Referee #1

The paper discusses the sensitivity of surface ozone and $PM_{2.5}$ in China to meteorological parameters. The information presented in the paper is useful to understand the interaction between pollution and meteorology, and regional difference in the sensitivity of emission control measures. I'd recommend the publication of the paper if the following comments are addressed:

(1) The method description is very brief, and the details in implementation may affect the interpretation of the results. In particular, I see one difficulty in this type of sensitivity simulation that a simple perturbation of individual parameters may lead to unphysical meteorological fields. For example, increasing/decreasing T by 1 K under some conditions may turn saturated/unsaturated air into unsaturated/saturated, but since only T is perturbed, no cloud is dissipated/formed in response to changing T. Another example, a simple perturbation of wind speed may generate a wind field that violates the physics, and is inconsistent with the pressure field that feeds into the air quality simulation, which may lead to spurious sensitivities in the result. Even more difficult is to perturb wind direction, though I notice the authors did not assess the wind direction sensitivity. In general, I'd like to see if and how this type of issues is handled by the authors. The current method description is too brief to tell the exact implementation. Other useful details to include are if the perturbations are done for the entire atmosphere or only in the boundary layer, if they are done for the whole day uniformly or only in the daytime.

Responses: To clarify how we perturb the meteorological parameters, we added the following sentences in the method section:

"All perturbations were implemented uniformly in space on the modeling domain and in time through the modeling periods. The perturbations on temperature, wind speed, and absolute humidity were made in all layers. To separate the effects of individual meteorological parameters, only one parameter was changed in each case while all other parameters were kept unchanged. Therefore, cloud dissipating or forming in response to changing temperature was not considered in the simulations. When perturbing horizontal wind speed, to avoid unphysical situations that mass would not be conserved, the vertical wind speed was adjusted in the vertical transport calculation based on the air density changes to conserve mass."

(2) The responses of emissions to meteorological parameters are not included in the assessment. The responses of emissions to meteorology is a significant contributor to the overall meteorological sensitivity of ozone and $PM_{2.5}$. To name a few, the effect of T on biogenic emissions, the effect of T on soil NOx emissions, the cloud cover/convection on lightning NOx emissions, the effect of T on power plant NOx emissions (high T leads to higher electricity demand in summer). Because emissions are held unchanged in the simulations, these effects are not included, which makes the analysis incomplete and less informative. This caveat needs to be discussed in the paper.

Responses: Thanks for the comments. In the method section, we added the following sentences:

"It is worthwhile to note that some meteorological parameters could have significant impacts on emissions, such as the effect of T on biogenic VOC and soil NOx emissions, the cloud cover/convection on lightning NOx emissions, the effect of T on power plant NOx emissions (high T leads to higher electricity demand in summer), which would affect air quality. Therefore, the sensitivities in this study only include the 'direct' effects of individual meteorological parameters on air quality. A full evaluation of the impacts of climate/weather changes on air quality should consider effects of the emissions changes."

(3) Evaluation against observations. The O3-T slope from model simulations is often found to be much lower than that derived from observations, suggesting that model tends to underestimate the sensitivity of O3 to meteorology. The current paper provides no evaluations of how good the model in use could reproduce the observed chemical-met relationship. Note this evaluation is different from evaluation of chemical concentrations, and is perhaps more relevant for the current work.
Responses: Thank you for your valuable advice. We conducted the evaluation of the O3-T relationship, following the method in Rasmussen et al. (2012). We have no O_3 observations in January (O_3 observations became available from March 2013 in China), so we only evaluated the results in July in the five cities as in the manuscript. We found that CMAQ overestimated the O_3-T relationship (CMAQ: 2.4 ppb/K vs. observation: 0.8 ppb/K, shown in Figure S1). Please note that we only have 1 month data and we use daily MDA8 O3 and daily maximum temperature in the evaluation, while a much more meaningful evaluation should be performed to use monthly averaged MDA8 O3 and monthly average temperature over a long-term period. We added above evaluation and discussion in the revised manuscript.

(4) In abstract and elsewhere (such as Line 282), the authors compare the different sensitivities. For instance, the paper says in Line 282 that "the sensitivity of O3 to T is obviously higher than that of WS, AH, and PBLH". This is to compare apples to oranges, because these sensitivities are in different units! The delta concentrations of O3 or PM2.5 from two simulations apparently depend on how much you perturb, and it is meaningless to compare which one is bigger unless the perturbations are carefully defined to relate to the variations of individual parameters.
Responses: We deleted the description of the comparison among different meteorological parameters because of the different unit problem.

Referee #2

The authors here have studied the effect of perturbation of meteorology on PM2.5 and O3 concentration across China. Overall, the manuscript was well written, method is sound, results are valid. I would recommend it to be published after addressing the following issues.

1. The authors here conduct the sensitivity analysis by perturbing the value of one meteorological parameter and keeping value of others constant. In real world when a meteorological parameter changes, a corresponding change in other parameters also takes place. This will affect the entire results.

Responses: For 'real world' meteorology changes, climate/weather forecasting models are usually utilized to predict how an entire set of meteorological parameters will change under certain scenarios and to estimate the impacts on air quality. Despite the large uncertainties in predicting 'real world' climate changes, another problem with this method is that it is impossible to isolate the effects of individual meteorological parameters.

Sensitivity studies are commonly used to achieve this objective by perturbing one parameter at a time and keeping other parameters unchanged. This method may not reflect 'real world' changes, but can provide information that the first method cannot provide, and this method has been applied in several studies, such as Dawson et al. (2007a), Dawson (2007b), and Horne et al. (2017).

2. Even if we assume that the authors are trying to only depict the sensitivity of PM2.5 and O3 on perturbation of meteorological parameters, the above said knowledge won't be handy to the authorities when trying to implement emission control in such scenarios. Since perturbation of one meteorological parameter will result in corresponding change in other parameters and since the current simulation is only based on assumption that only one parameter will change at any given time, the results from current sensitivity analysis won't be of any use.

Responses: The results are useful for implementing emission controls in several aspects. First, the results help identify the major meteorological factors to which PM2.5 and O3 have the largest sensitivities. For example, our results indicate that in July O3 is very sensitive to temperature but not so sensitive to PBL height in Beijing. Therefore, additional emission controls would be needed if temperature is predicted to increase in future, but not necessary if PBL height is predicted to increase (while temperature is predicted no significant increase).

Second, the results show that the PM2.5 sensitivities to these meteorological parameters are mainly through secondary components (SO_4^{2-}, NO_3^-, NH_4^+, and SOA). Therefore, more emission controls on the precursors of the secondary components would be needed in future to overcome the adverse impacts of meteorological condition changes on PM2.5.

Third, this study aims to isolate the effects of individual meteorological parameters on air quality. It is very straightforward to quantify the combined effects of changes in several meteorological parameters. As an example, we conducted an additional simulation to test the impact of all perturbations (T+1.0K, WS-10%, AH+10%, PBLH-20%, CLW+10%, and PCP+10%) on O3 and PM2.5 in January and July, and the results was shown in Fig.S8 in the revised manuscript.

3. Solar radiation apart from temperature is also one of the main factors affecting O3 why haven't the authors studied sensitivity of O3 concentration to change in solar radiation.

Responses: Solar radiation affects photolysis rates. In CMAQ, the photolysis rates are calculated in-line. First the clear-sky photolysis rates are calculated using the clear-sky actinic flux. Then photolysis rates are corrected to account for the effects of cloud and particle extinction. The actinic flux is calculated in real time as a function of time of day, longitude, latitude, altitude, and season, therefore is not perturbed in this study.

4. The authors doesn't mention on what basis they change the meteorological parameters i.e. on what basis is the magnitude of change in parameters considered.

Responses: The magnitude ranges of perturbations are based on IPCC AR5 report and the study of Dawson et al. (2007) and the references therein. For each parameter, three positive and three negative perturbations were then designed within its range to have a more comprehensive examination on the sensitivity of $PM_{2.5}$ and O_3 to this parameter. We add the above information in the method section.

5. Line 179-182, the authors discuss regarding effect of Temperature on Ozone in Ozone forming regime. Any references to suggest that the said areas in China are in ozone forming or ozone consumption regimes?

Response: The net O3 formation areas and the net O3 loss areas are classified based on the O3 concentrations (shown Fig. 2). The background O3 is about 35 ppb, therefore, areas with O3 concentrations over 35 ppb is the net O3 formation areas, and areas with O3 concentrations less than 35 ppb is the net O3 loss areas. We added the explanation in the revised manuscript.

6. In Figures S8-S13, the authors estimate the quantitative sensitivity of O3 and PM2.5 concentrations to change in individual meteorological parameters by linear fitting of the changes. The authors should also report the corresponding R-squared, slope and significance values, it would help to understand the rate of change of PM2.5 or O3 per change in meteorological parameters and if at all the rate of change is statistically significant.

Responses: Thanks for your suggestion. We added these metrics in Fig. S8-S13.

