# 1 Impacts of urbanization on air quality and related health risks in

# 2 a city with complex terrain

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Abstract: Urbanization affects air pollutants via 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 air quality management is usually achieved by regulating anthropogenic emissions. Air pollution in Chengdu was mainly caused by PM<sub>2.5</sub> and O<sub>3</sub> from 2015 to 2021. PM<sub>2.5</sub> pollution tended to appear in cold months (November to February) owing to the blocking of air and the stable atmospheric layer, while O<sub>3</sub> pollution was likely to occur in warm months (April to August) because of the high temperature and strong sunlight dominated by high-pressure systems. From 2015 to 2021, the 7-year annual averages of premature mortalities from all non-accidental causes (ANAC) due to PM<sub>2.5</sub> and O<sub>3</sub> were 9386 [95% confidence intervals (CI): 6542–11726] and 8506 (95%CI: 4817–11882), respectively. Based on the characteristics of PM<sub>2.5</sub> and O<sub>3</sub>, six numerical experiments were conducted to investigate the impacts of urban expansion and emission growth on health risks of air pollutants. The results show that urban land use led to an increase in air temperature and the boundary layer height compared to cropland, which was conducive to the diffusion of PM<sub>2.5</sub>. Thus, the monthly average surface PM<sub>2.5</sub> concentrations decreased by 10.8 µg m<sup>-3</sup> (7.6%) in January. However, the monthly average daily maximum 8 h average (MDA8) O<sub>3</sub> concentrations increased by 10.6 μg m<sup>-3</sup> (6.0%) in July owing to the stronger photochemical production and better vertical mixing during daytime. In this case, premature mortalities from ANAC due to PM<sub>2.5</sub> decreased by 171 (95%CI: 129–200, or about 6.9%) in January, and those due to O<sub>3</sub> increased by 203 (95%CI: 122–268, or about 9.5%) in July. As for the effects of emission growth, the monthly average PM<sub>2.5</sub> and MDA8 O<sub>3</sub> concentrations increased by 23.9 μg m<sup>-3</sup> (16.8%) and 4.8 μg m<sup>-3</sup> (2.7%) when anthropogenic emissions were taken into account. Premature mortalities from ANAC due to PM<sub>2.5</sub> and O<sub>3</sub> then increased by 388 (95%CI: 291–456, or about 15.7%) and 87 (95%CI: 54–112, or about 4.1%), respectively. From a health risk perspective, the effects of urban land use on health risks of PM<sub>2.5</sub> are about half that of anthropogenic emissions, whereas the effects of urban land use on health risks of O<sub>3</sub> can be 2 times that of anthropogenic emissions. This reminds us that, in addition to regulating anthropogenic emissions, urban planning is also important for urban air quality, especially for secondary pollutants like O<sub>3</sub>.

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 are ozone (O<sub>3</sub>), fine particulate matter (PM<sub>2.5</sub>, particulate matter with an aerodynamic diameter of 2.5 μm or less), sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>, which is NO + NO<sub>2</sub>). 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 4.2 million premature deaths worldwide annually (https://www.who.int/healthtopics/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 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<sub>3</sub> 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<sub>3</sub> concentration by 4.2%–8.5%. Liao et al. (2015) conducted a similar study in the Yangtze River Delta, and found that urbanization increased 2-m temperature, planetary boundary layer and surface O<sub>3</sub> concentration but decreased surface PM<sub>10</sub> (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).

Urban areas are centers of resource utilization and are a major contributor to air pollutants and greenhouse gas emissions (Karl et al., 2019; Qian et al., 2022). 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 total greenhouse gases. Air pollutants that originate from anthropogenic sources can 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 the root cause of poor air quality in urban areas, and thereby efforts have been made to reduce anthropogenic emissions to achieve the goal 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, 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 as the atmospheric dispersion

is limited in mountainous areas (Zardi and Whiteman, 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 daytime, 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. Examples can be found in Mexico City (Molina et al., 2010), Hong Kong (Guo et al., 2013), 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).

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. 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). 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). Luo et al. (2021) reported that Chengdu's urban area has increased by four times from 1996 to 2016. Because of the substantial anthropogenic emissions from human activities and the poor atmospheric diffusion capacity associated with terrain, Chengdu is one of the most polluted cities in China and has suffered from severe PM<sub>2.5</sub> and O<sub>3</sub> 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 investigate the impacts of urbanization on air pollutant concentrations and the corresponding health risks in Chengdu. We also compare the impacts of urban expansion with emission growth. First, the basic characteristics of air pollutants in Chengdu from 2015 to 2021 are analyzed. Then, the impacts of urbanization on air pollutant concentrations are investigated using the WRF-Chem model. Finally, premature mortalities attributable to changes in air pollutant concentrations are estimated using the standard damage function. The rest of this paper is organized as follows. Section 2 introduces the data, the model configurations and the experimental designs.

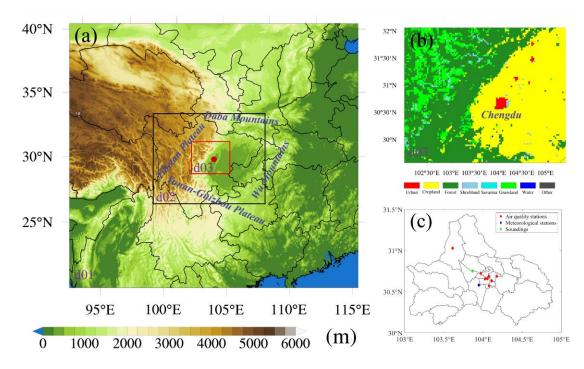


Figure 1. (a) Map of three nested WRF-Chem domains with terrain heights, (b) domain 3 with land cover maps and (c) locations of air quality stations, meteorological stations and soundings in Chengdu. The red dot in (a) shows the location of Chengdu.

