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# Large scale control of surface ozone by relative humidity observed during warm seasons in China

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## Abstract

Rising air pollution by surface ozone ( $O_3$ ) in China has induced extensive efforts to control ozone generation in major urban and industrial areas, yet mechanisms ruling the ozone production and loss are not well understood. In particular, ozone levels are strongly influenced by meteorological factors such as relative humidity, but this has been explored only in local situations, and the effect of relative humidity on ozone levels in warm seasons on a large scale in China is still unknown. Here we studied surface ozone, relative humidity, temperature, and other meteorological variables in 74 major cities in China during 2017–2018, focusing on the warm seasons in seven regions. Results show that ozone levels decrease with increasing relative humidity in all cities, with an average correlation coefficient of  $-0.58$ , ranging from  $-0.17$  in Zhangjiakou to  $-0.84$  in Hengshui. At high relative humidity levels, above 75%, average ozone levels ranged from 44.6 to 122.5  $\mu\text{g m}^{-3}$ , which is lower than Chinese quality threshold of hourly average ozone level of 200  $\mu\text{g m}^{-3}$ . The decreases of ozone with relative humidity were more pronounced at high temperature, above 30 °C, than below 25 °C. The increases of ozone with temperature were more pronounced at low relative humidity, below 40%. Overall, our findings reveal that mechanisms ruling surface ozone levels are similar on a large scale. This is promising to design common methods of climate engineering to protect human health.

**Keywords** Ozone · Relative humidity · Temperature · Warm season · China

## Introduction

Tropospheric ozone ( $O_3$ ) is a major oxidant and greenhouse gas related partly to pollution. Ozone has negative impacts on human health, regional climates, agricultural yields, and forest productivity (Yu et al. 2006; Shao et al. 2006; Booker et al. 2009; Monks et al. 2015). Anthropogenic ozone is produced by photochemical reactions of nitrogen oxides ( $\text{NO}_x$ ) and volatile organic compounds (VOCs) in the presence of sunlight (Seinfeld and Pandis 2016). High near-surface  $O_3$  levels occur in many urban and industrial areas across China. The establishment of an atmospheric monitoring network since 2000 in China has allowed regular ozone monitoring. Hourly monitoring data in 74 major cities disclosed an increase of daily average maximum 8-h  $O_3$  concentrations from about 69.5 ppbv in 2013 to 75 ppbv in 2015 (<http://www.mep.gov.cn>). Moreover, a 4.2% increase of mean  $O_3$  concentrations in 336 prefecture-level cities was observed from 2015 to 2016 (Li et al. 2019). Recently, maximum 8-h  $O_3$  concentrations

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exceeding the China National Ambient Air Quality Standard of  $160 \mu\text{g}/\text{m}^3$  (CNAAQ, GB3095-2012) have been observed in many regions in China, notably in Beijing-Tianjin-Hebei, Yangtze River Delta, and Pearl River delta (Wang et al. 2017).

Extensive efforts have been made to control surface ozone by reducing ozone precursors and controlling meteorological conditions (Ou et al. 2016; Wang et al. 2017; Chen et al. 2019a; Yang et al. 2019). Parameters ruling ozone levels include emissions, solar radiation, precipitation, temperature (T), relative humidity (RH), wind speed and direction, and cloud cover (Camalier et al. 2007; Davis et al. 2011; Reddy and Pfister 2016; Wang et al. 2017; Chen et al. 2019b; Li et al. 2019). Yu (2019) proposed a fog geoengineering method to increase atmospheric moisture and, in turn, abate near-surface  $\text{O}_3$  levels. In this study, we focused on the effects of relative humidity on surface  $\text{O}_3$ .

Relationships between  $\text{O}_3$  and relative humidity have been investigated in modeling studies. For instance, maximum daily temperature and average relative humidity had the largest effects in 74 cities in the US according to a city-specific generalized linear model (Davis et al. 2011). The role of water vapor in the reduction of surface  $\text{O}_3$  levels has also been demonstrated by simulations with Climate-chemistry models (CCMs) (Thompson et al. 1989; Johnson et al. 2001; Murazaki and Hess 2006; Doherty et al. 2013). A 19% increase of water vapor could lead to approximately 4–5% decrease of  $\text{O}_3$  concentrations (Doherty et al. 2013). Similarly, the increasing relative humidity from 9 to 87% can reduce  $\text{O}_3$  concentrations by 30% (Wang et al. 2016). The decrease of ozone with relative humidity has been explained by the following processes (Table S1):

- $\text{O}_3$  photolysis produces  $\text{O}(^1\text{D})$ .  $\text{O}(^1\text{D})$  then reacts with a water molecule ( $\text{H}_2\text{O}$ ) to generate two OH radicals, which favor ozone decomposition.
- Relative humidity favors heterogeneous reactions of  $\text{O}_3$  with particles in aerosols (He et al. 2017a; b). For instance, humidity enhance ozone uptake by increasing aerosol size and decreasing aerosol viscosity (Han et al. 2019).
- Ozone decomposition is enhanced by heterogeneous bonding on aqueous aerosol surfaces (Kotelnikov and Stepanov 2019).
- Wet aerosols formed under high humidity reduce ultraviolet actinic flux and inhibit photochemical reaction rates (Deng et al. 2010; Liu et al. 2011).
- Elevated relative humidity decreases temperature by water evaporation (Olszyna et al. 1997).
- Relative humidity inhibits the chain length of hydroxyl radicals (OH and  $\text{HO}_2$ ) and  $\text{NO}_2$ , thus limiting  $\text{O}_3$  generation (Reichert et al. 2003; Wang et al. 2016).

- High relative humidity promotes the reaction of gaseous  $\text{N}_2\text{O}_5$  with aqueous NaCl particles, thus greatly reducing  $\text{O}_3$  generation (Leu et al. 1995; Jia and Xu 2015).

Decreases of ozone with relative humidity have been reported in some regions and are commonly observed in summer seasons (Olszyna et al. 1997; Dueas et al. 2002; Elminir 2005; Camalier et al. 2007; Tu et al. 2007). A study in a coastal Spanish site showed that, together with temperature and wind speed, relative humidity was one of three dominant meteorological factors influencing ozone levels (Dueas et al. 2002). Pearson correlation and stepwise multiple regression analysis also showed negative correlations between ozone-RH (Manju et al. 2018). In Nanjing, a report shows a maximum  $\text{O}_3$  level of 43.4 ppbv below 40% relative humidity, versus the lowest value of 10.9 ppbv above 80% RH (Tu et al. 2007). Relative humidity was also found as the strongest influencing factor in central and southern parts of eastern China (Han et al. 2020). Relative humidity appeared more important than radiation and temperature to assess the adverse effects of ozone on vegetation (Gong et al. 2021). Overall, previous investigations focused on local situations or several cities during the same time period. Therefore, here we present a study of ozone, relative humidity and temperature in different warm seasons at a large geographical scale in the major 74 cities in China.

## Experimental

Monitoring data of  $\text{O}_3$  and its precursors ( $\text{NO}_2$ , CO) were collected from the website of the China National Environmental Monitoring Center (CNEMC, <http://www.mee.gov.cn/hjzl/dqbj/>). We focused on hourly  $\text{O}_3$  concentrations measured at all monitoring sites in 74 major cities in China during 2017–2018. The monitoring data in each city consist of urban and background sites. The average  $\text{O}_3$  concentrations at all sites in each separate city were used and regarded as the city-level  $\text{O}_3$  values. Meteorological variable data on an hourly basis including temperature (T,  $^\circ\text{C}$ ), relative humidity (RH, %), precipitation (Prec, mm), and wind speed (WS, m/s) were provided by the China Meteorological Administration (<http://data.cma.cn/site/index.html>). Only hours with both valid  $\text{O}_3$  and RH values were used for analysis. The data of leaf area indices (LAI) in the summer seasons and topography height (HGT\_M) in China were derived from MODIS products (<https://modis.gsfc.nasa.gov/data/>).