7. Does the authors perturb meteorology parameters only for China in the domain? As per spatial variation figures, the domain also constitutes parts of south-east Asia?

All perturbations were implemented uniformly in space on the modeling domain and in time through the modeling periods. The perturbations on temperature, wind speed, and absolute humidity were made in all layers. We have added above explanation in the method section.

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Sensitivity Analysis of the Surface Ozone and Fine Particulate Matter to Meteorological Parameters in China

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Abstract

Meteorological conditions play important roles in the formation of ozone (O_3) and fine particulate matter ($PM_{2.5}$). China has been suffering from serious regional air pollution problems, characterized by high concentrations of surface O_3 and $PM_{2.5}$. In this study, the Community Multiscale Air Quality (CMAQ) model was used to quantify the sensitivity of surface O_3 and $PM_{2.5}$ to key meteorological parameters in different regions of China. Six meteorological parameters were perturbed to create different meteorological conditions, including temperature (T), wind speed (WS), absolute humidity (AH), planetary boundary layer height (PBLH), cloud liquid water content (CLW) and precipitation (PCP). Air quality simulations under the perturbed meteorological conditions were conducted in China in January and July of 2013. The changes in O_3 and $PM_{2.5}$ concentrations due to individual meteorological parameters were then quantified. T has ~~the greatest impact~~a great influence on the daily maximum 8-h average O_3 (O_3-8h) concentrations, which leads to O_3-8h increases by 1.7 ppb K^{-1} in January in Chongqing and 1.1 ppb K^{-1} in July in Beijing. WS, AH, and PBLH have a smaller but notable influence on O_3-8h with maximum change rates of 0.3, -0.15, and 0.14 ppb $\%^{-1}$, respectively. T, WS, AH, and PBLH have important effects on $PM_{2.5}$ formation of in both January and July. In general, $PM_{2.5}$ sensitivities are negative to T, WS, and PBLH and positive to AH in most regions of China. The sensitivities in January are much larger than in July. $PM_{2.5}$ sensitivity to T, WS, PBLH, and AH in January can be up to -5 μg m^{-3} K^{-1} , -3 μg m^{-3} $\%^{-1}$, -1 μg m^{-3} , and +0.6 μg m^{-3} $\%^{-1}$, respectively, and in July can be up to -2 μg m^{-3} K^{-1}, -0.4 μg m^{-3} $\%^{-1}$, -0.14 μg m^{-3} $\%^{-1}$, and +0.3 μg m^{-3} $\%^{-1}$, respectively. Other meteorological factors (CLW and PCP) have negligible effects on O_3-8h (less than 0.01 ppb $\%^{-1}$) and $PM_{2.5}$ (less than 0.01 μg m^{-3} $\%^{-1}$). The results suggest that surface O_3 and $PM_{2.5}$ concentrations can

change significantly due to changes in meteorological parameters and it is necessary to consider these effects when developing emission control strategies in different regions of China.

Keywords: sensitivity, meteorological conditions, fine particulate matter, ozone, CMAQ model

1. Introduction

China has serious air pollution problems and fine particulate matter ($PM_{2.5}$) and ozone (O_3) are the two major air pollutants (Lin et al., 2010; Hu et al., 2016; Lu et al., 2019; Wu et al., 2019). The annual average $PM_{2.5}$ concentrations were higher than 50 μg m^{-3} in 26 out of the total 31 provincial capital cities in mainland China during 2013-2014 (Wang et al., 2014a), and the national 4th highest daily maximum 8-hour average O_3 (O_3-8h) is 86.0 ppb during the warm-seasons (April–September) in 2013-2017, which is 6.3–30% higher than that in other industrialized regions of the world (Lu et al., 2018). $PM_{2.5}$ alone caused 0.87-1.36 million deaths every year in China, and long-term exposure to O_3 was responsible for an extra 254 000 deaths (Apte et al., 2015; Cohen et al., 2017; Hu et al., 2017b; Silver et al., 2018). China has made remarkable improvement in air quality during recent years (Zhang et al., 2017; Zhao et al., 2017; China, 2018; Zheng et al., 2018), however, air pollution is still severe, making it the fourth-ranked healthy risk factor (Stanaway et al., 2018).

Surface $PM_{2.5}$ and O_3 concentrations are determined by atmospheric processes of emissions, transport and dispersion, chemical transformation (due to gas-phase, aqueous-phase and aerosol chemistry), and dry and wet deposition. These processes are affected by meteorological conditions. Studies have shown that the surface O_3 and $PM_{2.5}$ concentrations are sensitive to different meteorological parameters. For example, Dawson et al. (2007b) have investigated the sensitivity of surface O_3 to different meteorological parameters in the eastern United States (US) using the comprehensive air quality model with extensions (CAM_X). The results showed that temperature (T) had the greatest influence on daily O_3-8h of 0.34 ppb K^{-1}, followed by absolute humidity (AH) of 0.025 ppb $\%^{-1}$. Bernard et al. (2001) also confirmed that T presented a notable positive correlation with the surface O_3 concentration. The effects of meteorological parameters on $PM_{2.5}$ are even more complicated. Tran and Mölders (2011) showed that elevated $PM_{2.5}$ concentrations tended to occur under the condition of calm wind, low T and relative humidity in Fairbanks, Alaska. Olvera Alvarez et al. (2018) used a land use regression model to analyze the effects of different meteorological parameters on $PM_{2.5}$ in El Paso, Texas and obtained the same conclusion in winter, but in spring, the high $PM_{2.5}$ level was associated with high wind speed (WS) and low humidity. Dawson et al. (2007a) studied the effects of individual meteorological parameters in the Eastern US and found that $PM_{2.5}$ concentration decreased markedly as the increased precipitation (PCP) in winter, but in summer, the main meteorological factors affecting the $PM_{2.5}$ concentration were T, WS and planetary boundary layer height (PBLH). Dawson et al. (2009) simulated the effects of climate change on regional and urban air quality in the Eastern US, and found $PM_{2.5}$ concentration decreased by 0.3 μg m^{-3} in January mostly due to increasing in PCP and increased by 2.5 μg m^{-3} in July largely due to decreasing in PBLH and WS. Horne and Dabdub (2017) altered various meteorological parameters to investigate their effects on O_3, $PM_{2.5}$ and secondary organic aerosols (SOA), and found that the T predominated the effects of meteorology in California.

Many studies have proved that meteorological conditions play very important roles in air pollution events in China. Studies found that the pollutant concentrations could vary up to several times, due to meteorological changes with the same emission sources (Zhang et al., 2010; Xing et al., 2011; Zheng et al., 2015; Cai et al., 2017; Liu et al., 2017; Ning et al., 2018; Yang et al., 2018; Li et al., 2019b). For example, Xing et al. (2011) studied that the difference between the effects of 2007 and 2008 meteorological conditions on air quality during the 2008 Beijing Olympics. They found higher humidity in August 2008 was beneficial to the formation of SO_4^{2-} by up to ~60%, and lower T prevented the evaporation of NO_3^- by up to ~60%. Liu et al. (2017) reported that the monthly mean $PM_{2.5}$ concentrations in the Jing-Jin-Ji (JJJ) area in December 2015 increased by 5%~137% due to the unfavorable weather conditions such as low WS and high humidity.

A few studies investigated the relationships between air quality and meteorological conditions in China. Zhang et al. (2015) conducted a correlation analysis between air quality and meteorology in three megacities Beijing, Shanghai and Guangzhou in China. The result showed that air pollutants were significantly negatively correlated with WS, and O_3 had a positive correlation with T. Yin et al. (2016) found that the relationship between WS and $PM_{2.5}$ has complicated influence, with higher PM at low and high WS than in light to moderate winds in Beijing from 2008 to 2014. Xu et al. (2018) examined the variations of $PM_{2.5}$ concentration in January 2017 in China compared to that in January 2016 and found meteorological conditions of low WS, high humidity, low PBLH and low PCP contributed to $PM_{2.5}$ concentration worsening by 29.7%, 42.6% and 7.9% in the JJJ region, the Pearl River Delta (PRD) region and the Cheng-Yu Basin (CYB) region, respectively. Ma et al.

(2019) analyzed the effects of meteorology on air pollution in the Yangtze River Delta (YRD) region during 2014-2016 and found $PM_{2.5}$ was highly negatively correlated to WS, while O_3 concentration was positively correlated to T but negatively related to relative humidity. Zhu et al. (2017) reported that the surface concentrations of O_3 increased by 2-6 ppb in January and 8-12 ppb in July 2014 in PRD, mainly due to the increase in T and the decrease in NOx emissions.

These studies have investigated the impacts of meteorological conditions on $PM_{2.5}$ and O_3 in certain regions of China, however, quantitative sensitivity of $PM_{2.5}$ and O_3 to meteorological parameters has not been examined. The objective of this study is to quantify the sensitivity of O_3 and $PM_{2.5}$ to different meteorological parameters in winter and summer in different regions of China. The paper is constructed as following, Section 2 describes the method used to estimate the sensitivity, and Section 3 presents the effects of each meteorological variable on O_3 and $PM_{2.5}$ in China and in five representative cities. Conclusions are then summarized in Section 4.

