We are very grateful to the evaluations from the reviewers, which have allowed us to clarify and improve the manuscript. Below we addressed the reviewer comments, with the reviewer comments in black and our response in blue.

Reply for the referee comment#1

General comments: This is an interesting and well written paper. They study evaluates organic aerosol in CESM2.1 over the United States by comparing the model to long term surface measurements and aircraft campaigns. The authors find that the model overestimate organic aerosol during summer. Moreover, the model comparison with flight campaigns reveal that the model underestimate organic aerosol in the upper air. The authors conclude that these results could be explained by too high monoterpene SOA yields which result in too strong SOA production close to monoterpene sources. The topic of this paper falls well in the scope of ACP. The scientific methods in the paper are sound and well explained. The authors explain and discuss the results in the figures and tables in a clear and interesting manner. I recommend the paper for publication in ACP after the following comments have been addressed.

General Response: We greatly appreciate the referee for his/her time and efforts devoted to the review of our submission. We realize that most of the comments are due to the missing details of model description. We will present these details in this document as shown in the following responses.

Specific comments and responses:

Comment#1: The model used in this study, CAM6-Chem differs from the standard CAM6 since it has a more advanced chemistry. My impression is that CAM6 is the standard atmospheric model in CESM2.1. Could you describe to what extent CAM6-Chem is used in comparison to CAM6? It would be beneficial to better clarify the differences between CAM6 and CAM6-Chem in the methods section. In part of the method, you describe the changes in CESM2.1. It would be nice to refer to CAM6 or CAM6-Chem instead, as the text is currently written it is difficult to know if the VBS scheme is included in both CAM6 and CAM6-Chem or only in the latter. Moreover, you have evaluated CAM6-Chem, but it would be interesting to know how well CAM6 performs in comparison to CAM6-Chem with respect to organic aerosol. What are the differences in performance between CAM6-Chem and CAM6 in terms of organic aerosol?

Response: CESM2 (versions 2.0 and 2.1) supports two atmospheric model configurations, the Whole Atmosphere Community Climate Model version 6 (WACCM6) with 72 vertical layers up to about 150 km and the Community Atmosphere Model version 6 (CAM6) with 32 vertical layers up to about 40 km. CAM6 has simplified chemistry and simplified OA scheme, while CAM6 with comprehensive chemistry and comprehensive OA scheme are called CAM6-Chem. The differences between CAM6 and CAM6-Chem are included in the methods section. Due to the simplified chemistry and simple OA scheme, CAM has been used to explore physical processes, like cloud and precipitation processes (English et al., 2014), while CAM-Chem has been used to simulate specific chemical species and explore its climate effect (Schwantes et al., 2020; Tilmes et al., 2019; Jo et al., 2020). Our study focuses on the simulation performance of OA presented with VBS scheme, which is only included in CAM-Chem, not in CAM. We have changed the term "CESM2.1" to "CAM6-Chem" in method section and latter parts to avoid misunderstanding according to this comment. More detailed description of the differences between CAM and CAM-chem are reported in Tilmes et al. (2019), which comprehensively compared the difference between CAM6, WACCM, and WACCM6-Chem. WACCM6-Chem has almost exactly the same chemistry with very minor difference. Regarding the different performance of organic aerosol between CAM6-chem and CAM: CAM uses prescribed aerosol without comprehensive chemistry or SOA scheme and the simulation of aerosol is only used to serve radiative forcing and cloud, thus CAM is not compared to observation for organic aerosol.

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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 explicit chemistry, Atmos. Chem. Phys. Discuss., 2020, 1-57, 10.5194/acp-2020-543, 2020.

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.

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.

Conment#2: The model is only compared to observations over the United States. Could you comment on the limitations of this and if the model has been evaluated in other locations in any other studies.

