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Decadal changes in anthropogenic source contribution of PM$_{2.5}$ pollution and related health impacts in China, 1990–2015

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Abstract. Air quality in China has changed dramatically in response to rapid development of the economy and to policies. In this work, we investigate the changes in anthropogenic source contribution to ambient fine particulate matter (PM$_{2.5}$) air pollution and related health impacts in China during 1990–2015 and elucidate the drivers behind the decadal transition. We estimate the contribution of five anthropogenic emitting sectors to ambient PM$_{2.5}$ exposure and related premature mortality over China during 1990–2015 with 5-year intervals, by using an integrated model framework of a bottom-up emission inventory, a chemical transport model, and the Global Exposure Mortality Model (GEMM). The national anthropogenic PM$_{2.5}$-related premature mortality estimated with the GEMM for nonaccidental deaths due to noncommunicable diseases and lower respiratory infections rose from 1.26 million (95 % confidence interval (CI) [1.05, 1.46]) in 1990 to 2.18 million (95 % CI [1.84, 2.50]) in 2005; then, it decreased to 2.10 million (95 % CI [1.76, 2.42]) in 2015. In 1990, the residential sector was the leading source of the PM$_{2.5}$-related premature mortality (559 000, 95 % CI [467 000, 645 900], 44 % of total) in China, followed by industry (29 %), power (13 %), agriculture (9 %), and transportation (5 %). In 2015, the industrial sector became the largest contributor of PM$_{2.5}$-related premature mortality (734 000, 95 % CI [615 500, 844 900], 35 % of total), followed by the residential sector (25 %), agriculture (23 %), transportation (10 %), and power (6 %). The decadal changes in source contribution to PM$_{2.5}$-related premature mortality in China represent a combined impact of socioeconomic development and clean-air policy. For example, active control measures have successfully reduced pollution from the power sector, while contributions from the industrial and transportation sectors have continuously increased due to more prominent growth in activity rates. A transition in fuel consumption has dominated the decrease in the contribution from residential sector. Meanwhile, the contribution from the agriculture sector has continuously increased due to persistent NH$_3$ emissions and enhanced formation of secondary inorganic aerosols under an NH$_3$-rich environment.

1 Introduction

Ambient air pollution is one of the most harmful environmental issues arising from development. It is a major risk factor to public health and is linked to various adverse health outcomes (Lim et al., 2012; GBD 2015 Risk Factors Collaborators, 2016). Globally, ambient air pollution caused millions of deaths (Lelieveld et al., 2015; Cohen et al., 2017; Burnett et al., 2018), ranging from 4.2 million (Cohen et al., 2017) to 8.9 million (Burnett et al., 2018) in 2015, depending on the adoption of risk functions. The associated economic costs were valued at between 3.8 % (World Bank, 2007) and 9.9 % of the gross domestic product (GDP; World Bank, 2016). The largest number of deaths occurred in China (Cohen et al., 2017; Burnett et al., 2018), with a combination of severe air pollution and high population density (Lelieveld et al., 2015; J. Liu et al., 2016b). Urgent actions are needed

to reduce air pollution and improve public health (Zhang et al., 2012).

Driven by the rapid socioeconomic development and by environmental policies (Zheng et al., 2018), air quality in China has changed dramatically over the past decades (Xing et al., 2015). Between 1990 and 2015 the country increased its total energy consumption by a factor of 3.5, thermal power generation by a factor of 7.6, pig iron production by a factor of 10, and civil vehicle population by a factor of 28 (National Bureau of Statistics, 2016). Consequently, China has experienced increasingly adverse impacts from worsening air quality (Xing et al., 2015) and associated diseases for decades (Lim et al., 2012; Cohen et al., 2017). Over large areas, the ambient concentrations of fine particulate matter with an aerodynamic diameter equal to or smaller than 2.5 μm ($PM_{2.5}$) far exceeded the World Health Organization (WHO) air quality guidelines as well as China's national air quality standards (Wang et al., 2014, 2015; Zhang and Cao, 2015). During 2013–2014, only 3 out of the 31 provincial capital cities in China had the $PM_{2.5}$ annual concentration below the national standard grade II of $35\,\mu g\,m^{-3}$, and the highest concentration of $144\,\mu g\,m^{-3}$ in occurred Shijiazhuang, which was over 3 times higher than the standard (Wang et al., 2014).

China began responding to air pollution in the 1970s. The Law on the Prevention and Control of Atmospheric Pollution was formulated in 1987. Starting from 2005, national targets for sulfur dioxide (SO_2) and nitrogen oxide (NO_x) emission reductions were successively included in the 11th and 12th five-year plans (FYPs), and abatements have been achieved in the power, industrial, and transportation sectors in recent years (Huo et al., 2015; Liu et al., 2015; F. Liu et al., 2016; van der A et al., 2017). In 2013, to tackle the severe and widespread air pollution, the Chinese Government launched the national *Air Pollution Prevention and Control Action Plan*. Even more aggressive measures were implemented to reduce $PM_{2.5}$ concentrations by up to 25 % in major metropolitan areas by 2017 (Zheng et al., 2018; Cheng et al., 2019).

The complex interactions between economic development and environmental regulations have caused air pollutant emissions over China to change significantly in the past 25 years. After growing rapidly during the early stages of economic development (Lu et al., 2011; Zhao et al., 2013), SO_2 and NO_x emissions (observed by satellite instruments) peaked in 2007 and 2011–2012 (Krotkov et al., 2016; F. Liu et al., 2016; van der A et al., 2017). Studies on the historical $PM_{2.5}$ air pollution and related health impacts and the decadal transition of source contributions provide opportunities to evaluate the effectiveness of past policies and set the direction for prioritized control strategies in the future.

In China, annual estimates of mortality attributable to $PM_{2.5}$ exposure ranged from 1.10 to 1.37 million during 2010–2015 (Lelieveld et al., 2015; J. Liu et al., 2016b; Hu et al., 2017; Zheng et al., 2017), and studies have identified industrial and residential sectors and coal burning activities as the leading contributors to premature deaths attributable to $PM_{2.5}$ during 2010–2013 (Hu et al., 2017; Ma et al., 2017; Gu et al., 2018). However, constrained by the availability of long-term emission datasets, limited efforts have been made to explore the decadal transitions of source contributions to $PM_{2.5}$ air pollution and related premature mortality in response to socioeconomic development and clean-air policy. Recently, Zheng et al. (2019) have evaluated the emission source contributions to $PM_{2.5}$-related mortality during 2005–2015 and revealed the leading role of the agricultural and industrial sectors. But studies with longer temporal coverage to illustrate the decadal transition of different source sectors are still lacking. The changing patterns of sectoral emissions will result in varied source contributions at different stages of development (Zheng et al., 2018). Learning from the past is helpful for formulating future policies to optimize benefits.

In this study, we combine a state-of-the-art Chinese long-term emission dataset with a regional air quality model and the Global Exposure Mortality Model (GEMM; Burnett et al., 2018) and investigate the decadal changes in the anthropogenic source contribution of $PM_{2.5}$ air pollution and related health impacts in China from 1990 to 2015 for every 5-year period. We quantify the contribution from the power, industrial, residential, transportation, and agricultural sectors in each year; illustrate different transition routes for different source categories; and highlight the opportunities for further mitigation.

2 Materials and methods

2.1 Bottom-up emission inventories

The bottom-up emission inventory used in this study for mainland China during 1990–2015 was obtained from the Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org/, last access: 21 June 2020). The emissions from other Asian countries and regions were taken from the MIX emission inventory (M. Li et al., 2017b). The MEIC model, developed by Tsinghua University, is a technology-based, bottom-up, anthropogenic emission model. By integrating a dynamic methodology with up-to-date activity and local emission factors, it can provide model-ready emission inventories from 1990 to the present. The MEIC model includes more than 700 anthropogenic emission sources and quantifies emissions for 10 pollutants and greenhouse gases: SO_2, NO_x, carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), ammonia (NH_3), carbon dioxide (CO_2), $PM_{2.5}$, PM_{10}, black carbon (BC), and organic carbon (OC). The road transportation emissions were estimated at the county level and distributed across grids with the China Digital Road-network Map (Zheng et al., 2014). Emissions from coal-fired power plants were constructed from the unit-based China coal-fired

Power plant Emissions Database (CPED), which improved the spatial and temporal resolution of power emissions (Liu et al., 2015). In addition, the model has an improved speciation framework to generate anthropogenic NMVOC emissions for various chemical mechanisms (Li et al., 2014). Model-ready emissions of SO_2, $PM_{2.5}$, NO_x, NH_3, CO, OC, BC, and NMVOCs with the Carbon Bond version 5 (CB05) mechanism (Whitten et al., 2010) chemical mechanism were generated for the period from 1990 to 2015 in 5-year intervals using the MEIC and used as the Models-3 Community Multiscale Air Quality (CMAQ) input to simulate the $PM_{2.5}$ concentrations over China.