2. Methods

The sensitivity of O_3 and $PM_{2.5}$ associated with changes in meteorological parameters was quantified using the Community Multiscale Air Quality (CMAQ) model version 5.0.2. The meteorological parameters include T, WS, AH, PBLH, PCP, and cloud liquid water content (CLW). A base case was firstly simulated with meteorological fields predicted by the Weather Research and Forecasting (WRF) model v3.7.1 (http://www.wrf-model.org/) using the NCEP FNL Operational Model

Global Tropospheric Analyses dataset as the initial and boundary conditions. The base case has been described in a previous study and the model configurations of the base case were reported there (Hu et al., 2015). The WRF predicted meteorological parameters and the CMAQ predicted surface O_3 and $PM_{2.5}$ have been evaluated against observations at 422 sites in 60 major cities in China, and the accuracy of the model performance has been validated (Hu et al., 2016).

A suit of perturbation scenarios was created, and in each scenario, a certain meteorological parameter was perturbed to a certain extent. The details of the perturbation scenarios are listed in Table 1. Among those changes, the T was absolute changes, and other parameters are relative variations. The magnitude ranges of perturbations are based on IPCC AR5 report and the study of Dawson et al. (2007) and the references therein. For each parameter, three positive and three negative perturbations were then designed within its range to have a more comprehensive examination on the sensitivity of $PM_{2.5}$ and O_3 to this parameter. All perturbations were implemented uniformly in space on the modeling domain and in time through the modeling periods. The perturbations on temperature, wind speed, and absolute humidity were made in all layers. To separate the effects of individual meteorological parameters, only one parameter was changed in each case while all other parameters were kept unchanged. Therefore, cloud dissipating or forming in response to changing temperature was not considered in the simulations. When perturbing horizontal wind speed, to avoid unphysical situations that mass would not be conserved, the vertical wind speed was adjusted in the vertical transport calculation based on the air density changes to conserve mass.

Then the CMAQ model was re-run to predict the air quality under the perturbed meteorological condition. The emissions and other inputs were kept unchanged in each perturbed meteorological scenarios, therefore the difference of O_3 and $PM_{2.5}$ concentrations between each of the perturbation case and the base case was due to the change in the specific meteorological parameter, and the sensitivity of O_3 and $PM_{2.5}$ to individual meteorological parameters could be quantitatively determined. It is worthwhile to note that some meteorological parameters could have significant impacts on emissions, such as the effect of T on biogenic VOC and soil NOx emissions, the cloud cover/convection on lightning NOx emissions, the effect of T on power plant NOx emissions (high T leads to higher electricity demand in summer), which would affect air quality. Therefore, the sensitivities in this study only include the ‘direct’ effects of individual meteorological parameters on air quality. A full evaluation of the impacts of climate/weather changes on air quality should consider effects of the emissions changes.

The modeling domain covers East Asia, including entire China, with a horizontal resolution of 36×36 km^2. The base case and perturbation cases were conducted in January and July in 2013, representing the winter and the summer conditions, respectively. In addition to the regional analysis, five representative megacities were selected, i.e., Beijing, Shanghai, Guangzhou, Chongqing, Xi’an (Fig.1). These cities are located in the North China Plain (NCP), YRD, PRD, CYB, and Guanzhong Plain, respectively, where serious air pollution problems often occur. In this study, O_3-8h was used in the O_3 analyses, and 24h average $PM_{2.5}$ was used in the $PM_{2.5}$ analyses, if not specifically stated. The O_3-T relationship is examined using the method in Rasmussen et al. (2012) in the five cities. Observed and predicted O_3-T relationships were estimated using the daily observed and predicted O_3-8h concentrations and daily maximum T in July (O_3 observations became available from March 2013 in China, so no O_3 observations in January). The results show that CMAQ overestimated the O_3-T relationship (CMAQ: 2.4 ppb/K vs. observation: 0.8 ppb/K, shown in Figure S1). Please note that we only have 1 month data and we use daily O_3-8h and daily maximum temperature in the evaluation, while a much more meaningful evaluation should be performed to use monthly averaged O_3 and monthly average temperature over a long-term period Rasmussen et al. (2012).

3. Results and Discussion

3.1 Impacts of meteorological parameters on surface O_3

Figs. 2(a) and 2(b) show the spatial-distribution of the predicted monthly average O_3-8h concentrations in January and July, respectively. In January, the highest average concentrations are about 70 ppb in the Sichuan Basin, and the concentrations in southern and eastern China are generally higher than those in northern China. In July, the highest average concentrations are over 80ppb in the large areas of NCP and YRD, CYB, and Guangzhou areas in the PRD.

Fig. 3 shows the spatial distribution of the concentration changes of O_3-8h in January and July due to change of T + 1.0 K, WS - 10%, AH +10% PBLH - 20%,CLW + 10% case, and PCP + 10%, respectively. Fig. S1-S3 shows the results due to other extent changes in these parameters. When T increases 1.0 K (Fig. 3(a)), O_3-8h increases 1-2 ppb in most area of eastern and central China in January and in NCP and YRD in July, which is consistent with the high O_3 spatial distribution in the base case (shown in Fig. 2). O_3-8h decreases up to 4 ppb in January in Northeast China, and up to 2 ppb in the Southwest border of China and the East China Sea, which are the areas of low O_3 concentrations (generally less than the background O_3 concentration of 35 ppb). Therefore, the effect of T on O_3 is dependent on the O_3 formation regime. An increase in T promotes O_3 formation chemistry in net O_3 formation areas (O_3 concentrations greater than 35 ppb), but accelerates O_3 consumption chemistry in the net O_3 loss areas (O_3 concentrations less than 35 ppb).

Fig. 3(b) shows that the differences of O_3-8h in January and July when WS is 10% less than the base case in 2013. The influence of wind on O_3 concentration is complex, but generally, slower WS decreases O_3 in January in most parts of China, particularly in Sichuan by up to 3 ppb, but increases O_3 in July by a few ppb over the most areas in eastern and central China. Therefore, the impact of WS on O_3 appears opposite in winter and summer. Weaker winds slow down the dispersion of NOx and VOCs, which is conducive to O_3 formation in summer when the vertical mixing is strong, but increases O_3 titration in the surface in winter due to weaker vertical mixing.

Fig. 3(c) displays that the surface O_3 is expected to decrease generally less than 1 ppb when AH increases by 10% (relative change) both in January and July in most land areas of China except in the northeast area. Fig. 3(d) shows that a 20% decrease of PBLH leads to O_3-8h decreases by a few ppb in most area in January, while in July O_3-8h increases in eastern and central regions, especially in YRD, CYB and areas in Hubei-Hunan-Jiangxi in the central China. Sensitivity of O_3 to CLW and PCP is relatively small. Fig. 3(e) demonstrates that O_3-8h changes -0.03 ppb to 0.03 ppb in January and July for a 10% increase in CLW. Fig. 3(f) demonstrates that a 10% increase in PCP results in -0.1 to 0.2 ppb changes in O_3-8h. O_3 changes due to the six meteorological factors with different extents of perturbation (Figs. S1-S3) shows the similar trends and spatial patterns.

3.2 Impacts of meteorological parameters on surface $PM_{2.5}$

Figs. 2(c) and 2(d) show the spatial distribution of the monthly average surface $PM_{2.5}$ concentrations in January and July. $PM_{2.5}$ in January reaches over 200 μg m^{-3} in JJJ, SYB, central China, and urban areas in the Northeast China. $PM_{2.5}$ is much lower in July, generally lower than 50 μg m^{-3}, but is high (up to 70 μg m^{-3}) in areas in the JJJ, YRD, and central China regions.

Fig. 4 shows the spatial distribution of $PM_{2.5}$ changes due to the same changes of meteorological factors, as in Fig. 3. The $PM_{2.5}$ results of other cases of the sensitivity study are shown in Figs. S4-S6 of the Supplementary Materials. The results indicate that in January, a 1.0 K increase in T leads to up to 5-6 μg m^{-3} decrease of $PM_{2.5}$ in JJJ and central China; in July, a 1.0 K increase in T causes $PM_{2.5}$ increase by about 1 μg m^{-3} in southern China but decrease by 1-3 μg m^{-3} in the JJJ and east coast region. A 10% decrease in WS causes $PM_{2.5}$ increase up to over 40 μg m^{-3} in January and up to 5 μg m^{-3} in July. A 10% relative increase in AH leads to $PM_{2.5}$ increase of up to 6 μg

m^{-3} in January and up to 2 μg m^{-3} in JJJ and northeast regions but slightly decrease of less than 1 μg m^{-3} in southern China in July. A 20% decrease of PBLH causes $PM_{2.5}$ increases by up to 20 μg m^{-3} in January and up to 4 μg m^{-3} in July. The impact of CLW and PCP on $PM_{2.5}$ is small, and generally increase in CLW increases surface $PM_{2.5}$ and increase in PCP decreases $PM_{2.5}$.

