We are very grateful to the evaluations from the reviewers, which have helped us to clarify and improve the manuscript. Below we addressed the comments, with the comments in black and our response in blue. All the line numbers refer to the first version of the uploaded manuscript.

General comments and responses:

Comment#1. The argument that the model overestimation of OA at the surface and in summer is caused by an overrepresentation of monoterpene SOA production needs to be better substantiated or the limitations of this assertion better discussed. To be clear, I think the authors make a good suggestion by pointing to the large monoterpene SOA burden and the correlation with model bias. However, the correlation with model bias is not enough to make this argument. For instance, the authors point out that isoprene SOA also has a positive correlation with model bias. I don't completely follow why isoprene SOA was dismissed as a reason (Line 365). Isoprene also appears to have a seasonal cycle that peaks in the summer and the authors note the correlation with isoprene and model bias is also positive. Could the over prediction be due to both monoterpene and isoprene SOA yields? Additionally, the authors state in the Conclusions (Line 362) that the other POA and SOA components cannot explain the model bias; however, this does not appear to be explicitly shown in the Results section. I do see that monoterpene SOA dominants the OA composition in the summer (and I agree this is a good candidate for the cause of model overestimation), but I do not see a discussion that the other SOA species could not also contribute to the model overestimation. Again to be clear, I agree that the monoterpene SOA yield is a good suggestion for the cause of the model overestimation, but I feel this argument needs more context. This paper could be improved with a better discussion of this argument and its limitations or an additional simulation with altered monoterpene yields that reduced the model bias.

Response: We thank the reviewer for pointing out the limitation of the assertion and we agree that more discussion is necessary to clearly explain and describe the result. Our conclusion is that monoterpene-derived SOA is the largest contributor to modeling bias, thus it is urgently needed to improve the related chemical mechanism in CAM-chem. This conclusion was derived through two steps: first, we found that the spatiotemporal characteristics of simulation bias (Sect. 3.1: the eastern U.S. showed larger bias than the western U.S., summer showed larger bias than winter) indicated the biogenic VOCs played a dominant role over anthropogenic VOCs, because anthropogenic emissions of VOCs and POA wouldn't lead to such spatiotemporal characteristics. Second, the first finding inspired us to conduct sensitivity simulations to quantitatively estimate the contributions from every precursor through each chemical pathway to SOA (Sect. 3.2), and we found the monoterpene-derived SOA consisted 43% of the total SOA in summer, while none of the other precursors contributed more than 20%. Consequently, the 68% overestimation in summer shall most likely be attributed to monoterpene-related uncertainty. This comment reminds us that monoterpene might be a good candidate but other possibilities cannot be completely excluded. Just like the referee pointed out, the secondary largest contributor is isoprene-derived SOA, consist 17.0 % of total SOA in summer. Thus theoretically, the uncertainty in isoprene-derived SOA can also play an important role in the modeling bias. According to this comment, we have added a few brief discussions and rephrased our assertions in the manuscript. We also added discussion about the potential uncertainties related with isoprene-SOA treatment at line 329-340 with Figure 7. We have revised the paragraph at line 360-368 in the Sect. 4 as follows.

"Our analysis suggests that it is likely that simulated monoterpene SOA production is parameterized with too high yields and may be the most influential factor that affects the modeling bias for three reasons: first, monoterpene SOA contributes most (46.3 %) to the total SOA in summertime, while other anthropogenic POA or BVOCs have substantially smaller contributions. The large contribution of monoterpene SOA simulated by CAM6-Chem is consistent with other measurement and modeling studies, but the current VBS configuration adopted from GEOS-Chem may require further adjustment. Isoprene may also play an important role in modeling uncertainty but the influence is likely less significant than monoterpene as the isoprene-derived SOA consists 17.0 % of total SOA in summer. Second, the simulation bias showed a strong spatiotemporal correlation with monoterpene emission as demonstrated by the large overestimation in summer over eastern US, and larger overestimation of OA is found at places with higher NO_x condition under same monoterpene emissions level. Third, overestimation of OA at surface layer and underestimation of OA and monoterpene in the free troposphere suggests that both the production and photolytic removal processes might be parameterized too strong."

Comment#2. A large limitation of this study is that it focuses only on SOA production as opposed to any other chemical or physical processes in the model. Could the over estimation of model OA at the surface and underrepresentation of OA aloft point to an issue with vertical transport or removal? What about evaporation of OA - is this included in the model? I believe this paper would benefit from an explicit discussion of this limitation and how it affects the results.

Response: We agree with the reviewer the simulated mass concentration of OA is affected by many processes in addition to the chemical production of SOA. Other processes may also affect SOA, such as the biogenic and anthropogenic emissions, photolytic removal, dry and wet depositions, mixing, and transport. Modeling treatment for any of these processes may induce uncertainty to the final simulated mass concentrations of SOA. Lifetime of OA is about less than one week (Tsigaridis et al., 2014) owing to the rapid reaction rates of VOCs and the oxidants (OH, O_3, NO_3), thus the chemical production may play a more important role than other processes in the model. We intended to explore the remaining uncertainties within the chemical production because research community has been focusing on this topic during the past decades due to continuously improved understandings and new findings from lab and chamber studies. Development of SOA chemical production treatment in CAM-chem has been discussed in Tilmes et al. (2019) and Emmons et al. (2020), and there is also another paper by Jo et al. (2021) which reported the most recent update of SOA chemistry in CESM. But none of these studies provide a thorough evaluation of the model against surface observations at long-term scale, and a thorough understanding of the modeling bias is in absence. This is the major reason motivating our study, because it will help to clearly understand the performance and identify the most important causes for remaining uncertainties with the SOA chemistry. In contrast, other processes such as the deposition and mixing treatment of CESM are relatively better understood based on previous studies (Tilmes et al., 2019; Tsigaridis et al., 2014). It is inapplicable to elucidate all the related emission, physical, and chemical processes to identify and evaluate every potential source of uncertainties in one publication because that would be too long. Also, currently there is no exactly right answer for the SOA related processes as our understanding is still under development (Shrivastava et al., 2017). Thus, whenever the model could be adjusted to better describe the observed concentration of SOA, we have to make sure it is do so for the right reason. Identify and narrow down the bias in SOA chemistry can help us to build a better baseline, to further evaluate the uncertainties from other factors one by one and step by step.