2.2 $PM_{2.5}$ exposure and source contributions

We used the Weather Research and Forecasting (WRF) model version 3.5.1 and the CMAQ model version 5.0.1 (https://www.cmascenter.org/cmaq/, last access: 21 June 2020) to simulate the $PM_{2.5}$ concentrations over China at a horizontal resolution of 36 km. Our WRF-CMAQ domain covers East Asia, including China, North and South Korea. Japan, and other Asian countries (Fig. S1 in the Supplement). The National Centers for Environmental Prediction Final Analysis (NCEP-FNL) data were used as meteorological initial and boundary conditions to drive the WRF model. Since NCEP-FNL data were only available after 2000, for the years 1990 and 1995, we used the meteorological data in 2000 to drive the WRF model. The meteorological fields simulated by the WRF and the emissions generated by the MEIC model were then used as the inputs to the CMAQ model. Biogenic emissions were calculated by MEGAN v2.1 (Guenther et al., 2012). In the CMAQ model, the CB05 gas-phase chemistry and AERO6 aerosol chemistry with the ISORROPIA II thermodynamic equilibrium module (Fountoukis and Nenes, 2007) were used. The boundary conditions were generated with the global transport model GEOS-Chem (Bey et al., 2001). For each year, the CMAQ model ran continuously for the entire year; the 7 d at the end of the previous December were used for model spin-up. Details concerning the model configuration were provided in our previous study (Zheng et al., 2015). To evaluate the performance of the WRF-CMAQ model, we conducted a detailed validation of the meteorological field simulations of the WRF and $PM_{2.5}$ mass concentrations by the CMAQ. More details are provided in the Supplement (Table S1, Figs. S2 and S3).

In addition to CMAQ standard simulations between 1990 and 2015 at 5-year intervals, we conducted five zero-out sensitivity simulations for each year by subtracting the emissions of each source sector from the total emissions. The source sectors comprised power, industry, residential, transportation, and agriculture. Then, the contribution of each source sector was determined by the difference between the standard and sensitivity simulations. Since we focused on the contribution and relative importance of anthropogenic source sectors, we conducted another "clean" simulation for each

year by removing all the Chinese anthropogenic emissions in the total model emissions, to exclude the contributions of boundary conditions and emissions from biogenic sources, dust, sea salt, and other countries. Finally, to manage mass conservation, we implemented a grid-level normalization for the six sensitivity simulations to match the total $PM_{2.5}$ concentrations in the standard simulations.

2.3 Estimates of health impacts attributable to long-term $PM_{2.5}$ exposure

Long-term exposure to $PM_{2.5}$ has adverse impacts on human health; the most serious impact is death. Besides $PM_{2.5}$, ozone also has adverse health effects. In this study, we focus on the premature mortality due to long-term $PM_{2.5}$ exposure and did not consider the impact of ozone. The premature mortality attributable to $PM_{2.5}$ can be determined by the application of the GEMM functions (Burnett et al., 2018). The GEMM was developed based only on cohort studies from 16 countries with outdoor air pollution that covers the global exposure range. It predicts hazard ratios between the long-term exposure to $PM_{2.5}$ and the nonaccidental deaths due to noncommunicable diseases and lower respiratory infections (NCD+LRI) and deaths to five specific causes (5-COD). The hazard ratio function of the GEMM is described in Eq. (1).

$$
\text{GEMM}(z)
$$
$$
= \exp\left\{ \left[\theta \ln\left(\frac{z}{\alpha}+1\right)\right] \middle/ \left[1 + \exp\left(-\frac{z-\mu}{\nu}\right)\right]\right\}, \quad (1)
$$

where $z = \max(0, PM_{2.5} - cf)$, with cf representing the counterfactual $PM_{2.5}$ concentration of $2.4\,\mu g\,m^{-3}$, below which no additional risk is assumed. The parameters θ, α, μ, and ν describe the shape of the hazard ratio faction.

Following Burnett et al. (2018), we estimated the premature mortality attributable to $PM_{2.5}$ for both NCD+LRI and 5-COD. The latter includes death due to ischemic heart disease (IHD), stroke, lung cancer (LC), chronic obstructive pulmonary disease (COPD), and LRI. The premature mortality attributable to $PM_{2.5}$ was calculated for population subgroups by year, age, sex, and cause using Eq. (2), and the uncertainty and 95 % confidence interval were estimated through the standard error (SE) of parameter θ in Eq. (1).

$$
\Delta\text{Mort}_{\text{yr, s, a, c}}(z)
$$
$$
= y_{0_{\text{yr, s, a, c}}} \times \text{Pop}_{\text{yr, s, a}} \times \left[1 - 1/\text{GEMM}_{\text{a, c}}(z)\right], \quad (2)
$$

where $\Delta\text{Mort}_{i, j, k, l}(z)$ is the premature mortality attributable to $PM_{2.5}$ exposure level z in year yr for sex s, age group a, and cause c; $y_{0_{\text{yr, s, a, c}}}$ is the baseline mortality rate in year yr for sex s, age group a, and cause c; $\text{Pop}_{\text{yr, s, a}}$ is the population exposed to $PM_{2.5}$ in year yr for sex s and age group a; and $\text{GEMM}_{\text{a, c}}(z)$ is the hazard ratio associated with $PM_{2.5}$ exposure at level z for cause c in age group a.

The national baseline mortality rates of NCD, IHD, stroke, LC, COPD, and LRI by age and sex and population estimates

by age and sex for each year were obtained from the Global Burden of Disease Study 2016 (Global Burden of Disease Collaborative Network, 2017a, b). The year-specific population distributions were obtained from the Gridded Population of the World version 4 (GPWv4; http://sedac.ciesin.columbia.edu/data/collection/gpw-v4; Doxsey-Whitfield et al., 2015), which has a horizontal resolution of $0.0083° \times 0.0083°$. The 1990–2015 gridded population and annual average $PM_{2.5}$ concentration ($36\,km \times 36\,km$) simulated by the CMAQ were then regridded to a uniform domain over China for mortality estimation at a horizontal resolution of $0.1° \times 0.1°$. The $PM_{2.5}$-related premature mortality contributed by each sector was estimated using the gridded relative source contributions determined by the CMAQ sensitivity simulations.

Besides, to identify the driving factors underlying the changes in long-term health impact, we calculated the contributions from four factors to the net changes in $PM_{2.5}$-related premature mortality between the neighboring years, i.e., population growth, population aging, baseline mortality rates, and $PM_{2.5}$ exposure. Following the previous decomposition method (Cohen et al., 2017), we calculated the average factor contributions through all the change sequences in the four factors.

3 China's air pollution regulations and responses of emissions

In recognition of the importance of air pollution prevention, China has implemented air pollution controls since the 1970s. Table 1 lists the development sequence of the major air quality regulations in China; Fig. S4 shows the timetable of the emission standards implemented in the major sectors during the past decades, and Table 2 presents the annual emissions of SO_2, NO_x, $PM_{2.5}$, and NH_3 contributed by the agriculture, industry, power, residential, and transportation sectors. Historically, coal has been dominant in the energy system, contributing between 60 % and 70 % of the primary energy. Coal burning has caused high levels of SO_2 and total suspended particulates (TSP) in the air. In 1987, the Law on the Prevention and Control of Atmospheric Pollution was formulated with the intent of reducing emissions from industry and from coal burning. Subsequently, the law was revised in 1995 and again in 2000. During this period, the Two Control Zones policy (with acid rain and SO_2 pollution control zones) was established, and SO_2 emission controls were implemented to mitigate the acid rain and SO_2 pollution problems. A series of emission standards was established in the power (GB13223-1991, GB13223-1996, and GB13223-2003) and industrial sectors (GB4915-1996, GB 9078-1996, GB4915-2004) to reduce the SO_2, particulate matter, and NO_x emissions (Fig. S4). In the transportation sector, China followed the vehicle emission standard system developed by the European Commission. Since 1998 China has taken a

