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Fine particulate matter pollution in the Sichuan Basin of China from 2013 to 2020: Sources, emissions, and mortality burden

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ABSTRACT

Fine particulate matter ($PM_{2.5}$) pollution is a critical air quality concern which poses threats to public health. Despite strict air pollution control measures implemented in China since 2013, $PM_{2.5}$ exceedances and region-wide $PM_{2.5}$ episodes are still frequently observed in the Sichuan Basin (SCB) located in southwestern China. Here, we examine ambient $PM_{2.5}$ pollution within the SCB from 2013 to 2020, focusing on emission sources, trends, and health outcomes. By integrating ambient measurements, emission inventories, and the health impact model, our findings reveal a notable decrease in $PM_{2.5}$ levels across the basin, with the Chengdu Plain showing a significant reduction of 56 μg/m^3 in 2020 compared to 2013. Despite these improvements, it is still challenging for densely populated cities to attain the national air quality standards. We highlight a 46.8 % reduction in $PM_{2.5}$ emissions from 2013 to 2020, driven largely by decreased emissions from residential and industrial sources, which accounted for an average of 38.6 % and 50.3 % of total reduced emissions, respectively. In contrast, the decreases of NO_x emissions (26.0 %) were less pronounced compared to $PM_{2.5}$ due to modest reductions from industrial and transportation sectors. Health impact assessments at 1 km × 1 km using the GEMM model attributes 157,637 deaths to long-term $PM_{2.5}$ exposure in the SCB for 2017, with stroke and ischemic heart disease identified as leading causes. Further analysis indicates that significant variations in population density could greatly amplify the health impacts of long-term $PM_{2.5}$ exposure, highlighting the need to prioritize $PM_{2.5}$ reduction strategies specifically targeting megacities to maximize health benefits. These findings underscore the critical need for ongoing emission reduction efforts and the implementation of targeted pollution control measures to further improve air quality and reduce mortality burden in the SCB.

1. Introduction

Elevated fine particle matter ($PM_{2.5}$) is a prominent risk factor for illness and death (Kim et al., 2015; Russell & Brunekreef, 2009). Epidemiological studies have identified robust associations between long-term exposure to $PM_{2.5}$ and adverse health outcomes (Burnett et al., 2018; Li et al., 2018). In the Global Burden of Disease (GBD) assessment for 2019, long-term exposures to $PM_{2.5}$ was estimated to contribute to 4.1 million deaths globally, with respiratory and cardiovascular diseases, cancer, and diabetes being the main contributors (Murray et al., 2020; Sang et al., 2022), which has arisen as major public health crises (Anenberg et al., 2019).

Since the 2000 s, rapid industrialization and urbanization powered by fossil fuel consumption have resulted in a substantial increase in anthropogenic air pollutants emissions, subsequently leading to severe air quality issues across China (Du et al., 2024; Liu et al., 2022; Richter et al., 2005). To improve air quality and safeguard human health, the Chinese government has dedicated efforts to address high ambient $PM_{2.5}$ levels and mitigate haze pollution through the Air Pollution Prevention and Control Action Plan (APPCAP) starting in 2013 (Zheng et al., 2018). Among highly developed city clusters in China, the Sichuan Basin of China (SCB) is known for its persistent air quality challenges associated with high $PM_{2.5}$ levels in winter (Yang et al., 2020). Unlike the Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), and Pearl River Delta

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(PRD) regions, which are located in relatively flat plains with dense urban clusters and well-identified pollution transport pathways, the SCB is characterized by the deep basin topography that traps pollutants within its boundaries (Zhang et al., 2019). This geographical configuration amplifies the effects of pollutant emissions driven by rapid industrialization and urbanization (Shu et al., 2021).

The Qinling Mountains to the north, the Daba Mountains to the northeast, Yunnan-Guizhou Plateau to the south, and Longmen Mountains to the northwest often confine air pollutants during winter within the topographic depression of the SCB (Wu et al., 2021). Such shadow cast by the surrounding mountains resulted in unique meteorological conditions featured by low wind speed and high relative humidity (Zhao et al., 2018). In addition, frequent stagnant conditions in combination with profound inversion layers in wintertime triggered by the shallow and highly stable boundary layer frequently depicted in the SCB further exacerbate $PM_{2.5}$ pollution (Liu et al., 2021). Previous studies have identified that the deep basin topography, and unfavorable meteorological conditions, in conjunction with substantial primary air pollutants emitted from fossil-fuel driven infrastructures result in the SCB regularly experiencing episodes of $PM_{2.5}$ pollution (Zhang et al., 2019; Zhou et al., 2019).

Recently pioneer studies have revealed the spatial and temporal distribution of $PM_{2.5}$ and gaseous pollutants across the SCB (Li et al., 2021). For instance, Wu et al., (2022) adopted satellite NO_2 columns and emission inventory for inferring NO_x emission in the SCB from 2010 to 2020 and found NO_x emission reduced by 26.5 % in 2020 compared with 2012. Using ground-based ambient measurement data, Zhao et al., (2018) studied variations of $PM_{2.5}$, PM_{10}, SO_2, and NO_2 between 2015 and 2017, and reported that the Chengdu Plain and southern SCB are most susceptible to poor air quality. Chemical composition of ambient $PM_{2.5}$ have also been determined for several cities within the SCB (Chengdu, Zigong, Yibin, etc) (Chen et al., 2014; Fu et al., 2024; Kong et al., 2020; Wang et al., 2017). However, source identification of ambient PM was conducted over a few years, ranging from 2013 to 2017, making spatial changes of PM afterwards 2017 remain elusive. While several high PM episodes occurred in wintertime were analyzed in the literature which demonstrate the critical role of synoptic-scale meteorological conditions in contributing to short-term extremely high $PM_{2.5}$ levels, limited episodic studies are difficult to assess intra-urban spatial patterns of $PM_{2.5}$ across the SCB (Yang, et al., 2020).

Given the health risk posed by long-term exposure to ambient $PM_{2.5}$, continuous regulation efforts toward compliance of $PM_{2.5}$ standard could result in substantial co-benefits for air pollution improvements and associated health impacts (Zhang et al., 2021). Most of previous investigations on health effects from $PM_{2.5}$ improvements were performed at national scale with coarse model resolution (27 km × 27 km or coarser), resulting in insufficient understanding on regional scale as exposure maps at a fine scale are crucially needed (Geng et al., 2021; Zheng et al., 2021; Liu et al., 2022). Using the standard health damage function, Zhan et al., (2023) quantified the all-causes premature mortality induced by $PM_{2.5}$ exposure in Chengdu from 2015 to 2021 and reported that premature death due to $PM_{2.5}$ was as high as 9386 over the period. However, little is known about health risks from $PM_{2.5}$ throughout the SCB at the fine-scale. On the basis of these studies, it is essential to understand the sources and emissions of $PM_{2.5}$ from anthropogenic sectors and quantitively assess the mortality burden attributed to high levels of $PM_{2.5}$ exposure in the SCB.

In this study, we identify geospatial and temporal changes in $PM_{2.5}$ and gaseous pollutants concentrations throughout the SCB based on ambient measurements from 2013 to 2020. Further, a high-resolution emission inventory with detailed anthropogenic sectors is used for probing emission patterns of air pollutants over the study period. The mortality burden induced by long-term $PM_{2.5}$ exposure is determined using the updated Global Exposure Mortality Model (GEMM) with epidemiological data.

2. Methodology

2.1. Ground-level observation

Hourly observations of air pollutant levels in the SCB were obtained from the national monitoring network established by the China National Environmental Monitoring Center (CNEMC), which began sampling across the SCB in 2013 and evolved with newly established monitoring stations in the following years. The hourly air quality data spanned from January 1, 2013, to December 31, 2020, and annual average pollutant concentrations for cities were calculated by averaging measurements across all monitoring stations within each city. Data quality control was performed in accordance with the Technical Regulations on Ambient Air Quality Assessment (GB3095-2012). It should be noted that the reference conditions for gaseous pollutants were standard conditions (273 K, 1013 hPa) prior to August 31, 2018, and were subsequently changed to 298 K and 1013 hPa after September 1, 2018.

For meteorological parameters, hourly meteorological observations were obtained from the China Meteorological Data Service Center (CMDSC) that undergo rigorous data accuracy check (https://data.cma.cn/). The locations of SCB, and meteorological and air quality monitoring stations are presented in Fig. 1 and Wu et al., (2022).