Under the comprehensive consideration of the spatial patterns of  $\text{O}_3$  and national administrative divisions in China, the following 74 cities were geographically grouped into seven representative divisions, namely Northeast China (Dalian, Haerbin, Shenyang, Changchun), North China (Baoding, Beijing, Cangzhou, Chengde, Handan, Hengshui,

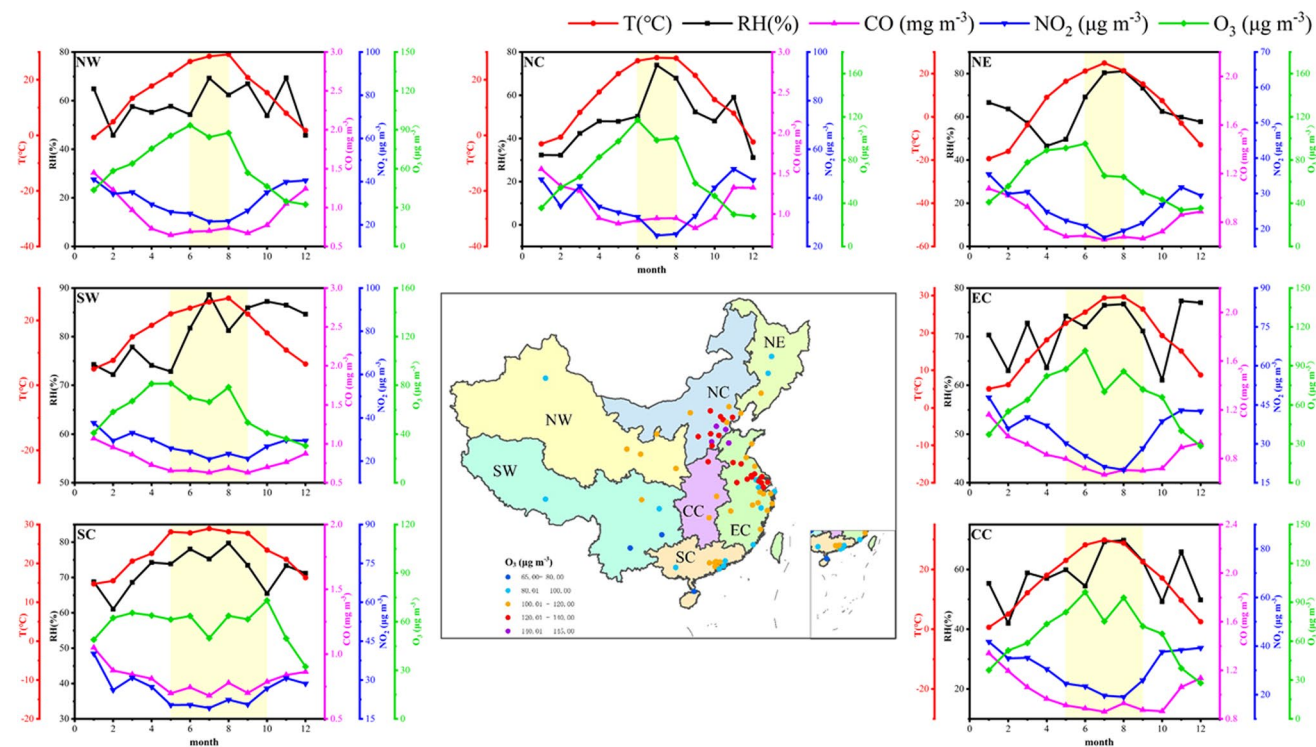
Huhehaote, Langfang, Qinhuangdao, Shijiazhuang, Taiyuan, Tangshan, Tianjin, Xingtai, Zhangjiakou), Central China (Wuhan, Changsha, Zhengzhou), East China (Changzhou, Fuzhou, Hangzhou, Hefei, Huzhou, Huai'an, Jinan, Jiaxing, Jinhua, Lishui, Lianyungang, Nanchang, Nanjing, Nantong, Ningbo, Qingdao, Quzhou, Xiamen, Shanghai, Shaoxing, Suzhou, Taizhou, Taizhouj, Wenzhou, Wuxi, Suqian, Xuzhou, Yancheng, Yangzhou, Zhenjiang, Zhoushan), South China (Dongguan, Foshan, Guangzhou, Haikou, Huizhou, Jiangmen, Nanning, Shenzhen, Zhaoqing, Zhongshan, Zhuhai), Southwest China (Chengdu, Guiyang, Kunming, Chongqing, Lasa), and Northwest China (Lanzhou, Wulumuqi, Xian, Xining, Yinchuan), as seen in Fig. 1 and Table S2. Then, concentrations and meteorological data averaged across all cities located in each division were used. While conducting correlation analyses between  $O_3$  and RH, we chose different study periods for the seven regions due to various warm seasons for each region, as based on monthly variations of temperature in Fig. 1. The national warm season typically spans April–September. But obviously the northern regions have lower temperatures than southern regions in May and September. So summer season

(June–August) is regarded as the warm season in northern areas in China. Since  $O_3$  concentrations in South China typically peak in October (Lu et al. 2018), also shown in Fig. 1, the study period of cities in South China was expanded to October. Therefore, the final chosen study periods were June to August for Northeast China (NE), North China (NC), and Northwest China (NW), May to September for East China (EC), Central China (CC), and Southwest China (SW), May to October for South China (SC). Due to geographical proximity of Fuzhou and Xiamen cities to South China, time periods of these two cities were kept in correspondence with the cities in South China.

## Results and discussion

### Seasonal variations of $O_3$ , $NO_2$ and CO

Average levels of ozone ( $O_3$ ), nitrogen dioxide ( $NO_2$ ), carbon monoxide (CO), relative humidity (RH), and temperature (T) in various Chinese regions in 2018 are shown in Fig. 1, with warm seasons shadowed in orange. Results show



**Fig. 1** Monthly variations of average concentrations of ozone ( $O_3$ ), nitrogen dioxide ( $NO_2$ ), and carbon monoxide (CO), and values of relative humidity (RH), and temperature (T) for Chinese areas in 2018. The orange-shaded areas denote chosen warm seasons: June to August for Northeast China (NE), North China (NC), and Northwest China (NW), May to September for East China (EC), Central China (CC), and Southwest China (SW), May to October for South China

(SC). Results show that six divisions displayed similar variation patterns of monthly average  $O_3$  concentrations peaking in May or June, whereas South China reached seasonal peak in October when temperature was not maximum but RH was relatively low. Coincident high  $O_3$  and low RH were generally observed in warm seasons for all the regions

that annual mean O<sub>3</sub> concentrations are heterogeneous in 74 cities, with values ranging from 65.8 μg m<sup>-3</sup> in Haikou, Hainan province, to 143.8 μg m<sup>-3</sup> in Cangzhou, Hebei province. The highest O<sub>3</sub> levels were mainly observed in North and East China and have been explained by greater emissions of ozone precursors in industrialized and urbanized regions. Indeed, in North China, power plants, coal combustion industries, and biomass burning sources produce more NO<sub>x</sub> and VOCs emissions (Zhao et al. 2012; Chai et al. 2014). Higher NO<sub>2</sub> levels are also observed in cities having more vehicle emission sources (Wang et al. 2010a, b).

NO<sub>2</sub> and CO levels in all Chinese regions display similar variations, with maximum values in winter and late autumn, and minimum values in summer, late spring, and early autumn (Fig. 1). The low concentrations of these ozone precursors in warmer seasons can be attributed to active photochemistry and stronger vertical atmospheric mixing. Whereas the high levels in colder seasons may be a result of larger consumption of heating fuels, as well as weak solar radiation, low wind speed and mixing layer height, enhancing NO<sub>2</sub> and CO concentrations (Wang et al. 2010a; Tai et al. 2010).

Considerable seasonal variations of O<sub>3</sub> are observed in all Chinese regions, with high levels in summer, late spring, and early autumn, and low levels in winter (Fig. 1). Maximum ozone concentrations range from 73.1 μg m<sup>-3</sup> in South China to 117.2 μg m<sup>-3</sup> in North China. In general, monthly variations of O<sub>3</sub> can be explained by prevailing meteorological conditions and anthropogenic activities (Reddy and Pfister 2016). Peak values of ground-level O<sub>3</sub> tend to appear in the warm seasons with intense solar radiation, high temperature, stagnant high-pressure conditions, as observed previously (Ou et al. 2015; He et al. 2017b).

Nevertheless, we also observed inconsistent fluctuations in variation patterns. For example, monthly average O<sub>3</sub> concentrations displayed similar variations in six regions, peaking in May or June. Whereas South China displayed a seasonal ozone maximum in October when temperature was not maximum. This is likely due to the sharp decline of relative humidity from about 80% in August to 65% in October in South China. Similar findings have been shown in previous studies conducted in the Pearl River delta, with October being the most polluted month, different from other regions (Bell et al. 2005; Zanutti and Schwartz 2008). This phenomenon has also been attributed to the adverse effect of the Asian summer monsoon on O<sub>3</sub>, with O<sub>3</sub> peaking before and after monsoon in the Yangtze River and Pearl River deltas (Lu et al. 2018; Han et al. 2020). Although higher temperature and longer sunshine duration in summer favors O<sub>3</sub> formation in South China, larger rainfall amounts in summer versus autumn can retard O<sub>3</sub> formation (Shao et al. 2009).

