

Article

Trends and Source Contribution Characteristics of SO₂, NO_x, PM₁₀ and PM_{2.5} Emissions in Sichuan Province from 2013 to 2017

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Abstract: As one of the most populated regions in China, Sichuan province had been suffering from deteriorated air quality due to the dramatic growth of economy and vehicles in recent years. To deal with the increasingly serious air quality problem, Sichuan government agencies had made great efforts to formulate various control measures and policies during the past decade. In order to better understand the emission control progress in recent years and to guide further control policy formulation, the emission trends and source contribution characteristics of SO₂, NO_x, PM₁₀ and PM_{2.5} from 2013 to 2017 were characterized by using emission factor approach in this study. The results indicated that SO₂ emission decreased rapidly during 2013–2017 with total emission decreased by 52%. NO_x emission decreased during 2013–2015 but started to increase slightly afterward. PM₁₀ and PM_{2.5} emissions went down consistently during the study period, decreased by 26% and 25%, respectively. In summary, the contribution of power plants kept decreasing, while contribution of industrial combustion remained steady in the past 5 years. The contribution of industrial processes increased for SO₂ emission, and decreased slightly for NO_x, PM₁₀ and PM_{2.5} emissions. The on-road mobile sources were the largest emission contributor for NO_x, accounting for about 32–40%, and its contribution increased during 2013–2015 and then decreased. It was worth mentioning that nonroad mobile sources and natural gas fired boilers were becoming important NO_x contributors in Sichuan. Fugitive dust were the key emission sources for PM₁₀ and PM_{2.5}, and the contribution kept increasing in the study period. Comparison results with other inventories, satellite data and ground observations indicated that emission trends developed in this research were relatively credible.



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1. Introduction

The Sichuan province is located in the hinterland of southwest China, and consists of 18 prefecture-level cities and three autonomous prefectures (see Figure 1). With the support of “Western Development Strategy”, Sichuan has made remarkable achievements in social and economic development in recent years. By 2017, the provincial GDP reached RMB 3268.05 billion, ranking the sixth among the 31 provinces in China mainland [1]. Meanwhile, the energy consumption, vehicle population and construction area in Sichuan increased rapidly, and accounted for approximately 5% of the country's total in 2017 [2]. However, the rapid urbanization and industrialization during the last decade also led to a dramatic increase of emissions. As a result, the Sichuan basin, geographic center of Sichuan province, has been facing severe air pollution, and has become one of the four traditional regions with frequent haze events [3–6].

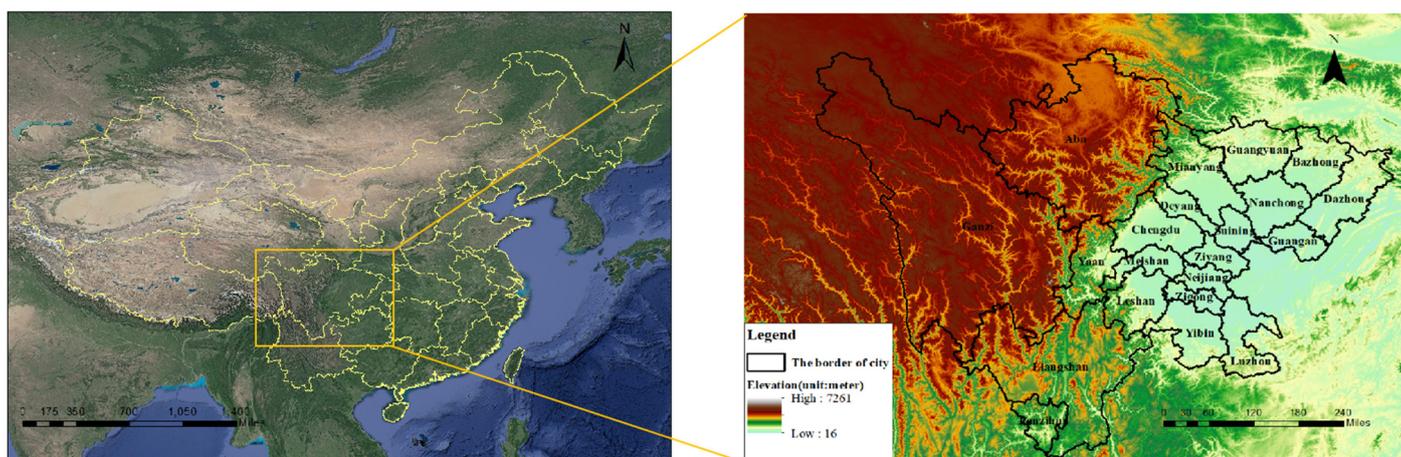


Figure 1. Geographical map and administrative division of Sichuan province.

To deal with the increasingly serious air quality problem, both national and local government agencies have made great efforts to formulate various control measures and policies during the past decade. The air pollution control from 2013 to 2017 was the most remarkable and systematic. At the national level, the State Council issued the Air Pollution Prevention and Control Action Plan in September 2013, with a goal to reduce PM₁₀ concentration in prefecture-level and above cities by more than 10% in 2017 compared with 2012. Then the Sichuan government promulgated the Sichuan Clean Air Action Plan (referred to as the Sichuan Action Plan). A series of stringent policies and measures were implemented during 2013–2017 in support of the Sichuan Action Plan, such as raising emission standards of industry and vehicles, eliminating outdated industrial capacity, phase-out of high-emission and old vehicles, and strengthening the control of fugitive dust and field burning of crop residues. These policies and control measures were summarized in Table 1. After implementing Sichuan Action Plan, the air quality of Sichuan has been improved greatly, which has been confirmed by both satellite-based and ground-based observations [7–9].

Table 1. Major policies and control measures implemented during 2013–2017 in Sichuan.

Emission Sources	Policies and Control Measure
Power plants Industrial combustion Industrial processes	<ol style="list-style-type: none"> 1. Installing flue gas desulfurization (FGD), denitrification facilities, and particulate matter control devices. 2. Enacting the “ultralow emission” standard for large-scale coal-fired power plants, which requests the emission limits for SO₂, NO_x, and particulates to be 35, 50, and 10 mg/m³, respectively. 3. Phase-out of industrial coal boilers less than 10 t/h in the urban area. 4. Installing FGD for coal boilers larger than 20 t/h. 5. Closing industrial enterprises with high energy consumption and emissions. 6. Installing denitrification facilities for cement plants. 7. Strengthening industrial emission standards for power plants, industrial boilers, steel industry, flat glass production, cement production, and brick production.
Vehicle sources	<ol style="list-style-type: none"> 1. Upgrading motor vehicle emission standards for all new vehicles (the National IV and V standards were implemented in 2013 and 2017, respectively). 2. Phase-out of yellow-label vehicles, including gasoline vehicles that are below National I standard, and diesel vehicles that are below National III standard. 3. Improving fuel quality and public transportation facilities.
Fugitive dust Field burning of crop residues	<p>Strengthening the control of dust resulting from paved road and construction sites.</p> <p>Reinforcing the control of field burning of crop residues.</p>

The air pollution is usually affected by source emissions, adverse meteorological conditions, and regional transportation [10–13]. Among them, source emissions have been

proved to be the most important driving force for the air quality deterioration, and the emission reduction is always one of the most effective ways to alleviate air pollution [14]. The improvement of air quality in Sichuan was remarkable during 2013–2017, and the source emission characteristics in this region may also change significantly. Although currently there are several nationwide inventories that addressed emission trends in Sichuan, the emissions were estimated based the top-down approach, and cannot properly or accurately represent the regional emission characteristics. How the related control measures affect the emission contribution characteristics in Sichuan is not yet clear. Taken in this sense, there is an urgent need for analyzing pollutant emission trends, in order to better understand the emission control progress due to the implementation of Sichuan Action Plan and to guide further control policy formulation. Thus, the objectives of this study are to characterize emission trends of primary pollutants (SO₂, NO_x, PM₁₀ and PM_{2.5}) from 2013 to 2017 and to assess the impacts of control measures on source characteristics in Sichuan.

