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 (O_3) across the remote Tibetan Plateau during 2005-2018

Type of Manuscript: article

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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 O_3 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 O_3 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 O_3 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 O_3 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 O_3 concentration over China, and explained the 66% spatial variability of O_3 level in Tibetan Plateau. Apart from these statistical models, some classical CTMs were also applied to estimate the O_3 concentration in the remote area. Both of Liu et al. (2018) and Lin et al. (2018) used CMAQ to estimate the O_3 level across China, while the R^2 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 O_3 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 O_3 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 O_3 concentrations: : :.".

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

Comment 8. L25: It should be "the estimated O_3 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 O_3 level: : :"

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

Comment 11. L90-91: It should be "the global distributions of O_3 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 O_3 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 O_3 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 O_3 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: “surfacce” => “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.

Reviewer #2: This paper attempts to create a machine-learned statistical model to estimate 8-h surface ozone concentrations where few measurements exist (Tibetan plateau). The model formulation (RF-GAM) is interesting and the subject fits within the scope of ACP. There are several flaws with the presentation of this work, namely the lack of significance tests and alternate performance metrics in addition to English grammatical errors throughout. However, I am optimistic the authors can correct these issues and I believe this work will be a very important addition to the scientific literature of air pollution in rural regions.

Response: Thank for reviewer’s suggestions. I have revised the manuscript based on the reviewer’s suggestions carefully. The detailed responses are as follows:

Comment 1: Does the paper address relevant scientific questions within the scope of ACP? This paper attempts to create a machine-learned statistical model to estimate 8-h surface ozone concentrations for the Tibetan plateau. The RF-GAM model is interesting and the subject fits within the scope of ACP.

Response: Thank for reviewer’s suggestions. The journal (ACP) focuses on the Earth's atmosphere and the underlying chemical and physical processes. The modelling of atmospheric components in the atmosphere is a hot topic of ACP, and many relevant references have been published in this journal. Therefore, we believe that the paper addresses relevant scientific questions within the scope

of ACP.

Comment 2: Does the paper present novel concepts, ideas, tools, or data? The machine learning model formulation is interesting and the application of the GAM to remove autocorrelation in the time series data is novel.

Response: Thank for reviewer's suggestions. The paper presents a novel machine learning model named RF-GAM, which shows excellent performance in predicting the 8-h O_3 concentration over Tibetan Plateau. The development of this model is the major novelty of this present study. Besides, the present study fills the gap of statistical estimation 8-h O_3 level in a remote region, and provides useful datasets for epidemiological studies and air quality management. The implication is very important. Moreover, we also

Comment 3: Are substantial conclusions reached? Not particularly, the performance metrics presented in the work are not convincing. The authors present RMSE as an absolute measure of performance yet the best performing model had an RMSE of 14.41 μg/m^3, which is greater than 10% error relative to the WHO 8-h ozone critical value (100 μg/m^3) they are using to determine nonattainment. There is very little discussion about significant differences between performance due to model architecture and between seasons and years. Greater discussion of model uncertainty needs to be had. This study addresses an important topic that lacks much attention in scientific literature, but the performance of the model is still considerably lacking.

Response: Thank for reviewer's suggestions. The differences of performance between years has been added in the Table 1. The predictive performances in different seasons were also shown in Table 2. The contents have been expanded to discuss the temporal variation of predictive accuracy of O_3 concentration. The uncertainty of the present study is also added in the revised version: "The determination of nonattainment days showed some uncertainties owing to the predictive error of modelled O_3 concentration. First of all, meteorological data used in RF-GAM model were collected from reanalysis data and these gridded data often showed some uncertainties, which could increase the uncertainty of O_3 estimation. Second, the O_3 column amount used in the present study reflected vertical O_3 column amount rather than surface O_3 concentration. Thus, it could decrease the predictive performance of surface O_3 level". Besides, some important references were also added in the revised version.

Comment 4: Are the scientific methods and assumptions valid and clearly outlined? Scientific

methods and data preprocessing are outlined well. Figure 2 presents a workflow method for the machine learning model, though including an actual architectural schematic of the final model would be better (e.g. what does the random forest look like in terms of the number of trees, nodes, etc.). Furthermore, none of the other machine learning models were given an architectural model description, i.e., how many nodes in the neural network, activation functions, etc. Machine learning is a burgeoning field with many nascent applications. Therefore, this paper would benefit going into greater detail about what other machine learning methods they tried in detail so that others could learn from their attempts.

Response: Thank for reviewer's suggestions. We have added some detailed information especially key parameters about various machine learning models in the revised version. Based on the iteration result, the optimal n_{tree} and m_{try} reached 500 and 5, respectively. For XGBoost method, the optimal tree depth and minimum child weight was set as 8 and 6, respectively. The activation functions of GRNN, BPNN, and ElmanNN were sigmoid function. The number of nodes in BPNN and ElmanNN were 5 and 4, respectively. The number of nodes in GRNN was equal to the sample size.

Comment 5: Are the results sufficient to support the interpretations and conclusions? (cite sections) There must be a greater statistical argument presented to support the interpretations of the authors. Oftentimes the authors present only an R^2 metric or an RMSE value to evaluate the goodness of fit among conditions. These performance measures are relatively obfuscating as even though ozone might increase or decrease between temporal/spatial extents, these changes, I suspect, are statistically insignificant. There are practically no significance tests conducted. These performance metrics must be put into greater context. Perhaps including confidence intervals (rather than only standard deviations) would also help. For the variable importance section 3.2, the authors should present an error covariance matrix for the inputs. For random forest algorithms, the variable importance determined by the model is meaningless if the variables are co-linear. Furthermore, oftentimes the explanations for the concentration of ozone are too speculative (e.g. lines 334-351). There are numerous mentions of NO_x and VOCs contributing to ozone loss or formation, yet this analysis cannot address such concerns directly as the model does not take into account these variables. These explanations are well thought out but exceed the scope of what this model can answer. Perhaps place them in the discussion rather than with the results. In addition to RMSE, an absolute metric, perhaps the authors should consider reporting a relative metric such as percent error

in tandem. R^2 values of 0.60 and RMSE values of 14 μg/m^3 are not readily obvious/interpretable if this is adequate performance. Also, conducting significance tests between different models might be beneficial. I am not convinced that RF and XGBoost are markedly different. The addition of the GAM seems like a different model architecture than typical machine learning models. That is, why not attach the GAM to the XGBoost and see how that performs? It is fine if the authors wanted to create an RF-GAM model from the outset, but the comparison between the other models does not seem very robust.

Response: Thank for reviewer's suggestions. We employed one-way ANOVA method to compare the predictive values of eight models. The results are as follows:

Source	SS	df	MS	F	p value
Groups	24542.6	7	3506.09	9.01	<0.01
Error	59209467.7	152154	389.14		
Total	59234010.3	152161			

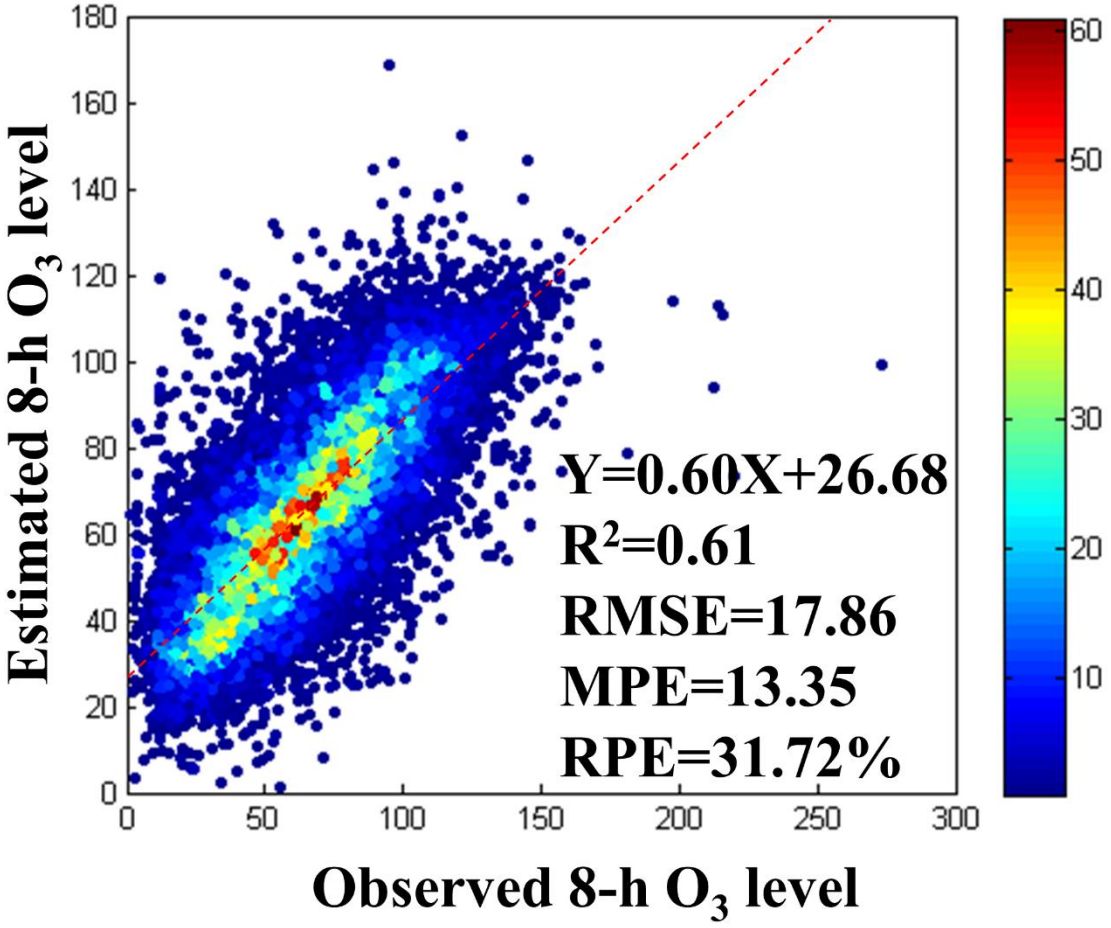
Based on the result of one-way ANOVA, we found that the RF-GAM model showed significantly better predictive performance compared with other seven models.

Indeed, the traditional statistical model required that all of the independent variables should be deviated from the autocorrelation. The multicollinearity of variables might decrease the reliability of these models. However, the random forest method did not consider the autocorrelation of all the variables because the algorithm was nonlinear.

