Dear editor,

Here we submit our revised manuscript for consideration to be published on Atmospheric

Chemistry and Physics

The further information about our manuscript is as follows:

Topic: Developing a novel hybrid model for the estimation of surface 8-h ozone (O₃) across the remote Tibetan Plateau during 2005-2018

Type of Manuscript: article

Authors: Rui Li^a, Yilong Zhao^a, Ya Meng^a, Wenhui Zhou^a, Ziyu Zhang^a, Hongbo Fu^{a-c*}

*Corresponding author:

Hongbo Fu; Address: Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China; Tel.: (+86)21-5566-5189; Fax: (+86)21-6564-2080; Email: <u>fuhb@fudan.edu.cn</u>

Firstly, we acknowledge the suggestions of editor and two reviewers, and are also grateful to your efficient serving. We have updated the manuscript on the basis of these valuable comments. Our responses were listed as following:

Reviewer #1: The subject is appropriate to Atmospheric Chemistry and Physics. This paper developed a novel hybrid model named random forest-generalized additive model (RF-GAM) to estimate the surface 8-h O_3 levels across the remote Tibetan Plateau during 2005-2018. This model displayed excellent prediction performances of O_3 spatiotemporal variations when compared with other seven machine learning models, and can be applied in the remote regions with sparse monitoring sites. This study is a valuable work. Therefore, I recommend clearly the acceptance for publication of this manuscript after revisions. Several editorial comments for improving the information content and presentation of the paper are listed as follows:

Response: Thank for reviewer's suggestions. I have revised the manuscript carefully based on reviewer's suggestions.

Comment 1: When you analyzed spatiotemporal variations of simulated 8-h O₃ concentrations and nonattainment days at the monitoring sites, why do you not use observed values for comparison? It may verify the results and rich your discussions.

Response: I agree with reviewer's suggestion. The measured mean 8-h O₃ concentrations were

added in Table 4. The relationships between measured O_3 concentrations and simulated O_3 levels have been added in the discussion part.

Comment 2: Since Community Multiscale Air Quality (CMAQ) model and other three-dimensional models have been widely applied to the simulation of surface O₃ concentrations, you'd better add a paragraph to summarize these results and compare your results with others to reveal the superiority of your novel model by adding more references such as Eder, B., and S. Yu, 2006. A performance evaluation of the 2004 release of Models-3 CMAQ. Atmospheric Environment, 40: 4811-4824. **Response:** Thank for reviewer's suggestion. We have added a paragraph to discuss the advantage of RF-GAM model developed in our study compared with previous studies. The detailed explanations are as follows:

To date, some previous studies also simulated the surface O₃ concentration in Tibetan Plateau using statistical models (Zhan et al., 2018). For instance, Zhan et al. (2018) employed the RF-STK model to estimate the surface O₃ concentration over China, and explained the 66% spatial variability of O₃ level in Tibetan Plateau. Apart from these statistical models, some classical CTMs were also applied to estimate the O₃ concentration in the remote area. Both of Liu et al. (2018) and Lin et al. (2018) used CMAQ to estimate the O₃ level across China, while the R² values in most of cities were lower than 0.50. In terms of the predictive performance, the RF-GAM model in our study showed the significant advantages compared with previous studies. It should be noted that our RF-GAM model could outperform most of current models, chiefly because of (1) accounting for the temporal autocorrelation of surface O₃ concentration; and (2) the use of high-quality satellite data.

Comment 3. As for the temporal variations of the simulated 8-h O_3 concentrations and nonattainment days as depicted in Fig. 7 and Fig. 10, the change pattern from 2005 to 2018 is not very obvious. Would you mind showing interannual variations in a clearer way?

Response: Thank for reviewer's suggestions. The interannual variation has been added in the Tab. S1, which was clearly shown in the form of table.

Comment 4. L225-227 Regarding the other five models (i.e., RF, generalized regression neutral

network (GRNN), backward propagation neural network (BPNN), Elman neural network (ElmanNN), and extreme learning machine (ELM)), you should put all related results from them into SI because you already prove that RF-GAM model is the best and it is unnecessary to present the results of other worse ones in the main text part.