The changes in the total $PM_{2.5}$ mass concentrations are determined by the changes in the chemical components of $PM_{2.5.}$ Fig. S7 displays the fraction of $PM_{2.5}$ species (elemental carbon (EC), primary organic carbon (POC), secondary organic aerosol (SOA), sulfate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+)) in five representative cities. Secondary inorganic aerosols (SO_4^{2-}, NO_3^-, NH_4^+) are the major PM components, accounting for over 50% of $PM_{2.5}$ in January and about 40% in July. Fig. 5 and Fig. 6 show the changes of the major $PM_{2.5}$ components due to the same changes of meteorological factors as in Fig. 4 in January and in July, respectively. The results show that the effects of the meteorological parameters on the total $PM_{2.5}$ (shown in Fig. 4) are mainly due to their effects on SO_4^{2-}, NO_3^-, and NH_4^+ in January, and due to the changes in SO_4^{2-}, NO_3^-, NH_4^+, and SOA in July. In general, PBLH, WS, and PCP are negatively correlated to SO_4^{2-}, NO_3^-, and NH_4^+ formation, but AH and CLW are positively correlated to these components. SOA concentrations are much higher in July than in January due to the contribution from biogenic emissions (Hu et al., 2017a). SOA formation is affect by reaction rates (positively affected by T), availability of oxidants (such as changes in O_3), and hydrogen ion strength (affected by changes in SO_4^{2-}, NO_3^-, NH_4^+). SOA concentrations mainly increase in south

China.

It is worthwhile noting that the effects of T on SO_4^{2-} and NO_3^- (changes of NH_4^+ is determined by changes of SO_4^{2-} and NO_3^-). Both in January and July, increase in T decreases SO_4^{2-} and NO_3^- in the major areas of eastern China. The NO_3^- decreases is expected because volatile NH_4NO_3 favors more in gas phase in higher temperature, and this result is consistent with studies in other regions (Dawson et al., 2007a;Horne and Dabdub, 2017). SO_4^{2-} is found to increase with T increase in those studies because faster gas- and aqueous- phase reactions of SO_4^{2-}. However, our finding of SO_4^{2-} in China is opposite. The CMAQ-Sulfur Tracking Model (CMAQ-STM) was further used to track the SO_4^{2-} formation from different processes. The results confirm that the SO_4^{2-} production from gas- and aqueous- phase increases with T increase. But meanwhile SO_4^{2-} production from heterogeneous reactions is reduced more when T is increased. Heterogeneous SO_4^{2-} formation has been proposed as a major SO_4^{2-} formation pathway during China haze events (Wang et al., 2014b; Gen et al., 2019; Huang et al., 2019; Li et al., 2019a) and in this study it accounts for up to ~75% of total SO_4^{2-} production. The treatment of heterogeneous SO_4^{2-} formation currently is modeled as a surface-controlled uptake process, in which the formation rate is determined by the aerosol surface area and the uptake coefficient of SO_2 on particle surface (Ying et al., 2014). When T is increased, the particle surface area decreases (as particle mass concentration decreases due to a combined effect of other components), resulting in decrease in the heterogeneous SO_4^{2-} formation.

An additional simulation was run to illustrate the combined effects of perturbations in all meteorological parameters (T+1.0K, WS-10%, AH+10%, PBLH-20%, CLW+10%, and PCP+10%) on O_3 and $PM_{2.5}$ in January and July. The results are shown in Fig. S8. The average O_3-8h concentration in this combined-change simulation drop by ~2 ppb in January, except in the northeast. In July, O_3 in eastern China and the Sichuan basin rose by 2 ppb. The changes in $PM_{2.5}$ resulting from this combined-change simulation were significantly higher, compared to the basecase concentrations, even increased by up to 50 μg m^{-3} in January.

3.3. Quantitative sensitivity of O_3 and $PM_{2.5}$ to individual meteorology parameters

The quantitative sensitivity of O_3 and $PM_{2.5}$ concentrations to individual meteorological parameter is calculated by linear fitting of the changes in monthly-average concentrations under all of the six perturbed cases of the meteorological parameter. Figs. S8-S10 in Supplementary Materials show the calculation examples of T, WS, and AH on O_3 at the five major cities of Beijing, Chongqing, Guangzhou, Shanghai and Xi'an, and Figs. S11-S13 shows the examples for the $PM_{2.5}$ cases. Fig.7 demonstrates the sensitivities of O_3-8h and $PM_{2.5}$ and its components to each meteorological parameter in the five cities. In January, T has a positive impact on O_3 in all cities, and the largest impact is in Chongqing with a rate of +1.69 ppb K^{-1}. In July, O_3 also shows a strong positive sensitivity to T in Beijing with +1.06 ppb K^{-1} and in Shanghai with +0.98 ppb K^{-1}, but has a small negative sensitivity (-0.15 ppb K^{-1}) in Xi'an and a moderate negative sensitivity (-0.74 ppb K^{-1})

in Guangzhou. The O_3 sensitivity to T in Guangzhou in July shows a highly nonlinear trend and is very different from other cities (Fig. S8(c)). More studies are needed to investigate the effects of T on O_3 pollution in the YRD region during summertime. WS and PBLH both have positive effects on O_3-8h in January, the effects vary significantly among cities, with 0.004-0.3 ppb $\%^{-1}$ for WS and 0.04-0.14 ppb $\%^{-1}$ for PBLH. AH has a negative effect on O_3-8h in January, ranging from -0.01 to -0.15 ppb $\%^{-1}$. But in July, the impacts of WS, AH, and PBLH are negative in most cities, with a range of -0.05 to -0.18, -0.05 to -0.13, and -0.02 to -0.07 ppb $\%^{-1}$, respectively. Generally speaking, T, WS, AH and PBLH led to rather lager O_3 changes~~the sensitivity of O_3 to T is obviously higher than that of WS, AH, and PBLH~~. The sensitivity of O_3 to CLW and PCP is even minimal (less than 0.~~0~~1 ppb $\%^{-1}$) and mostly negative.

Negative sensitivities are found for surface $PM_{2.5}$ concentrations to T, WS, PBLH, and PCP, and positive sensitivities for $PM_{2.5}$ to AH and CLW. The sensitivity of T in the five cities ranges from -1.5 to -3.6 μg m^{-3} K^{-1} in January and -0.3 to -1.65 μg m^{-3} K^{-1} in July. $PM_{2.5}$ is also very sensitive to WS in January, with a range of -0.8 to -2.97 μg m^{-3} $\%^{-1}$, while the sensitivity (-0.03 to -0.19 μg m^{-3} $\%^{-1}$) becomes much smaller in July. The sensitivity to PBLH is -0.12 to -0.58 μg m^{-3} $\%^{-1}$ in January and -0.003 to -0.23 μg m^{-3} $\%^{-1}$ in July. The sensitivity to AH is 0.16 to 0.30 μg m^{-3} $\%^{-1}$ in January and 0.05 to 0.27 μg m^{-3} $\%^{-1}$ in July. Sensitivity to CLW and PCP is small in January and July, mostly less than 0.01μg m^{-3} $\%^{-1}$. The $PM_{2.5}$ sensitivities can be explained by the major components of SO_4^{2-}, NO_3^-, and NH_4^+ in January and by SO_4^{2-}, NO_3^-, NH_4^+, and SOA in July.

Fig. 8 shows the spatial variations of the sensitivity of O_3-8h and $PM_{2.5}$ to the meteorological parameters. The sensitivity of O_3-8h to temperature is more significant in Sichuan and southern provinces of China in January, and in NCP and YRD in July, up to +2 ppb K^{-1} in both January and July. O_3-8h sensitivity to WS is diverse in space, and is generally positive in Sichuan and southern provinces in January; and it is negative in east China but positive in west China. O_3-8h sensitivity to AH is generally negative in both months in most regions of China, except the northeast in January and southwest in July. O_3-8h sensitivity to PBLH is mostly positive in January but becomes negative in YRD, CYB, NCP, and central China in July. O_3-8h sensitivity to CLW and PCP is negligible.

Fig. 9 displays the spatial variations of the sensitivity of surface $PM_{2.5}$ to the meteorological parameters. $PM_{2.5}$ sensitivities to the meteorological parameters are more consistent in January and July than the cases of O_3, i.e., negative sensitivity to T, WS, PBLH, PCP, and positive to AH and CLW in most regions of China in both months. On the other hand, $PM_{2.5}$ sensitivities are more profound in January than in July. $PM_{2.5}$ sensitivity to T is up to -5 μg m^{-3} K^{-1} in January and up to -2 μg m^{-3} K^{-1} in July. $PM_{2.5}$ sensitivity to WS is up to -3 μg m^{-3} $\%^{-1}$ in January, and up to -0.4 μg m^{-3} $\%^{-1}$ in July. $PM_{2.5}$ sensitivity to PBLH is up to -1 μg m^{-3} $\%^{-1}$ in January, and up to -0.14 μg m^{-3} $\%^{-1}$ in July. $PM_{2.5}$ sensitivity to AH is up to +0.6 μg m^{-3} $\%^{-1}$ in January, and up to 0.3 μg m^{-3} $\%^{-1}$ in July. The sensitivities to CLW and PCP is small, compared to the other four meteorological parameters. $PM_{2.5}$ sensitivity to T is negative in most land areas of China in January and in NCP and YRD in July because of the negative effects of T on SO_4^{2-}, NO_3^-, and NH_4^+, as discussed in previous sections. $PM_{2.5}$ sensitivity to T is positive in south China in July due to more SOA with higher T.

