Author's response

Manuscript: "Enhanced summertime ozone and SOA from biogenic volatile organic compound (BVOC) emissions due to vegetation biomass variability during 1981–2018 in China" (acp-2021-675)

Responses to reviewers' comments are listed below, respectively. Revised portions are marked in red.

Response to Referee #1

1.The validation for WRF/Chem simulation hasn't been conducted. Even the WRF/Chem model is a very powerful and advanced model for simulating regional atmospheric environment, simulation errors are always present. The MS should show how much error in the base to help readers to get how real the model results are. Evaluations for ozone and $PM_{2.5}$ in the base run are necessary, in my opinion.

Response: Thank you so much for your suggestion. In the revised manuscript, we add the validation of air quality simulations by WRF-Chem.

In this study, we focused on the contribution of historical BVOC emissions caused by vegetation biomass variability, so the anthropogenic emissions in all scenarios were fixed using the MIX Asian anthropogenic source emission inventory. In the base run, both anthropogenic and BVOC emissions were inputted to conduct O_3 and SOA simulation in June 2018. The observations of daily maximum 8-h (MDA8) O_3 and daily average $PM_{2.5}$ at 1588 sites in China were applied to evaluate the WRF-Chem simulation in the base run.

Line 143–153 of revised manuscript, "The observed daily maximum 8-h (MDA8) O_3 and daily average $PM_{2.5}$ concentrations at 1588 sites in June 2018 in China were applied to evaluate the WRF-Chem simulations in the control run (as listed in Table 1) in this study. The observations were from the daily updated national air quality released by the China National Environmental Monitoring Centre (http://www.cnemc.cn/). The verification statistics are shown in Table S1. Notably, $PM_{2.5}$ had no systematic bias between the observation and simulation, while the model predicted O_3 concentrations were lower than measurements. The errors could be mainly attributed to the anthropogenic emissions data used in this study as described in Section 2.3." is added.

Table S1. Verification statistics of meteorology and air quality simulations.

Variable	Year	Mean		MB	MAE	RMSE
		Observation	Simulation			
T2 (K)	2008	295.84	295.48	0.36	2.47	3.30
	2018	295.83	294.60	1.24	2.46	3.30
MDA8 O_3 (ppb)	2018	58.83	36.39	22.44	36.65	45.42
$PM_{2.5}$ (μg m^{-3})	2018	29.29	29.81	-2.71	21.31	30.51

T2: temperature at 2 m; MB: mean bias; MAE: mean absolute error; RMSE: root mean square error.

2. The MS considers REA results to validate the MEGAN performance, which maybe the most direct verification for BVOCs emissions. It is good. But can the MS supply the information of sampling location? And the conclusion that "the estimation is higher than measurement with an average mean bias of 1.11 mg m^{-2} h^{-1}" should be noted that it estimated higher with a mean bias of 1.1.1 mg/m^2/hr in these REA sampling regions, but not for the whole China in averaged. And it should be better to show the comparison between HCHO and MEGAN results first, followed by the comparison between REA and MEGAN results.

Response: Thank you very much for your valuable suggestion. In revised manuscript, we have added specific information on sampling sites and provided a clearer explanation about the use of flux measurement to evaluate BVOCs simulations limited to these sampling regions. Additionally, we adjusted the order of the flux measurements evaluation and HCHO concentration evaluation.

Line 186–203, "The emission simulations were validated by using the measurements of BVOC emission flux and formaldehyde (HCHO) concentration. The flux measurements of

