Fine Particulate Matter and Ozone Pollution in the 18 Cities of the Sichuan Basin in Southwestern China: Model Performance and Characteristics

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ABSTRACT

The Sichuan Basin (SCB) is located in southwestern China and has a total population of 108.1 million across 18 cities, including the 2 largest in western China (Chengdu and Chongqing). As most air quality monitoring stations are located in urban areas, we simulated the PM_{2.5} (i.e., particulate matter with an aerodynamic diameter < 2.5 µm) and ozone (O₃) in the entire SCB during winter (December 2014–February 2015) and summer (June–August 2015) by using the Weather Research and Forecasting (WRF) and the Community Multi-scale Air Quality (CMAQ) models. The simulated concentrations of 24-h PM_{2.5} and its major components generally agree with observations during both seasons, but the simulated 1-h and 8-h O₃ are acceptable only for summer. Increasing in severity from the rim of the SCB to its inner areas, the PM_{2.5}, as well as its major components, exhibits hotspots near the central urban areas of Chongqing and Chengdu, with concentrations of 150–200 µg m⁻³ and 40–60 µg m⁻³ during winter and summer, respectively. The 1-h and 8-h O₃ exhibit no hotspots in the urban centers of Chongqing and Chengdu but show elevated levels in some rural and suburban areas (55–70 ppb and 65–80 ppb, respectively), including those on the western and southwestern rim of the SCB, and downwind of the urban center of Chongqing. Despite the great spatial variations in the PM_{2.5} and O₃ concentrations, the vast majority of the basin fails to meet the WHO guidelines for 24-h PM_{2.5} (25 µg m⁻³) and 8-h O₃ (~47 ppb) on > 70% of the days during winter and > 40% of the days during summer, respectively. Based on the aforementioned spatial patterns of the PM_{2.5} and O₃ concentrations, and the wind directions within the basin, strictly controlling emissions originating in the SCB may greatly reduce PM_{2.5} and O₃ concentrations within the basin.

Keywords: Chengdu; Chongqing; Spatio-temporal variations; Air pollution.

INTRODUCTION

Air pollution is one of the most severe environmental problems in China. While haze is still a severe problem in China (Wang and Hao, 2012; Zhang *et al.*, 2012a; Zhang *et al.*, 2012b; Aunan *et al.*, 2018), ozone (O₃) has become an

increasingly important pollutant in recent years (Verstraeten *et al.*, 2015; Anger *et al.*, 2016; Ma *et al.*, 2016; Sun *et al.*, 2016; Wang *et al.*, 2017; Shen *et al.*, 2019a), as a result of increased emissions of volatile organic compounds (VOCs) in the VOC-limited regions (Shen *et al.*, 2019b), decreased nitrogen oxides (NO_x) in some VOC-limited regions (Li *et al.*, 2019), and slowed aerosol sink of hydroperoxy radicals that stimulates O₃ production (Li *et al.*, 2019). Air pollution has caused significant adverse effects on human health. It was estimated that more than 1 million premature deaths each year in China were attributable to outdoor air pollution (Lelieveld *et al.*, 2015). In order to mitigate air

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pollution, the Ministry of Environmental Protection of the People's Republic of China (MEP) has grouped the areas that are most affected by air pollution in the country into 3 regions and 10 city clusters (MEP, 2013; Fig. S1), including (1) Beijing-Tianjin-Hebei (BTH), (2) Yangtze River Delta (YRD), (3) Pearl River Delta (PRD), (4) central regions of Liaoning Province, (5) Shandong Province, (6) Wuhan and its peripheral localities, (7) Changsha, Zhuzhou, and Xiangtan in Hunan Province, (8) the Chengdu-Chongqing city cluster in the Sichuan Basin (SCB), (9) western coast of the Taiwan Strait, (10) central and northern regions of Shansi Province, (11) Guanzhong Plain in Shanxi Province, (12) parts of Gansu and Ningxia Province, and (13) Urumqi in Xinjiang Province. In total, these areas cover 1.32 million km² and have a population of 675 million, accounting for 14% of China's land, 48% of the country's population, 71% of national gross domestic product (GDP), 52% of national coal consumption, and 50% of the national emission of sulfur dioxide (SO₂), NO_x, particulate matter (PM), and volatile organic compounds (VOCs) (MEP, 2013).