Overall, our findings show that high ground-level O<sub>3</sub> levels are generally accompanied by high temperature and

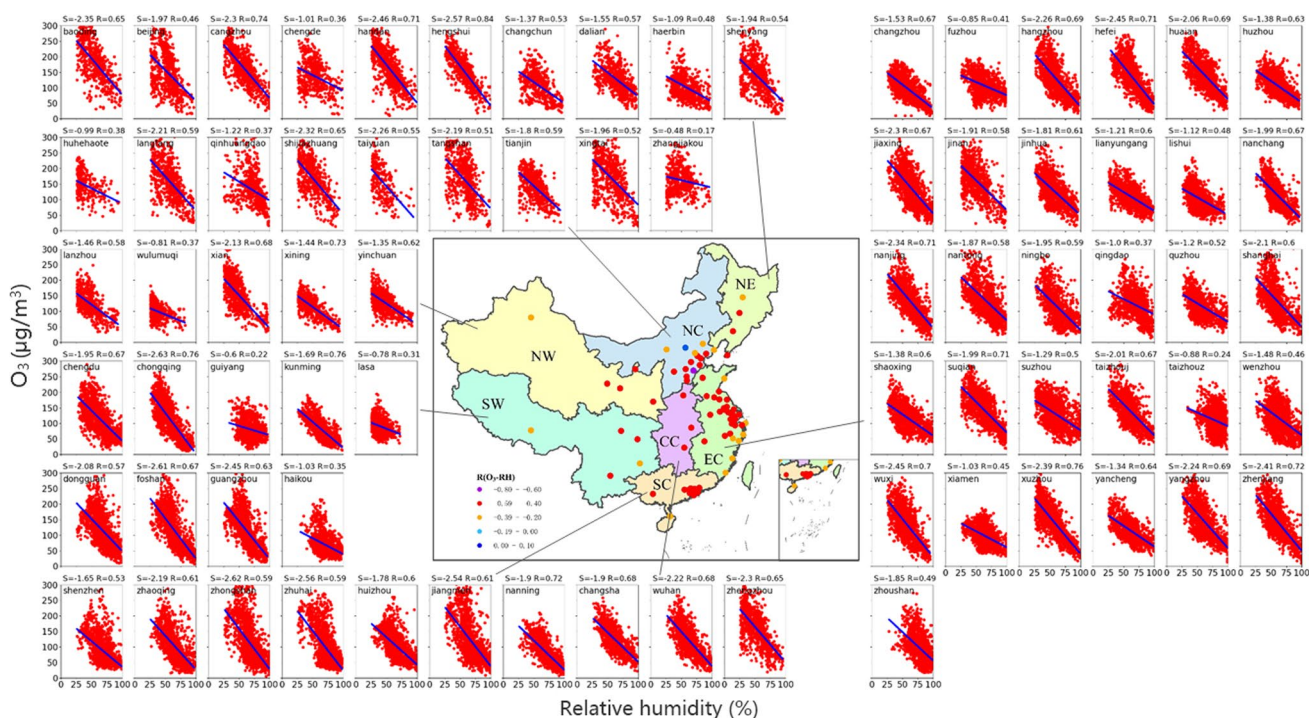
low relative humidity in all regions, whereas high NO<sub>2</sub> and CO concentrations appear under low temperature and high relative humidity. Since ozone peaked in warm seasons, we focused further research on warm seasons highlighted in orange in Fig. 1 to investigate relationships between ozone and relative humidity.

### Correlation between ozone and relative humidity

Figure 2 displays correlations of O<sub>3</sub> versus relative humidity (RH) during the noontimes, 11:00–16:00 local time, during warm seasons of 2017 and 2018 in the major 74 cities in China. We excluded data of rainfall time to avoid the influence of precipitation scavenging. We selected noontimes when O<sub>3</sub> formation is higher due to stronger solar radiation, according to Monks et al. (2015). Moreover, data with low relative humidity, below 25% and temperature, below 15 °C, were excluded owing to little effect of low humidity and temperature on ozone. Correlation coefficient (R) and slope of linear regression equation (S) were used to represent the relationships. Unlike prior studies choosing the same time periods for all cities, here we focused on dates of warm seasons, which are different in the seven regions.

Results show an overall decrease of ozone levels with relative humidity in all major cities, with R coefficient ranging from -0.84 in Hengshui city to -0.17 in Zhangjiakou city, and slope values varying widely from -2.57 to -0.48 μg m<sup>-3</sup>/1%RH (Fig. 2). The average R coefficient for all cities is -0.58 ± 0.14. Moreover, hourly O<sub>3</sub> levels are strongly correlated with RH in most cities, with correlation coefficient below -0.5 in 57 cities. Also, we did not observe significant heterogeneity of correlations between Southern and Northern regions, contrary to previous research in China and abroad (Camalier et al. 2007; Kavassalis and Murphy 2017; Cheng et al. 2018). This discrepancy is explained by the fact that analyses of previous investigations compared data at the same dates for different locations, whereas here we used only data of warm seasons, which occur at different dates in different regions. For example, greater responses were observed in the southern versus northern areas in 39 US cities from May to September (Camalier et al. 2007). We also studied the effect of other meteorological factors on ozone variations (Fig. S5). Results show that the relative humidity effect is comparable to or even stronger than temperature, and far exceeds wind speed. In summary, we found that almost all cities have large negative correlations between RH and ozone in warm seasons.

The different influences of relative humidity on ozone observed in different cities and regions (Fig. 2) can be explained by chemical and meteorological drivers. For instance, low temperatures in Southwestern areas throughout the year reduces O<sub>3</sub> formation, while high radiation intensity



**Fig. 2** Correlations between noontime  $O_3$  and relative humidity (RH), 11:00–16:00 local time, excluding data points with precipitation and low RH and T ( $RH \leq 25\%$ ,  $T \leq 15^\circ C$ ) in warm seasons of 2017 and 2018 for the major 74 cities in China. Results without exclusion are

shown in figure S3. S denotes the slope of linear regression equation, and R denotes the correlation coefficient. Results show that almost all cities have strong correlations with average R value of  $-0.58 \pm 0.14$

and long solar duration favors  $O_3$  production via enhancing the photolysis rates of  $NO_2$  and reaction rates of NO and  $O_3$  (Gaur et al. 2014). Also, long-range transport of pollutants from high emission areas to low emission regions can change precursor concentrations and further affect ozone-RH correlations.

Moreover, rising relative humidity increases the opening of tree stomata and, in turn, should induce the removal of ozone from air as a dry ozone sink (Kavassalis and Murphy 2017). To check this hypothesis, we tested the correlation of leaf area indices (LAI) with correlation coefficients of ozone and RH (Figure S1, Table S2). Our results indicate that the dry deposition of ozone in trees do not play a significant influence on the strong negative correlations between ozone and RH, in our conditions.