BVOCs conducted in China were collected (Bai et al., 2015, 2016, 2017). The gridded BVOC emission estimated by MEGAN were extracted where the flux measurement sites were located to do the comparison (Fig. S1). The modeled fluxes of BVOCs in this study capture the spatial variability of observations better with a correlation coefficient of 0.84. But the estimation is higher than measurement with an average mean bias of 1.11 mg m^{-2} h^{-1}, mainly because of the differences in time between them. Isoprene is the main compound in BVOC species, accounting for nearly half of total BVOC emissions in China. It undergoes chemical and photochemical reactions in the atmosphere, and the oxidation product is mainly HCHO (Bai and Hao, 2018; Orlando et al., 2000). In forest areas and in summer, biogenic isoprene is the dominant source of HCHO, so satellite HCHO column concentration is widely used to constrain isoprene emissions (Opacka et al., 2021; Palmer et al., 2003; Stavrakou et al., 2018; Wang et al., 2021; Zhang et al., 2021). In this study, we used the HCHO vertical column detected by Ozone Monitoring Instrument (OMI) to validate the spatial variability of isoprene estimates. The monthly OMI HCHO data from the EU FP7 project QA4ECV product (Quality Assurance for Essential Climate Variables; http://www.qa4ecv.eu) was used in this study. The result of the statistical analysis with a confidence interval of 99% indicates that the monthly averaged OMI HCHO vertical column in June 2018 is significantly correlated to the model-estimated isoprene emissions." is revised to "The emission simulations were validated by using the formaldehyde (HCHO) concentration and measurements of BVOC emission flux. Isoprene is the main compound in BVOC species, accounting for nearly half of total BVOC emissions in China. It undergoes chemical and photochemical reactions in the atmosphere, and the oxidation product is mainly HCHO (Bai and Hao, 2018; Orlando et al., 2000). In forest areas and in summer, biogenic isoprene is the dominant source of HCHO, so satellite HCHO column concentration is widely used to constrain isoprene emissions (Opacka et al., 2021; Palmer et al., 2003; Stavrakou et al., 2018; Wang et al., 2021; Zhang et al., 2021). In this study, we used the HCHO vertical column detected by Ozone Monitoring Instrument (OMI) to validate the spatial variability of isoprene estimates. The monthly OMI HCHO data from the EU FP7 project QA4ECV product (Quality Assurance for Essential Climate Variables; http://www.qa4ecv.eu) was used in this study. The result of the statistical analysis with a confidence interval of 99% indicates that the monthly averaged OMI HCHO vertical column in June 2018 is significantly correlated to the model-estimated isoprene emissions. The flux measurements of BVOCs conducted in China were collected (Bai et al., 2015, 2016, 2017). The details of these measurements are provided in Table S3, including the location of sampling sites and observed BVOC emission fluxes. The gridded BVOC emission estimated by MEGAN were extracted where the flux measurement sites were located to do the comparison (Fig. S1). The modeled fluxes of BVOCs in this study capture the spatial variability of observations better with a correlation coefficient of 0.84. But the estimation is higher than measurement with an average mean bias of 1.11 mg m^{-2} h^{-1} in these sampling sites, mainly because of the differences in time between them.".

Table S3. Detailed descriptions of the flux measurements used in this study.

Site	Location	Isoprene		Monoterpene		Sesquiterpene		References
		Obs	Sim	Obs	Sim	Obs	Sim	
Qianyanzhou	26°44′48″N, 115°04′13″E	0.07	3.25	0.81	0.17	0.01	0.06	Bai et al. (2017)
Taihuyuan	30°18′N, 119°34′E	3.35	7.06	0.01	0.14	0.00	0.02	Bai et al. (2016)
Changbai Mountain	42°240′N, 128°60′E	0.95	4.03	0.14	0.72	0.19	0.05	Bai et al. (2015)

Obs: observation; Sim: simulation; the units of flux measurements are mg m^{-2} h^{-1}.

References:

Bai, J. H. and Hao, N.: The relationships between biogenic volatile organic compound (BVOC) emissions and atmospheric formaldehyde in a subtropical Pinus plantation in China, Ecology and Environmental Sciences, 27(6): 991–999, https://doi.org/10.16258/j.cnki.1674-5906.2018.06.001, 2018.

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Bai, J., Guenther, A., Turnipseed, A., Duhl, T., Yu, S., and Wang, B.: Seasonal variations in whole-ecosystem BVOC emissions from a subtropical bamboo plantation in China, Atmos. Environ., 124, 12–21, https://doi.org/10.1016/j.atmosenv.2015.11.008, 2016.

Bai, J., Guenther, A., Turnipseed, A., Duhl, T., and Greenberg, J.: Seasonal and interannual variations in whole-ecosystem BVOC emissions from a subtropical plantation in China, Atmos. Environ., 161, 176–190, https://doi.org/ 10.1016/j.atmosenv.2017.05.002, 2017.