Among the 3 regions and 10 city clusters, the Chengdu-Chongqing city cluster in the SCB has the largest area (0.22 million km²) and the second largest population (108.1 million) (MEP, 2013). Within the basin, there are 16 cities belonging to the Chengdu-Chongqing city cluster and the other 2 cities are located in the north of the SCB (Fig. 1). The information of area, population, GDP, and air pollutant concentrations in each city are provided in Table S1. Due to the basin landform and high anthropogenic emissions within the basin, air pollution is severe and the major air pollutants are O₃ and PM_{2.5} (i.e., PM with an aerodynamic diameter < 2.5 μm) (Ning *et al.*, 2018; Zhao *et al.*, 2018a, b; Zhang *et al.*, 2019). In 2015, annual PM_{2.5} concentrations measured in the 18 cities were in the range of 21–74 μg m⁻³ (Table S1), about 2–7 times of the World

Health Organization (WHO) guideline of 10 μg m⁻³ (WHO, 2006). Annual 90th percentiles of 8-h O₃ in the 18 cities were measured in the ranges of 32–86 ppb in 2015, with 5 cities having values over 70 ppb. According to the measurements at the national air quality stations (NAQs; Fig. 1), PM_{2.5} concentrations were highest in winter but lowest in summer, and non-attainment days due to PM_{2.5} pollution were more frequent in winter (Ning *et al.*, 2018; Zhao *et al.*, 2018b). In contrast, O₃ was the major pollutant in summer (Ning *et al.*, 2018; Zhao *et al.*, 2018b). However, as almost all the NAQs are located in the urban and suburban areas, the characteristics of air pollution have not been investigated in detail for the entire basin, particularly for the vast rural areas.

Chemical transport models (CTMs) have been applied to study air pollution for China as a whole (Hu *et al.*, 2016; Li *et al.*, 2016; Zhu and Liao, 2016) and for several regions of China, such as the YRD (Liao *et al.*, 2015), NCP (Wang *et al.*, 2012), and PRD (Fan *et al.*, 2014). In previous nationwide CTM simulations, model performance on the SCB region was evaluated only for Chengdu and Chongqing, as observations in most of the cities in the basin were not available before 2015. For example, Hu *et al.* (2016) applied the Community Multi-scale Air Quality (CMAQ) (Byun and Schere, 2006) and the Weather Research Forecasting (WRF) (University Corporation for Atmospheric Research (UCAR), 2017) modeling system to predict air pollutant concentrations for the whole of China. The results show that 1-h and 8-h O₃ and 24-h PM_{2.5} from March to December 2013 were over-predicted but PM₁₀ (i.e., PM with an aerodynamic diameter < 10 μm) was under-predicted in Chengdu and Chongqing. In addition to the limitations of atmospheric models and emission inventories, the biases in air pollutant predictions might be associated with the coarse horizontal resolution (36 km × 36 km) (Hu *et al.*, 2016).

Fig. 1. (a) Locations of simulation domains and meteorological stations and (b) locations of the 18 cities in the SCB and national air quality stations (NAQs). Almost all the NAQs are located in the 18 SCB urban centers. The upwind and downwind regions are defined based on wind directions within the SCB, as shown in Figs. 5(a) and S4(a). Upwind cities: 1) Bazhong, 2) Dazhou, 3) Guangyuan, 4) Mianyang, and 5) Nanchong. Downwind cities: 6) Chengdu, 7) Deyang, 8) Guang'an, 9) Leshan, 10) Luzhou, 11) Meishan, 12) Neijiang, 13) Suining, 14) Yibin, 15) Zigong, and 16) Ziyang. Other cities: 17) Ya'an and 18) Chongqing.

The SCB is located in the transition region from central China to the Qinghai-Tibetan Plateau (QTP) with great elevation changes; thus, the landform is complex and using coarse horizontal resolutions may not accurately predict wind speed (WS), wind direction (WD), and other meteorological parameters. Given the potential high risk from air pollution to human health, there is a need to use a higher horizontal resolution to systematically model the characteristics, sources, and health impacts of air pollution for the SCB.