We agree with the reviewer that vertical transport may affect the simulation results of VOCs and SOA. CESM can generally well reproduce the vertical profiles of meteorology variables (He et al., 2015). A recent sensitivity study (Gaubert et al., 2020) demonstrated the systematic underestimation of CO vertical profile in Asia was mainly due to bias in anthropogenic emission instead of the vertical transport scheme. In our case, the model showed larger upper air underestimations in the Eastern U.S. (EUS) than the Western U.S. (WUS), and likewise for surface overestimations. If vertical transport played an important role, it would more likely induce systematic bias leading to similar scale of errors in EUS and WUS. Thus, the discrepancy in vertical profile is more likely related to VOCs chemistry rather than transport issue.

Evaporation of OA is represented in the CESM model. One of the main functions of VBS is to calculate the thermodynamic equilibrium between gas-phase precursors (SOAG) and SOA, based on the volatilities of different species which are categorized into 5 groups (details were listed in Table 4) in CESM. Evaporation is not treated for primary emissions of OA (POA) in current version of CESM.

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Comment#3. The sensitivity simulations turn off chemical reactions one at a time; however, these chemical mechanisms are not necessarily linear (or additive). How does this assumption affect your results?

Response: We agree with the reviewer that many of the reactions are nonlinear, directly removing one reaction may drive the full mechanism to a different chemical regium and induce uncertainty to our estimation. To avoid the nonlinear-related uncertainty, in each of the sensitivity simulations we turned off a specific reaction by artificially set the SOA precursors (SOAG) yield to zero. This means the reaction was still simulated together with all other reactions, but the related SOAG was not produced. The related reactants (mainly OH, O3, and NO3) remains unaffected. For example, the reaction of monoterpene (MTERP) oxidized by OH is represented as:

MTERP + OH -> MTERP + OH + 0.0508*SOAG0 + 0.1149*SOAG1 + 0.0348*SOAG2 + 0.0554*SOAG3 + 0.1278*SOAG4

In the sensitivity simulation, the reaction was represented as:

MTERP + OH -> MTERP + OH + 0.0000*SOAG0 + 0.0000*SOAG1 + 0.0000*SOAG2 + 0.0000*SOAG3 + 0.0000*SOAG4

Gas phase MTERP and OH were still cycled through the sensitivity simulation, and since the precursors (SOAG) didn’t affect each other or get involved in chemical transformation, nonlinear uncertainty was not induced to the full chemical mechanism in each sensitivity simulation.

Comment#4. The Introduction Section would benefit from further discussion on SOA oxidation and chemistry (including the VBS scheme) as well as a literature review of previous studies focused on model/measurement comparison of OA. Additionally, a number of statements are lacking citations (see Specific Comments for examples). This topic (of OA representation in models) has been explored previously. As I mentioned at the start of this review, the continued challenge of representing OA in models certainly warrants continued study. That said, I believe this study could be improved by including a literature review of previous measurement/modeling studies in the Introduction. I note that the authors do point to and comment on previous studies in the Methods and Results Sections (which is great). However, I think the manuscript would be improved by clearly discussing relevant previous work in the introduction. This would improve the ability of readers to follow the comparisons in later sections. One such example of this, is that Hodzic et al. (2016), which is cited throughout this work, seems to argue for stronger SOA production rates and stronger SOA sinks. Conversely, this study seems to argue for the opposite. This is an interesting comparison that could use more context.

Response: We agree with the reviewer that a clearly discussion of relevant previous work in the introduction section would be helpful to present a big picture of the whole SOA modeling study. The first version of CAM-chem was introduced by Lamarque et al. (2012) in which the SOA chemistry was represented with the two-product method (Lack et al., 2004; Heald et al., 2008). The next big update was reported by Tilmes et al. (2019), in which the two-product method was replaced by VBS following the work by Hodzic et al. (2016). Although CAM-chem has been applied in many studies including the AeroCom program (Tsigaridis et al., 2014), the model hasn't been thoroughly evaluated against long-term ground-based measurements. Tilmes et al. (2019) validated the model with flight campaign data, which was a very commonly used and well-accepted method for global model evaluation. This method gives a snapshot of the mode performance since campaigns are limited in time period and space coverages. As the global models are getting finer grid resolution and more detailed description of atmospheric chemistry processes, it is necessary to reveal the performance with more details to help understand the remaining uncertainty after lots of new model developments. We have added this in the revised manuscript.

We greatly appreciate the time and efforts the referee devoted to review our submission. Hodzic et al. (2016) indeed argued for stronger SOA production and stronger sink through photolytic depletion, but that VBS implementation was based on GEOS-Chem. Since the gas-phase chemical mechanisms, dry and wet deposition schemes, heterogenous chemistry schemes are all different between GEOS-Chem and CAM-chem, even the same configuration of VBS may lead to different simulation results from two distinct modeling platforms. So, the stronger production configuration demonstrated in Hodzic et al. (2016) may not necessarily lead to good performance in CAM-chem, as has been revealed through our study in line 293-304. To identify the exact causes for different VBS performance the two models would require a comprehensive comparison between them. We are planning to do such a comparison in the future, but this manuscript is trying to understand the uncertainties in CAM-chem, and an explicit comparison is out of the scope of this study. We agree with the reviewer that a brief comparison and explanation is necessary in the context, and we have added such discussions in the introduction section.

Please see the modification of the paragraph at line 49-57 as follows.