# 2 Data and methods

### 2.1 Air quality and meteorological data

Air pollutants, including PM<sub>2.5</sub>, PM<sub>10</sub>, O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub> and CO, are monitored by the National Environmental Monitoring Center 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<sub>3</sub> measurements are reported in the unit of μg m<sup>-3</sup> 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 pollutant concentrations reported in this paper are the average results of measurements at all monitoring sites. The daily PM<sub>2.5</sub> concentrations are obtained by averaging observations over 24 hours of the day. The daily maximum 8 h average (MDA8) O<sub>3</sub> concentrations are calculated only

on days with more than 18 h of O<sub>3</sub> measurements.

Surface meteorological data, including 2-m air temperature (T<sub>2</sub>), 2-m dew point temperature (TD<sub>2</sub>), 10-m wind speed (WS<sub>10</sub>) and 10-m wind direction (WD<sub>10</sub>), 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 (station 56187) are also acquired from this website. These sounding data contain temperature, dew point temperature 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).

### 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 height of the lowest model level 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) Final (FNL) reanalysis data with a 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) with a grid resolution of 0.25° × 0.25°. It should be noted that we empirically cut the PM<sub>2.5</sub> emissions by about 20% to avoid overestimation of PM<sub>2.5</sub> in the model. 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)

MADE/SORGAM (Schell et al., 2001)

Aerosol module

To estimate the impacts of urbanization, six numerical simulations are designed (Table 2). The year of the numerical simulations is 2017 since the MEIC emission inventory is currently updated to 2017. Taking into account the computational cost, January is the representative of cold months with frequent PM<sub>2.5</sub> pollution, while July is the representative of warm months with frequent O<sub>3</sub> pollution (Section 3.1). Jan\_Base is a baseline simulation using the MODIS land use and the MEIC emission inventory over all three domains. The land cover maps in domain 3 are particularly shown in Figure 1b. Jan\_noCD is a sensitivity simulation, in which the urban land use of Chengdu is replaced by cropland to examine the impacts of urban expansion. Jan\_noEmi is another sensitivity simulation, in which the anthropogenic emissions in Chengdu are shut down to identify the impacts of emission growth. The above three numerical experiments use 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 simulations are conducted in this study.

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 to PM<sub>2.5</sub> and O<sub>3</sub> 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.,

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

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

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

where  $\beta$  is the concentration-response function that relates a unit change in air pollutant concentrations to a change in health endpoint incidence. In practice,  $\beta$  usually represents the percentage increase in daily mortality associated with a 10  $\mu$ g m<sup>-3</sup> increase in daily PM<sub>2.5</sub>/MDA8 O<sub>3</sub> concentrations. C is the exposure concentration, which is the daily average concentration for PM<sub>2.5</sub> and the MDA8 O<sub>3</sub> concentration for O<sub>3</sub>. C<sub>0</sub> is the threshold concentration. When C is not greater than C<sub>0</sub>, the value of C–C<sub>0</sub> is 0.

In this study,  $C_0$  is 10  $\mu$ g m<sup>-3</sup> for daily PM<sub>2.5</sub> (Song et al., 2015), and 75.2  $\mu$ g m<sup>-3</sup> for MDA8 O<sub>3</sub> (Liu et al., 2018).  $\beta$  and  $y_0$  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<sub>2.5</sub>- and O<sub>3</sub>-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  $\beta$ , premature mortalities are presented as means and 95% confidence intervals (CI) based on  $\beta$  at 95% CI in this study. In addition, it should be noted that we use the average 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. Thus, our results are for Chengdu as a whole and do not address the spatial distribution of premature mortalities.

**Table 3.** Daily  $\beta$  and  $y_0$  values for ANAC, CVD, RD and COPD.

Disease	$\beta^*$ for PM <sub>2.5</sub>	$\beta$ for MDA8 O <sub>3</sub>	<i>y</i> 0
ANAC	0.22 (0.15, 0.28)	0.24 (0.13, 0.35)	1.687×10 <sup>-5</sup>
CVD	0.27 (0.18, 0.36)	0.27 (0.10, 0.44)	3.880×10 <sup>-6</sup>
RD	0.29 (0.17, 0.42)	0.18 (-0.11, 0.47)	1.841×10 <sup>-6</sup>
COPD	0.38 (0.23, 0.53)	0.20 (-0.13, 0.53)	1.623×10 <sup>-6</sup>

 $^*\beta$  is expressed as the percentage increase (posterior mean and 95% confidence intervals) in daily mortality associated with a 10  $\mu$ g m<sup>-3</sup> increase in daily PM<sub>2.5</sub>/MDA8 O<sub>3</sub> concentrations.

#### 3 Results and discussions

### 3.1 PM<sub>2.5</sub> and O<sub>3</sub> pollution in Chengdu

According to Chinese ambient air quality standards, PM<sub>2.5</sub> pollution occurs when daily PM<sub>2.5</sub> concentrations are greater than 75 μg m<sup>-3</sup>, and O<sub>3</sub> pollution occurs when MDA8 O<sub>3</sub> concentrations are greater than 160 μg m<sup>-3</sup>. As shown in Figure 2, Chengdu is suffering from severe PM<sub>2.5</sub> and O<sub>3</sub> pollution in recent years. There were 97, 101, 68, 53, 33, 43 and 37 PM<sub>2.5</sub> pollution episodes, and 61, 48, 42, 40, 42, 71 and 48 O<sub>3</sub> pollution episodes in Chengdu from 2015 to 2021. In China, the annual evaluation criterion for PM<sub>2.5</sub> is the annual average concentration, and for O<sub>3</sub> it is the 90<sup>th</sup> percentile of MDA8 O<sub>3</sub> concentration. The annual average concentrations of PM<sub>2.5</sub> were 60.7, 59.9, 52.6, 47.2, 40.6, 40.8 and 40.1 μg m<sup>-3</sup>, and the 90<sup>th</sup> percentile of MDA8 O<sub>3</sub> concentrations were 183.0, 167.0, 168.0, 164.0, 171.5, 188.9 and 167.1 μg m<sup>-3</sup> in Chengdu from 2015 to 2021. This

suggests that  $PM_{2.5}$  pollution improved significantly while  $O_3$  pollution did not.  $O_3$  pollution control in Chengdu should be taken seriously in the future. In addition,  $PM_{2.5}$  and  $O_3$  pollution had clear seasonal preferences, that is,  $PM_{2.5}$  pollution tended to appear in cold months (November to February) while  $O_3$  pollution preferred 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 high temperature and strong sunlight contribute to the elevated  $O_3$  concentrations in warm months.

