Impacts of urbanization on air quality and related health risks in a city with complex terrain

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Abstract: Urbanization affects air pollutants via~~by~~ urban expansion and emission growth, and thereby inevitably changes the health risks of air pollutants. However, the health risks related to urbanization are rarely estimated, especially for cities with complex terrain. In this study, a highly urbanized city with severe air pollution and complex terrain (Chengdu) is selected to explore this issue. The effects of urban expansion are further compared with emission growth since ~~as~~ air quality management is usually achieved by~~is mainly to~~ regulating~~e~~ anthropogenic emissions. Air pollution in Chengdu was~~is~~ mainly caused by $PM_{2.5}$ and O_3 from 2015 to 2021. $PM_{2.5}$ pollution tended~~s~~ to appear in cold months (November to February) owing~~due~~ to the ~~secondary circulation forced by complex terrain~~blocking of air and the frequent temperature inversion, while O_3 pollution was~~is~~ likely to occur in warm months (April to August) because of high temperature and strong sunlight dominated by high-pressure systems. From 2015 to 2021, the 7-year annual average of~~total~~ premature mortalities from all non-accidental causes (ANAC) due~~attributed~~ to $PM_{2.5}$ and O_3 were~~exposure are~~ 9386 (95%CI: 6542~11726)~~9386~~ and 8506 (95%CI: 4817~11882)~~7743~~, respectively. Based on the characteristics of $PM_{2.5}$ and O_3, six numerical experiments were~~are~~ conducted to investigate the impacts of urban expansion and emission growth on health risks of air pollutants. The results show that urban land use~~expansion~~ led to~~causes~~ an increase in air temperature and the boundary layer height, which was~~is~~ conducive to the diffusion of $PM_{2.5}$. Thus, the monthly

average surface $PM_{2.5}$ concentrations could decrease by 10.8~~1.7~~ μg m^{-3} (7.6%) in January. However, the monthly average daily maximum 8 h average (MDA8) O_3 concentrations could increase by 10.6 μg m^{-3} (6.0%) in July owing~~due~~ to the stronger photochemical production and better vertical mixing during~~ the~~ daytime. In this case~~Correspondingly~~,~~ the total~~ premature mortalities from ANAC due~~attributed~~ to $PM_{2.5}$~~ exposure~~ decreased by ~~182~~ 171 (95%CI: 129~200, or about 6.9%) ~~(6.9%)~~ in January, and~~but~~ those due~~attributed~~ to O_3 ~~exposure~~ increased by 203 (95%CI: 122~268, or about 9.5%)~~203 (9.5%)~~ in July. As for the effects of emission growth, the monthly average $PM_{2.5}$ and MDA8 O_3 concentrations could~~an~~ increase by 23.9 μg m^{-3} (16.8%)~~26.6 μg m^{-3}~~ and 4.8 μg m^{-3} (2.7%)~~4.8 μg m^{-3}~~ when anthropogenic emissions were~~are~~ taken into account. ~~The total~~ Ppremature mortalities from ANAC due~~attributed~~ to $PM_{2.5}$ and O_3~~ exposure~~ then increased by 388 (95%CI: 291~456, or about 15.7%)~~424 (16.0%)~~ and 87 (95%CI: 54~112, or about 4.1%)~~87 (4.1%)~~, respectively. From a health risk perspective, t~~T~~he effects of urban land use~~expansion~~ on health risks of $PM_{2.5}$ are~~is~~ about half that of anthropogenic emissions, ~~.~~ w~~W~~hereas the effects of urban land use~~expansion~~ on health risks of O_3 can be 2~~.3~~ times that of anthropogenic emissions. This reminds us that, in addition to regulating anthropogenic emissions, urban planning~~the development of cities~~ is also important for ~~the~~ urban air quality, especially for secondary pollutants like O_3.~~ apart from the emissions reduction.~~

Key Words: urbanization; land use; anthropogenic emissions; air quality; health risk;

1 Introduction

Air pollutants are substances that damage humans, plants and animals drastically when present in the atmosphere in sufficient concentration (Baklanov et al., 2016; Kinney, 2018; Pautasso et al., 2010). The most common air pollutants found in air are ozone (O_3), fine particulate matter ($PM_{2.5}$, particulate matter with an aerodynamic diameter of 2.5 μm or less), sulfur dioxide (SO_2) and nitrogen oxides (NO_x, which is NO + NO_2). These air pollutants threaten human health in many parts of the world, evoking a series of health risks including cardiovascular diseases, respiratory diseases and chronic obstructive pulmonary disease (Brauer et al., 2016; Lelieveld et al., 2013; Manisalidis et al., 2020). According to the World Health Organization (WHO), exposure to ambient air pollutants is associated with~~results in~~ 4.2~~about 4.2~~ million premature deaths annually~~globally~~

~~per year~~ (https://www.who.int/health-topics/air-pollution#tab=tab_2).

Most of those premature deaths occur in urban areas as urban areas currently host more than 50% of the population (over 3.5 billion people). This proportion ~~and this number~~ is projected to increase to 70% by 2050 due to ongoing urbanization (UNDESA, 2018~~)~~). Urbanization since the industrial revolution in the 19th century has led to a profound modification of land use via urban expansion (Seto et al., 2012). Natural surfaces are replaced by impervious surfaces, then the surface physical properties (e.g.., albedo, thermal inertia and roughness) and processes (e.g., the exchange of water, momentum and energy) are modified. These changes in surface physical properties and processes exert an important influence on urban meteorology and air quality, which has been widely acknowledged in previous studies. Wang et al. (2009) explored the impacts of urban expansion on weather conditions and its implication on O_3 concentration in the Pearl River Delta, and pointed out that urban land use changes can cause an increase in 2-m temperature by 1.0%~3.7%, an increase in planetary boundary layer height by 5.9%~6.3% and an increase in surface O_3 concentration by 4.2%~8.5%. Liao et al. (2015) conducted a similar study in the Yangtze River Delta, and found that urbanization increases 2-m temperature, planetary boundary layer and surface O_3 concentration but decreases surface PM_{10} (particulate matter with an aerodynamic diameter of 10 μm or less) concentration. Similar conclusions about the impacts of urbanization on meteorology and air quality have also been reported in the Beijing-Tianjin-Hebei region (Yu et al., 2012) and the Sichuan Basin (Wang et al., 2021, 2022a).

~~What's more, u~~Urban areas are centers of resource utilization and are a major contributor to air pollutants~~t~~ and greenhouse gas emissions (Karl et al., 2019; Qian et al., 2022) ~~and greenhouse gas emissions~~. According to the UN-Habitat (https://unhabitat.org/topic/energy), cities consume about 75% of global primary energy and emit ~~50%-60% of the world's greenhouse gas. Urbanization, especially in the rapid development stage, leads to increase in anthropogenic emissions.~~ Air pollutants originate from anthropogenic sources can ~~sometimes~~ accumulate and degrade urban air quality under unfavorable meteorological conditions characterized by weak winds, which leaves urban dwellers vulnerable to air pollution (Holman et al., 2015; Lin and Zhu, 2018). Excessive emissions are ~~considered to be~~ the root cause of poor air quality in urban areas, ~~and by effectively decreasing anthropogenic emissions, the health of urban dwellers can be improved. Urbanization can increase urban land use and anthropogenic emissions, and by effectively decreasing anthropogenic emissions, the health of~~ urban air pollution control. Urbanization can increase urban land use and anthropogenic emissions, which will affect the concentrations as well as health risks of air pollutants. However, the health risks related to urbanization are rarely estimated, especially for those cities with complex terrain.

This is of great concern to policymakers and can inspire future air quality control strategies. .

Although building in the mountains is not as easy as in the plains~~. In addition to emissions, urban air quality is also closely related to meteorology (Qian et al., 2017~~2022~~). Under calm and clear weatherconditions, local circulationsmountain-valley breezes driveninduced by unequal heating and cooling of the valleys and mountainsthe thermal contrast of the topography, such as mountain-valley breezes and sea-land breezes, are likely to form and play an important role in air quality urban environment (Crosman and Horel, 2010; Zhan and Xie, 2022). Because of historical, political and economic reasons~~, about 12% of the global population (over 720 million people) resides in mountainous areas for historical, political, strategic and economic reasons. It is thus important to understand the fate of air pollutants in mountainous cities, where air pollution is usually more severe than in flat locations~~ since mountainous terrain strongly alters the boundary layer structure, resulting in much more complicated diffusion conditions~~ as the atmospheric dispersion is limited in mountainous areas (Zardi and Whiteman, 2013~~Chow et al., 2013~~). The mountain-plain wind, resulting from horizontal temperature differences between air over mountain massifs and the air over the surrounding plains, is a key feature of the climatology of mountainous regions (Whiteman, 2000), which is important in determining the transport and dispersion of air pollutants. During daydurtime, the plain-to-mountain wind (plain wind) brings low-level air into the mountain massifs, whereas the mountain-to-plain wind (mountain wind) brings air out of the mountain massifs during nighttime. This wind system can often recirculate urban air pollutants and worsen air quality. ~~Many notably pollution episodes appear in valley bottoms, along mountain slopes and in mountain basins. These include~~ Examples can be found in~~examples like~~ Mexico City (Molina et al., 2010), Hong Kong (Guo et al., 2013), ~~the~~ Seoul (Ryu et al., 2013), the Salt Lake Valley (Baasandorj et al., 2017), the Colorado Front Range (Bahreini et al., 2018), the Alps (Karl et al., 2019) and the Taiwan Island (Lee et al., 2019).~~ Although the principles behind these examples apply to mountainous areas around the world, the phenomenon being described depends on the particular region (Whiteman, 2000; Oke et al., 2017). And a common principle is that diurnal wind systems driven by mountainous terrain can recirculate urban air pollutants and worsen air quality.~~

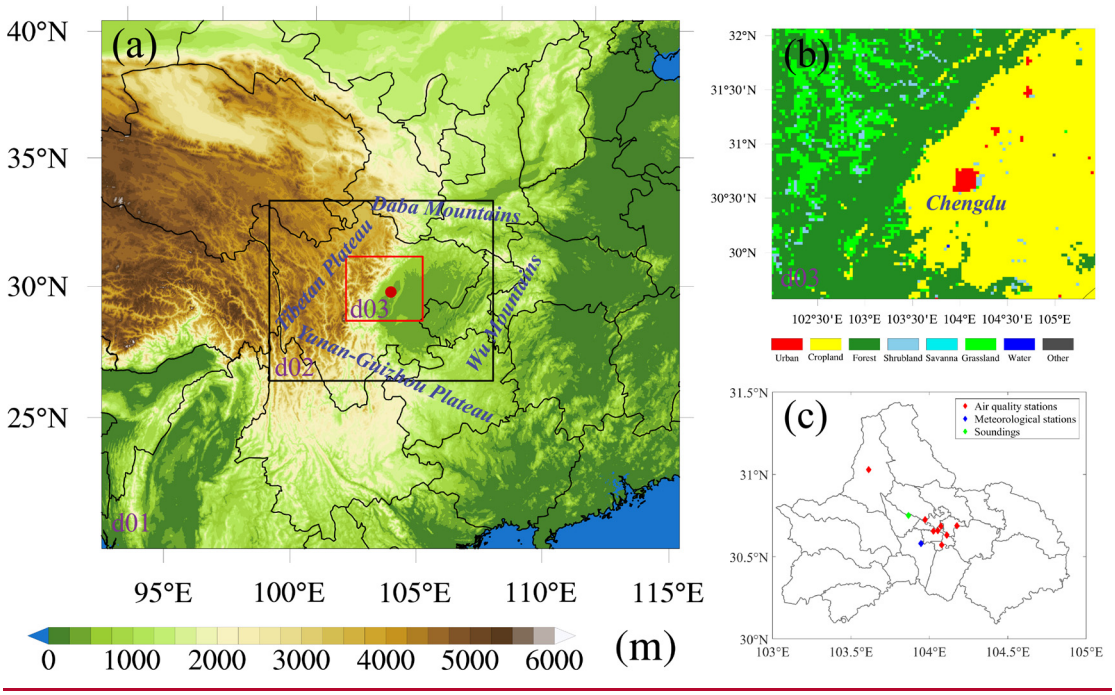
~~The world has been undergoing urbanization since the industrial revolution in the 19th century (Seto et al., 2012), which directly leads to changes in land use via urban expansion. Natural~~

~~surfaces are replaced by impervious surfaces, then land surface physical properties (e.g., albedo, thermal inertia and roughness) and processes (e.g., the exchange of water, momentum and energy) are modified, hence altering the urban meteorology and air quality. This has been widely investigated using numerical models. For example, Liao et al. (2015) reported that urban expansion can cause an increase in 2-m temperature by 0.9–2.3 °C, a decrease in 10-m wind speed by 0.6–1.2 m s^{-1} and an increase in planetary boundary layer height by 100–425 m in the Yangtze River Delta. These changes in meteorology further reduce surface PM_{10} concentrations by 15.3–57.6 μg m^{-3} but increase O_3 concentrations by 1.7–8.3 ppbv. Changes in concentrations of air pollutants inevitably affect their health risks. However, the health risks related to urbanization are rarely estimated, especially for those cities with complex terrain. This is of great concern to policymakers and can inspire future air quality control strategies in mountainous areas.~~

~~——~~Chengdu (104.01°E, 30.70°N) is the largest city in western China, occupying an area of 12,390 square kilometers with a population of more than 20 million. ~~Chengdu has the most complex terrain in the world.~~ Located in the west of the Sichuan Basin, this city is surrounded by the Tibetan Plateau to the west, the Wu Mountains to the east, the Yunnan-Guizhou Plateau to the south and the Daba Mountains to the north (Figure 1a). ~~In addition, the urbanization of Chengdu has developed rapidly~~Chengdu has experienced rapid urbanization over the past few decades with a surge in urban construction lands and a loss of cropland (Dai et al., 2021; Figure 1b). ~~The fast urbanization process is generally accompanied by a surge in urban construction lands and the loss of cropland (Figure 1c).~~ Luo et al. (2021) reported that Chengdu's urban area has increased by four times from 1996 to 2016. ~~Due to~~Because of the substantial anthropogenic emissions from human activities ~~(Figure 1b)~~ and the poor atmospheric diffusion capacity ~~caused by~~ associated with ~~complex~~ terrain, Chengdu is one of the most polluted cities in China and has suffered from severe $PM_{2.5}$ and O_3 pollution in recent years (Shu et al., 2021; Yang et al., 2020; Zhan et al., 2019). Complex terrain, rapid urbanization and severe air pollution make Chengdu an ideal place to study the impact of urbanization on health risks of air pollutants in mountainous areas. The results could also provide valuable insight for other cities with complex terrain in the world.

In this study, we ~~systematically~~ investigate~~evaluate~~ the impacts of urbanization on air pollutants~~quality~~ and the corresponding health risks in Chengdu. We also compare the impacts of

urban ~~expansion~~ expansion with emission growth~~emission growth~~. First, the basic characteristics of air pollutants in Chengdu from 2015 to 2021 are analyzed~~illustrated~~. Then, the impacts of urbanization on air pollutants are investigated ~~by~~ using the WRF-Chem model. Finally, premature mortalities attributable~~ed~~ to changes in air pollutants are estimated ~~by~~ using the standard damage function. The rest of this paper is organized as follows. Section 2 introduces the ~~observation~~ data, the model configurations and the experimental designs. Section 3 shows the main results and discussions. The conclusions are given in Section 4.



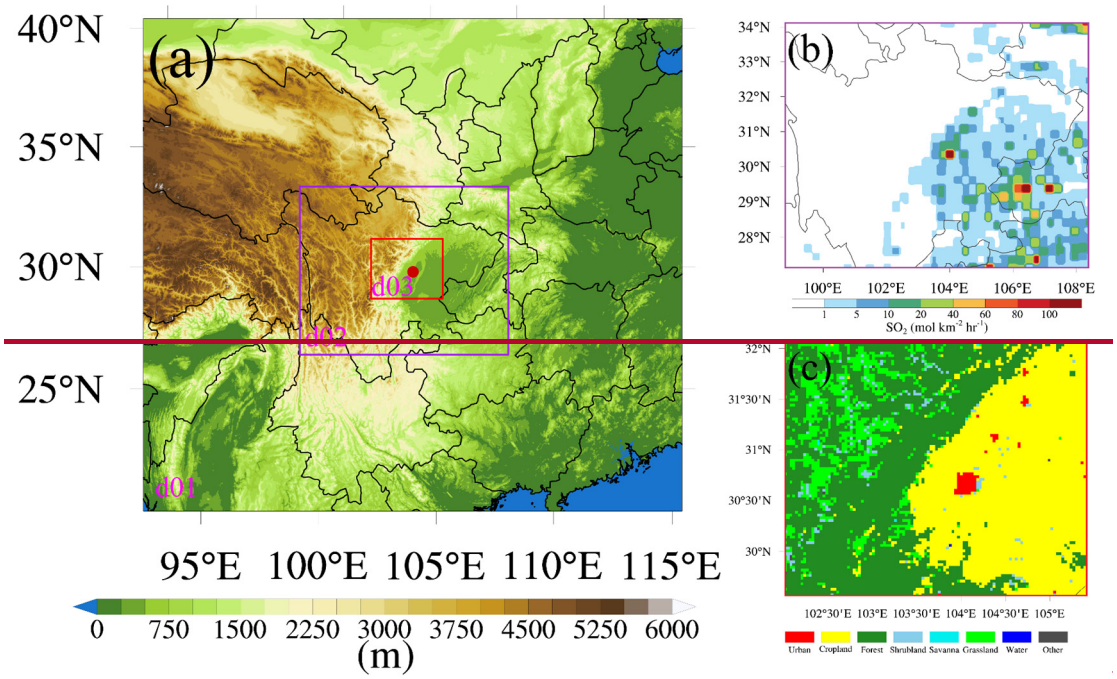


Figure 1. (a) ~~Three nested WRF-Chem~~Map of three nested WRF-Chem **domains** ~~, including (a) domain 1~~ **with terrain heights, (b)** ~~domain 2 with SO_2 emissions and (c)~~ **domain 3 with land cover maps** and (c) locations of air quality stations, meteorological stations and soundings in Chengdu**. The red dot in (a) shows the location of Chengdu.**

2 Data~~Materials~~ and methods

2.1 Air quality and meteorological~~Air pollutants and meteorological~~ data

Air pollutants, including $PM_{2.5}$, PM_{10}, O_3, NO_2, SO_2 and CO, are monitored by the National Environmental Monitoring Center ~~(NEMC)~~ of China. These data are hourly issued on the national urban air quality real-time publishing platform (http://106.37.208.233:20035/). The monitoring data are strictly in accordance with the national monitoring regulations (http://www.cnemc.cn/jcgf/dqhj/). It should be noted that the O_3 measurements are reported in the unit of μg m^{-3} at the standard atmospheric conditions (273.15 K, 1 atm) before September 2018, and at 298.15 K conditions afterward. There are eight air quality stations throughout Chengdu (Figure 1c), and the urban hourly pollutants are the average results of measurements at all monitoring sites. The daily $PM_{2.5}$ concentrations are obtained by averaging observations over 24 hours of the day. The daily maximum 8 h average (MDA8) O_3 concentrations are calculated only on days with more than 18 h of O_3 measurements. ~~This nationwide observation network consists of more than 2300 stations distributed~~

~~over 450 cities in China.~~

Surface meteorological data, including 2-m air temperature (T_2), ~~relative humidity (RH)~~2-m dew point temperature (TD_2), ~~and~~ 10-m wind speed (WS_{10}) and 10-m direction (WD_{10}), are taken from the website of the University of Wyoming at station ZUUU (http://weather.uwyo.edu/surface/). To verify upper-air fields, the sounding observations at Wenjiang (~~103.87°E, 30.75°N~~station 56187) ~~in Chengdu~~ are also acquired from this website. These sounding data contain temperature, dew point temperature ~~relative humidity~~ and wind speed, etc. at different pressure layers with a time resolution of 12 h (00:00 and 12:00 UTC), and are often plotted on a Skew-T diagram (https://www.ncl.ucar.edu/Applications/skewt.shtml#ex2).