Opacka, B., Müller, J.-F., Stavrakou, T., Bauwens, M., Sindelarova, K., Markova, J., and Guenther, A. B.: Global and regional impacts of land cover changes on isoprene emissions derived from spaceborne data and the MEGAN model, Atmos. Chem. Phys., 21, 8413–8436, https://doi.org/10.5194/acp-21-8413-2021, 2021.

Orlando, J. J., Nozière, B., Tyndall, G. S., Orzechowska, G. E., Grazyna, E., Paulson, S. E., and Rudich Y.: Product studies of the OH- and ozone-initiated oxidation of some monoterpenes, J. Geophys. Res., 105, 11561–11572, https://doi.org/10.1029/2000JD900005, 2000.

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Wang, H., Wu, Q. Z., Guenther, A. B., Yang, X. C., Wang, L. N., Xiao, T., Li, J., Feng, J. M., Xu, Q., and Cheng, H.: A long-term estimation of biogenic volatile organic compound (BVOC) emission in China from 2001–2016: the roles of land cover change and climate variability, Atmos. Chem. Phys., 21, 4825–4848, https://doi.org/10.5194/acp-21-4825-2021, 2021.

Zhang, M., Zhao, C., Yang, Y., Du, Q., Shen, Y., Lin, S., Gu, D., Su, W., and Liu, C.: Modeling sensitivities of BVOCs to different versions of MEGAN emission schemes in WRF-Chem (v3.6) and its impacts over eastern China, Geosci. Model Dev., 14, 6155–6175,

https://doi.org/10.5194/gmd-14-6155-2021, 2021.

3. The ozone and SOA from BVOCs due to vegetation biomass variability increase during 1981-2018. I can't understand why they will not show the increasing trend. Because almost all the model inputs are the same, but the vegetation biomass increase. The scenario settings are not so reasonable, and that weaken the significance of the results to policy management. Two real historical simulations should be better.

Response: Thank you for your suggestion. The vegetation change is the main driver of interannual variations of BVOC emissions (Li et al., 2020; Wang et al., 2021). The large-scale afforestation activities in recent years lead to the rapid increase of vegetation leaf biomass and therein BVOC emissions. In our study, we aim to explore the impacts of interannual BVOC emission variations on O_3 and SOA formation caused by vegetation biomass variability during 1981–2018. But we are sorry for the unclear statement of the increasing trend of O_3 and SOA from BVOCs impacted by vegetation biomass increase in the study.

During 1981–2018, due to the changed BVOC emissions caused by vegetation leaf biomass variability, both O_3 and SOA formations showed significant increasing trends ($p<0.05$ at the 95% confidence interval) by t-test. In the revised manuscript, line 344, "At the 95% confidence level, the increasing trend for O_3 is significant." is added. Line 389–290, "The national SOA enhanced at an annual rate of 0.01 μg m^{-3}." is revised to "The national SOA enhanced at an annual rate of 0.01 μg m^{-3}, showing a significantly increasing trend ($p<0.05$).". Line 438–439, "The interannual variation of BVOC emissions caused by increasing leaf biomass results in O_3 and SOA concentrations increasing at average rates of 0.11 ppb yr^{-1} and 0.008 μg m^{-3} yr^{-1}, respectively." is revised to "The interannual variation in BVOC emissions caused by increasing in leaf biomass results in significant increases ($p<0.05$) of O_3 and SOA concentrations at average rates of 0.11 ppb yr^{-1} and 0.008 μg m^{-3} yr^{-1}, respectively.".

Because our study mainly explores the sensitivity of O_3 and SOA generation by BVOC emissions to changes in vegetation leaf biomass, all the model inputs are the same except the increased vegetation biomass. But our results can conclude that the BVOC emissions caused by vegetation leaf biomass variability is an important factor affecting the generation of O_3 and

SOA. This study clarifies the impact of changes in vegetation leaf biomass and is of great significance for future researches on vegetation management and precise prevention and control of air pollution in China in the context of fighting climate change. But, as you addressed, it is critical to do real historical simulations using annual meteorology and anthropogenic emissions to understand the multivariate effects on O_3 and SOA formation. This is explained in line 454–455, "Future work can update the anthropogenic emission inventory and use dynamic meteorological data to explore multivariate effects and provide more accurate data for evaluating the roles of biogenic emissions in air quality.".