2. Materials and Methods

The methodological approaches used in this study include (1) methods for emission estimation; (2) activity data processing and determination of emission factors; (3) verification of emission trends; (4) methods for uncertainty analysis. The details are given in the following:

2.1. Methods for Emission Estimation

Referring to the EMEP/EEA air pollutant emission inventory guidebook [15], National Emissions Inventory (NEI) developed by the United States Environmental Protection Agency [16] and guideline for emission inventory issued by China government, the emission sources were classified into eight groups, as listed in Table 2. They were stationary combustion, industrial processes, on-road mobile sources, nonroad mobile sources, fugitive dust, field burning of crop residues, catering sources, and waste disposal sources.

Table 2. Emission source categorization in Sichuan province.

Category	Subcategory	Category	Subcategory
Stationary combustion sources	Power plants	On-road mobile sources	Buses
	Industrial combustion		Light duty trucks (LDT)
Industrial processes	Residential combustion	Nonroad mobile sources	Medium duty trucks (MDT)
	Building materials industry		Heavy duty trucks (HDT)
	Steel industry		Low-speed trucks (LST)
	Nonferrous metallurgy industry	Fugitive dust	Tricycles
	Petroleum refining and coking industry		Motorcycles
	Chemical industry		Plane
	Pulp and paper industry		Marine
	Mining industry	Field burning of crop residues	Construction machinery
	Pharmaceuticals industry		Agricultural machinery
	Alcoholic beverage industry	Catering sources	Construction dust
Food industry	Paved road dust		
On-road mobile sources	Other industries	Waste disposal sources	Sewage treatment
	Light duty vehicles (LDV)		Waste disposal
	Medium duty vehicles (MDV)		
	Heavy duty vehicles (HDV)		
	Taxis		

A bottom-up approach was used for emission sources where detailed activity data were available, such as power plants, industry combustion, industrial processes, and waste disposal sources. For residential combustion, mobile source, fugitive dust, field burning of crop residues, and catering sources, with only city-wide or region-wide statistical data available, the top-down approach was adopted instead. The commonly used methodology

for emission estimation includes material balance algorithm, emission factor method, real-world measurement, and model estimation. Among them, emission factor method is the most widely used, which is based on corresponding emission factors and activity data by the following equation:

$$E_p = \sum_i A_i \times EF_{i,p} \quad (1)$$

where p and i represent the pollutant type and specific sector, respectively; E is the annual emission of a given pollutant; A is the activity level and EF denotes the emission factor. Activity data usually refers to fuel consumption by types for stationary combustion, product output or raw materials for industrial processes, or vehicle kilometers traveled (VKT) for on-road mobile sources and others [17].

2.2. Activity Data Processing and Determination of Emission Factors

2.2.1. Activity Data Processing

Most of the activity data in this study were obtained from official statistics. The activity data of power plants, industrial combustion, industrial processes, and waste disposal were collected from Sichuan Provincial Pollutant Statistical Report (SPPSR) in 2013–2017. The SPPSR is an official statistic with detailed information for industry plants, including location (latitude and longitude), fuel type, fuel consumption, sulfur content, ash content, boiler type and capacity, stack parameter, control device, removal efficiency, product output, and raw material. More than 7000 enterprises are included in the SPPSR for each year, and can be used to calculate emissions for industry sources. For on-road mobile sources, 11 vehicle types were considered including six types of passenger cars and trucks (gross weight: light, medium, heavy), buses, taxis, tricycles, low-speed trucks, and motorcycles. The number of vehicles was collected from three data sets, including updated data of the first National Census of Pollution Sources (NCPS) in 2010, the SPPSR in 2011–2015, and the field survey according online system. Detailed activity data processing for vehicle sources were described in previous studies [18]. For other emission sources, activity data were mainly collected from the Sichuan Statistical Yearbook 2014–2018 [1], Sichuan Transport Yearbook 2014–2018 [19], and field surveys conducted in Sichuan province. Detailed activity information and the related data sources are summarized in Table 3.

Table 3. Activity data for major emission sources.

Sector	Subsector	Major Activity Data	Sources
Power plants	—	Fuel consumption by fuel types	SPPSR (2013–2017)
Industrial combustion	—	Fuel consumption by fuel types	SPPSR (2013–2017)
Residential combustion	—	Fuel consumption by fuel types	Sichuan Statistical Yearbook 2014–2018 [1]
Industrial processes	—	Product output and raw materials consumption	SPPSR (2013–2017)
On-road mobile sources	—	Vehicle populations and yearly VKT	NCPS (2010), SPPSR (2011–2015), field survey
	Plane	Full landing and take-off (LTO)	Field survey
Nonroad mobile sources	Marine	Oil consumption	Sichuan Statistical Yearbook 2014–2018 [1]
	Construction machinery	Oil consumption	Sichuan Statistical Yearbook 2014–2018 [1]
	Agricultural machinery	Oil consumption	Sichuan Statistical Yearbook 2014–2018 [1]

Table 3. Cont.

Sector	Subsector	Major Activity Data	Sources
Fugitive dust	Paved road dust	Roads length and traffic flow	Sichuan Transport Yearbook 2014–2018 [19], field survey
	Construction dust	Construction area	Sichuan Statistical Yearbook 2014–2018 [1]
Field burning of crop residues	—	Crop production	Sichuan Statistical Yearbook 2014–2018 [1]
Catering sources	—	Number of restaurants and consumption of cooking oil	Field survey
Waste disposal	—	Amount of sewage treatment and waste disposal	SPPSR (2013–2017)

2.2.2. Determination of Emission Factors

Most of the emission factors in this study were directly cited from the Technical Manual for the Compilation of Air Pollutant Emission inventory [20]. While, SO_2 and PM emission factors for fuel combustion sources can be calculated by Equations (2) and (3) based on material balance method.

$$EF_{SO_2} = 2 \times S \times (1 - sr) \times (1 - \eta) \quad (2)$$

where EF_{SO_2} is the emission factor of SO_2 , S and sr represent the sulfur content and sulfur retention in ash, η is the removal efficiency.

$$EF_{PM} = 2 \times Aar \times (1 - ar) \times f_{PM} \times (1 - \eta) \quad (3)$$

where EF_{PM} is the emission factor of PM_{10} or $PM_{2.5}$; Aar and ar represent the ash content and ash retention in bottom ash, respectively; f_{PM} is the PM_{10} (or $PM_{2.5}$) fractional abundance in PM ; η is the removal efficiency. S and Aar for each plant were collected from SPPSR (2013–2017), while sr , ar , and f_{pm} were derived from [21]. In addition, emission factors of on-road mobile sources, paved road dust, and catering sources were derived from local measurement. Emission factors for on-road mobile sources were estimated by International Vehicle Emission (IVE) model, a widely used tool for estimating vehicle emission factors for the developing countries. Vehicle-fleet technology distribution and vehicle activities were the major inputs for the IVE model, which were derived from the investigation and field measurement conducted in cities of Sichuan [21]. As for the paved road dust, PM emission factors were estimated using Equation (4) [22].

$$EF = k(sL)^{0.91} \times (W)^{1.02} \times \left(1 - \frac{P}{4N}\right) \quad (4)$$

where EF is the emission factor of size-specific PM; sL is the silt loading, g/m^2 ; W is the mean weight of the vehicle fleet, tons; k is a constant in g/VKT , set as 0.62 and 0.15 for PM_{10} and $PM_{2.5}$, respectively; P is the number of “wet” days with at least 0.254 mm of precipitation for a certain period; N is the number of days for a certain period. Silt loading and vehicle weight were estimated by local measurement and survey data [23]. The numbers of “wet” days were counted based on the meteorological data from China Meteorological Data Service Center (<http://data.cma.cn/>). Emission factors for catering sources were estimated from the measured pollutants concentration, flue gas flow, and oil consumption in a certain period of time [24].