~~In this study, the data quality control are performed as follows. First, the data indicated as missing are set as invalid. Second, the urban values are calculated by averaging observations at all monitoring sites in Chengdu. Third, the daily maximum 8 h average (MDA8) O_3 concentrations are calculated only on days with more than 18 h of O_3 concentrations measurements.~~

2.2 WRF-Chem model and experimental designs

WRF-Chem is the Weather Research and Forecasting (WRF) model coupled with Chemistry, in which meteorological and chemical variables use the same coordinates, transport schemes and physics schemes in space and time (Grell et al., 2005). WRF-Chem version 3.9.1 is employed in this study. As shown in Figure 1a, three nested domains are used with the grid spacing of 27, 9 and 3 km, respectively. 32 sigma levels are extending from the surface to 100 hPa in the vertical direction with 12 levels located below 2 km to resolve the boundary layer processes. The size of the lowest vertical grid is about 25 m. The MODIS-based land use data set as default in WRF are selected. The domains and main options for physical and chemical parameterization schemes are listed in Table 1. The National Centers for Environmental Prediction (NCEP) ~~global final~~Final (FNL) reanalysis data with a ~~horizontal~~ resolution of 1° × 1° at 6 h time intervals are adopted as the initial and boundary conditions for meteorological fields. Anthropogenic emissions are provided by the Multi-resolution Emission Inventory for China (MEIC) ~~in 2017~~ with a grid resolution of~~at~~ 0.25° × 0.25° ~~resolution~~. Biogenic emissions are calculated online using the Guenther scheme (Guenther et al., 2006).

Table 1. The domains and main options for WRF-Chem.

Items	Contents
Domains (x, y)	(94, 86), (109, 88), (112, 94)
Grid spacing (km)	27, 9, 3
Center	(104°E, 31°N)
Time step (s)	90
Microphysics	Purdue Lin scheme (Chen and Sun, 2002)
Longwave radiation	RRTM scheme (Mlawer et al., 1997)
Shortwave radiation	Goddard shortwave scheme (Matsui et al., 2018)
Surface layer	Monin-Obukhov scheme (Janjic, 1994)
Land-surface layer	Unified Noah land-surface model (Tewari et al., 2014)
Planetary boundary layer	Mellor-Yamada-Janjic TKE scheme (Janjic, 1994)
Cumulus parameterization	Grell 3D ensemble scheme (Grell and Devenyi, 2002)
Gas-phase chemistry	RADM2 (Stockwell et al., 1990)
Photolysis scheme	Fast-J photolysis (Fast et al., 2006)
Aerosol module	MADE/SORGAM (Schell et al., 2001)

To estimate~~investigate~~ the impacts of ~~urban expansion and anthropogenic emissions~~urbanization, six numerical simulations~~experiments~~ are designed (Table 2). The year of the numerical simulations is 2017 since the MEIC emission inventory~~anthropogenic emissions~~ is~~are~~ currently updated to 2017. Taking into account the computational cost~~Moreover~~, January is the representative of the cold months with frequent $PM_{2.5}$ pollution, while July is the representative of~~s~~ the warm months~~hot season~~ with frequent O_3 pollution (Section 3.1). Jan_Base~~ simulation~~ is a baseline simulation using the MODIS land use~~-~~ and the MEIC~~ anthropogenic~~ emission inventory over all three domains. The~~SO_2 emissions in domain 2 and~~ land cover maps in domain 3 are particularly ~~illustrated~~ shown in Figure 1b~~1b and 1c~~. Jan_noCD is a sensitivity simulation, in which the urban land use~~surface~~ of Chengdu is replaced by cropland to examine the impacts of urban expansion~~expansion~~. Jan_noEmi is another~~also a~~ sensitivity simulation, in which the anthropogenic emissions in Chengdu are shut down to identify the impacts of emission growth~~anthropogenic emissions~~. The above three numerical experiments use~~d~~ the same configurations (Table 1) running

from 00:00 UTC December 28, 2016 to 00:00 UTC February 1, 2017 with the first 96 h as spin-up time. July_Base, July_noCD and July_noEmi are the same as Jan_Base, Jan_noCD and Jan_noEmi, but run from 00:00 UTC June 27 to 00:00 UTC August 1, 2017 with the first 96 h as spin-up time.

Table 2. Six numerical ~~experiments~~ simulations are conducted in this study.

Scenarios	Description
Jan_Base	Baseline simulation in January
Jan_noCD	Replacing urban land use of Chengdu with cropland in January
Jan_noEmi	Shutting down anthropogenic emissions in Chengdu in January
July_Base	Baseline simulation in July
July_noCD	Replacing urban land use of Chengdu with cropland in July
July_noEmi	Shutting down anthropogenic emissions in Chengdu in July

2.3 Health risks estimation

Daily premature mortalities attributable~~ed~~ to ~~–~~$PM_{2.5}$ and O_3 exposure from all non-accidental causes (ANAC), cardiovascular diseases (CVD), respiratory diseases (RD) and chronic obstructive pulmonary diseases (COPD) are estimated using the standard damage function (Anenberg et al., 2010; Zhan et al., 2021):

$$\Delta M = y_0 (\frac{RR-1}{RR}) \text{Pop}, \tag{4}$$

where ΔM is the daily premature mortality, y_0 is the daily baseline mortality rate, RR is the relative risk, $(RR-1)/RR$ is the attributable fraction, and Pop is the exposed population. RR is calculated as follows:

$$RR = \exp(\beta(\mathrm{C} - \mathrm{C}_0)), \tag{5}$$

where β is the concentration-response function that relates a unit change in air pollutant concentrations to a change in health endpoint incidence~~concentration-response function~~. In practice, β represents the percent change of mortality per 10 μg m^{-3} increase of daily $PM_{2.5}$/MDA8 O_3 concentration with 95% confidence intervals (95% CI)~~, which represents the percentage increase in health effect per 1 μg m^{-3} $PM_{2.5}$ and MDA8 O_3 increment~~. C is the exposure concentration, which is the daily average concentration for $PM_{2.5}$ and the MDA8 O_3 concentration for O_3. ~~and~~ C_0 is the

threshold concentration. When C is not greater than C_0, the value of $C–C_0$ is 0.

In this study, C_0 ~~for $PM_{2.5}$~~ is 10 μg m^{-3} for daily $PM_{2.5}$ (Song et al., 2015), and~~, and~~ ~~for MDA8 O_3 is~~ 75.2 μg m^{-3} for MDA8 O_3 (Liu et al., 2018). ~~The~~ β and y_0 ~~values~~ for ANAC, CVD, RD and COPD are summarized in Table 3 (Chen et al., 2017; Yin et al., 2017). The populations of Chengdu provided by the National Bureau of Statistics of China are 16.853 million, 18.582 million, 19.188 million, 19.183 million, 20.409 million, 20.947 million and 20.938 million from 2015 to 2021.~~–~~

We first calculate the $PM_{2.5}$- and O_3-induced daily premature mortalities using the methods mentioned above, and then add up the daily premature mortalities for the whole year/month to get the total premature mortalities. Since the largest uncertainty among the factors that determine premature mortalities usually comes from β, premature mortalities are presented as means and 95% CI based on β at 95% CI in this study. In addition, it should be noted that we use the average of air pollutant concentration at all monitoring sites to represent air pollutant concentration in Chengdu. Correspondingly, the total population of Chengdu is used as the exposed population. So our results are for Chengdu as a whole and do not address the spatial distribution of premature mortalities.

Table 3. Daily β (95%CI) and y_0 values for ANAC, CVD, RD and COPD. ~~This table is cited from Wang et al. (2021).~~

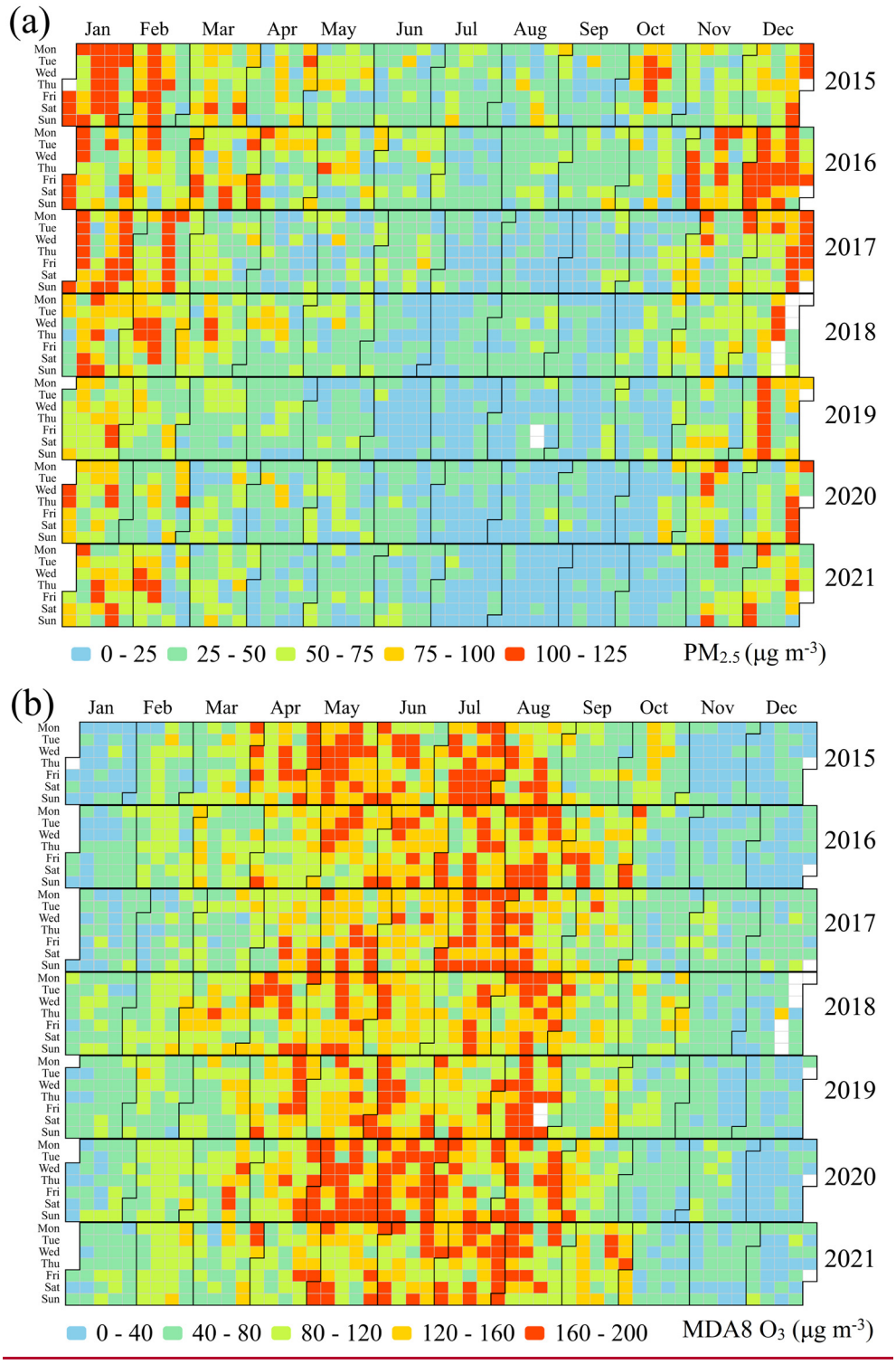
Disease	β for $PM_{2.5}$ (%)	β for MDA8 O_3 (%)	y_0
ANAC	0.22 (0.15, 0.28)	0.24 (0.13, 0.35)	1.687×10^{-5}
CVD	0.27 (0.18, 0.36)	0.27 (0.10, 0.44)	3.880×10^{-6}
RD	0.29 (0.17, 0.42)	0.18 (–0.11, 0.47)	1.841×10^{-6}
COPD	0.38 (0.23, 0.53)	0.20 (–0.13, 0.53)	1.623×10^{-6}

3 Results and discussions

3.1 $PM_{2.5}$ and O_3 pollution in Chengdu

~~$PM_{2.5}$ and O_3 are two crucial air pollutants~~According to Chinese ambient air quality standards, $PM_{2.5}$ pollution occurs when daily $PM_{2.5}$ concentrations are greater than 75 μg m^{-3}, and O_3 pollution occurs when MDA8 O_3 concentrations are greater than 160 μg m^{-3} ~~that account for air pollution~~. As shown in Figure 2, Chengdu is suffering from severe $PM_{2.5}$ and O_3 pollution in recent years. ~~The~~

~~Chinese ambient air quality standards for $PM_{2.5}$ and MDA8 O_3 are 75 μg m^{-3} and 160 μg m^{-3}, respectively. T~~~~As shown in Figure 2, Chengdu is suffering from severe $PM_{2.5}$ and O_3 pollution in recent years. T~~~~here were~~~~are~~ 97, 101, 68, 53, 33, 43 and 37 $PM_{2.5}$ pollution episodes, and 61, 48, 42, 40, 42~~26~~, 71~~50~~ and 48~~27~~ O_3 pollution episodes in Chengdu from 2015 to 2021. In China, the annual evaluation criterion for $PM_{2.5}$ is the annual average concentration, and it for O_3 is the 90th percentile of MDA8 O_3 concentration. The annual average concentrations of $PM_{2.5}$ were~~are~~ 60.7, 59.9, 52.6, 47.2, 40.6, 40.8 and 40.1 μg m^{-3}, and ~~those of~~ the 90th percentile of MDA8 O_3 concentrations were ~~MDA8 O_3 are~~ 183.0~~95.3~~, 167.0~~96.4~~, 168.0~~95.8~~, 164.0~~101.3~~, 171.5~~86.8~~, 188.9~~92.0~~ and 167.1~~89.6~~ μg m^{-3} in Chengdu from 2015 to 2021~~, respectively~~. This suggests that ~~In terms of the annual average concentrations,~~ $PM_{2.5}$ pollution had~~ has~~ improved significantly while O_3 pollution had not~~has not~~. O_3 pollution control in Chengdu should be taken seriously in the future. In addition, $PM_{2.5}$ and O_3 pollution ~~have~~ had clear seasonal preferences, that was~~is~~, $PM_{2.5}$ pollution tended~~s~~ to appear in cold months (November to February) while O_3 pollution preferred~~s~~ to appear in warm months (April to August). High $PM_{2.5}$ concentrations in cold months may be associated with the consumption of fossil fuels for heating and frequent temperature inversion. The h~~The h~~igh temperature and strong sunlight contribute to the elevated O_3 concentrations in warm months~~Elevated O_3 concentrations in warm months are contributed to the high temperature and strong sunlight~~.


(a)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Mon Tue Wed Thu Fri Sat Sun
2015
2016
2017
2018
2019
2020
2021
0 - 25
25 - 50
50 - 75
75 - 100
100 - 125
PM2.5 (μg m-3)
(b)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Mon Tue Wed Thu Fri Sat Sun
2015
2016
2017
2018
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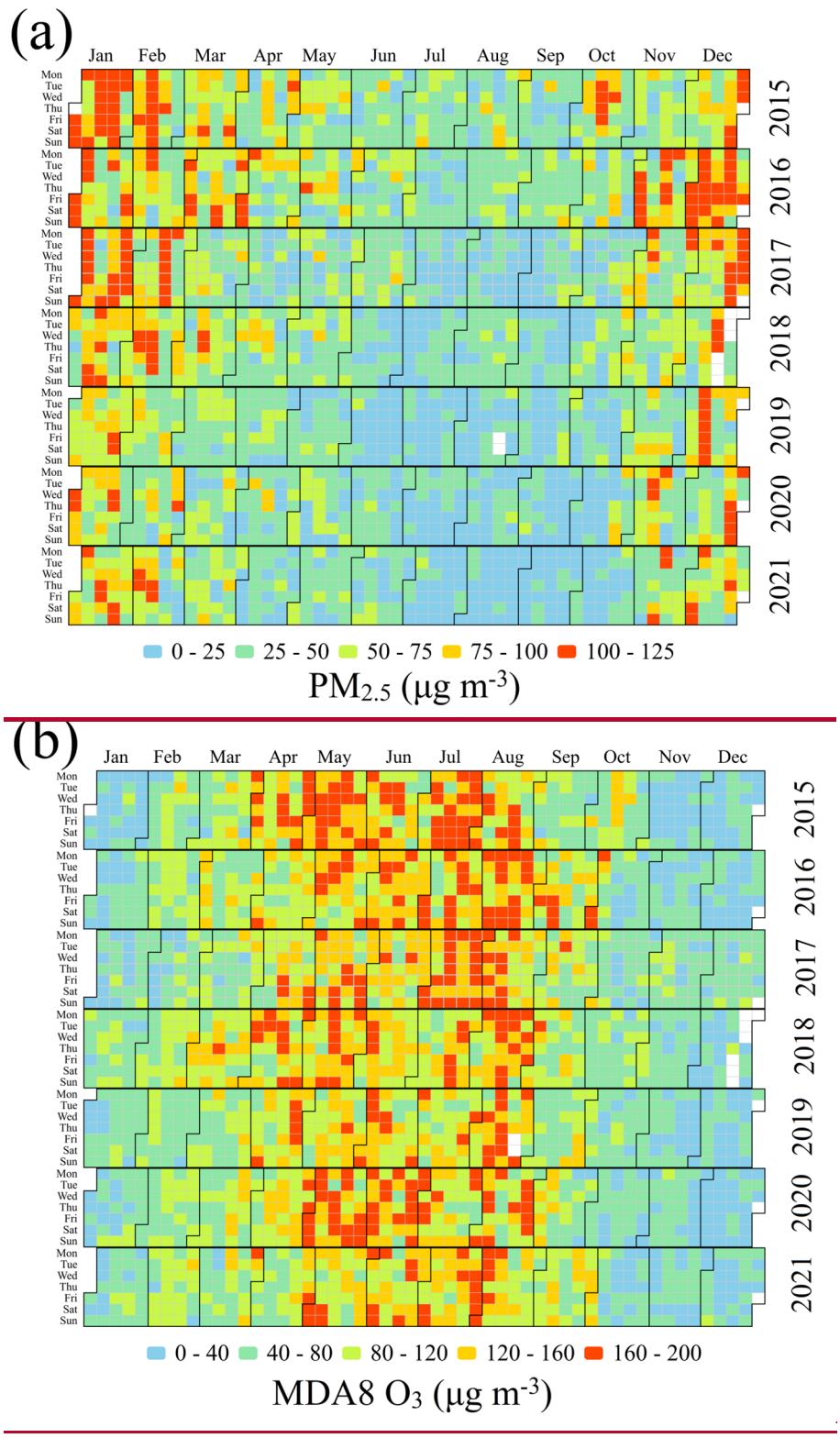
2021
0 - 40
40 - 80
80 - 120
120 - 160
160 - 200
MDA8 O3 (μg m-3)


(a)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Mon Tue Wed Thu Fri Sat Sun
2015 2016 2017 2018 2019 2020 2021
0 - 25 25 - 50 50 - 75 75 - 100 100 - 125
PM2.5 (μg m-3)
(b)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Mon Tue Wed Thu Fri Sat Sun
2015 2016 2017 2018 2019 2020 2021
0 - 40 40 - 80 80 - 120 120 - 160 160 - 200
MDA8 O3 (μg m-3)

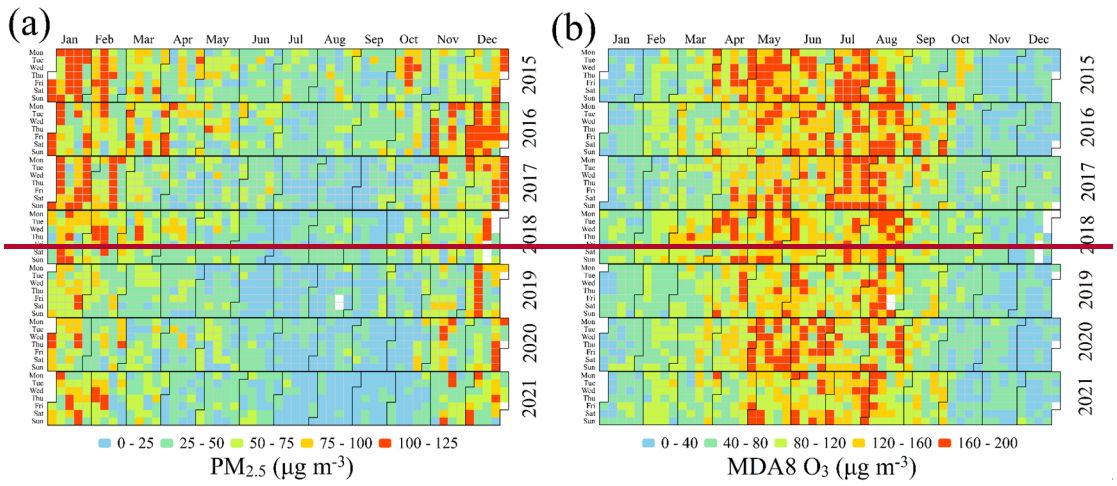


Figure 2. Heat maps of~~Distribution of~~ (a) daily ~~average~~ $PM_{2.5}$ and (b) MDA8 O_3 concentrations in Chengdu from 2015 to 2021.