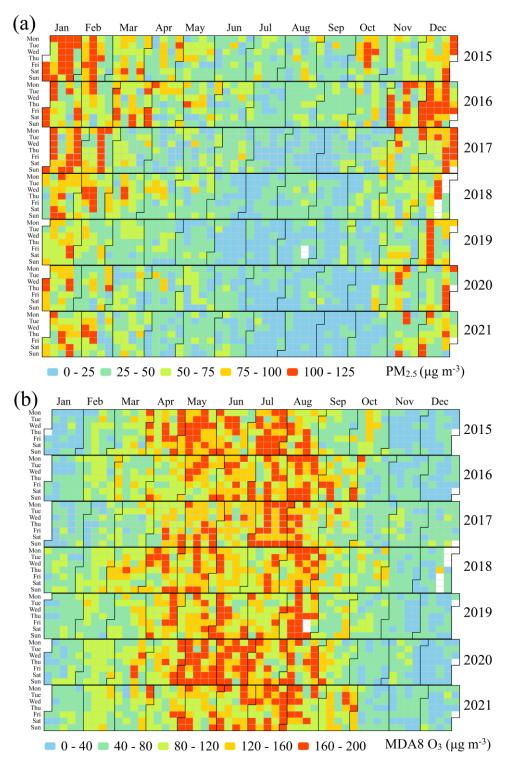


Figure 2. Heat maps of (a) daily  $PM_{2.5}$  and (b) MDA8  $O_3$  concentrations in Chengdu from 2015 to 2021.

# 3.2 Premature mortality attributable to PM<sub>2.5</sub> and O<sub>3</sub>

Severe  $PM_{2.5}$  and  $O_3$  pollution are responsible for a large number of premature mortalities in Chengdu. From 2015 to 2021, the premature mortalities from ANAC due to  $PM_{2.5}$  were 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), 8298 (95%CI: 5759–10402) and 8072 (95%CI: 5606– 10115), with the 7-year annual average of 9386 (95%CI: 6542-11726). The highest health risk among the diseases was from CVD with a 7-year annual average of 2609 (95%CI: 1788-3384), followed by COPD with a 7-year annual average of 1485 (95%CI: 941-1983) and RD with a 7-year annual average of 1321 (95%CI: 804-1840). This was mainly associated with the daily baseline mortality rate of different diseases (Table 3). Although Chengdu's population has been increasing by 24.2% from 2015 to 2021, premature mortalities due to PM<sub>2.5</sub> have generally declined (Figure 3a) owing to reduced PM<sub>2.5</sub> concentrations in recent years (Section 3.1). The premature mortalities from ANAC due to O<sub>3</sub> were 7657 (95%CI: 4345–10672), 8025 (95%CI: 4537–11227), 7870 (95%CI: 4451–11005), 8824 (95%CI: 4967–12397), 7919 (95%CI: 4483-11065), 10085 (95%CI: 5749-13999) and 9163 (95%CI: 5185-12809) from 2015 to 2021, with a 7-year annual average of 8506 (95%CI: 4817–11882), about 90% of that due to PM<sub>2.5</sub>. Unlike the overall reduction in premature mortalities due to PM2.5, the premature mortalities due to O3 increased slightly (Figure 3a), further indicating the urgent need for powerful O<sub>3</sub> control strategies in Chengdu.

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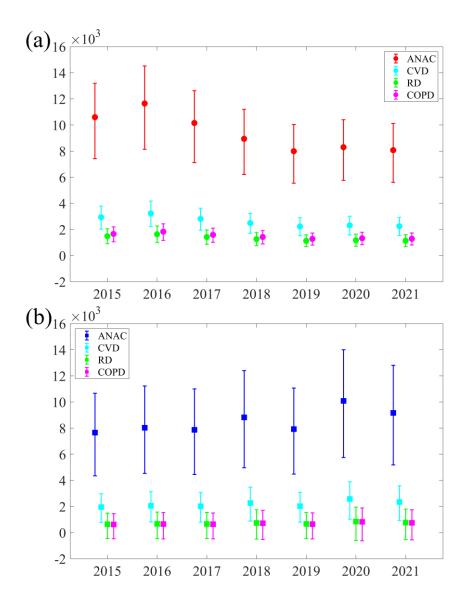


Figure 3. Premature mortality from ANAC, CVD, RD and COPD attributable to (a) PM<sub>2.5</sub> and (b) O<sub>3</sub> in Chengdu from 2015 to 2021. The dots represent the mean estimate, and the whiskers represent 95% confidence intervals.

# 3.3 Impacts of urbanization on PM<sub>2.5</sub> and O<sub>3</sub>

### 3.3.1 Meteorological conditions in January and July

In this study, January and July 2017, when PM<sub>2.5</sub> and O<sub>3</sub> pollution episodes are likely to occur (Figure 2), are selected to study the role of urbanization. In January 2017, Chengdu experienced PM<sub>2.5</sub> pollution for 23 out of 31 days with a monthly average concentration of 128.8 µg m<sup>-3</sup>. From the perspective of atmospheric circulations, westerly winds prevailed over Chengdu due to the large north-south geopotential height gradient at 500 hPa (Figure 4a). However, the westerly winds were

blocked by the Tibetan Plateau and thereby the dispersion of  $PM_{2.5}$  was limited. At 700 hPa, the southwestern air flow originating from the Bay of Bengal could reach Chengdu (Figure 4b). This warm advection was conducive to the formation of a stable layer near 700 hPa (Figure 4c and d), which made the vertical diffusion of  $PM_{2.5}$  difficult. The blocking of air and the stable layer were two important reasons for frequent  $PM_{2.5}$  pollution episodes during this period (Hu et al., 2021; Ning et al., 2018).



