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Revealing the origin of fine particulate matter in the Sichuan Basin from a source-oriented modeling perspective

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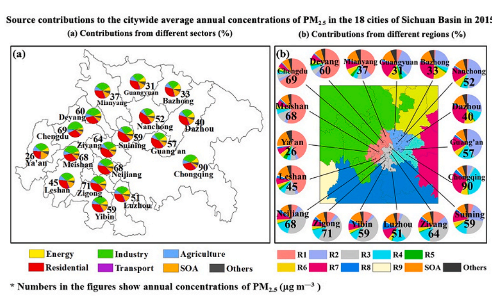
HIGHLIGHTS

- Annual $PM_{2.5}$ concentrations exceed China's standard in ~60% of the SCB areas.
- >60% of annual $PM_{2.5}$ in the urban SCB are from industrial and residential sources.
- SCB emissions contribute 20–86 μg m^{-3} to annual $PM_{2.5}$ in the urban SCB.
- Non-SCB emissions contribute up to 87 μg m^{-3} to 24-h $PM_{2.5}$ in the urban SCB.
- Non-SCB emissions should be also controlled for the SCB to meet $PM_{2.5}$ standards.

GRAPHICAL ABSTRACT



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ABSTRACT

The Sichuan Basin (SCB) with 18 cities is one of the regions that are greatly affected by $PM_{2.5}$ (i.e., particulate matter (PM) with an aerodynamic equivalent diameter less than or equal to 2.5 μm) in China. In this study, we used the Weather Research Forecasting (WRF) model and a source-oriented version of the Community Multiscale Air Quality (CMAQ) model to quantify the contributions from different sectors and regions to $PM_{2.5}$ for the SCB in 2015. The annual $PM_{2.5}$ concentrations in the 18 SCB urban centers (i.e., the central urban areas) are 42–112 μg m^{-3}, much higher than the World Health Organization (WHO) guideline (10 μg m^{-3}) and having 20–86, 6–17, and 6–10 μg m^{-3} due to SCB, non-SCB, and unidentified emissions, respectively. Non-SCB emissions can contribute up to 87 μg m^{-3} to 24-h $PM_{2.5}$ concentrations for an urban center. Industrial and residential activities are the largest sectors for annual $PM_{2.5}$ concentrations in the urban centers, and each of them contributes ~25%–50%. The combined residential and industrial contributions (>~60%) are always much higher than that from each of the other sources on $PM_{2.5}$ pollution and extreme pollution days (>75 and > 150 μg m^{-3}, respectively). This study suggests that China's standard for annual $PM_{2.5}$ (35 μg m^{-3}) in most of the SCB cities might be

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achieved mainly through controlling SCB emissions (particularly those from industrial and residential activities); however, to meet the WHO guideline and to reduce $PM_{2.5}$ pollution days and extreme pollution days, both SCB and non-SCB emissions should be greatly reduced.

1. Introduction

Many cities located in basins and valleys are suffering from air pollution across the world, such as those in the Mediterranean Basin in Europe (Kanakidou et al., 2011), the Los Angeles Basin (Langford et al., 2010) and Uinta Basin (Whiteman et al., 2014) in the United States, the Guanzhong Basin (Bei et al., 2017) and Lanzhou city (Chu et al., 2008) in China, and the Kathmandu Valley in Nepal (Panday and Prinn, 2009). In order to improve air quality in the basins and valleys, there is a need to quantify the source contributions to air pollutant concentrations, including how much are due to local and non-local emissions and how much are due to different sectors (e.g., industries, agriculture, power plants, transportation, and residential activites). The Sichuan Basin (SCB) in southwestern China are one of these regions (Fig. 1; Ning et al., 2018a, 2018b).

The SCB has a total area of 0.26 million km^2 and a total population of 67.5 million in 2017 (National Bureau of Statistics of China (NBSC), 2018). Within the SCB, there are 18 cities and the Chengdu−Chongqing Region (CCR, including 15 SCB cities) is the most developed region in western China. The CCR is also one of the Chinese regions that are largely affected by air pollution with ozone (O_3) and particulate matter with an aerodynamic equivalent diameter less than or equal to 2.5 μm ($PM_{2.5}$) as the major pollutants. In 2017, annual average $PM_{2.5}$ concentrations (21–66 μg m^{-3}) were observed much higher than the World Health Organization (WHO) guideline (10 μg m^{-3}) in all the 18 SCB cities and were higher than the Chinese National Ambient Air Quality Standard (CNAAQS; 35 μg m^{-3}) in 16 SCB cities (NBSC, 2018). Among these cities, ten had annual $PM_{2.5}$ concentrations higher than 50 μg m^{-3} in 2017 (NBSC, 2018), and extreme $PM_{2.5}$ events (24-h $PM_{2.5}$>150 μg m^{-3}) still occurred in the SCB (Chen et al., 2019). $PM_{2.5}$ concentrations in the SCB have large spatial and seasonal variations, with two hotspots near the urban areas of Chengdu and Chongqing (Qiao et al., 2019a) and with the highest and lowest concentrations in winter and summer, respectively (Ning et al., 2018, 2018b).

To formulate air pollution control strategies, it is imperative to understand the sources of air pollutants, including the partition of various sectors (e.g., industries, power plants, transportation, and residential activities). Some studies have revealed source contributions to $PM_{2.5}$ for a few SCB cities or the entire SCB by using receptor-based models or chemical transport models (CTMs), but the source contribution results to $PM_{2.5}$ varied greatly among studies (Tian et al., 2013; Zhang et al., 2013a; Chen et al., 2017; Shi et al., 2017). From 2010 to 2011 and based on the Positive Matrix Factorization (PMF) modeling, crustal dust, biomass burning, vehicular emissions, secondary sulfate and nitrate aerosols accounted for 14%, 28%, 24%, and 31% of the annual average $PM_{2.5}$ concentration in Chengdu, respectively (Zhang et al., 2013a). From 2009 to 2011 in urban Chengdu, the contributions from soil dust, cement dust, vehicular exhaust, secondary sulfate, and secondary nitrate were 30%, 5%, 25%, 25%, and 9%, respectively, according to a PMF-HCA study (i.e., Hierarchical Clustering Analysis; Tian et al., 2013). A PMF modeling study reported that windblown dust, vehicular emissions, and secondary aerosols were the largest contributors in urban Chongqing in 2012 (~15%, ~30%, and ~30%, respectively; Chen et al., 2017). Based on a source-oriented Community Multi-scale Air Quality (CMAQ) model simulation for 2013, residential, industrial, and agricultural activities were the largest sources and contributed 27%, 25%, and 11% to the annual average $PM_{2.5}$ concentration in Sichuan Province, the east and west of which are the 17 SCB cities and the rural QTP, respectively. Residential, industrial, and agricultural activities were also estimated to be the largest contributors in Chongqing Municipality in 2013 (25%, 29%, and 13%, respectively; Shi et al., 2017). The annual contributions from transportation were≤10% in Chengdu and Chongqing (Shi et al., 2017). The aforementioned large differences in the source apportionment results among studies might be associated with that (1) CTMs studies used horizontal spatial resolutions of several kilometers, and this might under-estimate the transportation contributions for central urban areas; (2) receptor-based modeling studies used observations made in central urban areas, where transportation emissions might be higher; (3) there were certain uncertainties in the emission inventories; and (4) receptor-based determine source