"Modeling discrepancies largely come from the lack of a consensus in the representation of chemical composition and formation processes of OA among different models (Tsigaridis et al., 2014; Goldstein and Galbally, 2007). Although laboratory and chamber studies have explicitly revealed thousands of new reactions and new species related to VOCs and SOA, these reactions and species are usually simplified and grouped into a few functions and lumped to fewer species in the models to make it possible for numeric simulating. In addition, many unclear SOA formation processes have to be approximated as the knowledge is still under development (Kanakidou et al., 2004; Hallquist et al., 2009). Thus, different models may use different simplified functions, definitions for lumped species, and approximation methods to represent the same SOA related processes. SOA production was represented with the two-product method (Lack et al., 2004; Heald et al., 2008) in the earlier model (Lamarque et al., 2012). Since the late 2000s, Volatility Basis Set (VBS) methods (Donahue et al., 2006; Robinson et al., 2007) have been widely adopted by different models due to the advantages over two-products method for considering the volatility (Lack et al., 2004; Heald et al., 2008). Since the 2010s, pilot studies started to include reactive uptake of isoprene epoxydiols (IEPOX) formation of SOA through aqueous-phase reactions into regional (Shrivastava et al., 2019; Karambelas et al., 2014) or global models (Marais et al., 2016; Zheng et al., 2020; Jo et al., 2019). Despite the tremendous efforts in the early stage, models still underpredicted measured SOA mass concentration by 1-2 orders of magnitude. Hodzic et al. (2016) therefore suggested corrected stronger yields of SOA formation which took into account the influence of vapor wall losses in chamber studies and were considered in Tilmes et al. (2019). However, due to the different gas-phase chemistry, dry and wet deposition schemes, and heterogenous chemistry schemes, the simulated OA may be different for the same VBS configuration in different models (Hodzic et al., 2016; Tilmes et al., 2019). These attempts not only validated the parameterizations and chemical pathways derived from measurement studies, but also extended the understanding of SOA formation on a scale broader than the chamber. Once formed, most of OA undergoes chemical aging via SOA formation (Zhang et al., 2007) with volatility and hygroscopicity changing, but such processing is poorly understood due to inadequate relevant observations. While some models consider species-dependent aging reactions and the subsequent volatility change, some simply apply a constant aging rate (Tsigaridis et al., 2014; Jo et al., 2013; Donahue et al., 2006; Zhao et al., 2016). In the remote areas of the United States (U.S.), OA and organic carbon (OC) concentrations show opposite bias in half of participating models (Tsigaridis et al., 2014), indicating the models lack a consensus representation of SOA production and depletion."

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Specific comments and responses:

Comment#1. I do not think an appropriate color scale was chosen for Figure 1. While red and blue are certainly appropriate for opposite ends of a diverging color scale, it is not immediately obvious to me that green should be opposite yellow. I suggest the authors use a standard diverging color scale (or simply shades of red and blue on each side).

Response: We intended to use warm colors for positive bias and cool colors for negative bias. We have changed the figure to use red and blue as shown below.

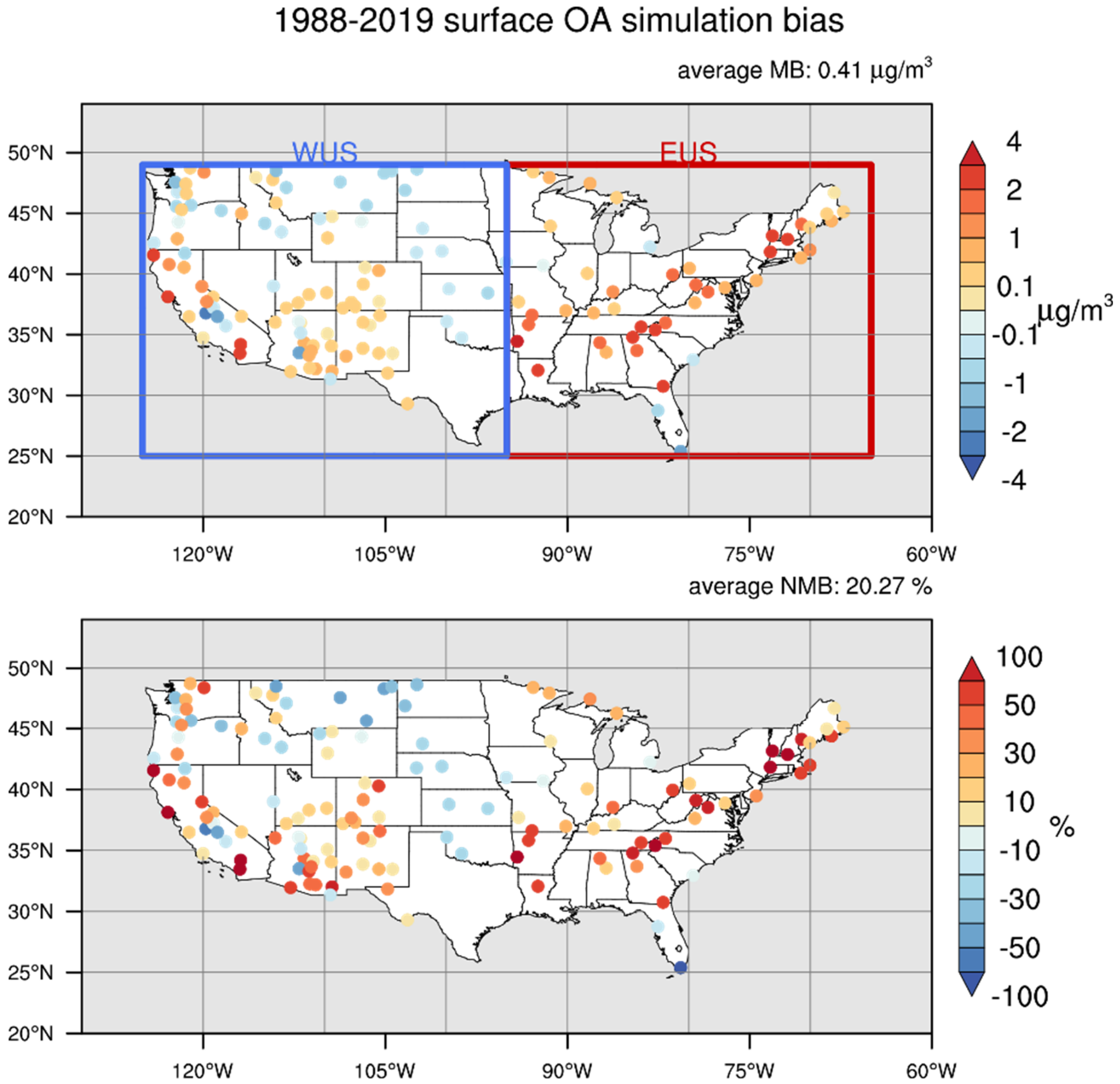

Figure 1: 1998–2019 CAM-Chem-SD surface OA concentration mean bias (top figure, unit: µg/m^3) and normalized mean bias (bottom figure, unit: %) compared to IMPROVE data. CONUS is divided into two subdomains, EUS (red box) and WUS (blue box).