series of measures to address vehicle emissions, including implementing and updating vehicle emission standards for new vehicles and phasing out old, high-emission vehicles (Huo et al., 2015). In 2000 and 2011, China implemented the national state I emission standards for light-duty gasoline vehicles and heavy-duty diesel vehicles, respectively. However, these measures did not keep up with the rapid growth of the economy and fossil fuel use, and the national emissions of SO_2, NO_x, and $PM_{2.5}$ increased by 142 %, 207 %, and 54 % from 1990 to 2005. Since 2005, national targets for reducing SO_2 and NO_x emissions have been included in the 11th and 12th five-year plans (FYPs). Installation of flue gas desulfurization (FGD) systems, selective catalytic reduction (SCR), and selective noncatalytic reduction (SNCR) equipment in coal-fired power plants and high-emission industries (such as iron and steel and cement); phaseout of small power plants; and more stringent vehicle emission standards have been mandated to achieve these targets (Liu et al., 2015; F. Liu et al., 2016; van der A et al., 2017; Zheng et al., 2018). In response, national SO_2 emissions dropped from 33.0 to 27.8 Mt by 16 % in the period of 2005–2010, which was even greater than the target reduction rate of 10 %. But NO_x emissions kept growing during the 11th FYP due to limited end-of-pipe measures and started to drop during the 12th FYP. In 2012, $PM_{2.5}$ was included as an indicator in the new ambient air quality standards (GB 3095–2012). Subsequently, in 2013, China issued the national *Air Pollution Prevention and Control Action Plan* (hereafter the Action Plan), which for the first time made a commitment to reducing ambient $PM_{2.5}$ concentrations by up to 25 % during 2013–2017. To fulfill the air quality targets, the government proposed 10 pollution control measures and implemented a series of more stringent emission standards for the power, industrial, and transportation sectors (Fig. S4) that have significantly reduced air pollutant emissions and substantially improved the air quality in China (Zheng et al., 2018; Cheng et al., 2019; Xue et al., 2019). As a result, the SO_2, NO_x, and $PM_{2.5}$ emissions in 2015 had decreased dramatically by 39 %, 10 %, and 22 %, compared with the levels in 2010. The 13th FYP was published recently. In addition to SO_2 and NO_x emissions, control targets were also established for VOC (volatile organic compound) emissions.

4 Results

4.1 Trends in annual average anthropogenic $PM_{2.5}$

As shown in Table 2, during 1990–2015, the SO_2, NO_x, $PM_{2.5}$, and NH_3 emissions in China increased from 13.6, 6.4, 8.9, and 7.2 to 16.9, 23.7, 9.1, and 10.5 Mt, with peaks occurring in 2005, 2010, 2005, and 2015, respectively. In response, the ambient $PM_{2.5}$ concentration over China has changed markedly during the 25-year study period. Figure 1 illustrates the annual average anthropogenic $PM_{2.5}$ con-

Table 1. Milestones of major air quality regulations in China over the past decades. BTH is Beijing–Tianjin–Hebei; YRD is Yangtze River Delta; PRD is Pearl River Delta.

Year	Policy	Targets and/or measures
1987	Issued atmospheric pollution prevention law	Aimed at emission control from industry and coal burning
1995	Revised atmospheric pollution prevention law	Included acid rain and SO_2 pollution controls
2000	Revised atmospheric pollution prevention law	Established two control zones – acid rain and SO_2 pollution; implemented SO_2 total emission control, vehicle emission control, and road dust control in the two control zones
2005	11th Five-Year Plan	Reduce national SO_2 emissions by 10 %
2010	12th Five-Year Plan	Reduce national SO_2 and NO_x emissions by 8 % and 10 %
2012	Ambient air quality standards	Included $PM_{2.5}$ as an indicator in the standard; hold the annual average concentration of SO_2, NO_2, CO, maximum daily 8 h average ozone, $PM_{2.5}$, and PM_{10} below $60\,\mu g\,m^{-3}$, $40\,\mu g\,m^{-3}$, $4\,mg\,m^{-3}$, $160\,\mu g\,m^{-3}$, $35\,\mu g\,m^{-3}$, and $70\,\mu g\,m^{-3}$, respectively, to meet the Class II standard
2013	*Air Pollution Prevention and Control Action Plan*	By 2017, reduce the inhalable particle concentration in cities at the prefecture level and above by over 10 %; reduce the $PM_{2.5}$ concentrations in the BTH, YRD, and PRD regions by 25 %, 20 %, and 15 %, respectively; control the annual $PM_{2.5}$ in Beijing to below $60\,\mu g\,m^{-3}$
2015	13th Five-Year Plan	Reduce $PM_{2.5}$ concentration by 18 % at the prefecture level and above in cities that failed to meet the $PM_{2.5}$ air quality standard; reduce national SO_2 and NO_x emissions by 15 % and 15 %, respectively; control VOC emissions in key regions and key sectors to reduce national emissions by 10 %

centrations in China from 1990 to 2015. The overall spatial pattern looked similar for all the years. High levels of $PM_{2.5}$ concentrations occurred in northern, central, and eastern China, covering most of the populous city clusters such as the Beijing–Tianjin–Hebei (BTH) region, the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and the Sichuan–Chongqing region. Despite a short-term slowdown caused by the Asian economic crisis in 1997, a pronounced increase in nationwide $PM_{2.5}$ concentrations occurred during 1990–2005, driven by the dramatic growth in SO_2, NO_x, NH_3, and primary $PM_{2.5}$ emissions (Table 2). The population-weighted $PM_{2.5}$ concentration increased from $36.0\,\mu g\,m^{-3}$ in 1990 to $63.5\,\mu g\,m^{-3}$ in 2005. From 2005 to 2015, under the air quality regulations during the 11th and 12th FYPs and the recent Action Plan (Table 1), the national SO_2 and $PM_{2.5}$ emissions were reduced by 49 % and 33 %, respectively (Table 2). During the same period, NO_x emissions rose by 20 % but reached their peak during 2012–2013. As a result, the annual average $PM_{2.5}$ concentrations have decreased, and the population-weighted $PM_{2.5}$ concentration dropped to $49.9\,\mu g\,m^{-3}$ in 2015. During the entire period from 1990 to 2015, the effectiveness of control policies was offset by increased emissions from expanding development; therefore, the annual $PM_{2.5}$ concentrations increased substantially in most of the regions. However, the more aggressive control measures resulted in improved air quality in some metropolitan areas, such as Beijing, the Yangtze River Delta, and the Pearl River Delta.

4.2 Trends in premature mortality attributable to $PM_{2.5}$

As shown in Fig. 2, the national premature mortality attributable to anthropogenic $PM_{2.5}$ exposure estimated with GEMM NCR+LRI functions rose from 1.26 million (95 % confidence interval (CI) [1.05, 1.46]) in 1990 to 2.18 million (95 % CI [1.84, 2.50]) in 2005; then, it slightly decreased to 2.10 million (95 % CI [1.76, 2.42]) in 2015. When adopting the GEMM 5-COD function, the excess deaths rose from 0.94 million (95 % CI [0.58, 1.24]) in 1990 to 1.70 million (95 % CI [1.13, 2.13]) in 2005; then, it decreased to 1.62 million (95 % CI [1.12, 2.01]) in 2015. The mortality impacts of $PM_{2.5}$ exposure based on mortality rates of NCD+LRI were 27 %–33 % higher than those based on mortality rates of 5-COD, indicating that the $PM_{2.5}$ exposure may contribute to mortality from causes other than these five-specific causes of death as we already knew (Burnett et al., 2018). The overall trend of $PM_{2.5}$-related premature mortality reflected the trend of national population-weighted $PM_{2.5}$ concentrations but exhibited a milder decline when the $PM_{2.5}$ concentrations fell after 2005, due to the impetus from other demographic factors (Fig. 3). From 1990 to 2015, premature mortality caused by LRI remained relatively stable, and prema-

Table 2. SO_2, NO_x, $PM_{2.5}$, and NH_3 emissions; population-weighted $PM_{2.5}$ concentration; and $PM_{2.5}$-related premature mortality contributed by the agricultural (Agr), industrial (Ind), power (Pow), residential (Res), and transportation (Tra) sectors.