2.2. Anthropogenic emission inventory

To reveal the historical changes of air pollutant emissions from anthropogenic activities, the Multi-resolution emission inventory for China (MEIC) developed by Tsinghua University for the period from 2013 to 2020 is adopted in this study (Wu et al., 2024). The MEIC provides timely and accurate estimates for air pollutant emissions and multiple strategies are employed by the MEIC to optimize the emission estimates, including utilizing a localized emission factor database and categorizing emission sources in approximately 800 sectors (Li et al., 2017). Specifically, there are 5 major anthropogenic source categories (including power, agriculture, transportation, industrial, and residential sources) in the MEIC inventory with a grid resolution of $0.25^\circ \times 0.25^\circ$ (Zheng et al., 2018). To further probe the sector-based anthropogenic emission changes, the detailed MEIC inventory with 22 sub-sectors under major source category are used in this work (listed in Table 1). Extensive studies based on inverse modeling and top-down constrained emission inventory comparison have shown that the MEIC inventory well captures the inter-annual changes of air pollutant emissions and exhibits strong capability in reflecting the sector-based emission variability over time (Zheng et al., 2021).

2.3. Global exposure mortality model (GEMM)

The Global Exposure Mortality Model (GEMM), which incorporates risk data from 41 cohort studies of outdoor air pollution on non-accidental mortality, has been extensively used in previous studies to estimate the disease burden associated with long-term $PM_{2.5}$ exposure (Geng et al., 2021; Kang et al., 2024; Pozzer et al., 2023; Wu et al., 2021). It is worth noting that GEMM significantly broadens the range of exposure concentrations by including cohorts from China (Burnett et al., 2018; Yin et al., 2017).

Here, we adopt annual averaged $PM_{2.5}$ concentrations in 2017 from the dataset developed by Hammer et al., (2020) with a grid resolution of $0.01^\circ \times 0.01^\circ$, to estimate $PM_{2.5}$-related disease burden using concentration–response functions (CRFs) from the updated GEMM documented in McDuffie et al., (2021). Specifically, the disease burden from 6 mortality endpoints (including Chronic Obstructive Pulmonary Disease (COPD), type II diabetes (DM), Lower Respiratory Infections (LRI), Lung Cancer (LC), Ischemic Heart Disease (IHD) and Stroke) linked to long-term $PM_{2.5}$ exposure were estimated. Cause-specific relative risk (RR) on concentration (C) in GEMM were calculated through Eq. (1)

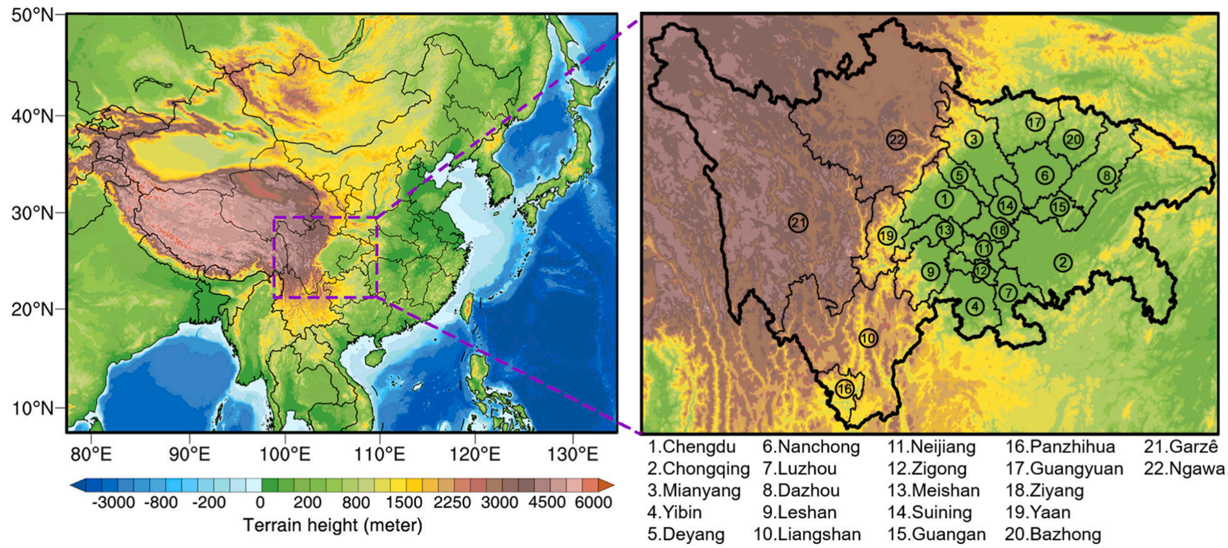

Fig. 1. Terrain map of the SCB in China and cities within the SCB.

Table 1
Major source category and detailed sub-sectors in MEIC emission inventory.

Major source category	Sub-sectors under major source category
Power	Power generation
Industrial	Heating
	Industrial boilers
	Cement
	Coking
	Steel
	Petrochemicals
	Oil and gas storage and transportation
	Industrial painting
	Printing and dyeing
	Architectural coatings
	Other industrial industries
Residential	Civilian coal burning
	Civilian biomass combustion
	Civilian chemical use
	Other civilian sources
Transportation	Gasoline vehicles
	Diesel vehicles
	Motorcycles
	Non-road mobile sources
Agriculture	Fertilizer application,
	Livestock breeding

$$RR\quad(C) = e^{\frac{\theta \times \ln\left(\frac{z}{\alpha}+1\right)}{1+e^{\left(-\frac{z-\mu}{\nu}\right)}}} \tag{1}$$

where θ, α, μ and ν determine the shape of the $PM_{2.5}$-mortality relationships. For each health endpoint, the attributable fraction (AF) of mortality due to long-term $PM_{2.5}$ exposure is further calculated using Eq. (2)

$$\mathrm{AF}\quad(\mathrm{C}) = 1 - \frac{1}{\mathrm{RR}\ \ (\mathrm{C})} \tag{2}$$

Lastly, the AF for each age group and disease ($AF_{age,\ disease}$) were subsequently multiplied by population and baseline mortality data specific to each age group for each disease. These results were then summed across all relevant age groups (m) and diseases (n) to determine the total disease burden associated with exposure to $PM_{2.5}$, as shown in Eq. (3).

$$PM_{2.5}\ \text{Mortality} = \sum_{disease}^{n}\sum_{age}^{m} P_j \times PS_s \times \mathrm{BMR}_{age,\ disease} \times \mathrm{AF}_{age,\ disease} \tag{3}$$

where P_j is the total population amount in grid j, PS_s denotes the fraction of a population subgroup s to the total population, BMR represents the national baseline mortality incidence rate of each disease for population subgroup s.

2.4. Satellite NO_2 columns

Satellite-derived NO_2 vertical column density (NO_2 VCD) is widely used to identify fine-scale NO_x hotspots and constrain NO_x emissions. In this study, TROPOspheric Monitoring Instrument (TROPOMI) NO_2 VCD is adopted to elucidate the spatial pattern of NO_x emissions across the SCB (Van Geffen et al., 2020). TROPOMI is an atmospheric monitoring spectrometer launched by the European Space Agency (ESA) for Sentinel-5P (Veefkind et al., 2012), with a spatial resolution of 7 km × 3.5 km (improved to 5.5 km × 3.5 km after August 6, 2019), which extends the capabilities of measuring NO_2 VCD at a space view due to its broad spatial coverage and high retrieval accuracy (Goldberg et al., 2019; Sun et al., 2021). TROPOMI typically passes over at 13:30 local time for achieving daily global coverage. To enhance data accuracy, pixels with cloud coverage greater than 30 %, solar zenith angles exceeding 70°, and quality assurance values (qa_value) less than 0.7 are excluded to reduce errors caused by cloud coverage and other factors.