Comment#2. The units of the color scale in Figure 1 could be more obvious. I suggest including the units as the color bar label itself or at least in the caption.

Response: We apologize for improper color label and missing units in Fig. 1. We modify color labels and include the units of each subplot in the Fig. 1 and its caption. The caption is revised as shown in the response for specific comment#1.

Comment#3. The legend of Figure 4 is much too small to be readable.

Response: We apologize for the unclear legends in Fig.4. We now use bigger size of legend and higher DPI for the whole figure, thus readers can zoom in it if necessary. Please see the figure below.

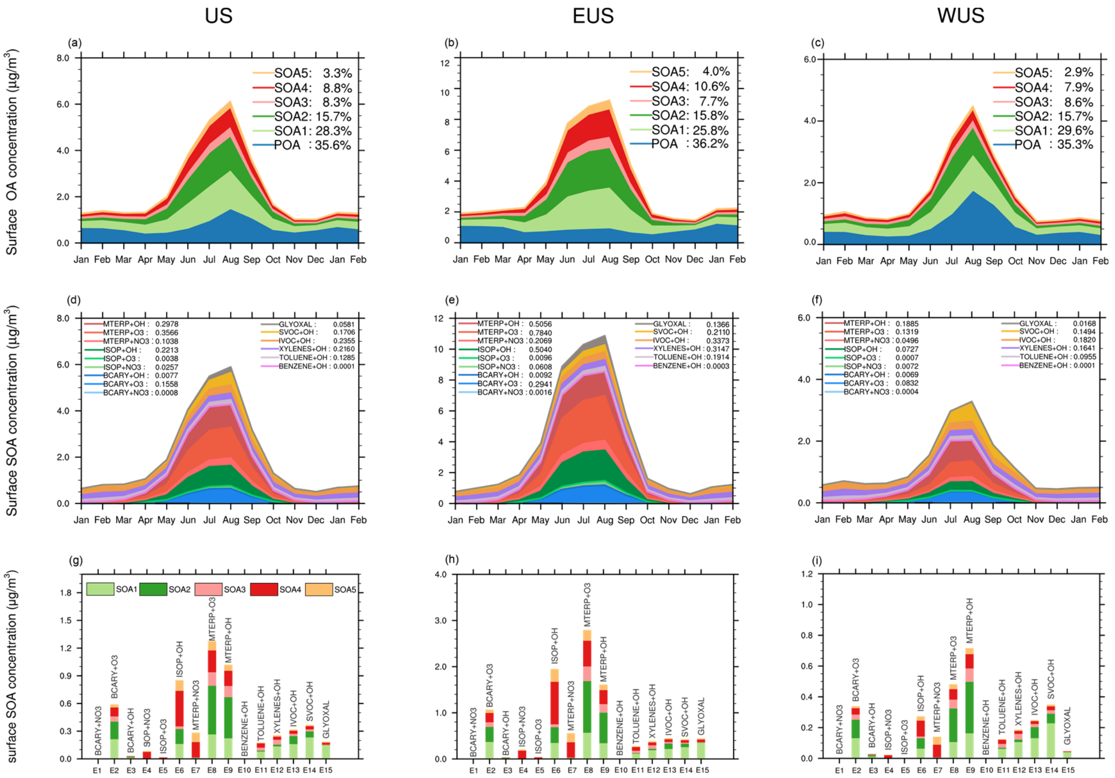

Figure 4: (a)~(c) Seasonal variation of surface OA concentration from CAM-Chem-climo simulation at all IMPROVE sites located over (a) CONUS, (b) EUS and (c) WUS. The contribution of POA and SOA in five VBS bins are shown in filled areas. The 14-month average contribution of POA and SOA are shown as the number in the legend. (d)~(f) Seasonal variation of SOA formed by 15 different pathways over (d) CONUS, (e) EUS and (f) WUS. The contribution of 15 reactions are shown in filled areas and 14-month average contribution is shown as the number in the legend. (g)~(i) the contributions from each pathway to the five SOA bins over (g) CONUS, (h) EUS and (i) WUS in 2010 July. SOA concentration in 5 bins are shown in filled bars.

Comment#4. The panel labels in the caption of Figure 4 are inconsistent. Sometimes the panel label follows the description while other times it precedes the description.

Response: We apologize for the inconsistent panel label of Fig. 4. We modify the description of Fig.4 followed by the panel label. Please see the response of comment#3.

Comment#5. The caption in Figure 7 points to the wrong color (I think it should read "black line" instead of red).

Response: Thanks for your comments. We modify the caption of Fig. 7. Please see the figure and its caption below.

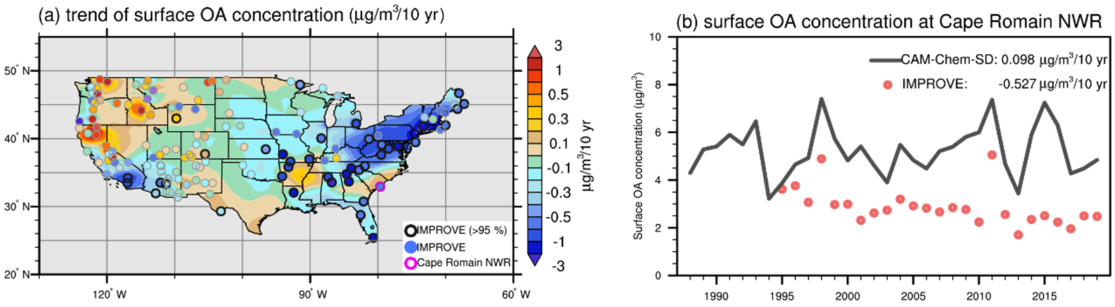

Figure 7: 1988–2019 JJA (a) simulated surface OA decade trend and IMPROVE surface OA decade trend at selected IMPROVE sites (filled circles). Black circles indicate sites with statistically significant trends with 95 % confidence according to the student's T-test; (b) simulated (black lines) and observed (red dots) surface OA concentration at Cape Romain NWR site (shown as purple circle in Fig. 7(a)). Simulated and observed OA decade trends (unit: μg/m^3/10 year) in summer at the site are shown as the numbers in the legend.