Indeed, some explanations for the concentration of ozone were speculative, and thus we revised many redundant or speculative explanations especially for the contribution of NO_x and VOCs to ozone formation. However, some explanations were well-founded because it could be supported by the Fig. S4. Besides, the effects of meteorological factors on ozone formation were confirmed by some previous studies and Fig. S5-S11, which was reliable.

In addition to RMSE, we also added a new indicator named relative prediction error (RPE) to assess the predictive accuracy (Fig. 3 and Fig. 4). We have employed the XGBoost method to combine the GAM to estimate the surface O_3 concentration (see the following figure), and found that the hybrid model showed the worse performance compared with RF-GAM model. The R^2 value of XGBoost-GAM model was significantly lower than that of RF-GAM model. Besides, both of

RMSE and MPE for XGBoost-GAM model were also significantly higher than those of RF-GAM. Thus, we believed that RF-GAM was more appropriate to estimate the O_3 concentration in Tibetan Plateau.



Comment 6: Is the overall presentation well structured and clear? The overall structure and flow of the paper are coherent, though oftentimes the English is not clear.

Response: Thank for reviewer's suggestion. We have significantly revised the language throughout the paper.

Comment 7: Is the language fluent and precise? This study needs considerable corrections to numerous grammatical and language errors. There are far too often awkward phrasings and mixed tenses.

Response: Thank for reviewer's suggestion. We have significantly revised the language throughout the paper and corrected many grammar errors.

Comment 8: Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? Many of the figures are too small to view. Needs to be increased in size.

Response: I agree with reviewer's suggestions. We have redrawn the Fig. 7 and Fig. 10 to increase the figure size.

Comment 9: Are the number and quality of references appropriate? Great overview and command of the literature.

Response: Thank for reviewer's suggestions. We have added some latest references in the revised version though many relevant references have been added.

Following are some line comments, though not all-inclusive:

Comment 10: a few times you use the word 'beneficial' to describe conditions that create pollution, perhaps consider different word choices as this is a bit awkward.

Response: I agree with reviewer's suggestions. "beneficial" was changed into "caused", "triggered", or "promoted".

Comment 11: Also found on line 127. 102-103: "decision tree models such as random forest (RF) and extreme gradient boosting (XGBoost) strike a perfect balance between prediction performance and computing cost". This seems to be a wide sweeping claim without a reference. Decision trees may be faster to train than neural networks, but they are also slower to evaluate. That is, decision trees are slow to train and slow to test, whereas neural networks are very slow to train and very fast to test. Consider revising your claim.

Response: I agree with reviewer's suggestions. (Line 102-103) The sentence has been changed into "Among these machine learning algorithms, decision tree models such as random forest (RF) and extreme gradient boosting (XGBoost) generally showed fast training speed and excellent prediction accuracy".

Comment 12: 135-138: awkward, confusing phrasing

Response: Thank for reviewer's suggestions. (Line 136-139) The sentence has been changed into "Unfortunately, these scarce monitoring sites in Tibetan Plateau cannot capture real O_3 pollution status especially in the remote areas (e.g., Northern part of Tibetan Plateau) because each site only possessed limited spatial representativeness".

Comment 13: Line 190: Still not sure why you included GDP as an input variable. You write that you integrated it because "these data were available each five years". Please provide a scientific explanation for the inclusion of this variable.

Response: Thank for reviewer's suggestion. GDP was included in the original model because many

previous studies confirmed that GDP might be linked with the O_3 pollution (Li et al., 2020 JCP). It was well known that the hotspot of O_3 pollution focused on the area with the higher VOCs and NO_x (e.g., industrial points and residential areas). These regions often displayed the relatively higher GDP compared with other regions.

Developing a novel hybrid model for the estimation of surface 8-h ozone (O_3) across the remote Tibetan Plateau during 2005-2018

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Abstract

We developed a two-stage model named random forest-generalized additive model (RF-GAM) based on satellite data, meteorological factors, and other geographical covariates to predict the surface 8-h O_3 concentrations across the remote Tibetan Plateau. The 10-fold cross-validation result suggested that RF-GAM showed the excellent performance with the highest R^2 value (0.76) and lowest root mean square error (RMSE) (14.41 μg/m^3) compared with other seven machine learning models. The predictive performance of RF-GAM model showed significantly seasonal discrepency with the highest R^2 value observed in summer (0.74), followed by winter (0.69) and autumn (0.67), and the lowest one in spring (0.64). Additionally, the unlearning ground-observed O_3 data collected from open websites were applied to test the transferring ability of the novel model, and confirmed

that the model was robust to predict the surface 8-h O_3 concentration during other periods ($R^2 = 0.67$, RMSE = 25.68 μg/m^3). RF-GAM was then used to predict the daily 8-h O_3 level over Tibetan Plateau during 2005-2018 for the first time. It was found that the estimated O_3 concentrations displayed a slow increase from 64.74 ± 8.30 μg/m^3 to 66.45 ± 8.67 μg/m^3 from 2005 to 2015, whereas it decreased from the peak to 65.87 ± 8.52 μg/m^3 during 2015-2018. Besides, the estimated 8-h O_3 concentrations exhibited notably spatial variation with the highest values in some cities of North Tibetan Plateau such as Huangnan (73.48 ± 4.53 μg/m^3) and Hainan (72.24 ± 5.34 μg/m^3), followed by the cities in the central region including Lhasa (65.99 ± 7.24 μg/m^3) and Shigatse (65.15 ± 6.14 μg/m^3), and the lowest one in a city of Southeast Tibetan Plateau named Aba (55.17 ± 12.77 μg/m^3). Based on the 8-h O_3 critical value (100 μg/m^3) scheduled by World Health Organization (WHO), we further estimated the annually mean nonattainment days over Tibetan Plateau. It should be noted that most of the cities in Tibetan Plateau shared ~~with~~ the excellent air quality, while several cities (e.g., Huangnan, Haidong, and Guoluo) still suffered from more than 40 nonattainment days each year, which should be paid more attention to alleviate local O_3 pollution. The result shown herein confirms the novel hybrid model improves the prediction accuracy and can be applied to assess the potential health risk, particularly in the remote regions with sparse monitoring sites.

Keywords: Surface O_3 level; satellite data; random forest; generalized additive model; Tibetan Plateau

1. Introduction

Along with the rapid economic development and urbanization, the anthropogenic emissions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) displayed high-speed growth. The chemical reactions between NO_x and VOCs in the presence of sunlight ~~was beneficial to~~triggered

45 the ambient ozone (O_3) formation (Wang et al., 2019; Wang et al., 2017). As a strong oxidant,
46 ambient O_3 could play a negative role on human health through aggravating the cardiovascular and
47 respiratory function (Ghude et al., 2016; Marco, 2017; Yin et al., 2017a). Apart from the effect on
48 human health, O_3 also posed a great threaten on vegetation growth (Emberson, 2017; Feng et al.,
49 2015; Qian et al., 2018; Feng et al., 2019). Moreover, the tropospheric O_3 can perturb the radiative
50 energy budget of the earth-atmosphere system as the third most important greenhouse gas next to
51 carbon dioxide (CO_2) and methane (CH_4), thereby changing the global climate (Bornman et al.,
52 2019; Fu et al., 2019; Wang et al., 2019). Recently, the particulate matter less than 2.5 μm ($PM_{2.5}$)
53 concentration showed the persistent decrease, while the O_3 issue has been increasingly prominent
54 in China (Li et al., 2017b; Li et al., 2019b). Therefore, it was critical to accurately reveal the
55 spatiotemporal variation of O_3 pollution and assess its heath risk in China.

56 A growing body of studies began to investigate the spatiotemporal variation of O_3 level
57 worldwide. Wang et al. (2014b) demonstrated that the 8-h O_3 concentration in nearly all of the
58 provincial cities experienced the remarkable increase during 2013-2014. Following the work, Li et
59 al. (2017) reported that the annually mean O_3 concentration over China increased by 9.18% during
60 2014-2016. In other Asian countries except China, Vellingiri et al. (2015) performed long-term
61 obervation and found that the O_3 concentration in Seoul, South Korea displayed gradual increase in
62 the past decades. In the Southeast United States, Li et al. (2018) observed that the surfac~~e~~e O_3
63 concentration ~~also~~ displayed the gradual decrease in the recent ten years. Although the number of
64 ground-level monitoring sites have been increasing globally, the limited monitoring sites still cannot
65 accurately reflect the fine-scale O_3 pollution status because each site shows small spatial
66 representativeness (0.25-16.25 km^2) (Shi et al., 2018). Furthermore, the number of monitoring

sites in many countries (e.g., China and the United States) displays uneven distribution characteristic at the spatial scale. In China, most of these sites focus on North China Plain (NCP) and Yangtze River Delta (YRD), while West China extremely lacks ~~of~~the ground-level O_3 data, which often increases the uncertainty of health assessment. Therefore, many studies used various models to estimate the O_3 concentration without monitoring sites. Chemical transport models (CTMs) were often considered as the typical methods to predict the surface O_3 level. Zhang et al. (2011) employed the Geos-Chem model to simulate the surface O_3 concentration over the United States, suggesting that the model could capture the spatiotemporal variation of surface O_3 concentration at a large spatial scale. Later on, Wang et al. (2016) developed a hybrid model named land use regression (LUR) coupled with CTMs to predict the surface O_3 concentration in the Los Angeles Basin, California. In recent years, these methods were also applied to estimate the surface O_3 level over China. Liu et al. (2018) used Community Multiscale Air Quality (CMAQ) model to simulate the nationwide O_3 concentration over China in 2015. Nonetheless, the high-resolution O_3 prediction using CTMs might be widely deviated from the measured value owing to the imperfect knowledge about the chemical mechanism and the higher uncertainty of emission inventory. Moreover, the continuous emission data of NO_x and VOCs were not always open access, which restricted the long-term estimation of surface O_3 concentration using CTMs.