In this study, a modified WRF/CMAQ modeling system was used to simulate PM_{2.5} and O₃ in winter (December 2014–February 2015) and summer (June–August 2015) for the entire SCB. The model performance is evaluated, the spatio-temporal variations of the 2 pollutants and PM_{2.5} components are analyzed, the non-attainment conditions are determined, and air pollution is compared among the 18 SCB cities. This is the first study that comprehensively investigates the air quality status as well as variations of criteria air pollutants and PM_{2.5} components for all the SCB cities. This study provides the basis for our other studies on the source apportionment (Qiao *et al.*, 2019), health effects, and control strategies of air pollutants in the SCB.

METHODS AND MATERIALS

Model Description and Application

The WRF/CMAQ modeling system was applied for the SCB using nested domains. The 36-km domain (197 × 129 grid cells) covers all of China and surrounding countries in East Asia, while the 12-km domain (118 × 118 grid cells) covers the entire SCB and its adjacent regions (Fig. 1). Meteorological inputs were generated using the WRF model version 3.9 with initial and boundary conditions (IC and BC) from the U.S. National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Model Global Tropospheric Analyses dataset (1° × 1° resolution; available at <http://dss.ucar.edu/datazone/dsszone/ds083.2/>). Detailed WRF model configurations are listed in Table S2. The CMAQ model version 5.0.1 was used to model air pollutant concentrations and modifications were made to improve the prediction of secondary inorganic and organic aerosols, including (1) providing more detailed treatment of isoprene oxidation chemistry SARPC-11 gas-phase photochemical mechanism (Ying *et al.*, 2015); (2) allowing surface-controlled reactive uptake of dicarbonyls, isoprene epoxydiol (IEPOX), and methacrylic acid epoxide (Crippa *et al.*, 2018) for secondary organic aerosol (SOA) formation (Li *et al.*, 2015; Ying *et al.*, 2015); (3) updating SOA yields based on vapor wall-loss correction (Zhang *et al.*, 2014); and (4) improving nitrate and sulfate formation through heterogeneous reactions of NO₂ and SO₂ (Wang *et al.*, 2014). More details of these modifications can be found in the cited references and the references therein. The model has been used in some studies for Asian countries (Hu *et al.*, 2016; Kota *et al.*, 2018).

Other input files used to run the CMAQ model include the IC and BC of chemicals and the anthropogenic and natural emissions. The default CMAQ IC and BC profiles were used for the 36-km simulation and the outputs of the

36-km simulation were used to generate the chemical IC and BC files for the 12-km simulations. Anthropogenic emissions files were generated based on the Emission Database for Global Atmospheric Research (EDGAR) version 4.3, which provided the datasets of carbon monoxide (CO), NO_x, SO₂, ammonia (NH₃), VOCs, PM_{2.5}, PM₁₀, elemental carbon (EC), and organic carbon (OC) (Crippa *et al.*, 2018). As the EDGAR database provides only monthly total emissions of air pollutants, an in-house preprocessor was used to generate hourly emissions based on monthly, weekly, and temporal allocation profiles as mentioned in Wang *et al.* (2014) and references within. Biogenic emission files were simulated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther *et al.*, 2012). Open burning emission files were generated by using the Fire Inventory from the National Center for Atmospheric Research (NCAR) (FINN) version 1.5 (Wiedinmyer *et al.*, 2011). Dust and sea salt emission files were generated in-line, and the dust emission module was updated to be compatible with the 20-category Moderate Resolution Imaging Spectroradiometer (MODIS) land use data (Hu *et al.*, 2016). The re-gridded emissions of individual species were mapped to model species needed by the SAPRC photochemical mechanism (Carter, 2010) and the AERO6 aerosol module. As shown in Fig. S2, anthropogenic emissions are much higher inside the SCB than in its neighboring regions. Seasonally, PM_{2.5}, SO₂, and NO_x emissions are higher in winter than in summer, but NH₃ emissions are generally similar between the two seasons.

