., 2010). With the lack of continuous ground-based COS concentration observations, COS concentrations in the bulk air are assumed to be spatially invariant over the globe but to vary annually in this study, which may lead to significant biases in COS simulations. Currently, several recent studies have simulated COS vegetation fluxes based on atmospheric transport model-derived COS concentration data within the big-leaf framework (Kooijmans et al., 2021; Maignan et al., 2021; Abadie et al., 2023). These COS fluxes simulated based on big-leaf**

models were in turn used to drive atmospheric transport models (Remaud et al., 2023; Ma et al., 2023). Within an atmosphere inversion framework, recent studies indicate an underestimation of the biosphere COS sink in high-latitude regions of the Northern Hemisphere (NH) (Ma et al., 2021; Remaud et al., 2023). Larger underestimations of ecosystem COS exchange based on big-leaf model at high latitudes have also been confirmed at the site scale, and the underestimations of COS are consistent with biases in GPP for some sites (Kooijmans et al., 2021). Interestingly, Luo et al. (2018) demonstrated that the reason for the underestimation of GPP by the big-leaf model is that it fails to accurately describe the instantaneous radiation distribution in the canopy, and the underestimation increases with the increase of LAI. As we all know, the NH high-latitude regions have relatively high LAI (Fang et al., 2019). Therefore, the deficiency of the big leaf model in simulating radiation distribution may contribute to the existence of the missing COS sink in the NH high latitude in summer, and this deficiency amplified by the larger LAI. In fact, the spatial distribution of LAI (i.e., GLOBMAP LAI) retrieved through remote sensing not only in NH high-latitude regions but also in central Africa aligns with the spatial distribution of the missing sink revealed by the "objective" inversion conducted by Ma et al. (2021) (as illustrated in Figure 7 in Ma et al. (2021)), which further validate the reasonableness of this hypothesis. Therefore, conducting COS simulations under the two-leaf framework at a global scale holds the promise of providing insights into the global COS vegetation sink and benefiting the simulation of the spatial and temporal distribution of COS concentrations. Thus, it is necessary to conduct regional and global COS simulations within the two-leaf model framework in the future." (line 547-568)

Technical corrections and Typing errors

Response: We sincerely appreciate the careful review and detailed comment provided by the reviewer.

Line 1: The title "two-leaf" could be two-leaf without "".

Response: Corrected.

Line 5: change to the affiliation only without currently at. If the co-author is currently only at this affiliation, please indicate with a superscript.

Response: Thank you for your comment. We have modified the affiliation accordingly.

Line 18: 'two-leaf' to two-leaf, and elsewhere.

Response: Corrected.

Line 19: "through the fusion of COS data" to "through the data assimilation of COS flux measurements".

Response: Thanks for your comment. We have made modifications to the sentence accordingly.

Line 27: GPP should be one keyword. Model-data fusion is not accurate, use data assimilation.

Response: Thanks for your comment. We have replaced the key word "Model-data fusion" with “data assimilation” and included “GPP” as a key word.

Line 55: “not only the model variables like GPP are expected to be optimized” to “not only the model variables like GPP are expected to be improved”.

Response: Corrected.

Line 57: “through assimilating the COS data” to “through assimilating the COS flux measurements”.

Response: Corrected.

Line 58: here more related papers should be cited, e.g. Zhu et al., 2023.

Response: Thank you for your comment. We have now added a citation to this manuscript.

Line 65: “LSM” to “a LSM”.

Response: Corrected.

Line 72-75: it is too vague to read. Please rewrite what you are going to do in details.

Response: Thanks for your comment. We have revised the sentences as follows: “**To address these questions, we utilized ecosystem COS flux data to optimize GPP across various ecosystems based on the coupling of COS modeling with the two-leaf based Biosphere-atmosphere Exchange Process Simulator (BEPS). Through Monte Carlo simulations, we conducted a global parameter sensitivity analysis to explore the sensitivity of COS and GPP simulations to model parameters related not only to photosynthesis but also to water and energy. The interaction and identifiability of these parameters were quantified using Monte Carlo optimized parameter sets. Additionally, the effectiveness of COS in constraining model uncertainty in simulated COS and GPP are evaluated.**” (line 84-89)

Line 90: “two-leaf” to two-leaf.

Response: Corrected.

Line 95: the model description is not clear enough. Suggest move details to the main text from appendix A1.

Response: Thanks for your valuable suggestion. To facilitate readers' understanding of our two-leaf COS and GPP simulation framework, we have moved the relevant model descriptions from Appendix A1 and Appendix A2 to the main text.

Table 1: Is there missing data in a whole year? How do you deal with the missing data?

Response: Thanks for your comment. The hourly COS flux observational data from these sites exhibit varying degrees of gaps, as illustrated in Figure 4. In Figure 4, we use the size of scatter points to represent the number of COS observational data points, aiming to provide readers with a rough understanding of the time periods and extent of missingness in the COS data. Additionally, for the majority of sites (except FI-Hyy and US-Ha1), the COS observational data time series are less than one year, typically around one month. Therefore, we have assigned different x-axis labels for different sites in Figure 4.