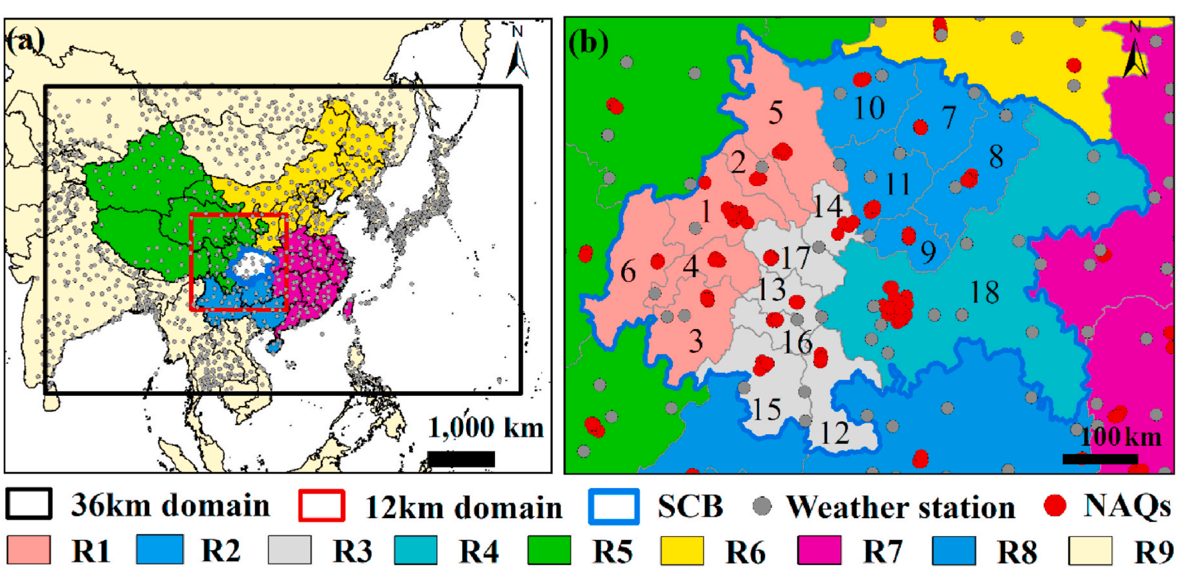

Fig. 1. (a) Locations of the simulation domains and meteorological stations and (b) locations of the nine emission regions and national air quality stations (NAQs). Regions 1–4 (R1-4) and R5-9 are the cities within and outside the SCB. The SCB cities are: 1-Chengdu, 2-Deyang, 3-Leshan, 4-Meishan, 5-Mianyang, 6-Ya'an, 7-Bazhong, 8-Dazhou, 9-Guang'an, 10-Guangyuan, 11-Nanchong, 12-Luzhou, 13-Neijiang, 14-Suining, 15-Yibin, 16-Zigong, 17-Ziyang, and 18-Chongqing.

contributions from factors, each of which may include several sectors or a partial contribution from an individual sector.

To improve air quality on a regional scale, it is also important to understand how much pollutants are from local and non-local emissions. By using the HYSPLIT and PSCF models (i.e., Hybrid Single Particle Lagrangian Integrated Trajectory and Potential Source Contribution Function, respectively), studies have revealed the impacts on Chengdu and Chongqing's air quality from air pollutant transport, such as dust storms from northwestern China (Zhao et al., 2010), inter-city transport within the SCB (Liao et al., 2017; Qiao et al., 2019b), and biomass burning in rural areas (Chen and Xie, 2014). Even in winter when precipitation and wind speed were lowest among the four seasons, 21%–51% and 39%–66% of citywide average $PM_{2.5}$ concentrations in each of the 18 SCB cities were due to local and non-local emissions, respectively (Qiao et al., 2019b). However, there is a lack of systematic analyses on the contributions from different sectors and regions specific for each SCB city under different $PM_{2.5}$ pollution conditions for an entire year.

Therefore, we used the Weather Research Forecasting (WRF) model and a source-oriented CMAQ model to systematically understand the sources of $PM_{2.5}$ for all the SCB cities for the entire year of 2015. The main objective of this study is to quantify the contributions to $PM_{2.5}$ and its major components from different sectors and regions under different $PM_{2.5}$ concentrations in the SCB, particularly for $PM_{2.5}$ pollution days (24-h $PM_{2.5}$ >75 μg m^{-3}) and extreme pollution days (24-h $PM_{2.5}$ >150 μg m^{-3}). As the SCB is surrounded by the mountains and plateaus which may inhibit air pollutants to transport into and out of the SCB, we hypothesize that it might be sufficient to make $PM_{2.5}$ concentrations in the 18 SCB cities achieve the Chinese national standards and the WHO guidelines through controlling the SCB emissions alone. The methodology of this study would be helpful for other cities in basins and valleys to quantify air pollutants' sources.

2. Materials and methods

2.1. Model description

The Weather Research Forecasting model (WRF; version 3.9.1; Skamarock et al., 2008) and a source-oriented version of the CMAQ model (based on version 5.0.1; Zhang et al., 2012; Qi et al., 2014; Hu et al., 2015; Shi et al., 2017; Qiao et al., 2019a) are used to simulate meteorological conditions and air pollutants, respectively. The model was modified to improve the prediction of secondary inorganic and organic aerosols and the modifications have been described in Qiao et al. (2019a) and references therein. This version of the source-oriented CMAQ model is capable of simultaneously tracking primary particulate matter (PPM) and secondary inorganic aerosols (SIA, including ammonia, sulfate, and nitrate ions (NH_4^+, SO_4^{2-}, and NO_3^-, respectively)) from nine sectors or regions. For PPM, non-reactive tracers are used to track the PPM emitted from different sectors/regions. For SIA, multiple reactive source-tagged gas and particle-phase species are added to represent the species from different emission sources. Aerosol and cloud modules are also expanded to include these tagged species. For example, NO_2_X1 and NH_3_X2 represent NO_2 and NH_3 released from two sectors or regions, respectively, and the two species can form (NH_4_X2) (NO_3_X1) through the following reactions: NO_2_X1 + OH → HNO_3_X1 and NH_3_X2 + HNO_3_X1 → (NH_4_X2) (NO_3_X1). Secondary Organic Aerosols (SOA) are included but its source contributions are not determined in the model, as SOA contributions to total $PM_{2.5}$ concentrations are relatively lower than that of PPM and SIA, particularly on $PM_{2.5}$ pollution days (see section 3.1.2).

2.2. Model application

Two nested domains are used to quantify source contributions to $PM_{2.5}$, PPM, and SIA for the 18 SCB cities for 2015 (Fig. 1a). The horizontal resolutions of the parent and nested domains are 36 km and 12 km, respectively. The overall modeling height is 20 km and is divided into 18 layers, with the layer closest to the land surface of ~35 m. Meteorological inputs, emissions files, and initial and boundary conditions (ICs and BCs) are used to run the source-oriented CMAQ model. Meteorological inputs were generated using the WRF model and the 6-hourly FNL (Final) Operational Global Reanalysis data from the U.S. National Center for Atmospheric Research (NCAR) with a spatial resolution of 1.0° × 1.0° (https://rda.ucar.edu/datasets/ds083.2/). The configurations of the WRF model can be found at the online supplementary materials of Qiao et al. (2019a). Biogenic emissions were estimated using the Emissions of Gases and Aerosols from Nature (MEGAN) model (version 2.1) and the WRF simulation results (Guenther et al., 2012). Emissions from open burning were estimated based on the Fire Inventory from the National Center for Atmospheric Research (NCAR FINN; Wiedinmyer et al., 2011). Windblown dust and sea salt emissions were determined on-line with the CMAQ simulations. Default ICs and BCs files were used for the 36-km simulations, and the outputs of the 36-km simulations were used to generate ICs and BCs for the 12-km simulations. For anthropogenic emissions, the Emission Database for Global Atmospheric Research (EDGAR) v4.3.1 was used (Crippa et al., 2018). The EDGAR inventory has a horizontal resolution of 0.1° × 0.1° and includes the air pollutant species of nitrogen oxides (NO_x), sulfur dioxide (SO_2), ammonia (NH_3), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), $PM_{2.5}$, PM_{10} (PM with an aerodynamic equivalent diameter less than or equal to 10 μm), elemental carbon (EC), and organic carbon (OC). The EDGAR emissions are grouped into energy, industries, residential activities, on-road and off-road transportation, and agriculture. More details about the emission file preparation can be found in Qiao et al. (2019a, 2019b). The EDGAR is used because that (1) we have found the model performance on $PM_{2.5}$ is acceptable for the SCB in our previous studies (Qiao et al., 2019a, 2019b) and (2) Hu et al. (2017) have already compared and discussed the simulations on $PM_{2.5}$ for entire China among different emission inventories, including EDGAR, Multi-resolution Emission Inventory for China (MEIC; Li et al., 2017), Regional Emission inventory in Asia version 2 (REAS2; Kurokawa et al., 2013), and the Emission Inventory for China by the School of Environment at Tsinghua University (SOE). The source apportionment results of this study are compared with those studies at the national scale based on the MEIC inventory (Ying et al., 2014; Hu et al., 2015; Shi et al., 2015; Qiao et al., 2018).