Response: In this manuscript we focused on evaluation over the United States mainly because it has the best public accessible long-term observation data to support the evaluation. The aim of this paper is to reveal the performance of CESM2.1 in OA simulation which has not been thoroughly discussed in other studies to the best of our knowledge. Tsigaridis et al. (2014) and Tilmes et al. (2019) validated the model against flight campaign data over North America, the Pacific, and Atlantic. Dong et al. (2018) validated the performance of CAM-chem over Europe and Asia for O_3, $PM_{2.5}$, PM_{10}, and aerosol optical depth (AOD) and reported the model showed comparable performance with other popular global models such as EMEP and GEOS5. Gaubert et al. (2020) validated the model performance for CO over South Korea against the KORUS-AQ flight campaign data. Similar to other global models, CAM-chem has been widely applied in global-scale studies thus it is usually validated against flight campaign data or satellite products (Gliß et al., 2021; Kim et al., 2019) other than with regional scale measurements. Pfister et al. (2020) indicated a regional refine version of CAM-chem will be applied for regional scale air quality studies, and we are expecting to perform such type of simulation and probe into the model performance at regional scale over East Asia.

Reference:

Dong, X., Fu, J. S., Zhu, Q., Sun, J., Tan, J., Keating, T., Sekiya, T., Sudo, K., Emmons, L., Tilmes, S., Jonson, J. E., Schulz, M., Bian, H., Chin, M., Davila, Y., Henze, D., Takemura, T., Benedictow, A. M. K., and Huang, K.: Long-range transport impacts on surface aerosol concentrations and the contributions to haze events in China: an HTAP2 multi-model study, Atmos. Chem. Phys., 18, 15581–15600, https://doi.org/10.5194/acp-18-15581-2018, 2018.

Gliß, J., Mortier, A., Schulz, M., Andrews, E., Balkanski, Y., Bauer, S. E., Benedictow, A. M. K., Bian, H., Checa-Garcia, R., Chin, M., Ginoux, P., Griesfeller, J. J., Heckel, A., Kipling, Z., Kirkevåg, A., Kokkola, H., Laj, P., Le Sager, P., Lund, M. T., Lund Myhre, C., Matsui, H., Myhre, G., Neubauer, D., van Noije, T., North, P., Olivié, D. J. L., Rémy, S., Sogacheva, L., Takemura, T., Tsigaridis, K., and Tsyro, S. G.: AeroCom phase III multi-model evaluation of the aerosol life cycle and optical properties using ground- and space-based remote sensing as well as surface in situ observations, Atmos. Chem. Phys., 21, 87–128, https://doi.org/10.5194/acp-21-87-2021, 2021.

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Pfister, G. G., Eastham, S. D., Arellano, A. F., Aumont, B., Barsanti, K. C., Barth, M. C., Conley, A., Davis, N. A., Emmons, L. K., Fast, J. D., Fiore, A. M., Gaubert, B., Goldhaber, S., Granier, C., Grell, G. A., Guevara, M., Henze, D. K., Hodzic, A., Liu, X., Marsh, D. R., Orlando, J. J., Plane, J. M. C., Polvani, L. M., Rosenlof, K. H., Steiner, A. L., Jacob, D. J., and Brasseur, G. P.: The Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICA), Bulletin of the American Meteorological Society, 101, E1743-E1760, 10.1175/BAMS-D-19-0331.1, 2020.

