Zhu et al. have greatly improved the clarity of their manuscript and mostly made adequate changes and responses to the reviewer comments on the first round. However, I still have two major concerns regarding the paper.

Continuing the discussion related to the US-Wrc site gradient fluxes. Note that there are two papers by Rastogi et al. in 2018 using gradient flux data from the same site; one has data from 2014 and COS fluxes are (for whatever reason) not published (https://doi.org/10.1029/2018JG004430) and the other reports fluxes from year 2015 (https://doi.org/10.5194/bg-15-7127-2018). Dataset for the latter (including gradient fluxes of COS) can be found from: https://zenodo.org/records/1516332
To reduce the considerable bias the authors currently have regarding the calculation of US-Wrc fluxes, I highly recommend to use the published COS gradient fluxes from this site, from year 2015, and to rerun the analysis once more using this dataset. This would considerably reduce bias and improve the analysis. These data can then also be used in the two-site assimilation, which, I still in its current state (when gradient fluxes are first calculated using simulations, which then are again used to simulate fluxes) I do not approve of. If proper gradient fluxes provided by Rastogi et al (in the link above) are used, only then the two-site assimilation is possible. Note that Hyytiälä forest also has flux measurements in 2015.

The authors argue that sensible heat flux (H) and latent heat flux (LE) as well as soil water content (SWC) are related to COS fluxes because COS fluxes are related to transpiration. However, transpiration is only one part of ET (evapotranspiration, highly related to LE) and the other part is evaporation, which has no relation to COS fluxes. Evaporation and SWC are also highly related to water availability (precipitation) as well as other environmental variables (radiation, temperature). In addition, it is definitely not only the leaf-scale energy demand that controls the sensible heat flux at ecosystem scale. You forget soil, atmospheric turbulence, input energy from the sun, ground heat flux, evaporation, precipitation, saturation of SWC… Yes, COS fluxes could be used to estimate transpiration, but anything further is overinterpretation. Thus, I still very highly recommend completely leaving out the LE, H and SWC simulations.

From the response document:
L400: "due to high value of observation" or rather underestimation by simulation?
Response: Could, of course, be either, but according to Kooijmans et al. (2021), the air depleted in COS can then suddenly be captured by the EC system when turbulence is enhanced in the morning.

- ➔ This is why we do storage correction to EC fluxes! Storage corrected fluxes do **not** have this problem. I am not saying that observations would be perfect, but they are "the best guess" we have. Thus, I suggest to reformulate accordingly.

Specific comments:

How is this manuscript related to a preprint that is simultaneously in review (Zhu et al., 2024)? The other study seems very much related, and should be cited in this study as well.

The abstract and conclusions are still missing concrete results. The authors use descriptive words such as “improved”-> improved by how much or by what metric? Describe in detail (using numbers) what were the most important results of your study (e.g. how much (in %) did the assimilation improve the prior simulation etc).

Merge Figs 3 and 4 in a similar way as Fig. 6.

Eq. 9-10: $F_{cos,biotic}$ is switched to F_{SWCg} and SWC to SWC_g between the equations. Please check that is consistent.

References:

Rastogi, B., Berkelhammer, M., Wharton,S., Whelan, M. E., Itter, M. S., Leen, J. B.,et al. (2018). Large uptake ofatmospheric OCS observed at a moistold growth forest: Controls andimplications for carbon cycleapplications.Journal of GeophysicalResearch: Biogeosciences,123,3424–3438. https://doi.org/10.1029/2018JG004430

Rastogi, B., Berkelhammer, M., Wharton, S., Whelan, M. E., Meinzer, F. C., Noone, D., and Still, C. J.: Ecosystem fluxes of carbonyl sulfide in an old-growth forest: temporal dynamics and responses to diffuse radiation and heat waves, Biogeosciences, 15, 7127–7139, https://doi.org/10.5194/bg-15-7127-2018, 2018.

Zhu, H., Xing, X., Wu, M., Ju, W., and Jiang, F.: Optimizing the terrestrial ecosystem gross primary productivity using carbonyl sulfide (COS) within a “two-leaf” modeling framework, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2023-3032, 2024.