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Impacts of coal burning on ambient $PM_{2.5}$ pollution in China

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Abstract. High concentration of fine particles ($PM_{2.5}$), the primary concern about air quality in China, is believed to closely relate to China's large consumption of coal. In order to quantitatively identify the contributions of coal combustion in different sectors to ambient $PM_{2.5}$, we developed an emission inventory for the year 2013 using up-to-date information on energy consumption and emission controls, and we conducted standard and sensitivity simulations using the chemical transport model GEOS-Chem. According to the simulation, coal combustion contributes 22 $\mu g\,m^{-3}$ (40 %) to the total $PM_{2.5}$ concentration at national level (averaged in 74 major cities) and up to 37 $\mu g\,m^{-3}$ (50 %) in the Sichuan Basin. Among major coal-burning sectors, industrial coal burning is the dominant contributor, with a national average contribution of 10 $\mu g\,m^{-3}$ (17 %), followed by coal combustion in power plants and the domestic sector. The national average contribution due to coal combustion is estimated to be 18 $\mu g\,m^{-3}$ (46 %) in summer and 28 $\mu g\,m^{-3}$ (35 %) in winter. While the contribution of domestic coal burning shows an obvious reduction from winter to summer, contributions of coal combustion in power plants and the industrial sector remain at relatively constant levels throughout the year.

1 Introduction

$PM_{2.5}$ (particulate matter with aerodynamic diameter less than or equal to 2.5 µm) was considered as the leading air pollutant in most key regions and cities in China, especially in the Beijing–Tianjin–Hebei (BTH) region and the Yangtze River Delta (YRD), according to the air quality status reports released by China's Ministry of Environmental Protection (MEP, 2014a, 2015). The annual mean $PM_{2.5}$ concentration in the BTH region was 102 $\mu g\,m^{-3}$ in 2013 and 93 $\mu g\,m^{-3}$ in 2014, while that in the YRD was 67 $\mu g\,m^{-3}$ in 2013 and 60 $\mu g\,m^{-3}$ in 2014 (MEP, 2014a, 2015), far beyond the World Health Organization (WHO) interim target-1 (35 $\mu g\,m^{-3}$) for annual mean $PM_{2.5}$ concentration and also the secondary class standard in China's new National Ambient Air Quality Standard (NAAQS, GB 3095-2012).

The high ambient $PM_{2.5}$ concentration is believed to closely relate to China's large primary energy consumption, especially coal consumption. According to the statistical review of world energy from BP P.L.C. (BP, 2015), China has become the largest energy consumer since 2009, and coal accounted for two-thirds of the total primary energy consumption. In the year 2010, coal was responsible for 81 % of the SO_2 emissions, 61 % of the NO_x emissions, 40 % of the primary PM_{10} emissions, and 34 % of the primary $PM_{2.5}$ emis-

sions in China (S. X. Wang et al., 2014b). As the most abundant and relatively cheap energy resource, coal is expected to be a dominant energy supply in China in the foreseeable future.

A number of studies have used atmospheric models to study the source contributions of ambient air pollution in China. Early studies (Wang et al., 2005; Hao et al., 2007) mainly focused on gaseous pollutants, including SO_2, NO_x, CO, and O_3. Later on, more studies (Bi et al., 2007; Cheng et al., 2007; Chen et al., 2007; Hao et al., 2007; Wang et al., 2008; Wu et al., 2009) placed emphasis on particulate matter, but mainly on PM_{10}. Recently, due to the frequent haze episodes characterized by extremely high $PM_{2.5}$ concentration in China, researchers are paying more and more attention to $PM_{2.5}$. Among these studies, most of them took advantage of 3-D chemical transport models like the Community Multi-scale Air Quality Model (CMAQ). H. Zhang et al. (2012) studied source contributions to sulfate and nitrate in $PM_{2.5}$ using the CMAQ model and reported that while the power sector is the largest contributor to inorganic components, the industry and traffic sector are also important sources. Some recent studies agreed that industrial and domestic sources were the most significant contributors to ambient $PM_{2.5}$ in most areas in China. L. T. Wang et al. (2014) studied a severe $PM_{2.5}$ pollution episode in January 2013 in North China using the CMAQ model and concluded that industrial and domestic sources, respectively, contributed 28 and 27 % to local $PM_{2.5}$ concentration in Hebei Province. D. Wang et al. (2014) conducted simulations with the same model and studied the same pollution episode but the city of Xi'an in northwestern China, also reporting that industrial and domestic activities are the two largest sources that account for 58 and 16 % of local $PM_{2.5}$ concentration, respectively. L. Zhang et al. (2015) used the GEOS-Chem model and indicated that residential and industrial sources in North China were responsible for 49.8 and 26.5 %, respectively, of the $PM_{2.5}$ concentration in Beijing. While most of the studies focused on developed metropolises or heavy pollution episodes, very few studies used atmospheric chemical transport models to study source contributions and their seasonal variation for the whole country throughout a year. In addition, while most researchers studied the total energy consumption in each sector or regarded coal combustion in all sectors as a whole, none of them distinguished coal burning in one sector from another. However, the utilization of coal and the end-of-pipe emission control policies are quite different in different sectors, which leads to different energy efficiency and thus different emissions. Therefore, contributions from coal burning in specific sectors should be identified individually, which is important for policy making.

In this study, we updated a previously developed emission inventory to the year 2013 using up-to-date information, and we conducted sensitivity simulations with the chemical transport model GEOS-Chem. In order to obtain a comprehensive understanding of the current contribution from coal combustion to $PM_{2.5}$ concentrations in China, we quantitatively identified source contributions from coal burning and their seasonal variations in each sector. Section 2 discusses the development of the emission inventory for the year 2013; Sect. 3 describes the method of simulation, GEOS-Chem model, and its evaluation; Sect. 4 discusses the model results; and the last section summarizes the conclusions.

2 Emission inventory

Our previous studies have developed the emission inventory of sulfur dioxide (SO_2), nitrogen oxide (NO_X), PM_{10}, $PM_{2.5}$, black carbon (BC), organic carbon (OC), nonmethane volatile organic compounds (NMVOCs), and ammonia (NH_3) for China for the year 2010 using a technology-based emission factor method (S. X. Wang et al., 2014b; Zhao et al., 2013a, b, c). The emissions from each sector in each province were calculated from the activity data (energy consumption, industrial products, solvent use, etc.), technology-based emission factors, and penetrations of control technologies. In this study, we updated the 2010 emission inventory to the year 2013 by incorporating the most recent information. The activity data and technology distribution for each sector were updated to 2013 according to the National Bureau of Statistics of China (NBS, 2014a, b, c) and a wide variety of technology reports (Fu et al., 2015; S. X. Wang et al., 2014b; CEC, 2011; ERI, 2010, 2009; THUBERC, 2009). The emission factors used in this inventory were described in Zhao et al. (2013b). The penetrations of removal technologies were updated to 2013 according to governmental bulletins and the evolution of emission standards (MEP, 2014b).