3. Results

3.1. Annual $PM_{2.5}$ and gaseous pollutants trends in SCB from 2013 to 2020

Fig. 2 presents annual $PM_{2.5}$ and NO_2 concentrations over cities within the SCB from 2013 to 2020, as well as TROPOMI NO_2 columns in 2019. For NO_2 columns, Chengdu and its surrounding cities, Meishan, Deyang, and Ziyang, form a high pollution area in the west border of the SCB, known as the Chengdu Plain pollution belt. Another highly polluted area is mainly concentrated in the central urban area of Chongqing and its surrounding regions, indicating a distinct regional characteristic of NO_x pollution in the SCB. The distribution of NO_2 column shows high concentration areas located in the Chengdu Plain city cluster (up to 12×10^{15} molecules/cm^2), Chongqing (up to 12×10^{15} molecules/cm^2), the Southern Sichuan city cluster (6×10^{15}

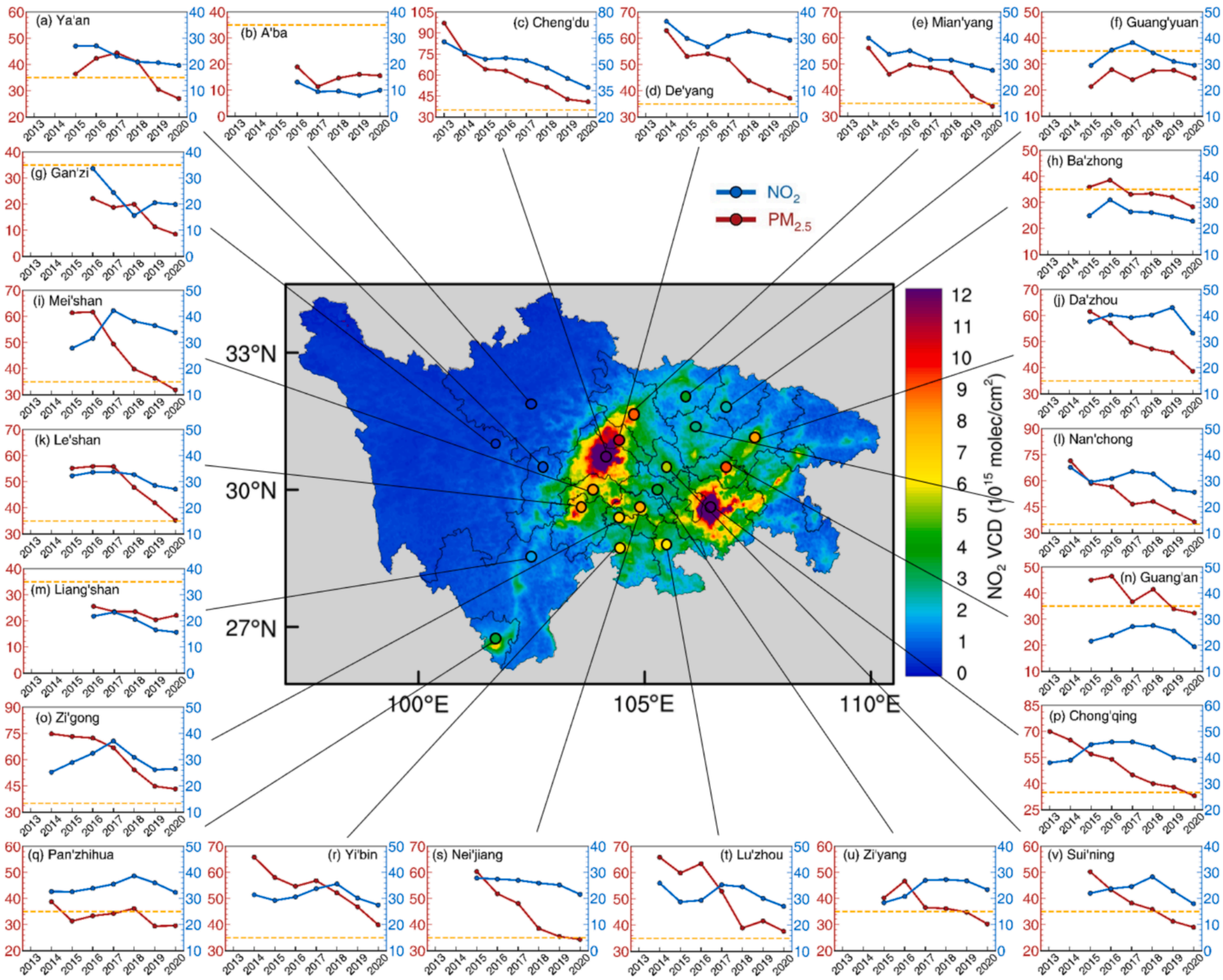

Fig. 2. (middle panel) Map of annual averaged tropospheric TROPOMI NO_2 vertical column density for 2019. (surrounding panels) Time series of NO_2, O_3, and O_x in ppb for each site for the 19 March to 30 June period from 2000 to 2020. annual mean $PM_{2.5}$ and NO_2 levels in μg/m^3 for cities within the SCB from 2013 to 2020.

molecules/cm^2), and the Northeastern Sichuan city cluster (4×10^{15} molecules/cm^2). Additionally, it is worth noting that densely populated cities (including Chengdu, Deyang, Leshan, Dazhou, and Yibin) which exhibit high NO_2 columns, consistently exceeded $PM_{2.5}$ air quality standard (annual average of 35 μg/m^3) from 2013 to 2020, which indicates a clear correlation between substantial NO_x emissions and elevated $PM_{2.5}$ concentrations.

In contrast to the consistent decline in $PM_{2.5}$ levels, the changes in ambient NO_2 levels across each city exhibit considerable temporal variability. In the Chengdu Plain, metropolitan areas such as Chengdu and Mianyang exhibited sustained NO_2 downward trends, recording substantial reductions of 41.6 % and 31.1 % throughout the study period, respectively. The southern Sichuan city cluster, however, displayed a distinctive pattern characterized by initial increases followed by subsequent decreases. For instance, annual average NO_2 concentrations peaked at 37.1 μg/m^3 in Zigong (2017) and 35.6 μg/m^3 in Yibin (2018) before the descent. While annual average NO_2 levels in several cities, including Chengdu, Mianyang, and Leshan, generally aligned with $PM_{2.5}$ concentration trends, the southern SCB city cluster (Yibin, Ziyang, and Zigong) experienced peak NO_2 levels in 2017 or 2018 – notably later than the implementation of APPCAP in 2013. This variable response of NO_2 levels across cities demonstrates the progressively evolving NO_x control measures over time. However, the interpretation of this phenomenon is complicated by the changing NO_x lifetime, inter-annual meteorological variability, and stringent pollution regulations, thereby calling for further investigation.

For inter-annual variations of $PM_{2.5}$ concentrations over time, it is seen that annual $PM_{2.5}$ concentrations in most SCB cities have steadily decreased since 2013 due to the implementation of the APPCAP, reaching the lowest levels in 2020. Specifically, $PM_{2.5}$ concentrations in Chengdu experienced the most significant reductions (as high as 57.9 %) in 2020 compared to 2013, and Chongqing and Nanchong saw decreases of 52.8 % and 49.1 %, respectively. In response to the outbreak of COVID-19, the Chinese government implemented first-level emergency responses in 2020, which resulted in reductions in fossil fuel use, industry production, and traffic volumes. These measures led to sharp reductions in emissions from transportation and industry sectors, resulting in significantly lower concentrations of $PM_{2.5}$ in most cities of the SCB in 2020 compared to 2019. Among them, large declines were observed in Dazhou and Guang'an in average annual NO_2 concentrations in 2020 compared to 2019, which dropped by 23.3 % and 23.9 %, respectively. Further, annual average $PM_{2.5}$ concentrations in Yibin, Leshan, and Dazhou experienced significant decreases in 2020, with a reduction of 17.5 %, 16.1 %, and 16.1 % compared to 2019, respectively. Unlike considerable reductions in other cities, a few SCB cities such as Chengdu, Panzhihua, and Zigong saw little changes for annual $PM_{2.5}$ levels in 2020 relative to 2019, and there was an increase in $PM_{2.5}$ levels in Liangshan. These abnormal patterns suggest that the economic rebound during the post COVID-19 period in 2020 may significantly offset the decreases of $PM_{2.5}$ in the lockdown period.