Fortunately, the daily satellite data enable~~s~~ the fine-scale estimation<u>s</u> of O_3 level at a regional scale due to broad spatial coverage and high temporal resolution (McPeters et al., 2015). Shen et al. (2019) confirmed that satellite retrieved O_3 column amount can accurately reflect the spatiotemporal distribution of surface O_3 level. Therefore, some studies tried to use

traditional statistical models coupled with high-resolution satellite data to estimate the ambient O_3 level. Fioletov et al. (2002) used the satellite measurement to investigate the global distribution of O_3 concentrations based on simple linear model. Recently, Kim et al. (2018) employed the integrated empirical geographic regression method to predict the long-term (1979-2015) variation of ambient O_3 concentration over United States based on O_3 column amount data. Although the statistical modelling of ambient O_3 concentration is widespread all around the world, most of these traditional statistical modelling only utilized the linear model to predict the ambient O_3 concentration, which generally decreased the prediction performance because the nonlinearity and high-order interactions between O_3 and predictors cannot be managed by a simple linear model.

As an extension of traditional statistical model, machine learning methods have been widely applied to estimate the pollutant level in recent years because of their excellent predictive performances. Among these machine learning algorithms, decision tree models such as random forest (RF) and extreme gradient boosting (XGBoost) ~~strike a perfect balance between prediction performance and computing cost~~generally showed fast training speed and excellent prediction accuracy (Li et al., 2020; Zhan et al., 2018). Furthermore, decision tree models can obtain the contributions of each predictor to air pollutants, which was beneficial to the parameter adaption and model optimization. Chen et al. (2018b) firstly employed RF model to simulate the $PM_{2.5}$ level in China since 2005. Following this work, we recently used the XGBoost model to estimate the 8-h O_3 concentration in Hainan Island for the first time and captured the moderate predictive performance ($R^2 = 0.59$) (Li et al., 2020). While decision tree model shows many advantages in predicting pollutant level, the spatiotemporal autocorrelation

of pollutant concentration is not concerned by these studies. Li et al. (2019a) confirmed that the prediction error by decision tree model varied greatly with space and time. Thus, it is imperative to incorporate the spatiotemporal variables into the original model to further improve the performance. To resolve the defects of decision tree models, Zhan et al. (2018) developed a hybrid model named RF-spatiotemporal Kriging (STK) to predict the O_3 concentration over China and achieved the better performance (Overall R^2 = 0.69, Southwest China R^2 = 0.66). Unfortunately, RF-STK model still showed some weaknesses in predicting O_3 concentration. First of all, the predictive performance of the STK model was strongly dependent on the number of monitoring sites and their spatial density. The model often showed worse predictive performance in the region with sparse monitoring sites (Gao et al., 2016). Moreover, the ensemble model cannot simulate the O_3 level during the periods without ground-measured data. In contrast, generalized additive model (GAM) not only considers the time autocorrelation of O_3 concentration, but also shows the better extrapolation ability (Chen et al., 2018a; Ma et al., 2015). Thus, the ensemble model of RF and GAM is proposed to predict the spatiotemporal variation of surface 8-h O_3 concentration.

Tibetan Plateau, the highest plateau around the world, shows the higher surface solar radiation compared with the region outside the plateau. It was well documented that high solar radiation ~~is beneficial to~~tended to generate large amount of OH radical, resulting in the O_3 formation via the reaction of VOC and OH radical (Ou et al., 2015). While the total O_3 column amount in Tibetan Plateau displayed the slight decrease since 1990s, the convergent airflow formed by subtropical anticyclones could bring ozone-rich air surrounding the plateau to the low atmosphere (Lin et al., 2008), thereby leading to the higher surface O_3 concentration over

the plateau. Most studies focused on the stratosphere-troposphere transport of O_3 in Tibetan Plateau, whereas limited effort was spared to investigate ground-level O_3 level over this region. To date, only several studies concerned about the spatiotemporal variation of surface O_3 concentration in this region based on field-observation data (Chen et al., 2019; Shen et al., 2014; Yin et al., 2017b). Unfortunately, these scarce monitoring sites in Tibetan Plateau cannot capture real O_3 pollution status especially in the remote areas (e.g., Northern part of Tibetan Plateau) because each site only possessed limited spatial representativeness. Apart from these field measurements, Liu et al. (2018) ($R = 0.60$) and Zhan et al. (2018) ($R^2 = 0.66$) used CTM and machine learning model to simulate the surface O_3 concentration over China in 2015, respectively. Both of these studies included the predicted O_3 level in Tibetan Plateau. Although they have finished the pioneering work, the predictive performances of both studies were not very excellent. Therefore, it was imperative to develop a higher quality model to enhance the modelling accuracy.

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Here, we developed a new hybrid method (RF-GAM) model integrating satellite data, meteorological factors, and geographical variables to simulate the gridded 8-h O_3 concentrations over Tibetan Plateau for the first time. Based on the estimated surface O_3 concentration, we clarified the long-term variation (2005-2018) of surface O_3 concentration and quantified the key factors for the annual trend. Filling the gap of statistical estimation 8-h O_3 level in a remote region, this study provides useful datasets for epidemiological studies and air quality management.

2. Materials and methods

2.1 Study area

Tibetan Plateau is located in Southwest China ranging from 26.00 to 39.58°N and from 73.33 to 104.78°E. Tibetan Plateau is surrounded by Taklamakan Desert to north, Sichuan Basin to southeast. The land area of Tibetan Plateau reaches 2.50 million km^2 (Chan et al., 2006). Based on the air circulation pattern, Tibetan Plateau can be roughly classified into the monsoon-influenced region and the westerly-wind influenced region (Wang et al., 2014a). The annually mean air temperature in most regions are below 0°C. The annually mean rainfall amount in Tibetan Plateau ranges from 50 to 2000 mm. The terrain conditions are complex and the higher altitude focused on the central region. Tibetan Plateau is generally treated as the remote region lack of anthropogenic activity and most of the residents focus on southeast and south parts of Tibetan Plateau. Tibetan Plateau ~~is consisted~~consists of 19 prefecture-level cities and their names and corresponding geographical locations are shown in Fig. 1 and Fig. S1.

2.2 Data preparation

2.2.1 Ground-level 8-h O_3 concentration

The daily 8-h O_3 data in 37 monitoring sites over Tibetan Plateau from May 13th , 2014 to December 31th, 2018 were collected from the national air quality monitoring network. The O_3 levels in all of these sites were determined using an ultraviolet-spectrophotometry method. The highest 8-h moving average O_3 concentration each day was calculated as the daily 8-h O_3 level after data quality assurance. The data quality of all the monitoring sites ~~were~~ was assured on the basis of the HJ 630-2011 specifications. The data with no more than two consecutive hourly measurement missing in each day was treated as the valid data.

2.2.2 Satellite-retrieved O_3 column amount

The O_3 column amounts (DU) during 2005-2018 were downloaded from the Ozone Monitoring

Instrument-O_3 (OMI-O_3) level-3 data with a 0.25° spatial resolution from the website of National Aeronautics and Space Administration (NASA) (https://www.nasa.gov/). The OMI-O_3 product shows global coverage and traverses the earth once a day. The O_3 column amount with cloud radiance fraction > 0.5, terrain reflectivity > 30%, and solar zenith angles > 85° should be removed. In addition, the cross-track pixels significantly influenced by row anomaly should be deleted.

2.2.3 Meteorological data and geographical covariates

The daily meteorological data were obtained from ERA-Interim datasets with 0.125° resolution. These meteorological data were consisted of 2 meter dewpoint temperature (d2m), 2 meter temperature (t2m), 10 meter U wind component (u10), 10 meter V wind component (v10), boundary layer height (blh), sunshine duration (sund), surface pressure (sp), and total precipitation (tp). The 30 m-resolution elevation data (DEM) was downloaded from China Resource and Environmental Science Data Center (CRESDC). The data of gross domestic production (GDP) and population density with 1 km resolution were also extracted from CRESDC. Population density and GDP in 2005, 2010, and 2015 were integrated into the model to predict the surface 8-h O_3 concentration over Tibetan Plateau because these data were available each five years. Additionally, the land use data of 30 m resolution (e.g., waters, grassland, urban, forest) were also extracted from CRESDC. At last, the latitude, longitude, and time were also incorporated into the model.

All of the explanatory variables collected were resampled to 0.25° × 0.25° grids to predict the O_3 level. The original meteorological data with 0.125° resolution were resampled to 0.25° grid. The land use area, elevation, GDP and population density in each grid were calculated using spatial clipping. Lastly, all of the predictors were integrated into an intact table to train the model.

2.3 Model development and assessment

The RF-GAM model was regarded as the hybrid model of RF and GAM. The RF-GAM model is a two-stage model that the prediction error estimated by the RF model was then simulated by GAM. The prediction results of RF and GAM were summed as the final result of RF-GAM model (Fig. 2). The detailed equation is as follows:

$$Z(s,t) = P(s,t) + E(s,t) \quad (1)$$

where Z(s,t) is the estimated 8-h O_3 level at the location s and time t; P(s,t) represents the 8-h O_3 concentration predicted by the RF model; E(s,t) denotes the prediction error by GAM.

In the RF model, a large number of decision trees were planted based on the bootstrap sampling method. At each node of the decision tree, a random sample of all predictors was applied to determine the best split among them. Following the procedure, a simple majority vote was employed to predict the 8-h O_3 level. The RF model avoided priori linear assumption of O_3 concentration and predictors, which was often not in good agreement with actual state. The RF model has two key parameters including n_{tree} (the number of trees grown) and m_{try} (the number of explanatory variables sampled for splitting at each node). The prediction performance of the RF model was strongly dependent on the two parameters. The optimal n_{tree} and m_{try} were determined based on the least out-of-bag (OOB) errors. Based on the iteration result, the optimal n_{tree} and m_{try} reached 500 and 5, respectively. Besides, the backward variable selection method was performed on the RF submodel to achieve the better performance. At each step of the predictor selection, the variable with the least important value was excluded from the next step. This one-variable-at-a-time exclusion method was repeated until only two explanatory variables remained in the submodel. Finally, all of the selected variables except the area of waters were integrated into the model to achieve the best prediction performance. The detailed RF model is as follows:

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$$O_3 = O_3\ column + Elevation + Agr + Urban + Forest + GDP + Grassland + Population + \Pr ec + T + WS + P + tsun + RH \quad (2)$$

where O_3 denotes the observed 8-h O_3 level in the monitoring site; the O_3 column represents the O_3 column amount in the corresponding grid; Elevation denotes the corresponding elevation of the site; Agr, Urban, Forest, Grassland are the agricultural land, urban land, forest land, and the grassland, respectively. Population represents the population density in the corresponding site. Prec, T, WS, P, tsun, and RH are precipitation, air temperature, wind speed, air pressure, sunshine duration, and relative humidity, respectively. Additionally, other five models including RF, generalized regression neutral network (GRNN), backward propagation neural network (BPNN), Elman neural network (ElmanNN), and extreme learning machine (ELM) also used the backward variable selection method. The R^2 value was treated as an important parameter to add or reduce the variable. The variable should be removed when the R^2 value of the submodel showed the remarkable decrease with the integration of this variable. Lastly, the optimal variable group was applied to establish the submodel.