References:

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Wang, H., Wu, Q. Z., Guenther, A. B., Yang, X. C., Wang, L. N., Xiao, T., Li, J., Feng, J. M., Xu, Q., and Cheng, H.: A long-term estimation of biogenic volatile organic compound (BVOC) emission in China from 2001–2016: the roles of land cover change and climate variability, Atmos. Chem. Phys., 21, 4825–4848, https://doi.org/10.5194/acp-21-4825-2021, 2021.

4. For the part of meteorological evaluation: using a table should be more clear.

Response: Thank you for your valuable suggestion. We add a table as Table S1 in the revised manuscript to show the statistical parameters of WRF simulation for metrological variables.

Correspondingly, line 93–95, "For 2008, the average mean bias (MB), mean absolute error (MAE), and root-mean-square error (RMSE) are 0.36, 2.47, and 3.30 K over China. For 2018, these statistics are 1.24, 2.46 and 3.30 K, respectively. The correlation coefficients between simulations and observations are 0.82 and 0.86 for the year 2008 and 2018, respectively. In general, the WRF simulation is considered reasonable for driving MEGAN." is revised to "We conducted the statistical verification of meteorological variables, as shown in Table S1, including the average mean bias (MB), mean absolute error (MAE), and root-mean-square error (RMSE). The results show that the WRF simulation is considered reasonable for driving MEGAN.".

Table S1. Statistics of meteorological and air quality variables.

Variable	Year	Mean		MB	MAE	RMSE
		Observation	Simulation			
T2 (K)	2008	295.84	295.48	0.36	2.47	3.30
	2018	295.83	294.60	1.24	2.46	3.30
MDA8 O_3 (ppb)	2018	58.83	36.39	22.44	36.65	45.42
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T2: temperature at 2 m; MB: mean bias; MAE: mean absolute error; RMSE: root mean square error.

5. Fig. 2: using blue/red color table should be better.

Response: Thank you for your suggestion. In the revised manuscript, Fig. 2 is revised.

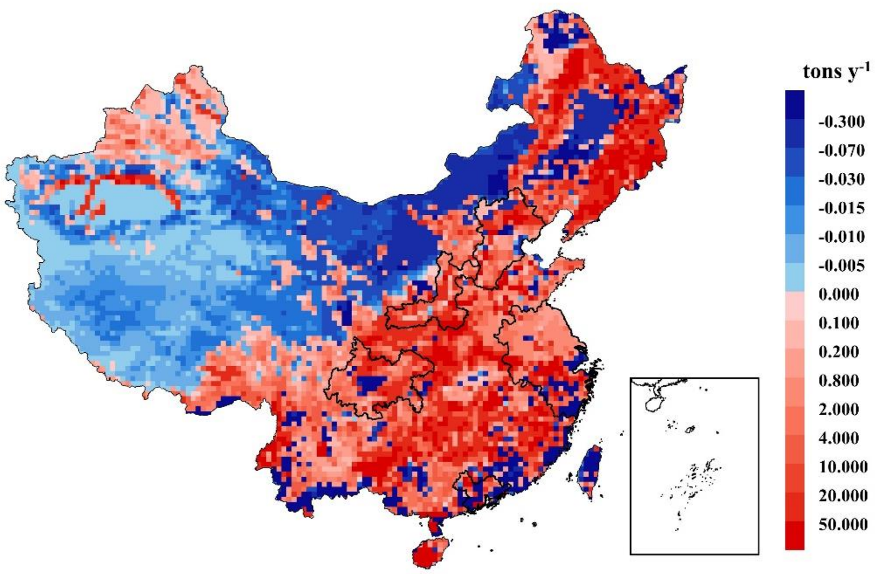

Fig. 2. Spatial distribution of interannual variations in BVOC emissions caused by leaf biomass changes.