Fig. 3 illustrates the changes of $PM_{2.5}$ chemical composition across cities within the SCB based on sampling source apportionment. It is seen

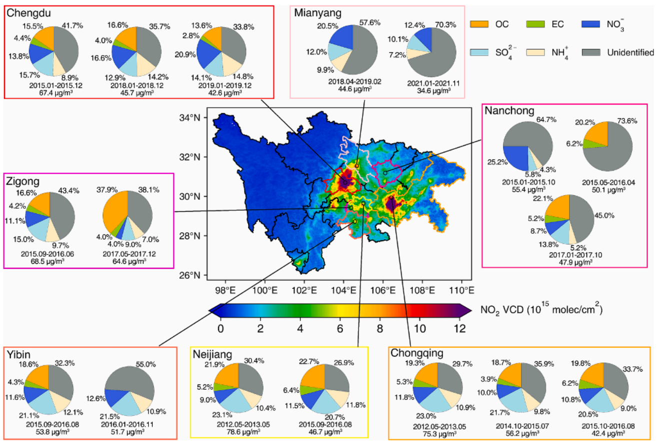

Fig. 3. (middle panel) Map of annual averaged tropospheric TROPOMI NO_2 vertical column density for 2019. (surrounding panels) Changes of $PM_{2.5}$ concentration (μg/m^3) and mass fractions of chemical components in major cities in the SCB over time.

that the vast majority of ambient $PM_{2.5}$ across the SCB is comprised of organic carbon (OC), elementary carbon (EC), and inorganic aerosols (including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), SNA). Specifically, SNA clearly dominates the components of $PM_{2.5}$ in densely populated areas, which account for ~40 % mass concentrations in both Chengdu and Chongqing. For instance, a major fraction of $PM_{2.5}$ in Chongqing, Neijiang, and Yibin consists of sulfate (>20 % in mass balance), compared to Mianyang where sulfate plays a relatively minor role and nitrate is much more significant in inorganic components. Moreover, the reduction of $PM_{2.5}$ levels driven by APPCAP changes regional aerosol composition which is clearly reflected in $PM_{2.5}$ constituents. Significant decreases in the proportion of SNA within $PM_{2.5}$ components in Mianyang are noted between 2018 and 2021 (particularly for nitrate aerosols). Conversely, nitrate aerosols have progressively evolved to become the dominant component of $PM_{2.5}$ in Chengdu, with their contribution surpassing that of OC in 2019. This finding implies that reducing nitrate aerosols plays a crucial role in decreasing $PM_{2.5}$ levels in Chengdu, which aligns with prior modeling studies based on CMAQ (Yang et al., 2024).

Regarding the compliance with China's National Ambient Air Quality Standard (NAAQS) of $PM_{2.5}$, annual $PM_{2.5}$ levels in most SCB cities were consistently in excess of $PM_{2.5}$ air quality standard (annual average of 35 μg/m^3) before 2020, indicating that $PM_{2.5}$ pollution remains as a critical environmental concern across the SCB, in spite of the stringent pollution control measures adopted afterwards 2013. More importantly, despite the substantial decreases of anthropogenic emissions induced by COVID-19 lockdown in 2020, annual $PM_{2.5}$ concentrations in Chengdu, Deyang, Dazhou, Luzhou, Yibin, and Zigong still did not attain the $PM_{2.5}$ air quality standard, demonstrating the urgent demand of much strict regulatory efforts for these cities. Moreover, it is worth mentioning that annual $PM_{2.5}$ levels in Chongqing in 2020 (33 μg/m^3) firstly met the $PM_{2.5}$ standard in 2020 during the period from 2013 to 2020, which implies that achieving the emission reduction goal aimed at attaining $PM_{2.5}$ standard can be difficult for Chongqing. In contrast, the West Sichuan Plateau city cluster as well as Guangyuan and Liangshan generally met NAAQS $PM_{2.5}$ standard with little changes of annual $PM_{2.5}$ concentrations over time. This pattern primarily stems from low anthropogenic emissions in these cities, characterized by less developed transportation infrastructure, lower population density, relatively modest industrial development, and consequently lower energy consumption compared to megacities (Chengdu and Chongqing). In spite of the remarkable reductions of annual $PM_{2.5}$ concentrations across the SCB between 2013 and 2020, all cities in the region consistently recorded annual $PM_{2.5}$ levels significantly exceeding 5 μg/m^3 throughout the study period. Notably, annual average $PM_{2.5}$ concentrations in major metropolitan areas including Chengdu and Chongqing remained five times higher than this threshold in 2020. This persistent phenomenon underscores the pressing challenge the SCB faces in meeting the World Health Organization's (WHO) updated air quality guidelines (AQGs) in September 2021, which recommended a limit on annual $PM_{2.5}$ exposure of 5 μg/m^3 (Pai et al., 2022).

3.2. Changes of anthropogenic emissions over the SCB

Fig. 4 illustrates the annual changes in primary emissions of $PM_{2.5}$, NO_x, and SO_2 across various sectors within the SCB from 2013 to 2020, using data from the MEIC inventory. Overall, anthropogenic emissions in the SCB have significantly decreased over the study period, with reductions estimated at 83.1 % for SO_2, 46.8 % for $PM_{2.5}$, and 26.0 % for NO_x.

The reduction of air pollutant emissions depends on the reduction magnitude of each sector (as seen in Fig. 5). Among these, SO_2 emissions saw the most substantial reduction. The total annual SO_2 emissions in the region were recorded at 2346.5 kt in 2013. In 2020, SO_2 emissions dropped dramatically to only 392.8 kt, marking an 83.1 % decrease from the emissions in 2013. This sharp decline is primarily due to significant reductions in emissions from power and industry sources. Specifically, emissions from the power sector decreased by 347.9 kt (87.2 %), while industrial emissions fell by 1426.0 kt (81.9 %). This is because stationary combustion sources of fossil fuels are the largest sources of SO_2 emissions, mainly due to coal combustion in power plants and industrial boilers (Zhou et al., 2018). Following the issuance of the APPCAP by the State Council in 2013, all provinces have intensified the control of SO_2 emissions, including desulfurization and the implementation of ultra-low emission standard for power plants, resulting in a notable decline in SO_2 emissions. It is important to note that SO_2 emissions from transportation sector were on an annual decrease with a considerable reduction of 38.7 % between 2013 and 2020. This reduction can be attributed to the implementation of the '13th Five-Year Plan', which focused on phasing out older vehicles and significantly enhancing the quality of automotive gasoline and diesel fuel. A key environmental benchmark in this improvement has been the reduction of sulfur content to just 10 ppm in both fuels. Specifically, SO_2 emission reductions were accomplished by complying with the ultralow emission standards. To meet these rigorous criteria, coal-fired power plants have widely adopted flue gas desulfurization (FGD) and selective catalytic reduction (SCR) systems at their facilities. The installation of these systems had a penetration rate of over 95 % by 2017 (Zheng et al., 2018).

Similarly, the total annual emissions of $PM_{2.5}$ decreased significantly, with total $PM_{2.5}$ emissions of 708.6kt in 2013 and 376.9kt in 2020, a decrease of 46.8 %. Among these sources, residential and industrial sectors are the primary contributors to $PM_{2.5}$ emissions. On average, residential sources accounted for 50.1 % of these emissions from 2013 to 2020, while industrial sources contributed 43.1 %. Compared to $PM_{2.5}$ emissions from residential and industrial sectors in 2013, the reductions in $PM_{2.5}$ emissions from residential and industrial sector in 2020 were as high as 38.6 % and 50.3 %. The significant reductions in $PM_{2.5}$ emissions of residential and industrial sectors were major factors in the substantial decrease in total annual $PM_{2.5}$ emissions observed in 2020.

From 2013 to 2020, the decline in NO_x emissions was steady but notably less pronounced than the reductions in $PM_{2.5}$ and SO_2 emissions, highlighting a considerable challenge in regulating NO_x emissions. During the study period, NO_x emissions in SCB decreased by 26.0 % while consistently staying above 1000 kt per year. As shown in Fig. 4, transportation and industrial sectors are identified as the primary contributors to NO_x emissions, accounting for 36.9 % and 47.2 % in 2020. While there are ongoing annual reductions in these sectors, the magnitude of reductions remains minor, highlighting significant potential for implementing more rigorous reduction strategies. On the contrary, a