Following the RF submodel, the prediction error estimated by the RF submodel was further modelled by the GAM. GAM could reflect the time autocorrelation of predictive error of RF model, and thus the ensemble model of RF and GAM might decrease the modelling error of one-stage model. All of the variables were incorporated into the models to establish the second-stage model, and the backward variable selection was also used to determine the optimal variable group.

The 10-fold cross-validation (CV) technique was employed to evaluate the predictive performances for all of the machine learning models. All of the training data set were randomly classified into 10 subsets uniformly. In each round of validation, nine subsets were used to train and the remaining subset was applied to test the model performance. The process was repeated 10 times until every subset has been tested. Some statistical indicators including R^2, Root Mean Square Error

(RMSE), Mean Prediction Error (MPE) and the slope were calculated to assess the model performance. The optimal model with the best performance was used to estimate the 8-h O_3 concentration in the past decades.

3. Results and discussion

3.1 The validation of model performance

Figure 3 and figure S2 show the density scatterplots of the fitting and 10-fold cross-validation results for eight machine learning models for China. The 10-fold cross-validation R^2 values followed the order of RF-GAM ($R^2 = 0.76$) > RF-STK ($R^2 = 0.63$) > RF ($R^2 = 0.55$) > GRNN ($R^2 = 0.53$) > BPNN ($R^2 = 0.50$) > XGBoost ($R^2 = 0.48$) > ElmanNN ($R^2 = 0.47$) > ELM ($R^2 = 0.32$). The RMSE values of RF-GAM, RF-STK, RF, GRNN, XGBoost, BPNN, ElmanNN, and ELM were 14.41, 17.79, 19.13, 19.41, 20.73, 20.06, 20.61, and 23.36 μg/m^3, respectively. Both of MPE and RPE showed the similar characteristic with RMSE in the order of RF-GAM (10.97 μg/m^3 and 26.50%) < RF-STK (13.48 μg/m^3 and 35.15%) < RF (14.71 μg/m^3 and 35.51%) < GRNN (14.89 μg/m^3 and 35.82%) < BPNN (15.43 μg/m^3 and 36.19%) < ElmanNN (15.75 μg/m^3 and 37.05%) < XGBoost (15.80 μg/m^3 and 38.13%) < ELM (18.23 μg/m^3 and 44.05%) (Fig. 3 and Fig. S2). Besides, the slope of the RF-GAM model was closer to 1 compared with other models. It was well documented that the RF model generally showed the better performance than other models because this method did not need to define complex relationships between the explanatory variables and the O_3 concentration (e.g., linear or nonlinear). Furthermore, the variable importance indicators calculated by the RF model can help user to distinguish the key variables from noise ones and make full use of the strength of each predictor to assure the model robustness. Although BPNN, GRNN, XGBoost, ElmanNN, and ELM have been widely applied to estimate the air pollutant concentrations

(Chen et al., 2018c; Zang et al., 2018; Zhu et al., 2019), these methods suffered from some weaknesses in predicting the pollutant level. For instance, both of BPNN and ElmanNN models could capture the locally optimal solution when the training subsets were integrated into the final model, which decreased the predictive performance of the model (Wang et al., 2015). Moreover, BPNN generally showed slow training speed, especially with the huge training subsets (Li and Park, 2009; Wang et al., 2015). ELM often consumed more computing resource and experienced the over-fitting issue due to the increase of sampling size (Huang et al., 2015; Shao et al., 2015). GRNN method advanced the training speed compared with BPNN model and avoided the locally optimal solution during the modelling process (Zang et al., 2019), whereas the predictive performance is still worse than that of RF model. XGBoost was often considered to be robust in predicting air pollutant level (Li et al., 2020), while the model did not display the excellent performance in the present study. It might be attributable to that the sampling size in the present study was not enough because the model generally showed the better performance with big samples. Moreover, we found that the two-stage model was superior to the one-way model in the predictive performance. The encouraging result suggested that the relationship between the predictors and the 8-h O_3 concentration varied with space and time. The two-stage model used the GAM method to further adjust the prediction error of the RF model, and considered the spatiotemporal correlation of predictor error in Tibetan Plateau. Although the STK model incorporated space and time into the model simultaneously, the RF-GAM model outperformed the RF-STK model. It was assumed that the STK model showed the higher uncertainty in predicting the O_3 concentration in the region with scarce sampling sites (Gao et al., 2016; Li et al., 2017a). Overall, the ensemble RF-GAM model showed the significant improvement in predictive performance.

The performances of RF-GAM displayed slight difference in each year during 2014-2018. As shown in Table 1, the R^2 value showed the highest value (0.76) in 2016, followed by that in 2018 (0.75), 2017 (0.73), 2015 (0.72), and the lowest one in 2014 (0.69). Both of RMSE and MPE exhibited the lowest values in 2014, while these parameters did not show significant variation during 2015-2018. The lowest R^2 value and the highest RPE focused on 2014 due to the least sample size, while the highest R^2 value and lowest RPE in 2016 was contributed by the maximum sample size. Geng et al. (2018) found that the predictive performance of machine learning model was strongly dependent on the number of training samples and sampling frequency. The lower RMSE and MPE in 2014 might be attributable to the lack of measured O_3 data in spring, which decreased the higher value of O_3 concentration. The performances of the RF-GAM model in four seasons were also assessed by 10-fold cross-validation (Table 2). The predictive performance of the RF-GAM model showed significantly seasonal difference with the highest R^2 value observed in summer (0.74), followed by winter (0.69) and autumn (0.67), and the lowest one in spring (0.64). However, both of RMSE and MPE displayed different seasonal characteristics with the R^2 value. Both of RMSE and MPE for RF-GAM followed the order of spring (15.32 and 11.94 μg/m^3) > summer (15.13 and 11.75 μg/m^3) > winter (14.58 and 11.44 μg/m^3) > autumn (13.23 and 10.52 μg/m^3). The lowest R^2 value in spring might be caused by multiple O_3 sources and complicate O_3 formation mechanisms. On the one hand, the O_3 in spring might be generated from the local anthropogenic emission or long-range transport (Li et al., 2017; Li et al., 2019b). On the other hand, a strong stratosphere-troposphere exchange process due to lower height of troposphere in Tibetan Plateau might lead to the higher O_3 concentration in spring (Skerlak et al., 2014). Unfortunately, both of long-range transport and stratosphere-troposphere exchange process were missing in the RF-GAM model, which restricted

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the accuracy of O_3 estimation in spring. The large estimation errors (e.g., RMSE ~~and~~ , MPE, and RPE) in spring and summer were attributable to the high 8-h O_3 concentration in these seasons, while the low prediction error observed in autumn was contributed by the low O_3 level.

Apart from the seasonal variation, we also investigated the spatial ~~variability~~ variabilities of the predictive accuracy for RF-GAM model. Tibetan Plateau was classified into five provinces and then the predictive performance of RF-GAM model in each province was calculated. Among the five provinces, Gansu province displayed the highest R^2 value (0.74), followed by Sichuan province (0.71), Qinghai province (0.70), Tibet autonomous region (0.69), and Yunnan province (0.54) (Table 3). The result shown herein was not in agreement with the previous studies by Geng et al. (2018), who confirmed that the predictive performance of machine learning model was positively associated with the sampling size. It was assumed that the spatial distribution of the sampling sites in Tibet was uneven and the sampling density is low, though Tibet possessed the maximum monitoring sites compared with other provinces. The prediction error (RMSE and MPE) did not exhibit the same characteristics with the R^2 value. The higher RMSE and MPE focused on Tibet autonomous region (14.81 and 11.24 μg/m^3) and Qinghai province (14.83 and 11.33 μg/m^3) due to the higher values of blh and sund. The lowest values of RMSE and MPE could be observed in Yunnan province, which was contributed by the higher rainfall amount. The highest RPE was concentrated on Yunnan province (25.85%), followed by Tibet (22.90%), Qinghai (22.65%), Sichuan (22.62%), and the lowest one in Gansu province (22.51%), which might be linked with the sample size.

Although 10-fold cross-validation verified that the RF-GAM model showed the better predictive performance in estimating the surface 8-h O_3 concentration, the test method cannot validate the transferring ability of the final model. The monitoring site in Tibetan Plateau before May, 2014 is

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very limited, and only the daily 8-h O_3 data in Lhasa from the open website (https://www.aqistudy.cn/historydata/) was available to compare with the simulated data. As depicted in Fig. 4, the R^2 value of unlearning 8-h O_3 level against predicted 8-h O_3 concentration reached 0.67, which was slightly lower than that of the 10-fold cross-validation R^2 value. Overall, the extrapolation ability of the RF-GAM model is satisfactory, and thus it was supposed that the model could be applied to estimate the O_3 concentration in other years. Both of RMSE and MPE for the unlearning 8-h O_3 level against the predicted 8-h O_3 concentration were significantly higher than those of the 10-fold cross-validation. It was supposed that Lhasa showed the higher surface 8-h O_3 concentration over Tibetan Plateau.

To date, some previous studies also simulated the surface O_3 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 O_3 concentration over China, and explained the 66% spatial variability of O_3 level in Tibetan Plateau. Apart from these statistical models, some classical CTMs were also applied to estimate the O_3 concentration in the remote area. Both of Liu et al. (2018) and Lin et al. (2018) used CMAQ to estimate the O_3 level across China, while the R^2 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 O_3 concentration; and (2) the use of high-quality satellite data.