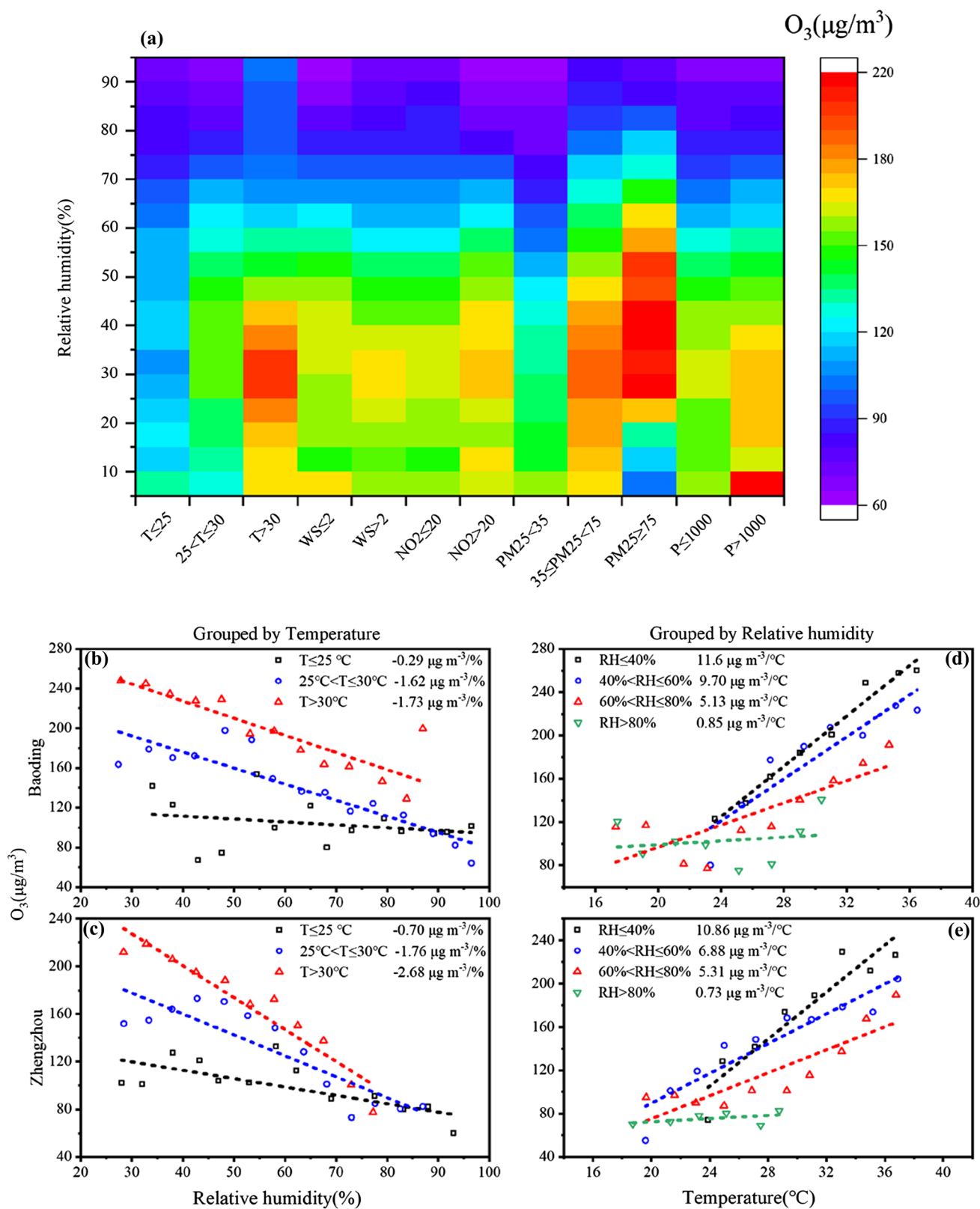
Table S3 shows the effects of high RH on  $O_3$  decreases in all cities. For RH over 75%, the average  $O_3$  concentrations varied from 44.6 to 122.5  $\mu g m^{-3}$  in these 74 cities, and the 90% percentile of hourly  $O_3$  concentrations ranged from 75.6 to 168.7  $\mu g m^{-3}$ , less than the quality standard of 200  $\mu g m^{-3}$ . For  $O_3$  concentrations exceeding 200  $\mu g m^{-3}$ , the mean RH values ranged from 31 to 66%, with 90% percentile of hourly RH values varying from 36 to 81%. This shows that  $O_3$  quality exceedance generally appeared under relatively low RH. In summary, our findings show that high

RH exerted great influence on  $O_3$  attainment in warm seasons for all 74 cities.

Overall, our findings show that ozone levels decreases with relative humidity in all Chinese regions, that there is no significant differences between southern and northern regions if only warm seasons are considered, and that dry ozone deposition on trees is not a major influencing factor in our conditions. Further effects of meteorological factors are studied below.

### Meteorological and chemical factors

We studied the effect of temperature, wind speed, surface pressure,  $NO_2$ , and particulate matter ( $PM_{2.5}$ ) concentrations, on ozone levels (Fig. 3a). Results show that  $O_3$  concentrations display larger variation ranges with RH at high temperature, low wind speed, high pressure, and high  $NO_2$  and  $PM_{2.5}$  levels. Ozone can exceed 160  $\mu g m^{-3}$  below 40% low RH, with  $PM_{2.5}$  over 35  $\mu g m^{-3}$  and  $NO_2$  over 20  $\mu g m^{-3}$ . This suggests the inducing effect of  $PM_{2.5}$  and  $NO_2$  on ozone, and implies that adverse health effects should be reduced by decreasing particulate matter and  $NO_2$  emissions. On the other hand, when relative humidity exceeds 60%,  $O_3$  concentrations are lower than 130  $\mu g m^{-3}$  under all conditions, except high  $PM_{2.5}$  levels, above 75  $\mu g m^{-3}$ .



This further demonstrates the important roles of high relative humidity for  $O_3$  decreases and the need of joint control of PM $_{2.5}$  and  $O_3$ .

We conducted a stratified analysis at four representative cities, Baoding in North China, Zhengzhou in Central China, Hangzhou in East China, and Chengdu in Southwest

**Fig. 3 a** The average  $O_3$  concentrations for each relative humidity (RH) bin (5% intervals) in all the major cities grouped by temperature (T), wind speed (WS), pressure (P),  $NO_2$ , and  $PM_{2.5}$  concentrations (units:  $^{\circ}C$ ,  $m\ s^{-1}$ , hPa,  $\mu g\ m^{-3}$ ,  $\mu g\ m^{-3}$ , respectively). Scatter plots of  $O_3$  against RH grouped by T during the noontime (11:00–16:00, local time) excluding precipitation in different warm seasons during 2017–2018 in **b** Baoding and **c** Zhengzhou. Points represent the average values of  $O_3$  concentration versus RH for each RH bin (5% intervals). Scatter plots of  $O_3$  against T grouped by RH during the same period as above in **d** Baoding and **e** Zhengzhou. Points represent the average values of  $O_3$  concentrations versus T for each T bin ( $2\ ^{\circ}C$  intervals). The slopes of linear regression equations are also provided in the plots ( $\alpha < 0.05$ ). Results show that high temperature strengthened negative dependence of  $O_3$  on RH, and high relative humidity not only lowered  $O_3$  levels, but also weakened the positive effects of T on  $O_3$

China to gain more knowledge on ozone response to relative humidity and temperature. These cities show high  $O_3$  pollution, high negative correlations between  $O_3$  and RH, with R below  $-0.5$ , and a large distribution range of hourly temperature and relative humidity data. Correlations of  $O_3$  versus RH at different temperature ranges are given in Figs. 3b, c, S4a, and S4b. Correlations of ozone versus temperature, grouped by relative humidity, are given in Figs. 3c, d, S4c, and S4d. Results show that the negative relationship between  $O_3$  and RH became stronger under higher temperature in Baoding city, with an average decreasing rate of  $-1.73\ \mu g\ m^{-3}/1\%RH$  above  $30\ ^{\circ}C$  compared to  $-0.29\ \mu g\ m^{-3}/1\%RH$  below  $25^{\circ}C$ . Similar variations were also found in Zhengzhou, Hangzhou and Chengdu with larger decreasing rates of  $O_3$  as RH rose at higher temperature. This result indicates that the increase of relative humidity is effective to decrease  $O_3$  concentrations at high temperature. The choice of the temperature range is therefore important to control  $O_3$  levels in mitigation practices.

We also observed that stronger increasing rates of  $O_3$  with temperature occurred under low relative humidity, below 40%, with 11.6, 10.86, 2.78, and  $9.19\ \mu g\ m^{-3}/^{\circ}C$  in Baoding, Zhengzhou, Hangzhou, and Chengdu, respectively, much higher than 0.85, 0.73, 0.04, and  $1.70\ \mu g\ m^{-3}/^{\circ}C$  in corresponding cities at high relative humidity, over 80%. Although under intermediate relative humidity, from 40 to 80%, increasing rates of  $O_3$  were comparable to and even greater in Hangzhou and Chengdu than under low RH, the weakening effects of high RH, above 80%, on  $O_3$  increase were evident in these two cities. This means that relative humidity can not only decrease ozone levels but also alleviate the positive effect of temperature on ozone generation. In summary, we found that high temperature strengthens the effects of relative humidity on ozone decrease, and that high relative humidity weakens the positive correlations between  $O_3$  and temperature.

## Conclusion

On the basis of observed pollutant concentrations and meteorological variables for the major 74 cities in China, we first studied seasonal variation patterns of  $O_3$ , then conducted correlation analyses of  $O_3$  and relative humidity during different warm seasons in 74 cities. Furthermore, the study discussed the effects of relative humidity and temperature on  $O_3$  using stratified analyses aimed at four cities, i.e., Baoding, Zhengzhou, Hangzhou, Chengdu.  $O_3$  levels displayed spatial differences among different regions, with higher levels mainly distributed in North China and East China. A considerable seasonal variation of  $O_3$  was recorded in each region, showing peak values in warm seasons with intense solar radiation, stagnant high-pressure conditions. We found the overall negative correlations between  $O_3$  and relative humidity in all cities, indicating the explanatory power of relative humidity to hourly variability of midday  $O_3$  in the warm seasons. Also, the role of high relative humidity in  $O_3$  attainment can be seen at all the cities. Then the stratified analyses show that high temperature can strengthen  $O_3$  dependence on relative humidity, and high relative humidity can weaken the positive impacts of temperature on  $O_3$ . Our results may have an implication for the great need of controlling the rising  $O_3$  pollution, especially in regions having drier climate conditions with low relative humidity and soil water content.