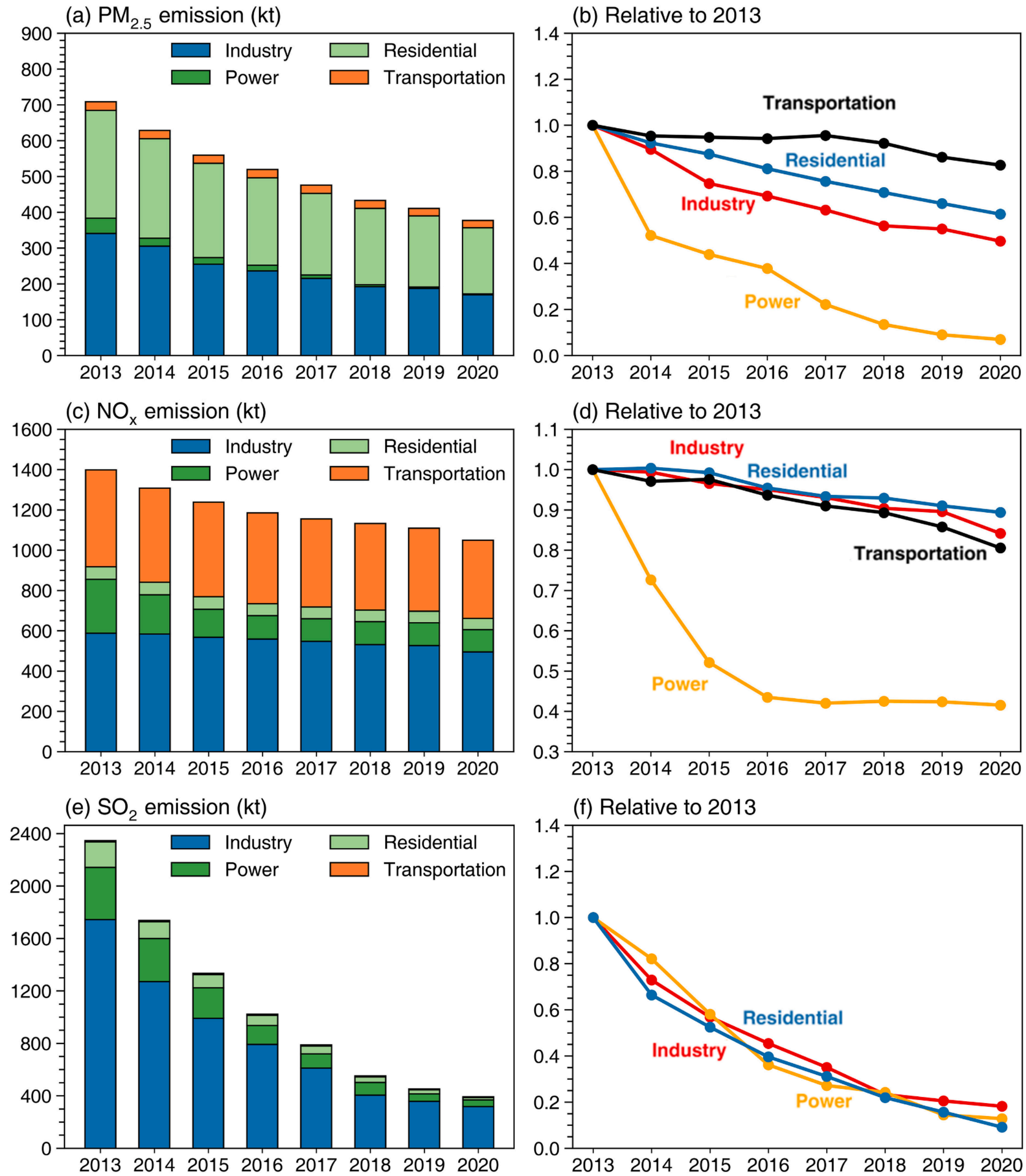

Fig. 4. Changes in anthropogenic emissions of primary $PM_{2.5}$, NO_x, and SO_2 (emission normalized to 2013 in (b)(d))(f)) in the SCB from 2013 to 2020.

substantial reduction in the power sector is depicted, where emissions declined by 60 % between 2013 and 2017. This was facilitated by stringent new emission standards and the widespread adoption of ultra-low emission technologies, which have set new benchmarks in emission controls within this sector. However, NO_x emissions from power sector experienced a rebound after 2017, primarily due to increased energy demands, while increased the power sector's emission load due to higher electricity consumption. NO_x emissions reductions from transportation sources were limited to just 19.4 % relative to 2013 levels by 2020. Meanwhile, data from the Ministry of Public Security shows that the ownership of vehicles in Chengdu over 5.98 million vehicles by the end of 2020, ranking it second in vehicle ownership across China. This highlights the transportation sector in Chengdu as a significant source of NO_x emissions for the SCB. In addition, it is worth noting that the total reduction of NO_x emissions (26.0 %) was less pronounced compared to primary $PM_{2.5}$ (46.8 %), which can be attributed to the changes induced by emission mitigation and additional emissions caused by growing activities among sub-sectors. Specifically, industrial sector is one of the largest contributors to both primary $PM_{2.5}$ and NO_x emissions. From 2013 to 2020, industrial primary $PM_{2.5}$ emissions decreased by 50.3 %, while industrial NO_x emissions only decreased by 16.0 %, as $PM_{2.5}$ is more easily removed by end-of-pipe control measures while industrial combustion sources lack effective NO_x controls (Zheng et al., 2018). Additionally, with upgraded emission standards and the deployment of ultra-low emission technology, power sector acted as the primary driver of NO_x emission reductions in the SCB from 2013 to 2020, while transportation emission, the dominant NO_x contributor, only decreased by 19.4 % over time. Sector-based decomposition analysis shows that emission changes in the industrial and transportation sectors, which dominate NO_x emissions, largely explain the modest NO_x reduction. Nevertheless, it should be acknowledged that strict transportation measures have reversed the increasing NO_x emission trends before 2013, the growing NO_x emissions due to vehicle growth between 2013 and 2020 in the SCB still make NO_x emission reduction challenging. This phenomenon pinpoints the critical need for comprehensive strategies that include enhancing NO_x reduction technologies in industrial applications and stricter vehicle emission standards.

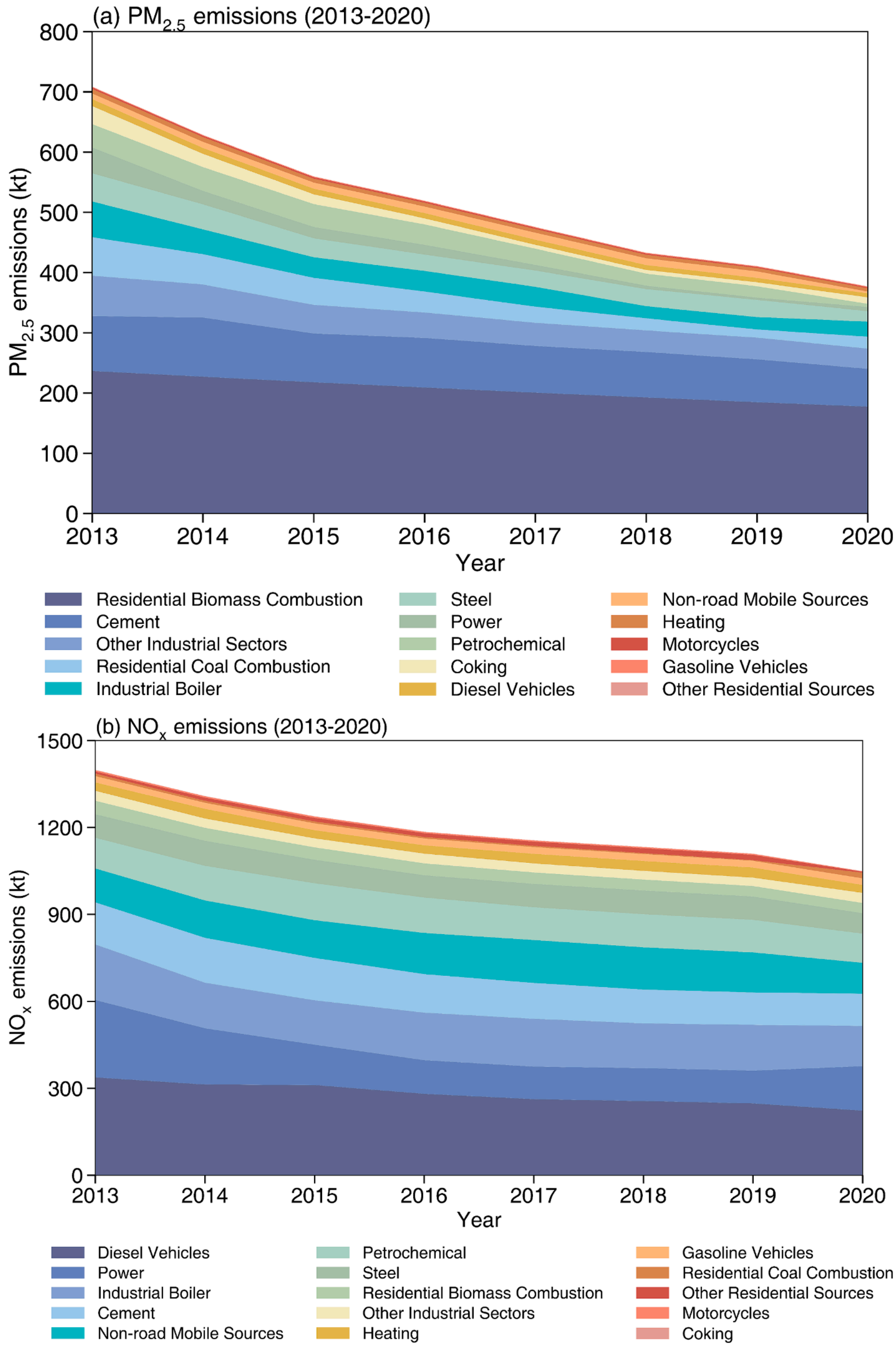

Fig. 5. Sector-based anthropogenic primary $PM_{2.5}$ (a) and NO_x (b) emissions in the SCB from 2013 to 2020.