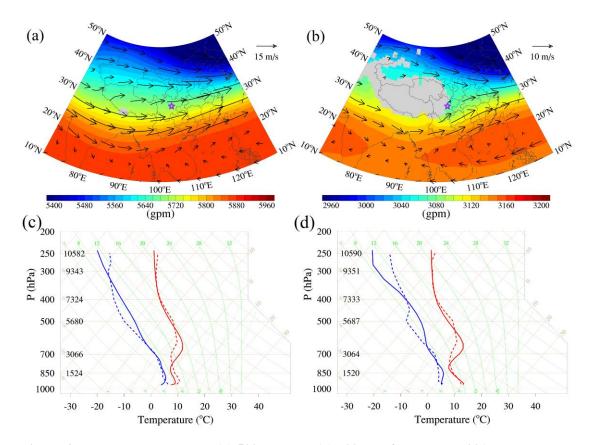


Figure 4. The weather charts at (a) 500 hPa and (b) 700 hPa for January 2017 are based on the NCEP FNL reanalysis data. The purple pentacles show the location of Chengdu. The skew-T diagram at (c) 00:00 UTC and (d) 12:00 UTC in January 2017. The red and blue solid lines are the simulated air temperature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

In July 2017, there were 19 days of O<sub>3</sub> pollution in Chengdu, and the monthly average MDA8 O<sub>3</sub> concentration was 172.9 μg m<sup>-3</sup>. At 500 hPa, Chengdu was dominated by strong high-pressure

systems, and thereby air temperature was high and wind speed was small (Figure 5a). The monthly average  $T_2$  was as high as 28.6 °C while the monthly average  $WS_{10}$  was only 1.6 m s<sup>-1</sup> during this period (Figure 6b). High temperature favored photochemical reactions of  $O_3$  while weak winds trapped  $O_3$ . Furthermore, the thickness of the stable layer in July was far less than that in January (Figure 4c and d; Figure 5c and d). Well-developed boundary layer facilitated vertical mixing of  $O_3$  within the boundary layer, which is an important way to maintain high surface  $O_3$  concentrations during the daytime (Aneja et al., 2000; Tang et al., 2017).



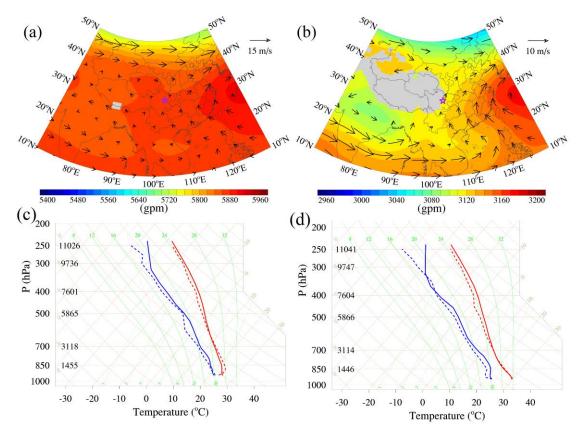


Figure 5. The weather charts at (a) 500 hPa and (b) 700 hPa for July 2017 are based on the NCEP FNL reanalysis data. The purple pentacles show the location of Chengdu. The skew-T diagram at (c) 00:00 UTC and (d) 12:00 UTC in July 2017. The red and blue solid lines are the simulated air temperature and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

# 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 troposphere. 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, simulated variables are compared observed variables, and the results are presented in Figure 6. The mean bias (MB) of the simulated and observed concentrations of PM<sub>2.5</sub> and O<sub>3</sub> are 12.7 μg m<sup>-3</sup> and 11.6 μg m<sup>-3</sup>, with the normalized mean bias (NMB) values of 9.9% and 12.0%, which are within the acceptable standards (NMB  $\leq \pm$  15%). The correlation coefficients (COR) of PM<sub>2.5</sub> and O<sub>3</sub> are 0.44 and 0.77, respectively. The statistical metrics for PM<sub>2.5</sub> and O<sub>3</sub> are similar to those in previous studies (Wang et al., 2022b; Wu et al., 2022), indicating that our model results for PM<sub>2.5</sub> and O<sub>3</sub> are reasonable and acceptable. With regard to the meteorological variables, T<sub>2</sub> 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 underestimate TD2 to some extent with the MB values are -1.5 °C and -2.6 °C in January and July, respectively. As for 10-m wind, poor simulation results are predictable in the case of low wind and complex terrain. The observed calm wind frequency was particularly high due to the starting speed of the anemometer (typically 0.5–1 m s<sup>-1</sup>), resulting in an overestimation of simulated WS<sub>10</sub> as in the studies of other scholars (Shu et al., 2021; Wu et al., 2022). This overestimation could also 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). The model, on the other hand, captures the shift in wind direction except for the case of calm wind. To sum up, the WRF-Chem model using our configuration has a good capability in simulating PM<sub>2.5</sub>, O<sub>3</sub> and meteorological variables in Chengdu, and thereby the simulations can be used for subsequent analysis.

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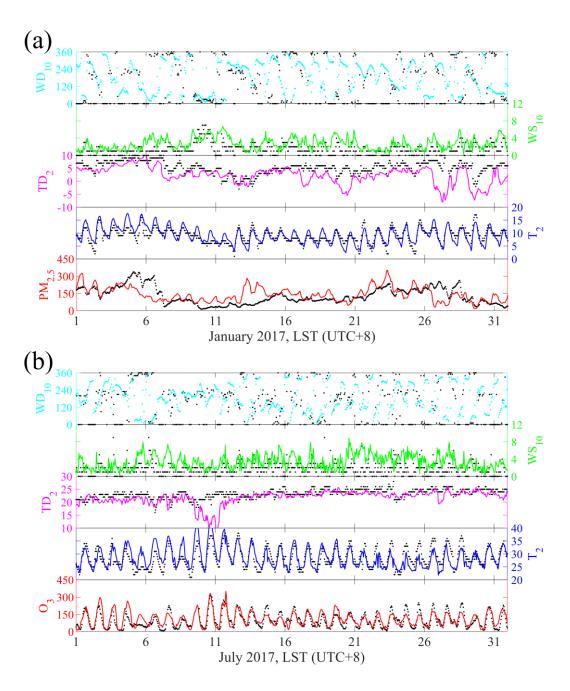


Figure 6. (a) Times series of  $PM_{2.5}$ ,  $T_2$ ,  $TD_2$ ,  $WS_{10}$  and  $WD_{10}$  for January 2017. (b) Times series of  $O_3$ ,  $T_2$ ,  $TD_2$  and  $WS_{10}$  and  $WD_{10}$  for July 2017. The black dots are observations. The colored lines and cyan dots are simulations in baseline simulations.