3.2 Premature mortality attributable~~buted~~ to $PM_{2.5}$ and O_3

Severe $PM_{2.5}$ and O_3 pollution are responsible for a ~~considerable~~ large number of premature mortalities~~mortalities~~ in Chengdu. From 2015 to 2021~~ As shown in Table 4~~, the premature mortalities from ANAC due~~attributed~~ to $PM_{2.5}$ were~~exposure are~~ 10596 (95%CI: 7420~13186), 11647 (95%CI: 8140~14518), 10154 (95%CI: 7116~12630), 8942 (95%CI: 6214~11198), 7992 (95%CI: 5540~10031)~~3~~, 8298 (95%CI: 5759~10402) and 8072 (95%CI: 5606~10115)~~from 2015 to 2021~~, with the 7-year~~an~~ annual average of 9386 (95%CI: 6542~11726). The highest health risk among the diseases was~~is~~ from CVD with a 7-year~~an~~ annual average of 2609 (95%CI: 1788~3384), followed by COPD with a 7-year~~an~~ annual average of 1485 (95%CI: 941~1983) and RD with a 7-year~~an~~ annual average of 1321 (95%CI: 804~1840). This was mainly associated with the daily baseline mortality rate of different diseases (Table 3). ~~Due to urbanization and administrative division adjustment,~~Although Chengdu's population has been increasing~~es by 24.2%~~ by 24.2% from 2015 ~~(16.853 million)~~ to 2021~~(20.938 million),~~ ~~In contrast, the total~~ premature mortalities ~~attributed to~~ due to $PM_{2.5}$ have generally declined (Figure 3a)~~falls by 23.8% from 2015 (10596) to 2021 (8072). The reduction in premature mortalities is mainly contributed to the reduction in $PM_{2.5}$ concentrations since the annual average $PM_{2.5}$ concentrations decrease from 60.7 μg m^{-3} in 2015 to 40.1 μg m^{-3} in 2021 (Section 3.1), suggesting that air pollution control can bring huge health benefits.~~ owing to reduced $PM_{2.5}$ concentrations in recent years (Section 3.1).

~~Unlike the overall reduction in premature mortalities due to $PM_{2.5}$, the premature mortalities~~

~~due to O_3 fluctuate.~~ The premature mortalities from ANAC ~~attributed to O_3 exposure are~~ due to O_3 were 7657 (95%CI: 4345~10672), 8025 (95%CI: 4537~11227), 7870 (95%CI: 4451~11005), 8824 (95%CI: 4967~12397)~~8556~~, 7919 (95%CI: 4483~11065)~~6367~~, 10085 (95%CI: 5749~13999)~~8300~~ and 9163 (95%CI: 5185~12809)~~7429~~ from 2015 to 2021, with a 7-year~~an~~ annual average of 8506 (95%CI: 4817~11882)~~7743~~, about 9~~8~~0% of that due~~attributed~~ to $PM_{2.5}$~~ exposure~~. ~~The total premature mortalities attributed to O_3 exposure in 2021 (7429) are only 3.0% lower than that in 2015 (7657). This is in line with the insignificant reduction of O_3 concentrations in Chengdu from 2015 to 2021, indicating that O_3 pollution control in Chengdu still has great potential and significance.~~ Unlike the overall reduction in premature mortalities due to $PM_{2.5}$, the premature mortalities due to O_3 increased slightly (Figure 3a), further indicating the urgent need for powerful O_3 control strategies in Chengdu.

~~**Table 4.** Premature mortality from ANAC, CVD, RD and COPD attributed to $PM_{2.5}$ and O_3 exposure.~~

~~Year~~	~~$PM_{2.5}$~~				~~MDA8 O_3~~			
	~~ANAC~~	~~CVD~~	~~RD~~	~~COPD~~	~~ANAC~~	~~CVD~~	~~RD~~	~~COPD~~
~~2015~~	~~10596~~	~~2935~~	~~1485~~	~~1660~~	~~7657~~	~~1957~~	~~643~~	~~624~~
~~2016~~	~~11647~~	~~3231~~	~~1635~~	~~1832~~	~~8025~~	~~2053~~	~~672~~	~~653~~
~~2017~~	~~10154~~	~~2812~~	~~1422~~	~~1589~~	~~7870~~	~~2013~~	~~659~~	~~641~~
~~2018~~	~~8942~~	~~2490~~	~~1262~~	~~1424~~	~~8556~~	~~2191~~	~~715~~	~~696~~
~~2019~~	~~7993~~	~~2230~~	~~1131~~	~~1280~~	~~6367~~	~~1630~~	~~532~~	~~518~~
~~2020~~	~~8298~~	~~2313~~	~~1173~~	~~1325~~	~~8300~~	~~2121~~	~~696~~	~~677~~
~~2021~~	~~8072~~	~~2249~~	~~1140~~	~~1287~~	~~7429~~	~~1902~~	~~621~~	~~604~~

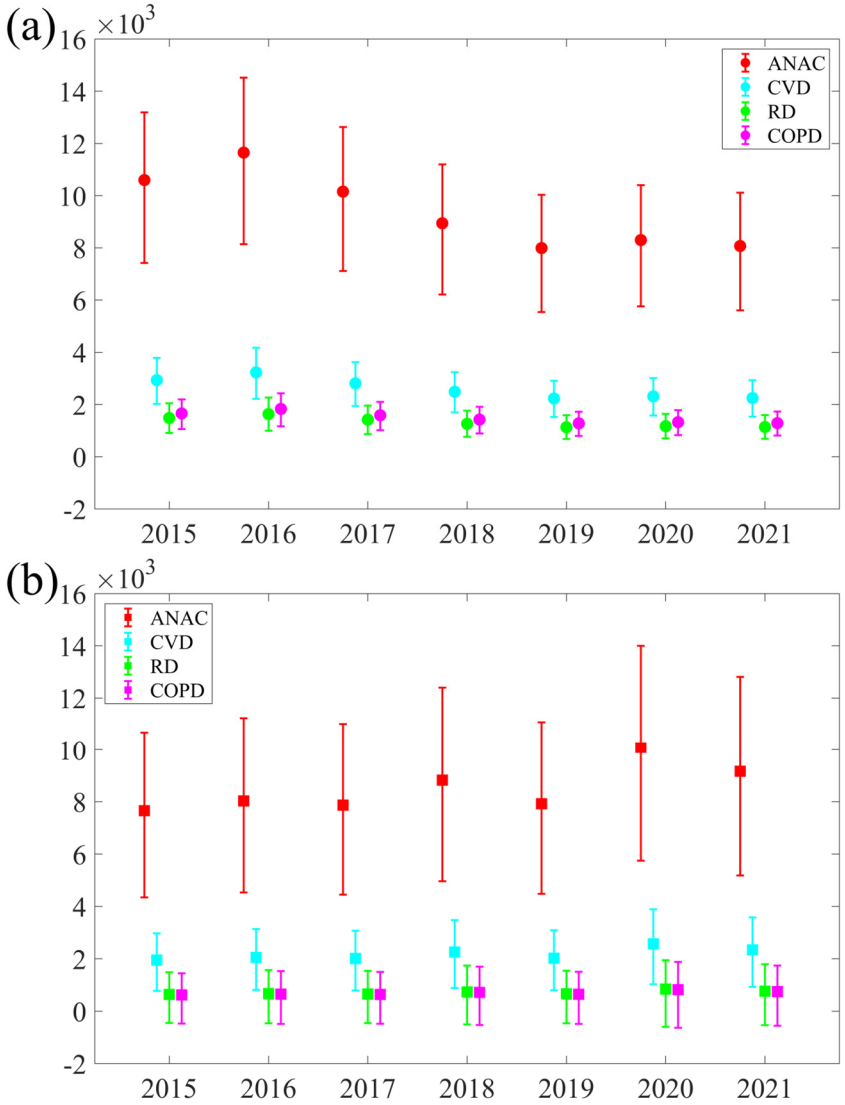


Figure 3. Premature mortality from ANA, CVD, RD and COPD attributable to $PM_{2.5}$ and O_3 in Chengdu from 2015 to 2021.

3.3 Impacts of urbanization on $PM_{2.5}$ and O_3

3.3.1 Meteorological conditions in January and July

In this study, January and July 2017, when ~~–~~$PM_{2.5}$ and O_3 pollution episodes are likely to occur (Figure 2), are ~~are chosen~~ selected~~for simulations~~ to study the role of urbanization~~and analysis when $PM_{2.5}$ and O_3 pollution are likely to occur (Figure 2)~~. In January 2017, Chengdu experienced~~s~~ $PM_{2.5}$ pollution for 23 out of 31 days with a monthly~~an~~ average concentration of 128.8~~5~~ μg m^{-3}. From the perspective of atmospheric circulations, westerly winds prevailed over Chengdu due to the large north-south geopotential height gradient at 500 hPa (Figure 4~~3~~a). However, the~~ cold~~ westerly winds

were ~~from the north are~~ blocked by the Tibetan Plateau and thereby the elimination of $PM_{2.5}$ was limited. ~~Instead~~ At 700 hPa, ~~, a low-pressure system, called the Southwest Vortex, appears to the left of~~the southwestern air flow originating from the Bay of Bengal could reach Chengdu (Figure 4b ~~Chengdu at 850 hPa (Figure 3b~~). ~~Warm and humid southerly air flows can reach Chengdu affected by this low-pressure system~~This warm advection was conducive to the formation of an inverse layer near 700 hPa (Figure 4c and d), which made the vertical diffusion of $PM_{2.5}$ difficult. ~~Hu et al., 2021; Ning et al., 2018). The dry air in the upper layer and moist air in the lower layer lead to a strong temperature inversion appearing from 700 hPa to 500 hPa (Figure 4a and b).~~ The blocking of ~~clod~~ air and the temperature inversion ~~make $PM_{2.5}$ pollution frequent during this period.~~ were two important reasons for frequent $PM_{2.5}$ pollution episodes during this period (Hu et al., 2021; Ning et al., 2018).

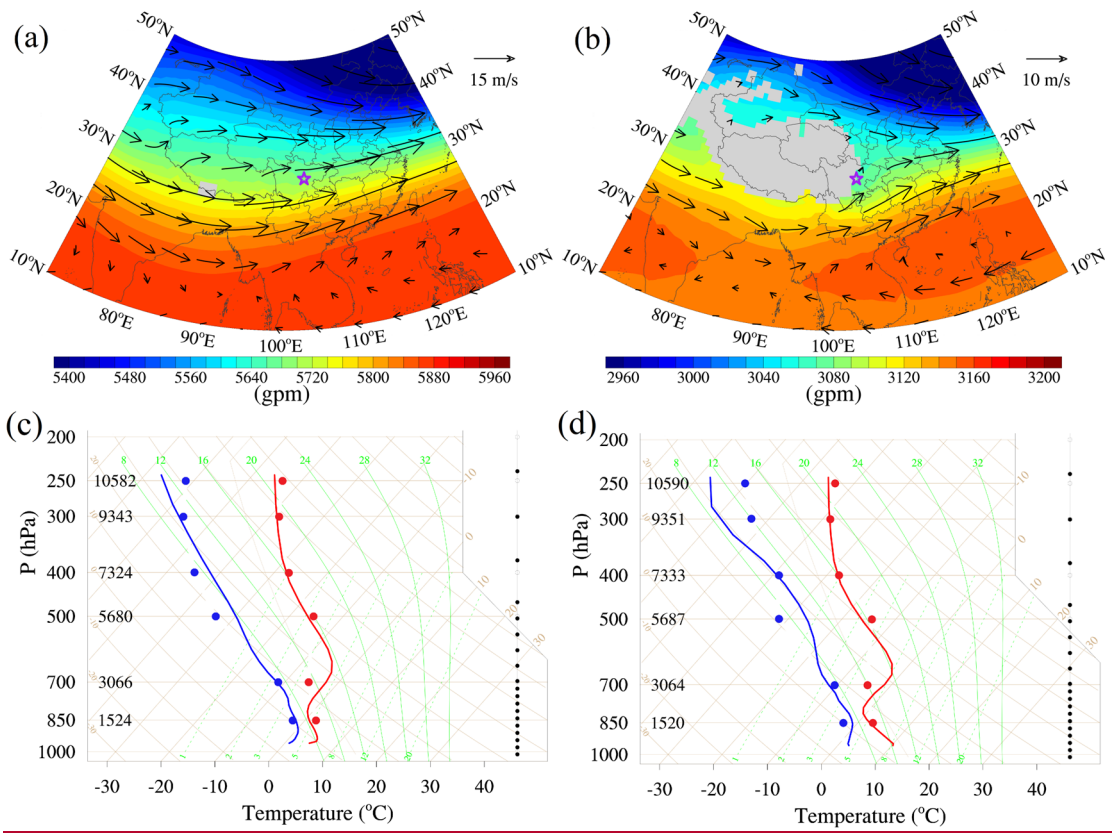


Figure 4. The weather charts at (a) 500 hPa and (b) 700 hPa for January 2017 are based on the NCEP FNL reanalysis data. The purple pentacles show the location of Chengdu. The skew-T diagram at (c) 00:00 UTC and (d) 12:00 UTC in January 2017. The red and blue lines are the simulated air temperature and dew point temperature in Jan_Base simulation, while the

red and blue points are the sounding temperature and dew point temperature. These results are monthly averages.

In July 2017, there were~~are~~ 19 days of O_3 pollution in Chengdu, and the monthly average MDA8 O_3 concentration was~~is~~ 172.9 μg m^{-3}. At 500 hPa, Chengdu was~~is~~ dominated by strong high-pressure systems, and thereby air temperature was~~is~~ high and wind speed was~~is~~ small (Figure 5~~3c~~a). The monthly average T_2 was~~is~~ as high as 28.6 °C while the monthly average WS_{10} was~~is~~ only 1.6 m s^{-1} during this period (Figure 5b)~~in July~~. ~~These meteorological conditions are conducive to the formation of O_3 pollution.~~High temperature favored photochemical reactions of O_3 while weak winds trapped O_3. Furthermore, the frequency and thickness of temperature inversion in July were~~are~~ far less than those in January (Figure 4c and d; Figure 5c and d~~4~~). Well-developed boundary layer facilitated vertical mixing of~~Then~~ O_3 ~~can be well mixed~~ within the boundary layer~~mixing layer during the daytime~~, which is an important way to maintain high surface O_3 concentrations during the daytime (Aneja et al., 2000; Tang et al., 2017).