There are some significant updates for NH_3 emissions in this inventory. For agricultural fertilizer application, the emissions of NH_3 in the previous study were based on predefined emission factors that lacked temporal or spatial details. In this inventory, we use an agricultural fertilizer modeling system that couples the regional air quality model CMAQ and an agroecosystem model (the Environmental Policy Integrated Climate model, EPIC) to improve the accuracy of spatial and temporal distribution (Fu et al., 2015). For livestock, the activity data were calculated by the amount of livestock slaughter per year in previous studies. However, the survival periods for livestock are different and not only 1 year; thus, the amount of slaughter cannot accurately stand for the amount of livestock. In this study, we use the amount of livestock stocks to calculate NH_3 emissions and improve the accuracy of the results.

In 2013, the anthropogenic emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC, and NH_3 in China were estimated to be 23.2, 25.6, 16.5, 12.2, 1.96, 3.42, 23.3, and 9.62 Mt, respectively. Table 1 shows emissions by sector and emissions originating from coal combustion, which indicates that in sectors of power plants and domestic fossil fuel combustion, the share of coal-burning emissions is almost over 90 %.

Table 1. Emissions by sector in 2013 in China (unit: kiloton).

	SO_2	NO_x	PM_{10}	$PM_{2.5}$	BC	OC	NMVOC	NH_3
Power plants	6275.4	6463.6	1034.2	612.1	8.1	14.9		
Coal[a]	6209.2	6091.2	1000.9	579.1	3.6	0.0		
Industrial combustion	7226.5	4399.8	1536.0	1030.1	142.8	41.2	133.5	
Coal[b]	5972.2	2969.4	1233.9	805.6	108.4	21.4	63.7	
Other industrial process	2061.1	2492.7	3173.2	1982.3	561.2	429.3	6297.4	215.0
Coal[c]	718.8	1758.9	1521.8	782.8	220.2	179.7	1188.8	
Cement	1704.0	2884.8	2985.1	1866.7	11.3	33.9		
Coal[d]	1270.8	2151.4	1224.4	843.1	8.4	25.3		
Steel	1859.8	532.6	1388.3	1024.2	37.7	48.2		
Coal[e]	1325.1	379.5	463.0	400.4	26.9	34.4		
Domestic fossil fuel combustion	2887.3	609.6	1320.9	974.4	448.1	348.5	4265.6	918.6
Coal	2692.6	554.0	1220.4	893.0	413.4	317.4	848.0	
Domestic biofuel combustion	72.4	477.9	2970.8	2878.0	503.7	1582.9		
On-road transportation	644.0	5138.2	121.2	114.8	52.4	33.5	2044.2	
Off-road transportation	329.5	2111.6	243.6	230.8	131.5	41.5	868.8	
Solvent use							8155.3	
Biomass open burning	90.2	527.1	1747.9	1441.6	57.7	576.6	1213.8	
Waste disposal							387.4	
Livestock farming								5489.8
Mineral fertilizer application								2997.9
National total emissions	23 150.2	25 638.0	16 521.2	12 155.1	1955.1	3423.1	23 366	9621.3
Emissions from coal combustion	18 188.7	13 904.4	6664.4	4304.0	780.9	578.2	2100.4	

[a] Coal here refers to emissions from coal in the corresponding sector in the row above.
[b, c, d, e] In this study industrial coal combustion includes emissions from these four sectors.

Table 2. Summary for simulation scenarios.

		Scenarios	Description	Meteorology
Standard scenario		STD	Standard emission for the year 2013	2012
Sensitivity scenarios	1	TC	Emissions from total coal burning removed	2012
	2	TCP	Emissions from coal burning in power plants removed	2012
	3	TCI	Emissions from coal burning in industry removed	2012
	4	TCD	Emissions from domestic coal burning removed	2012

Coal dominates the emissions in the industrial sector as well. In the year 2013, coal was responsible for 79 % of the SO_2 emissions, 54 % of the NO_x emissions, 40 % of the primary PM_{10} emissions, 35 % of the primary $PM_{2.5}$ emissions, 40 % of the BC emissions, and 17 % of the OC emissions.

3 Model and simulation

3.1 Simulation method

In this study, we conducted one standard simulation and four sensitivity simulations for ground-level $PM_{2.5}$ using the nested grid capability of GEOS-Chem for eastern Asia. The simulation scenarios are summarized in Table 2. In the standard simulation, we use the emissions for the year 2013 that are discussed in Sect. 2. To select the year of meteorology, we conducted standard simulation using the same emissions and different meteorology from the years 2010 to 2012 because the meteorological fields are not available for the whole year of 2013. We chose the year 2012 as our meteorological year, with which the simulation results best represented the mean $PM_{2.5}$ concentration from 2010 to 2012.

In sensitivity scenarios, we removed emissions from coal combustion in different sectors. In sensitivity scenario 1, we removed emissions from coal burning from all energy sectors (scenario for total coal burning, TC). In sensitivity scenarios 2 to 4, we respectively shut down emissions from total coal burning in power plants, industries, and domestic sectors (TCP, TCI, and TCD). All the meteorology used in the

sensitivity simulation was the same as the standard simulation. Used as spin-up were the 3 months before each simulation year. The differences between standard and sensitivity simulations are used to represent the contributions from coal combustion in each sector.

3.2 Model description

GEOS-Chem is a global chemical transport model that has been widely applied to study $PM_{2.5}$ over China (e.g., Brauer et al., 2012, 2015; Jiang et al., 2015; Kharol et al., 2013; van Donkelaar et al., 2010, 2015; Wang et al., 2013; Y. Wang et., 2014; Xu et al., 2015; L. Zhang et al., 2015; Q. Q. Zhang et al., 2015). The model is driven by assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS), including winds, temperature, clouds, precipitation, and other surface properties. GEOS-Chem (version 9-01-03) includes detailed HO_X–NO_x–VOC–ozone–BrO_X tropospheric chemistry originally described by Bey et al. (2001), with the addition of BrO_X chemistry by Parrella et al. (2012). Aerosol simulation is fully coupled with gas-phase chemistry, including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+) (Park et al., 2004; Pye et al., 2009), OC, BC (Park et al., 2003), sea salt (Alexander et al., 2005), and mineral dust (Fairlie et al., 2007). The aerosol thermodynamic equilibriums use the ISORROPIA II model (Fountoukis and Nenes, 2007) to calculate the partitioning of nitric acid and ammonia between gas and aerosol phases. The formation of secondary organic aerosol (SOA) includes the oxidation of isoprene (Henze and Seinfeld, 2006), monoterpenes, aromatics (Henze et al., 2008), and other reactive VOCs (Liao et al., 2007). In addition, we corrected errors in the model representation of too-shallow nighttime mixing depth following Walker et al. (2012) and introduced the production mechanism of sulfate on aerosol surface described in Wang et al. (2013). Aerosols interact with gas-phase chemistry in GEOS-Chem through the effect of aerosol extinction on photolysis rates (Martin et al., 2003) and heterogeneous chemistry (Jacob, 2000).