# 3.3.3 Spatiotemporal variations in PM<sub>2.5</sub> and O<sub>3</sub>

The spatiotemporal characteristics of PM<sub>2.5</sub> were first investigated based on the Jan\_Base simulation. PM<sub>2.5</sub> had a diurnal variation with high concentration at night and low concentration at noon, which was contrary to the boundary layer height (Figure 7a). The nocturnal atmospheric

boundary layer was often characterized by a stable boundary layer, and the boundary layer height was only ~320 m above ground. As a consequence, PM<sub>2.5</sub> was trapped and maintained on the ground. The daytime atmospheric boundary layer, also known as the convective boundary layer, could develop to ~1300 m above ground. Turbulence in the convective boundary layer could dilute PM<sub>2.5</sub> concentrations, resulting in low PM<sub>2.5</sub> 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 wind can easily form. During nighttime, the mountain wind was characterized by westerly and downslope flow at lower levels along the eastern slope of the Tibetan Plateau (Figure 7b and d). Converging with prevailing northeasterly wind, PM<sub>2.5</sub> pollution belt was likely to form and could spread hundreds of kilometers downstream. The daytime plain wind was nearly a reversal of the nighttime circulation, with easterly and upslope flow over the Sichuan Basin (Figure 7c and e). The upslope flow could draw PM<sub>2.5</sub> to a higher elevation, which could also facilitate vertical dispersion of PM<sub>2.5</sub> during the day.

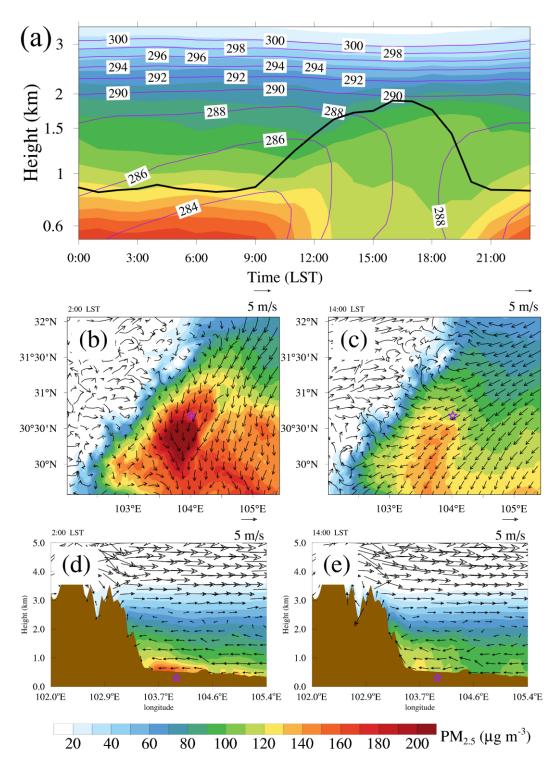


Figure 7. (a) Temporal-vertical cross sections of PM<sub>2.5</sub> (color shading), potential temperature (purple contour lines) and boundary layer height (thick black contour line) at Chengdu. Horizontal distributions of PM<sub>2.5</sub> with wind vectors at the lowest model level at (b) 2:00 and (c) 14:00 LST. East-west vertical cross sections of PM<sub>2.5</sub> 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 Jan\_Base simulation.

In terms of O<sub>3</sub>, it exhibited strong diurnal variation with an afternoon maximum and an early morning minimum (Figure 8a). After sunrise, the nocturnal residual layer was destroyed while the convective boundary layer developed as the surface heated up on account of the incoming radiation. The high-concentration O<sub>3</sub> in the residual layer was then transported downward (Hu et al., 2018). Meanwhile, O<sub>3</sub> could be generated by photochemical reactions between volatile organic compounds (VOCs) and NO<sub>x</sub> in the presence of sunlight. Through these two pathways, surface O<sub>3</sub> concentration increased rapidly in the morning (Zhan and Xie, 2022). By noon, O<sub>3</sub> was mixed within the convective boundary layer via strong turbulence. Strong photochemical production and vertical mixing could maintain high surface O<sub>3</sub> concentrations until late afternoon. The daytime plain wind drove the westward transport of O<sub>3</sub> and aggravated O<sub>3</sub> pollution along the eastern slope of the Tibetan Plateau (Figure 8c and e). After sunset, O<sub>3</sub> production ceased as the intensity of sunlight diminished. O<sub>3</sub> concentrations decreased substantially owing 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 8b). But O<sub>3</sub> in the nocturnal residual layer was still at a high level with values of more than 160 μg m<sup>-3</sup>. The nighttime mountain wind could carry rich-O<sub>3</sub> air eastward and enhanced O<sub>3</sub> concentrations aloft over the eastern slope of the Tibetan Plateau (Figure 8d). Compared with the Jan\_Base simulation, O<sub>3</sub> with a concentration of ~100 μg m<sup>-3</sup> had always existed over the Tibetan Plateau where PM<sub>2.5</sub> concentrations were quite low, indicating that the background concentration of O<sub>3</sub> was much higher than that of PM<sub>2.5</sub>. This can pose a huge challenge to O<sub>3</sub> pollution control in Chengdu.