Model Validation

Hourly observations of meteorological parameters and air pollutants were used to evaluate the model performance. The observations of wind speed (WS) and wind direction (WD) at 10 m above ground level (a.g.l.), ambient air temperature at 2 m a.g.l. (T2), and relative humidity (RH) were obtained from the National Climate Data Center (NCDC; <ftp://ftp.ncdc.noaa.gov/pub/data/noaa/>, last accessed on June 20, 2018). The meteorological data were available at 1,434 and 101 stations in the 36-km and 12-km domains, respectively (Fig. 1). PM_{2.5} and O₃ measurements were derived from the Air Quality Data Distribution Platform of China (<http://106.37.208.233:20035/>, last accessed on July 13, 2018), which provided data at 94 NAQs in the SCB (Fig. 1). Almost all the NAQs are located in the urban and suburban areas. Quality control measures were taken to remove possible problematic data points with observed hourly O₃ and PM_{2.5} greater than 250 ppb and 1500 µg m⁻³, respectively. The data points with a standard deviation of less than 5 ppb for O₃ and 5 µg m⁻³ for PM_{2.5} within 24 hours were also removed. To calculate 24-h PM_{2.5} for a given day in a city, citywide hourly PM_{2.5} data should be available for at least 20 hours (MEP, 2012a). To calculate 1-h and 8-h O₃ for a given day in a city, citywide hourly values should be available for at least 14 hours between 08:00 and 24:00 (MEP, 2012b). If hourly O₃ concentrations are available for < 14 hours but the calculated 1-h O₃ values are greater than the Chinese National Ambient Air

Quality Standards (CNAAQs), the 1-h O_3 values are still valid (MEP, 2012b).

RESULTS AND DISCUSSION

Model Validation

Meteorology

In order to assess the WRF model performance, the statistical metrics, including mean bias (MB), root mean square error (RMSE), and gross error (GE), were calculated based on the equations in Table S3 and then compared with the benchmarks suggested by Emery *et al.* (2001). When using the benchmarks to assess model performance, it should be noted that the benchmarks are considered as quantitative measures to compare with previous studies and model performance is affected by model parameterizations, landforms, meteorological reanalysis data, etc. As shown in Table S4, RH and T2 are both under-predicted (MBs < 0), and the under-predictions are greater in winter (MBs for RH and T2: -10.8 to -6.5% and -0.9 to -0.5 K, respectively) than in summer (MBs for RH and T2: -2.9 to -1.1% and -0.3 to -0.1 K, respectively). Compared to the benchmarks, the statistics of T2 are within the MB benchmark ($< \pm 0.5$ K) only in summer and there are no benchmarks for RH. In the two seasons, the MBs and RMSEs of WS (0.5 – 1.1 m s^{-1} and 1.8 – 2.3) are slightly larger than the benchmarks ($< \pm 0.5$ m s^{-1} and < 2.0 , respectively), but the GEs of WS meet the benchmark value (< 2.0 m s^{-1}) in both seasons. The MBs of WD (-5° to 6°) achieve the benchmark ($< \pm 10^\circ$), but its GEs (54 – 61°) are larger than the benchmark ($< 30^\circ$). Overall, the WRF model performance is acceptable and comparable to previous studies in China (Hu *et al.*, 2016; Hu *et al.*, 2017a). The biases in the WRF simulations can be partially explained by the complex landforms in the 12-km domain, which is located in the transition region from central China (mostly < 1000 m above sea level [a.s.l.]) to the QTP (> 3000 m a.s.l.) and the spatial resolution of the modeling.

PM_{2.5} and its Components

Fig. 2 shows the time series of simulated and observed 24-h PM_{2.5} in the urban centers of the 18 cities (i.e., the center of the NAQs in the urban area of a city) and Table S5 shows relevant evaluation statistics, including normalized mean biases (NMBs), normalized mean errors (NMEs), fractional biases (FBs), and fractional errors (FEs). In general, both the simulated and observed 24-h PM_{2.5} are much higher in winter than in summer in the urban centers, and the simulations capture the peaks of 24-h PM_{2.5} most of the time (Fig. 2). The NMBs of PM_{2.5} meet the criteria ($< \pm 30\%$; Emery *et al.*, 2017) in all the urban centers in both seasons, except for that of Guanyuan (41%), Mianyang (37%), Meishan (31%), Ziyang (48%), and Chongqing (42%) in winter and the NMB of Dazhou (-39%) in summer (Table S4). The 24-h PM_{2.5} simulations meet the goals of NME ($< 35\%$), FB ($< \pm 30\%$), and FE ($< 50\%$) in all the cities in both seasons, except for the NME of Ziyang (58%) in winter (Table S4). The possible causes leading to the uncertainties in PM_{2.5} simulations include but are not

limited to the bias in the emission inventories, uncertainties in the meteorological modeling, complex landforms, and spatial resolutions of the modeling. However, the time series in Fig. 2 and the statistics in Table S5 give confidence in the predictions of 24-h PM_{2.5} in both seasons for the SCB.