The domains and emissions are classified into nine regions, as the source-oriented CMAQ model is capable of tracking air pollutants from up to nine regions. Based on the wind direction (WD), $PM_{2.5}$ concentrations, and emission densities (Qiao et al., 2019a), the SCB areas are classified into four regions. Winds intrude the SCB mainly from the northern and eastern SCB, and these regions also have relatively lower emission densities, except for the central urban areas of Chongqing (Qiao et al., 2019a), thus the five northern cities and Chongqing are classified into Region 2 (R2) and R4, respectively. R1 includes the six cities in the western rim of the SCB and they have one of the two hotspots of $PM_{2.5}$ concentration in the SCB. R3 includes the five cities in the southern and central SCB, as winds travel anti-clockwise in the SCB and form a cyclone in R3. Based on the WD near the SCB and the spatial distributions of air pollutant emissions across China (Qiao et al., 2019a; Li et al., 2017), areas outside the SCB are categorized into R5, R6, R7, and R8, which are to the west, northeast, east, and south of the SCB, respectively. R9 includes other countries.

2.3. Model performance assessment

Meteorological parameters and $PM_{2.5}$ concentrations are compared between simulations and observations by using the indices, including Mean Bias (MB), Gross Error (GE), Root Mean Square Error (RMSE), Normalized Mean Bias (NMB), Normalized Mean Error (NME), Fractional Bias (FB), and Fractional Error (FE). The equations of these indices can be found in Table S1. To assess the WRF model performance, we used the ambient air temperature (T2) and relative humidity (RH) measured at ~2 m above ground level and the wind speed (WS) and wind direction (WD) measured at ~10 m above ground level from 101 national meteorological stations in the 12-km domain (National Climate Data Center (NCDC),ftp://ftp.ncdc.noaa.gov/pub/data/noaa/; Fig. 1a). To assess model performance on 24-h $PM_{2.5}$, hourly $PM_{2.5}$ concentrations were obtained from the 94 national air quality stations (NAQs) in the SCB (Air Quality Data Distribution Platform of China, http://106.37.208.233:20035/; Fig. 1b). As almost all the NAQs are located in the urban and sub-urban areas, citywide hourly concentrations and the citywide 24-h $PM_{2.5}$ concentrations for each city were calculated following the guidelines of the Ministry of Environmental Protection (MEP, 2012a, b).

3. Results and discussion

3.1. Model performance

3.1.1. Meteorology

The results of WRF model performance assessment for each month are shown in Table S2. To compare the WRF model performance between this and other studies, we used the benchmarks suggested by Emery et al. (2001). When using the benchmarks, it should keep in mind that they are based on the simulations with a variety of horizontal resolutions, model configurations, terrains, and periods, while the WRF model performance is affected by these factors. In this study, T2 is under-predicted and has MB and GE values (−1.0 to −0.2 K and 2.2–3.0%, respectively) exceeding the benchmarks (<±0.5 K and <2.0%, respectively) in nine and all the months. The MB values of WS (0.6–1.4 m s^{-1}) are higher than the benchmark (<±0.5 m s^{-1}) in all the months. The GE values of WS (1.4–2.1%) exceed the benchmark of <2.0% in three months, while the RMSE values (1.9–2.8 m s^{-1}) do not meet the benchmark of <2.0 m s^{-1} in ten months. The MB values of WD (−0.4°–10.2°) are within the benchmark (10°) in all the months except for February, but the GE values (52.5°–60.9°) in all the months are higher than the benchmark (30°). RH is under-predicted in all the months, with MB values of −7.3% to −1.6%. However, the WRF performance of this study is comparable to that of other studies for China (Wang et al., 2010, 2014; Zhao et al., 2013; Hu et al., 2016) and reflects WRF's current capability in reproducing the observed meteorological conditions for the SCB.

3.1.2. $PM_{2.5}$ and its components

Concentrations of 24-h $PM_{2.5}$ are compared between simulations and observations for the 18 SCB urban centers. The coordinate of a city's urban center is defined by averaging the coordinates of NAQs in the city's urban and suburban areas, as almost all the NAQs are located in urban and suburban areas. Fig. S1 shows that the simulated time-series of 24-h $PM_{2.5}$ in the 18 SCB urban centers are in good agreement with the observations and can capture the observed peaks most of the time. In Tables S2–S6, the values of NMB, NME, FB, and FE for the entire 2015 and for each season are compared with the simulation goals and criteria suggested by Emery et al. (2016). Goals indicate the statistical values that about a third of top-performing past applications have met and are the best a model can be expected to achieve (Boylan et al., 2006; Emery et al., 2016). Criteria are the statistical values that about two-thirds of past applications have met and are the accuracy that is considered to be acceptable for modeling applications (Boylan et al., 2006; Emery et al., 2016). As shown in Table S2, the simulations for the entire year meet the criteria or goals of NMB, NME, FB, and FE in all the 18 SCB cities, except for the NMB values of Chengdu, Guangyuan, Ziyang, and Chongqing and the NME of Ziyang. Seasonally, the model performance is best in spring and summer, with all the four indices achieving relevant criteria and goals in all the cities, except for Guangyuan's NMB in spring and Dazhou's NMB in summer. For fall and winter, the FB and FE values in all the cities achieve the criteria or the goals, but 9/6 cities in fall and 7/3 cities in winter do not meet the NMB/NME criteria. Overall, the model performance for the SCB is good for spring and summer and is acceptable for fall and winter.

The model performance on the major $PM_{2.5}$ components (including SIA, OC, EC, and others) is evaluated for Chengdu and Chongqing in Fig. S2, as observations are available from literature only for the two cities. Chengdu and Chongqing are the top two developed cities in the SCB and where the hotspots of annual $PM_{2.5}$ concentrations within the SCB are found (Fig. 2). Although the observations were made in 2011 and 2013, the major components in the two cities may not change significantly from 2011 to 2015. OC and SO_4^{2-} are simulated to be the largest contributors to annual $PM_{2.5}$ concentrations in the two cities (~25% from each component). This is in good agreement with the observations from Chen et al. (2016, 2017a), which found that OC and SO_4^{2-} accounted for ~30% and ~25% in the two cities, respectively. The simulated NO_3^- fraction is ~8% in Chengdu, lower than ~13%, which was observed by Zhang et al. (2013) and Chen (2016). The simulated fraction of NO_3^- is ~3% in Chongqing, the same as the observations from Yang et al. (2011) but lower than ~10%, which was reported by Chen et al. (2017a). The fractions of NH_4^+ are simulated to be 9% and 6% in Chengdu and Chongqing, respectively, and these values are close to the observations from Chen et al. (2016) and Yang et al. (2011). The EC fractions are simulated and observed to be ~5% in both cities. Overall, the model performance on the major $PM_{2.5}$ components is acceptable for Chengdu and Chongqing, and more observations are needed to assess model performance for other SCB cities.

3.2. Characteristics of $PM_{2.5}$ pollution

3.2.1. Spatial and seasonal variations

The annual and seasonal average $PM_{2.5}$ concentrations with wind vectors across the SCB are presented in Fig. 2 and Fig. S3, respectively. Annual average $PM_{2.5}$ concentrations increase from the rims (<40 μg m^{-3}) to the inner SCB areas (mostly in the range of 40–120 μg m^{-3}), with two hotspots (>80 μg m^{-3}) covering the urban centers of Chongqing and Chengdu and their adjacent regions (Fig. 2a). About 60% and all the SCB areas do not achieve the CNAAQS standard (35 μg m^{-3}) and the WHO guideline (10 μg m^{-3}) for annual average $PM_{2.5}$ concentration, respectively. The seasonal average $PM_{2.5}$ concentrations are highest and lowest in winter and summer, respectively (Fig. S3). For example, seasonal average $PM_{2.5}$ concentrations in the urban center of Chengdu are 66, 44, 76, and 133 μg m^{-3} in spring, summer, fall, and winter, respectively. In winter, 24-h $PM_{2.5}$ concentrations do not achieve the WHO guideline (25 μg m^{-3}) on >80% days in almost all the SCB areas and the concentrations do not meet the CNAAQS standard (75 μg m^{-3}) on >50% days in about half of the SCB areas (Fig. S3). In summer, 24-h $PM_{2.5}$ concentrations in most of the SCB areas meet the CNAAQS standard for >80% days, but the concentration in about 40% of the SCB areas do not achieve the WHO guideline for >50% days.