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3.2 Variable importance

The results of variable importance for key variables are depicted in Fig. 5. In the final RF-GAM model, it was found that time was the dominant factor for the 8-h O_3 concentration in Tibetan Plateau,

indicating that the ambient O_3 concentration displayed significantly temporal correlation. Following the time, meteorological factors served as the main factors for the O_3 pollution in the remote region. The sum of sund, sp, d2m, t2m, and tp occupied 34.43% of the overall variable importance. Among others, sund was considered to be the most important meteorological factors for the O_3 pollution. It was assumed that strong solar radiation and long duration of sunshine favored the photochemical generation of ambient O_3 (Malik and Tauler, 2015; Stähle et al., 2018). Tan et al. (2018) demonstrated that the chemical reaction between NO_x and VOCs was strongly dependent on the sunlight. Besides, the atmospheric pressure (sp) was also treated as a major driver for the O_3 pollution over Tibetan Plateau. Santurtún et al. (2015) have demonstrated that sp was closely linked to the atmospheric circulation and synoptic scale meteorological pattern, which could influence the long-range transport of ambient O_3. Apart from sund and sp, d2m and t2m played significant role on the O_3 pollution, which was in consistent with many previous studies (Zhan et al., 2018). Zhan et al. (2018) observed that cold temperature was not favorable to the O_3 formation. d2m can affect the surface O_3 pollution through two aspects. On the one hand, RH affected heterogeneous reactions of O_3 and particles (e.g., soot, mineral) (He et al., 2017; He and Zhang, 2019; Yu, 2019). On the other hand, high RH could increase the soil moisture and evaporation, and thus the water-stressed plants tended to emit more biogenic isoprene, thereby promoting the elevation of O_3 concentration (Zhang and Wang, 2016). It should be noted that the effect of precipitation on O_3 pollution was relatively weaker than those of other meteorological factors. Zhan et al. (2018) also found the similar result and believed that rain scavenging served as the key pathway for the O_3 removal only when O_3 pollution was very serious. The power of O_3 column amount on surface O_3 concentration seemed to be lower than those of most meteorological factors, suggesting that vertical transport of ambient

376 O_3 was complex. Although socioeconomic factors and land use types were not dominant factors for
377 the O_3 pollution in Tibetan Plateau, they still cannot be ignored in the present study because the
378 predictive performance would worsen if these variables were excluded from the model. It was
379 widely acknowledged that the anthropogenic emissions ~~of NO_x and VOCs~~ focused on the developed
380 urban areas with high population density especially in the remote plateau (Zhang et al., 2007; Zheng
381 et al., 2017). Compared with the urban land, the grassland played more important role on the O_3
382 pollution in Tibetan Plateau. It was thus supposed that the grassland was widely distributed on
383 Tibetan Plateau, which could release a large amount of biogenic volatile organic compounds
384 (BVOCs) (Fang et al., 2015). It was well known that photochemical reactions of BVOCs and NO_x
385 in the presence of sunlight caused ~~was beneficial to~~ the O_3 formation (Calfapietra et al., 2013; Yu et
386 al., 2006). Furthermore, Fang et al. (2015) confirmed that the BVOC emission in Tibetan Plateau
387 displayed a remarkable increase in the wet seasons.

388 3.3 The spatial distribution of estimated 8-h O_3 concentration over Tibetan Plateau

389 Figure 6 depicts the spatial distribution of the 8-h O_3 level estimated by the novel RF-GAM
390 model. The spatial distribution pattern modelled by the RF-GAM model showed the similar
391 characteristic with the result simulated by previous studies except North Tibetan Plateau (Liu et al.,
392 2018). The estimated 8-h O_3 concentration displayed the highest value in some cities of North
393 Tibetan Plateau such as Huangnan (73.48±4.53 μg/m^3) and Hainan (72.24±5.34 μg/m^3), followed
394 by the cities in the central region including Lhasa (65.99±7.24 μg/m^3) and Shigatse (65.15±6.14
395 μg/m^3), and the lowest one in some cities of Southeast Tibetan Plateau such as Aba (55.17±12.77
396 μg/m^3). The spatial pattern of 8-h O_3 concentration is highly consistent with the result predicted by
397 Liu et al. (2018) using CMAQ model, while it is not in agreement with the result estimated by Zhan

et al. (2018) using RF-STK model. The difference of the present study and Zhan et al. (2018) focuses on the North Tibetan Plateau, which lacks of monitoring site and remains the higher uncertainty. Firstly, it might be contributed by the weakness of RF-STK mentioned above. Moreover, Zhan et al. (2018) only used the ground-level measured data in 2015 to establish the model and the data in new sites since 2015 were not incorporated into the model, which could increase the model uncertainty (Zhan et al., 2018). As shown in Fig. 6, most of the cities in Qinghai province (e.g., Huangnan, Hainan, and Guoluo) generally showed the higher 8-h O_3 concentration over Tibetan Plateau, which was in a good agreement with the spatial distribution of O_3 column amount (Fig. S3). Besides, some cities in Tibet such as Shigatse and Lhasa also showed the higher 8-h O_3 levels. It was supposed that the precursor ~~(e.g., NO_x and VOCs)~~ emissions in these regions were significantly higher than those in other cities of Tibetan Plateau (Fig. S4). Zhang et al. (2007) used the satellite data to observe that the higher VOCs and NO_x emission focused on the residential area with high population density in the remote Tibetan Plateau. Apart from the effect of anthropogenic emission, the meteorological conditions could be also the important factors for the 8-h O_3 concentration. As shown in Fig. S5-S10, the higher blh and sp in the Northeast Tibetan Plateau ~~were beneficial to~~might promote the O_3 formation through the reaction of VOC and OH radical, leading to the higher 8-h O_3 concentration in these cities (Ou et al., 2015). In addition, the lower tp occurred in North Tibetan Plateau and Northeast Tibetan Plateau, both of which were unfavorable to the ambient O_3 removal (Yoo et al., 2014). In contrast, the higher tp observed in the Southeast Tibetan Plateau resulted in the slight O_3 pollution.

3.4 The temporal variation of the simulated 8-h O_3 concentration over Tibetan Plateau

The annually mean estimated 8-h O_3 concentration in Tibetan Plateau displayed the slow

increase from 64.74 ± 8.30 μg/m^3 to 66.45 ± 8.67 μg/m^3 2005 through 2015 (Table S1), whereas it decreased from the peak to 65.87 ± 8.52 μg/m^3 during 2015-2018 (Fig. 7). Based on the Mann-Kendall method (Fig. 8a), it was found that the surface O_3 concentration exhibited the slight increase as the whole, while the increase degree was not significant ($p > 0.05$). Besides, it should be noted that the O_3 concentrations in various regions showed different increase speed. As depicted in Fig. 8b, we found that the 8-h O_3 concentrations in North, West, and East Tibetan Plateau displayed significant increase trend by the speed of 1-3 μg/m^3 during 2005-2018. The middle region of Tibetan Plateau showed the moderate increase trend by the speed of 0-1 μg/m^3. However, the 8-h O_3 concentration in Shigatse and Sannan even displayed the decrease trend 2005 through 2018.

Besides, the 8-h O_3 concentrations in Tibetan Plateau displayed significantly seasonal discrepancy. The estimated 8-h O_3 level in Tibetan Plateau followed the order of spring (75.00±8.56 μg/m^3) > summer (71.05±11.13 μg/m^3) > winter (56.39±7.42 μg/m^3) > autumn (56.13±8.27 μg/m^3) (Fig. 9 and Table 4). The 8-h O_3 concentrations in most of prefecture-level cities showed the similarly seasonal characteristics with the overall seasonal variation in Tibetan Plateau. Based on the result summarized in Table S2, it was found that the key precursors of ambient O_3 generally displayed the higher emissions in winter compared with other seasons. However, the seasonal distribution of ambient O_3 concentration was not in accordance with the precursor emissions, suggesting that the meteorological factors might play more important roles on ambient O_3 concentration. It was well known that the higher air temperature in spring and summer ~~was~~ were closely related to the low sp and high sund, both of which ~~were beneficial to the~~promoted O_3 formation (Sitnov et al., 2017). Although summer showed the highest air temperature and the longest sunshine duration, the higher rainfall amount in summer decreased the ambient O_3 concentration

via wet deposition (Li et al., 2017b; Li et al., 2019b). Moreover, the highest blh occurred in spring, which was favorable to the strong stratosphere-troposphere exchange process in Tibetan Plateau (Skerlak et al., 2014). Therefore, the 8-h O_3 concentration in summer and winter were relatively lower than that in spring. Nonetheless, the 8-h O_3 levels in Diqing, Sannan, and Nyingchi displayed the highest values in spring (56.38±7.87, 73.90±5.97, and 73.22±2.77 μg/m^3), followed by winter (45.88±7.05, 61.71±4.32, and 62.24±3.63 μg/m^3) and summer (44.35±5.90, 61.00±5.86, and 59.60±2.33 μg/m^3), and the lowest ones in autumn (37.45±5.76, 54.70±3.13, and 53.84±2.06 μg/m^3). The lower O_3 level in summer than winter was mainly attributable to the higher precipitation observed in the summer of these cities (Fig. S11). In addition, it should be noted that the NO_x and VOCs emissions of South Tibetan Plateau (e.g., Sannan) exhibited the higher values in winter compared with other seasons.

3.5 The nonattainment days over Tibetan Plateau during 2005-2018

The annually mean nonattainment days in the 19 prefecture-level cities over Tibetan Plateau are summarized in Table 2. 100 μg/m^3 was regarded as the critical value for the 8-h O_3 level by World Health Organization (WHO). The nonattainment days denoted total days with the 8-h O_3 concentration higher than 100 μg/m^3. Although the annually mean 8-h O_3 concentrations in all of the cities over Tibetan Plateau did not exceed the critical value, not all of the regions experienced excellent air quality in the long period (2005-2018). Some cities of Qinghai province including Huangnan, Haidong, and Guoluo suffered from 45, 40, and 40 nonattainment days each year (Fig. 10 and Table 5). Besides, some cities in the South Tibetan Plateau such as Shigatse and Sannan also experienced more than 40 nonattainment days each year, suggesting that Tibetan Plateau was still faced of the risk for O_3 pollution. Fortunately, some remote cities such as Ali, Ngari, and Qamdo

did not experience the excessive O_3 pollution all the time, which was ascribed to the low precursor emissions and appropriate meteorological conditions. It should be noted that the nonattainment days in the region with high O_3 concentration showed the significantly seasonal difference, whereas the seasonal difference was not remarkable in the city with low O_3 pollution. As shown in Table 2, it should be noted that nearly all of the nonattainment days could be detected in spring and summer, which was in good agreement with the O_3 levels in different seasons, indicating that the O_3 pollution issue should be paid more attention in spring and summer.