In this study, we conducted simulations for ground-level $PM_{2.5}$ using the nested grid capability of GEOS-Chem for eastern Asia, which was originally described by Wang et al. (2004) and Chen et al. (2009). The nested domain for eastern Asia covers an area spanning from 70° to 150° E and from 11° S to 55° N, with a horizontal resolution of 0.5° latitude by 0.667° longitude. The boundary fields are provided by the global GEOS-Chem simulation, with a resolution of four latitudes by five longitudes, and are updated every 3 h. We assume that the organic mass / organic carbon ratio is 1.8 and relative humidity is 50 % for $PM_{2.5}$ in China.

The global simulations use emissions from the Global Emission Inventory Activity (GEIA) (Benkovitz et al., 1996), which is overwritten by the NEI05, EMEP, and INTEX-B inventories (Zhang et al., 2009) over the US, Europe, and eastern Asia, respectively. The CO emission we used in this study is from EDGAR v3, which is also overwritten by INTEX-B in the nested domain of eastern Asia. In the nested-grid simulation for eastern Asia, we use the emissions for the year 2013 (as discussed in Sect. 2) over China, with emissions over the rest of eastern Asia taken from the INTEX-B emission inventory. In addition, the simulation also includes open fire emissions from the GFED3 inventory (Giglio et al., 2010; van der Werf et al., 2010; Mu et al., 2011), lightning NO_x emissions calculated with the algorithm of Price and Rind (1992), and volcanic SO_2 emissions from the AEROCOM database (http://aerocom.met.no/download/emissions/AEROCOM_HC/volc/) implemented by Fisher et al. (2011).

3.3 Model evaluation

The GEOS-Chem model is driven by assimilated meteorological data from the NASA GEOS. Y. Wang et al. (2014) evaluated the important meteorological factors that are relevant to particle formation in the model, including temperature, relative humidity (RH), wind speed, and direction, using observation data from the National Meteorological Center (NMC) of China. It reported good spatial and temporal correlations with observed temperature, RH, and wind direction. The correlation of wind speed, however, was poorer as the model tends to overestimate in low speed conditions.

In this study, we conducted model evaluation using the surface $PM_{2.5}$ observation network of the China National Environmental Monitoring Center (CNEMC, http://106.37.208.233:20035). This monitoring program was initiated in January 2013, covering 74 major cities in China. Figure 1 compares simulated annual mean $PM_{2.5}$ concentrations with those observed in 74 major cities in China for the year 2013. As shown in Fig. 1a, the simulated ambient $PM_{2.5}$ concentration has a clear regional distribution with high values in the Sichuan Basin (SCB), North China Plain (NC), and middle Yangtze River area (MYR). The highest concentration occurs in the Sichuan Basin with an average value of 73.5 $\mu g\,m^{-3}$. Concentrations in the abovementioned severely polluted regions are generally above 60 $\mu g\,m^{-3}$. The observation data are compared with the concentrations in the grids where the city centers are located. The comparison shows that the model reproduces the spatial distribution well with a normalized mean bias (NMB) of −16.3 %. The correlation coefficient for annual mean concentration is 0.68. The slight underestimate mainly appears in the heavily polluted area in the NC region where observations are largely influenced by local emissions; however, current simulation cannot capture it due to relatively coarse resolution (H. Zhang et al., 2012). Figure 2 shows comparisons between simulated and observed seasonal mean concentrations. $PM_{2.5}$ concentration has an obvious seasonal variation, with the highest value in winter and the lowest in summer, which is correctly reproduced by the model. The largest bias occurred in winter with the value of −23.3 %. The inconsistency of meteorology also partly

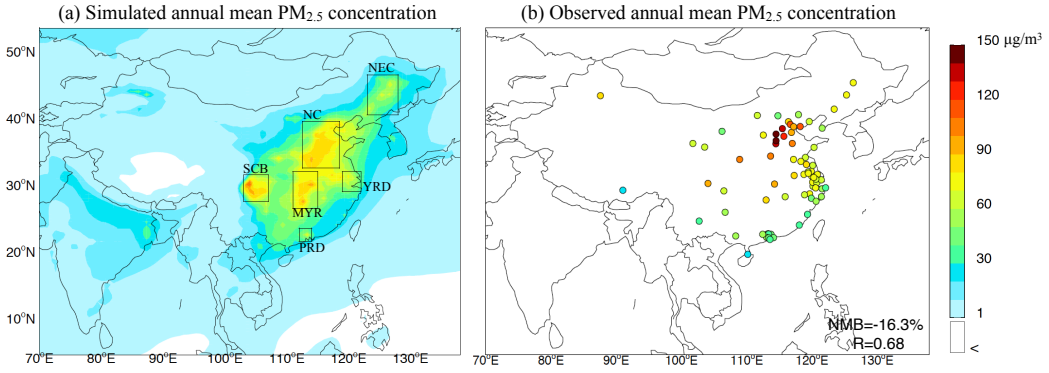

Figure 1. Simulated and observed annual mean $PM_{2.5}$ concentration in China. The six key regions include the Northeast China (NEC), North China (NC), Yangzte River Delta (YRD), Sichuan Basin (SCB), Middle Yangzte River (MYR), and Pearl River Delta (PRD).

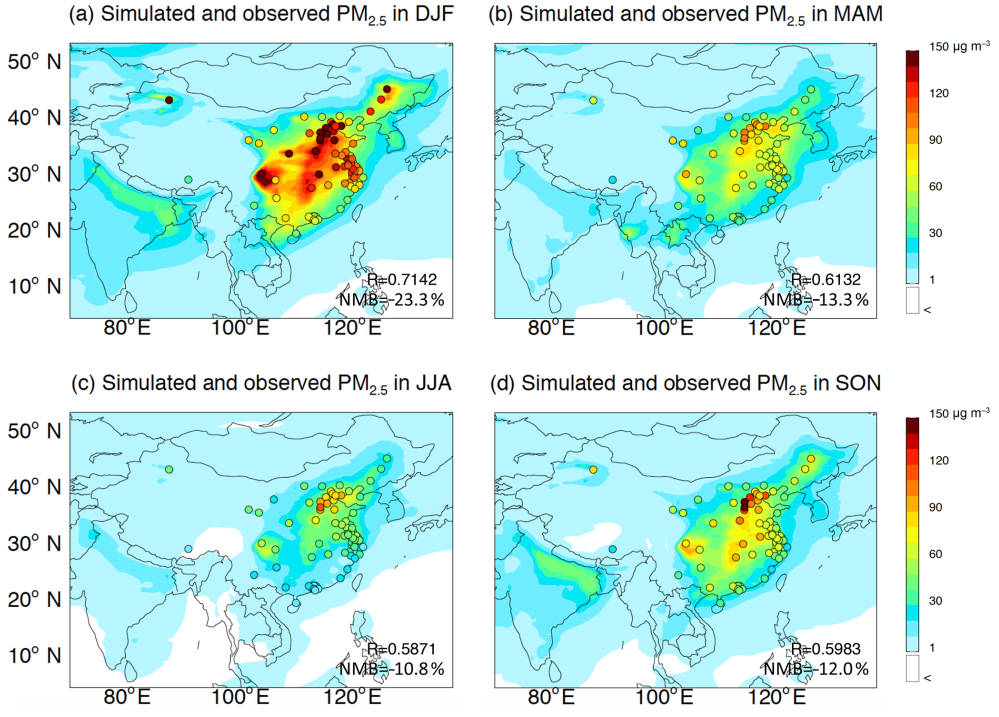

Figure 2. Simulated and observed seasonal $PM_{2.5}$ concentration in China.

accounts for the underestimate because the meteorological condition was more unfavorable in January 2013. Y. Wang et al. (2014) conducted simulations for January in 2012 and 2013 using the same emissions and found that the ground $PM_{2.5}$ concentration was 27 % higher in January 2013 than that in 2012. The model performs better in the other three seasons, with biases between −13.3 and −10.8 %. Correlation maps for each season are shown in Fig. 3. The $PM_{2.5}$ concentration in winter is more spread out in coordinates as it varies substantially across China, which has a larger correlation coefficient of 0.71. In other seasons, the correlation coefficients are around 0.6.