		Sector	1990	1995	2000	2005	2010	2015
Emissions (Mt (%))		Agr	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
		Ind	5.6 (41)	8.9 (45)	8.9 (43)	13.3 (40)	16.4 (59)	9.8 (58)
	SO_2	Pow	5.2 (38)	7.9 (40)	9.5 (46)	16.7 (51)	7.8 (28)	3.9 (23)
		Res	2.8 (21)	3.0 (15)	2.3 (11)	2.8 (9)	3.4 (12)	2.9 (17)
		Tra	0 (0)	0.1 (0)	0.1 (1)	0.2 (1)	0.2 (1)	0.3 (2)
		Agr	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
		Ind	1.9 (29)	2.6 (28)	2.9 (25)	5.4 (28)	9.1 (35)	9.7 (41)
	NO_x	Pow	2 (31)	3.1 (33)	3.5 (30)	6.7 (34)	8.6 (33)	5.1 (21)
		Res	0.8 (12)	0.8 (8)	0.7 (6)	1.0 (5)	1.0 (4)	0.9 (4)
		Tra	1.7 (27)	3.0 (31)	4.6 (39)	6.5 (33)	7.7 (29)	8.0 (34)
		Agr	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
		Ind	3.4 (39)	6.3 (52)	5.7 (51)	6.8 (50)	6.1 (52)	4.4 (48)
	$PM_{2.5}$	Pow	1.1 (12)	1.4 (12)	1.1 (10)	1.4 (10)	0.8 (7)	0.6 (7)
		Res	4.2 (47)	4.1 (34)	3.8 (34)	4.7 (35)	4.3 (37)	3.6 (40)
		Tra	0.2 (2)	0.3 (2)	0.5 (5)	0.7 (5)	0.5 (5)	0.5 (5)
		Agr	6.7 (93)	8.3 (94)	9.0 (94)	9.3 (93)	9.5 (93)	9.7 (93)
		Ind	0.1 (1)	0.1 (1)	0.2 (2)	0.3 (3)	0.3 (3)	0.4 (4)
	NH_3	Pow	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
		Res	0.4 (5)	0.4 (4)	0.3 (3)	0.4 (4)	0.4 (4)	0.3 (3)
		Tra	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
Population-weighted contribution ($\mu g\,m^{-3}$ (%))	$PM_{2.5}$	Agr	3.4 (10)	5.6 (12)	7.3 (14)	11.6 (18)	12.6 (22)	11.6 (23)
		Ind	10.3 (29)	17.6 (37)	18.3 (36)	21.9 (35)	22.1 (38)	17.5 (35)
		Pow	4.5 (12)	6.3 (13)	6.4 (13)	8.0 (13)	4.6 (8)	3.0 (6)
		Res	16.1 (45)	14.8 (31)	13.9 (28)	16.8 (26)	14.7 (25)	12.6 (25)
		Tra	1.7 (5)	2.9 (6)	4.7 (9)	5.2 (8)	4.6 (8)	5.2 (10)
$PM_{2.5}$-related premature mortality (thousands (%)) [95 %CI])	NCD+LRI	Agr	117.8 (9) [98.5, 136]	189.9 (12) [159.5, 218.4]	258.2 (14) [216.9, 296.6]	396.8 (18) [334.3, 454.7]	459.0 (21) [386.3, 526.7]	484.4 (23) [406.5, 557.4]
		Ind	362.4 (29) [302.6, 418.9]	602.2 (37) [505.3, 693]	649.6 (35) [545.7, 746.6]	731.4 (34) [616.8, 837.5]	797.7 (37) [671.3, 915.3]	734.0 (35) [615.5, 844.9]
		Pow	162.3 (13) [135.4, 187.9]	224.5 (14) [188.0, 258.8]	242.3 (13) [203.1, 279.2]	287.0 (13) [241.4, 329.5]	177.6 (8) [149.1, 204.2]	130.9 (6) [109.6, 150.9]
		Res	559.0 (44) [467.0, 645.9]	517.5 (32) [433.8, 596]	511.4 (28) [429.1, 588.5]	589.0 (27) [495.7, 675.8]	543.4 (25) [456.8, 624.1]	531.9 (25) [445.9, 612.5]
		Tra	60.3 (5) [50.4, 69.7]	100.7 (6) [84.5, 115.9]	169.9 (9) [142.7, 195.3]	176.8 (8) [148.9, 202.6]	168.2 (8) [141.5, 193.1]	218.3 (10) [183.1, 251.2]
	5-COD	Agr	88.8 (9) [54.5, 116.6]	144.2 (12) [89.4, 186.9]	197.8 (14) [125.7, 253.8]	309.3 (18) [206, 388.4]	359.9 (21) [246.9, 447.6]	373.2 (23) [258.8, 464.2]
		Ind	270.9 (29) [165.9, 356.6]	455.0 (37) [281.9, 591.0]	495.7 (35) [315.2, 636.6]	569.2 (34) [379.7, 713.3]	624.9 (37) [428.8, 777.2]	564.8 (35) [391.4, 703.1]
		Pow	120.6 (13) [73.7, 159.4]	168.9 (14) [104.3, 220.6]	184.4 (13) [116.7, 238.3]	223.5 (13) [148.4, 282]	139.0 (8) [95.1, 173.8]	100.5 (6) [69.6, 125.4]
		Res	417.8 (44) [256.1, 549.5]	389.8 (32) [241.2, 507.4]	389.1 (28) [246.9, 501.2]	457.9 (27) [304.5, 576.6]	424.9 (25) [291.2, 529.4]	408.6 (25) [283.2, 508.9]
		Tra	45.2 (5) [27.7, 59.4]	76.1 (6) [47.2, 98.8]	129.6 (9) [82.4, 166.5]	137.6 (8) [91.6, 172.7]	131.8 (8) [90.4, 164.1]	168 (10) [116.5, 209.1]
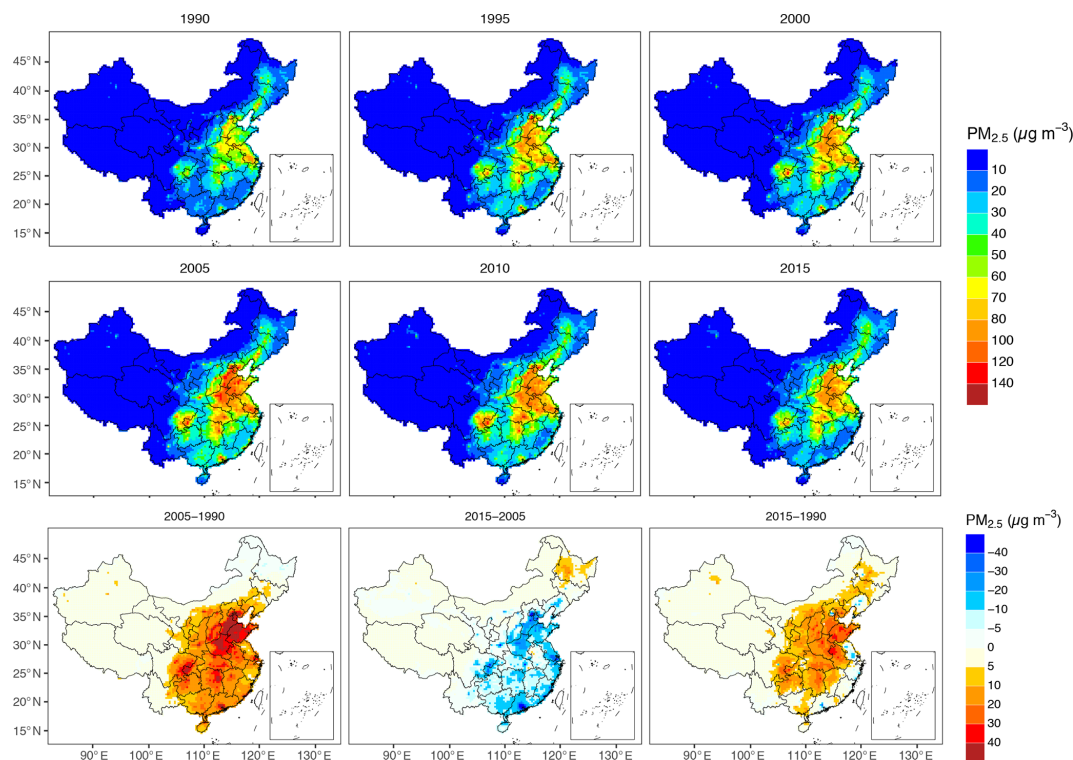
Figure 1. Annual average $PM_{2.5}$ concentrations and concentration changes from 1990 to 2015 ($\mu g\,m^{-3}$).

ture mortality caused by COPD declined, while premature mortality caused by IHD, LC, and stroke increased, reflecting the demographic and epidemiological transitions over time (Yang et al., 2013). The rapid rise of these noncommunicable diseases poses challenges to China's public health.

The trends in premature mortality attributable to $PM_{2.5}$ are driven by changes in air quality, demographic factors, and baseline mortality rates. As shown in Fig. 3, reductions in baseline mortality rates continuously contributed to the decrease in $PM_{2.5}$-related mortality, but these benefits were counterbalanced by increases resulting from population growth and aging. Before 2005, the deteriorating air quality contributed to the growth of $PM_{2.5}$-related mortality, whereas the notable improvements of air quality in the past 10 years have contributed to a decrease in $PM_{2.5}$-related mortality. Overall, however, the net reductions were rather small compared with the total premature mortality (for example, the reduction was only -2% from 2010 to 2015). With the population still growing and the accelerating aging of the population, it has become crucial to take further steps to sharply reduce $PM_{2.5}$ concentrations to effectively improve the related health benefits.

4.3 Source contributions to $PM_{2.5}$ air pollution and related premature mortality

We determined the source contributions from the power, industrial, transportation, residential, and agricultural sec-

tors to $PM_{2.5}$ air pollution and related premature mortality through a series of CMAQ sensitivity simulations. Figure 4 shows the spatial distribution of the $PM_{2.5}$ concentration contributed by each source sector in 1990, 2000, 2010, and 2015, and Fig. 5 shows the relative and absolute source contributions to the national population-weighted $PM_{2.5}$ concentration and related premature mortality from 1990 to 2015. The relative contributions of the source sectors to $PM_{2.5}$ concentrations and related premature mortality are similar (Table 2), and the nonaccidental mortality (NCD+LRI) was found to have enhanced statistical power to characterize the shape of the GEMM functions compared to specific causes of death (Burnett et al., 2018). Therefore, we discuss the source contributions from a health perspective estimated with GEMM NCD+LRI functions in more detail, and the estimates with GEMM 5-COD are also listed in Table 2.