**Supplementary Information** The online version contains supplementary material available at <https://doi.org/10.1007/s10311-021-01265-0>.

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## Supplementary Information

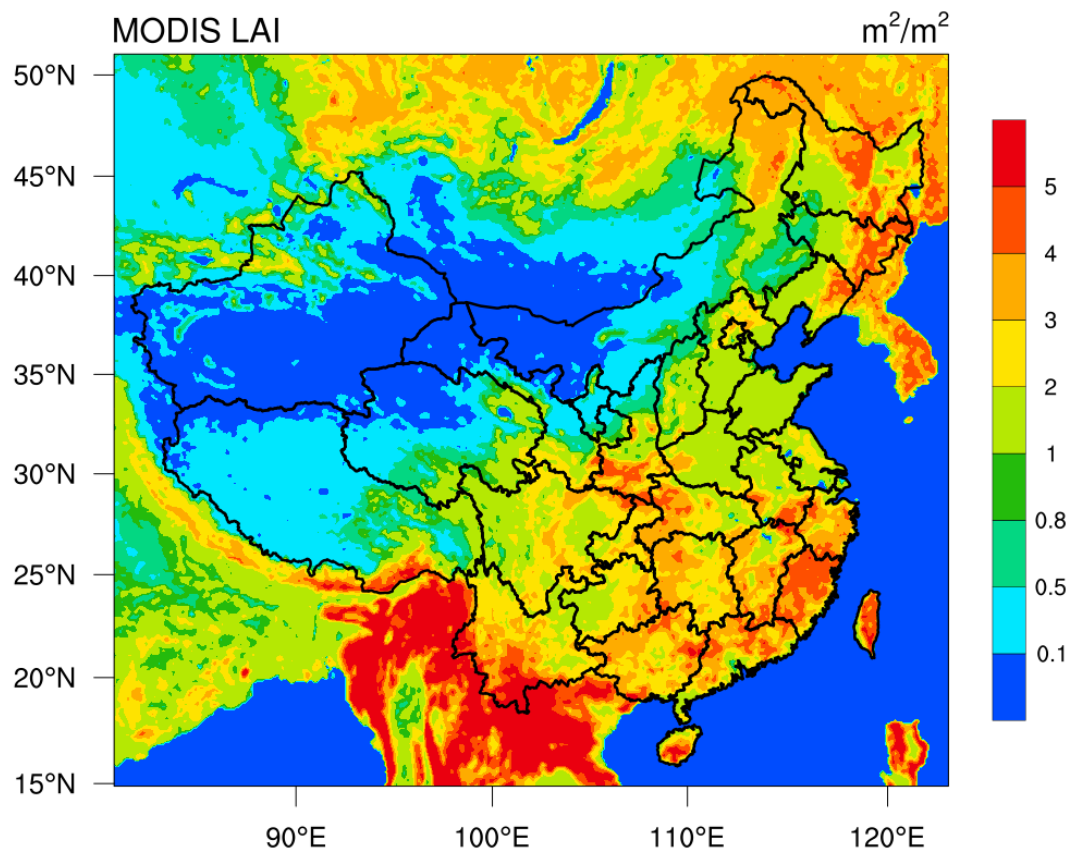


Fig. S1 Spatial distributions of leaf area indices (LAI) in summer season in China.

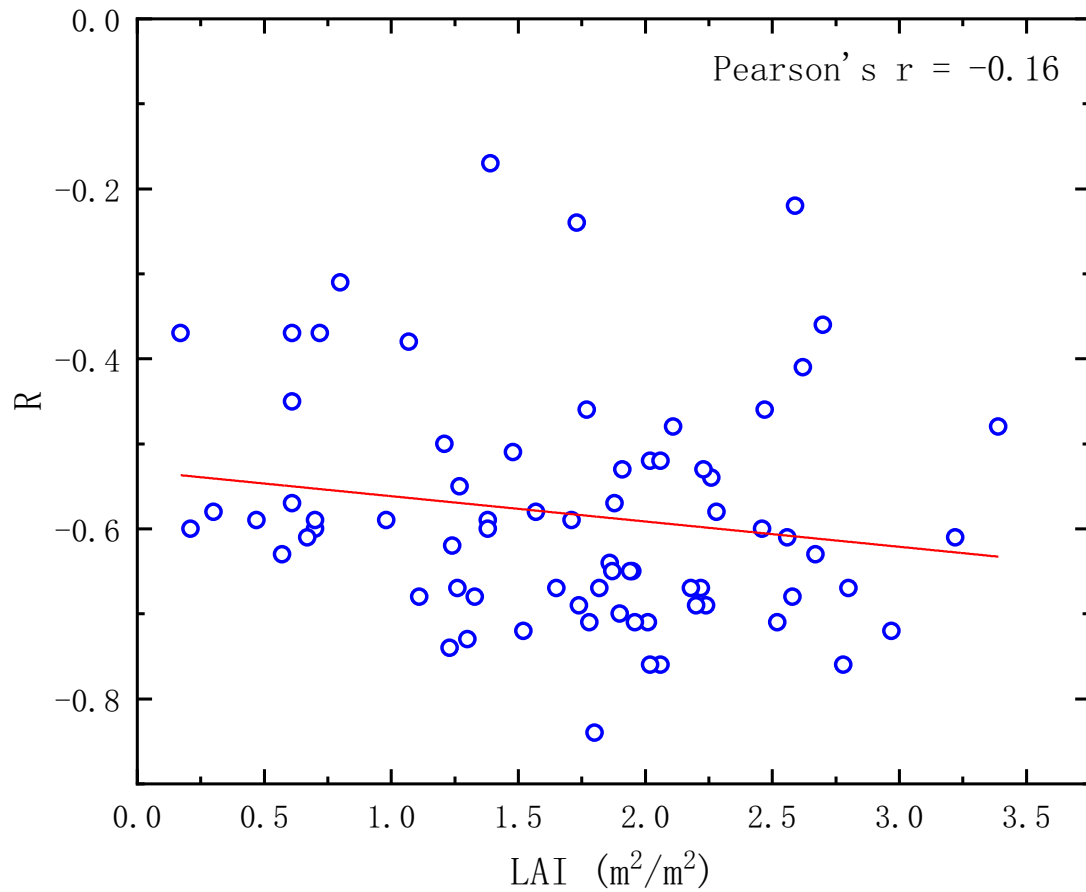


Fig. S2 Scatter plot and linear regression between leaf area indices (LAI) and correlation coefficients of  $O_3$  and RH (R) across all the 74 cities.