3.3. *Spatial changes of $PM_{2.5}$ and NO_x emissions in 2013–2020*

The spatial distribution of annual total primary $PM_{2.5}$ and NO_x emissions across the SCB from 2013 to 2020 as well as the changes over time are presented in Figs. 6 and 7. Evidently, hotspots of $PM_{2.5}$ and NO_x emissions are mainly found in the Chengdu Plain, urban Chongqing, and southern city cluster of SCB, with annual emissions ranging from 4 to 8kt. This spatial pattern clearly reflects the intensity of anthropogenic activities associated with on-road vehicles emissions and industrial processes. While the spatial distribution pattern of $PM_{2.5}$ and NO_x emissions in 2017 was broadly similar to that of 2013, notable basin-wide reductions for both $PM_{2.5}$ and NO_x in emission intensity are depicted due to the regulation efforts by APPCAP between 2013 and 2017. In addition, it is worth noting that regions with high emissions experienced the most significant reductions in emission levels (particularly for Chengdu and Chongqing), indicating the success of emission control strategies adopted since 2013. In 2019, it can be found that annual $PM_{2.5}$ emissions decreased compared with 2017, which results in primary $PM_{2.5}$ emissions over most areas of the SCB (more than 90 % emission grids) lower than 2kt. However, the decreasing trend of $PM_{2.5}$ and NO_x emissions were reversed in a few grids in 2019, featured by significant increases found in urban and rural areas (especially Leshan, Mianyang, and Chongqing). This abnormal pattern is primarily linked to enhanced NO_x emissions in power and industrial sectors in 2019 compared to 2017. As the result of the COVID-19 lockdown measures in 2020, both $PM_{2.5}$ and NO_x emissions were further reduced throughout

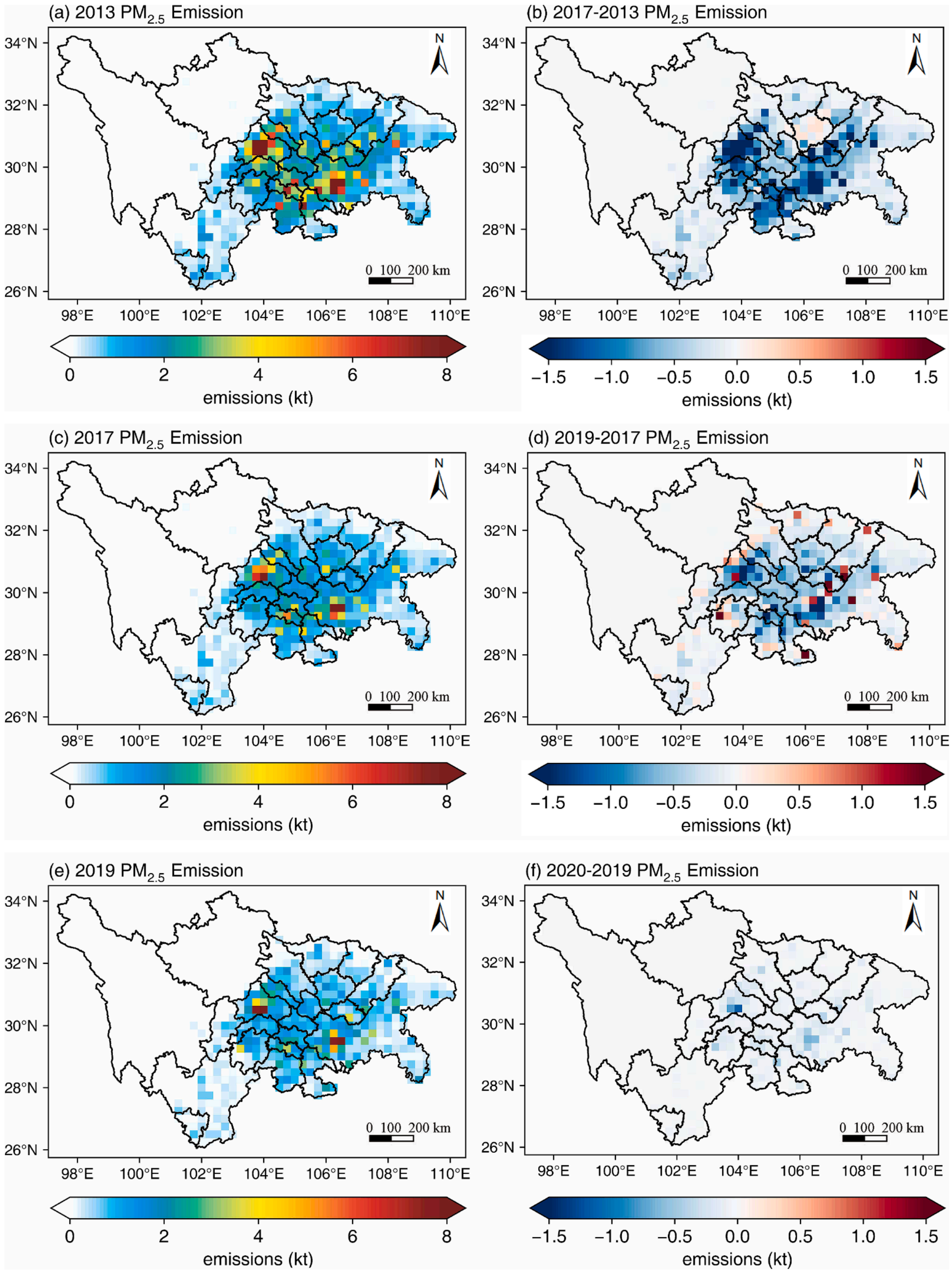

Fig. 6. Spatial map of annual $PM_{2.5}$ emissions in the SCB in 2013, 2017, and 2019 and emission changes between 2013 and 2017, 2017 and 2019, and 2019 and 2020.

the SCB. Interestingly, the reduction magnitude of $PM_{2.5}$ emissions between 2019 and 2020 was much less than prior years, implying that the potential capacity of emission reduction may be reached and further diminishing $PM_{2.5}$ emissions might be challenging for the SCB.

A comparison of emission changes across different years reveals further details. From 2013 to 2017, significant reductions in $PM_{2.5}$ and NO_x emissions were observed in multiple regions, particularly in the Chengdu Plain, highlighting the effectiveness of stringent pollution control measures and environmental regulations. However, slight increases in emissions were noted in some areas of the Southern Sichuan City Cluster, Northeastern Sichuan City Cluster, and eastern Chongqing. For the period between 2017 and 2019, significant reductions in $PM_{2.5}$ and NO_x emissions were seen in the Chengdu Plain and the central urban areas of Chongqing. However, certain regions outside these areas experienced increases in emissions. Notably, a comparison of the reductions in $PM_{2.5}$ and NO_x emissions show that $PM_{2.5}$ saw a more pronounced decline, while NO_x reductions were less significant. As illustrated above, high emission levels of $PM_{2.5}$ in 2019 were only concentrated in the central urban areas of Chengdu and Chongqing (as high as 8 kt annually), with other areas not exceeding 4 kt annually. On the other hand, NO_x high-emission areas remained widely distributed across the Chengdu Plain Urban Cluster, Southern Sichuan Urban Cluster, and southwestern Chongqing (reaching 8 kt/grid). As of 2020, annual total NO_x emissions have remained above 1000 kt, with only a