3.2.2. $PM_{2.5}$ components

The average composition of $PM_{2.5}$ in 2015 and under different 24-h $PM_{2.5}$ concentration ranges are presented in Fig. S4 for the SCB urban centers. The 24-h $PM_{2.5}$ concentration ranges are <25, 25–75, 76–115, 116–150, and >150 μg m^{-3}, which are used to classify air quality levels in China. In each SCB urban center, primary organic aerosol (POA) and SO_4^{2-} are the largest components of $PM_{2.5}$, having annual contributions of 15%–20% and 21%–30%, respectively, while the annual contributions from NO_3^-, NH_4^+, and SOA are ~5%–13% each. EC has the lowest contributions to annual average $PM_{2.5}$ concentrations in the urban centers (~5%). As 24-h $PM_{2.5}$ concentrations increase from <25 to >150 μg m^{-3}, the total contributions of SIA increase from 25%-40% to 37%–51% in the urban centers. Specifically, the NO_3^- contributions increase to ~15% and 6% in the urban centers of R1-3 and R4, respectively, while the NH_4^+ contributions increase to ~10% but the SO_4^{2-} contributions may either increase or decrease in the R1-4 urban centers.

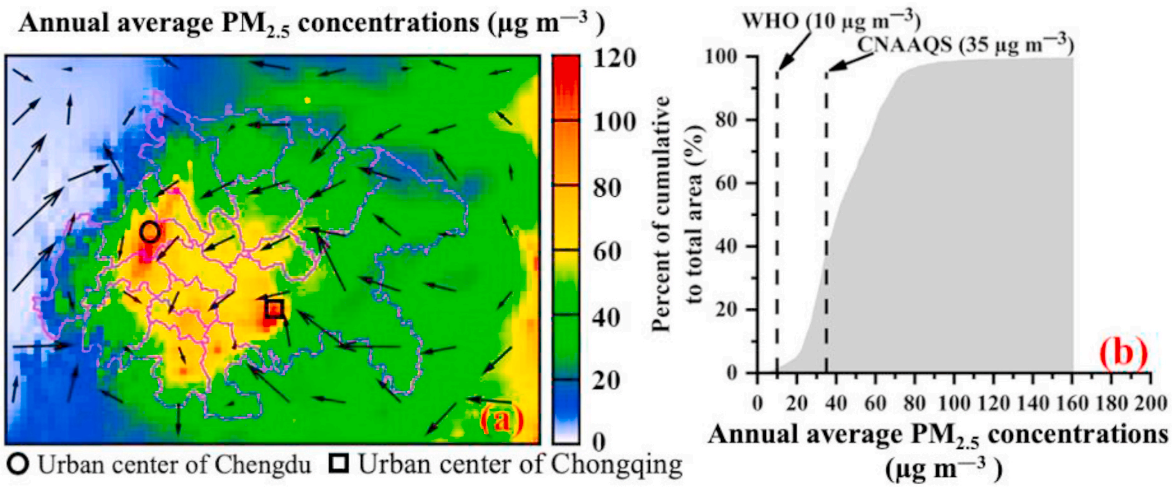

Fig. 2. (a) Spatial distributions of simulated annual average $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$) and (b) the percentage of cumulative to total area of annual average $PM_{2.5}$ concentrations in 2015. The black arrows and purple lines in (a) are wind vectors and the boundaries of each SCB city, respectively. The dash lines in (b) show the WHO guideline and the CNAAQS standard for annual $PM_{2.5}$ concentration.n

In contrast, the SOA contributions decrease as 24-h $PM_{2.5}$ concentrations increase, and they are ~5% when 24-h $PM_{2.5}$ are >75 $\mu g\ m^{-3}$. The contributions of POA and EC in the R1-4 urban centers are similar among the five 24-h $PM_{2.5}$ ranges (~18% and ~5%, respectively). All the above suggest that to reduce the $PM_{2.5}$ pollution days and the extreme pollution days, POA and SIA should be largely controlled.

3.3. Contributions from different sectors

3.3.1. Annual and seasonal averages

Fig. 3 and Fig. S5 show the contributions from different sectors to annual and seasonal average $PM_{2.5}$ concentrations for each of the SCB urban centers, respectively, as the urban centers have higher population densities and source contributions may have considerable spatial variations. Industrial and residential activities are the largest contributors to annual $PM_{2.5}$ concentrations (each about 20%–45%), followed by SOA (4%–9%), agriculture (~10%), and energy (5%–11%; Fig. 3). Transportation and other sources (including windblown dust, sea salt, biogenic emissions, and open burning) only account for ≤3% and ≤7% of the annual concentrations, respectively (Fig. 3). The fractional contributions from the sectors are similar among the four seasons, except that (1) SOA contributions are higher in summer (~10%–30%) than in other seasons (<10%) and (2) residential contributions are lower in summer (~15%–20%) than in other seasons (~20%–40%). However, the seasonal SOA contributions higher than 15% only occurred in the cities with summer $PM_{2.5}$ concentrations less than 30 $\mu g\ m^{-3}$. The higher SOA and lower residential contributions in summer are likely associated with higher biogenic emissions and lower residential energy use in the season, respectively. Thus, residential and industrial emissions should be the main sources to be controlled, and this is also supported by the source apportionment studies, which used the MEIC emission inventories (Qiao et al., 2018; Shi et al., 2018).

As the contributions from different sectors to $PM_{2.5}$ may have considerable spatial variations within each SCB city, the citywide average contributions from different sectors to annual concentrations of $PM_{2.5}$, PPM, and SIA are presented in Figs. S6–S8. Residential and industrial activities are still the largest sources of $PM_{2.5}$ (22%–42% and 23%–44%, respectively), and all the other sources have a combined contribution of 28%–38% in each city (Fig. S6). However, the sector contributions are different between PPM and SIA, as shown in Figs. S7 and S8 and supported by the studies based on the MEIC inventories (Qiao et al., 2018; Shi et al., 2018). For PPM, the annual citywide

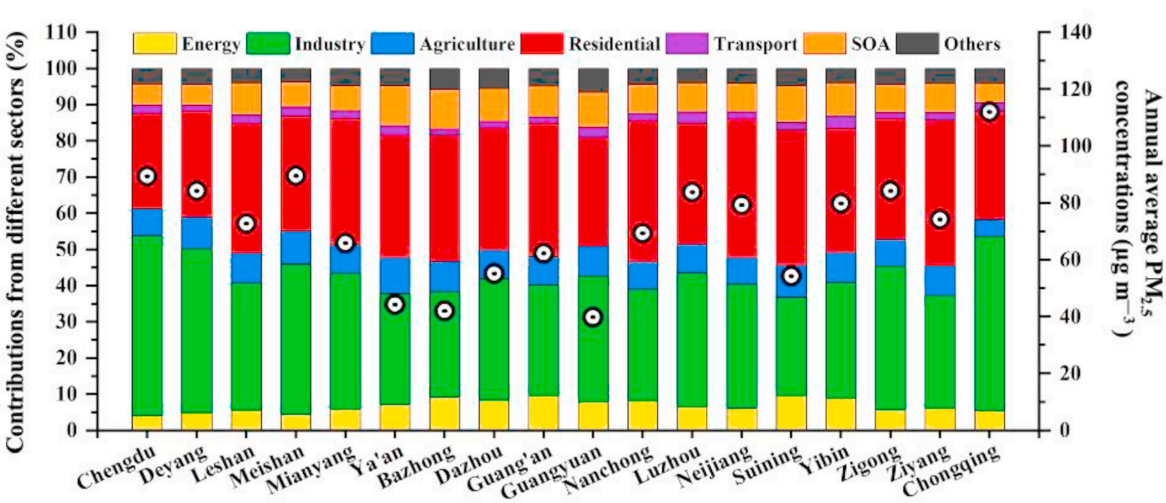

Fig. 3. Contributions from different sectors to annual average $PM_{2.5}$ concentrations in the 18 SCB urban centers in 2015. The bars and circles show the fractional contributions (%) and annual average $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$), respectively. Others include ICs, BCs, windblown dust, sea salt, biogenic emission, and open burning.

average contributions from residential and industrial activities are 48%–74% and 19%–48%, respectively, and the combined contributions from all the other sectors are ≤13% (Fig. S7). For SIA, the annual citywide average contributions are due to industries (33%–48%), followed by agriculture (~20%), residential activities (~15%), and energy (10%–22%; Fig. S8). Thus, to reduce annual citywide average concentrations of either $PM_{2.5}$, PPM, or SIA, industrial and residential emissions are the main sources to be controlled, and agricultural emissions should be also controlled to reduce SIA.