We also evaluated the monthly variation using averaged monthly mean concentrations in cities in each key region since analyses and discussions mainly focused on these six areas. The six key regions are shown with frames in Fig. 1a, which includes Northeast China (NEC; 123–128° E, 41–47° N), North China (NC; 113–119° E, 33–40° N), Yangtze River Delta (YRD; 119–122° E, 29.5–32.5° N), Middle Yangtze River (MYR; 111–115° E, 27–32.5° N), Sichuan Basin (SCB; 103–107° E, 28–32° N), and Pearl River Delta (PRD; 112–114° E, 22–24° N). Cities in each region share the similar weather conditions, terrain, and pollution levels. As shown in Fig. 4, the model generally reproduces the monthly variation well. The NMB ranges from −45 to 1 %,

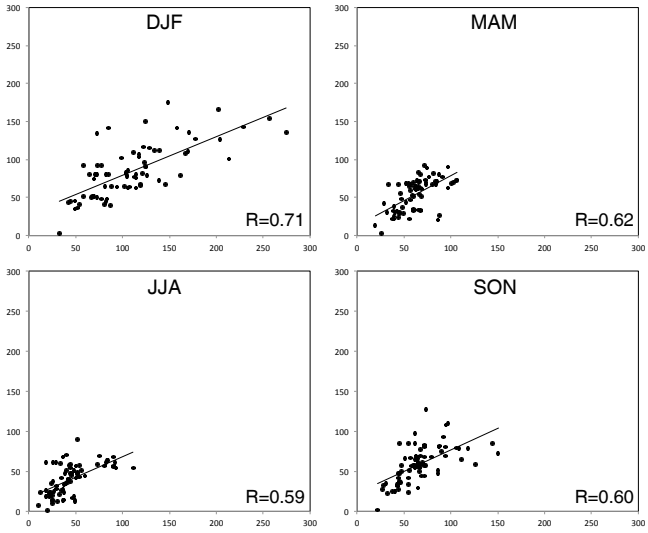

Figure 3. Correlation maps for each season.

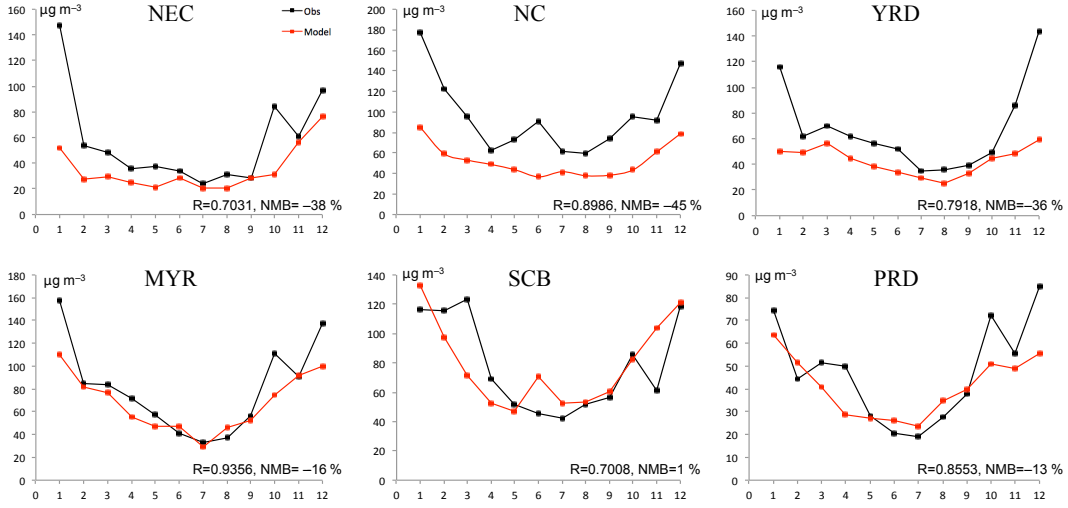

Figure 4. Monthly mean simulated and observed $PM_{2.5}$ in six key regions.

and the correlation coefficient varies between 0.7 and 0.94. The model performance is better in the MYR, SCB, and PRD than in NC, NEC, and the YRD. The large discrepancy is mainly due to the failure to capture the extremely high concentration in wintertime. The normalized mean errors (NMEs) of simulated $PM_{2.5}$ concentrations in NEC, NC, and the YRD regions are estimated to be 38, 45, and 36 %, which is the same as the values of the NMB since the model underestimated the $PM_{2.5}$ concentration throughout the year. In the MYR, SCB, and PRD regions, the NMEs are estimated to be 18, 21, and 22 %, which are higher than the estimated the NMB, especially in the SCB. Overall, the model can reproduce the monthly variation of ambient $PM_{2.5}$ concentration in these key regions.

The $PM_{2.5}$ composition shows a great diversity across China. Sulfate–nitrate–ammonium (SNA), BC, organic matter (OM), and crustal material constituted 7.1–57 %, 1.3–12.8 %, 17.7–53 %, and 7.1–43 %, respectively, in $PM_{2.5}$ mass in China, and the fractions of SNA in $PM_{2.5}$ (40–57 %) are much higher in eastern China (Yang et al., 2011). OM and mineral dust also play significant roles in $PM_{2.5}$ concentration. $PM_{2.5}$ speciation in China simulated by GEOS-Chem has been evaluated in some previous studies. Wang et al. (2013) reported annual biases of −10, +31, and +35 % for sulfate, nitrate, and ammonia, respectively, compared with observations at 22 sites in eastern Asia. Fu et al. (2012) indicated that annual mean BC and OC concentrations in rural and background sites were underestimated by 56 and 75 %. $PM_{2.5}$ speciation is also evaluated in this study using