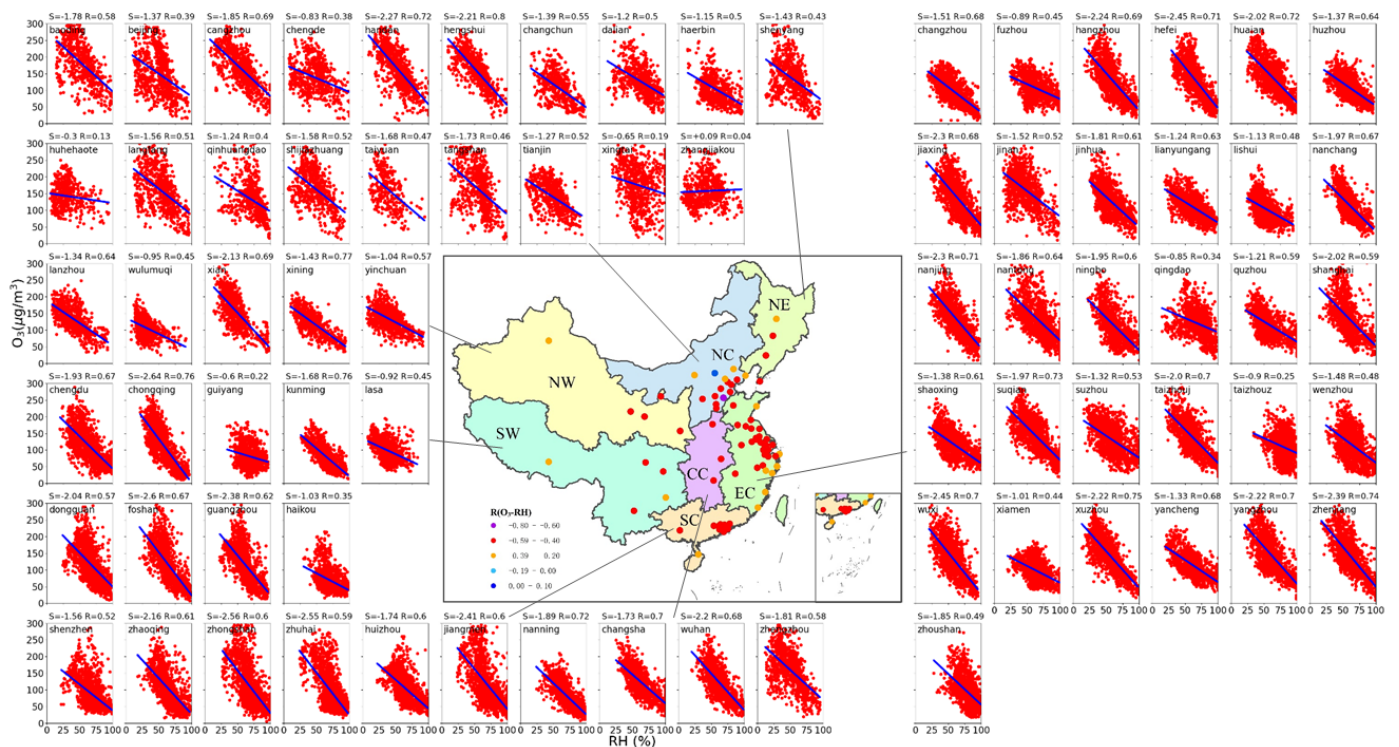


Fig. S3 Correlations between noontime  $O_3$  and relative humidity (RH) (11:00 to 16:00, local time) by excluding data points

with precipitation and low T ( $T \leq 15^\circ\text{C}$ ) in different warm seasons of 2017 and 2018 for the major 74 cities in China but for all RH values when compared to Fig. 2.

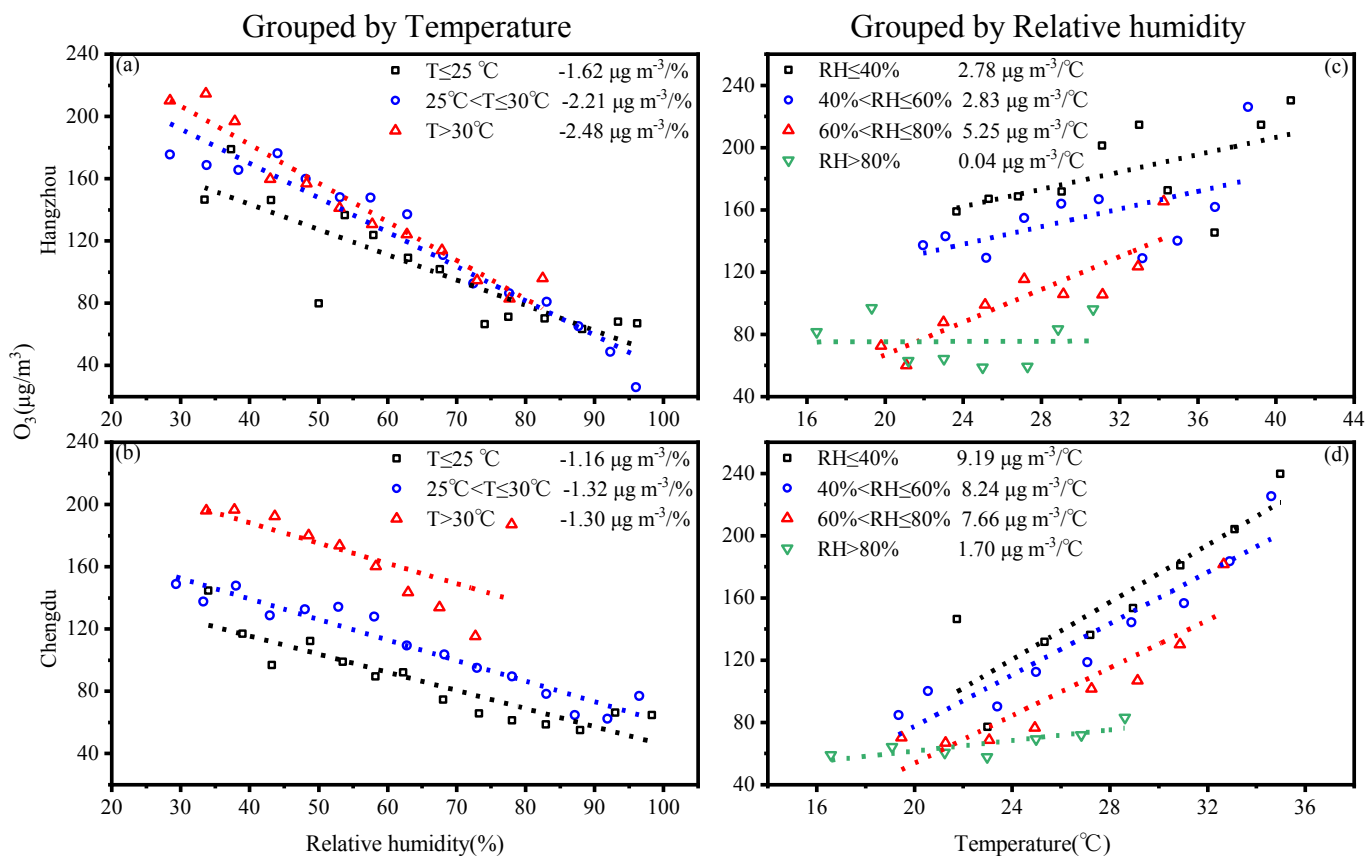


Fig. S4 Scatter plots of  $\text{O}_3$  against relative humidity (RH) grouped by T during the noontime (11:00 to 16:00, local time) excluding precipitation in different warm seasons during 2017-2018 in (a) Hangzhou and (b) Chengdu. Points represent the average values of  $\text{O}_3$  concentration versus RH for each RH bin (5% intervals). Scatter plots of  $\text{O}_3$  against T grouped by RH during the same period as above in (c) Hangzhou and (d) Chengdu. Points represent the average values of  $\text{O}_3$  concentrations versus T for each T bin ( $2^\circ\text{C}$  intervals). The slopes of linear regression equations are also provided in the plots ( $\alpha < 0.05$ ).