3.3.2. Contributions under different 24-h $PM_{2.5}$ concentrations

As industrial and residential activities are the largest contributors, we present the contributions from the two sectors and combine the contributions from all the other sources for each day in Fig. 4. When 24-h $PM_{2.5}$ concentrations in the R1-R4 urban centers increase from <25 to >150 μg m^{-3}, the contributions of residential activities increase from 13%-26% to 35%–43%; in contrast, the contributions due to industries decrease from 48%, 38%, 39%, and 60%–39%, 29%, 32%, and 45% in the R1-4 urban centers, respectively (Fig. 4). Fig. S9 shows that residential and industrial contributions are also much higher than that of other sources on the pollution days in each season, except that (1) there are no identified pollution days in R2 and R3 in summer, (2) industries is the sole dominant source on the summer pollution days in R4, and (3) residential, SOA, and industrial contributions are 18%, 25%, and 45% on the sole summer pollution day in R1 (Fig. S9). Thus, residential and industrial emissions should be the main sources to be controlled to prevent pollution days.

3.4. Contributions from different regions

3.4.1. Annual and seasonal averages

Spatial variations in the SCB and non-SCB contributions to annual average $PM_{2.5}$ concentrations are presented in Fig. 5, and the contributions in each season are shown in Fig. S10. The SCB emissions have very limited impacts on annual $PM_{2.5}$ concentrations outside the SCB (<~10 μg m^{-3}; Fig. 5a). While, the impacts from non-SCB emissions on the annual $PM_{2.5}$ concentrations in the SCB decrease from ~30 to <5 μg m^{-3} from the eastern to western SCB (Fig. 5be), as the QTP to the west of the SCB has low anthropogenic emissions. In general, the annual $PM_{2.5}$ concentrations due to SCB emissions are higher than the CNAAQS annual standard (35 μg m^{-3}) and the WHO guideline (10 μg m^{-3}) in ~30% and ~70% of the SCB areas, respectively (Fig. 5d). The annual $PM_{2.5}$ concentrations due to non-SCB emissions do not exceed the CNAAQS standard in almost all the SCB but are higher than the WHO guideline in ~70% of the SCB areas (Fig. 5e). Others (including ICs, BCs, windblown dust, sea salt, and SOA) have combined contributions mostly in the range of 5–10 μg m^{-3} across the SCB. The seasonal spatial patterns (Fig. S10) are similar to that to the annual pattern (Fig. 5a–c), and the seasonal concentrations due to SCB and non-SCB emissions are higher in winter (up to ~140 and ~50 μg m^{-3}, respectively) than in the other seasons. Thus, to achieve the CNAAQS standard (35 μg m^{-3}), it is more important to reduce SCB emissions than non-SCB emissions; However, to achieve the WHO guideline (10 μg m^{-3}), both SCB and non-SCB emissions should be greatly controlled.

The citywide average regional contributions to annual concentrations of $PM_{2.5}$, PPM, and SIA are presented in Figs. S6–S8. These results also suggest that the fractional contributions from non-SCB emissions

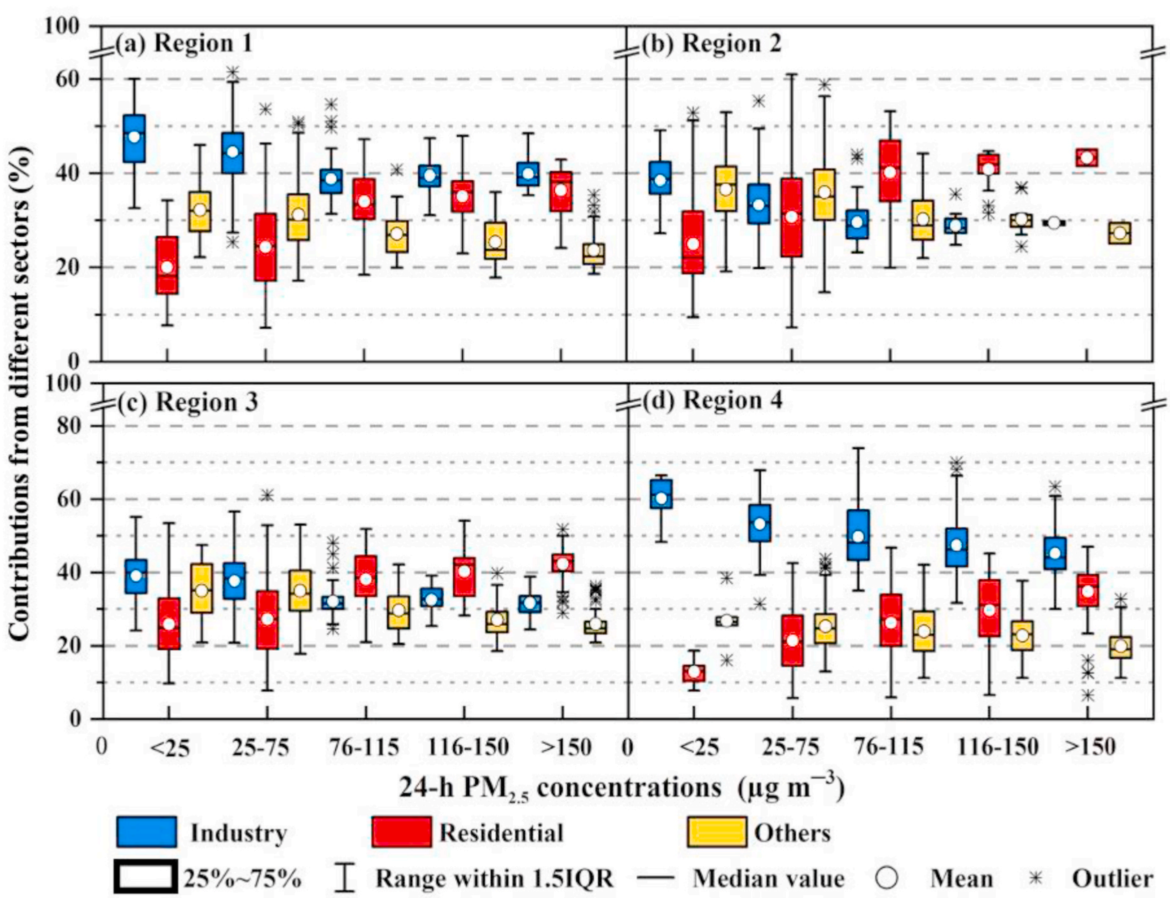

Fig. 4. Contributions from industries, residential activities, and others to $PM_{2.5}$ under different 24-h $PM_{2.5}$ concentration ranges in the urban centers of R1-4 in 2015. Others include ICs, BCs, windblown dust, sea salt, biogenic emission, open burning, energy, agriculture, transportation, and SOA. The classifications of R1-4 are shown in Fig. 1 and are explained in section 2.2. The number of days under each 24-h $PM_{2.5}$ concentration ranges in each city and region are summarized in Table S8.