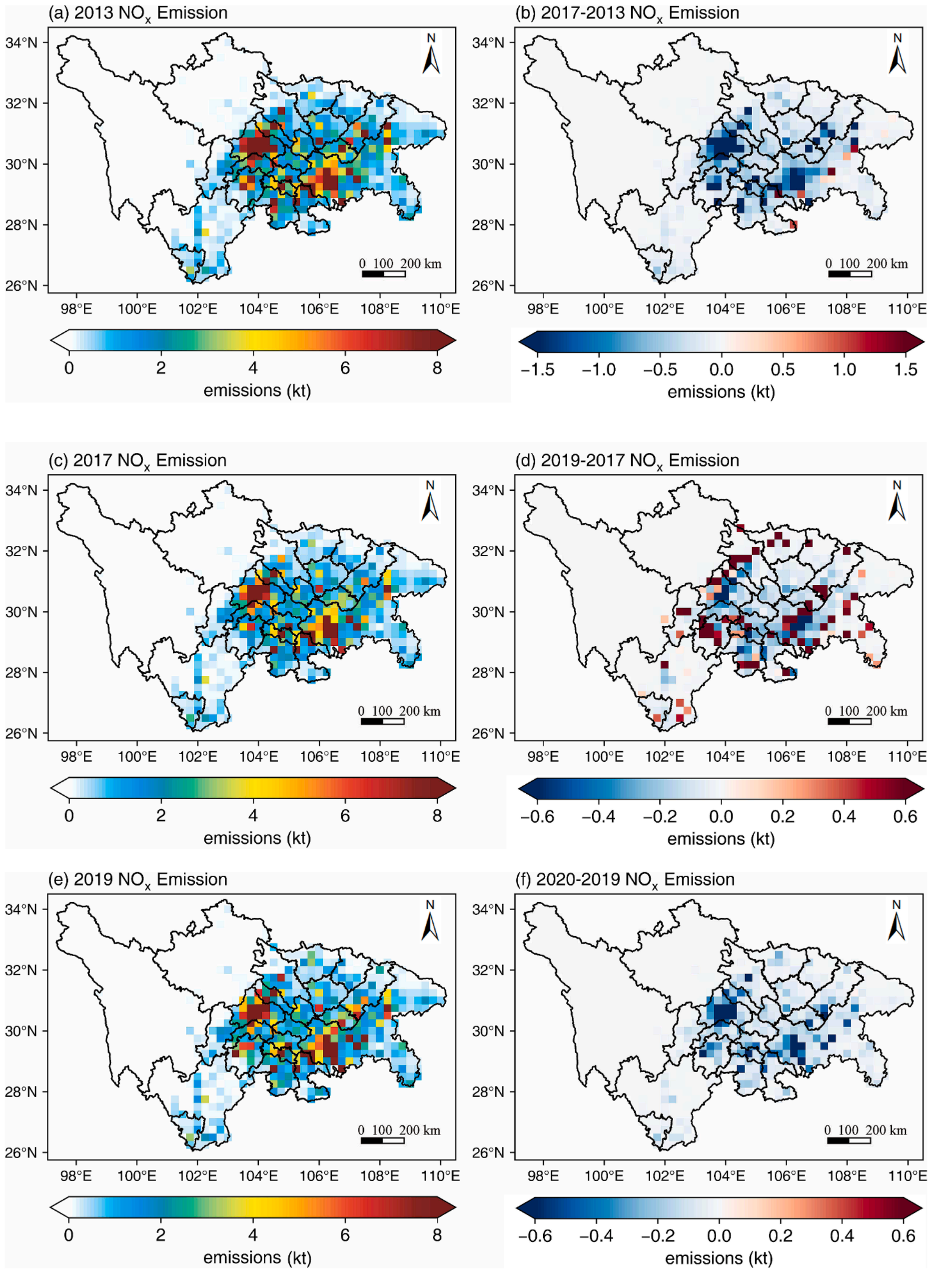

Fig. 7. Spatial map of annual NO_x emissions in the SCB in 2013, 2017, and 2019 and emission changes between 2013 and 2017, 2017 and 2019, and 2019 and 2020.

slight decrease, which reflects the region's large heavy industrial and infrastructures.

3.4. Mortality burden of $PM_{2.5}$ pollution

Fig. 8 presents the ambient $PM_{2.5}$ mortality burden and fractional disease contributions throughout the SCB, as well as the sector-based relative contributions to city-level $PM_{2.5}$ emissions. Basin-wide total $PM_{2.5}$-associated mortality burden was as high as 157,637 deaths in 2017, aligning well with previous estimates of 141,630 premature deaths estimated by Wu et al., (2021) and Geng et al., (2021). While these earlier studies examined nationwide $PM_{2.5}$-induced mortality using coarser spatial resolution data, their findings corroborate our basin-specific assessment. In terms of mortality burden, stroke and Ischemic Heart Disease (IHD) were the leading causes of deaths, resulting in 60,533 and 42,404 deaths respectively, which represent 38.4 % and 26.9 % of the total excess mortality in the SCB. In addition, Chronic Obstructive Pulmonary Disease (COPD) and Lung Cancer (LC) contribute a substantial fraction of mortality, accounting for 18.2 % (28,690) and 11.7 % (18,444). Compared to abovementioned endpoints, the burden associated with deaths from Lower Respiratory Infectious Diseases (LRI) is much smaller-contributing to approximately 2.3 % of total mortality burden in the SCB.

The disease burden in a specific location depends on the interplay of air quality, population density, and baseline mortality rates (Davidson et al., 2020). The spatial pattern of $PM_{2.5}$ mortality burden reveals a distributing pattern of annual average $PM_{2.5}$ concentrations and associated mortality across various urban areas within the SCB, highlighting significant attributable incidence in densely populated regions. Notably, populous areas including Chengdu Plain, the Southern Sichuan city cluster, and urban center of Chongqing exhibited substantial attributable deaths linked to high levels of $PM_{2.5}$ exposure. In particular, the density of traffic in highly urbanized areas (urban center of Chengdu and Chongqing) and the ubiquity of on-road vehicle emissions largely