Comment#6. The units in Figure 7a are never stated. Please be explicit about units in all figures.

Response: We apologize for the typo in the figures. We include units in Fig. 7 and its caption. Please see the response of comment#5.

Comment#7: Table 3 is difficult to read. Could the different regions be grouped in a more obvious way?

Response: We modify Table 3 by adding extra space between different regions, as shown below.

Table 3: The correlation coefficient (CC), mean bias (MB) and normalized mean bias (NMB) between observations (five field campaigns and IMPROVE surface measurements) and CAM-Chem-SD.

Observations		**CC**	**Mean Obs. (μg/m^3)**	**Mean Sim. (μg/m^3)**	**MB (μg/m^3)**	**NMB (%)**
			IMPROVE			
CONUS	Annual	0.40	2.07	2.48	0.41	20.27
	Spring	0.67	1.65	1.57	-0.08	-4.81
	Summer	0.37	2.90	4.87	1.97	68.78
	Fall	0.34	2.23	2.23	-0.01	0.13
	Winter	0.70	1.49	1.22	-0.27	-19.05
EUS	Annual	0.79	2.64	3.72	1.08	40.82
	Spring	0.64	2.49	2.66	0.17	6.71
	Summer	0.60	3.26	7.52	4.26	131.15
	Fall	0.69	2.63	2.88	0.25	9.70
	Winter	0.82	2.25	1.82	-0.43	-19.11
WUS	Annual	0.36	1.78	1.89	0.11	10.49
	Spring	0.77	1.23	1.04	-0.19	-15.36
	Summer	0.48	2.72	3.66	0.94	34.83
	Fall	0.35	2.03	1.91	-0.12	-5.85
	Winter	0.73	1.12	0.91	-0.21	-18.00
			Aircraft			
CalNex		0.43	2.06	1.46	-0.60	-29.01
DC3		0.12	2.99	0.72	-2.17	-72.75
SENEX		0.33	7.09	4.22	-2.87	-40.54
SEAC4RS		0.10	6.90	1.93	-4.97	-71.97
FRAPPE		0.27	3.05	2.42	-0.63	-20.64

Comment#8. Please add units to Table 3.

Response: We add the units into Table 3 as shown in the response of comment#7.

Comment#9. This is a minor comment - the citations need spaces after the semicolon.

Response: Thanks for this comment. We add spaces after the semicolon in the citations.

Comment#10. The sentence at Line 51 is unclear. Are you comparing the model representation to other model processes or stating the reasons why model representation of OA is challenging?

Response: We apologize for the unclear description. In this sentence, we intended to state that model discrepancies are due to different representations of OA among the models, and this is because our knowledge of VOCs and SOA related chemistry is still under development and lacks a consensus understanding. We have revised the sentence as shown in the response for general comment#4.

Comment#11. Lines 51-57 should be edited or revised for clarity. I am not sure what is meant by these sentences.

Response: We totally agree with the unclear information at Line 51-57. In this paragraph, we want to clarify the model discrepancies comes from different SOA representations among models which are lack of some processes compared to chamber studies. We have revised the sentence as shown in the response for general comment#4.

Comment#12. Citations are needed for the comments on OA and climate impacts (such as on Line 41 and Line 60).

Response: We apologize for the missing citations for the comments on OA and climate impacts. Citations are included in the sentence at Line 41.

"The radiative forcing effect of OA has been assessed with climate models through tremendous efforts during the past decades (Ghan et al., 2012; Myhre et al., 2013; Sporre et al., 2020; Chen and Gettelman, 2016), yet the limited capability of climate models in terms of simulating the productions and depletions of OA induce large uncertainties."

Citations are also included in the sentence at Line 60.

"The model has been widely applied for OA climate effect assessment purpose (A. Gettelman et al., 2019; Jo et al., 2021; Glotfelty et al., 2017; Tilmes et al., 2019) and a significant portion of improvements have been implemented in the latest version regarding the chemical mechanisms (Tilmes et al., 2019), but evaluation of simulated OA concentration hasn't been well documented or thoroughly discussed."

Reference:

A. Gettelman, M. J. Mills, D. E. Kinnison , R. R. Garcia, A. K. Smith1 , D. R. Marsh , S. Tilmes , F. Vitt , C. G. Bardeen , J. McInerny, H.-L. Liu , S. C. Solomon , L. M. Polvani , L. K. Emmons , J.-F. Lamarque , J. H. Richter , A. S. Glanville, J. T. Bacmeister, A. S. Phillips , R. B. Neale , I. R. Simpson , A. K. DuVivier , A. Hodzic , and Randel, a. J.: TheWhole Atmosphere Community Climate Model Version 6 (WACCM6), 10.1029/2019JD030943, 2019.

Chen, C.-C., and Gettelman, A.: Simulated 2050 aviation radiative forcing from contrails and aerosols, Atmospheric Chemistry and Physics, 16, 7317-7333, 10.5194/acp-16-7317-2016, 2016.

Ghan, S. J., Liu, X., Easter, R. C., Zaveri, R., Rasch, P. J., Yoon, J. H., and Eaton, B.: Toward a Minimal Representation of Aerosols in Climate Models: Comparative Decomposition of Aerosol Direct, Semidirect, and Indirect Radiative Forcing, Journal of Climate, 25, 6461-6476, 10.1175/jcli-d-11-00650.1, 2012.

Glotfelty, T., He, J., and Zhang, Y.: Improving organic aerosol treatments in CESM/CAM5: Development, application, and evaluation, J Adv Model Earth Syst, 9, 1506-1539, 10.1002/2016MS000874, 2017.

Jo, D. S., Hodzic, A., Emmons, L. K., Tilmes, S., Schwantes, R. H., Mills, M. J., Campuzano-Jost, P., Hu, W., Zaveri, R. A., Easter, R. C., Singh, B., Lu, Z., Schulz, C., Schneider, J., Shilling, J. E., Wisthaler, A., and Jimenez, J. L.: Future changes in isoprene-epoxydiol-derived secondary organic aerosol (IEPOX SOA) under the Shared Socioeconomic Pathways: the importance of physicochemical dependency, Atmos. Chem. Phys., 21, 3395-3425, 10.5194/acp-21-3395-2021, 2021.