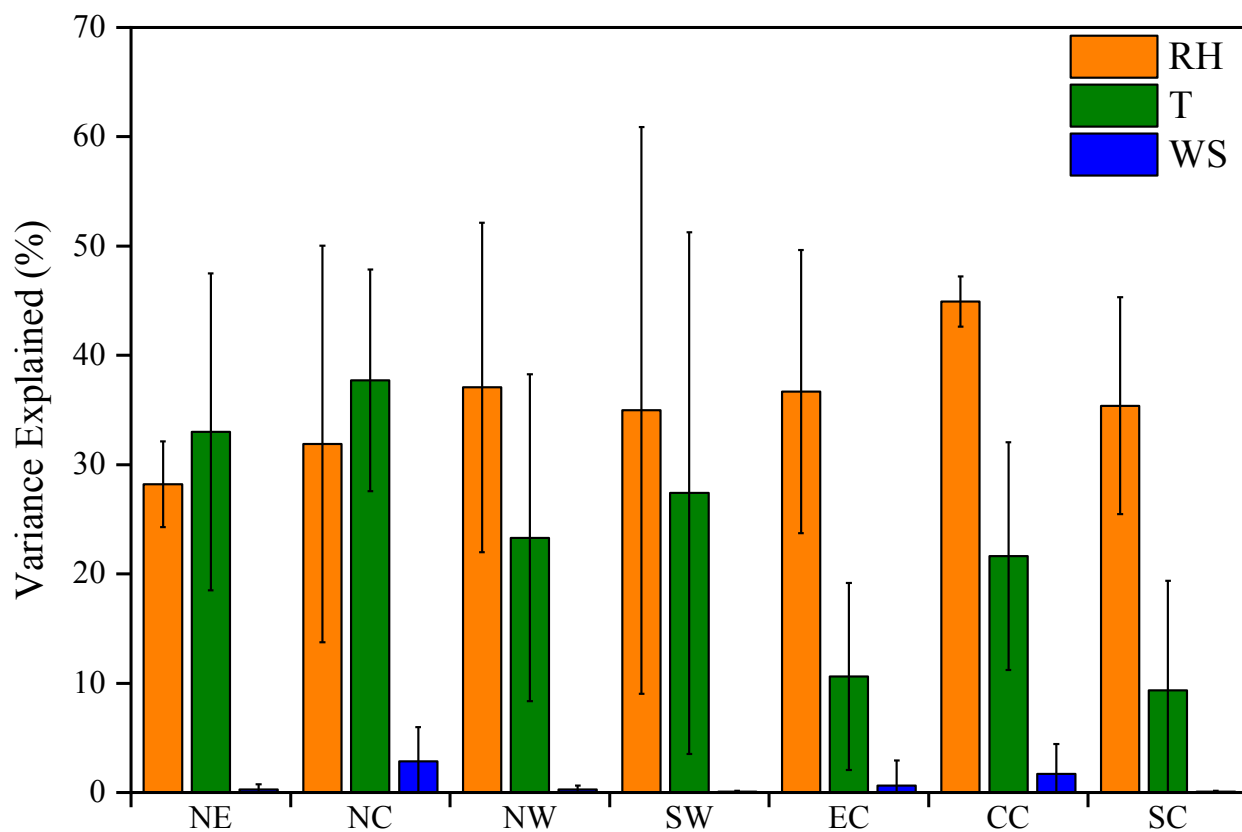


Fig. S5 Ozone variance attributed to meteorology conditions (relative humidity (RH), temperature (T), wind speed (WS)) during the noontime (11:00 to 16:00, local time) excluding precipitation in warm seasons in 2017-2018 averaged across all cities in seven regions. The variance explained is equal to the square of the correlation coefficient between  $O_3$  and each factor multiplied by 100. The error bars represent standard deviation from the mean.

Table S1 Related water vapor-involved chemical reactions of O<sub>3</sub> formation (Yu, 2019).

O <sub>3</sub> decomposition by water vapor	hydroxyl radicals termination reactions	NO <sub>2</sub> termination reactions
$O_3 + hv \rightarrow O(^1D) + O_2$	$HO_2 + CO + nH_2O \rightarrow CO_2 + OH$	$O(^3P) + NO_2 + M \rightarrow NO_3 + M$
$O(^1D) + H_2O \rightarrow 2OH$	$HO_2 + NO + nH_2O \rightarrow \text{nonradical products}$	$NO_2 + O_3 \rightarrow NO_3 + O_2$
$O_3 + OH \rightarrow HO_2 + O_2$	$OH + CO + nH_2O \rightarrow \text{nonradical products}$	$NO_2 + NO_3 + M \rightarrow N_2O_5 + M$
$O_3 + HO_2 \rightarrow OH + 2O_2$		$2NO_2 + H_2O \rightarrow HONO + HNO_3$
		$N_2O_5 + H_2O \rightarrow 2HNO_3$
		$N_2O_{5(gas)} + NaCl \rightarrow ClNO_2 + NaNO_3$

Table S2 Average leaf area indices (LAI) in summer season, topography height (HGT\_M) and at each of 74 major cities.

Region	City	LAI (m <sup>2</sup> /m <sup>2</sup> )	HGT_M (m)	R	Region	City	LAI (m <sup>2</sup> /m <sup>2</sup> )	HGT_M (m)	R
Northeast China	Dalian	0.61	28.34	-0.57	East China	Changzhou	1.65	10.08	-0.67
	Haerbin	2.11	160.38	-0.48		Fuzhou	2.62	146.11	-0.41
	Shenyang	2.26	93.31	-0.54		Hangzhou	1.74	40.92	-0.69
	Changchun	1.91	225.63	-0.53		Hefei	1.96	30.96	-0.71
North China	Baoding	1.95	9.38	-0.65		Huzhou	0.57	10.86	-0.63
	Beijing	1.77	99.48	-0.46		Huaian	2.24	10.05	-0.69
	Cangzhou	1.23	6.25	-0.74		Jinan	1.57	200.80	-0.58
	Chengde	2.70	573.66	-0.36		Jiaxing	2.22	7.02	-0.67
	Handan	2.01	42.66	-0.71		Jinhua	2.56	166.92	-0.61
	Hengshui	1.80	16.84	-0.84		Lishui	3.39	423.98	-0.48
	Huhehaote	1.07	1243.88	-0.38		Lianyungang	0.70	22.56	-0.6
	Langfang	1.71	8.34	-0.59		Nanchang	1.82	18.14	-0.67
	Qinhuangdao	0.17	19.13	-0.37		Nanjing	1.78	32.89	-0.71
	Shijiazhuang	1.94	55.65	-0.65		Nantong	2.28	3.60	-0.58
	Taiyuan	1.27	1059.15	-0.55		Ningbo	0.98	30.16	-0.59
	Tangshan	1.48	51.95	-0.51		Qingdao	0.72	80.12	-0.37
	Tianjin	1.38	5.08	-0.59		Quzhou	2.06	94.15	-0.52
	Xingtai	2.02	43.76	-0.52		Xiamen	0.61	58.70	-0.45
Zhangjiakou	1.39	1221.46	-0.17	Shanghai		0.21	2.58	-0.6	
South China	Dongguan	1.88	54.88	-0.57		Shaoxing	1.38	17.28	-0.6
	Foshan	1.26	16.69	-0.67	Suzhou	1.21	7.51	-0.5	
	Guangzhou	2.67	104.44	-0.63	Taizhouz	1.73	41.10	-0.24	
	Haikou	0.00	1.70	-0.35	Taizhouj	2.80	17.88	-0.67	
	Huizhou	2.46	91.05	-0.6	Wenzhou	2.47	123.08	-0.46	
	Jiangmen	0.67	10.09	-0.61	Wuxi	1.90	9.70	-0.7	
	Nanning	2.97	146.45	-0.72	Suqian	2.52	14.67	-0.71	
	Shenzhen	2.23	78.31	-0.53	Xuzhou	2.06	42.81	-0.76	
	Zhaoqing	3.22	133.32	-0.61	Yancheng	1.86	3.23	-0.64	
	Zhongshan	0.70	8.05	-0.59	Yangzhou	2.20	2.27	-0.69	
	Zhuhai	0.47	16.43	-0.59	Zhenjiang	1.52	5.10	-0.72	
Northwest China	Lanzhou	0.30	1802.68	-0.58	Zhoushan	0.00	8.10	-0.49	
	Wulumuqi	0.61	754.52	-0.37	Southwest China	Chengdu	2.18	591.93	-0.67
	Xian	1.33	456.64	-0.68		Guiyang	2.59	1240.98	-0.22
	Xining	1.30	2569.29	-0.73		Kunming	2.78	2100.75	-0.76
Yinchuan	1.24	1138.26	-0.62	Chongqing		2.02	356.03	-0.76	
Central China	Wuhan	1.11	21.65	-0.68		Lasa	0.80	4387.26	-0.31
	Changsha	2.58	65.72	-0.68					
	Zhengzhou	1.87	90.75	-0.65					



Table S3 Statistical values of O<sub>3</sub> concentrations under RH<sub>≥</sub>75% and RH under O<sub>3</sub>≥200 μg m<sup>-3</sup>. Min, Max, Mean, SD, 90% denotes the minimum, maximum, mean, standard deviation, and 90% percentile of corresponding parameters, respectively. NaN represents no values, that is, no O<sub>3</sub> concentrations exceeds 200 μg m<sup>-3</sup> for these cities. The average O<sub>3</sub> concentrations varied in the range of 44.6 and 122.5 μg m<sup>-3</sup> with 90% percentile of hourly O<sub>3</sub> values ranging from 75.6 to 168.7 μg m<sup>-3</sup> in all the 74 cities under RH over 75%, lower than China National Ambient Air Quality Standard (CNAAQs, GB3095-2012, 200 μg m<sup>-3</sup>), showing the important role of high RH in O<sub>3</sub> attainment in warm seasons at all cities.