In general, the industrial sector is the prime source of $PM_{2.5}$-related premature mortality. This sector's contribution grew from 29% in 1990 to 37% in 2010, driven by the increasing demand for industrial production, but fell to 35% in 2015 under strengthened emission control measures during the Action Plan. The residential sector was the dominant source in 1990 but has had a decreasing trend over the past 25 years. The relative contribution of the power sector was almost stable from 1990 to 2005 but then began to decrease, owing to active emission control over the past 10 years. In contrast, the transportation and agricultural sec-
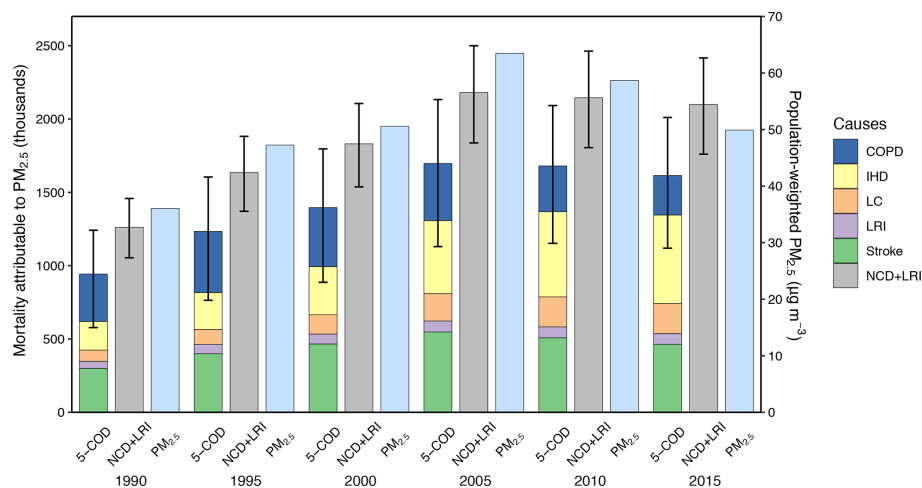
Figure 2. Historical trends of national population-weighted $PM_{2.5}$ concentrations ($\mu g\,m^{-3}$) and $PM_{2.5}$-related premature mortality shared by disease causes (thousands) estimated with GEMM NCD+LRI and GEMM 5-COD functions.
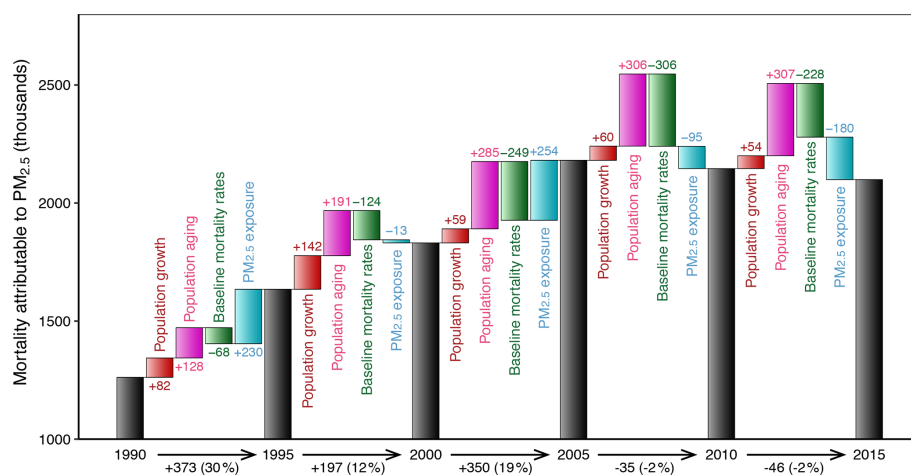
Figure 3. Contribution of factors to the changes in national $PM_{2.5}$-related premature mortality estimated with GEMM NCD+LRI functions for each 5-year interval (in thousands). The length of each bar reflects the contribution of each factor.

tors have experienced increasing trends; their source contributions to $PM_{2.5}$-related mortality increased from 5 % and 9 %, respectively, in 1990 to 10 % and 23 %, respectively, in 2015. These growing contributions indicate the necessity for attention when planning future mitigations.

Historically, the power sector is the largest coal consumer and is considered to be an important emission source in China. Since 1990, driven by the growing demand for electricity, the power sector has prominently increased its emissions and became the leading source of SO_2 and NO_x emissions in 2005 (Table 2). As a result, the population-weighted $PM_{2.5}$ concentration contributed by the power sector increased from 4.5 $\mu g\,m^{-3}$ in 1990 to 8.0 $\mu g\,m^{-3}$ in 2005. Sulfate, nitrate, and ammonium (SNA) were the major chemical components, accounting for approximately 80 % of the total $PM_{2.5}$ mass concentrations (Fig. S5). Correspondingly,

the absolute contribution of the power sector to $PM_{2.5}$-related mortality increased from 162 300 (95 % CI [135 400, 187 900]) in 1990 to 287 000 (95 % CI [241 400, 329 500]) in 2005. However, after 2005, a series of control measures were actively enforced in the power plants to meet the new emission standards (GB13223-2003, GB13223-2011), including installations of FGD systems in the 11th FYP, SCR and SNCR systems in the 12th FYP, and more stringent measures under the Action Plan (Zheng et al., 2018), which have continuously reduced power plant SO_2 and NO_x emissions. Until 2015, the power sector accounted for only 130 900 (95 % CI [109 600, 150 900]) or 6 % of the $PM_{2.5}$-related deaths.

The industrial sector was the largest contributor to $PM_{2.5}$ air pollution and related mortality during 1995–2015, responsible for between 602 200 and 797 700 (34 %–37 %)
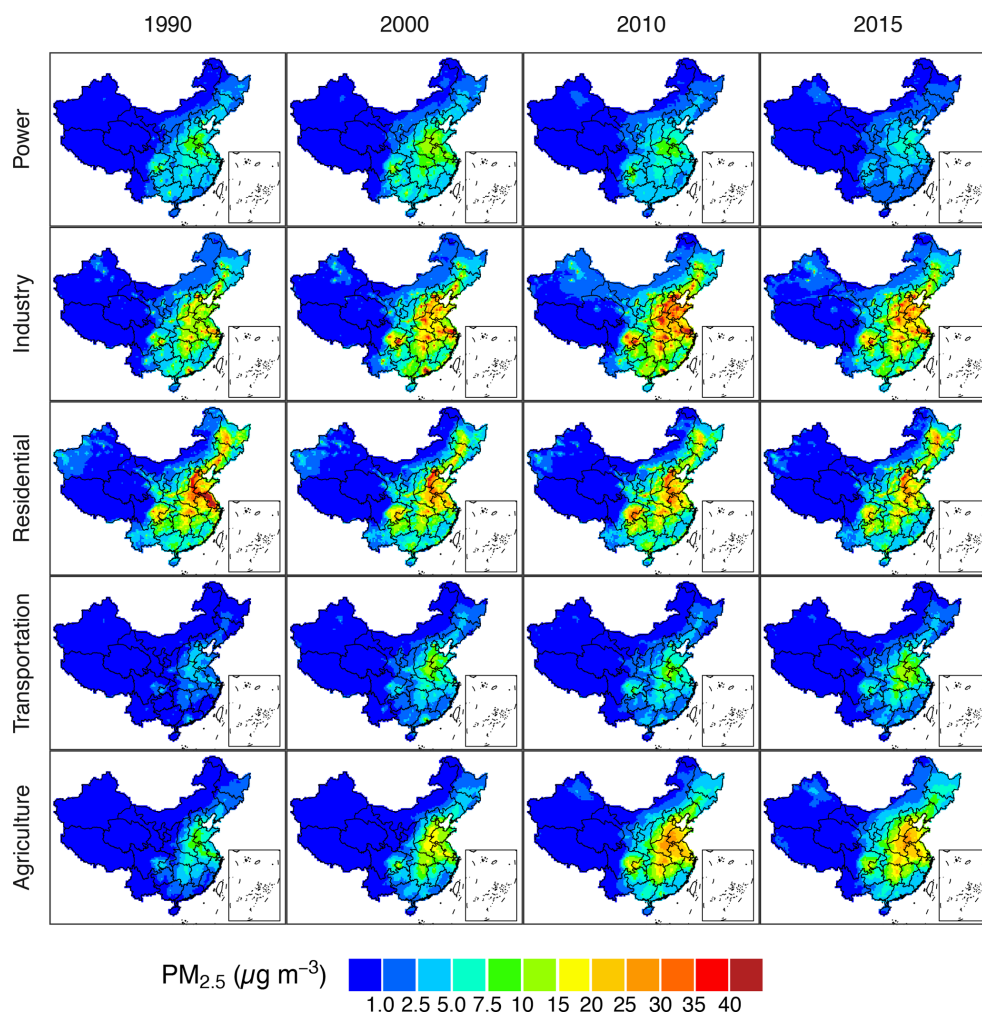
Figure 4. Source contributions to annual average $PM_{2.5}$ concentration ($\mu g\,m^{-3}$) in 1990, 2000, 2010, and 2015.