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Tsigaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., Balkanski, Y., Bauer, S. E., Bellouin, N., Benedetti, A., Bergman, T., Berntsen, T. K., Beukes, J. P., Bian, H., Carslaw, K. S., Chin, M., Curci, G., Diehl, T., Easter, R. C., Ghan, S. J., Gong, S. L., Hodzic, A., Hoyle, C. R., Iversen, T., Jathar, S., Jimenez, J. L., Kaiser, J. W., Kirkevåg, A., Koch, D., Kokkola, H., Lee, Y. H., Lin, G., Liu, X., Luo, G., Ma, X., Mann, G. W., Mihalopoulos, N., Morcrette, J.-J., Müller, J.-F., Myhre, G., Myriokefalitakis, S., Ng, N. L., O'Donnell, D., Penner, J. E., Pozzoli, L., Pringle, K. J., Russell, L. M., Schulz, M., Sciare, J., Seland, Ø., Shindell, D. T., Sillman, S., Skeie, R. B., Spracklen, D., Stavrakou, T., Steenrod, S. D., Takemura, T., Tiitta, P., Tilmes, S., Tost, H., van Noije, T., van Zyl, P. G., von Salzen, K., Yu, F., Wang, Z., Wang, Z., Zaveri, R. A., Zhang, H., Zhang, K., Zhang, Q., and Zhang, X.: The AeroCom evaluation and intercomparison of organic aerosol in global models, Atmos. Chem. Phys., 14, 10845–10895, https://doi.org/10.5194/acp-14-10845-2014, 2014.

Technical corrections and responses:

Comment#3: Line 71:"over previous versions" sounds a bit odd.

response: We have revised the sentence at line 71 as shown below.

CESM2 (versions 2.0 and 2.1) includes 2 versions of model top, the Whole Atmosphere Community Climate Model version 6 (WACCM6) with 72 vertical layers up to about 150 km and the Community Atmosphere Model version 6 (CAM6) with 32 vertical layers up to about 40 km. CAM6 has simplified chemistry and simplified OA scheme, while CAM6 with comprehensive chemistry and comprehensive OA scheme are called CAM6-Chem which is updated compared to previous versions.

Comment#4: Line 138-141: This is a very long sentence, please split it up.

Response: We have split the sentence at line 139-141 as shown below.

To exclude the influence of potential extreme meteorology condition or emission inputs, these sensitivity runs are configured with FC2010climo component set and Newtonian relaxation time of three hours. The FC2010climo component set is as same as FCSD component set except that the emissions are a 10-year average used for each year of the simulation.

Comment#5: Line 169: "prominently overestimates in" is there a word missing here? What is overestimated?

Response: We apologize for the missing word in the sentence at line 169. We have corrected the sentence as shown below.

In EUS, the simulation prominently overestimates surface OA concentration in summer by 4.26 $\mu g/m^3$ (131.15 %) but successfully reproduces the temporal change with a strong correlation with observations of 0.60 (Table 3) as shown in Fig. 2(c).

Comment#6: Line 170: "with a strong correlation with observations of 0.60 as shown in Fig. 2c" The correlation coefficients are not shown in Fig 2 but rather in Table 3. Please refer to the table or both the figure and table. The same problem with referring to figure 2 instead of table 3 occur on line 174.

Response: We apologize for the problem at line 170 and line 174. Please see the response of comment#5 for the modification of the sentence at line 170. The modification of the sentence at line 174 is shown below.

The model shows smaller bias in WUS but also a poor correlation of 0.36 (Table 3) in summer as shown in Fig. 2(e) and 2(f).

Comment#7: Line 170: "As compared with CONUS domain, simulation at" are there missing words in this part of the sentence?

Response: We apologize for the missing words in the sentence at line 170. The "simulation" is referred to the "surface OA concentration from the simulation". We have modified the sentence as follows.

As compared with CONUS domain, surface OA concentration from the simulation at EUS shows an even greater overestimation during warmer months as shown in Fig. 2(d).

Comment#8: Line 295-299. This is a very long sentence that should probably be split up. Also, the English in this sentence needs to be checked.

Response: We totally agree that the long sentence is poorly readable. We have split and modified the long sentence in line 295-299 as follows.

It is certainly reasonable to take the wall-loss effect into account when making the chamber measurements. But it also should be noticed that those measurements were conducted under artificial environment with predefined chemical species that may vary significantly from the real meteorology condition and atmospheric chemistry regime. Thus, the parameters reported in the chamber studies need to be carefully interpreted and adjusted when applied in atmospheric models.