4. Conclusions

Meteorological conditions can have a great influence on surface O_3 and $PM_{2.5}$ concentrations. In this study, the sensitivities of O_3-8h and $PM_{2.5}$ to T, WS, AH, PBLH, PCP, and CLW are quantitatively estimated in January and July, respectively in China. The response of O_3-8h to T is important ~~is most sensitive to T~~ and the sensitivity can be up to +2 ppb K^{-1} in both January and July, and the sensitivity is dependent on the O_3 chemistry formation or loss regime, i.e., positive in the net O_3 formation areas, and negative in the O_3 consumption areas. In general, $PM_{2.5}$ sensitivities are negative to T, WS, PBLH, and PCP and positive to AH and CLW in most regions of China in both January and July. The sensitivities in January are much larger than in July. $PM_{2.5}$ sensitivities to T, WS, AH, and PBLH are important. The $PM_{2.5}$ sensitivities to these meteorological parameters are through major effects on SO_4^{2-}, NO_3^-, NH_4^+, and SOA. The sensitivities of O_3 and $PM_{2.5}$ to CLW and PCP are negligible. The results show that O_3 and $PM_{2.5}$ concentrations in China are greatly affected by meteorological conditions, therefore changes in these meteorological parameters due to climate change or inter-annual meteorological variations could potentially alter O_3 and $PM_{2.5}$ concentrations significantly, and it should consider these effects when developing emission control strategies. The results also show that the O_3 and $PM_{2.5}$ sensitivities to meteorological parameters have substantial spatial variations. Future studies can further investigate how the changes in meteorological conditions affect the effectiveness of emission control plans in reaching the designed air quality objectives in the different regions of China.

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Tables and Figures

Table 1. Meteorological perturbations imposed in this study

Meteorological Parameter	Changes in Values Examined
Temperature (T)	±0.5K，±1.0K，±1.5K
Wind speed (WS)	±5%，±10%，±20%
Absolute Humidity (AH)	±5%，±10%，±20%
Boundary layer height (PBLH)	±10%，±20%，±30%
Cloud liquid content (CLW)	±5%，±10%，±20%
Precipitation (PCP)	±5%，±10%，±20%

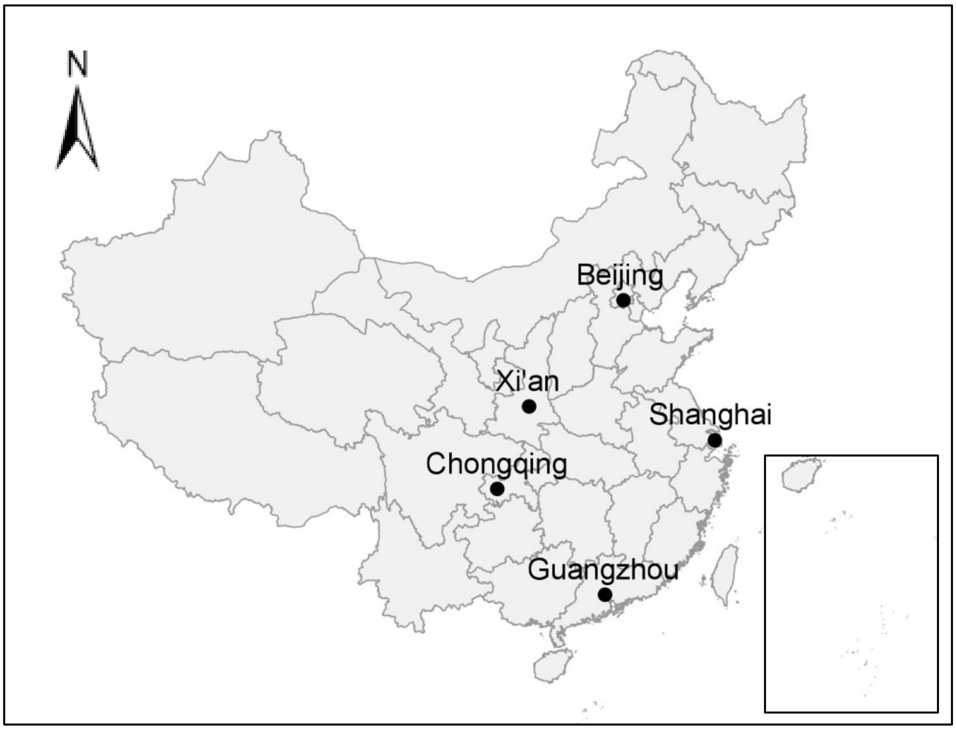

Fig.1 Location map of China and the five cities.

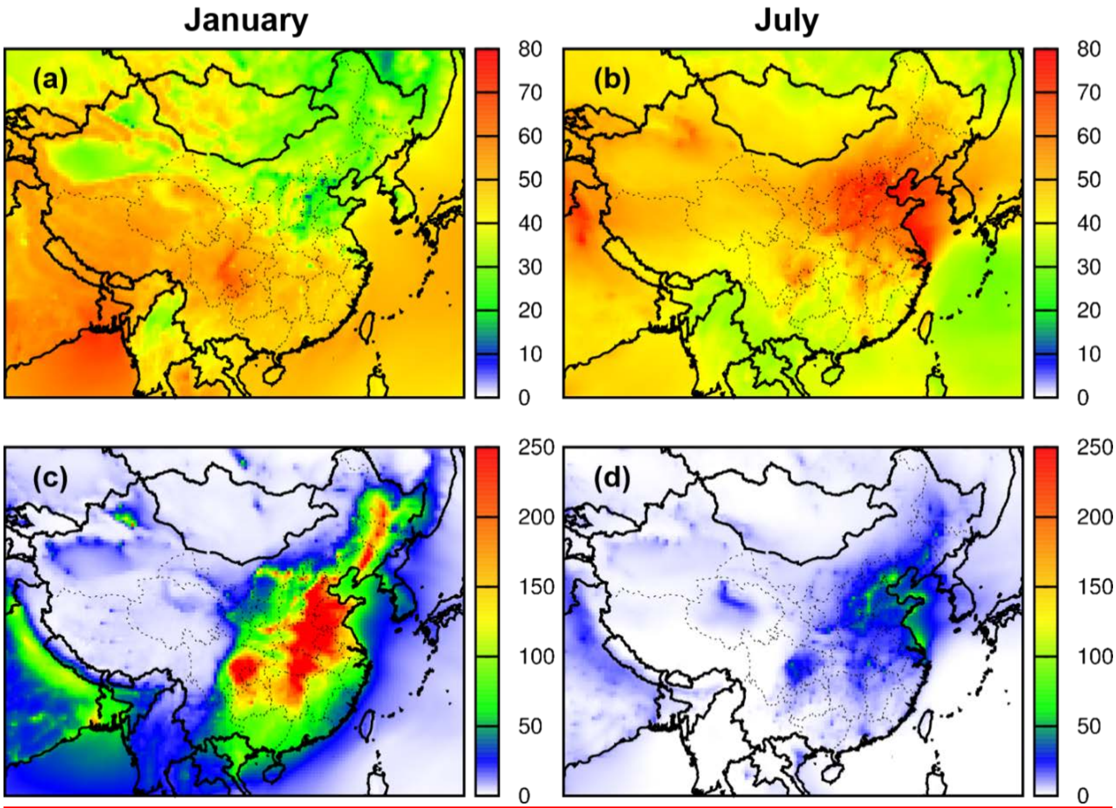

Fig.2 Spatial distributions of monthly average O_3-8 h (ppb) in (a) January and (b) July, and monthly average $PM_{2.5}$ (μg m^{-3}) in (c) January and (d) July 2013.