deaths. Industrial emissions are emitted from both stationary combustion and industrial processes. Cement plants, iron and steel plants, and industrial boilers are the major contributors. Driven by the relentless growth of industrial production, the industrial SO_2 and NO_x emissions increased continuously from 1990 to 2010 (Table 2). But the industrial $PM_{2.5}$ emissions exhibit a different trend: they first increased from 1990 to 1995 and then stabilized during 1995–2010, since the $PM_{2.5}$ emission mitigation from the switch from shaft kilns to precalciner kilns just counterbalanced the emission growth from other industrial sectors (M. Li et al., 2017a). Driven by those emission changes, the population-weighted $PM_{2.5}$ concentration contributed by the industrial sector increased from 10.3 to 22.1 $\mu g\,m^{-3}$ from 1990 to 2010 (Table 2), and the premature mortality contributed by the industrial sector increased prominently from 362 400 (95 % CI [302 600, 418 900]; 29 %) in 1990 to 797 700 (95 % CI [671 300, 915 300]; 37 %) in 2010. SNA and other unspeciated primary $PM_{2.5}$ emissions are the major chemical components of population-weighted $PM_{2.5}$, accounting for 42 %–

58 % and 31 %–48 % of the mass concentration, respectively (Fig. S5). Until recently, the growing trend of industrial contributions has been effectively reversed by a series of enhanced control measures under the Action Plan, including more stringent industrial emission standards; the elimination of outdated industrial capacity; the phasing out of small, polluted factories; and the elimination of small coal-fired industrial boilers (Zheng et al., 2018). In 2015, the industrial NO_x emissions flattened out and SO_2 and $PM_{2.5}$ emissions were remarkably reduced by 40 % and 28 % compared with their levels in 2010 (Table 2), which consequently drove down the premature mortality shared by the industrial sector to 734 000 (95 % CI [615 500, 844 900]; 35 %).

The residential sector is another major emitter of anthropogenic pollutants, including $PM_{2.5}$, BC, OC, and NMVOCs, due to poor combustion efficiency and lack of emission controls (M. Li et al., 2017a). The prime cause of residential emissions is the consumption of fossil fuels and biofuels for heating and cooking. It is a major ambient air pollution source, especially during the winter heating season in

northern China (Li et al., 2015; J. Liu et al., 2016a). On average, the residential sector was the second-largest source of population-weighted $PM_{2.5}$ in China. The major chemical components are organic matter (OM) and BC, which comprise approximately 46 % and 16 %, respectively, of the $PM_{2.5}$ concentration contributed by the residential sector (Fig. S5). In 1990, the residential sector was the leading contributor, accounting for 44 % of the $PM_{2.5}$-related deaths, but it experienced an overall decreasing trend over time. There are several reasons for this decrease. First, China has undergone an accelerating urbanization process. Hundreds of millions of rural people have migrated from the countryside into the cities. The urbanization-induced migration reduced emissions due to the switch from solid fuels to cleaner fuels after migration (Shen et al., 2017). Second, driven by the socioeconomic development, clean-energy transitions from solid fuels to clean fuels such as natural gas and electricity gradually happened in rural households (Chen et al., 2016), resulting in a decrease in residential emissions. Third, from 2013 to 2017, China strove to replace the direct use of coal with electricity and gas-powered heating in millions of households in northern China to mitigate air pollution in the countryside (Zheng et al., 2018; Cheng et al., 2019). Therefore, the contribution of the residential sector gradually declined from 44 % in 1990 to 25 % in 2015 (Fig. 5).

The transportation sector is a growing source and is a major NO_x, NMVOC, and BC emitter. Before 2000, China was in the prestage of the current vehicle emission standard system (Fig. S4), and emission control efficiencies were very limited in the transportation sector. Driven by vehicle population growth, NO_x and $PM_{2.5}$ emissions increased by 167 % and 229 % from 1990 to 2000, and the population-weighted $PM_{2.5}$ contributed by the transportation sector increased from 1.7 to 4.7 $\mu g\,m^{-3}$ (Table 2). Nitrate and BC are the major chemical components, accounting for 55 % and 16 % of the $PM_{2.5}$ mass concentration contributed by the transportation on average (Fig. S5). The $PM_{2.5}$-related mortality caused by transportation emissions increased from 60 300 (95 % CI [50 400, 69 700]; 5 %) in 1990 to 169 900 (95 % CI [142 700, 195 300]; 9 %) in 2000 (Fig. 5). Then between 2000 and 2001, China implemented the national state I emission standards for light-duty gasoline vehicles and heavy-duty diesel vehicles. Subsequently, the emission standards were strengthened to stage II, stage III, and stage IV standards during 2003–2015 (Fig. S4). With these policy measures, vehicle emissions did not increase as rapidly as the number of vehicles, and the NMVOC and $PM_{2.5}$ emissions from this sector have been declining since 2005 (Huo et al., 2015). NO_x emissions have stabilized but are still growing. From 2000 to 2010, the contribution of the transportation sector to $PM_{2.5}$-related mortality was relatively stable; however, by 2015, when pronounced reductions were achieved in the power and industrial sectors, the prominence of the transportation sector increased, with a contribution of 218 300

(95 % CI [183 100, 251 200]) $PM_{2.5}$-related deaths, or 10 % of the total, revealing a growing threat to public health.

The agricultural sector is another growing source with a surge of NH_3 emissions. During 1990–2015, the $PM_{2.5}$-related mortality shared by the agricultural sector increased from 117 800 (95 % CI [98 500, 136 000]; (9 %) to 484 400 (95 % CI [406 500, 557 400]; 23 %; Fig. 5). This increase was driven not only by the rising NH_3 emissions but also by the abruptly growing NO_x emissions. NH_3 is an important precursor of ambient $PM_{2.5}$ that exists in the secondary forms of ammonium sulfate and ammonium nitrate. Over 90 % of NH_3 emissions stem from agricultural activities, including synthetic nitrogen fertilizer applications and livestock manure management. Because of the increasing demand for agricultural products and the lack of effective control measures, agricultural NH_3 emissions increased from 6.7×10^6 t in 1990 to 9.7×10^6 t in 2015. Consequently, the population-weighted $PM_{2.5}$ concentration contributed by the agricultural sector increased from 3.4 to 12.6 $\mu g\,m^{-3}$ during 1990–2010. However, it subsequently decreased to 11.6 $\mu g\,m^{-3}$ in 2015 due to NO_x emission reductions imposed by the Action Plan (Table 2). Ammonium, nitrate, and sulfate were the major chemical components of the agricultural population-weighted $PM_{2.5}$, accounting for over 98 % of the mass concentration (Fig. S5). Sulfate and nitrate are formed through the oxidation and neutralization of precursor gases SO_2, NO_x, and NH_3. $(NH_4)_2SO_4$ is the preferential species due to its stability; the semivolatile NH_4NO_3 is formed when excess NH_3 is available beyond sulfate requirements (Seinfeld et al., 2006). In the sensitivity simulations, when the agricultural NH_3 emissions were removed, most nitrate formation was inhibited due to lack of NH_3. In contrast, sulfate can exist in the form of acid particles. Therefore, the amount of nitrate is much higher than the amount of sulfate in the $PM_{2.5}$ (Fig. S5) contributed by the agriculture sector.