Region	City	O <sub>3</sub> (μg m <sup>-3</sup> ) under RH <sub>≥</sub> 75%			RH (%) under O <sub>3</sub> ≥200 μg m <sup>-3</sup>		
		[Min, Max]	Mean ± SD	90%	[Min, Max]	Mean ± SD	90%
Northeast China	Dalian	[32.9, 190.8]	93.5 ± 29.3	131.9	[29, 70]	47 ± 11	66
	Haerbin	[29.2, 157.4]	70.7 ± 31.5	109.5	[27, 54]	43 ± 10	52
	Shenyang	[15.5, 126.1]	59.0 ± 31.9	99.3	[26, 69]	43 ± 10	56
	Changchun	[20.6, 151.5]	58.9 ± 21.7	79.4	[28, 59]	45 ± 10	57
North China	Baoding	[36.6, 199.7]	110.8 ± 38.1	157.2	[26, 71]	44 ± 11	58
	Beijing	[16.2, 131.5]	70.5 ± 28.7	111.2	[26, 63]	46 ± 10	57
	Cangzhou	[26.7, 189.0]	95.1 ± 34.5	136.9	[26, 73]	42 ± 11	58
	Chengde	[15.2, 163.0]	88.1 ± 34.2	128.9	[27, 62]	42 ± 9	54
	Handan	[31.5, 167.3]	91.1 ± 30.0	125.9	[26, 68]	40 ± 10	54
	Hengshui	[19.3, 133.3]	80.3 ± 27.7	110.9	[26, 67]	38 ± 8	47
	Huhehaote	[61.8, 159.9]	101.2 ± 33.0	149.0	[26, 39]	32 ± 4	36
	Langfang	[26.3, 172]	82.7 ± 35.6	125.2	[27, 73]	46 ± 11	60
	Qinhuangdao	[16.6, 302.2]	110.7 ± 43.4	162.1	[28, 82]	63 ± 14	78
	Shijiazhuang	[13.5, 174.1]	85.5 ± 37.4	136.8	[26, 70]	40 ± 10	52
	Taiyuan	[80.3, 134.5]	103.4 ± 20.7	123.3	[26, 49]	34 ± 6	43
	Tangshan	[13.0, 211.2]	90.3 ± 51.8	168.7	[27, 76]	48 ± 13	65
	Tianjin	[27.1, 185.4]	77.0 ± 31.4	99.7	[26, 62]	39 ± 7	47
	Xingtai	[15.8, 226.0]	98.0 ± 51	165.1	[27, 78]	46 ± 13	63
Zhangjiakou	[77.8, 182.8]	122.5 ± 25.9	155.4	[26, 71]	45 ± 9	58	
Northwest China	Lanzhou	[37.0, 107.2]	75.7 ± 22.4	105.7	[26, 52]	39 ± 8	46
	Wulumuqi	[39.7, 107.1]	67.0 ± 22.3	90.5	NaN		
	Xian	[34.7, 124.6]	76.5 ± 23.0	109.9	[26, 54]	36 ± 7	47
	Xining	[34.8, 108.0]	64.8 ± 16.8	85.9	NaN		
	Yinchuan	[49.3, 110.9]	82.6 ± 14.6	99.3	[26, 38]	31 ± 4	37
Southwest China	Chengdu	[15.8, 187.2]	69.2 ± 25.8	103.6	[32, 69]	46 ± 9	58
	Guiyang	[23.6, 160.7]	76.2 ± 27.9	114.9	NaN		
	Kunming	[13.7, 115.1]	53.5 ± 19.6	81.1	NaN		
	Chongqing	[10.9, 151.1]	44.6 ± 23.0	75.6	[28, 55]	41 ± 7	50
	Lasa	NaN			NaN		
East China	Changzhou	[10.6, 125.9]	53.1 ± 24.0	88.5	NaN		
	Fuzhou	[26.4, 180]	91.2 ± 26.6	128.2	NaN		
	Hangzhou	[18.4, 148.7]	70.7 ± 28.1	113.9	[27, 71]	44 ± 9	54
	Hefei	[20.9, 168.6]	76.2 ± 28.4	116.1	[31, 68]	52 ± 8	62
	Huzhou	[31.6, 188.2]	70.6 ± 24.1	98.0	[39, 49]	43 ± 5	47
	Huaian	[27.8, 189.0]	94.4 ± 32.1	134.7	[26, 67]	43 ± 10	57
	Jinan	[10.9, 222.9]	79.9 ± 43.8	129.2	[26, 84]	42 ± 11	55
	Jiaxing	[22.7, 216.0]	86.0 ± 35.6	136.8	[27, 75]	50 ± 10	64
	Jinhua	[15.7, 144.0]	78.8 ± 28.4	114.1	[28, 70]	42 ± 9	55

	Lishui	[24.7, 134.0]	$73.1 \pm 21.4$	103.0	[30, 49]	$40 \pm 7$	46
	Lianyungang	[29.4, 160.0]	$80.0 \pm 22.8$	109.2	[34, 44]	$38 \pm 4$	42
	Nanchang	[21.6, 177.4]	$73.1 \pm 26.9$	103.7	[33, 50]	$40 \pm 6$	49
	Nanjing	[9.7, 187.2]	$80.8 \pm 34.4$	126.0	[28, 67]	$47 \pm 9$	58
	Nantong	[24.8, 308.0]	$94.7 \pm 38.0$	142.0	[26, 93]	$50 \pm 12$	69
	Ningbo	[22.3, 200.6]	$78.7 \pm 30.9$	115.6	[29, 90]	$47 \pm 10$	59
	Qingdao	[14.3, 241.2]	$99.9 \pm 39.5$	151.7	[43, 89]	$58 \pm 11$	73
	Quzhou	[31.0, 155.0]	$88.5 \pm 27.1$	128.9	[28, 47]	$37 \pm 5$	43
	Xiamen	[32.5, 170.7]	$75.8 \pm 23.7$	106.3	NaN		
	Shanghai	[24.5, 302.1]	$84.9 \pm 41$	139.2	[26, 91]	$47 \pm 10$	58
	Shaoxing	[35.0, 172.5]	$83.0 \pm 23.9$	115.6	[28, 49]	$38 \pm 6$	45
	Suzhou	[31.7, 233.4]	$97.4 \pm 37.9$	153.0	[26, 77]	$45 \pm 14$	68
	Taizhouz	[18.0, 231.7]	$103.1 \pm 37.7$	151.3	[50, 92]	$66 \pm 12$	81
	Taizhouj	[33.0, 227.3]	$90.0 \pm 32$	125.7	[26, 81]	$44 \pm 12$	58
	Wenzhou	[20.3, 208]	$80.3 \pm 37.3$	136.1	[31, 75]	$51 \pm 8$	60
	Wuxi	[14.4, 187.9]	$64.0 \pm 30.5$	100.9	[26, 67]	$42 \pm 9$	54
	Suqian	[19.8, 191]	$93.0 \pm 33.5$	138.0	[26, 71]	$44 \pm 9$	57
	Xuzhou	[16.8, 175.4]	$71.5 \pm 26.0$	101.7	[26, 67]	$40 \pm 8$	50
	Yancheng	[35.0, 155.5]	$83.5 \pm 20.5$	107.8	[27, 52]	$37 \pm 7$	48
	Yangzhou	[13.0, 212.4]	$87.5 \pm 30.1$	126.5	[26, 76]	$46 \pm 10$	58
	Zhenjiang	[9.3, 193.3]	$83.1 \pm 34.1$	132.6	[26, 69]	$46 \pm 10$	60
	Zhoushan	[17.3, 221.3]	$81.1 \pm 37.3$	132.2	[49, 80]	$64 \pm 9$	74
Central China	Wuhan	[21.8, 152.3]	$68.9 \pm 25.7$	102.9	[30, 66]	$46 \pm 7$	53
	Changsha	[22.7, 163.6]	$81.7 \pm 26$	117.8	[27, 65]	$42 \pm 8$	52
	Zhengzhou	[26.6, 149.1]	$79.3 \pm 27.3$	107.4	[26, 66]	$39 \pm 9$	54
South China	Dongguan	[9.8, 228.2]	$72.1 \pm 38.1$	119.6	[42, 85]	$57 \pm 8$	70
	Foshan	[7.4, 212.0]	$58.6 \pm 30.9$	93.2	[28, 84]	$52 \pm 9$	61
	Guangzhou	[13.3, 181.7]	$60.7 \pm 35.3$	114.4	[40, 67]	$52 \pm 6$	60
	Haikou	[20.2, 131.4]	$56.7 \pm 22.8$	89.2	[51, 68]	$62 \pm 6$	67
	Huizhou	[23.0, 219.8]	$70.5 \pm 32.8$	114.6	[46, 95]	$60 \pm 11$	69
	Jiangmen	[9.0, 288.3]	$73.1 \pm 38$	114.0	[28, 93]	$53 \pm 12$	66
	Nanning	[8.0, 139.0]	$53.8 \pm 23.5$	85.1	[59, 61]	$60 \pm 1$	61
	Shenzhen	[27.7, 208.5]	$67.9 \pm 31.8$	110.3	[38, 84]	$57 \pm 11$	71
	Zhaoqing	[16.0, 231.3]	$64.8 \pm 41.4$	123.3	[37, 87]	$53 \pm 10$	64
	Zhongshan	[6.0, 270.3]	$70.6 \pm 34.7$	112.1	[30, 76]	$52 \pm 10$	66
	Zhuhai	[16.8, 250.3]	$70.4 \pm 35.4$	110.8	[26, 81]	$55 \pm 14$	73