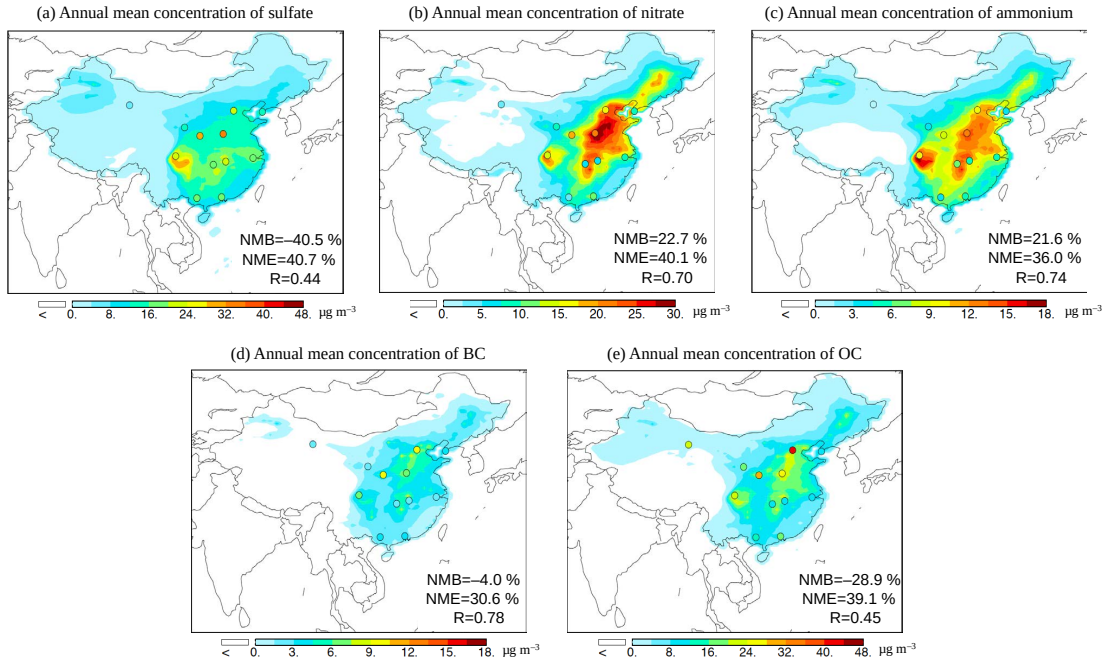

Figure 5. Simulated and observed $PM_{2.5}$ composition in China.

the observed concentration of aerosol compositions averaged from 2012 to 2013 in 12 cities across China (X. Y. Zhang et al., 2015), as shown in Fig. 5. The information of each site is described in detail in X. Y. Zhang et al. (2012). The underestimate of sulfate mainly occurs in the two cities of Zhengzhou and Xi'an, two orange spots in central and northern China, as these two sites are located in an urban area. Nitrate and ammonia are overestimated by around 20 %, which is a common issue in most chemistry transport models (CTMs.) OC is underestimated by 28.9 % due to the incomplete mechanism of SOA simulation. The NME is calculated between 30 and 41 %. The correlation coefficients range between 0.44 and 0.78.

4 Source contributions to ambient $PM_{2.5}$ concentration

4.1 Annual mean source contributions

Figure 6 shows the spatial distribution of annual mean source contributions from coal burning. As shown in Fig. 6a, the contribution from total coal burning has a similar spatial distribution with the annual mean $PM_{2.5}$ concentration, which indicates the large influence of coal burning on air quality. Table 3 also shows a higher percentage contribution in areas with higher $PM_{2.5}$ concentrations such as the NC, MYR, and SCB regions. The national average contribution from total coal burning, which is an average of concentrations in 74 major cities, is up to 22.5 $\mu g\,m^{-3}$, accounting for almost 40 % of the total $PM_{2.5}$ concentration. In the six key regions, coal burning contributes 34.5–50.2 % of the total ambient $PM_{2.5}$ concentration. The largest contribution occurs in the SCB, which reaches 36.9 $\mu g\,m^{-3}$ on average due to the dense population, large emissions, and unfavorable terrain that tends to trap the emissions and secondary pollutants in this area. The highest contribution is up to 56.9 $\mu g\,m^{-3}$, occurring in the southwestern city of Chengdu. Following the SCB, coal-burning contributions in the MYR and NC are also above the national average, with average values of 30.8 $\mu g\,m^{-3}$ (45.1 %) and 26 $\mu g\,m^{-3}$ (40.5 %), respectively. Among the six key regions, coal combustion in the PRD shows the smallest contribution of 12.6 $\mu g\,m^{-3}$, yet still accounting for 35 % of the local $PM_{2.5}$ concentration. In addition to the key regions, coal burning contributes to around 25 $\mu g\,m^{-3}$ (more than 50 %) of the local $PM_{2.5}$ in cities like Baotou and Hohhot in Inner Mongolia, an autonomous region near the middle northern border, as it is one of the largest production areas of coal and a large amount of raw coal is burnt for energy supply. In the northwestern city of Ürümqi, coal burning is also a large contributor that accounts for around 40 % of the local $PM_{2.5}$ concentration as there are no other large anthropogenic sources of air pollutants there.

Among all the subsectors in coal combustion, industrial coal burning is the most significant contributor, followed by coal burning in power plants and the domestic sector, which is shown in Fig. 6b–d and Table 3. The contribution from industrial coal burning is up to 9.6 $\mu g\,m^{-3}$ (17 %) on national average (average of 74 major cities), while those from coal burning in power plants and the domestic sector are 5.6 $\mu g\,m^{-3}$ (9.8 %) and 2.2 $\mu g\,m^{-3}$ (4 %), respectively. The contribution from each sector differs in different regions.

Table 3. Annual mean absolute contributions ($\mu g\,m^{-3}$) and percentage contributions from coal burning.

	Mean $PM_{2.5}$	Total coal-burning contributions	Contributions from coal burning in		
			power plant	industry	domestic
National average*	56.7	22.5 39.6 %	5.6 9.8 %	9.6 17.0 %	2.2 4.0 %
NEC	34.5	13.2 38.3 %	3.6 10.4 %	5.3 15.3 %	1.8 5.3 %
NC	64.3	26.0 40.5 %	7.7 12.0 %	10.8 16.8 %	1.9 2.9 %
YRD	52.2	18.0 34.5 %	5.1 9.8 %	7.6 14.6 %	0.7 1.4 %
MYR	68.3	30.8 45.1 %	6.9 10.1 %	14.0 20.5 %	2.7 3.9 %
SCB	73.5	36.9 50.2 %	5.6 7.6 %	19.0 25.9 %	4.0 5.5 %
PRD	36.2	12.6 35.0 %	2.7 7.5 %	5.7 15.8 %	0.9 2.5 %

* The national average is an average of concentrations in 74 grids where major city centers are located.

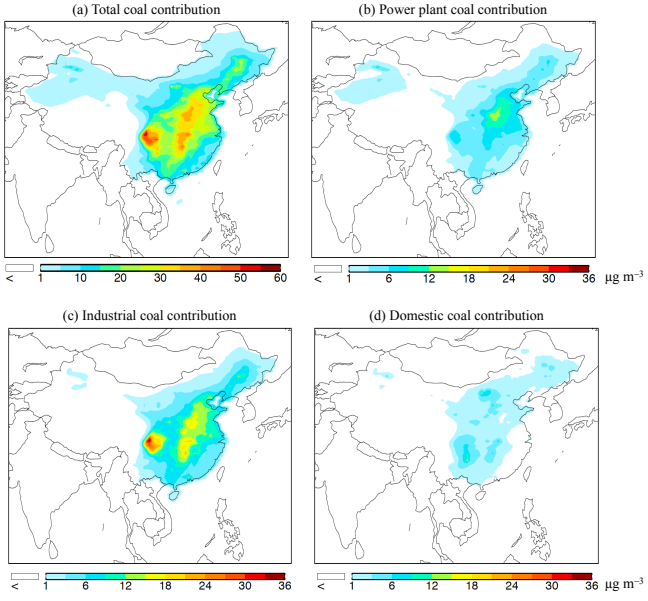

Figure 6. Annual mean contributions from coal combustion.