The model performance on PM_{2.5} components (including sulfate, nitrate, and ammonia ions (SO_4^{2-} , NO_3^- , and NH_4^+ , respectively), OC; EC; and others) is assessed for the urban centers of Chengdu and Chongqing (Fig. 3), as observations are available from literature for the 2 cities. In Chengdu, the two largest contributors to PM_{2.5} are simulated to be secondary inorganic aerosols (SIA; including SO_4^{2-} , NO_3^- , and NH_4^+ here; $\sim 40\%$ in total) and OC (28–34%), while the simulated EC fractions are $\sim 5\%$ in both seasons. These simulations are similar to the observations from Chen *et al.* (2016), which reported that SIA, OC, and EC accounted for 45%, 23–30%, and 5% of PM_{2.5} from 2012 to 2013, respectively. Within SIA, the simulated contributions from SO_4^{2-} , NO_3^- , and NH_4^+ to PM_{2.5} are 21%/30%, 12%/2%, and 10%/8% in Chengdu in winter/summer, and these values also agree with the observations of 20%/25%, 15%/8%, and $\sim 10\%$ (Chen *et al.*, 2016), respectively. Shi *et al.* (2017) reported SO_4^{2-} , NO_3^- , OC, and EC contributions (25%, 8%, 13%, and 7% in winter and 23%, 4%, 12%, and 9% in summer, respectively) similar to our simulations. In Chongqing, SIA and OC are also simulated to be the largest contributors to PM_{2.5}. The simulated SIA fractions in Chongqing (35% and 30% in winter and summer, respectively) are lower than the observations (45% and 50% in winter and summer, respectively) from Chen *et al.* (2017) but are higher than the annual mean fraction of 29% reported by Yang *et al.* (2011). The simulated OC fractions in Chongqing (38% and 33% in winter and summer, respectively) are higher than the observations (33% and 25% in winter and summer, respectively) from Chen *et al.* (2017) and are close to the annual mean of 33% reported by Yang *et al.* (2011). The EC fractions in Chongqing are similar between simulations ($\sim 12\%$) and observations ($\sim 10\%$; Chen *et al.*, 2012) in the two seasons. The discrepancies between the simulations and observations on PM_{2.5} components might be associated with the following: (1) The periods of observations and simulations are different; (2) emission inventories, meteorological modeling, etc. lead to the uncertainties of simulations on PM_{2.5} components; (3) locations of PM_{2.5} component measurements are not in the urban centers; and (4) concentrations of PM_{2.5} components may vary greatly within a urban center or a modeling grid cell (12 km $\times$ 12 km).

1-h and 8-h O_3

The time series of 8-h O_3 in the 18 urban centers are presented in Fig. 4, and the evaluation statistics of 8-h and 1-h O_3 are presented in Tables S6 and S7, respectively. Only 1-h and 8-h O_3 concentrations larger than 30 ppb are used to calculate NMB and NME, as a cutoff of 30–60 ppb is suggested by the U.S. Environmental Protection Agency (U.S. EPA, 2005), Hu *et al.* (2016), and Emery *et al.* (2017). The 8-h O_3 concentrations above 30 ppb in summer

Fig. 2. Simulated and observed 24-h $\text{PM}_{2.5}$ concentrations in the 18 SCB urban centers in summer (June–August 2015) and winter (December 2014–February 2015). “Obs.” shows observations. “Pred.” indicates the simulations at the grid cells covering the urban centers, while “Best” indicates the simulated values closest to the observations within 3×3 grid cell regions that surround the urban center.

are mostly captured in the cities, except for Ya'an (Fig. 4). Also, the simulated diurnal patterns in summer well replicate the time and concentrations of O_3 peaks in 11 cities in summer (Fig. S3). The NMBs and NMEs of 1-h and 8-h O_3 for summer in all the 18 cities achieve the criteria (NMB $< \pm 15\%$ and NME $< 25\%$) recommended by Emery *et al.* (2017), except for the NMBs and NMEs of 1-h and 8-h O_3 in Ya'an and the NMBs of 8-h O_3 in Zigong and Suining (Tables S6 and S7). However, 1-h and 8-h O_3 are both greatly over-predicted in winter, with NMBs and NMEs of 20–79%, almost all of which do not meet the corresponding criteria ($\pm 15\%$ and 25%, respectively). Thus, the model performance on 1-h and 8-h O_3 is acceptable for summer, when the photochemical production of O_3 due to anthropogenic emissions should be strongest in the basin.