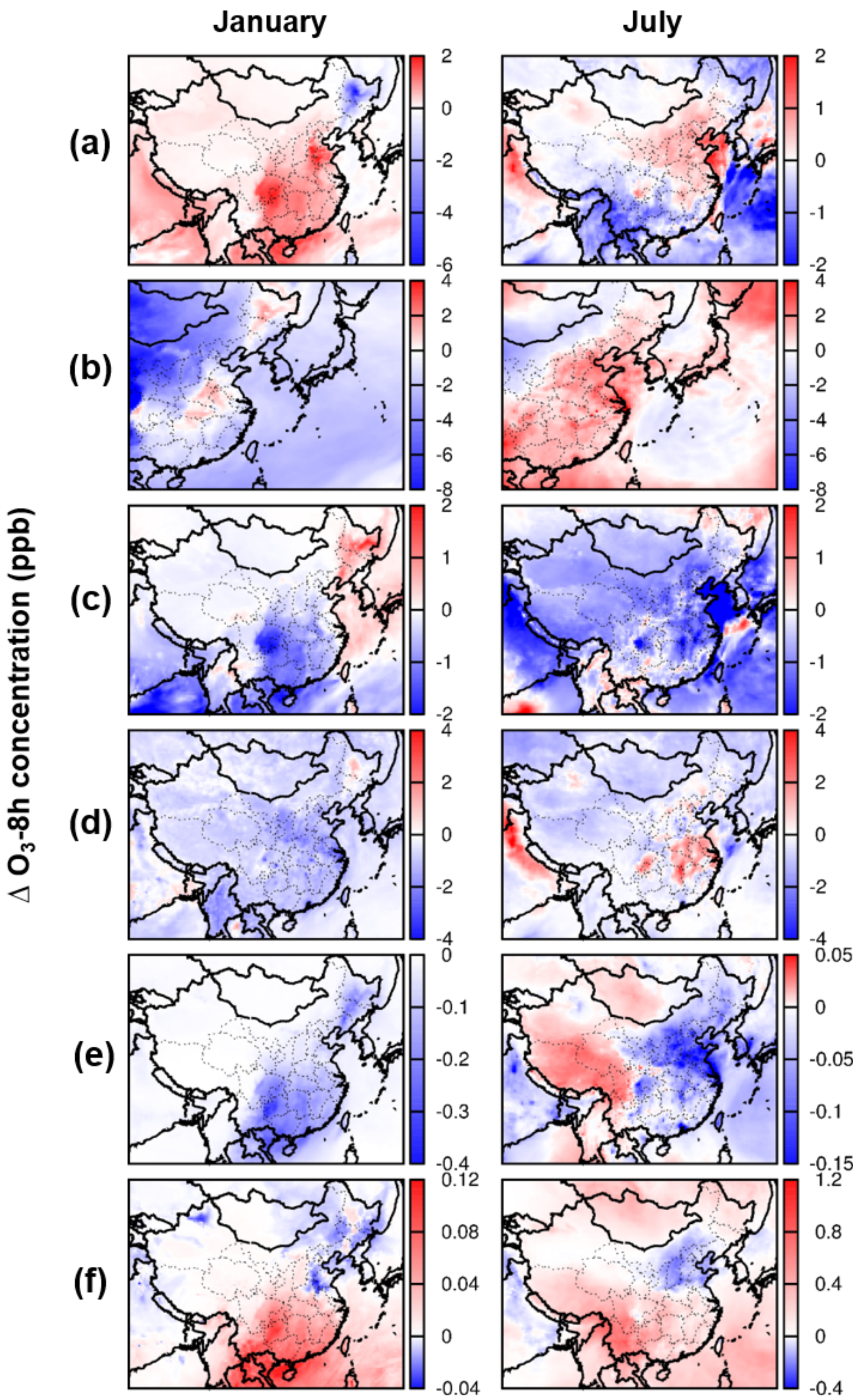

Fig.3 Changes in monthly average O_3-8h (ppb) in January and July, 2013 due to (a) T+1.0K, (b) WS-10%, (c) AH+10%, (d) PBLH-20%, (e) CLW+10%, and (f) PCP+10%.

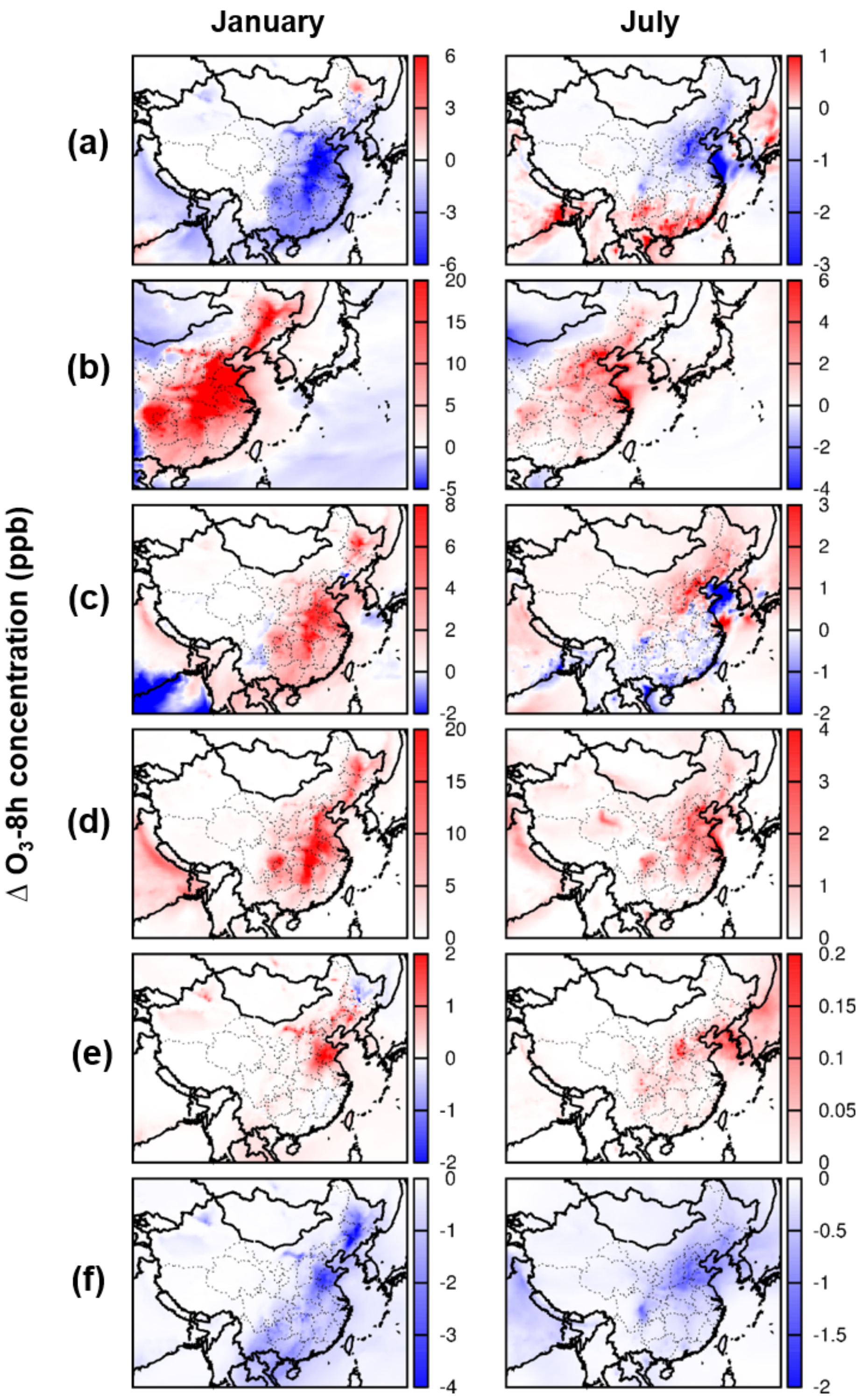

Fig.4 Changes in monthly average $PM_{2.5}$ concentration (μg m^{-3}) in January and July, 2013 due to (a) T+1.0K, (b) WS-10%, (c) AH+10%, (d) PBLH-20%, (e) CLW+10%, and (f) PCP+10%.