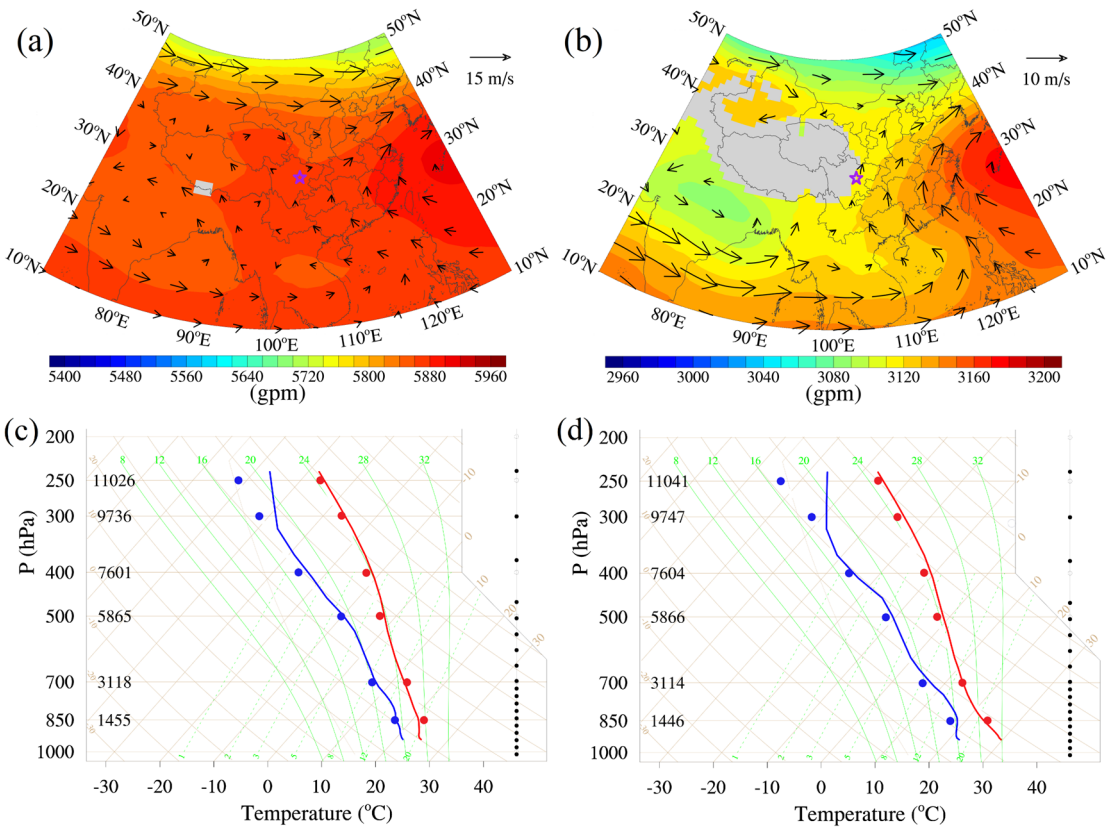
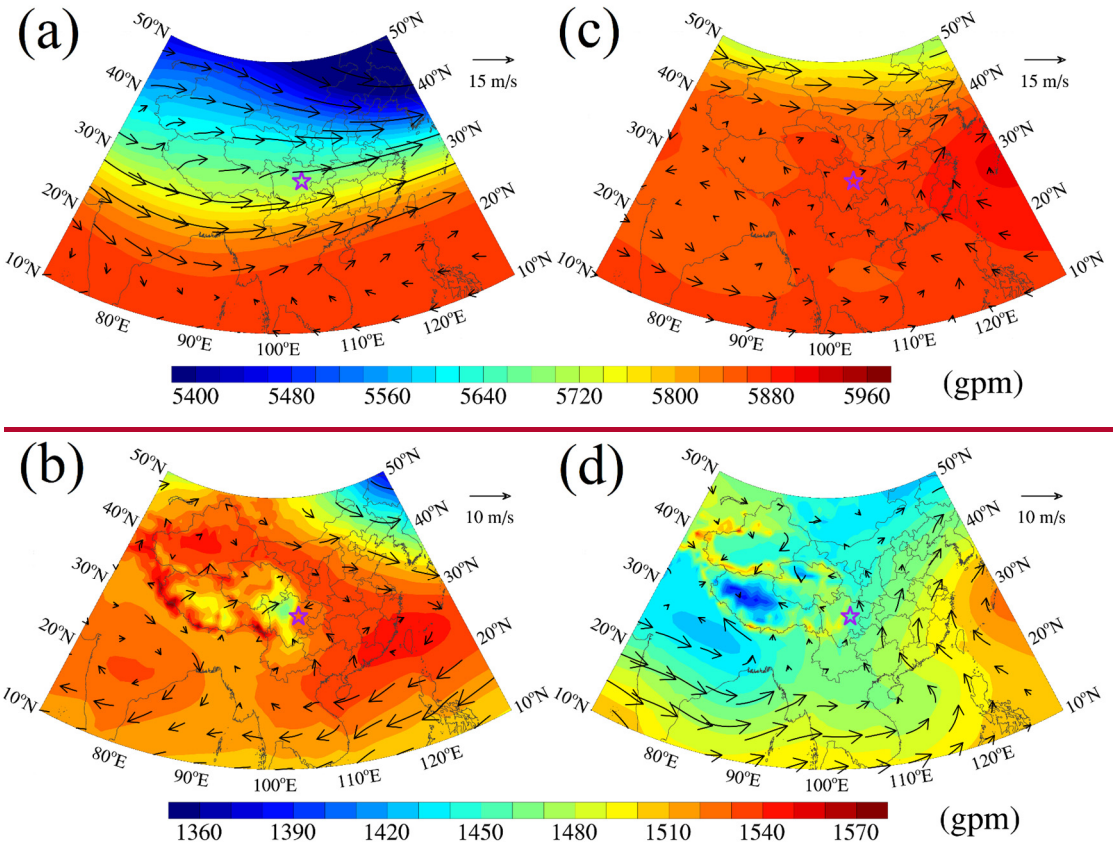
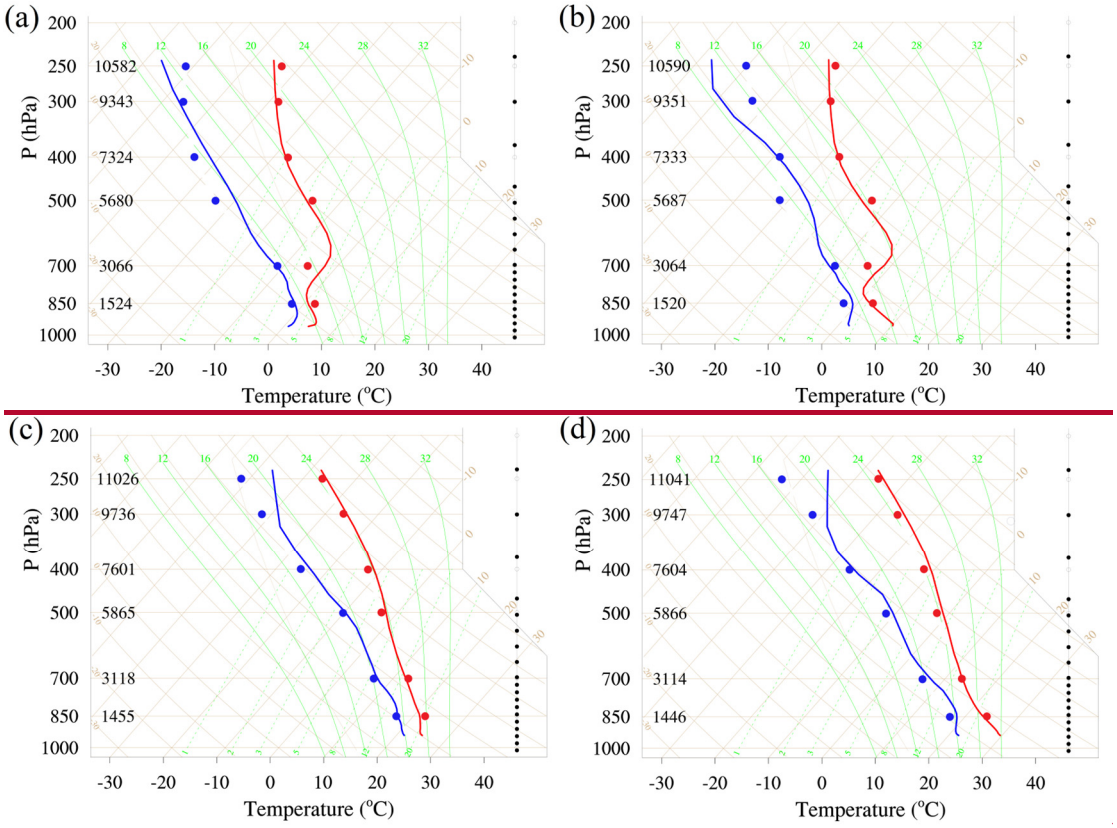
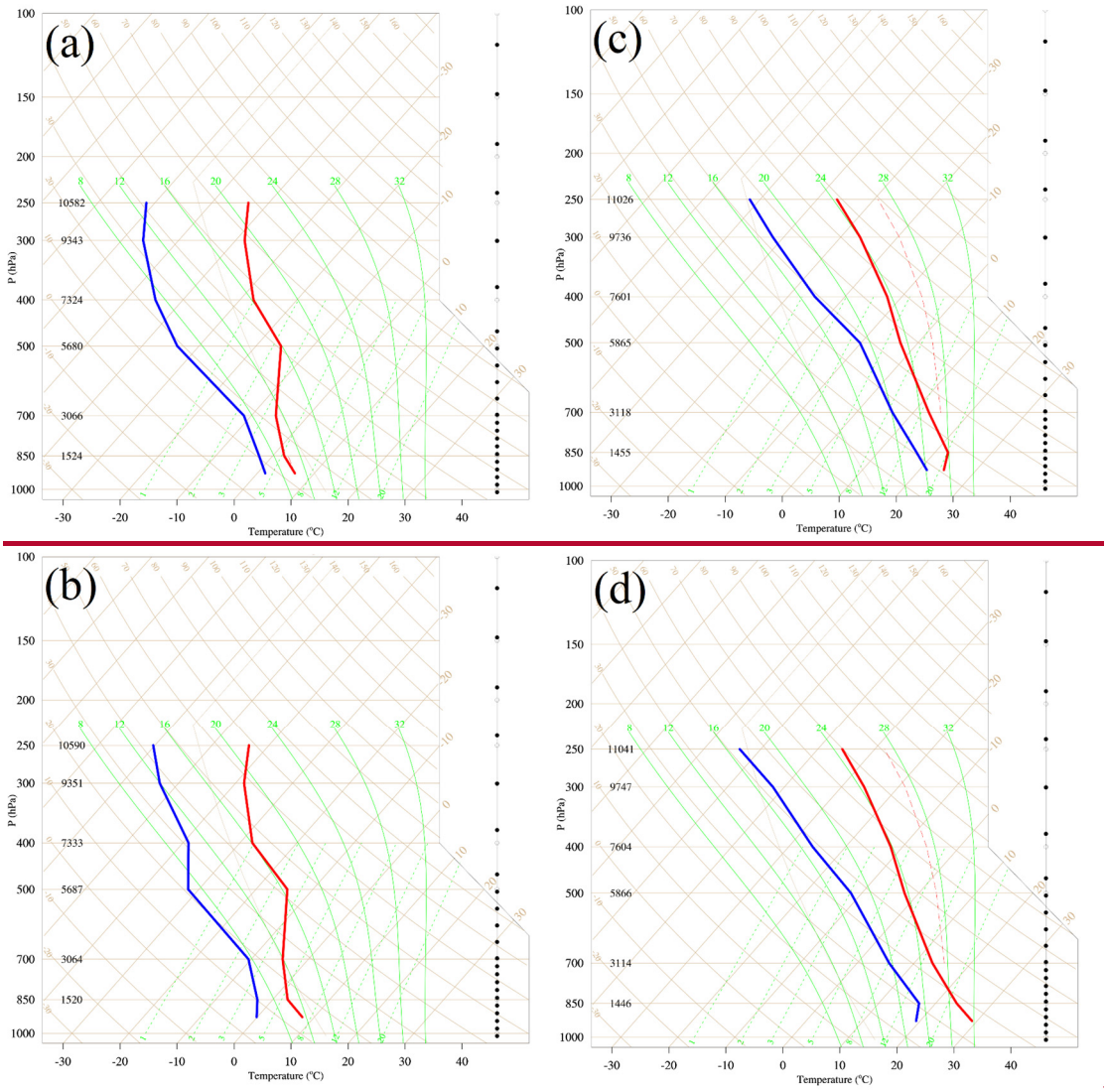


Figure 5. The weather charts at (a) 500 hPa and (b) 700 hPa for July 2017 are based on the NCEP FNL reanalysis data. The purple pentacles show the location of Chengdu. The skew-T diagram at (c) 00:00 UTC and (d) 12:00 UTC in July 2017. The red and blue lines are the simulated air temperature and dew point temperature in July Base simulation, while the red and blue points are the sounding temperature and dew point temperature. These results are monthly averages.



~~**Figure 3. The weather charts at (a) 500 hPa and (b) 850 hPa for January 2017. (c) and (d) are the same as (a) and (b), but for July 2017. The purple pentacles show the location of Chengdu. These weather charts are based on the NCEP global final analysis data.**~~


(a)
P (hPa)
200
250
300
400
500
700
850
1000
10582
9343
7324
5680
3066
1524
-30 -20 -10 0 10 20 30 40
Temperature (°C)
(b)
10590
9351
7333
5687
3064
1520
(c)
11026
9736
7601
5865
3118
1455
(d)
11041
9747
7604
5866
3114
1446

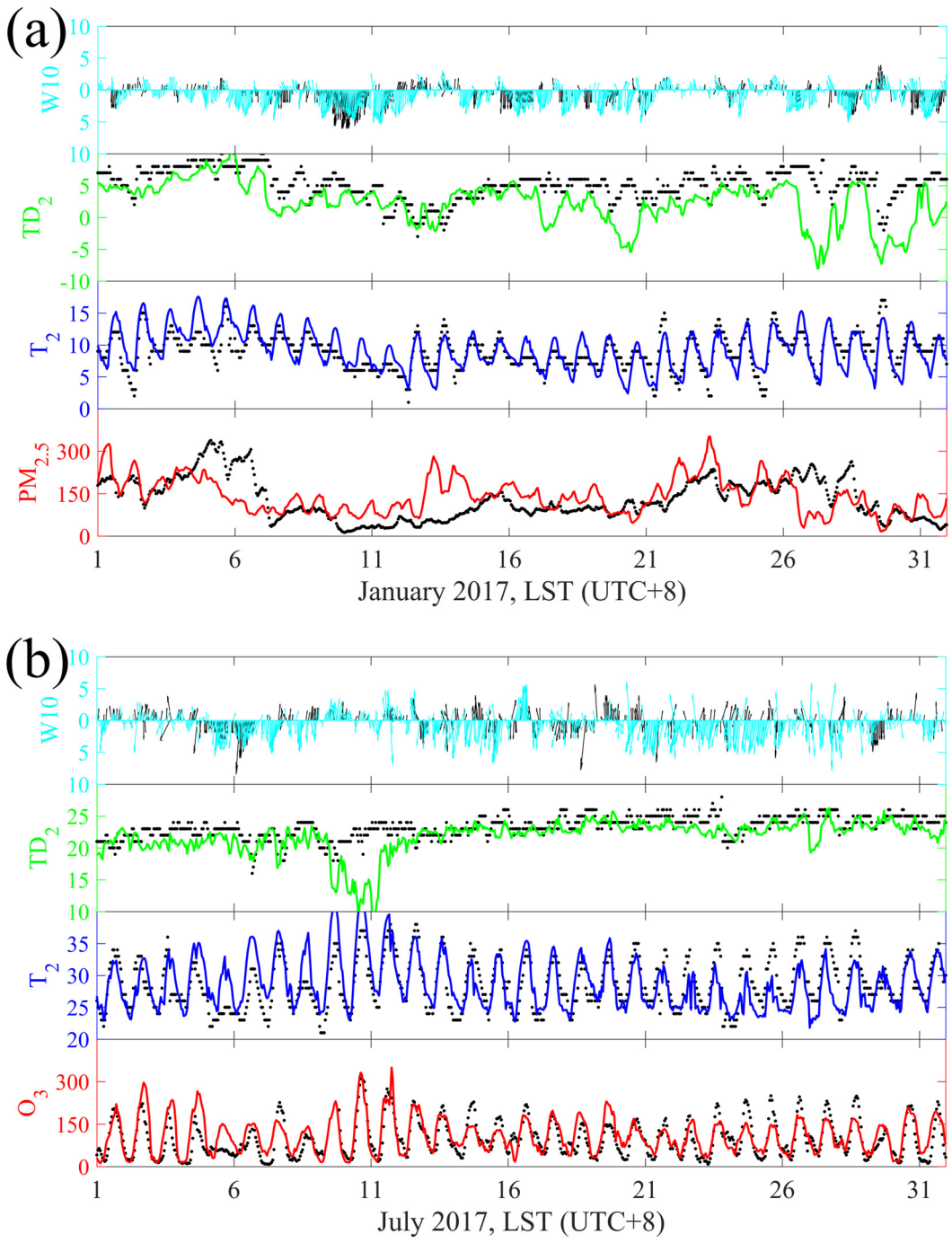


~~**Figure 4. The skew-T diagram at (a) 00:00 UTC and (b) 12:00 UTC in January 2017. (c) and (d) are the same as (a) and (b), but for July 2017. The red and blue lines indicate air temperature and dew point temperature, respectively.are the simulated air temperature and dew point temperature, while the red and blue points are the sounding temperature and dew point temperature.**~~

3.3.2 Evaluation of model performance

We first compare vertical profiles in the model with the sounding data to determine whether the model captures the vertical structure of the tropospheric atmosphere. As shown in Figure 4c-d and 5c-d, the WRF-Chem model can successfully simulate the changes in temperature and dew point temperature in the vertical direction, whether in January or July, day or night. Therefore, the vertical results in the model are reliable. Furthermore, simulations at the lowest model level are

compared with surface observations, and the results are presented in Figure 6. The mean bias (MB) of the simulated and observed concentrations of~~Simulated $PM_{2.5}$ concentrations, O_3 concentrations, air temperature, relative humidity and wind speed in baseline simulations are compared with the observations to verify the model performance (Figure 5). The magnitudes of simulated~~ $PM_{2.5}$ and O_3 are~~ reasonable with the small mean bias (MB) of~~ 12.7~~23.4~~ μg m^{-3} and 11.6 μg m^{-3}, with the normalized mean bias (NMB) values of 9.9% and 12.0%, which are within the acceptable standards (NMB < ± 15%)~~respectively~~. The~~ high~~ correlation coefficients (COR) of~~for~~ $PM_{2.5}$ (0.44) and O_3 (0.77) ~~indicate that simulations reproduce well the diurnal variation in pollutants~~are 0.44 and 0.77, respectively. The statistical metrics for $PM_{2.5}$ and O_3 are similar to those in previous studies (Wang et al., 2022b; Wu et al., 2022), indicating that~~Therefore,~~ our~~the~~ model~~ing~~ results for $PM_{2.5}$ and O_3 are~~ generally~~ reasonable and acceptable. With regard to the meteorological factors, T_2 is well simulated with low MB (0.2 and 0.1 °C) and high COR (0.76 and 0.70) values in both January and July. The simulations~~Our simulation~~ underestimate~~s~~ TD_2~~RH~~ to some extent with ~~(~~the MB values are ~~-14.3%~~ –1.5 °C and –2.6 °C~~-4.8%~~ in January and July, respectively~~), but the diurnal variation of RH is well represented (the COR values are 0.54 and 0.64)~~. As for 10-m wind~~WS_{10}~~, poor simulation results are predictable in the case of low wind and complex terrain. The WS_{10} in the model is overestimated ~~(the MB values are 1.3 and 1.7 m s^{-1})~~as in the studies of other scholars (Shu et al., 2021; Wu et al., 2022). It could~~, which may be related to the unresolved terrain features by the default surface drag parameterization causing an overestimate of wind speed in particular at low values~~be argued that the unresolved topographic features produce an additional drag to that generated by vegetation, but their effects are not considered in WRF (Jimenez and Dudhia, 2012)~~Jimenez and Dudhia, 2012)~~. The model, on the other hand, captures the shift in wind direction. ~~Due to the small change in weak wind, the COR for WS_{10} is not high.~~ To sum up~~In general~~, the WRF-Chem model using our configuration has a good capability in simulating $PM_{2.5}$, O_3~~air pollutants~~ and meteorological factors in Chengdu, and thereby the simulations can be used for subsequent analysis.


(a)
W10
TD2
T2
PM2.5
January 2017, LST (UTC+8)
(b)
W10
TD2
T2
O3
July 2017, LST (UTC+8)

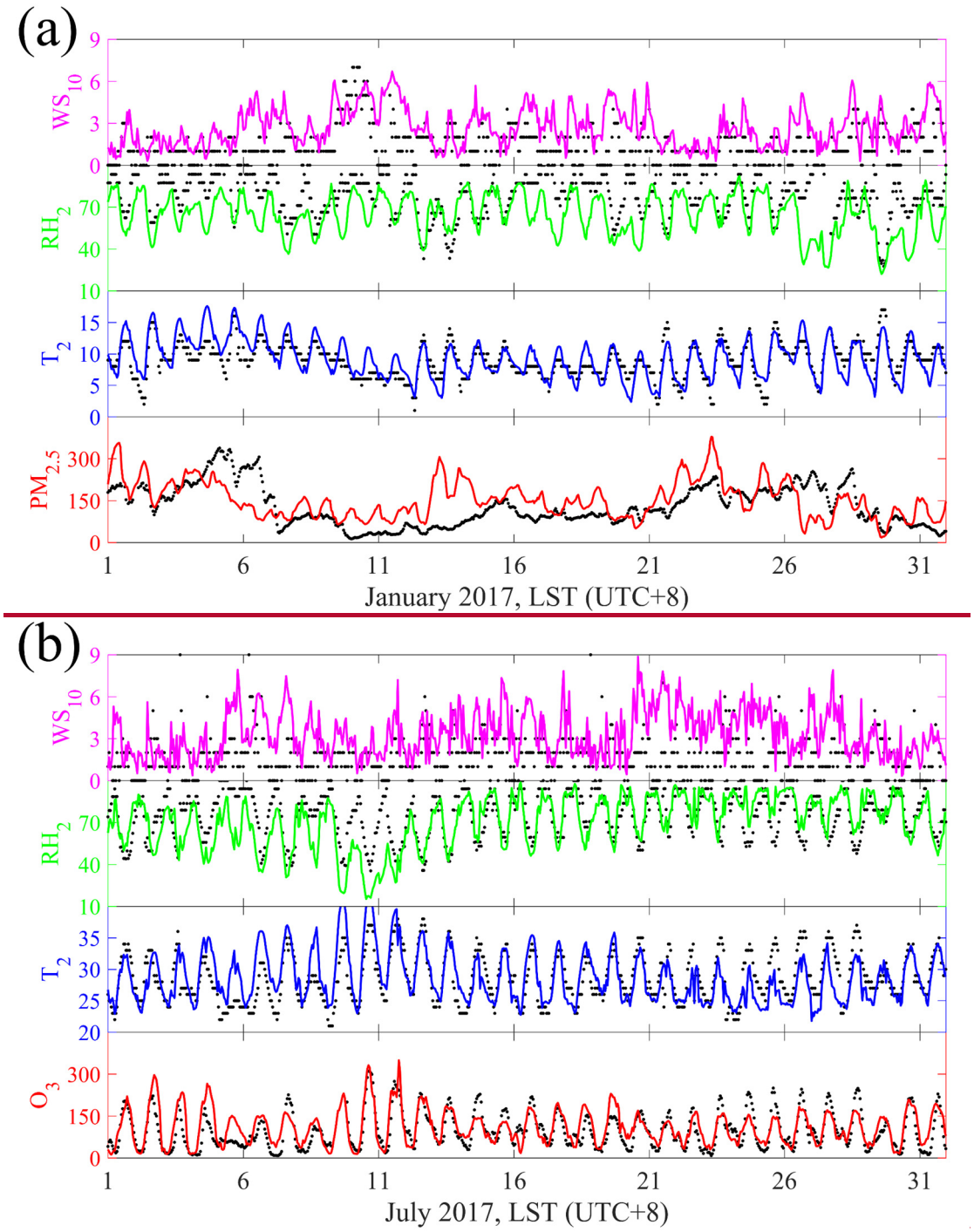
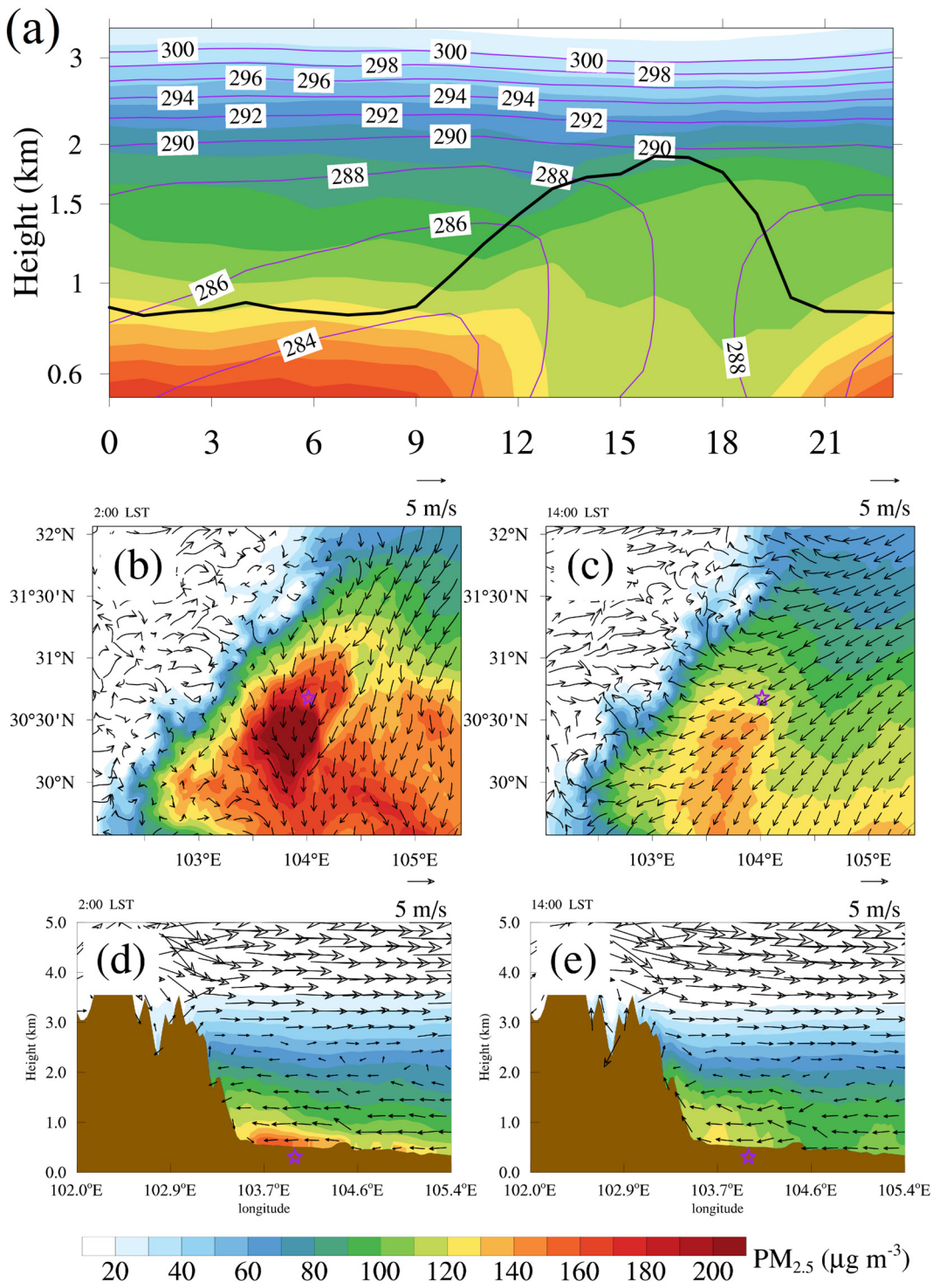