2.3. Verification of Emission Trends

In order to validate the reliability of emission trends, the estimated results of SO_2 , NO_x , PM_{10} and $PM_{2.5}$ were compared with other inventories, satellite data, and ground observations. The provincial emissions extracted from Multiresolution Emission Inventory for

China (MEIC, <http://meicmodel.org/>) and Emission Database for Global Atmospheric Research (EDGAR, <https://www.eea.europa.eu/themes/air/links/data-sources/emission-database-for-global-atmospheric>) were used to compare with emission trends developed in this study.

The provincial SO₂ and NO₂ column concentrations were extracted from data products by National Aeronautics and Space Administration (NASA), and from Ozone Monitoring Instrument (OMI) satellite with a spatial resolution which were used to compare with SO₂ and NO_x emissions, respectively. The aerosol optical depth (AOD) data with 1-km resolution for Sichuan province were acquired from Moderate-resolution Imaging Spectroradiometer (MODIS) aerosol product by NASA's Goddard Earth Sciences Distributed Active Archive Center. AOD is defined as the integral of the extinction coefficient of aerosol in the vertical direction, and represents the light attenuation by aerosols [3]. AOD reflects the extent of regional aerosol pollution and can be used to verify the PM₁₀ and PM_{2.5} emission trends. The verifications and evaluations of the OMI data products and MODIS AOD products have been conducted by several scholars, and the results showed significant correlation between the satellite-based data and the ground values [8,25]. Annual satellite-based data for Sichuan were calculated by averaging the corresponding daily values.

Annual air quality data for Sichuan province were collected from the website of Sichuan Ecological and Environmental Monitoring Center (<http://www.scnewair.cn:6112/publish/index.html>). The provincial average concentrations were calculated by averaging the values at all national monitoring sites, which were usually located in the urban area in Sichuan.

2.4. Methods for Uncertainty Analysis

The approaches for characterizing uncertainties of emission inventories including qualitative, semiquantitative, and quantitative approaches [26]. In this study, both qualitative and quantitative methods were used to evaluate the uncertainty in emission estimates depending on the data availability. Qualitative analyses involve listing and discussing the possible uncertainty sources that affect the estimation results. Quantitative approaches include the use of bootstrap simulation to quantify uncertainty in emission factors or activity data, and the use of Monte Carlo simulation to propagate uncertainties in model inputs to that in emission estimates. Detailed description about bootstrap simulation and Monte Carlo simulation can be found in another study [27].

3. Results and Discussion

The materials presented in this section include (1) emission trends in Sichuan province; (2) characterization of emission source contribution variation; (3) emission trends validation; (4) uncertainties in emission estimates. The details in this section are given in the following:

3.1. Emission Trends in Sichuan Province

SO₂, NO_x, PM₁₀ and PM_{2.5} emission trends of anthropogenic sources in Sichuan province from 2013 to 2017 are shown in Figure 2. SO₂, NO_x, PM₁₀ and PM_{2.5} emissions decreased by 52%, 12%, 26%, and 25%, while the GDP and energy consumption increased by 40% and 3%, indicating the effectiveness of control measures adopted by government in recent years. The great emission reduction in SO₂ indicated accurate source identification and effective emission control in the past 5 years. However, it should be also noticed that the annual decrease rate for SO₂ emission was falling down, indicating less room for further emission reduction. The NO_x emission increased slightly after 2015, probably due to the complicated source contribution in Sichuan province. Though PM_{2.5} and PM₁₀ emissions presented totally decreasing trends during the study period, the emissions remained steady since 2016, indicating the challenge of PM emission control. Generally, the different reductions of pollutant emissions were determined by the source sector distributions and related emission mitigation efforts. The trends for detailed emission sources will be discussed as follows.

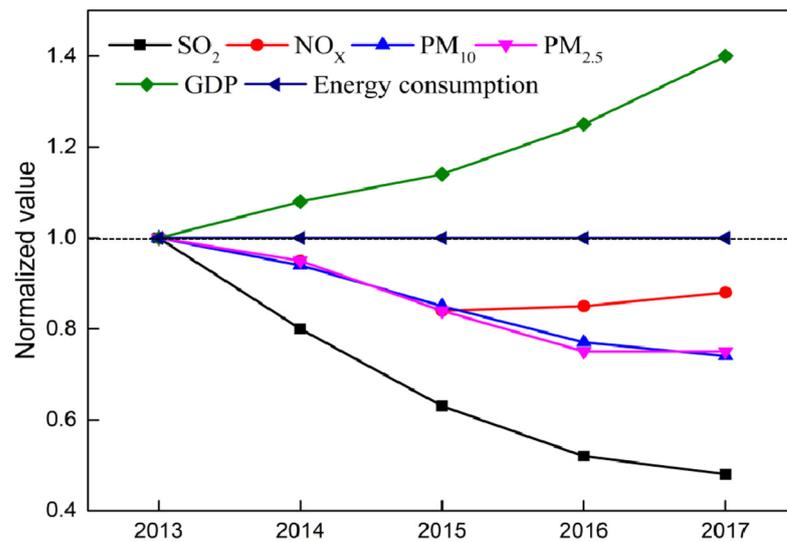


Figure 2. Trends in pollutant emissions, GDP, and energy consumption in Sichuan, all data were normalized to the year 2013.

3.2. Characterization of Emission Source Contribution Variation

In this section, the variations in source contribution characteristics of SO₂, NO_x, PM₁₀ and PM_{2.5} emissions during the 2013–2017 are discussed, meanwhile, possible impacts of policies and control measures on source characteristics are also identified.

3.2.1. SO₂ Emission Characteristics Variation

Figure 3 presents variation in SO₂ source contributions from 2013 to 2017. Apparently, power plants, industrial combustion, and industrial processes were the three major contributors, and showed different trends in contributions. The contribution of power plants showed a downward trend, accounting for 38% in 2013 and 11% in 2017 of provincial SO₂ emissions. The contribution of industrial combustion remained steady in the past 5 years, accounting for approximately 24–29%. However, the contribution of industrial processes increased from 25% in 2013 to 46% in 2017.

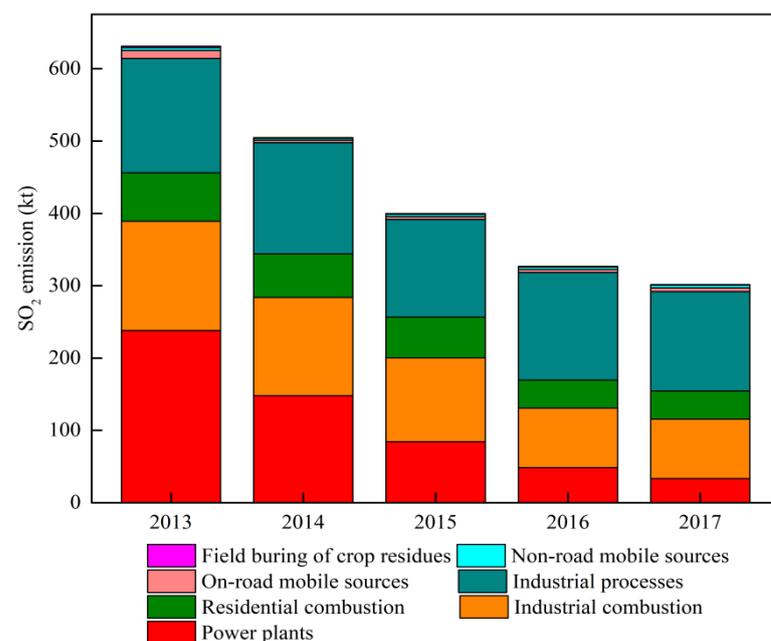


Figure 3. Changes in SO₂ emission in Sichuan during 2013–2017.