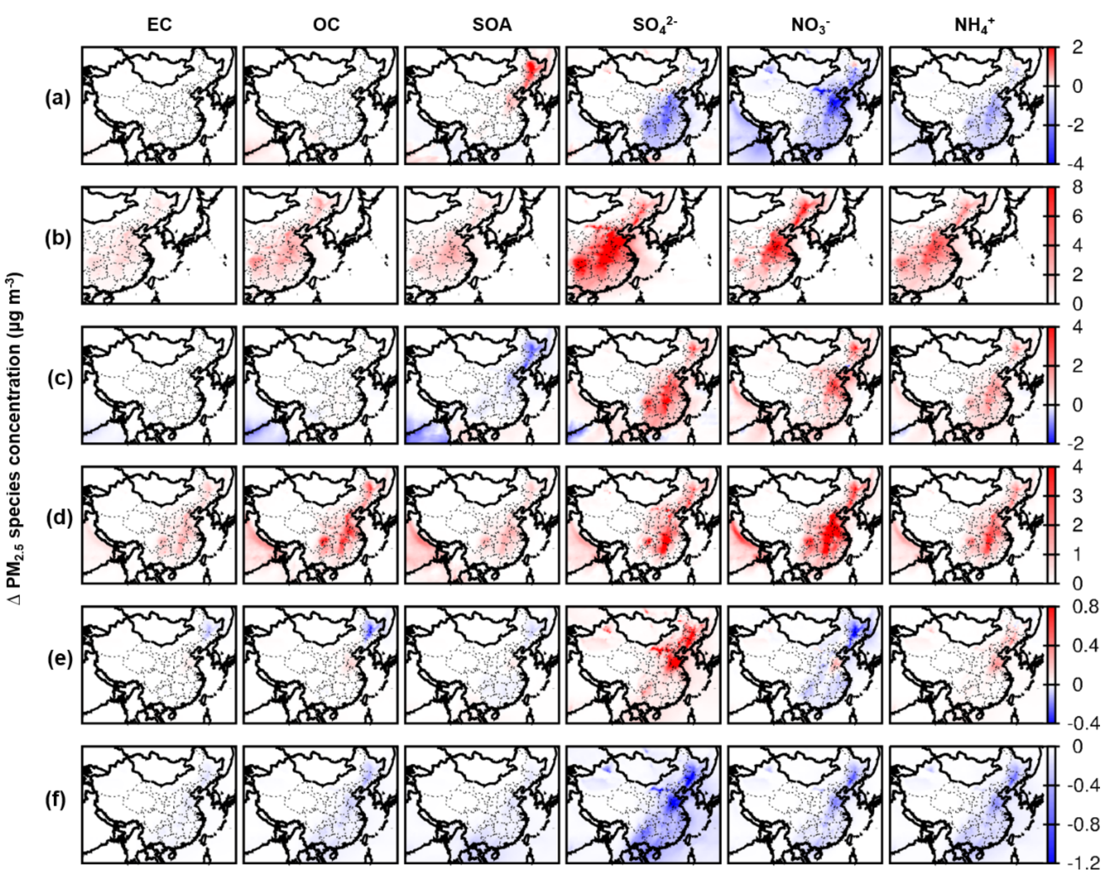

Fig.5 Changes in monthly average $PM_{2.5}$ component concentration (μg m^{-3}) in January due to (a) T+1.0K, (b) WS-10%, (c) AH+10%, (d) PBLH-20%, (e) CLW+10%, and (f) PCP+10%.

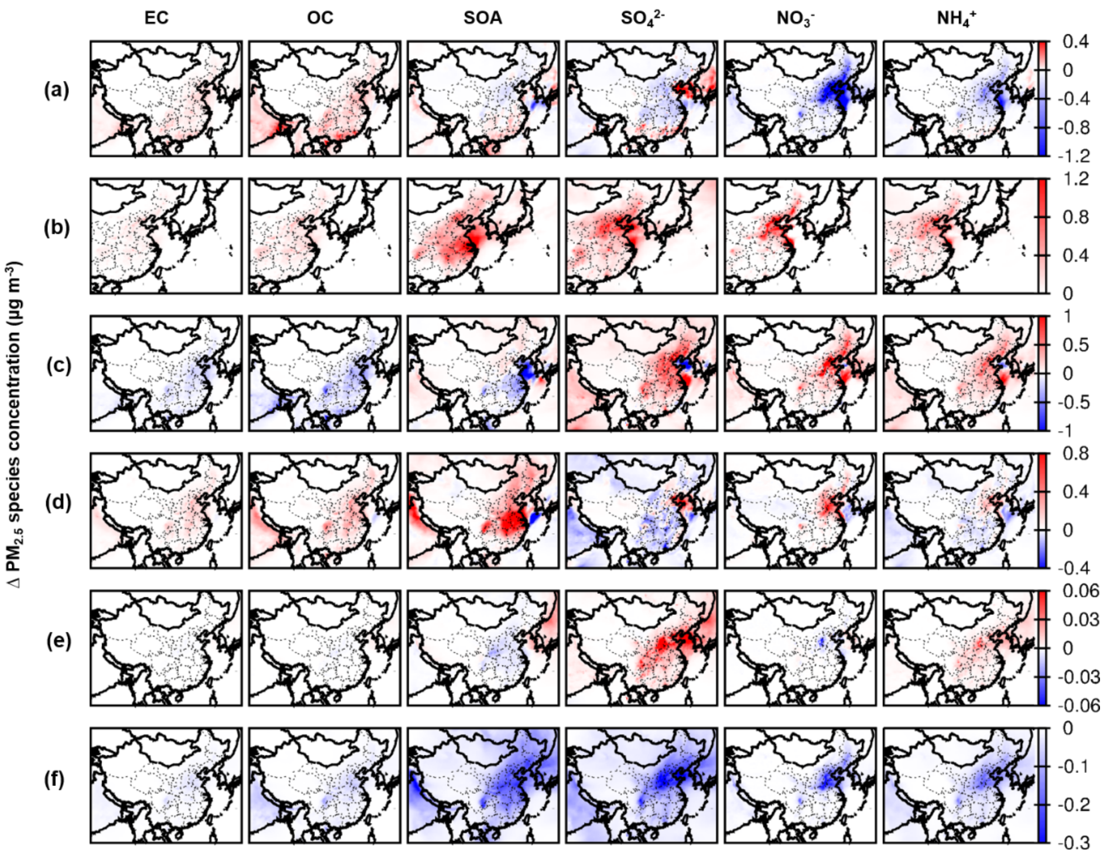

Fig.6 Changes in monthly average $PM_{2.5}$ component concentration (μg m^{-3}) in July due to (a) T+1.0K, (b) WS-10%, (c) AH+10%, (d) PBLH-20%, (e) CLW+10%, and (f) PCP+10%.

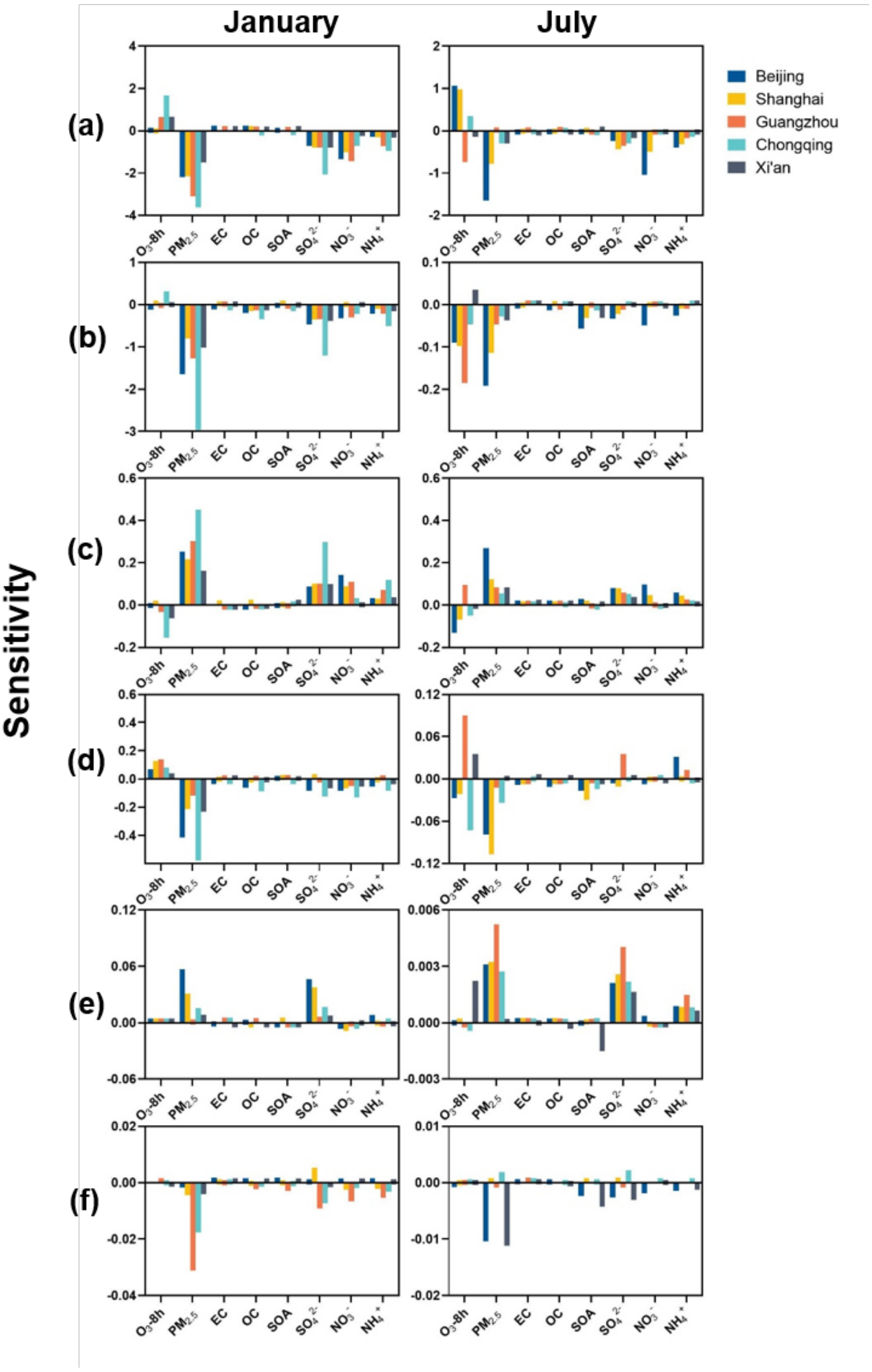

Fig.7 Sensitivity of O_3-8h, $PM_{2.5}$ and its components to meteorological parameter of (a) T, (b) WS, (c) AH, (d) PBLH, (e) CLW, and (f) PCP in five cities in China. The unit of sensitivity is ppb K^{-1} for O_3-8h to T, and is ppb $\%^{-1}$ for O_3-8h to other meteorological parameters; and the unit is μg m^{-3} K^{-1} for $PM_{2.5}$ and its components to T, and is μg m^{-3} $\%^{-1}$ for $PM_{2.5}$ and its components to other meteorological parameters.