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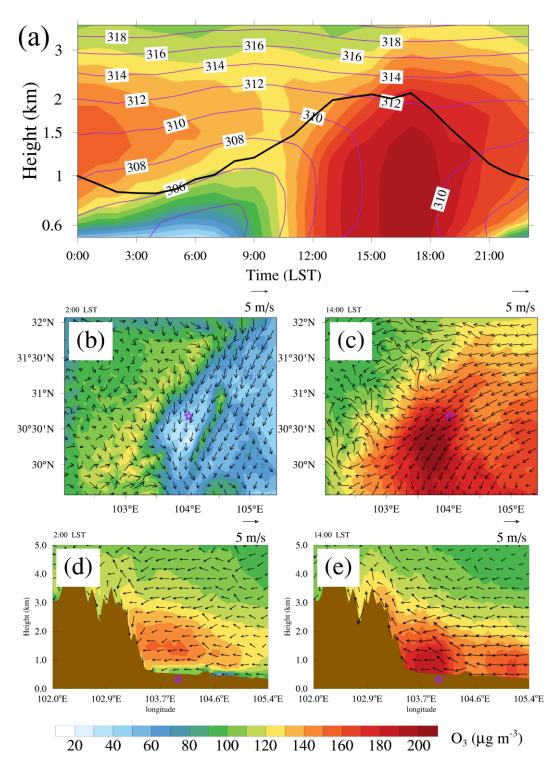


Figure 8. (a) Temporal-vertical cross sections of O<sub>3</sub> (color shading), potential temperature (purple contour lines) and boundary layer height (thick black contour lines) at Chengdu. Horizontal distributions of O<sub>3</sub> with wind vectors at the lowest model level at (b) 2:00 and (c) 14:00 LST. East-west vertical cross sections of O<sub>3</sub> 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.

### 3.3.4 Impacts of urban land use on PM<sub>2.5</sub> and O<sub>3</sub>

Modification of urban land use changes surface dynamic and thermal characteristics, affecting the exchange of energy, moisture and momentum and hence altering urban meteorology and air quality. As illustrated in Figure 9, surface  $PM_{2.5}$  concentrations in Jan\_Base simulation were lower than those in Jan\_noCD simulation, with the monthly average concentrations decreased by 10.8  $\mu$ g m<sup>-3</sup> (7.6%). Moreover, the decrease in  $PM_{2.5}$  concentrations was larger during nighttime than during daytime. The monthly average  $PM_{2.5}$  concentrations decreased by 13.9  $\mu$ g m<sup>-3</sup> (8.6%) at 2:00 LST (LST is UTC+8h) but only 3.0  $\mu$ g m<sup>-3</sup> (2.6%) at 14:00 LST (Figure 9a and b). The decrease in surface  $PM_{2.5}$  concentrations was mainly attributed to the modification of the boundary layer height. Urban land use can enhance surface heating and then increases air temperature. The vertical air movement is then enhanced by the warming up of air temperature, increasing the boundary layer height (Figure S1), which facilitates the vertical diffusion of surface  $PM_{2.5}$ .  $PM_{2.5}$  concentrations increased by 2–6  $\mu$ g m<sup>-3</sup> in the upper boundary layer (~1 km above ground) (Figure 9c and d), further confirming this point.

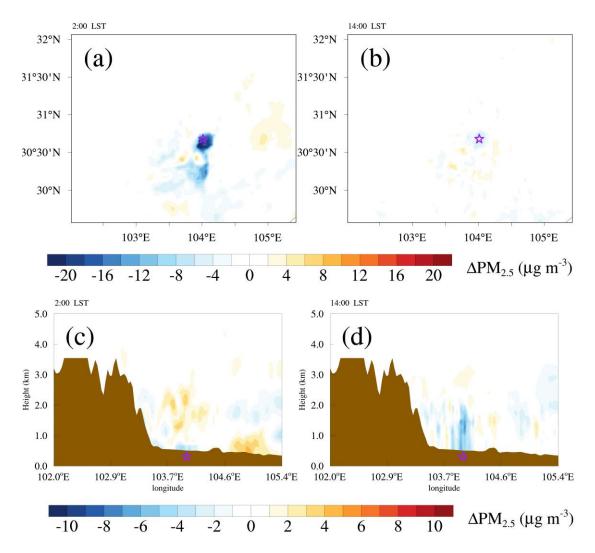


Figure 9. Horizontal distributions of the differences in PM<sub>2.5</sub> at the lowest model level at (a) 2:00 and (b) 14:00 LST. East-west vertical cross sections of the difference in PM<sub>2.5</sub> at (c) 2:00 and (d) 14:00 LST. Purple pentacles show the location of Chengdu. Brown-shaded areas represent 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 its precursors (VOCs and  $NO_x$ ). Due to the increase in upward air movement and boundary layer height induced by urban land use compared to cropland (Figure S2), like  $PM_{2.5}$ ,  $NO_x$  concentrations also decreased near the surface (Liao et al., 2015; Zhu et al., 2017). The decrease in  $NO_x$  near the surface resulted in an increase in surface  $O_3$  at night since the  $NO_x$  titration was weakened (Figure 10a and c). Although the elevated boundary layer diluted  $O_3$  concentrations to some extent, the nighttime  $O_3$ 

concentrations were mainly dominated by chemical effects and increased by 15.6  $\mu$ g m<sup>-3</sup> (16.0%) at 2:00 LST (Figure 10a). During daytime, the increased air temperature was conducive to the photochemical production of O<sub>3</sub>, and the well-developed convective boundary layer favored the vertical mixing of O<sub>3</sub>. O<sub>3</sub> concentrations would also increase (Figure 10b and d), with the monthly average value increasing by 5.4  $\mu$ g m<sup>-3</sup> (4.5%) at 14:00 LST. Since high O<sub>3</sub> concentrations were mainly concentrated in the afternoon, the monthly average MDA8 O<sub>3</sub> concentrations finally increased by 10.6  $\mu$ g m<sup>-3</sup> (6.0%) due to the effects of urban expansion.