Response: I agree with reviewer's suggestion. We have placed some other models in SI.

Comment 5. There are many English grammar errors in the manuscript. I only list some of

them here for your reference below (please make all necessary corrections before publication).

Response: Thank for reviewer's suggestions. We have revised the grammar errors throughout the manuscript based on the reviewer's suggestions.

Comment 6. L16: It should be "to predict the surface 8-h O₃ concentrations: : ..."

Response: I agree with reviewer's suggestion. "concentration" has been replaced by "concentrations".

Comment 7. L21: It should be "to predict the surface 8-h O₃ concentrations: : :..".

Response: We have corrected these errors based on reviewer's suggestions.

Comment 8. L25: It should be "the estimated O₃ mean concentrations: : :: : :"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 9. L44: It should be "The chemical reactions between NO_x and VOCs in the presence of sunlight were: : :"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 10. L85: It should be "the daily satellite data enable the fine-scale estimations of O₃ level: : :"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 11. L90-91: It should be "the global distributions of O₃ concentrations based on simple linear : : :."

Response: We have corrected these errors based on reviewer's suggestions.

Comment 12. L103-104: It should be "can obtain the contributions of each predictor to air pollutants: : :."

Response: We have corrected these errors based on reviewer's suggestions.

Comment 13. L146-147: It should be "simulate the gridded 8-h O₃ concentrations over Tibetan Plateau for the first time: : :.".

Response: We have corrected these errors based on reviewer's suggestions.

Comment 14. L162: It should be "lack of anthropogenic activities and most of the residents focus on southeast and south parts of: : :"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 15. L171: It should be "The data quality of all the monitoring sites was assured on.."

Response: We have corrected these errors based on reviewer's suggestions.

Comment 16. L175: It should be "The O_3 column amounts (DU) during 2005-2018 were downloaded from the Ozone Monitoring: : :".

Response: We have corrected these errors based on reviewer's suggestions.

Comment 17. L294-295: It should be "we also investigated the spatial variabilities of the predictive accuracy for RF-GAM model: : :...".

Response: We have corrected these errors based on reviewer's suggestions.

Comment 18. L337-337 about the effects of RH on the O₃ formation: Please add the following reference to have more discussions in terms of chemical mechanism: S. Yu, 2019. Fog geoengineering to abate local ozone pollution at ground-level by enhancing air moisture. Environ Chem Lett, 17(1), 565–580, doi: 10.1007/s10311-018-0809-5.

Response: We have corrected these errors based on reviewer's suggestions. Besides, we also citied the reference in the revised version.

Comment 19. L353-354: It should be "It was well known that photochemical reactions of BVOCs and NOx in the presence of sunlight were beneficial to the O3 formation..". Also please add more references about this such as: S. Yu, et al., 2006. Performance and diagnostic evaluations of a real-time ozone forecast by the Eta-CMAQ model suite during the 2002 New England Air Quality Study (NEAQS). Journal of the Air & Waste Management Association, 56:1459-1471.

Response: We have corrected these errors based on reviewer's suggestions. Besides, we also citied the reference in the revised version.

Comment 20. L397-398: It should be "Besides, the 8-h O₃ concentrations in Tibetan Plateau displayed significantly seasonal discrepancy: : ...".

Response: We have corrected these errors based on reviewer's suggestions.

Comment 21. P44: It should be "Table" in stead of "Tab.".

Response: We have corrected these errors based on reviewer's suggestions.

Comment 22. P2, L34: "shared with" => "shared"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 23. P3, L62: "surface" => "surface"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 24. P3, L63: It should be ": : :concentration displayed the gradual decrease in the recent ten years".

Response: We have corrected these errors based on reviewer's suggestions.

Comment 25. P4, L69: "lacks of" => "lacks"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 26. P8, L163: "is consisted of" => "consists of"

Response: We have corrected these errors based on reviewer's suggestions.

Comment 27. P20, L422, 434: "Tab. 2" should be "Table 4"

Response: We have corrected these errors based on reviewer's suggestions.