Myhre, G., Samset, B. H., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Chin, M., Diehl, T., Easter, R. C., Feichter, J., Ghan, S. J., Hauglustaine, D., Iversen, T., Kinne, S., Kirkevåg, A., Lamarque, J. F., Lin, G., Liu, X., Lund, M. T., Luo, G., Ma, X., van Noije, T., Penner, J. E., Rasch, P. J., Ruiz, A., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., Wang, P., Wang, Z., Xu, L., Yu, H., Yu, F., Yoon, J. H., Zhang, K., Zhang, H., and Zhou, C.: Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, Atmospheric Chemistry and Physics, 13, 1853-1877, 10.5194/acp-13-1853-2013, 2013.

Sporre, M. K., Blichner, S. M., Schrödner, R., Karset, I. H. H., Berntsen, T. K., van Noije, T., Bergman, T., O'Donnell, D., and Makkonen, R.: Large difference in aerosol radiative effects from BVOC-SOA treatment in three Earth system models, Atmospheric Chemistry and Physics, 20, 8953-8973, 10.5194/acp-20-8953-2020, 2020.

Tilmes, S., Hodzic, A., Emmons, L. K., Mills, M. J., Gettelman, A., Kinnison, D. E., Park, M., Lamarque, J. F., Vitt, F., Shrivastava, M., Campuzano-Jost, P., Jimenez, J. L., and Liu, X.: Climate Forcing and Trends of Organic Aerosols in the Community Earth System Model (CESM2), Journal of Advances in Modeling Earth Systems, 10.1029/2019ms001827, 2019.

Comment#13. Citations are needed for the sentences that begin on Line 49, 50, and 52.

Response: Please see the response for general comment#4 in which citations are included.

Comment#14. Line 80/81 - I think the papers by Donahue et al. (2006) and Robinson et al. (2007) should be included in the citation for the VBS.

Response: We apologize for missing citations for the VBS. The citations are included in Line 80/81.

“CAM6-Chem applies the Volatility Basis Set (VBS) scheme (Bergström et al., 2012; Hodzic et al., 2016; Shrivastava et al., 2011; Shrivastava et al., 2013; Donahue et al., 2006; Robinson et al., 2007) by lumping SOA precursors based on their volatility bins to simulate SOA production.”

Reference:

Bergström, R., Denier van der Gon, H. A. C., Prévôt, A. S. H., Yttri, K. E., and Simpson, D.: Modelling of organic aerosols over Europe (2002-2007) using a volatility basis set (VBS) framework: application of different assumptions regarding the formation of secondary organic aerosol, Atmospheric Chemistry and Physics, 12, 8499-8527, 10.5194/acp-12-8499-2012, 2012.

Donahue, N. M., Robinson, A. L., Stannier, C. O., and N., P. S.: Coupled partitioning, dilution, and chemical aging of semivolatile organics, Environ. Sci. Technol., 2006.

Hodzic, A., Kasibhatla, P. S., Jo, D. S., Cappa, C. D., Jimenez, J. L., Madronich, S., and Park, R. J.: Rethinking the global secondary organic aerosol (SOA) budget: stronger production, faster removal, shorter lifetime, Atmospheric Chemistry and Physics, 16, 7917-7941, 10.5194/acp-16-7917-2016, 2016.

Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop, A. P., Lane, T. E., Pierce, J. R., and Pandis, S. N.: Rethinking Organic Aerosols: Semivolatile Emissions and Photochemical Aging, Science, 315, 1259, 10.1126/science.1133061, 2007.

Shrivastava, M., Fast, J., Easter, R., Gustafson, W. I., Zaveri, R. A., Jimenez, J. L., Saide, P., and Hodzic, A.: Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach, Atmospheric Chemistry and Physics, 11, 6639-6662, 10.5194/acp-11-6639-2011, 2011.

Shrivastava, M., Zelenyuk, A., Imre, D., Easter, R., Beranek, J., Zaveri, R. A., and Fast, J.: Implications of low volatility SOA and gas-phase fragmentation reactions on SOA loadings and their spatial and temporal evolution in the atmosphere, Journal of Geophysical Research: Atmospheres, 118, 3328-3342, 10.1002/jgrd.50160, 2013.

Comment#15. Line 85 - minor typo with the comma

Response: We apologize for an extra space at Line 85. The sentence begun at Line 84 is modified to “Compared with the simple SOA scheme which proportionally calculated SOAG based on emissions of precursors (Liu et al., 2012), this VBS scheme was demonstrated to improve CESM performance with smaller bias of OA concentration over remote regions when evaluated against aircraft observations (Shrivastava et al., 2015; Tilmes et al., 2019).”.

Reference:

Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J. F., Gettelman, A., Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C., Ekman, A. M. L., Hess, P., Mahowald, N., Collins, W., Iacono, M. J., Bretherton, C. S., Flanner, M. G., and Mitchell, D.: Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5, Geoscientific Model Development, 5, 709-739, 10.5194/gmd-5-709-2012, 2012.

Shrivastava, M., Easter, R. C., Liu, X., Zelenyuk, A., Singh, B., Zhang, K., Ma, P.-L., Chand, D., Ghan, S., Jimenez, J. L., Zhang, Q., Fast, J., Rasch, P. J., and Tiitta, P.: Global transformation and fate of SOA: Implications of low-volatility SOA and gas-phase fragmentation reactions, Journal of Geophysical Research: Atmospheres, 120, 4169-4195, 10.1002/2014jd022563, 2015.

Tilmes, S., Hodzic, A., Emmons, L. K., Mills, M. J., Gettelman, A., Kinnison, D. E., Park, M., Lamarque, J. F., Vitt, F., Shrivastava, M., Campuzano-Jost, P., Jimenez, J. L., and Liu, X.: Climate Forcing and Trends of Organic Aerosols in the Community Earth System Model (CESM2), Journal of Advances in Modeling Earth Systems, 10.1029/2019ms001827, 2019.

Comment#16. Line 124 - Could you elaborate (briefly) on what a "specified dynamical" simulation is? In addition, please define "FCSD" and "FC2010climo".