Figure 6~~5~~. Times series of $PM_{2.5}$~~, O_3~~, T_2, TD_2~~RH~~ and 10-m wind for (a) January 2017~~WS_{10} for observations (black dots) and baseline simulations (colored lines)~~. And times series of O_3, T_2, TD_2 and 10-m wind for (b) July 2017. The black dots are observations and the colored lines are simulations in baseline simulations.

3.3.3 Spatiotemporal variations in $PM_{2.5}$ and O_3

~~Figure 6 shows the January-averaged spatiotemporal distribution of $PM_{2.5}$ in~~ The spatiotemporal characteristics of $PM_{2.5}$ were first investigated based on the Jan_Base simulation. $PM_{2.5}$ had~~s~~ a diurnal variation with high concentration at night and low concentration at noon, which was~~is~~ contrary to ~~the diurnal variation of~~ the boundary layer height (Figure 7a). The nocturnal atmospheric boundary layer was often characterized by a stable boundary layer~~At night~~, and~~,~~ the boundary layer height was~~is usually~~ only ~320 m~~low~~. ~~,~~ A~~A~~s a consequence, $PM_{2.5}$ was~~is~~ trapped and maintained on the ground. The daytime atmospheric boundary layer, also known as the convective boundary layer, could develop to ~1300 m.~~At noon,~~ T~~t~~urbulence in the convective boundary layer could~~an~~ dilute $PM_{2.5}$ concentrations, ~~through vertical mixing,~~ resulting in low $PM_{2.5}$ concentrations at surface. Chengdu is on the east side of the Tibetan Plateau, with a large elevation drop exceeding 3000 m over a short horizontal distance (Figure 1a). In this case, the mountain-plain~~valley~~ ~~breezes~~ wind can easily ~~develop~~ form~~in winter when atmospheric conditions are usually stagnant.~~ ~~,~~ During nighttime, the mountain wind was characterized by westerly and downslope flow at lower levels along the eastern slope of the Tibetan Plateau. ~~and are crucial for $PM_{2.5}$ in Chengdu. The $PM_{2.5}$ pollution zone tends to appear in the converging airflows associated with the mountain breezes and can spread hundreds of kilometers (Figure 6a-d). Driven by the near-surface northeasterly winds~~Coupled with the prevailing northeasterly wind, $PM_{2.5}$ was~~is~~ uplifted over the eastern~~windward~~ slope of the Tibetan Plateau, and the $PM_{2.5}$ pollution zone could spread hundreds of kilometers (Figure 7b and d) ~~(Figure 6e-h)~~. The daytime plain wind was nearly a reversal of the nighttime circulation, with easterly and upslope ~~Then the uphill airflows are restrained and overturned below 3 km, forming a vertical secondary circulation over Chengdu. Governed by the secondary circulation forced by the complex terrain, the southwesterly winds at 3 km can transport $PM_{2.5}$ downward, which could replenish the surface $PM_{2.5}$ and facilitate the accumulation and maintenance of surface $PM_{2.5}$.~~flow over the Sichuan Basin (Figure 7c and e). The upslope flow could draw $PM_{2.5}$ to a higher elevation, which could also facilitate vertical dispersion of $PM_{2.5}$ during the day. ~~–~~


(a)
Height (km)
3
2
1.5
1
0.6
300
296
298
294
292
290
288
286
284
0
3
6
9
12
15
18
21
5 m/s
2:00 LST
14:00 LST
(b)
(c)
32°N
31°30'N
31°N
30°30'N
30°N
103°E
104°E
105°E
(d)
(e)
Height (km)
5.0
4.0
3.0
2.0
1.0
0.0
102.0°E
102.9°E
103.7°E
104.6°E
105.4°E
longitude
20
40
60
80
100
120
140
160
180
200
PM2.5 (μg m-3)

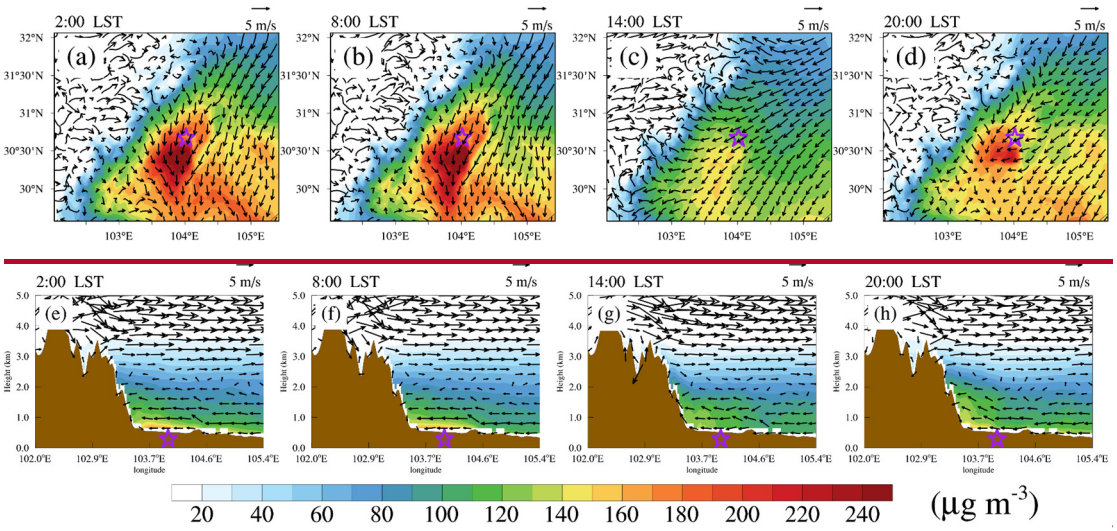
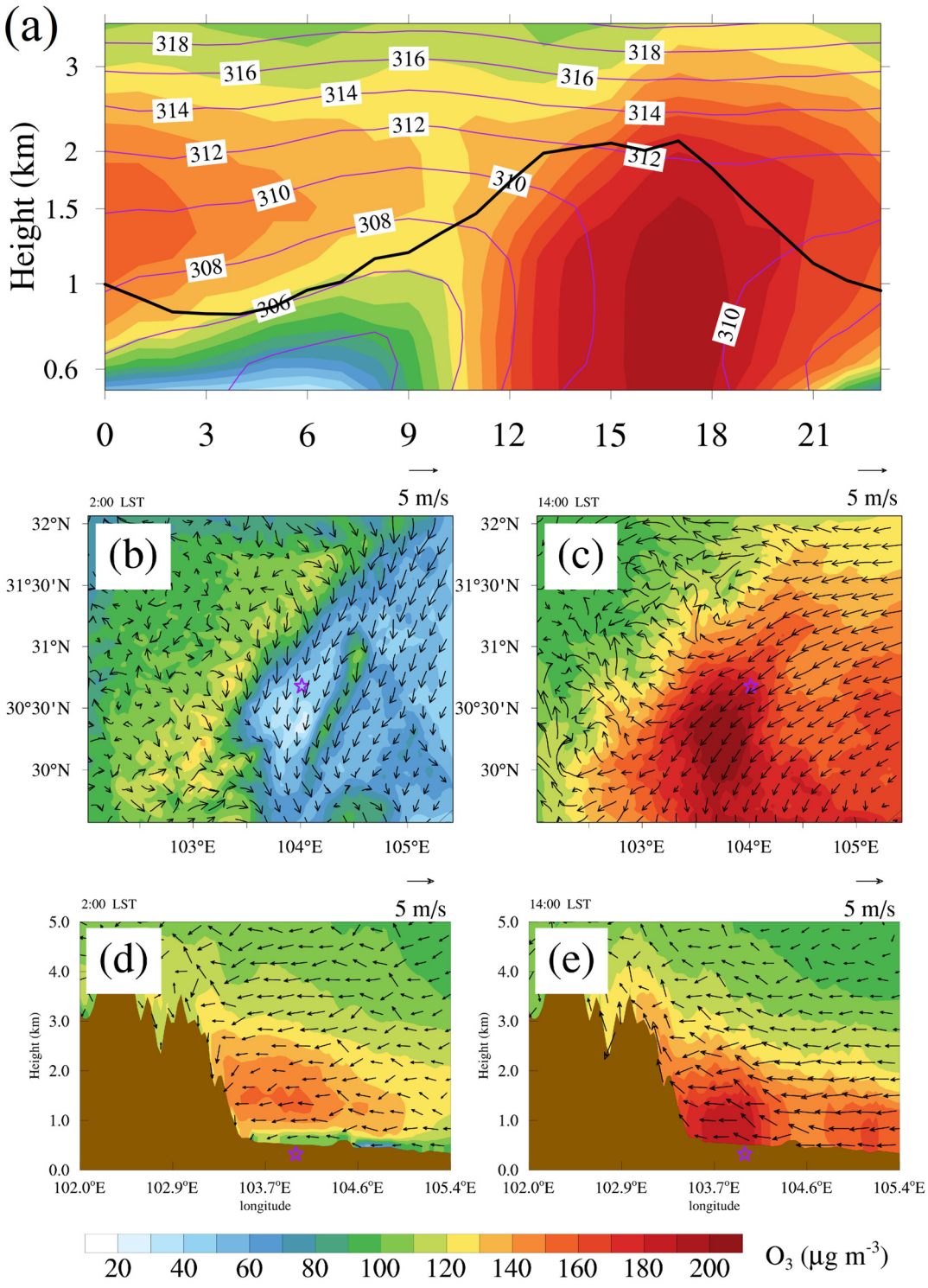


Figure 7~~6~~. ~~(a-d) Spatial distributions and (e-h) east-west vertical cross sections of $PM_{2.5}$ with wind fields at 2:00, 8:00, 14:00 and 20:00 LST (LST is UTC+8h) in Jan_Base simulation. Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain.~~ (a) Temporal-vertical cross sections of $PM_{2.5}$, potential temperature and boundary layer height at Chengdu. ~~(b-e)~~ Horizontal~~Spatial~~ distributions of $PM_{2.5}$ with wind vectors at the lowest model level at (b) 2:00 and (c) 14:00 LST. E~~and (d-e)~~ east-west vertical cross sections of $PM_{2.5}$ with wind vectors~~fields~~ at (d) 2:00 and (e) 14:00 LST ~~(LST is UTC+8h)~~. Purple pentacles show the locations of Chengdu. Brown-shaded areas represent the terrain. These results are the monthly average based on ~~from~~ Jan_Base simulation.

In terms of O_3, it exhibited~~s~~ strong diurnal variation with an afternoon maximum and an early morning minimum (Figure 8a~~7a-d~~). After sunrise, the nocturnal residual layer ~~is~~ was destroyed while the convective boundary layer ~~begins to form~~ developed as the surface ~~is~~ heated up on account of the incoming radiation. T~~his leads to downward mixing of O_3 from aloft~~ he high-concentration O_3 in the residual layer was then transported downstream (Hu et al., 2018~~; Zhan and Xie, 2022~~). Meanwhile, O_3 could be~~is also~~ generated by photochemical reactions between volatile organic compounds (VOCs) and NO_x in the presence of sunlight. Through these two pathways, surface O_3 concentration increased rapidly in the morning (Zhan and Xie, 2022)~~As a consequence, O_3 concentrations increase rapidly from morning to noon (Figure 7b, 7c, 7f and 7g)~~. By noon, O_3 was~~can be well~~ mixed within the convective boundary layer via strong turbulence. Strong

photochemical production and vertical ~~well~~ mixing could maintain~~ause~~ high surface O_3 concentrations until late afternoon. ~~(Figure 7c, 7d, 7g and 7h).~~ The daytime plain wind drove the westward transport of O_3 and aggravated O_3 pollution along the eastern slope of the Tibetan Plateau (Figure 8c and e). After sunset~~Thereafter~~, O_3 production ceased~~decreases~~ as~~since~~ the intensity of sunlight diminished~~s~~. ~~–~~ ~~After sunset,~~ O_3 concentrations decreased substantially owing ~~due~~ to surface deposition and nitrogen oxide titration ($O_3 + NO \rightarrow O_2 + NO_2$), and gradually reached their minimum in the early morning (Figure ~~7a, 7d, 7e and 7h~~8b). But O_3 in the nocturnal residual layer was~~is~~ still at a high level with values of more than 1640 μg m^{-3}. The nighttime mountain wind could carry air containing rich O_3 eastward downslope and enhanced O_3 concentrations aloft over the eastern slope of the Tibetan Plateau (Figure 8d). ~~Compared with the Jan_Base simulation, the secondary circulation forced by the complex terrain is not obvious. In addition~~Compared with the Jan_Base simulation, O_3 with a concentration of ~~about~~ ~100 μg m^{-3} had~~s~~ always existed over the Tibetan Plateau where~~, but~~ $PM_{2.5}$ concentrations were~~there are~~ quite low, indicating~~. This indicates~~ that the background concentration of O_3 was~~is~~ much higher than that of $PM_{2.5}$.~~,~~ This~~which~~ can~~may~~ pose a huge challenge to O_3 pollution control.


(a)
Height (km)
3
2
1.5
1
0.6
318
316
314
312
310
308
306
0
3
6
9
12
15
18
21
5 m/s
2:00 LST
14:00 LST
(b)
(c)
32°N
31°30'N
31°N
30°30'N
30°N
103°E
104°E
105°E
(d)
(e)
Height (km)
5.0
4.0
3.0
2.0
1.0
0.0
102.0°E
102.9°E
103.7°E
104.6°E
105.4°E
longitude
20
40
60
80
100
120
140
160
180
200
O3 (μg m-3)