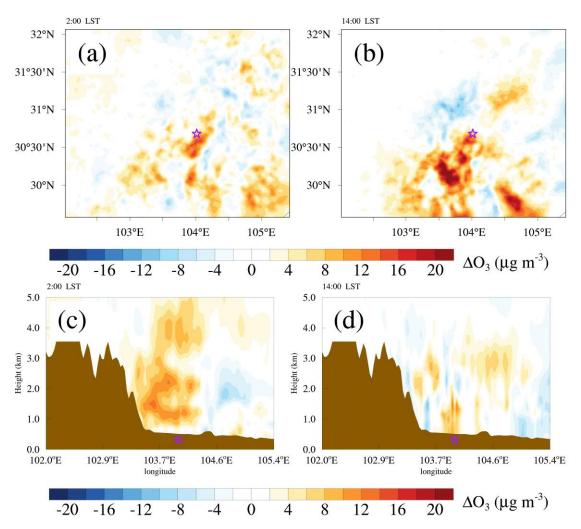


Figure 10. Horizontal distributions of the differences in O<sub>3</sub> at the lowest model level at (a) 2:00 and (b) 14:00 LST. East-west vertical cross sections of the difference in O<sub>3</sub> at (c) 2:00 and (d) 14:00 LST. Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain. These results are the difference between the monthly average of July\_Base and July\_noCD simulations (July\_Base minus July\_noCD).

### 3.3.5 Impacts of anthropogenic emissions on PM<sub>2.5</sub> and O<sub>3</sub>

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. Figure 11 shows the differences in PM<sub>2.5</sub> between the monthly average of Jan\_Base and Jan\_noEmi simulations (Jan\_Base minus Jan\_noEmi). PM<sub>2.5</sub> concentrations in Jan\_Base simulation were significantly higher than those in Jan\_noEmi simulation, with the monthly average concentration increased by 23.9 µg m<sup>-3</sup> (16.8%), more than twice the difference between Jan\_Base and Jan\_noCD simulations. Furthermore, the increases in PM<sub>2.5</sub> concentrations appeared throughout the boundary layer (Figure 11c and d) and could extend downstream for hundreds of kilometers (Figure 11a and b), indicating that reducing anthropogenic emissions is an effective way to reduce PM<sub>2.5</sub> concentrations.

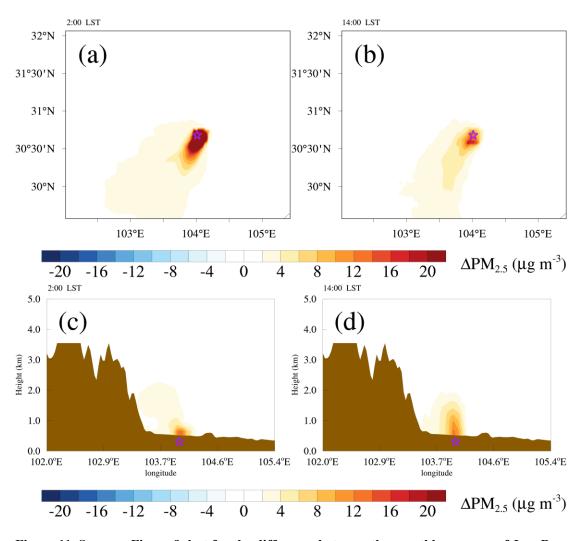


Figure 11. Same as Figure 9, but for the difference between the monthly average of Jan\_Base

## and Jan noEmi simulations (Jan Base minus Jan noEmi).

As for  $O_3$ , the monthly average  $O_3$  concentrations in July\_Base simulation were only 1.6  $\mu g$  m<sup>-3</sup> (1.4%) higher than those in July\_noEmis simulation at 14:00 LST (Figure 12b and d), which was much smaller than the change in PM<sub>2.5</sub>. 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 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), 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. Although the presence of anthropogenic emissions reduced the monthly average  $O_3$  concentrations by 3.0  $\mu$ g m<sup>-3</sup> (3.1%) at 2:00 LST, the monthly average MDA8  $O_3$  concentrations in July\_Base simulation were 4.8  $\mu$ g m<sup>-3</sup> (2.7%) higher than those in July\_noEmis simulation.

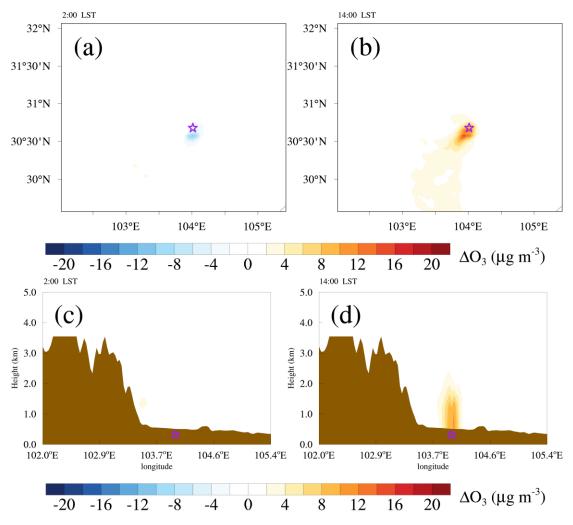


Figure 12. Same as Figure 10, but for the difference between the monthly average of July\_Base and July noEmi simulations (July Base minus July noEmi).

# 3.4 Health risks caused by urbanization

According to the above results, urban land use decreased the monthly average of PM<sub>2.5</sub> concentrations by 10.8 μg m<sup>-3</sup> (7.6%) but increased the monthly average of MDA8 O<sub>3</sub> concentrations by 10.6 μg m<sup>-3</sup> (6.0%). On the other hand, anthropogenic emissions increased both PM<sub>2.5</sub> and MDA8 O<sub>3</sub> concentrations, with monthly average values of 23.9 μg m<sup>-3</sup> (16.8%) and 4.8 μg m<sup>-3</sup> (2.7%), respectively. We then calculate the changes in premature mortalities under different simulation scenarios to assess the health risks from changes in PM<sub>2.5</sub> and O<sub>3</sub> concentrations. As shown in Figure 13, the premature mortalities from ANAC, CVD, RD and COPD due to PM<sub>2.5</sub> decreased by 171 (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%) and 23 (95%CI: 17–26, or about 6.2%) in January 2017 when Chengdu area