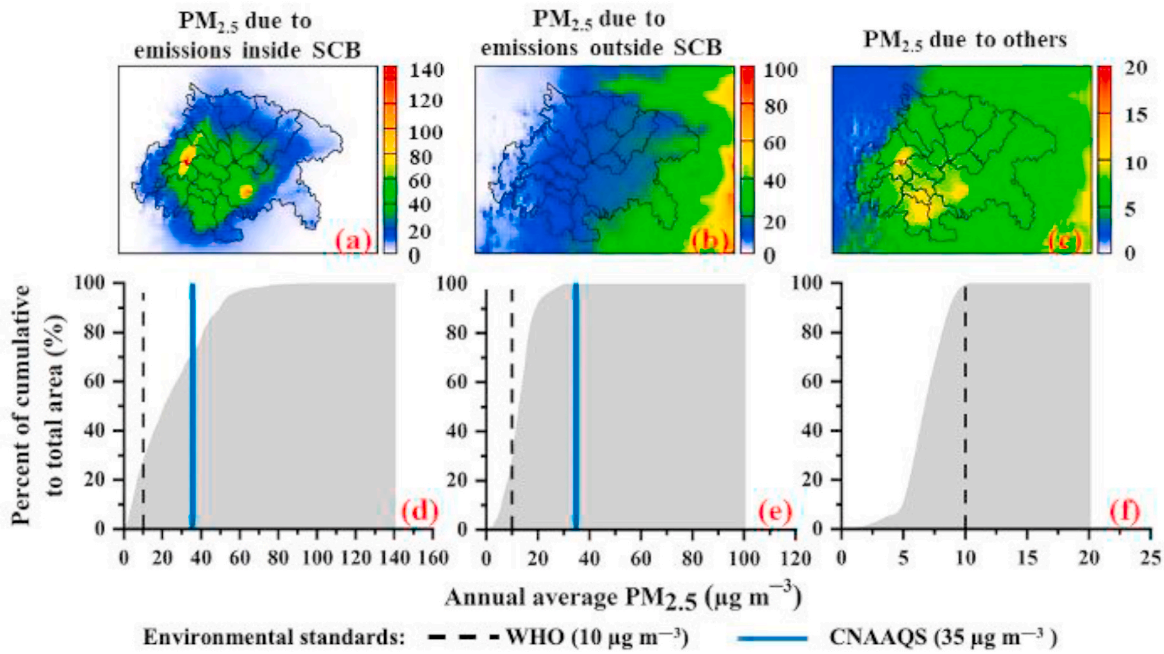

Fig. 5. Spatial distributions of simulated annual $PM_{2.5}$ concentrations (μg m^{-3}) due to (a) SCB emissions, (b) non-SCB emissions, and (c) others and the percentage of cumulative to total area of $PM_{2.5}$ concentrations due to (d) SCB emissions, (e) non-SCB emissions, and (f) others in 2015. The dash lines show the WHO guideline and the CNAAQS standard. Others include ICs, BCs, windblown dust, sea salt, and SOA.

decrease from the east to the west, and this spatial pattern is the same as that reported by Hu et al. (2015) and Ying et al. (2014), which are based on the MEIC inventory. Figure S11 shows that $PM_{2.5}$ concentrations in the urban centers in each season are more affected by SCB emissions (60%–80%) than by non-SCB emissions (10%–20%), except that the contributions from non-SCB emissions (20%–40%) are as important as that from SCB emissions in some R2 urban centers in spring, fall, and winter. For example, SCB and non-SCB emissions each account for ~40% in the three seasons. In the summer, SOA accounts for 15%–40% in the 18 urban centers, and the lower $PM_{2.5}$ concentrations, the higher fractional contributions from SOA. However, SCB and non-SCB emissions contribute 20-86 and 6-17 μg m^{-3} to the annual average $PM_{2.5}$ concentrations in the 18 SCB urban centers, respectively, and the contributions from non-SCB emissions are close to or higher than the WHO guideline of 10 μg m^{-3} (Table 1). Thus, it is important control both SCB and non-SCB emissions for the SCB cities to achieve the WHO guideline (10 μg m^{-3}).

Table 1
Regional contributions to annual average concentrations of $PM_{2.5}$ in the 18 SCB urban centers in 2015.

Region ID	Cities	$PM_{2.5}$ (μg m^{-3})	Regional contributions (μg m^{-3})			
			SCB emissions	Non-SCB emissions	SOA	Others[a]
R1	Chengdu	89	72	8	5	4
	Deyang	84	67	9	5	3
	Leshan	73	56	7	6	3
	Meishan	90	72	8	6	3
	Mianyang	66	49	10	5	3
	Ya'an	44	31	6	5	2
R2	Bazhong	42	20	15	5	2
	Dazhou	55	30	17	5	3
	Guang'an	62	39	15	5	3
	Guangyuan	40	22	12	4	2
	Nanchong	69	47	14	6	3
R3	Luzhou	84	61	13	7	3
	Neijiang	79	58	12	6	3
	Suining	54	33	14	5	2
	Yibin	80	60	10	7	3
	Zigong	84	63	11	7	3
	Ziyang	74	54	11	6	3
R4	Chongqing	112	86	15	6	4

[a] Include ICs, BCs, windblown dust, and sea salt.

3.4.2. *Contributions under different 24-h $PM_{2.5}$ concentrations*

The regional contributions under different 24-h $PM_{2.5}$ concentrations in the urban centers of R1-4 are shown in Fig. 6, and the data of each season are shown in Fig. S12. As 24-h $PM_{2.5}$ concentrations increase, the concentrations due to SCB emissions grow faster than that due to non-SCB emissions in general (Fig. 6). As 24-h $PM_{2.5}$ concentrations increase from <25 to >150 μg m^{-3} in the R1-R4 urban centers, the mean contributions from SCB emissions increase from 14 to 145 μg m^{-3} in R1, from 12 to 98 μg m^{-3} in R2, from 13 to 138 μg m^{-3} in R3, and from 16 to 178 μg m^{-3} in R4, while those from non-SCB emissions increase from 2 to 17 μg m^{-3} in R1, from 1 to 40 μg m^{-3} in R2, and from 2 to 26 μg m^{-3} in R3 and R4. Among the urban centers, the R2 urban centers are more affected by non-SCB emissions, as R2 has relatively lower anthropogenic emission densities across the SCB and is located in the north, where winds intrude the SCB (Qiao et al., 2019b). The concentrations due to SCB emissions are always higher than that due to non-SCB emissions on each day in each season, except for few episodes (Fig. 6 and Fig. S12). However, the largest contributions from non-SCB emissions in R1-4 are 51, 62, 70, and 87 μg m^{-3}, respectively, and these values are much higher than the WHO guideline of 35 μg m^{-3}. To prevent the pollution and extreme pollution days and reduce 24-h $PM_{2.5}$ concentrations less than the WHO guideline in the urban centers, the above data suggest that great efforts should be made to simultaneously reduce SCB and non-SCB emissions.