Contributions from coal burning in power plants and industry have similar spatial distributions to the annual mean $PM_{2.5}$ concentration. As shown in Fig. 6b, coal burning in power plants has the largest contribution in NC, with the highest value of 13.1 $\mu g\,m^{-3}$ (15 %) and an average of 7.7 $\mu g\,m^{-3}$ (12 %), due to the large number of power plants in this area. The smallest contribution occurs in the PRD with the value of only 2.7 $\mu g\,m^{-3}$ (7.5 %). In most key areas in China, coal burning in the power sector contributes to around 10 % of the local $PM_{2.5}$ concentration, which is a relatively minor source compared with industry due to higher energy efficiency and more stringent emission control policies in power sectors. Industrial coal burning, as shown in Fig. 6c, has the largest contribution in the SCB, with an average value of 19 $\mu g\,m^{-3}$ (25.9 %). The largest contribution occurs in the city of Chengdu, which is up to 35.8 $\mu g\,m^{-3}$, accounting for around one-third of the local $PM_{2.5}$. NC and the MYR are also significantly influenced by industrial coal burning, with contributions of 10.8 $\mu g\,m^{-3}$ (16.8 %) and 14 $\mu g\,m^{-3}$ (20.5 %), respectively. In other areas, including NEC, the YRD, and the PRD, the average contributions of coal burning in the industrial sector are generally less than 10 $\mu g\,m^{-3}$, accounting for around 15 % of the local $PM_{2.5}$ concentration. As shown in Fig. 6d, domestic coal burning has little contribution to ambient $PM_{2.5}$ in most areas in the six key regions. However, in some individual regions in Guizhou Province in the southwest and Inner Mongolia in North China, domestic coal burning contributes more than 10 $\mu g\,m^{-3}$, which accounts for more than 15 % in Guizhou and 25 % in Inner Mongolia where people tend to burn more raw coal for heating. In addition, the high sulfur content of coal in Guizhou Province also accounts for the large contribution.

In the nested simulation of eastern Asia, the contributions from outside the nested domain are also accounted for. In order to quantify the background concentration, we conducted another sensitivity simulation with all sources outside the domain shut off. The standard and sensitivity simulation results are shown in Fig. 7a and b, and the difference between them is analyzed as the contribution from outside the domain, which is shown in Fig. 7c. The maximum contribution from outside is up to 13.8 $\mu g\,m^{-3}$, which mainly occurs at the western and northwestern boundaries. The average contribution is 1.57 $\mu g\,m^{-3}$ in the simulation domain of eastern Asia. Within the boundary of China, the largest contribution occurs in the northeast, which is 7.35 $\mu g\,m^{-3}$. The average contribution from outside the nested domain is only 0.3 $\mu g\,m^{-3}$ within China.

4.2 Seasonal variation of coal contributions

Figure 8 shows the simulated seasonal mean $PM_{2.5}$ concentration (Fig. 8a and b) and source contributions from coal burning in winter (averaged from December to February) and in summer (averaged from June to August) (Fig. 8c–j), which is also summarized in Tables 4 and 5. As shown

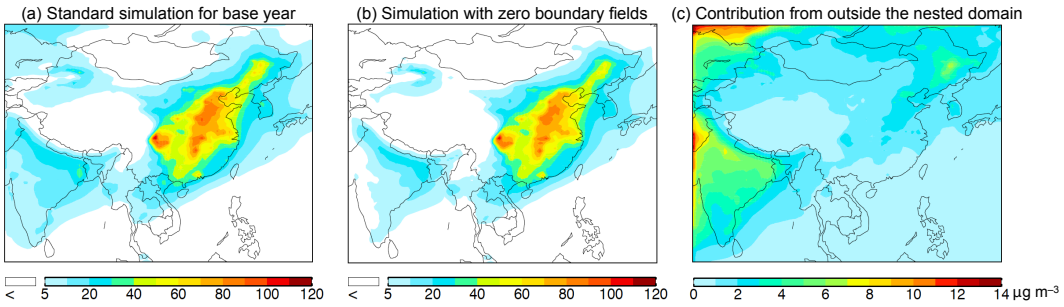

Figure 7. Annual mean contributions from outside the nested domain.

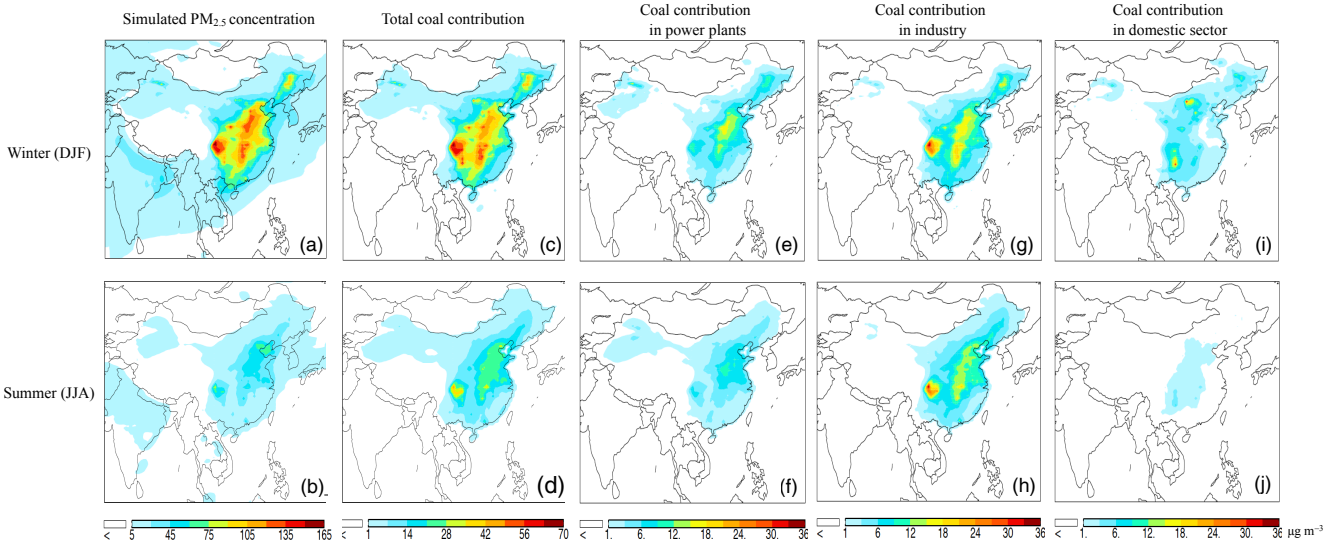

Figure 8. Seasonal contributions from coal burning in winter and summer.

in Fig. 8a and b, the ambient $PM_{2.5}$ concentration has obviously different distributions in winter and in summer. $PM_{2.5}$ in winter has a similar distribution with the annual mean, but with much higher values. The highest value still occurs in the SCB with an average of 118.8 $\mu g\,m^{-3}$ due to the large emission, unfavorable terrain, and weather conditions in winter. Following the SCB, the average concentrations in the MYR and NC regions are above 100 and 90 $\mu g\,m^{-3}$, respectively. There are also several populated cities in NEC where $PM_{2.5}$ are generally above 75 and up to 150 $\mu g\,m^{-3}$. $PM_{2.5}$ in summer has an obviously different distribution from winter with much lower concentrations and more even distribution throughout the country due to the stronger vertical mix, more wet deposition, and lower emissions. The largest concentration occurs in the NC region with 46.9 $\mu g\,m^{-3}$ on average, followed by the SCB with an average of 44.1 $\mu g\,m^{-3}$. In addition to the two regions above, $PM_{2.5}$ concentrations in other key regions are generally around or below 35 $\mu g\,m^{-3}$ on average.