Figures 4 and 5 present SO₂ emission trends and related activity data for power plants and industrial combustion. As important contributors to SO₂ emission, power plants and industrial combustion have reduced emissions obviously in the past 5 years, and decreased by 86% and 46%, respectively. The apportionment of high SO₂ removal efficiency increased year by year. Take power plants for example, plants with removal efficiency above 80% accounted for 30% in 2013 and 74% in 2017. In addition, there was higher removal efficiency in power plants than industrial combustion, this was mainly because FGD facilities were required to be installed in all power plants with strict supervision, while only required in large industrial boilers. Overall, installing and operating FGD facilities in power plants and large industrial boilers were main reasons for the rapid decrease of SO₂ emission. The coal consumption for power plants also showed a downward trend from 2013 to 2017, with a decrease rate of 58%, while electricity generation increased by about 35% in the past 5 years. It can be concluded that the energy efficiency in power plants has improved in recent years, which also contributed to the emission reduction. Additionally, the rapid decreasing of SO₂ emission from power plants might be attributed to the implementation of shutting down small and high-emitting power generation units. Although the energy consumption of the whole province increased slowly, the implementation of clean energy measures such as “coal to gas” or “coal to electricity” had led to a decline in coal consumption for industrial combustion year by year, and also led to the reduction of SO₂ emission.

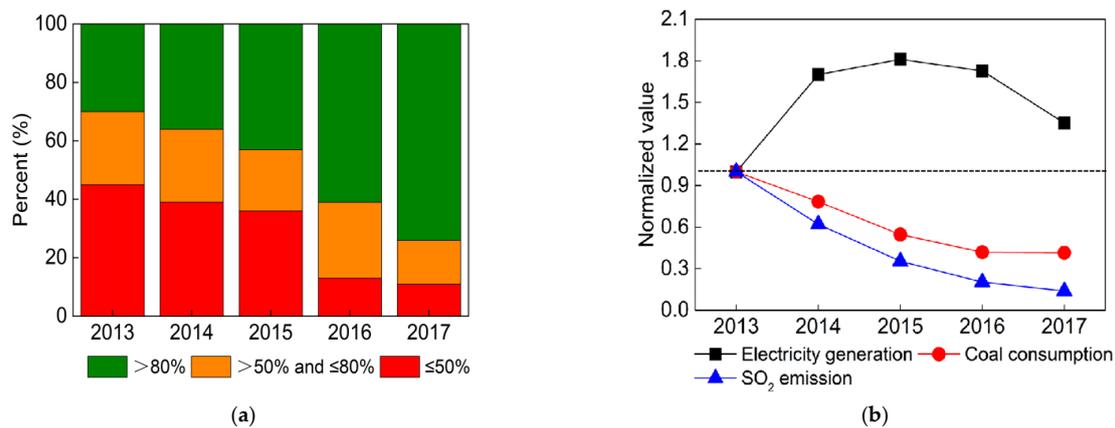


Figure 4. Trends in (a) desulfurization efficiency for power plants; (b) SO₂ emission and related activity data for power plants, the related data were normalized to the year 2013.

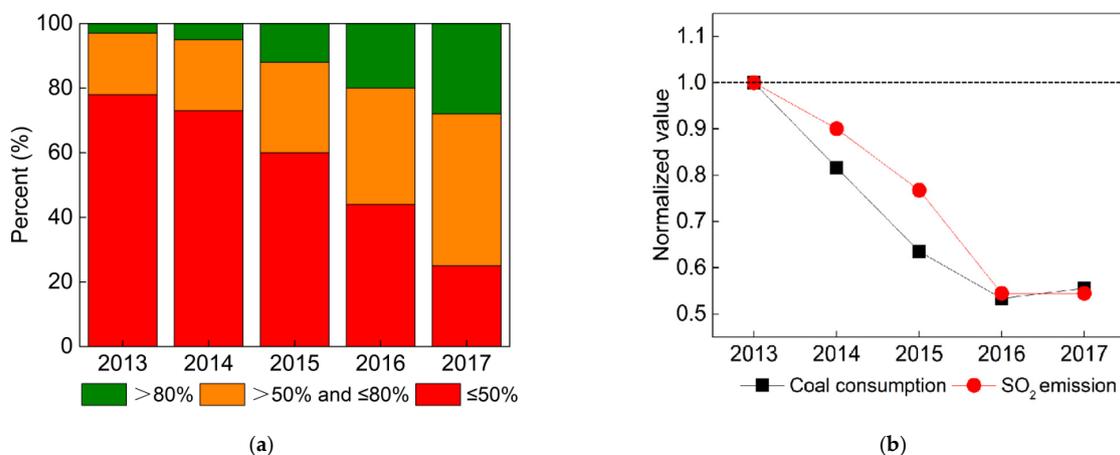


Figure 5. Trends in (a) desulfurization efficiency for industrial boilers (capacity ≥ 10 t/h); (b) SO₂ emission and coal consumption for industrial combustion, the related data were normalized to the year 2013.

Industrial processes were also important contributors to SO₂ emission. However, the decrease rate per half-decade for SO₂ emission from industrial processes was lower than that from power plants and industrial combustion. Figure 6 shows the SO₂ emission from major industries. It can be seen that the reduction of SO₂ emission for industrial processes was mainly from the steel industry. Although the steel production presented a downward trend in recent years [1,19], the implementation of the new emission standard has tightened the pollutants concentration limits strictly, and the improved desulfurization efficiency has played a positive role in SO₂ emission reduction. It was worth noticing that SO₂ emission from building materials industry and chemical industry both showed an upward trend. The rise of SO₂ emission in building materials industry were mainly from brick production, ceramic production, and glass production. For the SO₂ emission from chemical industry, sulfuric acid manufacture was the main reason for the increase.

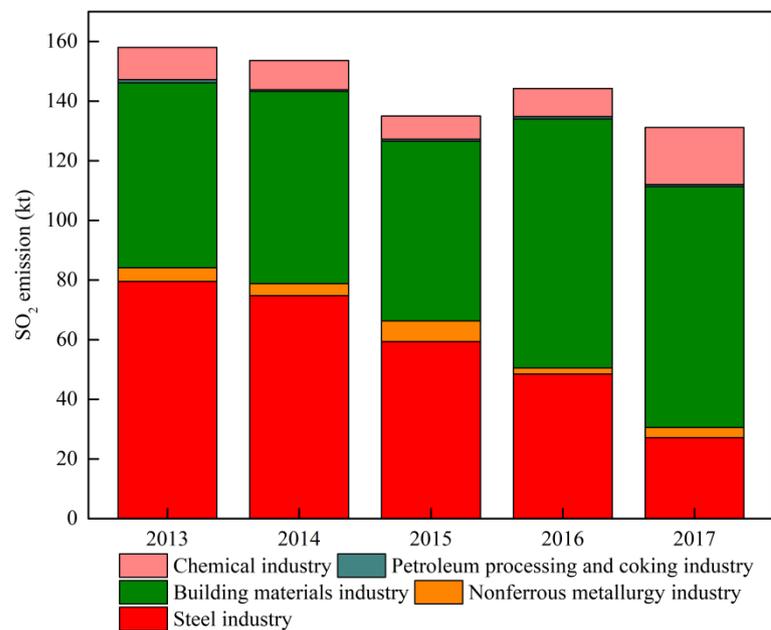


Figure 6. Trends in SO₂ emission from major industrial processes.

