Responses to Reviewer #1

We are grateful to the reviewer for the helpful comments and guidance that led to important improvements of the original manuscript. Our point-by-point responses are listed below. Reviewer's comments are in black font, and authors' responses are in blue. Page and line numbers refer to the manuscript egusphere-2023-372 (the ACPD version).

In this manuscript, Zheng et al. investigate the role of anthropogenic pollutants on the production of biogenic secondary organic aerosols. Using a chemistry-climate model, they perform decadal simulations for present day and pre-industrial periods and compare three schemes that simulate the production of secondary organic aerosols (SOAs) with increasing complexity: (1) Simple, (2) CMPX and (3) CMPX_ag. Over high-source regions of biogenic emissions such as the South-eastern United States and the Amazon, the more advanced scheme, CMPX_ag, outperforms the others and reproduces the observed seasonal variability and trend of summertime organic aerosols. By accounting for different NOx regimes (low vs. high) and for photochemical aging, the CMPX_ag scheme is more sensible to anthropogenic pollution, thus simulating an increased SOA production over present-day compared to the pre-industrial period, although land-cover change has driven a decrease in biogenic emissions. Hence, this study highlights the tight link between SOA production and anthropogenic pollution.

The paper is within the scope of ACP. It examines an important topic such as the sensitivity of biogenic SOA production to anthropogenic pollution, and addresses relevant scientific questions. The paper is well written, the abstract is concise and complete, the introduction is exhaustive and clear, the methods and modeling are well laid out, the literature is thoroughly referenced, and the results are presented in good clear figures. For this reason, I recommend publication after a few minor comments, listed below, have been addressed by the authors.

Sect. 2, Methods

Regarding the GFDL AM4.1 model and the modeling of the SOA formation, since the CMPX scheme depends on OH, O_3 and NO_x abundance, in my opinion the author should show how well the GFDL AM4.1 model reproduces these gases, or at least insert a sentence/paragraph that summarizes results from previous studies that evaluated the model performance.

To allow the traceability of results, I think it is important to provide details on the spatial resolution and the time-step of the GFDL AM4.1 model. I also suggest to precise the original temporal resolution of observational datasets (IMPROVE, SEARCH, ACTRIS and ARM) and if (and how) these data have been aggregated.

Moreover, I think it is important to explain in the Methods section how relative/percent trends, presented in Sect. 3 (e.g., pag.6, line 18), have been computed. If I correctly understood, I found this information in Fig. S4, in the Supplementary Material ("Changing rates *m* have units of % per year relative to their 2000-2016 averages").

In Page 5 Line 8, we add: "The surrogate TSOA products are implemented in addition to the original gas-phase monoterpene oxidation chemistry in AM4.1 and the implementation does not

doubt count reductions of OH, O_3 and NO_3. There is little difference in the concentration of these gases between the CMPX and CTRL simulations. The gas-phase chemistry has been validated in Horowitz et al. (2020) and in Figure S3 in which we show summertime O_3 and NO_2 in SEUS well reproduce their observed decreasing trend."

In Page 4 Line 11, we add: "AM4.1 has 49 vertical levels from surface to 1Pa (~80km). We conduct AM4.1 simulations at a horizontal resolution of 1°×1.25° latitude by longitude."

In Page 5 Line 35, we add: "IMPROVE and SEARCH report daily average organic carbon measurements every 3 days. … and calculate monthly average of organic aerosol (OA) across these sites for each network."

In Page 5 Line 39, we add: "In Section 3.1, we calculate the absolute trend of a variable as the slope of the regression line of the variable's values versus time, and we calculate the relative trend (represented by "m" in Figure 1) as the absolute trend divided by the variable's 2000-2016 average."

In Page 6 Line 2, we add: "We average the original hourly OA measurement to monthly mean data for these sites to compare with modeling results."

Pag. 4, line 17-18: In my opinion, I think it is important to specify that LAI values follow an annual cycle (prescribed at the 1992 level, as precised by the authors).

In Page 4 Line 19, we add: "LAI values follow an annual cycle of the year 1992 and PFTs are prescribed at the 1992 level."

Sect. 3, Results

Pag. 6, line 27: To avoid confusion among readers, figures should be referenced in the order they appear in the text. Here, the authors refer to the Supplementary Figure S5, while in the next paragraph they refer to Fig. S3. For this reason, I suggest the authors to i) revise the order supplementary figures are presented in the Supplementary Material, and ii) comment all the supplementary figures (or remove those that are not commented in the manuscript).

Thanks! We revise and correct all supplement figure numbers accordingly.

Pag. 7, ll. 21: I suggest to briefly recall the other vegetated regions that have been selected for evaluation and that have been presented in Sect. 2.3.

In Page 7 Line 21, we add: "… in other vegetated regions in the Amazon, Europe and US (Figure 2)."

Sect. 4, Summary

I think that the application of the CMPX_ag scheme could be also interesting for local-regional studies on SOA production. For this reason, although Zheng et al. performed their study at the global scale, I think it could be useful for readers to know the computational cost of including the CMPX_ag scheme in their runs, compared to the cost of using the Simple or the CMPX schemes. This information could be provided in the Methods Section, or it could be commented in the Summary when discussing about perspectives.

As we apply a simplified aging scheme without adding more tracers, the CMPX_ag scheme does not increase the computational cost significantly relative to the CMPX simulation. The difference in their runtimes is almost negligible. In Page 5 Line 16, we add: "Including the aging scheme in CMPX does not increase computational cost notably."

MINOR COMMENTS

Through the whole manuscript: space is missing before parenthetical citations (e.g., pag.2, line 2: "BVOCs(Guenther et al., 2012))". Please add these spaces.

Corrected.

Sect. 1, Introduction

Pag. 3, ll. 2: Definition of the acronym CMPX and CMPX_ag are missing. Please define them.

We remove the names of simulations here and save them for the Methods Section. The sentence now is: "We use three schemes (summarized in Table 1 and detailed in Methods) to investigate the AIBS from decadal to centennial time scales."

Sect. 3, Results

Pag. 8, ll. 18: I think "is" is missing before the adjective "consistent".

The words "which is" are omitted here and should be fine.

Figures

Fig. 1: In the figure caption, I suggest to recall region boundaries, which are precised in the text (pag. 5, ll. 37). As well, I think it could be useful to recall in the caption the meaning of the different acronyms (ISOA, TSOA, ASOA)

We add the region boundaries and the definition of ISOA, TSOA and ASOA in the figure caption.