4. Conclusion

Annual average $PM_{2.5}$ concentrations exceed the CNAAQS standard and the WHO guideline in ~60% and ~100% of the SCB areas in 2015. To develop relevant pollution mitigation strategies, it is crucial to understand the contributions from different sectors to $PM_{2.5}$. In this study, we used a source-oriented CMAQ model to quantify not just the source

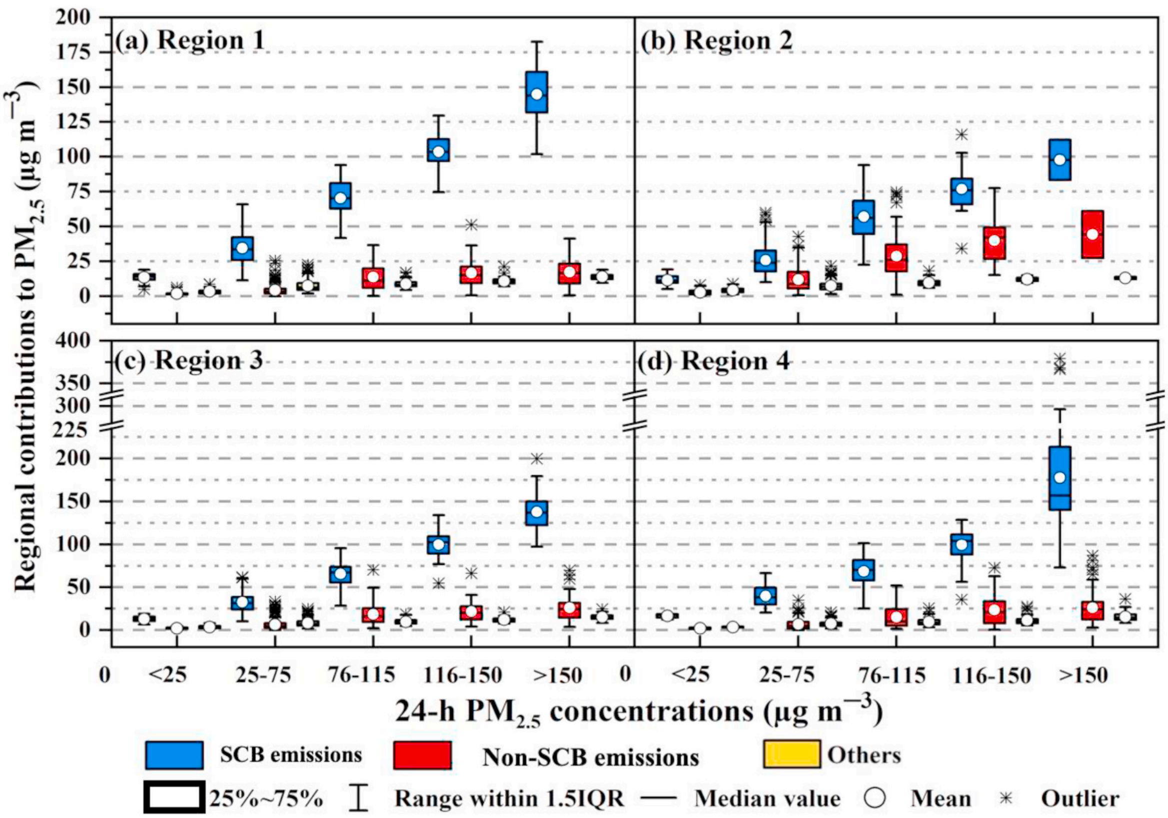

Fig. 6. Contributions from SCB emissions, non-SCB emissions, and others to $PM_{2.5}$ (μg m^{-3}) under different 24-h $PM_{2.5}$ concentration ranges in the urban centers of R1-4 in 2015. Others include ICs, BCs, windblown dust, sea salt, and SOA. The number of days under each 24-h $PM_{2.5}$ concentration ranges in each city and region are summarized in Table S8.

contributions to annual and seasonal average $PM_{2.5}$ concentrations but also the source contributions under different 24-h $PM_{2.5}$ concentrations for all the 18 SCB cities. The results suggest that industrial and residential activities are the largest sectors for annual $PM_{2.5}$ concentrations in the cities, and each of the two sectors has contributions of 25%–50% in the urban centers. On the pollution days and the extreme pollution days (24-h $PM_{2.5}$ concentrations >75 and 150 μg m^{-3}, respectively), residential and industrial emissions still have combined contributions (>~65%) much higher than that from other sources. In general, $PM_{2.5}$ concentrations in the SCB are more affected by SCB emissions than by non-SCB emissions, as annual $PM_{2.5}$ concentrations in the 18 SCB urban centers have 20–86 and 6–17 μg m^{-3} from SCB and non-SCB emissions, respectively. The impacts from non-SCB emissions on $PM_{2.5}$ concentrations decrease from the east to the west of the SCB. Annual average $PM_{2.5}$ concentrations due to non-SCB emissions do not exceed the CNAAQS standard in almost the entire SCB but are higher than the WHO guideline in ~70% of the SCB area. Furthermore, non-SCB emissions can contribute up to 87 μg m^{-3} for 24-h $PM_{2.5}$ in an urban center. In conclusion, the CNAAQS standard for annual $PM_{2.5}$ might be achieved in most of the SCB cities through mainly controlling SCB emissions (particularly those from industrial and residential activities); however, in order to achieve the WHO guideline for annual $PM_{2.5}$ and reduce $PM_{2.5}$ pollution and extreme pollution days, SCB and non-SCB emissions should be simultaneously controlled. As there are many cities in basins and valleys which are suffering from air pollution across the world, the methodology of this study would be helpful for these cities to quantify air pollutants' sources and help make relevant air pollution mitigation strategies.

CRediT authorship contribution statement

Xue Qiao: Research design, model simulations, and manuscript preparation. **Yanping Yuan:** Data curation, Formal analysis, Data analyses and visualization. **Ya Tang:** Data curation, Formal analysis, Research design and data analyses. **Qi Ying:** Model development and configuration. **Hao Guo:** Model simulations. **Yueying Zhang:** Data curation, Formal analysis, Visualization, Data analyses and visualization. **Hongliang Zhang:** Research design and model development.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2020.117896.

References

Bei, N., Zhao, L., Xiao, B., Meng, N., Feng, T., 2017. Impacts of local circulations on the wintertime air pollution in the Guanzhong Basin, China. Sci. Total Environ. 592, 373–390.

Boylan, J.W., Russell, A.G., 2006. PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. Atmos. Environ. 40, 4946–4959.

Chen, J., Zeng, J., Shi, C., Liu, R., Lu, R., Mao, S., Zhang, L., 2019. Associations between short-term exposure to gaseous pollutants and pulmonary heart disease-related mortality among elderly people in Chengdu, China. Environ. Health 18, 64.

Chen, Y., Xie, S., Luo, B., 2016. Composition and pollution characteristics of fine particles in Chengdu from 2012 to 2013. Acta Sci. Circumstantiae 36, 1021–1031.

Chen, Y., Xie, S., Luo, B., Zhai, C., 2017. Particulate pollution in urban Chongqing of southwest China: historical trends of variation, chemical characteristics and source apportionment. Sci. Total Environ. 584–585, 523–534.

Chen, Y., Xie, S.D., 2014. Characteristics and formation mechanism of a heavy air pollution episode caused by biomass burning in Chengdu, Southwest China. Sci. Total Environ. 473–474, 507–517.

Chu, P.C., Chen, Y.C., Lu, S.H., Li, Z.C., Lu, Y.Q., 2008. Particulate air pollution in Lanzhou, China. Environ. Int. 34, 698–713.

Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., van Aardenne, J.A., Monni, S., Doering, U., Olivier, J.G.J., Pagliari, V., Janssens-Maenhout, G., 2018. Gridded emissions of air pollutants for the period 1970-2012 within EDGAR v4.3.2. Earth Syst. Sci. Data 10, 1987–2013.

Emery, C., Liu, Z., Russell, A.G., Odman, M.T., Yarwood, G., Kumar, N., 2016. Recommendations on statistics and benchmarks to assess photochemical model performance. J. Air Waste Manag. Assoc. 67, 582–598.

Emery, C., Tai, E., Yarwood, G., 2001. Enhanced meteorological modeling and performance evaluation for two Texas episodes. In: Report to the Texas Natural Resources Conservation Commission. prepared by ENVIRON, International Corp., Novato, CA available at: http://www.tceq.state.tx.us/assets/public/implementation/air/am/contracts/reports/mm/EnhancedMetModelingAndPerformanceEvaluation.pdf.

Guenther, A.B., Jiang, X., Heald, C.L., Sakulyanontvittaya, T., Duhl, T., Emmons, L.K., Wang, X., 2012. The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions. Geosci. Model Dev. (GMD) 5, 1471–1492.

Hu, J., Wu, L., Zheng, B., Zhang, Q., He, K., Chang, Q., Li, X., Yang, F., Ying, Q., Zhang, H., 2015. Source contributions and regional transport of primary particulate matter in China. Environ. Pollut. 207, 31–42.

Hu, J.L., Chen, J.J., Ying, Q., Zhang, H.L., 2016. One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system. Atmos. Chem. Phys. 16, 10333–10350.