3.2.2. NO_x Emission Characteristics Variation

The variation of NO_x source contributions from 2013 to 2017 is depicted in Figure 7. Obviously, on-road mobile sources were the largest contributor, and the contribution increased during 2013–2015 and then decreased, accounting for about 32–40%. In addition, industrial processes and nonroad mobile sources also made great contributions to NO_x emission. Contributions of industrial processes fluctuated around 21–25% from 2013 to 2017 while contributions of nonroad mobile sources increased significantly, from 15% in 2013 to 24% in 2017. The most obvious decrease was the emission from power plants, with total emission decreased by 78%, and it only contributed approximately 4% in 2017. NO_x emission from industrial combustion decreased from 2013 to 2015, but increased slightly since 2016, and its contributions fluctuated around 10–12% in the past 5 years.

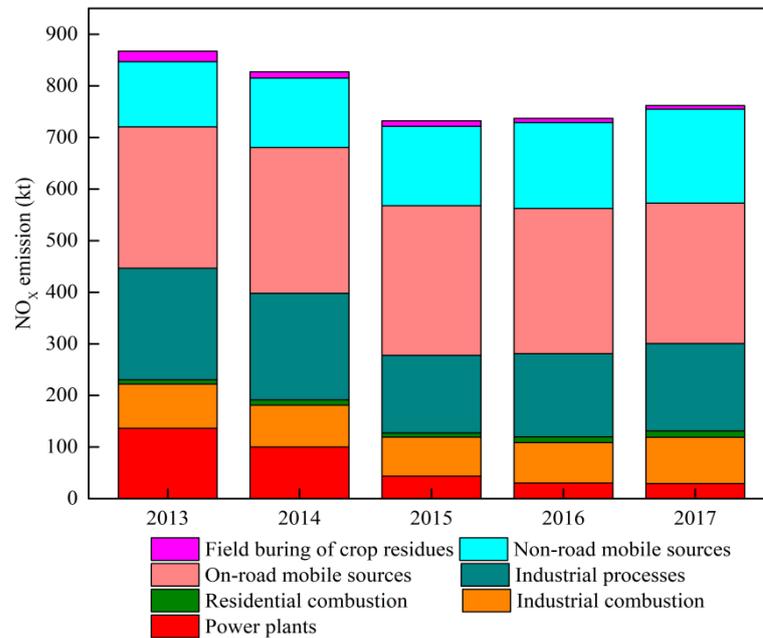


Figure 7. Changes in NO_x emission in Sichuan during 2013–2017.

Figure 8 showed trends in NO_x emission from on-road mobile sources and vehicle population from 2013 to 2017. The vehicle population kept rising in the study period, while the NO_x emission decreased from the year 2015. Heavy duty trucks were the main contributors for NO_x emission. The number of heavy duty trucks had increased by 27% in the past 5 years, while the related emission increased by 9%. It can be concluded that control measures like phase-out of high-emitting vehicles, upgrading vehicle emission standards, and popularizing clean fuel vehicles slowed down the annual growth rate of NO_x emission.

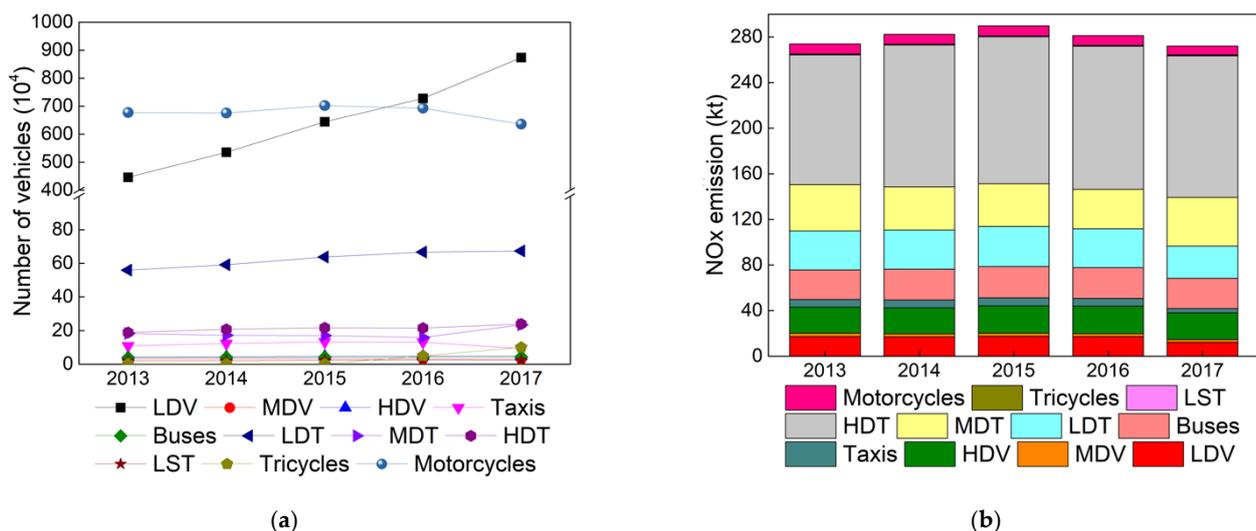


Figure 8. Trends in (a) number of vehicles; (b) NO_x emission from vehicle sources.

Figure 9 shows the changes in NO_x emission from major industrial processes. As can be seen, NO_x emission from cement industry and flat glass industry decreased significantly with the stable production output from 2013 to 2017 [1]. The reason can be attributed to the installation of flue gas denitration devices under strict supervision. Take cement industry as an example, the proportion of enterprises with denitration efficiency greater than 30% increased from 12.9% in 2013 to 56.4% in 2017. The NO_x emission from brick industry

increased by approximately 37% compared to 2013. Most of the brick plants were small or medium-sized, and few denitration facilities were installed due to the high costs, thus the emission increases were mainly affected by product output.

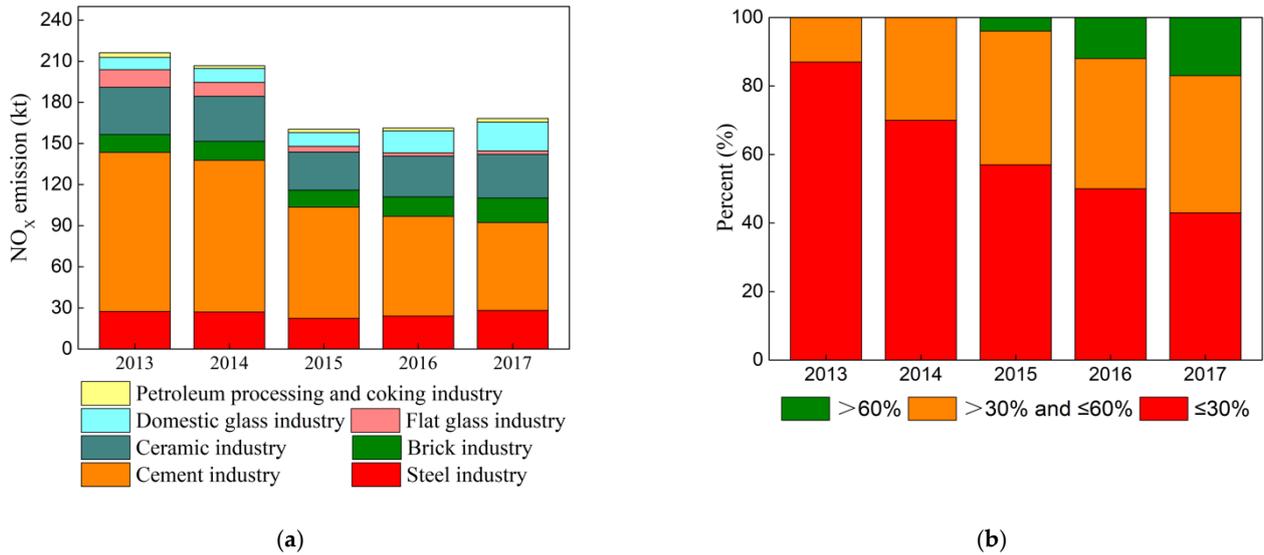


Figure 9. Trends in (a) NO_x emission from major industrial processes; (b) denitration efficiency for cement industry.