The determination of nonattainment days showed some uncertainties owing to the predictive error of modelled O_3 concentration. First of all, meteorological data used in RF-GAM model were collected from reanalysis data and these gridded data often showed some uncertainties, which could increase the uncertainty of O_3 estimation. Second, the O_3 column amount used in the present study reflected vertical O_3 column amount rather than surface O_3 concentration. Thus, it could decrease the predictive performance of surface O_3 level.

4. Summary and implication

In the present study, we developed a novel hybrid model (RF-GAM) based on multiple explanatory variables to estimate the surface 8-h O_3 concentration across the remote Tibetan Plateau. The 10-fold cross-validation method demonstrated that RF-GAM achieved excellent performance with the highest R^2 value (0.76) and lowest root mean square error (RMSE) (14.41 μg/m^3) compared with other model including RF-STK, RF, BPNN, XGBoost, GRNN, ElmanNN, and ELM models. Moreover, the unlearning ground-measured O_3 data validated that the RF-GAM model showed the better extrapolation performance (R^2=0.67, RMSE=25.68 μg/m^3). The result of variable importance suggested that time, sund, and sp were key factors for the surface 8-h O_3 concentration over Tibetan

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Plateau. Based on the RF-GAM model, we found that the estimated 8-h O_3 concentration exhibited notably spatial variation with the highest value in some cities of North Tibetan Plateau such as Huangnan (73.48±4.53 μg/m^3) and Hainan (72.24±5.34 μg/m^3) and the lowest one in some cities of Southeast Tibetan Plateau such as Aba (55.17±12.77 μg/m^3). Besides, we also found that the O_3 level displayed a slow increase from 64.74±8.30 μg/m^3 to 66.45±8.67 μg/m^3 2005 through 2015, while the O_3 concentration decreased to 65.87±8.52 μg/m^3 in 2018. The estimated 8-h O_3 level in Tibetan Plateau showed the significantly seasonal discrepancy with the order of spring (75.00±8.56 μg/m^3) > summer (71.05±11.13 μg/m^3) > winter (56.39±7.42 μg/m^3) > autumn (56.13±8.27 μg/m^3). Based on the critical value set by WHO, most of the cities in Tibetan Plateau shared with the excellent air quality, while several cities (e.g., Huangnan, Haidong, and Guoluo) still suffered from more than 40 nonattainment days each year.

The RF-GAM model for O_3 estimation has several limitations. First of all, the O_3 estimation in North Tibetan Plateau might show some uncertainties because the ground-level monitoring site is very scarce, and thus we cannot validate the reliability of predicted value in the region without monitoring site. Secondly, our approach did not include data on emission inventory, or traffic count because the continuous emissions of NO_x and VOCs were not open access. At last, we only focused on the temporal variation of surface O_3 concentration in recent ten years, and the short-term O_3 data cannot reflect the response of O_3 pollution to climate change. In the future work, we should combine more explanatory variables such as long-term NO_x and VOCs emissions to retrieve the surface O_3 level over Tibetan Plateau in the past decades.

Author contributions

This study was conceived by Rui Li and Hongbo Fu. Statistical modelling was performed by Rui

Li, Yilong Zhao, Ya Meng, Wenhui Zhou and Ziyu Zhang. Rui Li drafted the paper.

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Figure and table captions

Fig. 1 The geographical locations and annually mean 8-h O_3 concentrations in the ground-observed sites (red dots) over Tibetan Plateau during 2014-2018. The elevation data are collected from geographical and spatial data cloud at a 30-m spatial resolution.

Fig. 2 The workflow for predicting the spatiotemporal distributions of 8-h O_3 levels.

Fig. 3 Density scatterplots of model fitting and cross-validation result at a daily level. (a), (b), and (c) represent RF-GAM, RF-STK, and RF models, respectively. The red dotted line denotes the fitting linear regression line. The full names of MPE and RMSE are mean prediction error ($\mu g/m^3$) and root mean squared prediction error ($\mu g/m^3$), respectively.

Fig. 4 The transferring ability validation of RF-GAM method based on the measured daily 8-h O_3 concentration during December 2013-May 2014.

Fig. 5 The variable importance of predictors in the final RF-GAM model.

Fig. 6 The mean value of estimated 8-h O_3 concentration during 2005-2018 over Tibetan Plateau.

Fig. 7 The inter-annual variation of predicted 8-h O_3 level ($\mu g/m^3$) from 2005 to 2018 across Tibetan Plateau.

Fig. 8 The trend analysis of predicted 8-h O_3 concentration. (a) and (b) represent the result of Mann-Kendall method and discrepancy of estimated O_3 level during 2005-2018 across Tibetan Plateau.

Fig. 9 The seasonal variability of estimated 8-h O_3 level across Tibetan Plateau. (a), (b), (c), and (d) represent the predicted 8-h O_3 concentrations in spring, summer, autumn, and winter, respectively.

Fig. 10 The spatial distributions of nonattainment days in Tibetan Plateau during 2005-2018.

Table 1 The R^2 values, RMSE, MPE, and RPE of RF-GAM in different years during 2014-2018 over Tibetan Plateau.

Table 2 The R^2 values, RMSE, ~~and~~ MPE, and RPE of RF-GAM in four seasons over Tibetan Plateau.

Table 3 The predictive performances ~~R^2 values, RMSE, and MPE~~ of RF-GAM in different provinces over Tibetan Plateau.

Table 4 The estimated 8-h O_3 concentration in 19 prefecture-level cities over Tibetan Plateau during four seasons including spring, summer, autumn, and winter.

Table 5 The mean nonattainment days (8-h O_3 level >100 μg/m^3) in 19 prefecture-level cities over Tibetan Plateau each year.

Fig. 1

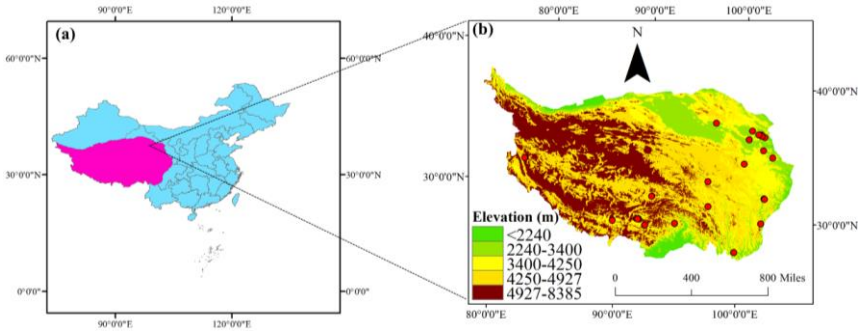
(a)
90°0'0"E
120°0'0"E
60°0'0"N
30°0'0"N
0°0'0"
(b)
80°0'0"E
90°0'0"E
100°0'0"E
40°0'0"N
30°0'0"N
N
Elevation (m)
<2240
2240-3400
3400-4250
4250-4927
4927-8385
0
400
800 Miles

Fig. 2

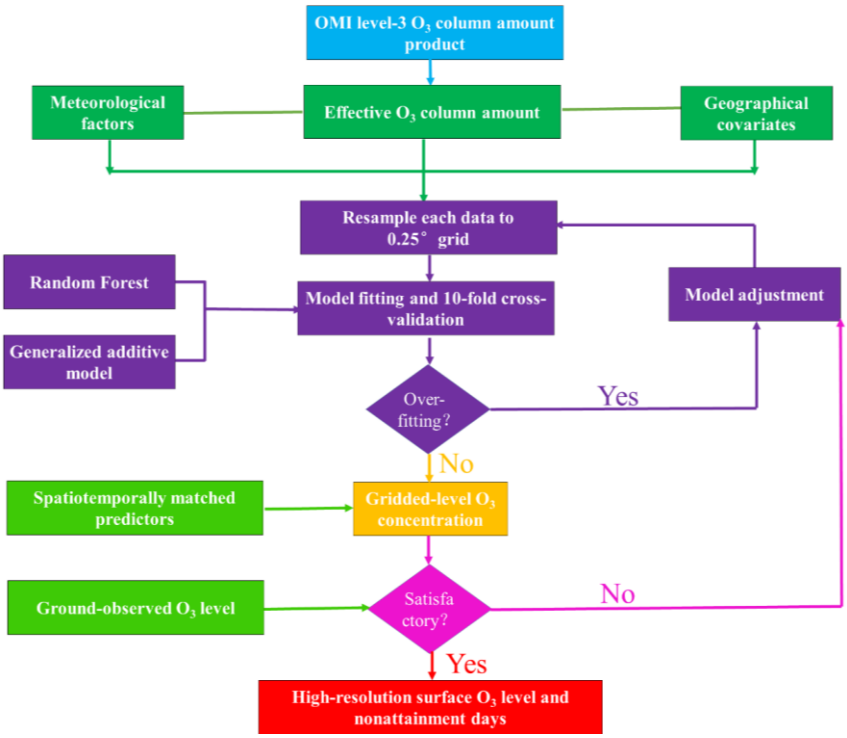
OMI level-3 O_3 column amount product
Meteorological factors
Effective O_3 column amount
Geographical covariates
Resample each data to 0.25° grid
Random Forest
Generalized additive model
Model fitting and 10-fold cross-validation
Model adjustment
Over-fitting?
Yes
No
Spatiotemporally matched predictors
Gridded-level O_3 concentration
Ground-observed O_3 level
Satisfa ctory?
No
Yes
High-resolution surface O_3 level and nonattainment days

Fig. 3

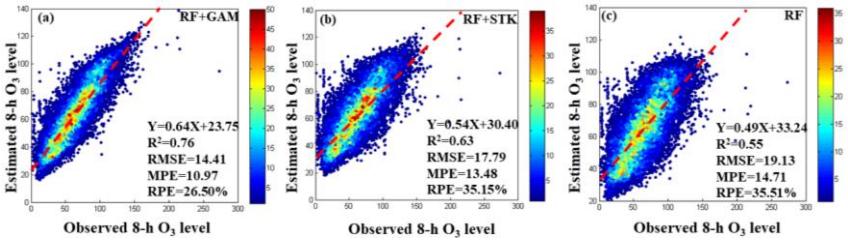
(a)
RF+GAM
Y=0.64X+23.75
R²=0.76
RMSE=14.41
MPE=10.97
RPE=26.50%
Estimated 8-h O3 level
Observed 8-h O3 level
(b)
RF+STK
Y=0.54X+30.40
R²=0.63
RMSE=17.79
MPE=13.48
RPE=35.15%
Estimated 8-h O3 level
Observed 8-h O3 level
(c)
RF
Y=0.49X+33.24
R²=0.55
RMSE=19.13
MPE=14.71
RPE=35.51%
Estimated 8-h O3 level
Observed 8-h O3 level