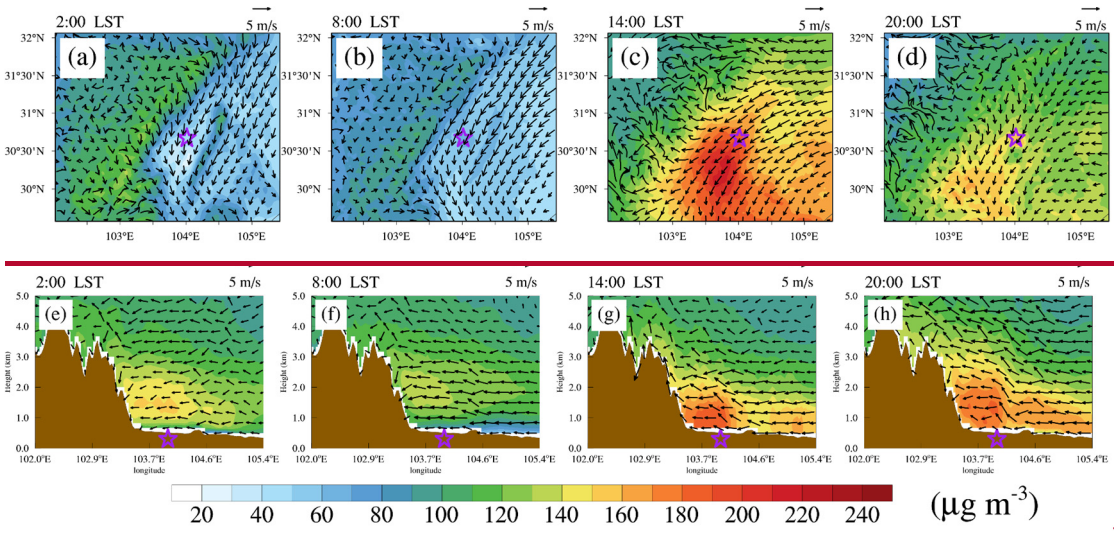


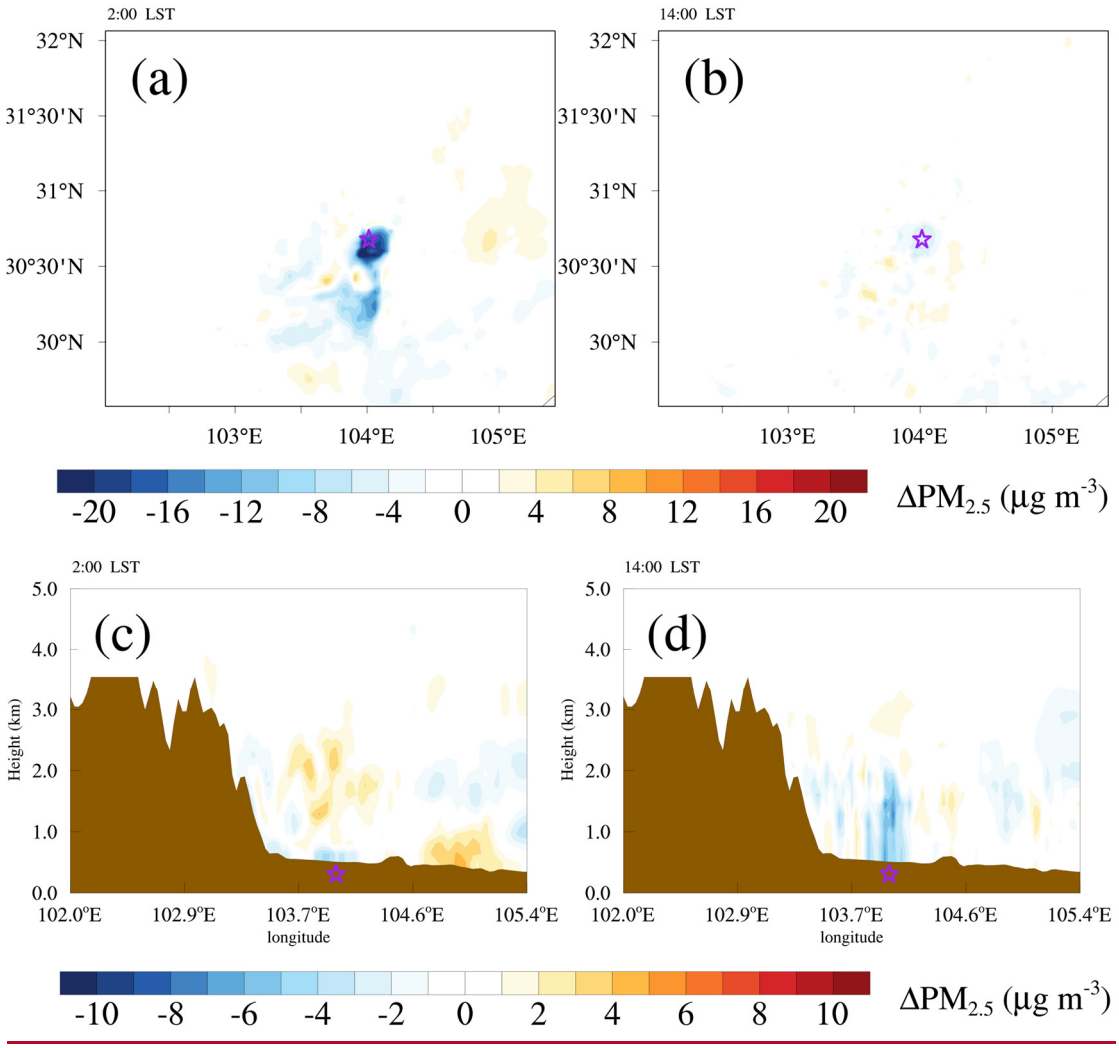
Figure 8~~7~~. (a) Temporal-vertical cross sections of O_3, potential temperature and boundary layer height at Chengdu. ~~(b-c) Spatial~~ Horizontal distributions ~~and (d-e) east-west vertical cross sections~~ of O_3 with wind vectors~~fields~~ at the lowest model level at (b) 2:00 and (c) 14:00 LST ~~(LST is UTC+8h)~~. East-west vertical cross sections of O_3 with wind vectors at (d) 2:00 and (e) 14:00 LST. Purple pentacles show the locations of Chengdu. Brown-shaded areas represent the terrain. These results are the monthly average based on July_Base simulation. ~~These results are from July_Base simulation.~~

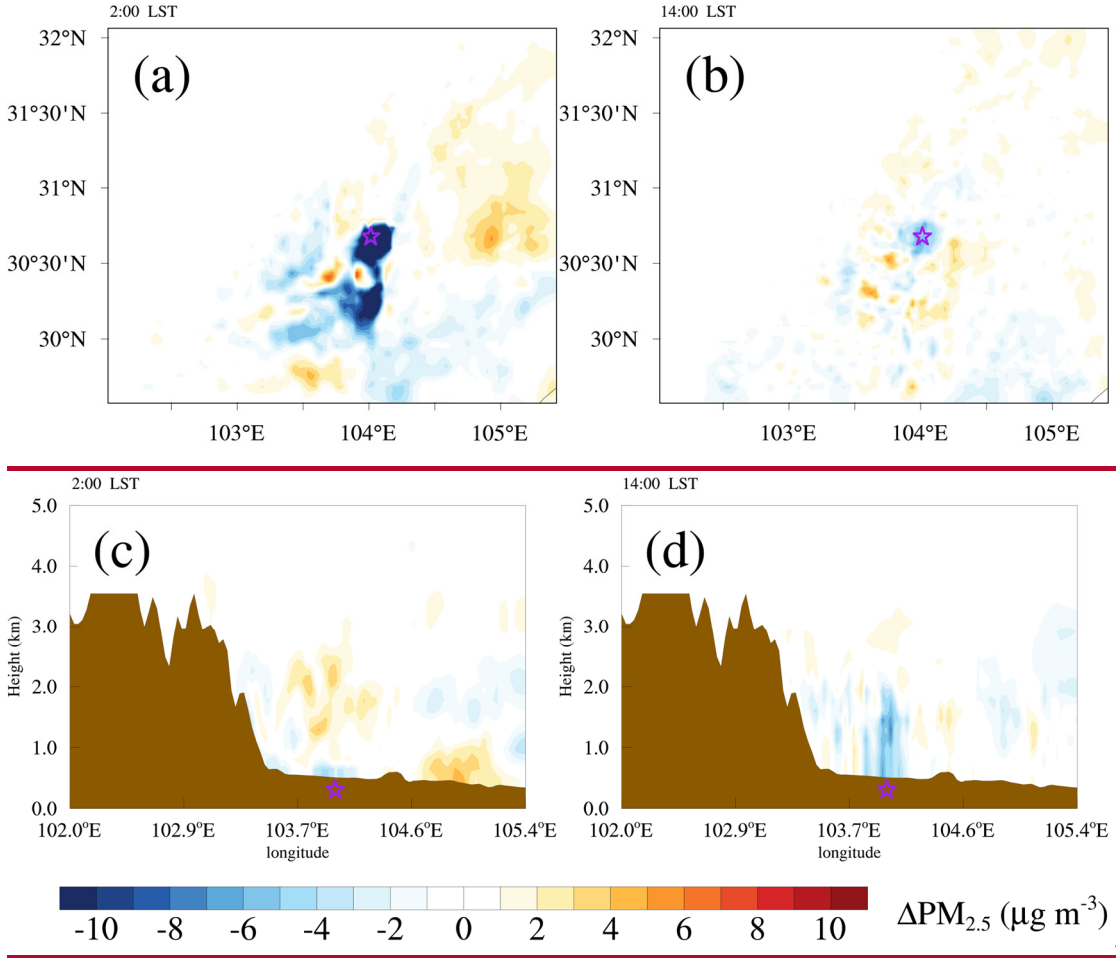
~~(a-d) Spatial distributions and (e-h) east-west vertical cross sections of O_3 with wind fields at 2:00, 8:00, 14:00 and 20:00 LST (LST is UTC+8h) in July_Base simulation. Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain.~~

3.3.4 Impacts of urban land use~~expansion~~ on $PM_{2.5}$ and O_3

Modification of urban land use changes surface dynamic and thermal characteristics, ~~and thereby affects the transportation and dispersion of air pollutants~~affecting the exchange of energy, moisture and momentum and hence altering urban meteorology and air quality. ~~Figure 8 shows the differences in $PM_{2.5}$ between Jan_Base and Jan_noCD simulations (Jan_Base minus Jan_noCD)~~As illustrated in Figure 9,~~.~~ ~~Results show that s~~urface $PM_{2.5}$ concentrations in Jan_Base simulation were~~are~~ lower than those in~~lower at all times compared with~~ Jan_noCD simulation,~~,~~ with the monthly average concentrations decreased~~ing~~ by 10.8~~1.7~~ μg m^{-3} (7.6%) ~~(Figure 8a-d)~~. Moreover~~Moreover~~, ~~,~~ the decrease in $PM_{2.5}$ concentrations was~~is~~ larger during ~~the~~ nighttime than

during~~ the~~ daytime. ~~Specially,~~ The monthly average~~surface~~ $PM_{2.5}$ concentrations could decrease by 13~~5~~.9~~0~~ μg m^{-3} (8.6%) at 2:00 LST (LST is UTC+8h) and~~and~~ only~~–~~ 3.0~~2~~ μg m^{-3} (2.6%) at 14:00 LST (Figure 9a and b). The decrease in surface $PM_{2.5}$ concentrations was~~is~~ mainly attributed to the modification of the boundary layer height. Urban land use can enhance surface heating and then increases~~leading to an increase in~~ air temperature~~, known as the urban heat island~~. The vertical air movement is then enhanced by the warming up of~~ surface~~ air temperature, resulting in an increase in the boundary layer height (Figure S1), which facilitates the vertical diffusion of surface $PM_{2.5}$. $PM_{2.5}$ concentrations increased by 2~~–~~6~~2–6~~ μg m^{-3} in the upper boundary layer (~1 km above the surface) (Figure 9c and d)~~8e-h)~~, further confirming this point.





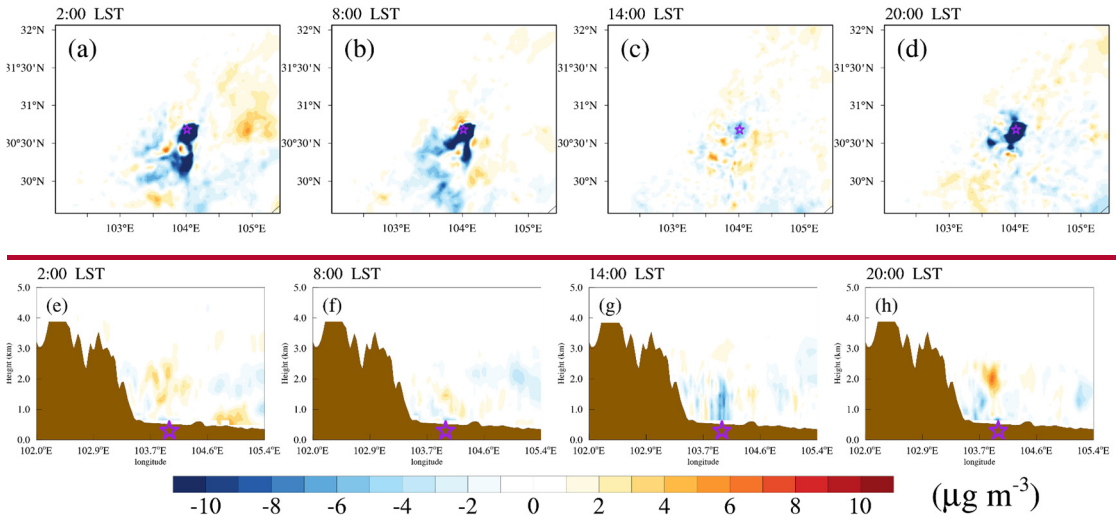
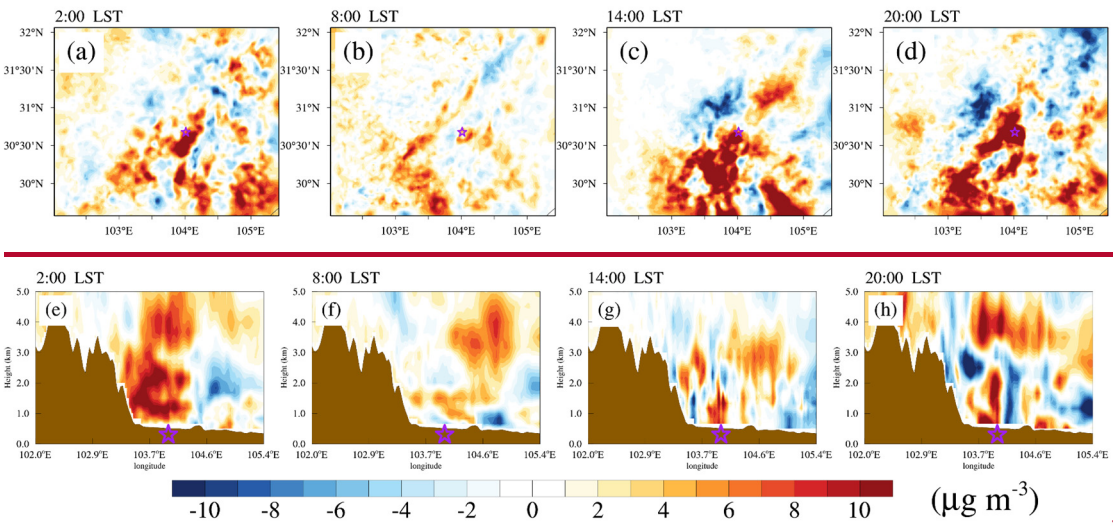
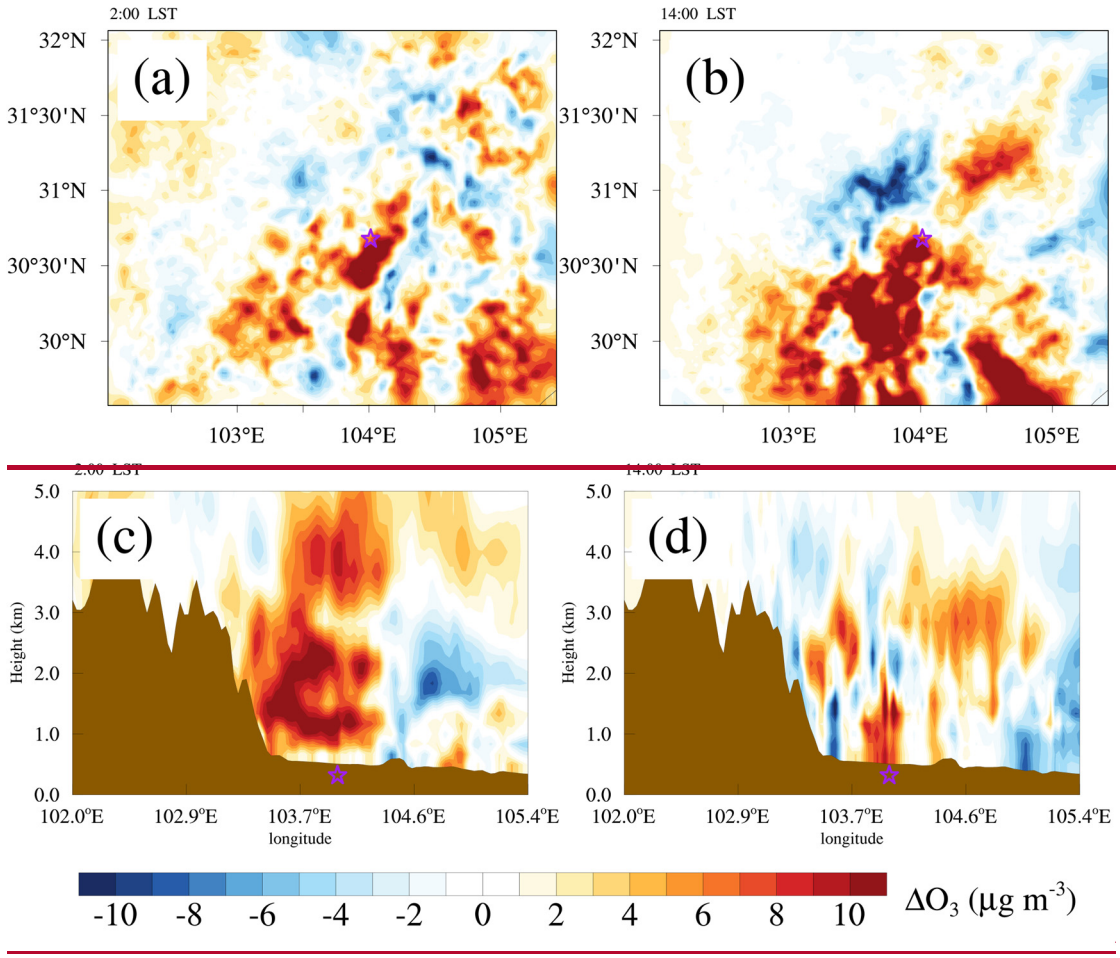


Figure 98. ~~(a-d)~~ Horizontal~~Spatial~~ distributions ~~and (e-h) east-west vertical cross sections~~ of the differences in $PM_{2.5}$ at the lowest model level at (a) 2:00 and (b) 14:00 LST~~between Jan_Base and Jan_noCD simulations (Jan_Base minus Jan_noCD)~~. East-west vertical cross sections of the difference in $PM_{2.5}$ at (c) 2:00 and (d) 14:00 LST. Purple pentacles show the

location of Chengdu. Brown-shaded areas represent the terrain. These results are the difference between the monthly average of Jan_Base and Jan_noCD simulations (Jan_Base minus Jan_noCD).

O_3 is a secondary air pollutant that is not only related to meteorological conditions but also to its precursors (VOCs and NO_x). Due to the increase in upward air movement and boundary layer height induced by urban land use (Figure S2), like $PM_{2.5}$, ~~$PM_{2.5}$ and~~ NO_x concentrations also decreased near the surface~~ but increase in the upper boundary layer~~ (Liao et al., 2015; Zhu et al., 2017). The decrease in NO_x near the surface resulted in~~results in~~ an increase in surface O_3 at night since the NO_x titration was~~is~~ weakened (Figure 10~~9~~a and ~~d~~c). Although the elevated boundary layer diluted~~s~~ O_3 concentrations to some extent, the nighttime O_3 concentrations were~~are~~ mainly dominated by chemical effects and~~ eventually~~ increased by 15~~a maximum of 25~~.6~~8~~ μg m^{-3} (16.0%) at 2:00 LST (Figure 10a). During~~ the~~ daytime, the increased air temperature was~~is~~ conducive to the photochemical production of O_3, and the well-developed convective boundary layer favored~~s~~ the vertical mixing of O_3. O_3 concentrations would~~ill~~ also increase (Figure 10~~9~~b and d~~e~~), with the~~the~~ monthly average value of 5~~4~~.4 μg m^{-3} (4.5%) at 14:00 LST.~~ in Chengdu.~~Since high O_3 concentrations were mainly concentrated in the afternoon~~ Finally~~, the monthly average MDA8 O_3 concentrations~~ in July can~~ finally increased by 10.6~~–~~ μg m^{-3} (6.0%) due to the effects of urban expansion.