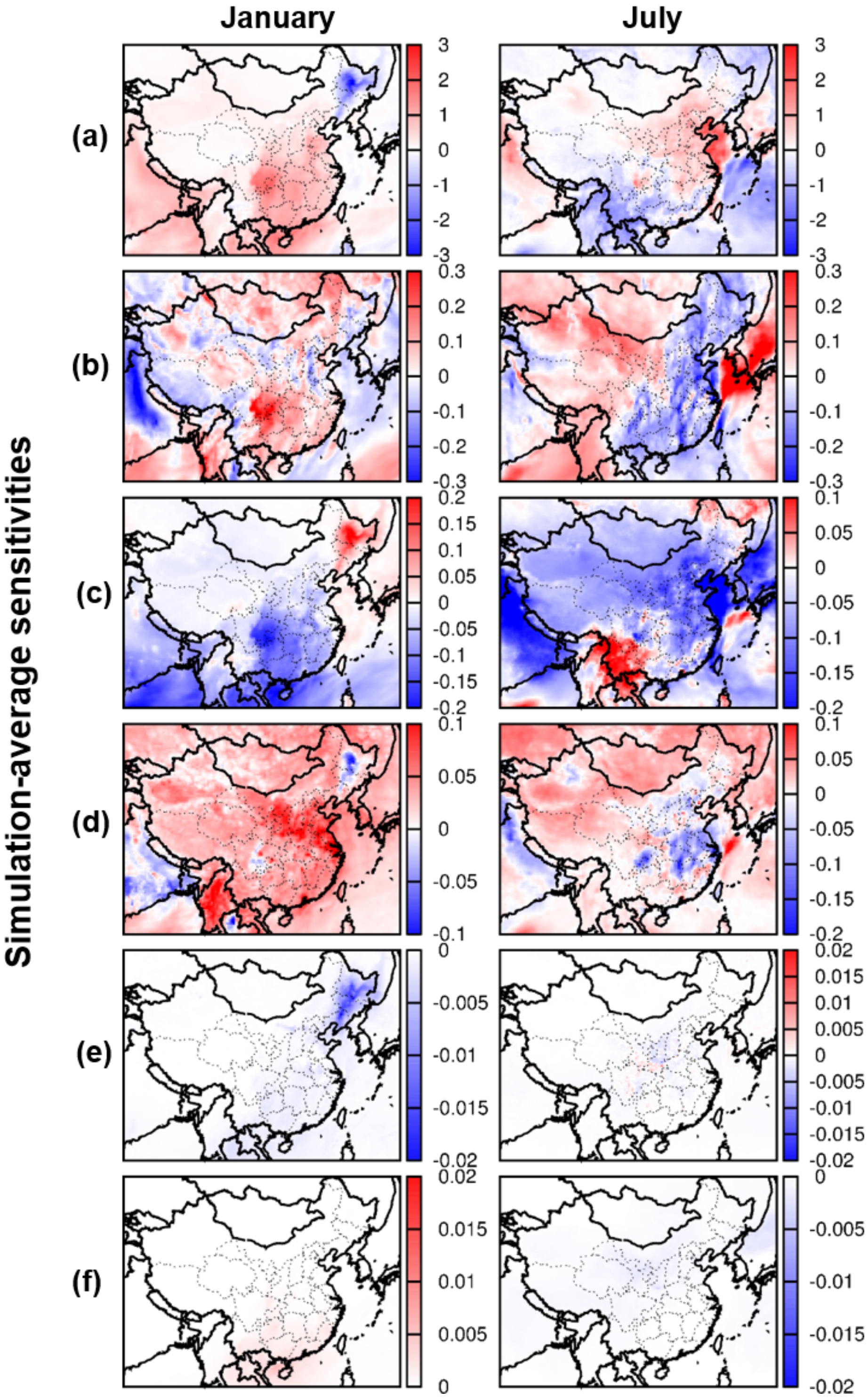

Fig.8 Sensitivity of O_3-8h mean to meteorological perturbations (a) T, (b) WS, (c) AH, (d) PBLH, (e) CLW, (f) PCP in China. The value in T is measured in ppb K^{-1}, and others is ppb $\%^{-1}$.

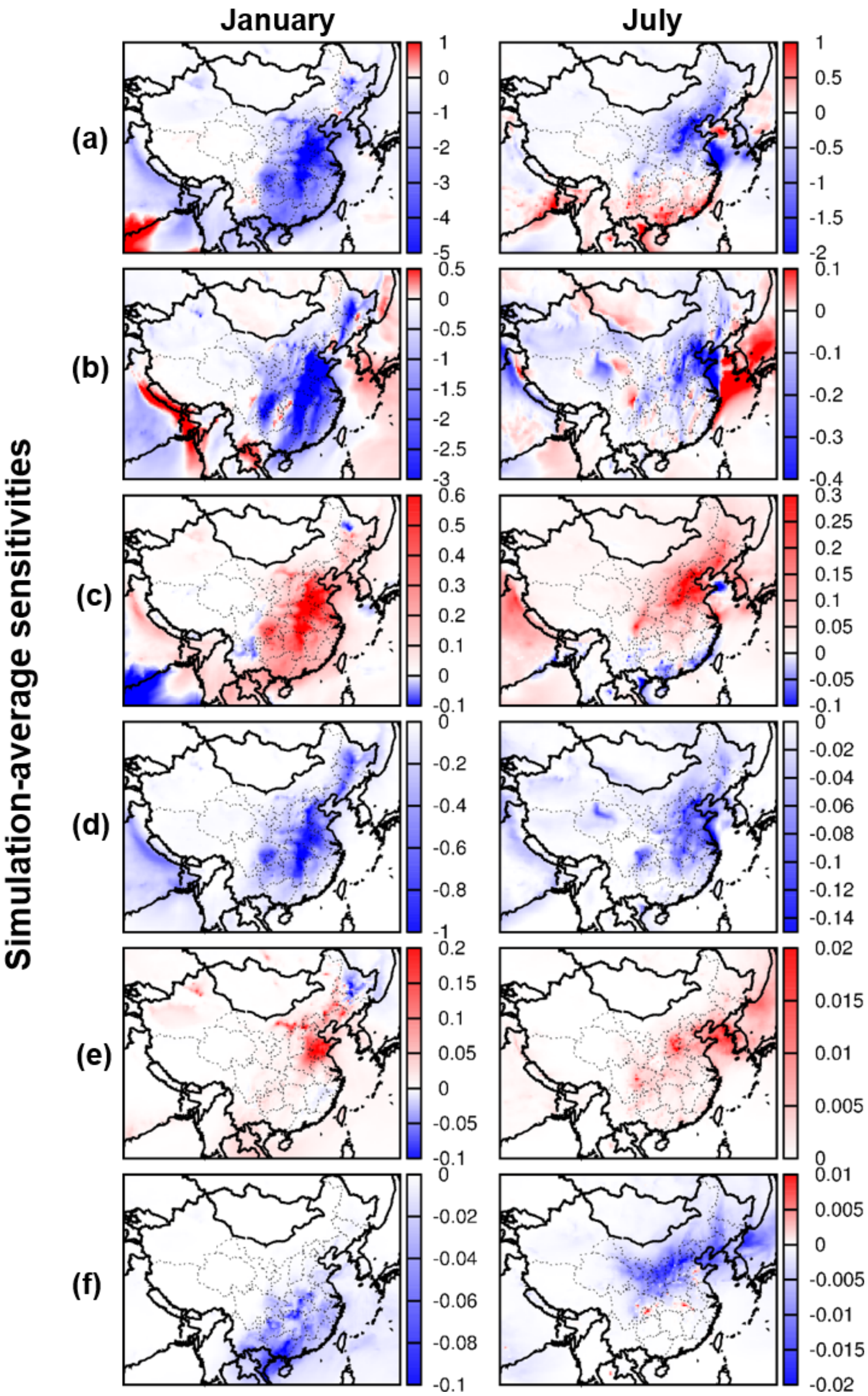

Fig.9 Sensitivity of $PM_{2.5}$ mean to meteorological perturbations (a) T, (b) WS, (c) AH, (d) PBLH, (e) CLW, (f) PCP in China. The value in T is measured in μg m^{-3} K^{-1}, and others is μg m^{-3} $\%^{-1}$.