5 Discussion

5.1 Comparison with other studies

We compared our estimates of anthropogenic source contributions to national $PM_{2.5}$-related premature mortality with those of previous studies in China (Table 3). Most of the studies focused on 1 particular year for the period of 2010–2014 (Lelieveld et al., 2015; Hu et al., 2017; Gu et al., 2018; Reddington et al., 2019), and one study investigated the changes in source contributions during 2005–2015 (Zheng et al., 2019), whereas our study has a longer temporal coverage of 25 years, from 1990 to 2015. The differences between studies could arise from the differences in emission inventories and air quality models. The residential sector was highlighted in all the studies, sharing a contribution ranging from 15 % (Zheng et al., 2019) to 42 % (Reddington et al., 2019). The industrial sector was also identified as the leading source
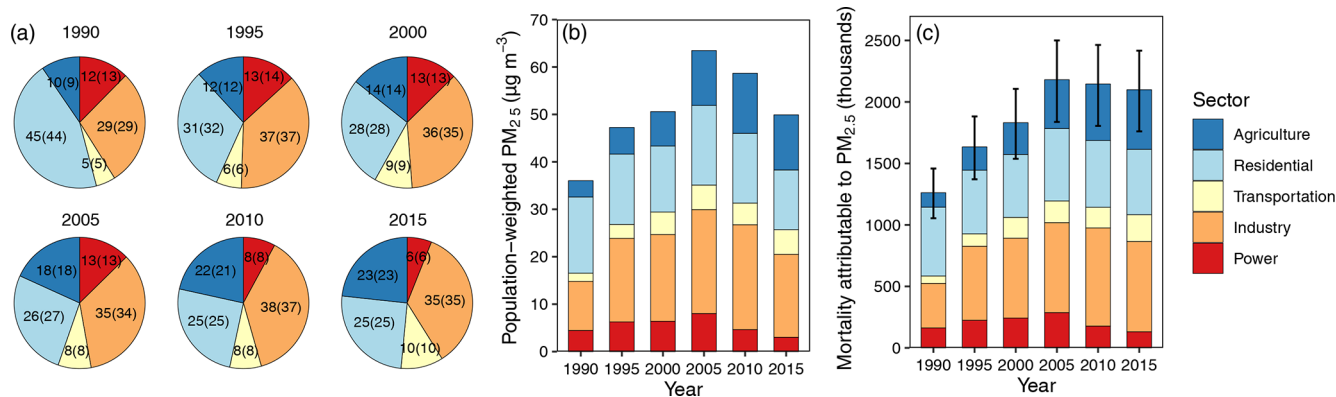
Figure 5. Relative and absolute source contributions to national population-weighted PM$_{2.5}$ concentrations and related premature mortality estimated with GEMM NCD+LRI functions. **(a)** Relative source contributions (%) to national population-weighted PM$_{2.5}$ concentrations (numbers outside brackets) and related premature mortality (numbers inside brackets). **(b)** Absolute source contributions to national population-weighted PM$_{2.5}$ concentrations (μg m^{-3}). **(c)** Absolute source contributions to national PM$_{2.5}$-related premature mortality (thousands) estimated with GEMM NCD+LRI functions.

in five out of the six studies. A decreasing trend in the contribution from the power sector during 2005–2015 was illustrated by all the studies. Large discrepancies occurred in the agricultural sector, with the contribution ranging from 0.1 % (Reddington et al., 2019) to over 30 % (Lelieveld et al., 2015; Zheng et al., 2019). The magnitude of the contribution from the transportation sector was relatively close in all the studies. Overall, our estimates of relative source contributions were generally consistent with previous studies, well within the ranges of their estimates, and closest to those from Hu et al. (2017) and Gu et al. (2018), who estimated the same order of source contributions as our study before 2015, i.e., industry, residential, agriculture, power, and transportation, from the highest to the lowest.

5.2 Uncertainties and limitations

Our study has a number of uncertainties and limitations. First, we estimated the long-term exposure of PM$_{2.5}$ using CMAQ model simulations rather than satellite-based estimates. In practice, the satellite-based estimates utilize information from satellite and observations, resulting in improvements in estimates (Ma et al., 2016; van Donkelaar et al., 2016; Xue et al., 2017). However, satellite remote sensing data were unavailable prior to 1998. If we had combined the CMAQ simulations for 1990–1995 and satellite-based estimates for 2000–2015, we would have introduced additional uncertainties, which may have perturbed the long-term trends of source contributions to PM$_{2.5}$ and weakened the information conveyed from the decadal sectoral emission changes. To illustrate the impacts, we calculated the PM$_{2.5}$-related mortality with both CMAQ simulations and satellite-based estimates of van Donkelaar et al. (2016) and Ma et al. (2016) for 3 overlapping years: 2005, 2010, and 2015. The results showed that the PM$_{2.5}$-related premature mor-
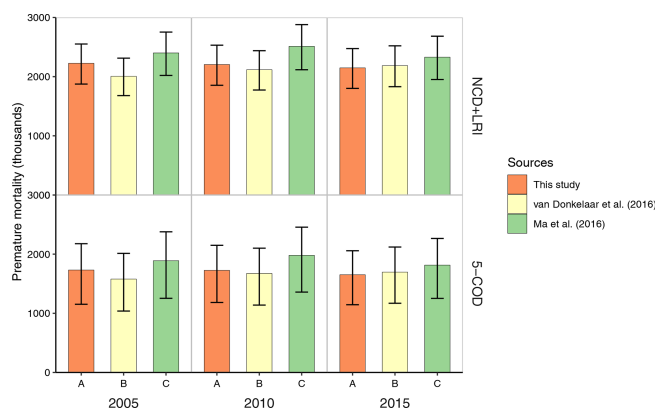
Figure 6. National PM$_{2.5}$-related premature mortality estimated by PM$_{2.5}$ estimates from CMAQ simulations (this study) and satellite-based methods (thousands).

tality estimated with the CMAQ simulations were comparable to those using satellite-based PM$_{2.5}$ (Fig. 6). These differences were acceptable considering the uncertainty ranges from the GEMM functions. Previous studies on long-term trends in the chemical composition of population-weighted PM$_{2.5}$ (C. Li et al., 2017) and on the attribution of source contributions to PM$_{2.5}$-related mortality to emission changes (Lelieveld et al., 2015; Ma et al., 2017; Gu et al., 2018; Zheng et al., 2019) also applied the chemical transport model simulations to estimate exposure.

Second, the uncertainty in this study also stems from PM$_{2.5}$ exposure–response functions. Globally, the excess mortality associated with PM$_{2.5}$ air pollution in 2015 ranged from 4.2 million (Cohen et al., 2017) to 8.9 million (Burnett et al., 2018), depending on the choice of exposure–response functions. Burnett et al. (2018) constructed the up-to-date GEMM functions based only on cohort studies of outdoor

Table 3. Comparison of anthropogenic source contributions to $PM_{2.5}$-related premature mortality with previous studies (%). The source sectors include Power (Pow), Industry (Ind), Residential (Res), Transportation (Tra), and Agriculture (Agr).

Year	Pow	Ind	Res	Tra	Agr	Model	Emissions	References
2010	20	9	36	3	32	Global EMAC	EDGAR	Lelieveld et al. (2015)*
2010	14	36	26	9	16	WRF-CMAQ	HTAP v2	Gu et al. (2018)*
2013	13	38	27	7	15	Source-oriented CMAQ	MEIC	Hu et al. (2017)*
2014	6	48	42	4	0.1	WRF-Chem	EDGAR-HTAP v2	Reddington et al. (2019)*
2005	14	33	19	8	26	WRF-CMAQ	Self-compiled emissions	Zheng et al. (2019)*
2015	7	28	15	15	35	WRF-CMAQ	Self-compiled emissions	Zheng et al. (2019)*
2005	13	34	27	8	18	WRF-CMAQ	MEIC	This study
2010	8	37	25	8	21	WRF-CMAQ	MEIC	This study
2015	6	35	25	10	23	WRF-CMAQ	MEIC	This study

* Relative anthropogenic source contributions are derived from a normalization across all anthropogenic source sectors.

air pollution which cover the global exposure range and estimated 2.47 million (GEMM NCD+LRI) and 1.95 million (GEMM 5-COD) deaths in China due to ambient $PM_{2.5}$ exposure. Our calculation with the GEMM risk functions illustrated 2.10 million and 1.62 million excess deaths related to the anthropogenic $PM_{2.5}$ exposure in 2015 by GEMM NCD+LRI and GEMM 5-COD functions. Our results were rather close to the estimates of Burnett et al. (2018) but prominently higher than the previous estimates of 0.87–1.37 million during 2010–2015 (Lelieveld et al., 2015; J. Liu et al., 2016b; Hu et al., 2017; Zheng et al., 2017, 2019), which applied the integrated exposure–response (IER) functions developed in the Global Burden of Disease (GBD) project (Burnett et al., 2014; Cohen et al., 2017). We conducted a sensitivity analysis by repeating the calculations with the IER risk functions used in GBD 2015 and estimated the anthropogenic $PM_{2.5}$-related mortality in China in 2015 was 0.97 million (Table 4), which was close to the estimate of 0.87 million in 2015 by Zheng et al. (2019). The sensitivity analysis shows that different $PM_{2.5}$ exposure–response functions exhibit large discrepancies in the estimates of $PM_{2.5}$-related mortality, which calls for more additional cohort studies in China to improve the health risk models and narrow the gaps between different studies. However, since our study focuses more on the relative fraction of source contributions to the population-weighted $PM_{2.5}$ and $PM_{2.5}$-related mortality, the results and conclusions do not change no matter which version of exposure–response functions we choose.