Coal combustion and natural gas combustion were the two major contributors to the NO_x emission from industrial combustion. Figure 10 presents the NO_x emission from industrial combustion with different fuel types during 2013–2017. The emission from coal fired boilers decreased significantly, while emissions from natural gas boilers continue to rise, and the annual growth rate has accelerated since the year of 2015. Therefore, the total NO_x emission from industrial combustion decreased during 2013–2015, and started to increase obviously afterward. The decrease of emission from coal fired boilers can be attributed to the implementation of clean energy transformation and installation of denitration devices in large boilers. The increase of NO_x emission from natural gas fired boilers can be attributed to the following reasons. Firstly, the natural gas consumption increased significantly in recent years. Secondly, the emission concentration of NO_x from natural gas boilers was relatively high, while almost all natural gas boilers in Sichuan do not install denitration devices, thus NO_x emission increased together with the increase of consumption.

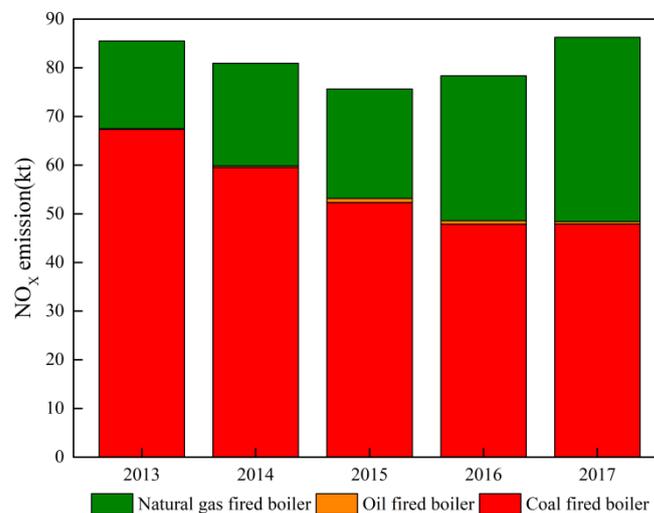


Figure 10. Trends in NO_x emission from fuel-based industrial combustion.

In general, NO_x emission reduction in power plants, coal fired boilers, cement industry and flat glass industry was highly effective in recent years. Meanwhile, phase-out of yellow-label vehicles also made an important contribution to the emission reduction. However, NO_x emission from natural gas combustion, brick industry, and nonroad mobile sources increased significantly, indicating these emission sources should be paid more attention in the future.

3.2.3. PM_{10} and $\text{PM}_{2.5}$ Emissions Characteristics Variation

Figure 11 presents the changes in PM_{10} and $\text{PM}_{2.5}$ source contributions from 2013 to 2017, respectively. Paved road dust and industrial processes were the two major contributors for PM_{10} emission, with average contributions of 32% and 29%, respectively. The contribution of industrial processes showed a declining trend (33% in 2013 and 28% in 2017), while the contribution of paved road dust kept growing (29% in 2013 and 35% in 2017). Besides, construction dust and field burning of crop residues were also important PM_{10} contributors. The contribution of construction dust increased from 13% in 2013 to 17% in 2017. Due to the strict burning prevention, the contribution of field burning of crop residues continued to decrease, from 12% in 2013 to 7% in 2017. Similar with PM_{10} emission, industrial processes and paved road dust also contributed significantly to $\text{PM}_{2.5}$ emission, with average contributions of 46% and 18%, respectively. The variation of $\text{PM}_{2.5}$ emission characteristics were just consistent with PM_{10} emission.

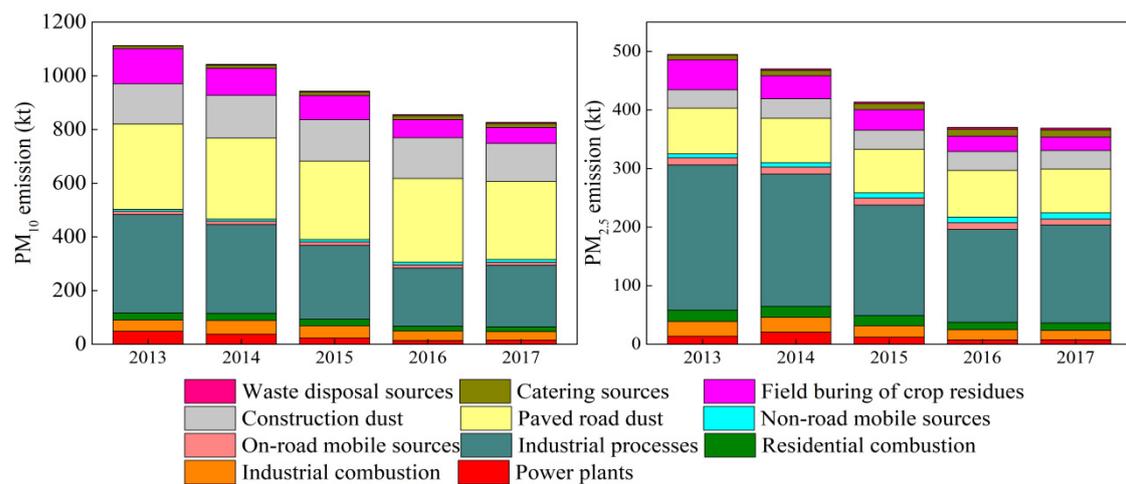


Figure 11. Changes in PM_{10} and $\text{PM}_{2.5}$ emissions in Sichuan during 2013–2017.

The $\text{PM}_{2.5}$ emissions of major industrial processes from 2013 to 2017 are presented in Figure 12. It can be concluded that the two major industries for $\text{PM}_{2.5}$ emission, building materials industry and steel industry both showed an obvious downward trend, with total emission decreasing by 45% and 46%, respectively. Emissions of industrial processes were mainly affected by product output and removal efficiency. The cement production in 2017 was close to that in 2013, while pig iron production in 2017 decreased by approximately 6% compared with 2013 [1]. For the comprehensive dust removal efficiency, as can be seen in Figure 13, the proportion of high removal efficiency in cement industry and steel industry increased obviously in recent years. Taking cement industry as an example, the proportion of enterprises with removal efficiency higher than 99% increased from 31% in 2013 to 51% in 2017. Generally, the improvement of removal efficiency was the main reason for the $\text{PM}_{2.5}$ emission reduction. Nevertheless, $\text{PM}_{2.5}$ emission from some industries increased significantly, such as the nonferrous metallurgy industry. Some nonferrous smelting industries still executed the comprehensive furnace emission standard, which has been established for a long time, and the concentration limits were relatively loose. Thus, the update of emission standards for nonferrous industries should be accelerated and the relevant emission concentration limits should be tightened.

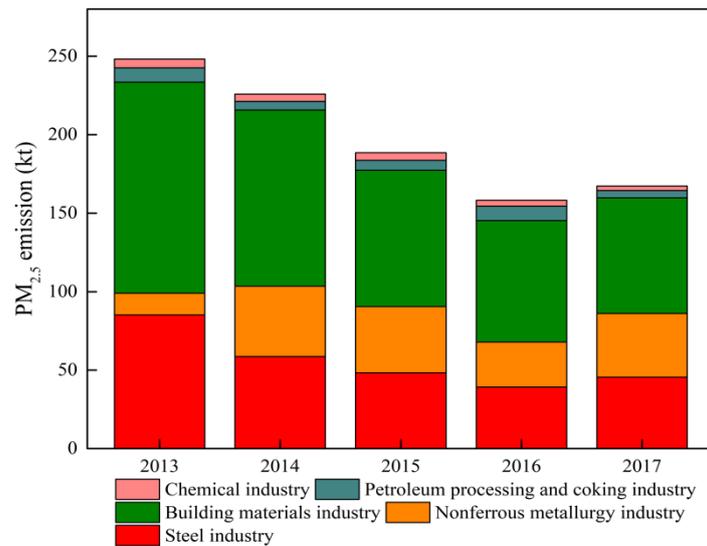


Figure 12. Trends in PM_{2.5} emission from major industrial processes.