2:00 LST
14:00 LST
(a)
(b)
(c)
(d)
32°N
31°30'N
31°N
30°30'N
30°N
103°E
104°E
105°E
Height (km)
5.0
4.0
3.0
2.0
1.0
0.0
102.0°E
102.9°E
103.7°E
104.6°E
105.4°E
longitude
-10
-8
-6
-4
-2
0
2
4
6
8
10
ΔO3 (μg m-3)

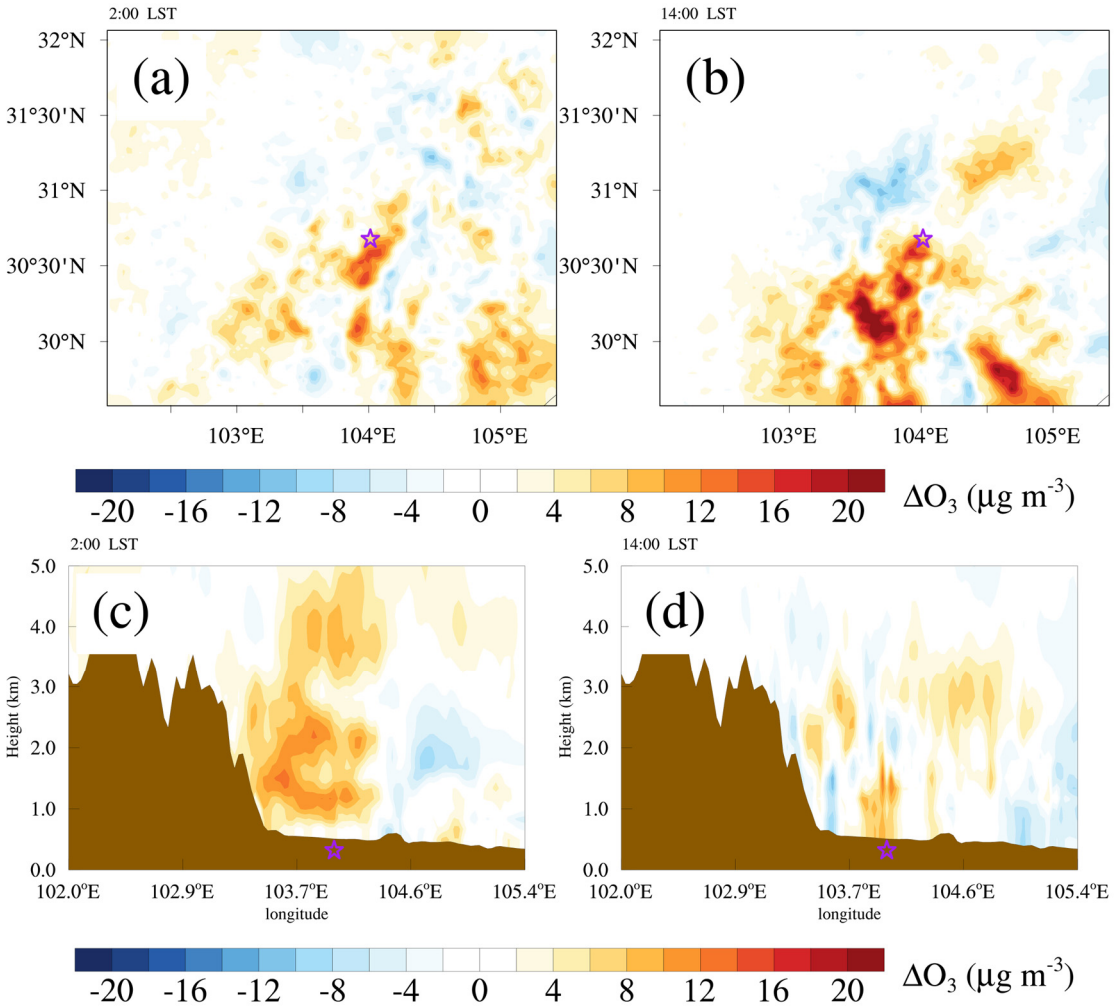
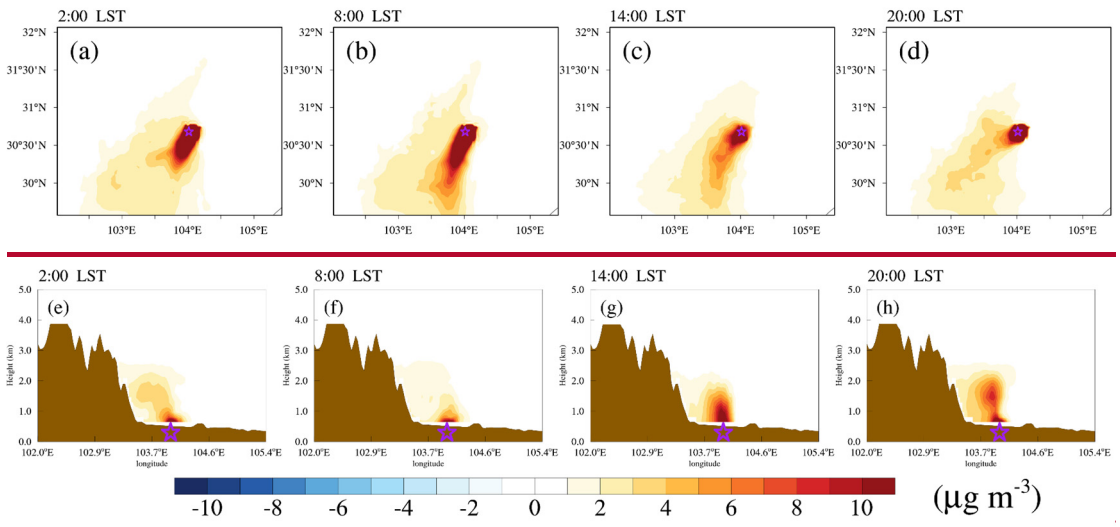


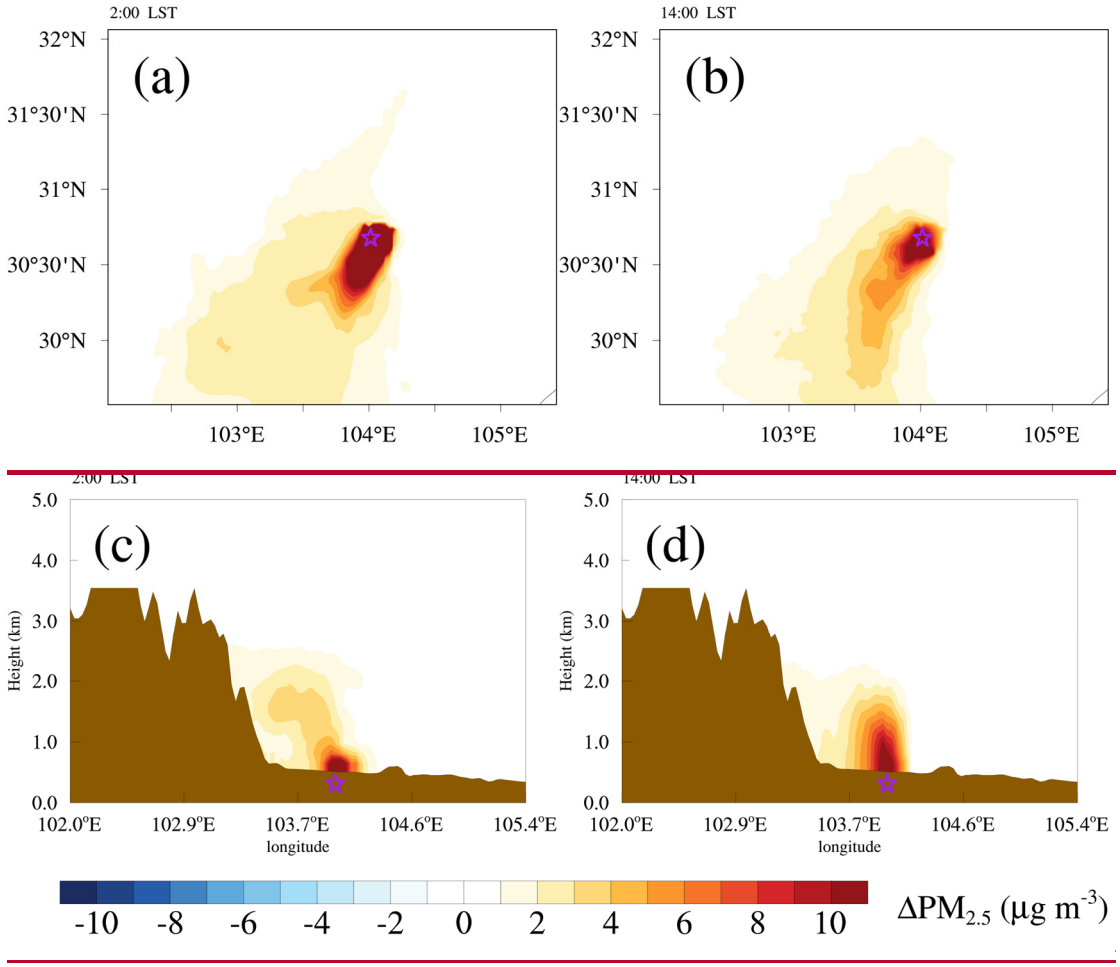
Figure 10. Horizontal distributions of the differences in O_3 at the lowest model level at (a) 2:00 and (b) 14:00 LST. East-west vertical cross sections of the difference in O_3 at (c) 2:00 and (d) 14:00 LST. Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain. These results are the difference between the monthly average of July_Base and July_noCD simulations (July_Base minus July_noCD).~~**Figure 9. (a-d) Spatial distributions and (e-h) east-west vertical cross sections of the differences in O_3 between July_Base and July_noCD simulations (July_Base minus July_noCD). Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain.**~~

3.3.5 Impacts of anthropogenic emissions on $PM_{2.5}$ and O_3

~~Air pollutants become part of the air once released, and they do not have a direct effect on airflows like temperature or radiatio~~Rising anthropogenic emissions of air pollutants and their precursors can significantly increase ambient air pollution. Therefore, the impacts of anthropogenic

emissions are more intuitive than urban land use~~expansion~~. Figure 11~~0~~ shows the differences in $PM_{2.5}$ between the monthly average of Jan_Base and Jan_noEmi simulations (Jan_Base minus Jan_noEmi). $PM_{2.5}$ concentrations in Jan_Base simulation were~~are~~ significantly higher than those in Jan_noEmi simulation~~s~~, with the monthly average concentration increased~~ing~~ by 23~~6~~.9~~6~~ μg m^{-3} (16.8%), more than twice the difference between Jan_Base and Jan_noCD simulations. Furthermore, the increases in $PM_{2.5}$ concentrations appeared~~s~~ throughout the boundary layer (Figure 11c~~0e~~ and d~~-h~~) and could~~an~~ extend downstream for hundreds of kilometers (Figure ~~10a~~11a and -b~~d~~), indicating that reducing anthropogenic emissions is an effective way to reduce $PM_{2.5}$ concentrations.




2:00 LST
14:00 LST
(a)
(b)
(c)
(d)
32°N
31°30'N
31°N
30°30'N
30°N
103°E
104°E
105°E
Height (km)
5.0
4.0
3.0
2.0
1.0
0.0
102.0°E
102.9°E
103.7°E
104.6°E
105.4°E
longitude
-10
-8
-6
-4
-2
0
2
4
6
8
10
ΔPM2.5 (μg m-3)

584

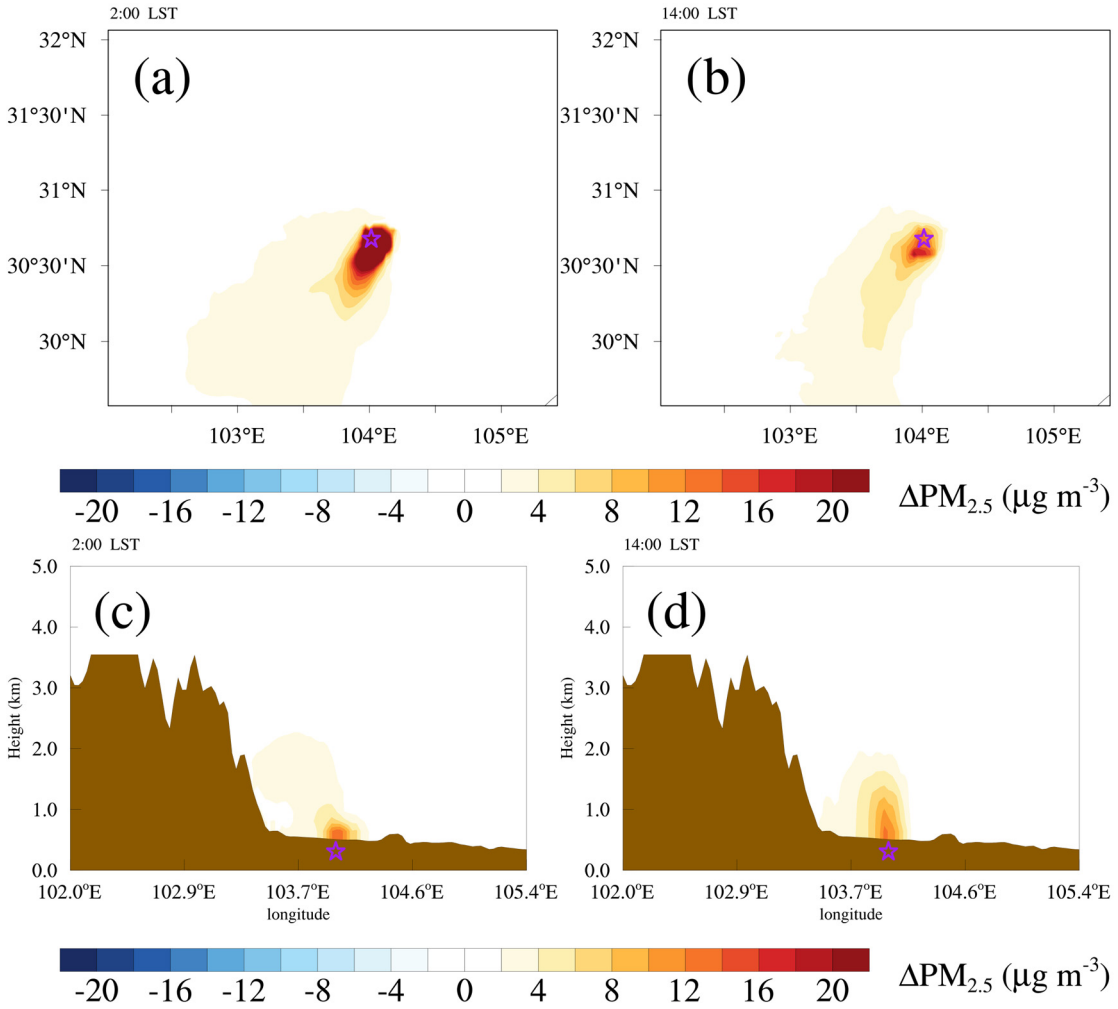
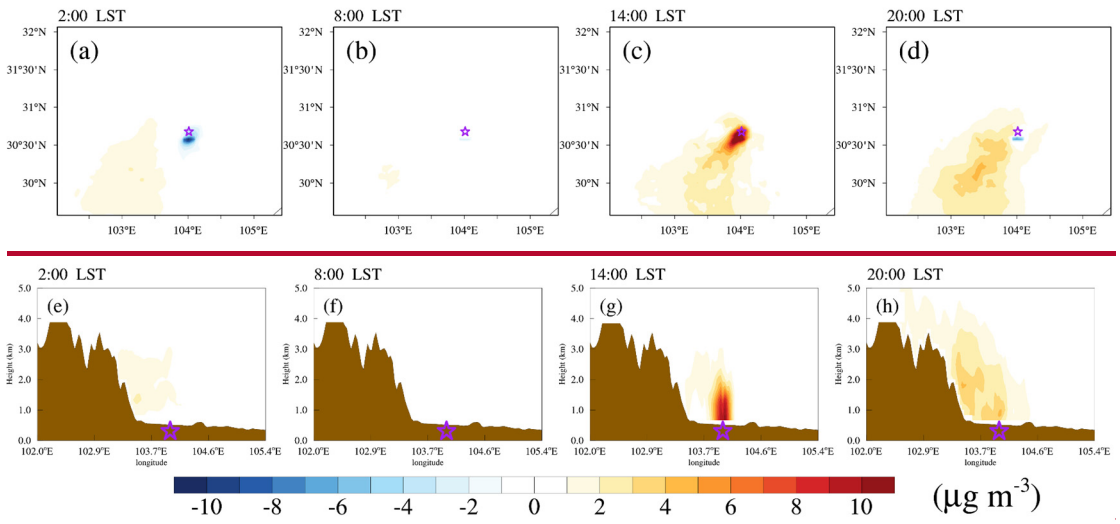
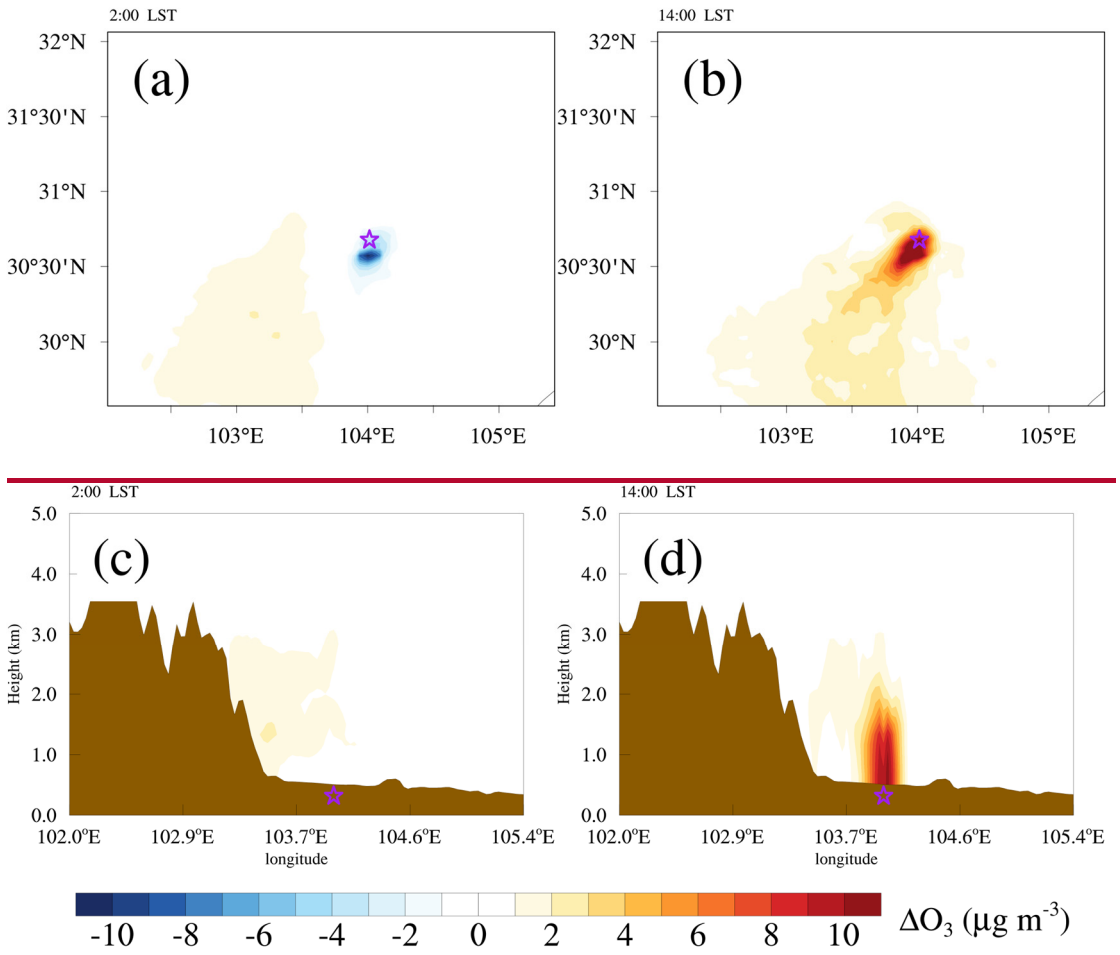


Figure ~~10~~11. Same as Figure 9, but for the difference between the monthly average of Jan_Base and Jan_noEmi simulations (Jan_Base minus Jan_noEmi). ~~(a-d) Spatial distributions and (e-h) east-west vertical cross sections of the differences in $PM_{2.5}$ between Jan_Base and Jan_noEmi simulations (Jan_Base minus Jan_noEmi). Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain.~~

As for O_3, O_3 concentrations in July_Base simulation were~~are~~ only 1~~5~~.6~~8~~ μg m^{-3} (1.4%) higher than those in July_noEmis simulation at 14:00 LST~~ due to the abundance of O_3 precursors~~ (Figure ~~11c~~12b and d), which was much smaller than the change in $PM_{2.5}$~~. However, O_3 concentrations decrease by 3.4 μg m^{-3} at 2:00 LST (Figure 11a)~~. This phenomenon may be related to the non-linear sensitivity of O_3 to VOCs and NO_x precursor emissions. O_3 formation regimes can be~~are often~~ classified into VOC-limited, NO_x-limited and transition regimes depending on the ratio of VOCs and NO_x (Jin et al., 2020; Lu et al., 2019). At low VOC/NO_x ratios (VOC-limited regime~~, usually~~

~~in urban areas~~), reducing the concentrations of NO_x would even lead to an increase in O_3 formation. Considering Chengdu remained VOC-limited regime during 2013 to 2020 (Tian et al., 2018; Wang et al., 2022), the effects of reducing NO_x emissions may be partially offset by changes in VOCs, and thereby a reasonable regulation framework that involves joint control of NO_x and VOC emissions is necessary to alleviate O_3 pollution. On the other hand, the presence of anthropogenic emissions reduced O_3 concentrations by 3.0 μg m^{-3} (3.1%) at 2:00 LST. Finally, the monthly average ~~Apart from the amount of anthropogenic emissions, a reasonable emission reduction path is also necessary to alleviate O_3 pollution. Since O_3 concentrations increase during the daytime,~~ MDA8 O_3 concentrations in July_Base simulation were~~are still~~ 4.8 μg m^{-3} (2.7%) higher than those in July_noEmis simulation.