In winter, coal burning contributes to 28.2 $\mu g\,m^{-3}$ (35.4 %) of total $PM_{2.5}$ concentration on the national level. Similar to the annual mean, coal-burning contribution in winter peaks in the SCB with an average of 50.3 $\mu g\,m^{-3}$ (42.3 %) and reaches the lowest in the PRD with 16.1 $\mu g\,m^{-3}$ (29 %). Among the coal-burning sectors, the contributions from power plants and industry also have similar spatial patterns to the annual mean distribution. Coal burning in industry, followed by that in power plants, is the largest contributor in both seasons. Domestic coal burning is a significant contributor in winter due to the large amount of emissions from heating supply. The high $PM_{2.5}$ concentration from the domestic sector mainly occurs in some areas in Guizhou Province in the southwest and Inner Mongolia in the north, where a large amount of raw coal is burnt for heating. The largest contribution reaches as much as 37.6 $\mu g\,m^{-3}$ in Inner Mongolia, which accounts for almost 40 % of the local $PM_{2.5}$ concentration.

In summer, the national average contribution from coal burning is estimated to be 17.8 $\mu g\,m^{-3}$ (46.2 %), which is less than two-thirds of the contribution in winter due to the favorable meteorological condition including stronger convection and more frequent wet deposition. Regional contribution ranges from 8.2 $\mu g\,m^{-3}$ in the PRD to 26.3 $\mu g\,m^{-3}$ in

Table 4. Seasonal absolute contributions ($\mu g\,m^{-3}$) and percentage contributions from coal burning in winter.

	Mean $PM_{2.5}$	Total coal-burning contributions	Contributions from coal burning in		
			power plant	industry	domestic
National average*	79.6	28.2 35.4 %	6.3 7.9 %	9.4 11.8 %	4.3 5.4 %
NEC	53.6	20.6 38.5 %	5.5 10.3 %	6.8 12.7 %	4.0 7.4 %
NC	90.0	31.8 35.3 %	9.2 10.2 %	10.6 11.8 %	3.1 3.4 %
YRD	66.2	19.5 29.5 %	4.8 7.2 %	6.7 10.1 %	1.2 1.7 %
MYR	104.9	40.2 38.3 %	9.3 8.9 %	14.0 13.4 %	3.8 3.6 %
SCB	118.8	50.3 42.3 %	7.4 6.3 %	18.9 15.9 %	7.3 6.2 %
PRD	55.4	16.1 29.0 %	2.2 4.0 %	5.4 9.8 %	1.8 3.2 %

* The national average is an average of concentrations in 74 grids where major city centers are located.

Table 5. Seasonal absolute contributions ($\mu g\,m^{-3}$) and percentage contributions from coal burning in summer.

	Mean $PM_{2.5}$	Total coal-burning contributions	Contributions from coal burning in		
			power plant	industry	domestic
National average*	38.4	17.8 46.2 %	5.2 13.4 %	9.0 23.4 %	1.0 2.5 %
NEC	20.3	8.9 44.1 %	2.7 13.3 %	4.8 23.4 %	0.5 2.5 %
NC	46.9	21.7 46.4 %	7.3 15.5 %	10.5 22.5 %	1.0 2.1 %
YRD	34.1	14.2 41.5 %	4.7 13.8 %	6.7 19.5 %	0.18 0.5 %
MYR	36.2	20.2 56.1 %	5.1 14.2 %	11.6 32.0 %	1.6 4.5 %
SCB	44.2	26.2 59.5 %	4.7 10.7 %	16.0 38.5 %	1.9 4.2 %
PRD	20.2	8.2 40.7 %	2.2 10.8 %	4.3 21.5 %	0.3 1.5 %

* The national average is an average of concentrations in 74 grids where major city centers are located.

the SCB, which is approximately half of the contributions in winter. The seasonal variation of contributions in inland areas (NEC, MYR, SCB) is more significant than those in coastal areas (NC, YRD, PRD). In coal-burning sectors, the absolute contributions from power plants and industry do not show very noticeable reductions in summer compared with those in winter as emissions from these two sectors are in a relatively constant status throughout the year and the nitrate reduction due to the high temperature in summer is counteracted by the enhancement of sulfate formation (H. Zhang et al., 2012). In contrast, the domestic sector contributes 1 $\mu g\,m^{-3}$ (2.5 %) on the national level in summer, which is 3–8 times less than that in winter.

4.3 Comparisons with other studies

The Natural Resources Defense Council (NRDC) launched the China Coal Consumption Cap Project in October 2013 and released the report "Coal Use's Contribution to Air Pollution in China" as part of the study results in October 2014 (NRDC, 2014). This study used the CAMx model with the Multi-resolution of Emission Inventory for China (MEIC) inventory and meteorology from the Weather Research and Forecasting (WRF) model to simulate coal contributions to ambient $PM_{2.5}$ in January, February, April, and October in the year 2012 in 333 main cities in China. In order to compare with the NRDC study, we extracted the simulated contribution in the 333 main cities during the same periods from our study results. Figure 9 represents the comparison in each province and shows that our study underestimates the coal contribution by 22 % compared to that in the NRDC study. This discrepancy is mainly generated from the different amounts of emissions that originate from coal in the two studies. According to the report, the NRDC study included both emissions directly from coal burning and emissions from industries closely related to coal burning. For example, air pollutants from industries like coke, steel, cement, and nonferrous metal are generated two ways: directly from coal combustion and from technological processes. As coal is used as fuel in these industries and is not likely to be substituted for in the near future, the NRDC study includes both parts as emissions from coal use. In our study, we include only the first part of the emissions as the contribution from coal, which is actually generated from coal burning. According to the report by the NRDC, coal combustion is responsible for 79 % of SO_2 emissions, 57 % of NO_x emissions, and 44 % of primary PM emissions, and the coal-related sources are responsible for 15, 13, and 23 % of the SO_2, NO_x, and

Table 6. Results of the uncertainty analysis of the emissions in China.