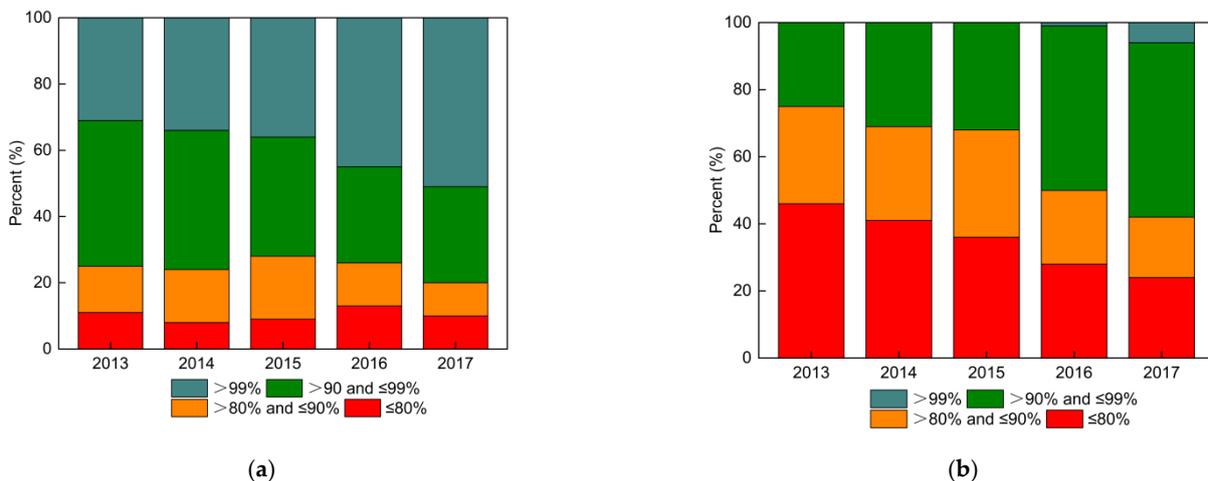


Figure 13. Trends in dust removal efficiency for (a) cement industry and (b) steel industry.

The PM_{2.5} emissions from fugitive dust remained steady in the past 5 years. Among them, emissions from paved road dust in 2017 only decreased by 4% compared to 2013, and the change of construction dust emissions was also small in the study period. Emissions from paved road dust were mainly affected by rainfall, traffic flow, road length, and road sweeping frequency. In recent years, the improvement of sweeping frequency of urban roads especially in heavy pollution days had led to emission reductions. Emissions from construction dust were influenced by the construction area and relative control measures. With the rapid development of urbanization, the construction area of Sichuan province increased from 473.8 km² in 2013 to 605.9 km² [1,19]. Meanwhile, control measures like enclosure, coverage, and flushing were usually used to reduce particulate matter emissions. However, most of these measures were administrative and cannot be quantified, thus the control effects were limited.

3.3. Emission Trends Validation

3.3.1. Comparison with Other Emission Inventories

A preliminary comparison was conducted between the derived emission trends with the results from MEIC and EDGAR. As shown in Figure 14, SO₂, PM₁₀ and PM_{2.5} emission trends in this study showed a good agreement with the results from MEIC. However, NO_x emission reported in MEIC showed a continuous downward trend, while that emission in

this study showed a slight increase from the year of 2015. The discrepancy can be attributed to the different source contribution variation, especially for mobile sources (including on-road mobile sources and nonroad mobile sources). NO_x emission from mobile sources in this study increased by 13% during 2013–2017, while decreased by 10% in MEIC. Emission control for the nonroad mobile sources was inadequate in Sichuan during 2013–2017, besides, nonroad equipment always has a relatively long useful life, thus the related emissions were expected to increase with the growth of activity data. Emissions reported in EDGAR during 2013–2015 remained relatively steady, this can be partially attributed to the underestimation of pollutant removal efficiency in a global scale. Meanwhile, estimates of SO₂ and NO_x emissions in this study were lower than those from MEIC and EDGAR. This discrepancy might be due to the use of different activity data and emission factors in the three inventories. As for PM₁₀ emission, estimated results in this study were higher than the values reported by MEIC and EDGAR. This discrepancy might be due to the missing emission sources in the global-scale and country-scale inventories, like fugitive dust and field burning of crop residues. However, when comparison was only based on power plants, industrial sources, residential sources, and mobile sources, estimation of PM_{2.5} emission in this study was lower than those from MEIC and EDGAR. This discrepancy might be due to the different estimation of removal efficiency for industrial sources and the different activity data processing for residential sources.

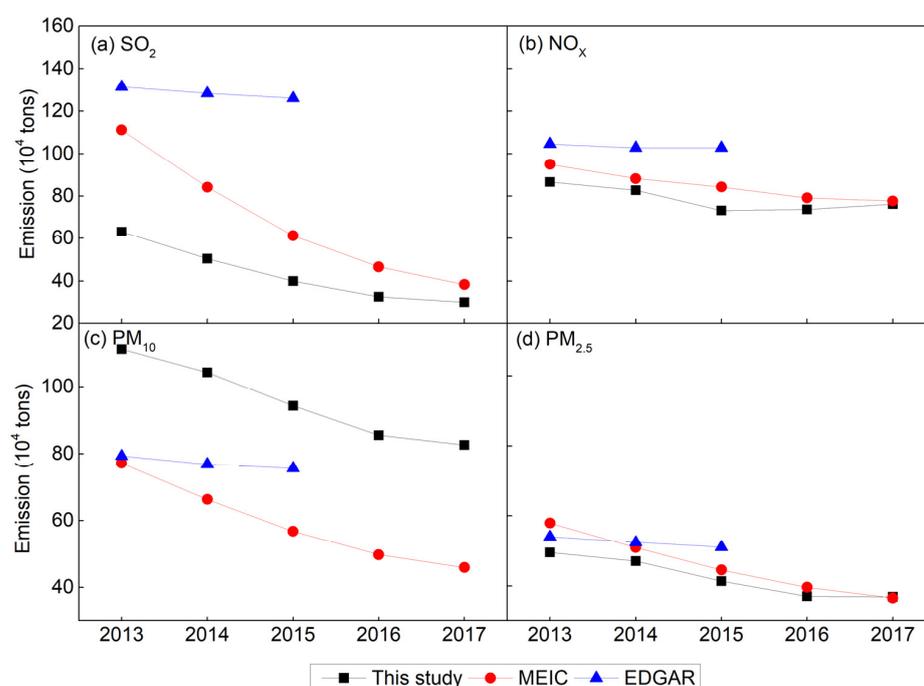


Figure 14. Comparison with other emission inventories.

3.3.2. Comparison with Satellite Data and Ground Concentrations

Figure 15 shows trends in emissions, satellite data, and ground concentrations from 2013 to 2017. Generally speaking, trends in SO₂ emission, satellite-based SO₂ column concentration, and SO₂ ground concentration presented broad agreement in temporal evolution. They all presented continuous downward trends (decreased by 52%, 25%, and 59% during 2013–2017, respectively), except that SO₂ column concentration in 2015 showed a slight increase. Since most of the national monitoring sites were located in the urban area, the lower decrease rate per half-decade of SO₂ column concentration might be due to the inadequate control of SO₂ pollution in suburb. Additionally, some SO₂ emission sources, like small and polluted enterprises in remote areas, might be missed in this study.