Third, uncertainties are also introduced by the meteorological conditions. In the study, we applied the NCEP-FNL meteorological data in 2000 to drive the WRF model for the years 1990 and 1995. Initially, we intended to combine the NCEP Climate Forecast System Reanalysis (NCEP-CFSR) for 1990–1995 and NCEP-FNL for 2000–2015, since the NCEP-CFSR was available before 2011 and the NCEP-FNL was available after 1999. However, when we ran the WRF-CMAQ model for the overlapping year 2000 with both products, we found the differences in the chemical compo-

sition of the population-weighted $PM_{2.5}$ could be as high as $1.0\,\mu g\,m^{-3}$, which may introduce extra bias into the trend of relative source contributions. Therefore, we applied the same meteorological product to drive the WRF model. To quantify the influence from the interannual variations in meteorological conditions, we carried out three sensitivity simulations with the same meteorology in year 2000 and the year-specific emissions in 2005, 2010, and 2015. The differences between the base and the sensitivity simulation denote the impacts of meteorology. It shows that the impacts of the interannual variations in meteorological conditions differ with regions and leads to an overall change in the population-weighted $PM_{2.5}$ concentration below 5 %. The interannual changes in $PM_{2.5}$ concentrations were dominated by the changes in emissions.

Fourth, uncertainties also arise from the uncertainties in the demographic factors. To present the long-term trends in $PM_{2.5}$-related premature mortality, we utilized the year-specific national population age structure and the cause-specific mortality shared in GBD 2016 (Global Burden of Disease Collaborative Network, 2017a, b). However, there are substantial provincial heterogeneities in age structures and mortality due to differences in social economics, geographic distributions, and health services that were not studied in this work. The existing data from statistics and other sources are not sufficient to provide such details. Besides, the variations in different versions of GBD cause-specific mortality data may also introduce uncertainties into the short-term contributions of the demographic factors in the decomposition analysis. For example, it was estimated that the health benefits contributed by the decrease in baseline mortality rates ranged from 11 000 (Ding et al., 2019) to 32 000 (Yue et al., 2020) from national air pollution control actions for the period of 2013 to 2017, with the same exposure–response function utilized for the same study period. However, since our study mainly focuses on the trends of $PM_{2.5}$ exposure and the transitions in relative source contributions,

Table 4. PM$_{2.5}$-related premature mortality contributed by the agricultural (Agr), industrial (Ind), power (Pow), residential (Res) and transportation (Tra) sectors. Estimated with IER risk function [unit: thousand (%)].

Model	Sector	1990	1995	2000	2005	2010	2015
	Agr	66.6 (9)	99.6 (12)	127.8 (14)	186.1 (18)	209.7 (21)	217.1 (23)
	Ind	206.7 (29)	318.1 (37)	323.2 (35)	341.9 (33)	365.2 (37)	330.8 (35)
IER	Pow	93.6 (13)	120.2 (14)	122.8 (13)	136.7 (13)	82.8 (8)	59.7 (6)
	Res	317.6 (44)	274.7 (32)	256.5 (28)	279.3 (27)	250.9 (25)	240.4 (25)
	Tra	34.3 (5)	53.1 (6)	84.5 (9)	83.0 (8)	77.2 (8)	98.2 (10)

the adoption of different versions of GBD baseline mortality data will not change the major results and conclusions.

Finally, we differentiated the contributions from various source categories with a series of zero-out sensitivity simulations. To address the mass nonconservation resulting from the zero-out method, we normalized the sensitivity simulations to match the total PM$_{2.5}$ concentrations in the standard simulation, which could introduce uncertainties. In fact, the zero-out method estimates the upper value for the contribution from gaseous precursors to secondary PM$_{2.5}$, especially for the agricultural sector, where NH$_3$ dominates most of the emissions. The agricultural contribution estimated in this study includes the nonagricultural nitrate. According to our estimation, the agricultural sector accounted for 21 %–23 % of the total premature mortality during 2010–2015, which is higher than the estimation (15 %) using the source-oriented CMAQ model (Hu et al., 2017) but lower than the estimation (32 %–35 %) that used the zero-out method in the global chemical transport model (Lelieveld et al., 2015) and the CMAQ model (Zheng et al., 2019; Table 3).

5.3 Policy implications

Ambient PM$_{2.5}$ pollution contributed substantially to premature mortality in China. To mitigate air pollution problems, the government launched air quality regulations decades ago. The transition of source contributions calls for adjustments to how policy focuses on emission sectors.

Pronounced emission abatements have been achieved in the power sector. Recently, the penetration rates of FGD and SCR–SNCR systems have reached over 95 %, and in 2017 approximately 71 % of current power plants operated close to the level of "ultralow emission" (Zheng et al., 2018). Thus, the potential to reduce future emissions is limited in the power sector. Currently, industry is the largest contributor. To fulfill the air quality targets, a series of measures have been implemented in the industrial sector, including phasing out outdated capacity; strengthening emission standards; and eliminating small, high-polluting factories (Zheng et al., 2018). However, the industry sector still accounted for 35 % of the anthropogenic PM$_{2.5}$-related premature mortality in 2015. Further reductions by the industrial sector are essential to achieving the air quality targets in the 13th FYP. The residential sector was once the largest contributor to

PM$_{2.5}$-related premature mortality, but its contribution has decreased over time. This decreasing trend was accelerated when millions of households switched from direct coal use to electricity and gas-fired heating during 2013–2017. The wider promotion of clean-fuel use in suburban and rural households promises to further reduce emissions from the residential sector.

Although emissions from the power, industrial, and residential sectors are decreasing, contributions from the transportation and agricultural sectors have gradually increased. Emissions from the transportation sector have stabilized in recent years because strengthened policy measures have counterbalanced the growing vehicle population. If there were no emission standards during 2000–2012, the vehicle emission levels in China would have increased by 2–6 times the levels measured in 2000 (Huo et al., 2015). Given that vehicle ownership per 1000 people in China is still much lower than that in developed countries, implementing more stringent standards and accelerating the phaseout of older high-emission vehicles are important to further reduce vehicle emissions. In fact, China is planning to implement the latest national VI emission standards for light-duty vehicles from 2020 and for diesel-fueled heavy-duty vehicles from 2021, which makes even larger emission reductions in the near future likely. Currently, the agricultural sector lacks mitigation policies, and growing NH$_3$ emissions are predicted to grow in the future. The NH$_3$ emissions growth will offset the air quality benefits from SO$_2$ and NO$_x$ reductions (Wang et al., 2013) and pose challenges for future air quality management. Technologies to improve manure management and reduce ammonia applications in fields and optimizing human diets are potential strategies to mitigate ammonia emissions from the agricultural sector (Zhao et al., 2017). Thus, the emphasis of future mitigations should also be put on controlling vehicle and agricultural emissions.

6 Conclusions

Ambient PM$_{2.5}$ pollution contributed substantially to premature mortality in China. In this study, we investigated the decadal changes in the anthropogenic source contribution of PM$_{2.5}$ air pollution and related health impacts in China. Accompanied with the development of the economy and the

implementation of environmental policies, the emissions of major air pollutants have changed dramatically during the past 25 years. The SO_2, NO_x, $PM_{2.5}$, and NH_3 emissions increased from 13.6, 6.4, 8.9, and 7.2 Mt in 1990 to 16.9, 23.7, 9.1, and 10.5 Mt in 2015, with peaks in 2005, 2010, 2005, and 2015, respectively. In response, the population-weighted $PM_{2.5}$ concentration increased from 36.0 μg m^{-3} in 1990 to 63.5 μg m^{-3} in 2005 and then gradually decreased to 49.9 μg m^{-3} in 2015. We estimated that the total premature mortality attributable to anthropogenic $PM_{2.5}$ with GEMM NCD+LRI functions rose from 1.26 million (95 % CI [1.05, 1.46]) in 1990 to 2.18 million (95 % CI [1.84, 2.50]) in 2005 and fell to 2.10 million (95 % CI [1.76, 2.42]) in 2015. Besides the influence of the changes in air quality, the reductions in baseline mortality rates contributed to the decrease in $PM_{2.5}$-related mortality, while the population growth and aging contributed to the increases in $PM_{2.5}$-related mortality.

Investigating the decadal transition in source contribution of $PM_{2.5}$ air pollution and related premature mortality can help to evaluate the effectiveness of past efforts and provide important insights for prioritizing strategies in the future. We found that the contributions of various sectors to anthropogenic $PM_{2.5}$-related premature mortality changed substantially during 1990–2015. In 1990, the residential sector was the leading source of $PM_{2.5}$-related mortality (44 % of total) in China, followed by industry (29 %), power (13 %), agriculture (9 %), and transportation (5 %). Whereas in 2015, the industrial sector became the largest contributor (35 %), followed by the residential sector (25 %), agriculture (23 %), transportation (10 %), and power (6 %). Limited potential to further reduce emissions remains in the power sector after effective controls since 2005. The industrial and residential sectors are still the leading contributors to $PM_{2.5}$-related premature mortality, despite their declining trends. The importance of the transportation and agricultural sectors are also highlighted with their continuously increasing contributions. Emphasis should be directed onto the last four sectors when planning future mitigations.

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