was urban land use rather than cropland. On the other hand, anthropogenic emissions in Chengdu increased premature mortalities from ANAC, CVD, RD and COPD due to PM<sub>2.5</sub> by 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%). With regard to O<sub>3</sub>, premature mortalities from O<sub>3</sub>-induced diseases all increased when urban land use and anthropogenic emissions were taken into account. Urban land use led to an increase in premature mortalities from ANAC, CVD, RD and COPD due to O<sub>3</sub> by 203 (95%CI: 122–268, or about 9.5%), 51 (95%CI: 22–71, or about 9.4%), 18 (95%CI: –14–35, or about 10.0%) and 17 (95%CI: –15–33, or about 9.7%) in July 2017 compared to cropland. When anthropogenic emissions in Chengdu were turned on, premature mortalities from ANAC, CVD, RD and COPD due to O<sub>3</sub> increased by 87 (95%CI: 54–112, or about 4.1%), 22 (95%CI: 10–29, or about 4.1%), 8 (95%CI: –7–14, or about 4.4%) and 7 (95%CI: –7–13, or about 4.0%), respectively. In summary, the total premature mortalities due to PM<sub>2.5</sub> and O<sub>3</sub> changed by about –6.9% and 9.5% affected by urban expansion, and these values changed to about 15.7% and 4.1% affected by emissions growth.



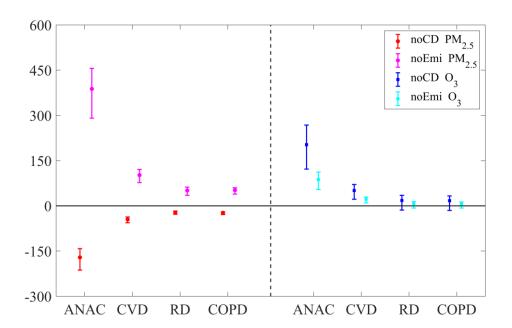


Figure 13. Differences in premature mortality from ANAC, CVD, RD and COPD due to  $PM_{2.5}$  (left of the dotted line) and  $O_3$  (right of the dotted line) between baseline and sensitivity simulations. The dots represent the mean estimate, and the whiskers represent 95% confidence intervals.

#### 4 Conclusions

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495 With the development in urbanization, urban land use and anthropogenic emissions increase, 496 which affects urban air quality and then health risks of air pollutants. In this study, the impacts of 497 urban land use and anthropogenic emissions on air pollutant concentrations and the related health 498 risks in Chengdu, a highly urbanized city with severe air pollution and complex terrain, are 499 quantified. Management of urban air pollution is usually achieved by reducing anthropogenic 500 emissions. Thus, the effects of urban expansion are further compared with those of emissions growth. 501 Chengdu is suffering from severe PM<sub>2.5</sub> and O<sub>3</sub> pollution in recent years. There were 97, 101, 68, 53, 33, 43 and 37 PM<sub>2.5</sub> pollution episodes, and 61, 48, 42, 40, 42, 71 and 48 O<sub>3</sub> pollution 502 episodes from 2015 to 2021. Severe PM<sub>2.5</sub> and O<sub>3</sub> pollution posed huge health risks. The 7-year 503 annual averages of premature mortalities from ANAC, CVD, RD and COPD due to PM2.5 were 504 505 9386 (95%CI: 6542-11726), 2609 (95%CI: 1788-3384), 1321(95%CI: 804-1840) and 1485 506 (95%CI: 941–1983), those due to O<sub>3</sub> were 8506 (95%CI: 4817–11882), 2175 (95%CI: 863–3320), 507 713 (95%CI: -492-1664) and 693 (95%CI: -517-1617). PM<sub>2.5</sub> and O<sub>3</sub> pollution had different 508 seasonal preferences. Owing to the blocking of air and the stable atmospheric layer, PM<sub>2.5</sub> pollution 509 tended to appear in cold months (November to February). However, O<sub>3</sub> pollution was likely to occur 510 in warm months (April to August) because of the high temperature and strong sunlight dominated 511 by high-pressure systems. PM<sub>2.5</sub> concentrations were high at night and low at noon, which was 512 contrary to the boundary layer height. O<sub>3</sub> exhibited strong diurnal variation with an afternoon maximum and an early morning minimum, which was related to photochemical reactions during 513 514 daytime and nitrogen oxide titration at night. 515 The urban land use of Chengdu was replaced by cropland in the WRF-Chem model to examine 516 the impacts of urban expansion. Urban land use led to an increase in air temperature and boundary 517 layer height compared to cropland, and decreased monthly averaged surface PM<sub>2.5</sub> concentrations by 10.8 μg m<sup>-3</sup> (7.6%). Higher temperature and boundary layer height increased O<sub>3</sub> concentrations 518 519 via stronger photochemical reactions and better vertical mixing during daytime. During nighttime, 520 dominated by the weakened chemical NO<sub>x</sub> titration, O<sub>3</sub> concentrations also increased. The monthly averaged MDA8 O<sub>3</sub> concentrations finally increased by 10.6 µg m<sup>-3</sup> (6.0%). In this case, when 521 522 Chengdu area was urban land use rather than cropland, the premature mortalities from ANAC due 523 to PM<sub>2.5</sub> exposure decreased by 171 (95%CI: 129–200, or about 6.9%) but those due to O<sub>3</sub> increased

by 203 (95%CI: 122–268, or about 9.5%). Anthropogenic emissions increased surface PM<sub>2.5</sub> significantly with the monthly average concentration increasing by 23.9 μg m<sup>-3</sup> (16.8%), more than twice the difference caused by urban land use. Owing to the non-linear sensitivity of O<sub>3</sub> to its precursors, O<sub>3</sub> concentrations increased at noon but decreased at night. In particular, the monthly average O<sub>3</sub> concentrations increased by 1.6 μg m<sup>-3</sup> (1.4%) at 14:00 LST but decreased by 3.0 μg m<sup>-3</sup> (3.1%) at 2:00 LST. Since O<sub>3</sub> concentrations in daytime were much higher than those at night, the monthly average MDA8 O<sub>3</sub> concentrations still increased by 4.8 μg m<sup>-3</sup> (2.7%). As a consequence, the premature mortalities from ANAC due to PM<sub>2.5</sub> increased by 388 (95%CI: 291–456, or about 15.7%), and those due to O<sub>3</sub> increased by 87 (95%CI: 54–112, or about 4.1%) with anthropogenic emissions in Chengdu.

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 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 although 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 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.

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