Spatial Variations: Air Pollutant Concentrations and Non-attainment Days

$\text{PM}_{2.5}$ and its Components

Spatial distributions of $\text{PM}_{2.5}$, $\text{PM}_{2.5}$ components, and

wind vectors are presented in Fig. 5 for winter and Fig. S4 for summer. The entire SCB experiences much higher $\text{PM}_{2.5}$ concentrations in winter (~ 50 – $200 \mu\text{g m}^{-3}$) than in summer (~ 5 – $60 \mu\text{g m}^{-3}$). The higher $\text{PM}_{2.5}$ concentrations in winter are partially due to the lower amount and frequency of precipitation and lower wind speed in the season according to observations (Ning *et al.*, 2018; Zhao *et al.*, 2018b). In general, $\text{PM}_{2.5}$ concentrations are much lower in the rims of the SCB ($< 75 \mu\text{g m}^{-3}$ and $< 20 \mu\text{g m}^{-3}$ in winter and summer, respectively) than in the inner SCB (100 – $200 \mu\text{g m}^{-3}$ and 20 – $60 \mu\text{g m}^{-3}$ in winter and summer, respectively). Two hotspots of $\text{PM}_{2.5}$ concentrations are found and they cover the urban centers of Chengdu and Chongqing and their adjacent regions in both seasons and have $\text{PM}_{2.5}$ concentrations of 150 – $200 \mu\text{g m}^{-3}$ in winter and 40 – $60 \mu\text{g m}^{-3}$ in summer. SO_4^{2-} , NO_3^- , NH_4^+ , primary organic aerosol (POA), and EC present spatial variations similar to that of $\text{PM}_{2.5}$ in both seasons. In contrast, no hotspots are found for SOA. In winter, SOA concentrations are also lower in the rims of the SCB ($< 4 \mu\text{g m}^{-3}$) than in

the inner areas ($4\text{--}10\ \mu\text{g m}^{-3}$), except that the southwestern rim of the SCB also has higher values ($6\text{--}10\ \mu\text{g m}^{-3}$). In summer, higher SOA concentrations ($4\text{--}8\ \mu\text{g m}^{-3}$) are

found in the inner and southwestern SCB, as well as some regions in the north and east of the SCB. In summary, $\text{PM}_{2.5}$ and most of its major components have concentrations

Fig. 3. Comparison of $\text{PM}_{2.5}$ components in Chengdu and Chongqing between the observations and simulations. The simulations of this study are the averages in the winter (December 2014–February 2015) and in the summer (June–August 2015).

Fig. 4. Simulated and observed 8-h O_3 concentrations in the 18 SCB urban centers in summer (June–August 2015) and winter (December 2014–February 2015). “Obs.” shows observations. “Pred.” indicates the simulations at the grid cells covering the urban centers, while “Best” indicates the simulated values closest to the observations within 3×3 grid cell regions that surround the urban center.

Fig. 5. Spatial variations of simulated wind vectors, $\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$), and $\text{PM}_{2.5}$ components ($\mu\text{g m}^{-3}$) in winter (December 2014–February 2015) over the SCB. The left and right crosses show the urban centers of Chengdu and Chongqing, respectively.

increasing from the rims to the inner areas of the SCB, have two hotspots near the urban centers of Chengdu and Chongqing, and present decreasing gradients from the hotspots of Chengdu and Chongqing to their downwind areas. The above spatial variations may reflect not just the emissions and accumulation of air pollutants within the SCB but also the trans-boundary transport of air pollutants. In a companion paper, we have revealed that 25–52% and 39–66% of $\text{PM}_{2.5}$ in each urban center are due to non-local emissions in summer and winter, respectively, and emissions outside the SCB can contribute up to 70% of $\text{PM}_{2.5}$ in winter for the eastern SCB rims where $\text{PM}_{2.5}$ concentrations are relatively low compared to that of other SCB regions (Qiao *et al.*, 2019).