Fig. 4

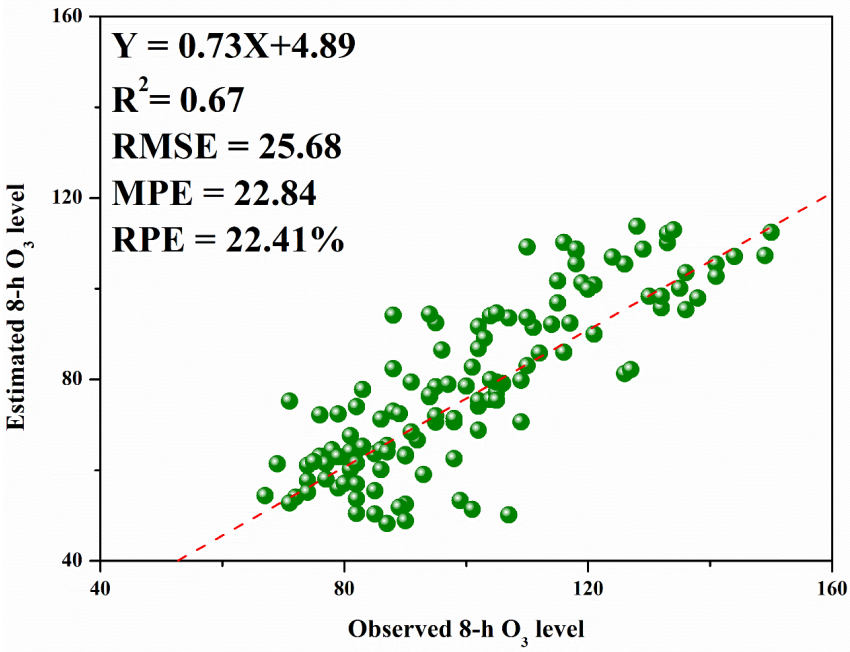
Y = 0.73X+4.89
R2= 0.67
RMSE = 25.68
MPE = 22.84
RPE = 22.41%
Estimated 8-h O3 level
Observed 8-h O3 level
160
120
80
40
40
80
120
160

Fig. 5

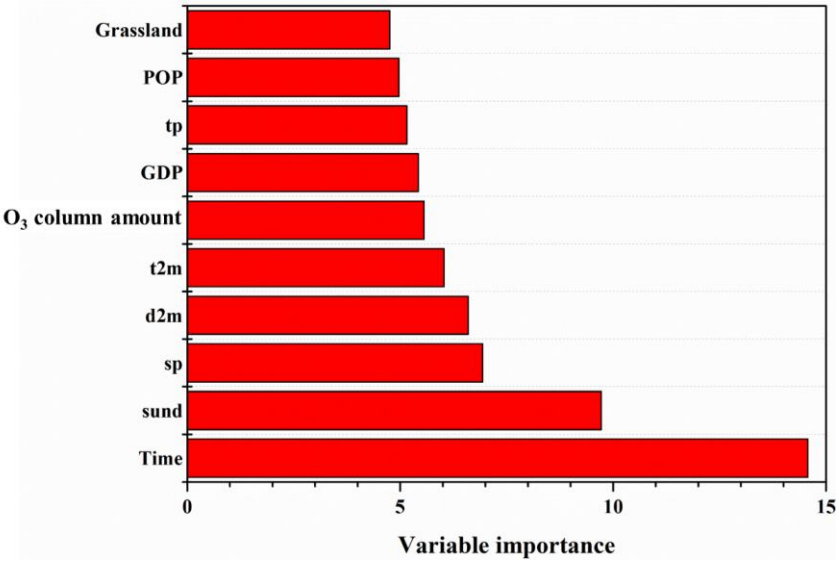
Grassland
POP
tp
GDP
O3 column amount
t2m
d2m
sp
sund
Time
0
5
10
15
Variable importance

Fig. 6

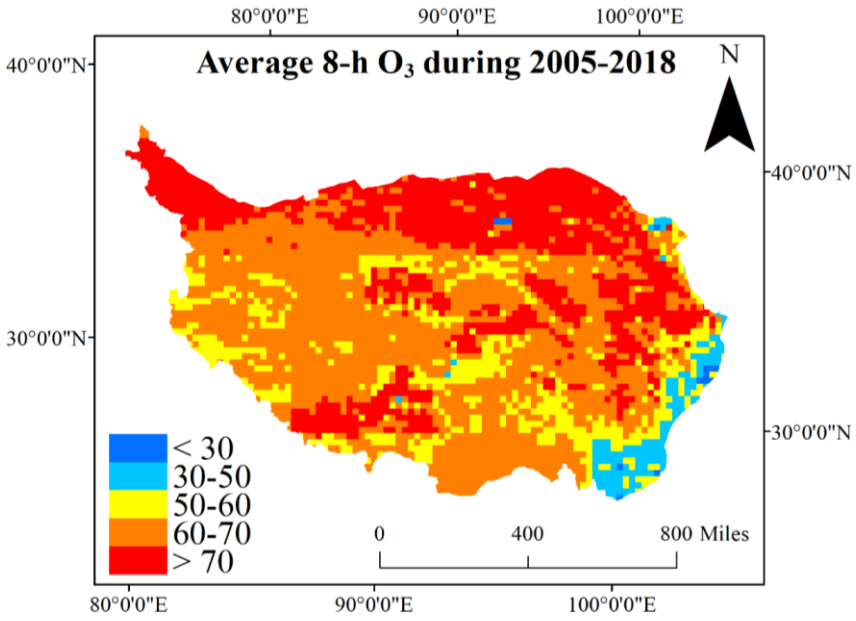
80°0'0"E
90°0'0"E
100°0'0"E
40°0'0"N
30°0'0"N
Average 8-h O3 during 2005-2018
N
< 30
30-50
50-60
60-70
> 70
0
400
800 Miles

Fig. 7

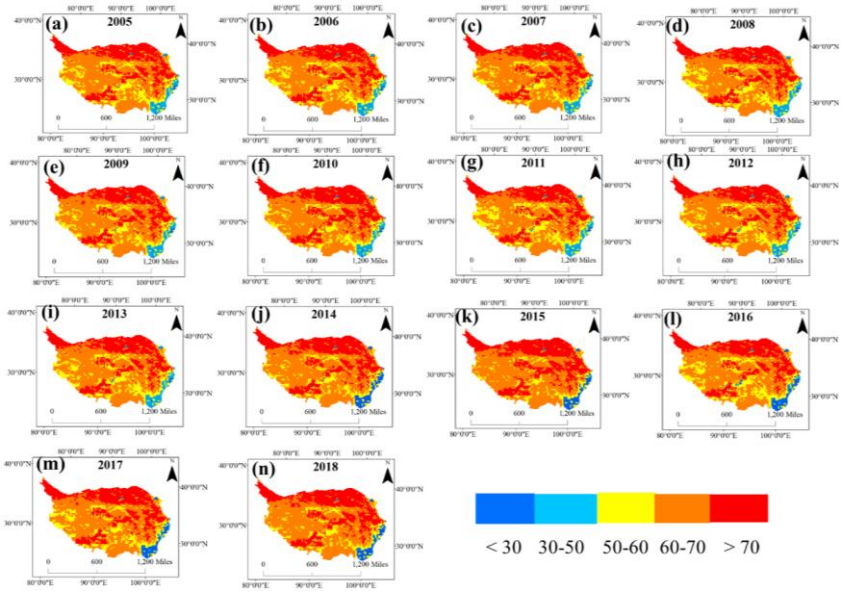
(a) 2005
(b) 2006
(c) 2007
(d) 2008
(e) 2009
(f) 2010
(g) 2011
(h) 2012
(i) 2013
(j) 2014
(k) 2015
(l) 2016
(m) 2017
(n) 2018
< 30
30-50
50-60
60-70
> 70

Fig. 8

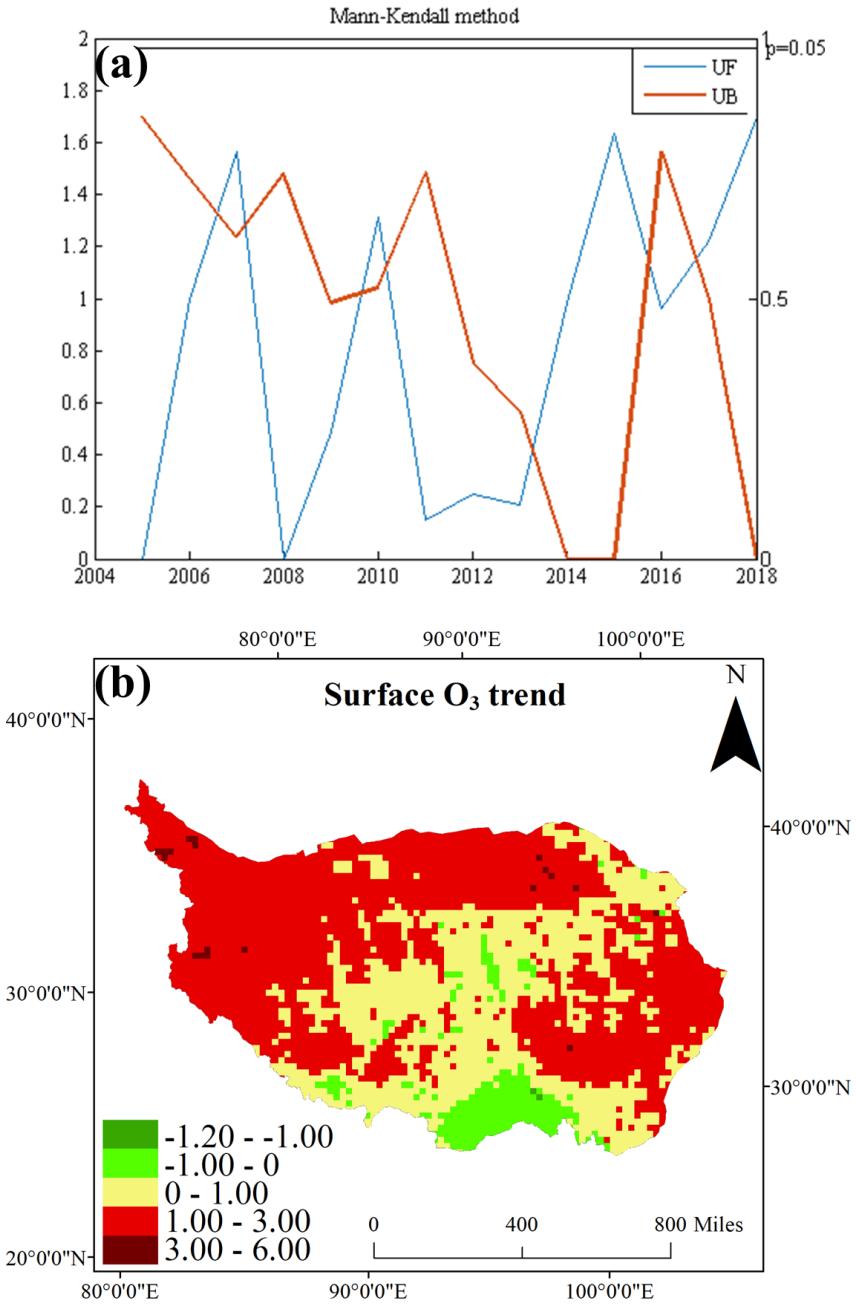
Mann-Kendall method
(a)
UF
UB
p=0.05
2
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
0.5
2004
2006
2008
2010
2012
2014
2016
2018
80°0'0"E
90°0'0"E
100°0'0"E
(b)
Surface O_3 trend
N
40°0'0"N
30°0'0"N
20°0'0"N
-1.20 - -1.00
-1.00 - 0
0 - 1.00
1.00 - 3.00
3.00 - 6.00
0
400
800 Miles