Response: A "specified dynamical" simulation in Line 124 is referred to the FCSD simulation where meteorological fields is read from the input meteorological fields from MERRA2 reanalysis dataset for detailed comparisons to field experiments and specific observations. The meteorological fields here are referred to temperature, horizontal winds, and surface fluxes.

"FCSD" and "FC2000climo" are defined component sets of CESM which define meteorological field, emissions, physical and chemistry schemes used in each module. The information of "FCSD" and "FC2000climo" are shown in the table below. These 2 component sets differ in terms of emission and meteorological fields used in simulation.

Component set	**FCSD**	**FC2000climo**
Emission	Historical emission depending on simulation period	Present day (year 2000) emission
Physics of atmosphere model	CAM6	CAM6
Chemistry of atmosphere model	CAM-Chem troposphere/stratosphere chemistry with simplified VBS-SOA	CAM-Chem troposphere/stratosphere chemistry with simplified VBS-SOA

Meteorological fields	Nudged with MERRA2 dataset	Free running
Land model	CLM5	CLM5

We have revised the sentence in line 125-127 as follows.

"This experiment (referred as CAM-Chem-SD) uses FCSD component set in which CAM6 physics, troposphere/stratosphere chemistry (MOZART-TS1) with VBS SOA scheme, historical emission, and offline meteorological field are applied. In details, Temperature, horizontal winds, and surface fluxes are nudged to Modern-Era Retrospective analysis for Research and Applications (MERRA2) fields for detailed comparisons to field experiments and specific observations."

Comment#17. L 152 - I do not think the word choice of "critical" simulation bias is correct. Do you mean "large" or "substantial"?

Response: We agree with the uncorrected word used in the sentence at line 151-153. In the sentence at line 151-153, we are supposed to show apparent simulation bias of surface OA concentration as shown in Fig.1. We modify "critical simulation bias" to "substantial simulation bias" at line 152.

Comment#18. L 173 - I suggest changing "descending" trend to "decreasing" trend.

Response: We totally agree that the trend of simulated surface OA concentration in WUS is not obviously decreasing and modify "descending tread" to "decreasing trend" at line 173.

Comment#19. L 173 - I think this is a really interesting point that needs a little clarification. Do you mean that the increasing wildfires are leading to increasing trends in observations but the wildfire emissions are not included in the model and so the trend is not represented?

Response: Wildfire emissions are included in the model simulation, but the climate-fire-ecosystem interactions are not well represented in the climate models (Zou et al., 2020). For example, Zou et al. (2020) pointed out that wildfire is driven by offline statistical regression or one-way coupled prognostic fire models, while feedback to weather, climate, and vegetation was neglected in CESM.

In the sentence at line 173, we are supposed to show the opposite trend of surface OA concentration between simulation and IMPROVE dataset in summertime over WUS region. Wildfires lead to large inter-annual variation of observed surface OA concentration as discussed in Malm et al. (2017). The simulated OA also has large inter-annual variation but did not shows increasing trend due to lower value after 2017. The wildfire emissions from the CMIP6 emissions (Feng et al., 2020) are included in CAM-Chem-SD. It needs to be emphasized that historical emissions are used in CAM-Chem-SD simulation from 1987 to 2014 and SSP585 emissions after 2014 as clarify at line 129-132. The SSP585 emissions is based on shared socioeconomic pathway 5 (SSP5) (O'Neill et al., 2017) and forcing levels of Representative Concentration Pathways 8.5 (RCP8.5), which means the emissions do not exactly match the observed condition. Therefore, both of the simulated and observed OA show increasing trend in summertime over WUS region before 2014, but opposite trend after 2014. We have rephrased this sentence in 171-173 to avoid misunderstanding as follows.

"In WUS, simulated OA shows a slow decreasing trend in summer (-0.02 μg/m3 per decade) while the observations indicate an ascending trend (0.23 μg/m3 per decade). The large inter-annual variation from 1999 to 2019 are shown in observed surface OA concentration mainly due to the influence of wildfires (Malm et al., 2017). The simulated surface OA concentration also has large inter-annual variation but do not shows increasing trend due to lower value after 2017. It needs to be emphasized that historical emissions are used from 1987 to 2014 and SSP585 emissions after 2014 in CAM-Chem-SD simulation, which means the emissions do not exactly match the observed condition after 2014."

Reference:

Feng, L., Smith, S. J., Braun, C., Crippa, M., Gidden, M. J., Hoesly, R., Klimont, Z., van Marle, M., van den Berg, M., and van der Werf, G. R.: The generation of gridded emissions data for CMIP6, Geoscientific Model Development, 13, 461-482, 10.5194/gmd-13-461-2020, 2020.

Malm, W. C., Schichtel, B. A., Hand, J. L., and Collett, J. L.: Concurrent Temporal and Spatial Trends in Sulfate and Organic Mass Concentrations Measured in the IMPROVE Monitoring Program, Journal of Geophysical Research: Atmospheres, 122, 10,462-410,476, 10.1002/2017jd026865, 2017.

O'Neill, B. C., Kriegler, E., Ebi, K. L., Kemp-Benedict, E., Riahi, K., Rothman, D. S., van Ruijven, B. J., van Vuuren, D. P., Birkmann, J., Kok, K., Levy, M., and Solecki, W.: The roads ahead: Narratives for shared socioeconomic pathways describing world futures in the 21st century, Global Environmental Change, 42, 169-180, 10.1016/j.gloenvcha.2015.01.004, 2017.

Zou, Y., Wang, Y., Qian, Y., Tian, H., Yang, J., and Alvarado, E.: Using CESM-RESFire to understand climate–fire–ecosystem interactions and the implications for decadal climate variability, Atmospheric Chemistry and Physics, 20, 995-1020, 10.5194/acp-20-995-2020, 2020.

Comment#20. L 184 - Is the current study not also influenced by the bias of evaporation of OA off filters as in Hodzic et al. (2016)?

Response: IMPROVE has been used in numerous studies and some of them reported possible measurement bias. For example, a summertime low bias of IMPROVE dataset, for about 25% (Kim et al., 2015), is due to evaporation of OA from the filter after collection (Zheng et al., 2020). But the official release of IMPROVE measurement doesn't recommend any adjustment, and many other studies directly adopt the data too. Thus, we include this information in the main text only to point out this potential bias within the measurement. Since the regional average NMB over CONUS (68.78 %) in summer is exceedingly larger than the bias of evaporation of OA off filters, considering the IMPROVE bias won't change the conclusion of this study.