The percentage of non-attainment days (i.e., the days that do not meet the CNAQS of $75 \mu\text{g m}^{-3}$ or the WHO guideline of $25 \mu\text{g m}^{-3}$) is shown in Fig. 6. In summer, only the $\text{PM}_{2.5}$ hotspots located near the central cities of Chengdu and Chongqing have non-attainment days > 10% based on the CNAQS. However, over half the area of the basin has $\text{PM}_{2.5}$ exceeding the WHO guideline for > 20% of summer days, and > 80% of summer days near the $\text{PM}_{2.5}$ hotspots do not meet the WHO guideline. In winter, over half the area of the basin has $\text{PM}_{2.5}$ exceeding the CNAQS for > 40% days and almost the entire basin has $\text{PM}_{2.5}$ larger than the WHO guideline for > 70% of winter days. All of the above suggests that the entire basin has $\text{PM}_{2.5}$ pollution based on the WHO guideline in winter.

1-h and 8-h O_3

Spatial variations of O_3 concentrations and the percentages of non-attainment days in summer are presented in Fig. 7. 1-h and 8-h O_3 concentrations are higher in the western and southwestern SCB and downwind from the urban center of Chongqing, with average concentrations of 65–80 ppb and 55–70 ppb, respectively. Only in some of these higher- O_3 areas do 8-h and 1-h O_3 exceed the corresponding

CNAQS for 10–40% and > 5% of days in summer, respectively. However, almost the entire basin and over half of the basin have 8-h O_3 concentrations larger than the WHO guideline (~ 47 ppb) for > 40% and > 60% of summer days, respectively. Although the trans-boundary transport of O_3 and its precursors has not been quantified yet, we have found that over 30% of the O_3 concentration in the forested areas of the western SCB from 09:00 to 15:00 in August 2013 is associated with anthropogenic NO_x emissions (Qiao *et al.*, 2019b), and this reflects the transport of O_3 precursors from urban to rural areas.

Comparison of the 18 Cities

In both summer and winter, near-surface winds intrude on the SCB from the north, east, and southeast and then travel anti-clockwise within the basin (Figs. 5 and S4). Based on the wind vectors, we approximately categorized the 18 cities into 3 groups (Fig. 1), including upwind cities (Bazhong, Dazhou, Guangyuan, and Mianyang), downwind cities (Chengdu, Deyang, Guangan, Leshan, Luzhou, Meishan, Nanchong, Neijiang, Suining, Yibin, Zigong, and Ziyang), and others (Ya'an and Chongqing).

The citywide average concentrations and attainments of $\text{PM}_{2.5}$ are compared among the 18 cities in Fig. 8. Citywide average $\text{PM}_{2.5}$ concentrations are generally higher in the downwind cities ($106.5 \pm 13.6 \mu\text{g m}^{-3}$ and $24.3 \pm 4.9 \mu\text{g m}^{-3}$) than in the upwind cities ($62.6 \pm 15.9 \mu\text{g m}^{-3}$ and $15.6 \pm 2.7 \mu\text{g m}^{-3}$) and the other cities ($16.0 \pm 10.5 \mu\text{g m}^{-3}$ and $66.0 \pm 28.9 \mu\text{g m}^{-3}$) in winter and summer, respectively. In summer, 24-h $\text{PM}_{2.5}$ concentrations in the 18 cities exceed the CNAQS for < 5% days but are higher than the WHO guideline for 12–55% days (except Ya'an). In winter, the percentages of non-attainment days are also much lower based on the CNAQS (18–80%) than based on the WHO guideline (65–100%). Although $\text{PM}_{2.5}$ concentration and its attainment both vary greatly among the cities, POA and SO_4^{2-} are the largest contributors to $\text{PM}_{2.5}$ in the 18 cities

Fig. 6. The percentage of the days that do not meet the CNAAQs ($75 \mu\text{g m}^{-3}$) and WHO guideline ($25 \mu\text{g m}^{-3}$) for 24-h $\text{PM}_{2.5}$ concentrations in summer (June–August 2015) and winter (December 2014–February 2015). The black lines show the SCB city boundaries. The left and right crosses show the urban centers of Chengdu and Chongqing, respectively.