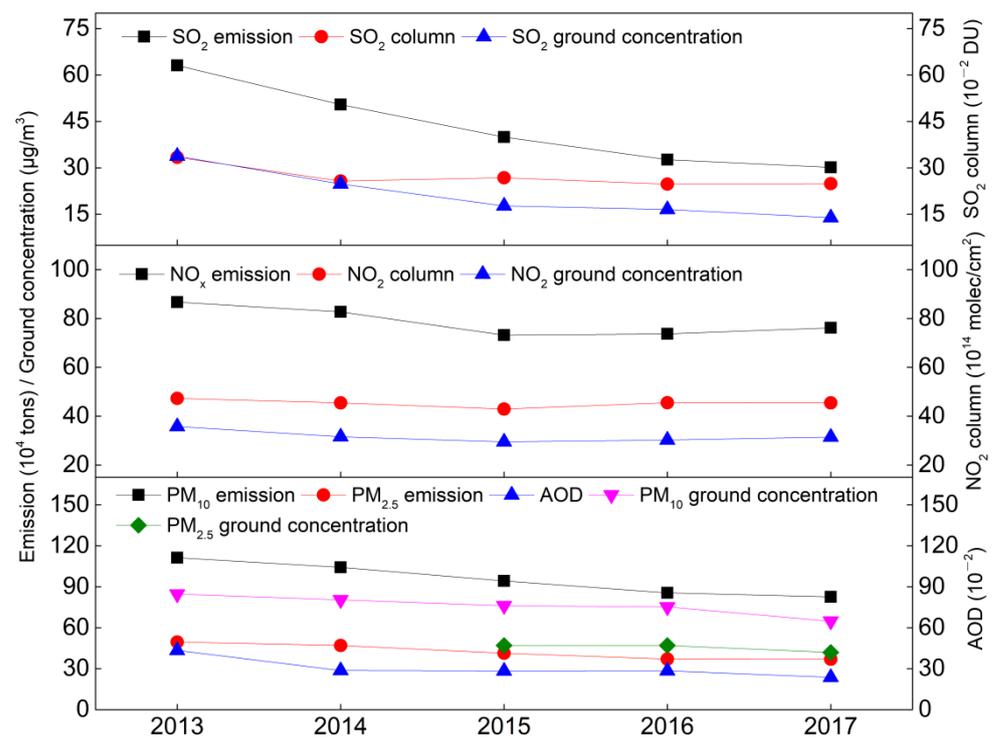


Figure 15. Trends in emissions, satellite data, and ground concentrations.

Basically, NO_x emission, satellite-based NO₂ column value, and NO₂ ground observation presented similar variation during study period, all decreased from 2013 to 2015 and then increased slightly. Similar to SO₂, the total decrease rate per half-decade of NO₂ column concentration was the lowest, which might be caused by the underestimated emissions for some areas, and continued improvement of emission inventory was needed in Sichuan.

As for PM₁₀ and PM_{2.5}, all of the corresponding emissions, satellite-based AOD values and ground concentrations showed downward trends during 2013–2017. In comparison, a general downward trend in PM₁₀ and PM_{2.5} emissions but a relative steady trend in AOD and PM₁₀ ground concentration was observed in 2014–2016. Meanwhile, the PM_{2.5} concentration in 2016 remained unchanged compared with the previous year. This discrepancy can be attributed to the following reasons: (1) the meteorology variations had important effects on air quality, especially for fine particulate matter; (2) PM_{2.5} was often transported directionally from one city to another city inside the basin [13], and might influence the regional PM_{2.5} pollution; (3) emission estimates of PM₁₀ and PM_{2.5} in this study also had some uncertainties due to the complicated source characteristics.

In a word, there was a relatively good agreement between emission trends, satellite-based data, and ground observations during 2013–2017. These results indicated that emission trends developed in this research were reasonable to a certain extent.

3.4. Uncertainties in Emission Estimates

In this study, uncertainties for power plants, industrial combustion, cement industry, steel industry, paved road dust, construction dust, and field burning of crop residues are quantified. Table 4 lists the uncertainty ranges at the 95% confidence interval for emissions estimated from major sources. Uncertainties in power plants and industrial combustion were relatively lower than those in other sources, due to use of the bottom-up approach, and having detailed fuel consumption activity data and domestic measurements of emission factors. For the cement production and steel industry, though a bottom-up approach was used for emission estimation, there were relatively higher uncertainties compared with power plants and industrial combustion. The reasons were mainly attributed to the lack of

detailed information for production processes, pollutant removal efficiency, and localized emission factors. Uncertainties in paved road dust and construction dust were relatively high, which arose not only from empirical emission factor models, but also from the activity data estimated based upon surveys.

Table 4. Uncertainty assessment of major emission sources estimated in Sichuan.

Emission Source	Uncertainty Range *			
	SO ₂	NO _x	PM ₁₀	PM _{2.5}
Power plants	(−9%,9%)	(−23%,20%)	(−28%,42%)	(−30%,47%)
Industrial combustion	(−15%,15%)	(−24%,27%)	(−32%,45%)	(−36%,46%)
Cement industry	(−35%,35%)	(−31%,41%)	(−44%,40%)	(−54%,43%)
Steel industry	(−41%,45%)	(−47%,50%)	(−46%,42%)	(−51%,46%)
Paved road dust			(−52%,89%)	(−54%,92%)
Construction dust			(−45%,77%)	(−47%,86%)
Field burning of crop residues	(−23%,25%)	(−28%,35%)	(−33%,56%)	(−37%,58%)

* Uncertainty ranges are quantitatively characterized on the 95% confidence interval.

The uncertainties for other emission sources were not quantified due to the absence of supporting data. For residential combustion, the uncertainties were mainly from the lack of city-based official statistics data of fuel consumption and local emission factors. For emissions of on-road mobile sources, the uncertainties can be introduced by emission factors calculated by IVE model, vehicle's annual travel distance, and the number of different types of vehicle based on survey. As for emission sources like nonroad mobile sources, catering sources, and waste disposal sources, uncertainties may be also high, since only a few studies about proper statistics of activity data and emission measurements have been conducted at present. Moreover, on-road mobile sources and nonroad mobile sources are considered as important uncertainty contributors to the total emission, especially for NO_x.

4. Summary and Conclusions

Emission trends and variations in source contribution of SO₂, NO_x, PM₁₀ and PM_{2.5} in Sichuan from 2013 to 2017 were characterized in this study. The emission trends results showed that SO₂ emission decreased sharply by approximately 52%. NO_x emission decreased during 2013–2015 but started to increase slightly in the following years, with a decrease rate per half-decade of approximately 12%. PM₁₀ and PM_{2.5} emissions presented continuous decrease during the study period, which declined by 26% and 25%, respectively. The source characterization results showed that contribution of power plants declined significantly, especially for SO₂ and NO_x emissions, indicating the effectiveness of related control measures. Industrial combustion was an important contributor to SO₂ and NO_x emissions, and the contribution remained steady during 2013–2017. Industrial processes were becoming more prominent for SO₂ emissions, and were still important contributors to NO_x, PM₁₀ and PM_{2.5} emissions, though the contribution decreased slightly. Worthy of attention was that emissions from natural gas fired boilers, nonferrous metallurgy and brick production increased obviously during study period and became important emission sources in Sichuan. On-road mobile sources were the largest NO_x contributor, and nonroad mobile sources gradually became major NO_x emission contributors in Sichuan, which need immediate control actions on them. Fugitive dust including paved road dust and construction dust contributed significantly to PM₁₀ and PM_{2.5} emissions, which should be paid more attention in the future.

Emission trends in this study showed a good agreement with the results from MEIC. Meanwhile, there was a broad agreement between emission trends, satellite-based data, and ground observations. The comparison results indicated that emission trends developed in this research were credible to some extent. However, there were relatively high uncer-

tainties in emission estimates due to the lack of detailed activity data and local emission factors. More work should be conducted on the estimation of industrial processes, fugitive dust, on-road mobile sources, and nonroad mobile sources in the future.

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