For most sites (except FI-Hyy), only the measured COS flux data are available and we did not do anything to deal with the missing data. Following the recommendations regarding the standardized processing of eddy covariance flux measurements of COS (Kohonen et al., 2020), both measured and gap-filled COS flux of FI-Hyy are provided in Vesala et al. (2022), and the latter were utilized in this study. We have also clarified this in the revised manuscript: "**Specifically, following the recommendations regarding the standardized processing of eddy covariance flux measurements of COS by Kohonen et al. (2020), both the measured and gap-filled COS flux observations are provided in Vesala et al. (2022), and the latter were utilized in this study.**" (line 184-186)

Section 2.3.1: how do you select the satellite LAI data to best match the field measurements?

Response: Thanks for your comment, the publications listed in **Table 1** provided in-situ LAI information for these sites, for example, the mean LAI values ($m^2\ m^{-2}$) during the campaign of AT-Neu, DK-Sor, ES-Lma and IT-Soy are provided in Table S1 of the supplementary material in Spielmann et al. (2019). Such information provides us with a reference for selecting LAI products. Now, we have rewritten the sentence to provide specific explanations for the selection of LAI, as follows: "**With reference to the observed LAI at these sites (Wehr et al., 2017; Rastogi et al., 2018; Spielmann et al., 2019; Kohonen et al., 2022), we used GLOBMAP products to drive the BEPS model at most sites (5/7) due to its good agreement with the observed LAI. Specifically, as the GLOBMAP product had considerably underestimated LAI at DK-Sor and was not consistent with the vegetation phenology at ES-Lma during the measurement campaign, GLASS LAI was used at these two sites.** (line 163-167)

Line 129: define ERA5.

Response: Thanks for your comment. Now the definition of ERA5, i.e., **European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis v5** is added. (line 174-175)

Section 2.4: Is it the optimization approach? If so, please rename the section title to show the method explicitly.

Response: Thanks for your valuable comment. We have renamed the section title as "**2.4 The Monte Carlo-based parameter optimization approach**". (line 193)

Line 159: please refer to literatures.

Response: Thanks for your comment. Now, we have supplemented the original sentence with additional references, and the revised sentence reads as: "The prior values and prior ranges for these parameters (**Table 2**) were chosen based on literature **(Jackson et al., 1996; Medlyn et al., 1999; Kattge et al., 2009; Miner et al., 2017; Ryu et al., 2018)** and default model settings." (line 219-220)

Line 164: define "behavioral and non-behavioral simulations".

Response: Thanks for your comment. We have rewritten the sentences to include a definition of behavioral and non-behavioral simulations. "**Subsequently, model realizations are grouped into behavioral and non-behavioral model runs and associated parameter sets based on the values of the single or multiple performance measures and the predefined**

threshold value (Houska et al., 2014). The former describes acceptable model realizations conditioned on the available observational data (Blasone et al., 2008; Beven and Binley, 2014). The latter describes parameter sets that produce behavior inconsistent with observed behavior." (line 199-203)

Line 207: "influence GPP modeling but have minimal impacts on COS modeling." To "influence GPP simulations but have minimal impacts on COS simulations."

Response: Corrected.

Figure 1: the parameters need to be explained in the figure caption.

Response: Thank you for your comment. Now, we have provided detailed descriptions of the parameters before presenting the results, as shown in **Table 2**.

Line 220: Here the text refers to Fig. 2?

Response: Yeah, thanks for your comment. We have thoroughly reviewed the manuscript to ensure accurate referencing of figures and tables.

Figure 2: there are many subplots in the figure, maybe make it bigger.

Response: Thanks for your comment. We have made every effort to present the figure as clearly as possible.

Figure 2: explain the parameters and PI in the figure caption.

Response: Thank you for your comment. Now, we have provided detailed descriptions of the parameters before presenting the results, as shown in **Table 2**. Additionally, the definition of PI has been added in the figure caption.

Line 281: here you refer to Fig. 3? Also Line 287-288.

Response: Yeah, thanks for your comment. We have thoroughly reviewed the manuscript to ensure accurate referencing of figures and tables.

Table 3: define reduction in percentage.

Response: Thank you for your comment. The definition of reduction has been added in the legend, as follows: **The reduction (%) of $RMSE_{mean}$ and range width is calculated as (1-posterior / prior) * 100.** (line 381-382)

Figure 3: the order of numbering is something wrong. IT-Soy should be (d).