Fig. 7. (a) 8-h O_3 concentrations and wind vectors, (b) 1-h O_3 concentrations, (c–d) percentages of the days that do not meet the CNAAQs and WHO guideline for 8-h O_3 , and (e) percentage of the days that do not meet the CNAAQs for 1-h O_3 in summer 2015. The concentrations and percentages are in $\mu\text{g m}^{-3}$ and %, respectively. The black lines show the SCB city boundaries. The left and right crosses show the urban centers of Chengdu and Chongqing, respectively.

in winter, accounting for 26–35% and 19–30% of $\text{PM}_{2.5}$, respectively (Fig. 9). In summer, SO_4^{2-} is still one of the most important contributors with contributions of 23–30% in the urban centers. Although the contributions from SOA (20–33%) in summer are similar to or as important as that from POA (19–24%) in the urban centers of Bazhong, Dazhou, Guangyuan, Leshan, Suining, Yibin, and Ya'an, these cities have average $\text{PM}_{2.5}$ concentrations lower than $30 \mu\text{g m}^{-3}$ in the season. In contrast to that of SOA, the contributions from NO_3^- decrease from 6–15% in winter to 1–8% in the 18 cities in summer, as NO_3^- would be easily transformed into NO_x when the air temperature is over 30°C (Chen *et al.*, 2003). Therefore, in order to reduce $\text{PM}_{2.5}$ concentrations in the basin, we suggest that POA and SO_4^{2-} should receive priority for being controlled in both seasons.

Citywide concentrations of 1-h and 8-h O_3 are slightly

higher in the downwind cities (58.9 ± 2.8 ppb and 54.7 ± 2.2 ppb) than in the upwind (53.5 ± 2.7 ppb and 50.6 ± 2.3 ppb) and other (55.0 ± 3.5 ppb and 52.0 ± 3.1 ppb) cities in summer, respectively (Fig. 8). 8-h O_3 concentrations in the cities do not meet the WHO guideline for 54–88% of summer days and are higher than the CNAAQs for 0–12% of summer days. 1-h O_3 does not meet the CNAAQs for < 5% of summer days in the 18 cities and there is no WHO guideline for 1-h O_3 . As citywide 8-h O_3 concentrations in the 18 cities exceed the WHO guideline for 50% of summer days, there is a need to develop relevant control measures for the basin.

CONCLUSIONS

In this study, the WRF/CMAQ modeling system was used to simulate the $\text{PM}_{2.5}$ and O_3 during winter and

Fig. 8. The citywide average concentrations and non-attainment days of (a) summer 24-h $\text{PM}_{2.5}$, (b) winter 24-h $\text{PM}_{2.5}$, (c) summer 8-h O_3 , and (d) summer 1-h O_3 in the 18 SCB cities.

Fig. 9. Percentage contributions of different components to the total $\text{PM}_{2.5}$ concentrations in the 18 SCB urban centers in summer (June–August 2015) and winter (December 2014–February 2015).

summer in the SCB, which contains the 2 largest cities in western China (namely, Chengdu and Chongqing). The $\text{PM}_{2.5}$ increases in concentration from the rim of the basin ($25\text{--}100\text{ }\mu\text{g m}^{-3}$ and $5\text{--}20\text{ }\mu\text{g m}^{-3}$ during winter and summer, respectively) to the inner areas ($100\text{--}200\text{ }\mu\text{g m}^{-3}$ and $20\text{--}60\text{ }\mu\text{g m}^{-3}$ during winter and summer, respectively)

and exhibits 2 hotspots ($150\text{--}200\text{ }\mu\text{g m}^{-3}$ and $40\text{--}60\text{ }\mu\text{g m}^{-3}$ during winter and summer, respectively) near the urban centers of Chengdu and Chongqing. Decreasing gradients for the $\text{PM}_{2.5}$ concentration are found in the regions downwind of these urban centers, reflecting the transport of air pollutants within the basin. POA and SO_4^{2-} are the

largest contributors to the PM_{2.5} during both winter and summer, with SOA being a third significant factor during the latter. Although noticeable hotspots are absent, the overall 1-h and 8-h O₃ concentrations (55–70 ppb and 65–80 ppb, respectively) are higher in some rural and suburban areas, including those located on the western and southwestern rim of the SCB and in regions downwind of the urban center of Chongqing. In spite of the large spatial variations in the PM_{2.5} and O₃ concentrations, the vast majority of the basin exceeds the WHO guidelines for 24-h PM_{2.5} and 8-h O₃ on > 70% of winter days and > 40% of summer days, respectively. Although the basin landform favors the accumulation of air pollutants within the SCB, it inhibits the transportation of pollution from outside. Thus, strictly controlling emissions originating inside the basin may greatly reduce PM_{2.5} and O₃ in this region.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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