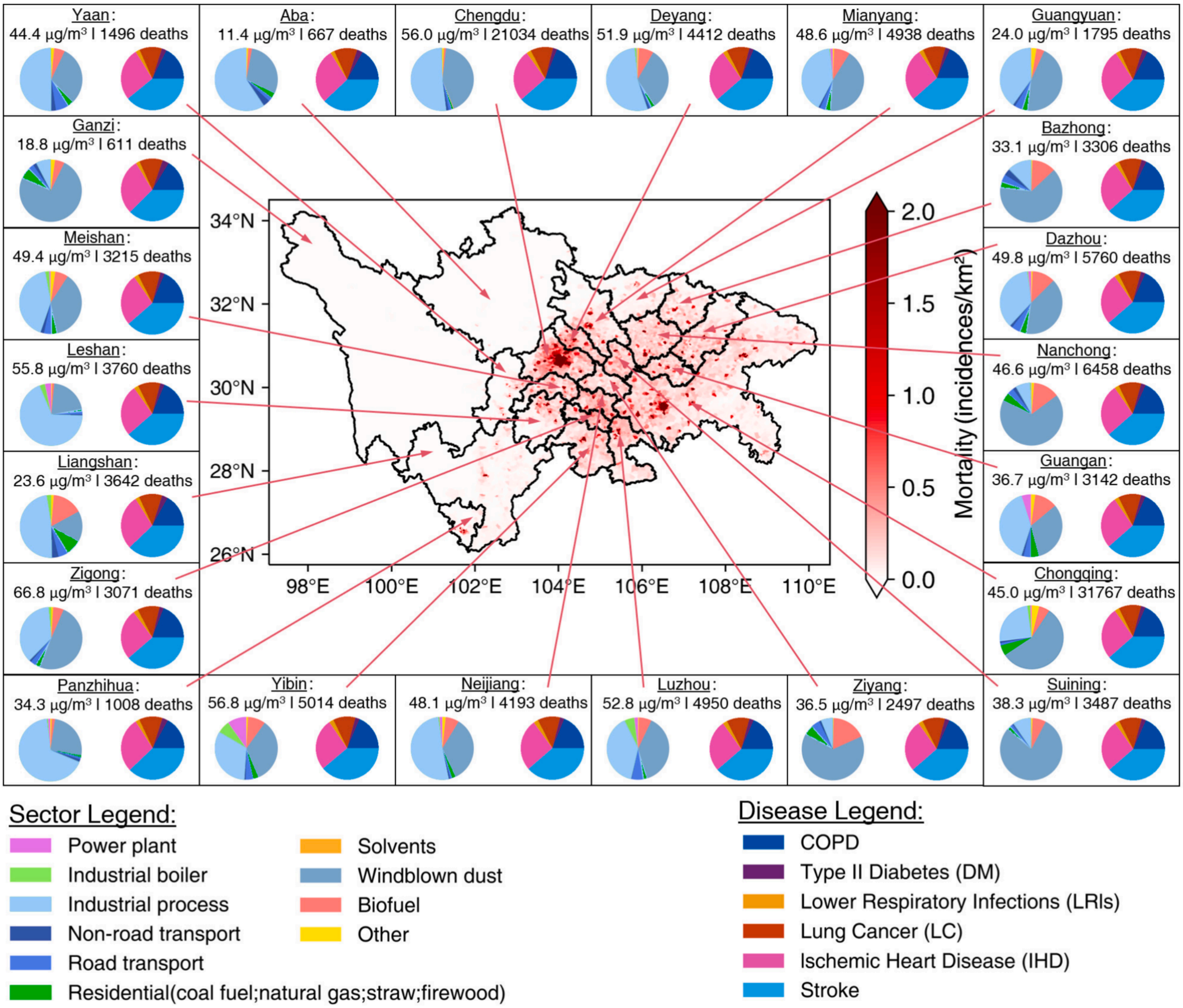

Fig. 8. Sector-based $PM_{2.5}$ emissions and absolute ambient $PM_{2.5}$ mortality burden and disease contributions for the SCB in 2017. (Note: The left pie charts are fractional sectoral source contributions of $PM_{2.5}$ emissions, and the right pie charts are relative disease contributions quantified by the revised GEMM model.).

explain the peaks in urban health burden and the large spatial gradient of estimated $PM_{2.5}$-burden that extends to suburban and rural areas. Further analysis shows a distinct spatial discrepancy between $PM_{2.5}$ concentrations and the number of attributable deaths (as seen in Fig. S1). For instance, Zigong suffered the highest annual average $PM_{2.5}$ concentration across the SCB at 66.8 μg/m^3, while the attributable deaths in Zigong were close to basin-average mortality burden. In Chengdu, the annual average $PM_{2.5}$ concentration was 56.0 μg/m^3, which resulted in 21,034 deaths. In contrast, with a slightly higher $PM_{2.5}$ level of 56.8 μg/m^3 in Leshan, it is estimated significantly fewer deaths (approximately 3,760 incidences). Such discrepancy clearly suggests that high attributable deaths are not solely determined by pollution levels but are also critically dependent on population density. As discussed in Sec 2.3 and Eq. (3), $PM_{2.5}$-attributable mortality is determined by four quantifiable variables and the changes in these variables contribute to the changes in $PM_{2.5}$-related mortality. This underscores the complexity of factors influencing health outcomes related to $PM_{2.5}$ exposure, including population density and age distribution. Areas with larger populations, such as Chengdu and Chongqing, not only have higher exposure levels but also higher population density, thus amplifying the public health impact of $PM_{2.5}$ exposure. Therefore, it is illustrated that population density acts as the most important factor in determining $PM_{2.5}$ disease burden across the SCB despite the relatively large differences of annual $PM_{2.5}$ levels among cities.

Although disease burden is quantitatively assessed using the GEMM model, it is important to acknowledge the uncertainty associated with $PM_{2.5}$ exposure and mortality estimates. First, the high-resolution dataset of $PM_{2.5}$ concentrations is subject to uncertainty introduced by the downscaling method which relies on satellite products and air quality model simulation (as detailed in Hammer et al., (2020)). A comparison of the annual $PM_{2.5}$ levels used in this study with ground-level $PM_{2.5}$ monitors and regional CMAQ model simulations over the SCB in 2017 at a resolution of 3 km × 3 km from Wu et al., (2023) shows good agreement and spatial consistency across the SCB. Additionally, uncertainties may stem from the limited number of epidemiological studies and the estimation methods used in the GEMM model. While local cohort studies conducted in the SCB could help reduce bias in estimates, the GEMM model remains widely recognized as the most advanced assessment tool, given its integration of Chinese cohort studies, robust theoretical foundation, and adaptability. Finally, other factors, such as total population, age distribution, and population distribution, may contribute to bias in $PM_{2.5}$-related mortality estimation. However, these factors are deemed minor in this work, as the most recent datasets are utilized, which are unlikely to significantly affect the major conclusions.

4. Discussion

In this study, the inter-annual changes of ambient $PM_{2.5}$ and anthropogenic emissions in the SCB over the period of 2013 to 2020 were revealed, presenting an insightful look into the dynamics of $PM_{2.5}$ pollution changes and health impacts. This analysis, grounded in a robust dataset comprising surface measurements and high-resolution bottom-up emission inventory underscores remarkable basin-wide decreases in $PM_{2.5}$ concentrations. In addition, the relationship between $PM_{2.5}$ exposure and mortality burden were quantified using the GEMM model at 1 km × 1 km, which lays the foundation for probing spatial gradient of exposure risk for vulnerable populations, thereby providing new insights from more detailed analysis at a fine-scale.

The results based on the abovementioned approach indicated that the most significant reduction of ambient $PM_{2.5}$ was found in the Chengdu Plain, where $PM_{2.5}$ levels dropped by 56 μg/m^3 in 2020 compared to 2013. The persistent decreases of $PM_{2.5}$ concentrations are mainly attributed to considerable reductions in both primary $PM_{2.5}$ emissions and gaseous pollutants (particularly NO_x) which are key precursors for secondary $PM_{2.5}$ formation. Sector-based analysis shows that $PM_{2.5}$ emissions saw a decline of 46.8 %, from 708.6kt in 2013 to 376.9kt in 2020. NO_x emissions exhibited slightly lower reductions with 1398.7kt in 2013 reduced to 1049.2kt in 2020.

Further analysis focused on the health impact of $PM_{2.5}$ exposure based on an updated GEMM model linked $PM_{2.5}$ exposure to 157,637 deaths in the SCB in 2017, with the majority of mortality burden attributed to stroke and ischemic heart disease. Compared to the mortality burden reported in prior studies, estimates of mortality burden here provide reasonable estimates of the magnitude of adverse health impacts, which are broadly consistent with prior studies targeted at city- and provincial- level (Geng et al., 2021). Moreover, this study further extends the literature beyond general coarse spatial scale toward high-resolution characterization of adverse outcomes of long-term $PM_{2.5}$ exposure across the SCB, which pinpoints the pathway and location of prioritized emission controls for decision-makers and stakeholders. Nevertheless, although high-resolution exposure has been identified through GEMM, the health benefits resulting from APPCAP and its underlying drivers over time deserve more attention and warrant in-depth investigation in the future.

The findings of this work highlight the successes of regulation efforts induced by APPCAP in substantially reducing ambient $PM_{2.5}$ concentrations. Despite substantial $PM_{2.5}$ decreases depicted across the SCB, attaining $PM_{2.5}$ air quality standard for densely populated cities (including Chengdu, Deyang, and Yibin) remains challenging due to ongoing industrialization and urbanization. Further reducing $PM_{2.5}$ and NO_x emissions poses great challenges to adaptation and calls for more aggressive mitigation given the approaching emission reduction capacity. In addition, the mortality burden caused by $PM_{2.5}$ exposure in the SCB is much higher than national average (McDuffie et al., 2021), which points to the demand for more stringent efforts to curb emissions. Concerning the highly varied $PM_{2.5}$ sources and the health impacts of $PM_{2.5}$ compositional diversity, future research should consider employing chemical transport models and adopting conclusive epidemiological data from emerging cohort studies to estimate source-oriented $PM_{2.5}$ and speciated health burdens of $PM_{2.5}$ at 1 km × 1 km to inform air quality management and safeguard human health (Apte & Manchanda, 2024).

CRediT authorship contribution statement

Xianyu Yang: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Resources, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Bingzheng Ben:** Writing – review & editing, Writing – original draft, Formal analysis, Data curation, Conceptualization. **Wenlei Wang:** Writing – review & editing, Visualization, Validation. **Bin Long:** Writing – review & editing. **Yan Xie:** Writing – original draft, Software, Resources, Project administration. **Kai Wu:** Writing – review & editing, Writing – original draft, Formal analysis, Data curation, Conceptualization. **Xiaoling Zhang:** Validation, Supervision, Resources, Project administration.

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.envint.2025.109366.

Data availability

Data will be made available on request.

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