Reference:

Kim, P. S., Jacob, D. J., Fisher, J. A., Travis, K., Yu, K., Zhu, L., Yantosca, R. M., Sulprizio, M. P., Jimenez, J. L., Campuzano-Jost, P., Froyd, K. D., Liao, J., Hair, J. W., Fenn, M. A., Butler, C. F., Wagner, N. L., Gordon, T. D., Welti, A., Wennberg, P. O., Crounse, J. D., St. Clair, J. M., Teng, A. P., Millet, D. B., Schwarz, J. P., Markovic, M. Z., and Perring, A. E.: Sources, seasonality, and trends of southeast US aerosol: an integrated analysis of surface, aircraft, and satellite observations with the GEOS-Chem chemical transport model, Atmospheric Chemistry and Physics, 15, 10411-10433, 10.5194/acp-15-10411-2015, 2015.

Zheng, Y., Thornton, J. A., Ng, N. L., Cao, H., Henze, D. K., McDuffie, E. E., Hu, W., Jimenez, J. L., Marais, E. A., Edgerton, E., and Mao, J.: Long-term observational constraints of organic aerosol dependence on inorganic species in the southeast US, Atmos. Chem. Phys. Discuss., 2020, 1-39, 10.5194/acp-2020-575, 2020.

Comment#21. Line 321 - Is this an entirely new model configuration and simulation? If so, I recommend including this model configuration in the Methods Section. If I understand correctly, this simulation includes SOA production schemes that were suggested as part of the model bias in the previous paragraph. This seems like an important result that should be given more discussion.

Response: Thanks for the reminder. This is an entirely new configuration and very recent update reported in Jo et al. (2021). The brief information of MOZART-TS2 mentioned in line 321 has been introduced at line 76-78. We add the description of CAM-Chem-SD (TS2) simulation at the end of line 134.

"Moreover, another simulation of CESM2.2 which is the latest released version of CESM, referred to CAM-Chem-SD (TS2), is conducted with MOZART-TS2 gas phase chemistry (Schwantes et al., 2020) from January 2013 to February 2014 with first 2 months as spin-up time. The FCSD component set is also used in CAM-Chem-SD (TS2). Except for the difference of gas phase chemistry, the SOA scheme is also improved in CAM-Chem-SD (TS2) compared with CAM-Chem-SD. The NOx dependence of SOA formation in CAM-Chem-SD (TS2) is not considered in CAM-Chem-SD. Thus, we compared CAM-Chem-SD and CAM-Chem-SD (TS2) to investigate the impact of NOx dependence on SOA formation."

We also modify Table 2 to include the experiment information of CAM-Chem-SD (TS2) as follows.

Table 2: CESM experiments used in this study

Index	**Experiment ID**
B1	CAM-Chem-SD
B2	CAM-Chem-SD (TS2)
B3	CAM-Chem-climo
E1	no ISOP+OH
E2	no ISOP+O3
E3	no ISOP+NO3
E4	no MTERP+OH
E5	no MTERP+O3
E6	no MTERP+NO3
E7	no BCARY+OH
E8	no BCARY+O3
E9	no BCARY+NO3
E10	no BENZENE+OH
E11	no TOLUENE+OH
E12	no XYLENES+OH
E13	no IVOC+OH
E14	no SVOC+OH
E15	no GLYOXAL

In Sect. 3.3, we are discussing the possible factors that leads to the simulation bias of surface SOA concentration. The 2 major factors are strong monoterpene mass yields which is discussed before line 299 and NOx dependence in VBS scheme. CAM-Chem-SD(TS2) experiment is conducted to show that less SOA is formed when NOx dependence is considered in VBS scheme. Therefore, we compare CAM-Chem-SD and CAM-Chem-SD(TS2) in the paragraph begun at line 299 to discuss the influence of NOx dependence in SOA concentration. It shows when NOx dependence is considered in VBS scheme, surface OA concentration decrease but is still larger than observation, which means the strong monoterpene SOA yield is the main factor that leads to modeling bias.

Reference:

Schwantes, R. H., Emmons, L. K., Orlando, J. J., Barth, M. C., Tyndall, G. S., Hall, S. R., Ullmann, K., St. Clair, J. M., Blake, D. R., Wisthaler, A., and Bui, T. P. V.: Comprehensive isoprene and terpene gas-phase chemistry improves simulated surface ozone in the southeastern US, Atmospheric Chemistry and Physics, 20, 3739-3776, 10.5194/acp-20-3739-2020, 2020.

Jo, D. S., Hodzic, A., Emmons, L. K., Tilmes, S., Schwantes, R. H., Mills, M. J., Campuzano-Jost, P., Hu, W., Zaveri, R. A., Easter, R. C., Singh, B., Lu, Z., Schulz, C., Schneider, J., Shilling, J. E., Wisthaler, A., and Jimenez, J. L.: Future changes in isoprene-epoxydiol-derived secondary organic aerosol (IEPOX SOA) under the Shared Socioeconomic Pathways: the importance of physicochemical dependency, Atmos. Chem. Phys., 21, 3395-3425, 10.5194/acp-21-3395-2021, 2021.

Comment#22. Line 356-357 is confusing. Is the second parenthetical placed correctly?

Response: We apologize for the misleading of the sentence at line 356-357. The sentence is modified to “Larger summer time bias is found over eastern US (overestimates by 131.15 %) where BVOCs emissions are more intensive than western US.”.

Comment#23. Line 368 states “...and photolytic removal processes might be too strong”. I do not follow why this is an argument in support of monoterpene SOA production being too high. It seems like it argues that bias is not entirely due to SOA production rates in contrast to the point of this paragraph.

Response: In this paragraph, we explained the three reasons of modeling bias in summertime. For the third reason, high SOA production rates lead to high SOA concentration, while strong photolytic removal processes lead to low SOA concentration. If both production and removal processes are configured too strong, the model is likely to show overestimation near source region (surface layer, near VOCs emission area) but underestimation in remote region (upper air, far from VOCs emission sources). Therefore, we conclude that CESM2.1 overestimated surface OA concentration and underestimated SOA concentration at higher altitude in summer.