2:00 LST
14:00 LST
(a)
(b)
(c)
(d)
32°N
31°30'N
31°N
30°30'N
30°N
103°E
104°E
105°E
Height (km)
5.0
4.0
3.0
2.0
1.0
0.0
102.0°E
102.9°E
103.7°E
104.6°E
105.4°E
longitude
-10
-8
-6
-4
-2
0
2
4
6
8
10
ΔO3 (μg m-3)

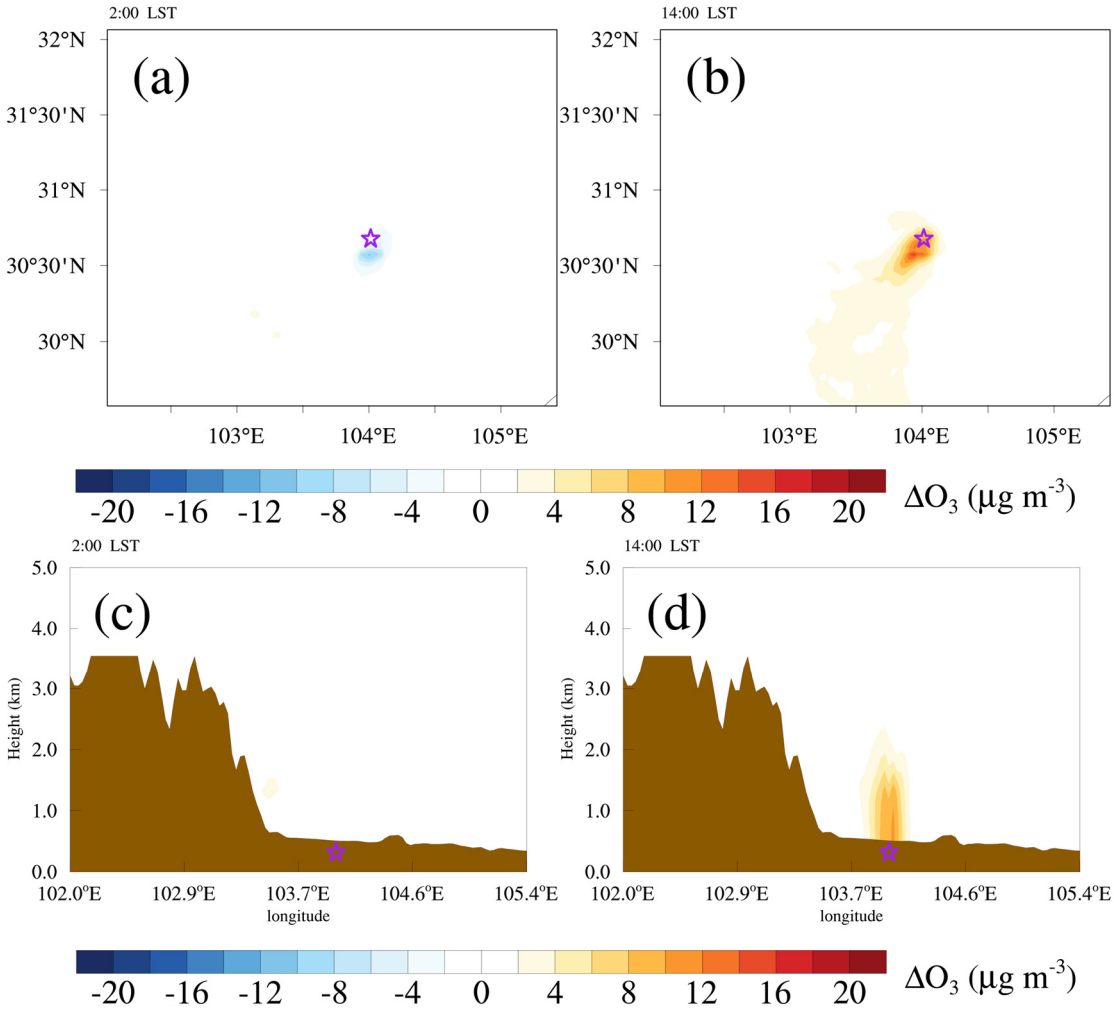


Figure ~~11~~12. Same as Figure 10, but for the difference between the monthly average of July Base and July noEmi simulations (July Base minus July noEmi). ~~(a-d) Spatial distributions and (e-h) east-west vertical cross sections of the differences in O_3 between July_Base and July_noEmi simulations (July_Base minus July_noEmi). Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain.~~

3.4 Health risks caused by urbanization

According to the above results, urban land use~~expansion~~ could~~an~~ decrease the monthly average of~~surface~~ $PM_{2.5}$ concentrations by 10~~1~~.8~~7~~ μg m^{-3} (7.6%) but increase the monthly average of MDA8 O_3 concentrations by 10.6 μg m^{-3} (6.0%). On the other hand, anthropogenic emissions could~~an~~ increase ~~surface~~ both $PM_{2.5}$ ~~by 26.6 μg m^{-3}~~ and MDA8 O_3 concentrations, with the monthly average values of 23.9 μg m^{-3} (16.8%) and ~~by~~ 4.8 μg m^{-3} (2.7%), respectively. We then calculate the changes in premature mortalities~~–~~ under different simulation scenarios to assess the health risks from

changes in $PM_{2.5}$ and O_3 concentrations~~ to access the health risks from changes in $PM_{2.5}$ and O_3 concentrations~~. As shown in Figure 13~~2~~, the premature mortalities from ANAC, CVD, RD and COPD due to $PM_{2.5}$ decreased by 1~~8~~71 (95%CI: 129~200, or about 6.9%), 45 (95%CI: 34~53, or about 6.7%), 22 (95%CI: 16~27, or about 6.5%)~~2 (6.9%), 47 (6.5%), 23 (6.4%) and 24 (6.1%)~~ and 23 (95%CI: 17~26, or about 6.2%) in January 2017 with the existence of Chengdu. While anthropogenic emissions in Chengdu increased premature mortalities from ANAC, CVD, RD and COPD due to $PM_{2.5}$ by~~ 424 (16.0%), 111 (15.4%), 55 (15.2%) and 56 (14.3%)~~ 388 (95%CI: 291~456, or about 15.7%), 102 (95%CI: 77~121, or about 15.1%), 51 (95%CI: 35~62, or about 15.0%) and 52 (95%CI: 39~60, or about 14.1%)~~, respectively~~. With regard to O_3, premature mortalities from ~~the~~ O_3-induced diseases all increased when urban land use and anthropogenic emissions were~~are~~ taken into account. Urban land use~~expansion~~ le~~ad~~d~~s~~ to an increase in~~of~~ premature mortalities from ANAC, CVD, RD and COPD due to O_3 by 203 (95%CI: 122~268, or about 9.5%~~9.5%~~), 51 (95.~~4~~%CI: 22~71, or about 9.4%), 18 (~~1~~95%CI: –14~35, or about 10.0%~~0.0%~~) and 17 (95.~~7~~%CI: –15~33, or about 9.7%) in July 2017~~, respectively~~. When anthropogenic emissions in Chengdu were~~are~~ turned on, premature mortalities from ANAC, CVD, RD and COPD due to O_3~~ can~~ increased by 87 (95%CI: 54~112, or about 4.1%~~4.1%~~), 22 (95%CI: 10~29, or about 4.1%~~4.1%~~), 8 (95~~4.4~~%CI: –7~14, or about 4.4%) and 7 (95~~4.0~~%CI: –7~13, or about 4.0%), respectively. In summary, ~~affected by urban expansion and anthropogenic emissions, changes in t~~the total premature mortalities due to $PM_{2.5}$ and O_3 changed by~~are~~ about ~~–~~6.9% and 9.5~~16.0~~% affected by urban expansion, and these values changed to~~, due to O_3 are~~ about 15.7~~9.5~~% and 4.1~~4.1~~% affected by emissions growth. ~~The effects of urban expansion on health risks are in the same order (1/2 to 2 times) as those induced by anthropogenic emissions.~~

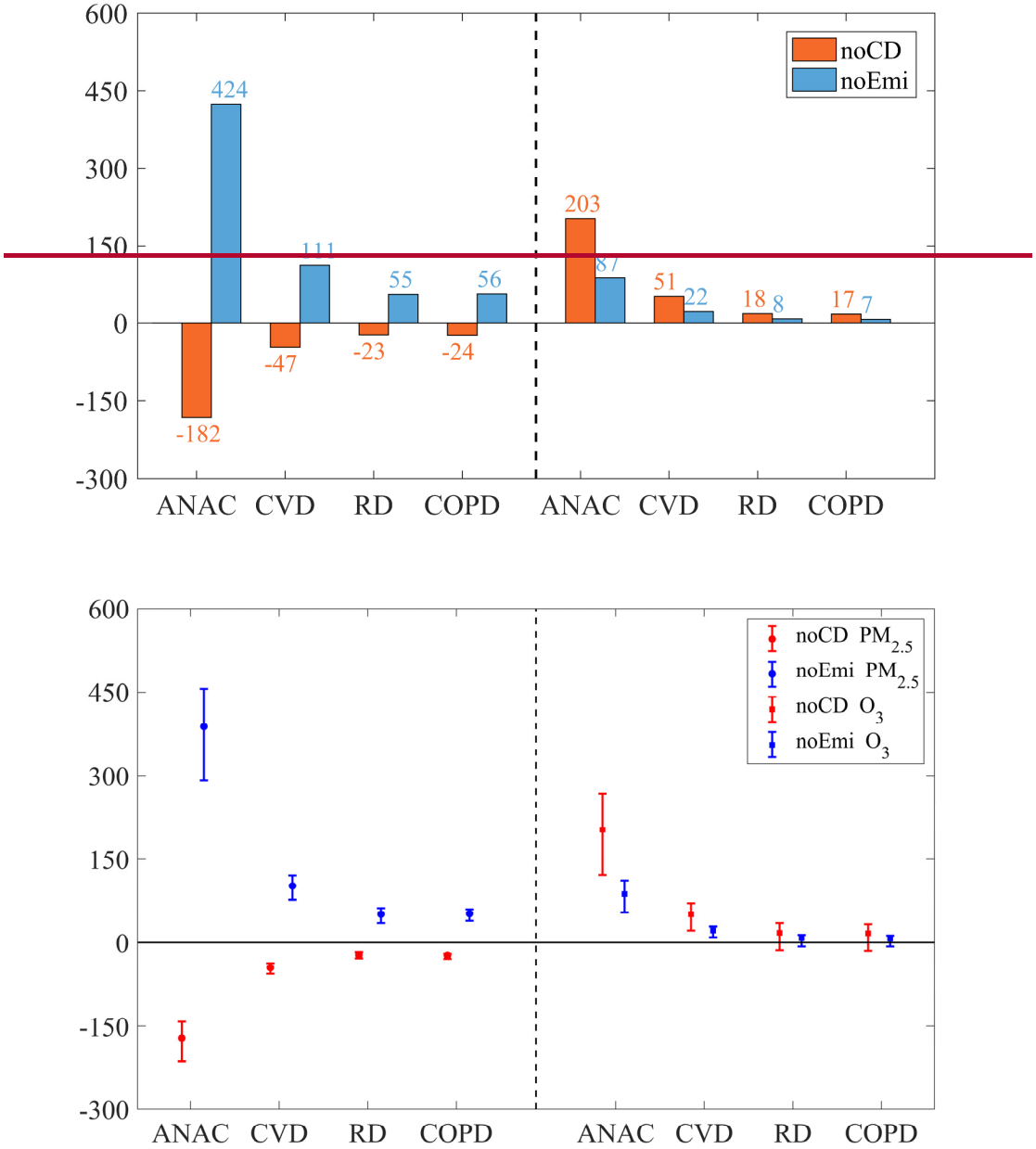


Figure 1~~3~~2. Differences in premature mortality from ANAC, CVD, RD and COPD due~~attributed~~ to $PM_{2.5}$ (left of dotted line) and O_3 (right of dotted line)~~ exposure~~ between baseline and sensitivity simulations. ~~The red bars are the differences between Jan_Base and Jan_noCD simulations (Jan_Base minus Jan_noCD), and the differences between July_Base and July_noCD simulations (July_Base minus July_noCD). The blue bars are the differences between Jan_Base and Jan_noEmi simulations (Jan_Base minus Jan_noEmi), and the differences between July_Base and July_noEmi simulations (July_Base minus July_noEmi).~~

4 Conclusions

With the development in urbanization, urban land use and anthropogenic emissions increase, which affects urban air quality and then health risks of air pollutants. In this study, the impacts of urban land use and anthropogenic emissions~~ization~~ on air quality and the related~~corresponding~~ health risks in Chengdu, a highly urbanized city with severe air pollution and complex terrain, are quantified. Management of urban air pollution is usually achieved by reducing anthropogenic emissions. Thus,~~So~~ the effects~~impacts~~ of urban expansion have been~~are~~ also~~further~~ compared with those of emissions growth~~anthropogenic emissions on health risks~~.

Chengdu is suffering from severe $PM_{2.5}$ and O_3 pollution in recent years. There were~~are~~ 97, 101, 68, 53, 33, 43 and 37 $PM_{2.5}$ pollution episodes, and 61, 48, 42, 40, 42~~26~~, 71~~50~~ and 48~~27~~ O_3 pollution episodes~~ in Chengdu~~ from 2015 to 2021. Severe $PM_{2.5}$ and O_3 pollution posed huge health risks. The 7-year annual average of premature mortalities from ANAC, CVD, RD and COPD due to $PM_{2.5}$ were~~are~~ 9386 (95%CI: 6542~11726), 2609 (95%CI: 1788~3384), 1321(95%CI: 804~1840) and 1485 (95%CI: 941~1983), those due to O_3 were~~are~~ 8506 (95%CI: 4817~11882)~~7743~~, 2175 (95%CI: 863~3320)~~1981~~, 713 (95%CI: –492~1664)~~648~~ and 693 (95%CI: –517~1617)~~30~~. $PM_{2.5}$ and O_3 pollution had~~ve~~ different seasonal preferences. Owing~~Due~~ to the ~~secondary circulation driven by complex terrain~~ blocking of air and the frequent temperature inversion, $PM_{2.5}$ pollution tended~~s~~ to appear in cold months (November to February). ~~–~~While O_3 pollution was~~is~~ likely to occur in warm months (April to August) because of high temperature and strong sunlight dominated by high-pressure systems. $PM_{2.5}$ ~~has a diurnal variation with high~~ concentrations were high at night and~~but~~ low~~ concentrations~~ at noon, which was ~~affected by~~contrary to the boundary layer height. While O_3 exhibited~~s~~ strong diurnal variation with an afternoon maximum and an early morning minimum, which was~~is~~ related to photochemical reactions during ~~the~~ daytime and nitrogen oxide titration at night.~~–~~

The urban land use of Chengdu was~~is~~ replaced by cropland in the WRF-Chem model to examine the impacts of urban expansion. Urban land use~~expansion~~ le~~a~~d~~s~~ to an increase in air temperature and boundary layer height, and decreased~~s~~ surface $PM_{2.5}$ concentrations by 10.8~~1.7~~ μg m^{-3} (7.6%)~~ in January 2017~~. Higher temperature and boundary layer height increased O_3~~As for O_3, the surface concentration increases by 4.4 μg m^{-3} at noon due~~ concentrations via ~~to~~ stronger photochemical reactions and better vertical mixing during daytime. During nighttime, dominated

by the weakened chemical NO_x titration, O_3 concentrations~~,~~ also~~and~~ increased~~s by 25.8 μg m^{-3} at midnight since the NO_x titration is weakened~~. MDA8 O_3 concentrations could finally increase~~s by 10.6 μg m^{-3} ~~in July 2017~~ (6.0%). In this case, w~~w~~hen urban land use was~~is~~ taken into account~~. In this case,~~, the premature mortalities from ANAC ~~, CVD, RD and COPD attributed~~due to $PM_{2.5}$ exposure decreased by~~–~~ 171 (95%CI: 129~200, or about 6.9%)~~182 (6.9%), 47 (6.5%), 23 (6.4%) and 24 (6.1%),~~ but those due~~attributed~~ to O_3~~ exposure~~ increased by 203 (95%CI: 122~268, or about 9.5%)~~203 (9.1%), 51 (9.4%), 18 (10.0%) and 17 (9.7%)~~. Anthropogenic emissions increased surface $PM_{2.5}$ significantly with the monthly average concentration increasing by 23~~6~~.9~~6~~ μg m^{-3} (16.8%), more than twice the difference caused by urban land use~~expansion~~. Owing~~Due~~ to the non-linear sensitivity of O_3 to its precursors, O_3 concentrations increased ~~during the daytime~~ at noon but decreased at night. In particular, the monthly average O_3 concentrations increased by 1~~5~~.6~~8~~ μg m^{-3} (1.4%) at 14:00 LST but decreased by 3.0~~4~~ μg m^{-3} (3.1%) at 2:00 LST~~ with anthropogenic emissions in Chengdu~~. Since O_3 concentrations in daytime were~~are~~ much higher than those at night, the MDA8 O_3 concentrations still~~still~~ increased by 4.8 μg m^{-3} (2.7%). As a consequence, the premature mortalities from ANAC due~~, CVD, RD and COPD attributed~~ to $PM_{2.5}$~~ exposure~~ increased by 388 (95%CI: 291~456, or about 15.7%)~~y 424 (16.0%), 111 (15.4%), 55 (15.2%) and 56 (14.3%)~~, and those due~~attributed~~ to O_3~~ exposure~~ increased by 87 (95%CI: 54~112, or about 4.1%) with anthropogenic emissions in Chengdu~~87 (4.1%), 22 (4.1%), 8 (4.4%) and 7 (4.0%)~~.

Our results show that the impacts of urban expansion (about –6.9% for $PM_{2.5}$ and about 9.5% for O_3) are in the same order~~ (1/2 to 2 times)~~ as those induced by emissions growth (about 15.7% for $PM_{2.5}$ and about 4.1% for O_3) on air pollutants. This suggests that al~~Al~~though the focus of air quality management is traditionally to regulate emissions, urban planning is an ancillary option and should also be considered in future air pollution strategies.

Data Availability Statement.

Air quality monitoring data are acquired from the official NEMC real-time publishing platform (http://106.37.208.233:20035/). Meteorological data are taken from the website of the University of Wyoming (http://weather.uwyo.edu/). The NCEP ~~global final analysis~~ FNL data were taken from the NCEP (https://doi.org/10.5065/D6M043C6/). The MEIC data are accessible at http://meicmodel.org/. These data can be downloaded for free as long as you agree to the official

instructions.

Author contributions.

CZ and MX had the original ideas, designed the research, collected the data and prepared the original draft. CZ did the numerical simulations and carried out the data analysis. MX acquired financial support for the project leading to this publication. HL, BL and ZW collected the data. TW, BZ, ML and SL reviewed the initial draft and checked the language of the original draft.

Competing interests.

The contact author has declared that neither they nor their co-authors have any competing interests.

Acknowledgements.

We are grateful to NEMC for the air quality monitoring data, to NCDC for the meteorological data, to NCEP for global final analysis fields and to Tsinghua University for the MEIC inventories. The numerical calculations were performed on the Blade cluster system in the High_–Performance Computing and Massive Data Center (HPC&MDC) of School of Atmospheric Sciences, Nanjing University. We thank the anonymous reviewers for their constructive comments and suggestions.

Financial support.

This work was supported by the National Nature Science Foundation of China (grant no. 42275102), the open research fund of Chongqing Meteorological Bureau (KFJJ-201607) and the Natural Science Foundation of Jiangsu Province (grant no. BK20211158).~~This work was supported by the open research fund of Chongqing Meteorological Bureau (KFJJ-201607), the Natural Science Foundation of Chongqing (cstc2021jcyj-msxmX1007), the Chongqing Science and Technology Commission technology innovation and application demonstration project (cstc2018jszx-zdyfxmX0003) and Innovation Team Fund of Southwest Regional Meteorological Center, China Meteorological Administration.~~

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