Fig. 9

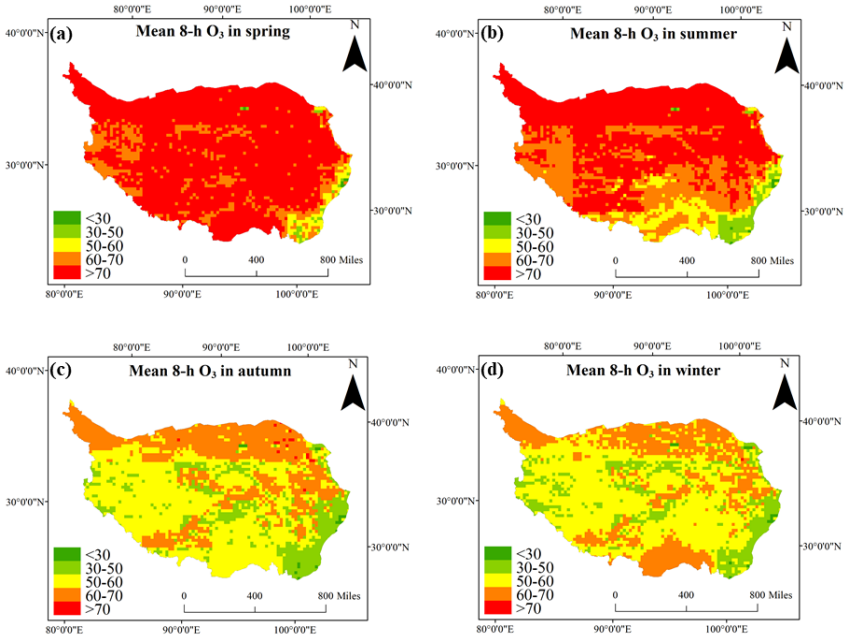
(a) Mean 8-h O_3 in spring
(b) Mean 8-h O_3 in summer
(c) Mean 8-h O_3 in autumn
(d) Mean 8-h O_3 in winter
80°0'0"E
90°0'0"E
100°0'0"E
40°0'0"N
30°0'0"N
N
<30
30-50
50-60
60-70
>70
0
400
800 Miles

Fig. 10

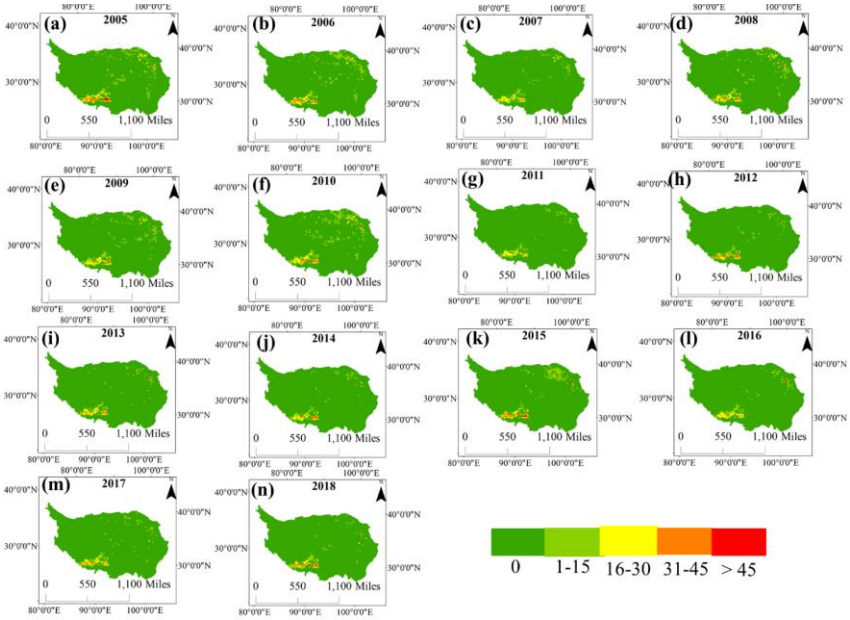
(a) 2005
(b) 2006
(c) 2007
(d) 2008
(e) 2009
(f) 2010
(g) 2011
(h) 2012
(i) 2013
(j) 2014
(k) 2015
(l) 2016
(m) 2017
(n) 2018
0 550 1,100 Miles
0 1-15 16-30 31-45 >45

Table 1

	2014	2015	2016	2017	2018
R^2	0.69	0.72	0.76	0.73	0.75
RMSE	13.65	14.56	14.28	14.52	14.35
MPE	9.53	10.82	10.84	10.95	10.93
RPE	23.27%	23.26%	23.02%	23.20%	23.09%

Table 2

	Spring	Summer	Autumn	Winter
R^2	0.64	0.74	0.67	0.69
RMSE	15.32	15.13	13.23	14.58
MPE	11.94	11.75	10.52	11.44
RPE	24.63%	22.35%	23.32%	23.24%

Table 3

	Tibet	Qinghai	Gansu	Sichuan	Yunnan
R^2	0.69	0.70	0.74	0.71	0.54
RMSE	14.81	14.83	13.65	13.23	12.49
MPE	11.24	11.33	10.88	10.08	10.20
RPE	22.90%	22.65%	22.51%	22.62%	25.85%

Table 4

	Province	Spring	Summer	Autumn	Winter	Annual	Measured O_3 level
Aba	Sichuan	65.61±14.30	59.46±14.32	45.55±12.03	47.95±10.55	55.17±12.77	47.75±19.47
Ngari	Tibet	71.34±3.12	70.10±3.57	53.14±3.67	51.84±3.69	62.21±3.34	53.34±24.46
Qamdo	Tibet	72.52±4.29	62.74±5.79	52.06±4.01	55.42±3.09	61.10±3.93	59.76±23.77
Diqing	Yunnan	56.38±7.87	44.35±5.90	37.45±5.76	45.88±7.05	46.22±6.51	47.81±21.63
Gannan	Gansu	76.77±9.73	73.27±10.67	54.74±8.33	54.72±6.95	65.60±8.91	68.86±25.45
Ganzi	Sichuan	69.38±10.99	61.45±11.58	48.49±8.79	50.94±6.62	58.06±9.48	38.07±19.08
Guoluo	Qinghai	80.12±5.12	76.13±5.83	58.86±5.71	57.38±4.66	68.77±5.25	80.04±23.90
Haibei	Qinghai	78.18±10.21	78.84±10.31	60.90±9.69	57.48±9.78	69.47±9.99	81.07±32.74
Haidong	Qinghai	74.20±10.34	73.70±9.12	53.61±8.11	51.02±9.60	63.84±9.21	44.28±34.96
Hainan	Qinghai	83.01±5.36	82.27±5.72	61.57±5.39	58.96±5.44	72.24±5.34	78.34±27.11
Haixi	Qinghai	79.39±6.88	79.48±7.79	60.78±7.48	57.71±6.99	69.99±7.24	80.60±27.17
Huangnan	Qinghai	85.21±4.98	83.01±4.66	61.95±4.18	60.62±4.49	73.48±4.53	74.83±22.63
Lhasa	Tibet	80.08±9.63	70.13±8.42	55.86±5.78	55.85±5.19	65.99±7.24	75.45±26.65
Nagqu	Tibet	74.59±5.13	70.46±6.69	54.60±5.16	53.53±4.83	63.83±5.23	44.79±28.75
Shigatse	Tibet	77.31±8.62	69.66±7.69	55.93±4.58	55.57±4.72	65.15±6.14	75.62±26.50
Sannan	Tibet	73.90±5.97	61.00±5.86	54.70±3.13	61.71±4.32	63.04±4.00	73.04±26.31
Xining	Qinghai	77.43±10.27	77.84±9.44	58.19±9.29	54.72±10.04	67.77±9.70	61.77±22.58
Yushu	Qinghai	77.35±5.55	73.34±6.37	56.12±5.53	55.02±5.01	66.05±5.50	57.14±31.98
Nyingchi	Tibet	73.22±2.77	59.60±2.33	53.84±2.06	62.24±3.63	62.40±2.20	66.61±26.71

Table 5

	Spring	Summer	Autumn	Winter	Annual
Aba	0	0	0	0	0
Ngari	0	0	0	0	0
Qamdo	0	0	0	0	0
Diqing	0	0	0	0	0
Gannan	0	1	0	0	1
Ganzi	13	2	0	0	15
Guoluo	19	21	0	0	40
Haibei	0	0	0	0	0
Haidong	22	18	0	0	40
Hainan	14	12	1	0	27
Haixi	1	1	0	0	2
Huangnan	23	22	0	0	45
Lhasa	12	7	0	0	19
Nagqu	24	14	0	0	38
Shigatse	28	13	0	0	41
Sannan	33	7	0	0	40
Xining	2	1	0	0	3
Yushu	0	0	0	0	0
Nyingchi	0	0	0	0	0