	NO_X	SO_2	$PM_{2.5}$	NMVOC
Power plants	±34 %	±30 %	±31 %	–
Industrial sector	±41 %	±49 %	±53 %	±63 %
Residential sector	±55 %	±51 %	±68 %	±65 %
Transportation	±66 %	±48 %	±52 %	±57 %
Solvent use	–	–	–	±78 %
Other sectors[a]	±177 %	±179 %	±216 %	±184 %
Total emissions[b]	[−31 %, 44 %]	[−29 %, 45 %]	[−39 %, 49 %]	[−42 %, 67 %]

[a] Other sectors mainly refer to open biomass burning. [b] The last line shows the average 90 % confidence intervals of the total emissions.

Table 7. Comparisons with other studies on recent air pollutant emissions in China (in kilotons).

	SO_2	NO_X	PM_{10}	$PM_{2.5}$	VOCs
This study	23 150	25 638	16 521	12 155	23 366
MEP (2014s)	20 439	22 273	–	–	–
Liu et al. (2016)	–	28 300	–	–	–
Xia et al. (2016)	23 014–26 884	28 002–28 817	–	–	–
Wu et al. (2016, 2012*)	–	–	–	–	29 850
Zhao et al. (2014, 2015*)	26 792	27 511	15 599	11 419	–

* The year of emission if different from the year of emission (2013) in our study.

PM emissions, respectively. Despite the different definition of coal contribution to air pollutant emissions, the NRDC and our study both predicted a high contribution to $PM_{2.5}$ concentration from coal, especially in the municipality of Chongqing and Sichuan Province in the SCB.

4.4 Uncertainty analysis

The uncertainties of the contribution estimates in this study may arise from the uncertainties of the emission inventory, model simulation, and non-linearity of the atmospheric chemistry. A Monte Carlo uncertainty analysis was performed on the emission inventory, as described in Zhao et al. (2013c) and S. X. Wang et al. (2014b). Table 6 shows the uncertainty analysis of the emissions in China. Among all the coal-consuming sectors analyzed in this study, the domestic sector is subject to the highest uncertainty, which may lead to more uncertainty in the $PM_{2.5}$ simulation and contribution estimates. Other studies on major pollutant emissions in China are summarized in Table 7. Emissions from Liu et al. (2016), Xia et al. (2016), and Wu et al. (2016) are also estimated using the bottom-up method, while those from Zhao et al. (2014) are projected emissions for 2015 based on the year 2010. The results of this study fall into the range of previous studies except for China's Ministry of Environmental Protection (MEP, 2014a), which is at low end. One major reason for low NO_x emission from MEP (2014a) is that it does not include the emissions from non-road vehicles.

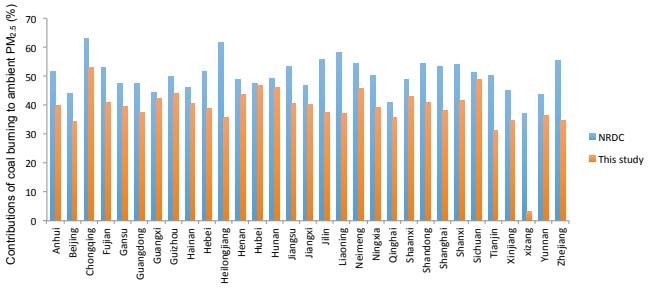

Figure 9. Comparison of coal contribution to $PM_{2.5}$ concentration between NRDC and this study.

Another important cause of uncertainty is the model simulation of the $PM_{2.5}$ composition. The coal contribution to sulfate is larger than that to nitrate since the share of coal-burning emissions of SO_2 is 79 % in this study, 25 % higher than that of NO_x emissions. Therefore, the actual coal-burning contribution to $PM_{2.5}$ is very likely to be larger than the estimates in this study due to the underestimation of sulfate concentration and overestimation of nitrate concentration by the model.

In addition, due to the nonlinear response of $PM_{2.5}$ concentration to precursor emissions, contributions from coal burning in each sector add up to less than the contribution from the total coal burning, which indicates the probable underestimation of the contribution in subsectors. The impact of nonlinearity of the atmospheric chemistry on $PM_{2.5}$

concentrations and their composition has been discussed in detail in previous studies (Zhao et al., 2013b; S. Wang et al., 2014a). There are some studies using different methods to study the source apportionment of ambient $PM_{2.5}$. As this study only focuses on coal-burning emissions in each sector, the results are not directly comparable to most similar studies except for results for the power sector as coal combustion dominates the emissions in the power plant sector. Zhao et al. (2015) used the extended response surface modeling technique to access the nonlinear response of fine particles to precursor emissions in each sector in the PRD region, reporting that local $PM_{2.5}$ concentration decreased less than 3 % (7.2 % in our study) in January and around 12 % in August (13.8 % in our study), when 90 % of emissions in power plants are reduced. Our results include the trans-boundary contributions as we shut off emissions across the country in the sensitivity simulation, which is one of the reasons causing the discrepancies. L. Zhang et al. (2015) took advantage of the adjoint capability of GEOS-Chem, reporting that power plants contributed 6 % to $PM_{2.5}$ concentration in Beijing, which is consistent with our study (6.9 %).

5 Conclusion

We updated China's emission inventory to the year 2013 using up-to-date information on energy statistics and emission control policies. The anthropogenic emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC, and NH_3 in China were estimated to be 23.2, 25.6, 16.5, 12.2, 1.96, 3.42, 23.3, and 9.62 Mt, respectively. Using the emission inventory, we conducted standard and sensitivity simulations for major coal-burning sectors to quantitatively identify the source contributions from coal burning using the chemical transport model GEOS-Chem. Results show that coal combustion contributes 22.5 $\mu g\,m^{-3}$ (40 %) of the total $PM_{2.5}$ concentration on national average (average of 74 major cities). The highest contribution occurs in the Sichuan Basin, which reached 36.9 $\mu g\,m^{-3}$ and accounts for more than 50 % of the local $PM_{2.5}$. Among the subsectors of coal combustion, industrial coal burning is the dominant contributor, with the largest contribution of 19 $\mu g\,m^{-3}$ (26 %) in the Sichuan Basin and the second largest of 14 $\mu g\,m^{-3}$ (20 %) in the Middle Yangtze River area, which indicates that coal combustion in industry should be prioritized when energy policies and end-of-pipe control strategies are applied, especially in middle–west regions in China, from the perspective of the whole country. Coal combustion in power plants shows the largest contribution in North China, with an average of 7.7 $\mu g\,m^{-3}$ (12 %). Domestic coal burning has the largest contribution in some regions in Guizhou Province in Southwest China and Inner Mongolia in North China, where combustion of raw coal should be substantially reduced, especially in winter. An obvious seasonal variation is also predicted. The absolute contributions due to coal combustion are estimated to be 28 $\mu g\,m^{-3}$ (35 %) in winter and 18 $\mu g\,m^{-3}$ (46 %) in summer on the national level. The seasonal differences are mainly due to the dramatic change of domestic emissions and more favorable meteorological conditions, including stronger convection and wet deposition in summer. While contribution from domestic coal shows a significant reduction from winter to summer, the absolute contributions from coal burning in power plants and industry remain at relatively steady levels throughout the year.

Data availability. Data in this work are available upon request.

The Supplement related to this article is available online at doi:10.5194/acp-17-4477-2017-supplement.

Competing interests. The authors declare that they have no conflict of interest.

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