Response: Thanks for your comment. In **Table 1**, the site characteristics were listed in alphabetical order. We intend to present the results in the same order (alphabetical order) in other figures, including **Figure 3** and **Figure 4** of the original manuscript. Therefore, in **Figure 3** and **Figure 4**, the subplot corresponding to IT-Soy has been assigned the label (e) instead of (d) based on alphabetical order of the site names, even though it is placed above the subplot corresponding to FI-Hyy for compact arrangement of the subplots. Regarding this matter, we have clarified in the legends: **The subplot numbers are assigned based on the alphabetical order of the site names.** (line 388-389)

Figure 3: it is confusing that some panels have x-axis labeled as year, while others are labeled as Day of year. Please make it in consistency.

Response: Thanks for your comment. Here, x-axis labels are assigned to the subplots according to the duration of the corresponding COS observational data. For the majority of sites (except FI-Hyy and US-Ha1), the duration of COS observational data is only about one month. In contrast, multi-year COS observations are available at FI-Hyy and US-Ha1. We are keenly aware of the importance of maintaining consistency in the labels of each subplot. However, given the significant differences in the duration of COS observational data across these sites, different x-axis labels (“Year” and “Day of year”) have been assigned to the subplots in Figure 3 and Figure 4 of the original manuscript.

Line 334: refer to Fig. 4.

Response: Corrected.

Line 341: refer to Fig. 4d.

Response: Corrected.

Figure 4: IT-Soy should be (d).

Response: Thanks for your comment. Regarding this matter, we have already provided detailed explanations earlier, please refer to the preceding sections. Also, we have clarified the assignment of the subplot numbers: “**The subplot numbers are assigned based on the alphabetical order of the site names.**” (line 420-421)

Table 2 and Table 4: why is RMSE reduction of COS range width is much larger than that of GPP?

Response: Thanks for your valuable comment. The reason for this phenomenon is the difference of the sensitivity of simulated COS and GPP to the model parameters. Specifically, the parameters m_{H_2O} and VJ_{slope} strongly influence GPP modeling but have minimal impacts on COS modeling. Therefore, even after COS assimilation, these two parameters still have a wide posterior range, thus resulting in a large posterior range for GPP simulation. On the contrary, parameters that are sensitive to COS (i.e., those that have a significant impact on the posterior range of COS simulation) are well-constrained. As a result, there is a considerable reduction in the range of COS simulation.

Figure 5 and Figure C1: move Figure C1 to main text. Or combine Figure 5 and Figure C1.

Response: Thanks for your comment. According to your suggestion, we have combined these two figures.

Line 390: “Knauer et al., 2020” is not in the Reference.

Response: Corrected.

Line 394: “Ma et al., 2022” is not in the Reference. check reference if all of them are properly cited in the main text.

Response: Thanks for your comment. We have thoroughly reviewed the manuscript to ensure precise citation.

Line 420: remove “To provide deeper insights into these interactions and highlight significantly correlated parameter combinations, we generated Fig. 6.”

Response: Thank you for your comment. We have modified the sentence accordingly.

Line 421: “This figure …” To “Figure 6 …”

Response: Corrected.

Figure 6: It is not easy to interpret the information from 3D view. Please try cross-section.

Response: Thanks for your comment. The design of this figure is inspired by Figure 4 from Beven and Binley (2014). Similar to Beven and Binley (2014), we employ 3D plots to further explore, visually, the parameter space. The difference lies in the fact that Beven and Binley (2014) uses parameter likelihood thresholds to identify behavioral parameter sets and plots likelihood threshold surfaces in parameter space. In this study, we use an acceptable sampling rate to identify behavioral parameter sets. Thus, what is depicted here are collections of behavioral parameter sets in parameter space. However, fundamentally, our goal is to explore, visually, the parameter space, akin to Beven and Binley (2014). Therefore, following the suggestion of the reviewer #2, we remained this figure but relocated it to the appendix.

Line 436: define PI before using it.

Response: Thanks for your comment. We have already defined PI in Section 2.7. (line 253)

Line 456: provide citation or the text you refer to.

Response: Done.

Line 474: “show a significant range of variation”, provide an estimate of the range.

Response: Thanks for your comment. In order to quantitatively describe the results, we revised the sentence as follows: **However, given the uncertainties of model parameters, structure and driving data etc., instances like at ES-Lma arise where the model runs with the 3 % highest RMSE for GPP instead exhibit good performance in terms of COS, with their RMSE values for COS all falling within the top 55 %. (line 526-528)**

Line 482: “COS modeling” to “COS simulation”

Response: Corrected.

Reference:

Zhu, H., Wu, M., Jiang, F., Vossbeck, M., Kaminski, T., Xing, X., Wang, J., Ju, W., and Chen, J. M.: Assimilation of Carbonyl Sulfide (COS) fluxes within the adjoint-based data assimilation system–Nanjing University Carbon Assimilation System (NUCAS v1.0), EGUsphere [preprint], https://doi.org/10.5194/egusphere-2023-1955, 2023

Response: Thank you for your suggestion regarding citing the manuscript. Now, we have already included a citation to this manuscript.

References:

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