Hu, J., Li, X., Huang, L., Qi, Y., Zhang, Q., Zhao, B., Wang, S., Zhang, H.J., 2017. Ensemble prediction of air quality using the WRF/CMAQ model system for health effect studies in China. Atmos. Chem. Phys. 17, 13103–13118.

Kanakidou, M., Mihalopoulos, N., Kindap, T., Im, U., Vrekoussis, M., Gerasopoulos, E., Dermitzaki, E., Unal, A., Koçak, M., Markakis, K., 2011. Megacities as hot spots of air pollution in the East Mediterranean. Atmos. Environ. 45, 1223–1235.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., Akimoto, H.J., 2013. Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2. Atmos. Chem. Phys. 13, 11019–11058.

Langford, A., Senff, C., Alvarez, R., Banta, R., Hardesty, R., 2010. Long-range transport of ozone from the Los Angeles Basin: a case study. Geophys. Res. Lett. 37, L06807.

Li, M., Zhang, Q., Kurokawa, J., Woo, J.H., He, K.B., Lu, Z.F., Ohara, T., Song, Y., Streets, D.G., Carmichael, G.R., Cheng, Y.F., Hong, C.P., Huo, H., Jiang, X.J., Kang, S. C., Liu, F., Su, H., Zheng, B., 2017. MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP. Atmos. Chem. Phys. 17, 935–963.

Liao, T., Wang, S., Ai, J., Gui, K., Duan, B., Zhao, Q., Zhang, X., Jiang, W., Sun, Y., 2017. Heavy pollution episodes, transport pathways and potential sources of PM2.5 during the winter of 2013 in Chengdu (China). Sci. Total Environ. 584–585, 1056–1065.

MEP (Ministry of Environmental Protection of China), 2012a. Ambient air quality standards. Ministry of Environmental Protection of the People's Republic of China. Accessed on. http://kjs.mee.gov.cn/hjbhbz/bzwb/dqhjbh/dqhjzlbz/201203/W020120410330232398521.pdf. (Accessed 10 January 2019).

MEP, 2012b. Technical regulation for ambient air quality assessment. Accessed on. http://kjs.mee.gov.cn/hjbhbz/bzwb/jcffbz/201309/W020131105548549111863.pdf. (Accessed 10 January 2019).

NBSC (National Bureau of Statistics of China), 2018. China Statistical Yearbook. http://www.stats.gov.cn/tjsj/ndsj/2018/indexeh.htm.

Ning, G., Wang, S., Ma, M., Ni, C., Shang, Z., Wang, J., Li, J., 2018a. Characteristics of air pollution in different zones of Sichuan Basin, China. Sci. Total Environ. 612, 975–984.

Ning, G., Wang, S., Yim, S.H.L., Li, J., Hu, Y., Shang, Z., Wang, J., Wang, J., 2018b. Impact of low-pressure systems on winter heavy air pollution in the northwest Sichuan Basin, China. Atmos. Chem. Phys. 18, 13601–13615.

Panday, A.K., Prinn, R.G., 2009. Diurnal cycle of air pollution in the Kathmandu Valley, Nepal: observations. J. Geophys. Res. Atmos. 114, D09305.

Qi, Y., Li, W., Zhang, H., 2014. Local and inter-regional contributions to $PM_{2.5}$ nitrate and sulfate in China. Atmos. Environ. 94, 582–592.

Qiao, X., Ying, Q., Li, X.H., Zhang, H.L., Hu, J.L., Tang, Y., Chen, X., 2018. Source apportionment of $PM_{2.5}$ for 25 Chinese provincial capitals and municipalities using a source-oriented Community Multiscale Air Quality model. Sci. Total Environ. 612, 462–471.

Qiao, X., Guo, H., Wang, P.F., Tang, Y., Ying, Q., Zhao, X., Deng, W.Y., Zhang, H.L., 2019a. Fine particulate matter and ozone pollution in the 18 cities of Sichuan Basin, southwestern China: model performance and characteristics. Aerosol. Air Qual. Res. 19, 2308–2319.

Qiao, X., Guo, H., Tang, Y., Wang, P., Deng, W., Zhao, X., Hu, J., Ying, Q., Zhang, H., 2019b. Local and regional contributions to fine particulate matter in the 18 cities of Sichuan Basin, southwestern China. Atmos. Chem. Phys. 19, 5791–5803.

Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Duba, M.G., Huang, X. Y., Wang, W., Powers, J.G., 2008. A Description of the Advanced Research WRF Version 3 (No. NCAR/TN-475+STR). University Corporation for Atmospheric Research.

Shi, Z.H., Li, J.Y., Huang, L., Wang, P., Wu, L., Ying, Q., Zhang, H.L., Lu, L., Liu, X.J., Liao, H., Hu, J.L., 2017. Source apportionment of fine particulate matter in China in 2013 using a source-oriented chemical transport model. Sci. Total Environ. 601, 1476–1487.

Tian, Y.Z., Wu, J.H., Shi, G.L., Wu, J.Y., Zhang, Y.F., Zhou, L.D., Zhang, P., Feng, Y.C., 2013. Long-term variation of the levels, compositions and sources of size-resolved particulate matter in a megacity in China. Sci. Total Environ. 463–464, 462–468.

Wang, L.T., Jang, C., Zhang, Y., Wang, K., Zhang, Q.A., Streets, D., Fu, J., Lei, Y., Schreifels, J., He, K.B., Hao, J.M., Lam, Y.F., Lin, J., Meskhidze, N., Voorhees, S., Evarts, D., Phillips, S., 2010. Assessment of air quality benefits from national air pollution control policies in China. Part I: background, emission scenarios and evaluation of meteorological predictions. Atmos. Environ. 44, 3442–3448.

Wang, L.T., Wei, Z., Yang, J., Zhang, Y., Zhang, F.F., Su, J., Meng, C.C., Zhang, Q., 2014. The 2013 severe haze over southern Hebei, China: model evaluation, source apportionment, and policy implications. Atmos. Chem. Phys. 14, 28395–28451.

Whiteman, C.D., Hoch, S.W., Horel, J.D., Charland, A., 2014. Relationship between particulate air pollution and meteorological variables in Utah's Salt Lake Valley. Atmos. Environ. 94, 742–753.

Wiedinmyer, C., Akagi, S.K., Yokelson, R.J., Emmons, L.K., Al-Saadi, J.A., Orlando, J.J., Soja, A.J., 2011. The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. Geosci. Model Dev. (GMD) 4, 625–641.

Yang, F., Tan, J., Zhao, Q., Du, Z., He, K., Ma, Y., Duan, F., Chen, G., 2011. Characteristics of $PM_{2.5}$ speciation in representative megacities and across China. Atmos. Chem. Phys. 11, 5207–5219.

Ying, Q., Wu, L., Zhang, H.L., 2014. Local and inter-regional contributions to $PM_{2.5}$ nitrate and sulfate in China. Atmos. Environ. 94, 582–592.

Zhang, H., Li, J., Qi, Y., Jian, Z.Y., Wu, D., Yuan, C., He, K., Jiang, J., 2012. Source apportionment of $PM_{2.5}$ nitrate and sulfate in China using a source-oriented chemical transport model. Atmos. Environ. 62, 228–242.

Zhang, Z.S., Tao, J., Xie, S.D., Zhou, L.D., Song, D.L., Zhang, P., Cao, J.J., Luo, L., 2013. Seasonal variations and source apportionment of $PM_{2.5}$ at urban area of Chengdu. Acta Sci. Circumstantiae 33, 2947–2952.

Zhao, B., Wang, S.X., Dong, X.Y., Wang, J.D., Duan, L., Fu, X., Hao, J.M., Fu, J., 2013. Environmental effects of the recent emission changes in China: implications for particulate matter pollution and soil acidification. Environ. Res. Lett. 8, 024031.

Zhao, Q., He, K., Rahn, K.A., Ma, Y., Jia, Y., Yang, F., Duan, F., Lei, Y., Cheng, G., Cheng, Y., Liu, H., Wang, S., 2010. Dust storms come to central and southwestern China, too: implications from a major dust event in Chongqing. Atmos. Chem. Phys. 10, 2615–2630.