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Environmental effects of the recent emission changes in China: implications for particulate matter pollution and soil acidification

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Abstract
Driven by a rapid increase of energy consumption and emerging pollution control policies, air pollutant emissions have changed dramatically in China during 2005–2010. This study developed a multi-pollutant emission inventory, and used the community multi-scale air quality (CMAQ) modeling system to evaluate the impact of the emission changes on particulate matter pollution and soil acidification. During 2005–2010, the emissions of SO_2, PM_{10} and $PM_{2.5}$ decreased by 14.9%, 15.1% and 11.7%, respectively. In contrast, the emissions of NO_X, NMVOC and NH_3 increased by 33.8%, 21.0% and 10.4%, respectively. The emission trends differed notably in different regions. Driven by emission changes, $PM_{2.5}$ concentrations decreased by 2–17 $\mu g\ m^{-3}$ in most of the North China Plain, the Yangtze River Delta and the Pearl River Delta, while increasing by 4.5–16 $\mu g\ m^{-3}$ in most of the Sichuan Basin and Eastern Hubei. The changes of $PM_{2.5}$ emissions led to the decline of primary $PM_{2.5}$ concentrations in most of Eastern China. As an effect solely of emission changes, nitrate concentrations increased across most of China; sulfate concentrations decreased in most of Eastern China, with the largest reduction in the North China Plain, while they increased in the Sichuan Basin and parts of the Pearl River Delta and Eastern Hubei. The concentrations of secondary inorganic aerosols (SIA) and the extinction coefficient increased in most of China, especially in the Sichuan Basin and Eastern Hubei, implying that the NO_X and NH_3 emissions should be reduced simultaneously in China. Combining the acidification effects of S and N, the exceedance of critical loads decreased across the country, but increased in the Sichuan Basin, the Pearl River Delta and Eastern Hubei, where the soil acidification was the most serious. Different control policies need to be implemented in different regions.

Keywords: emission inventory, particulate matter pollution, soil acidification, CMAQ
S Online supplementary data available from stacks.iop.org/ERL/8/024031/mmedia

1. Introduction

Driven by rapid economic development and intensive energy use, emissions of atmospheric pollutants have caused serious

environmental effects in China. Among these effects are high concentrations of primary and secondary particulate matter (Lin *et al* 2010, Remer *et al* 2008), and the world's highest levels of acid deposition observed in parts of southern China (Wang and Xu 2009, Zhao *et al* 2007). The high particulate matter concentrations are associated with severe haze pollution (Huang *et al* 2012, Zhang *et al* 2012a, 2012b), climate forcing (Solomon *et al* 2007), and human health (WB and SEPA 2007, Ho and Nielsen 2007).

During 2005–2010, the period of the 11th Five Year Plan, Chinese energy consumption increased by 6.6% annually, compared to an annual average increase of 5.6% during 1980–2005. Under strong environmental pressure, the Chinese government took aggressive steps to improve energy efficiency and reduce emissions of primary aerosols and sulfur dioxide (SO_2) during this period. Small, inefficient power generating units and industrial facilities were replaced with larger and more efficient ones. The ratio of thermal power plants equipped with flue gas desulfurization (FGD) increased from 12% in 2005 to 82.6% in 2010 (The State Council of the People's Republic of China 2011). These measures were expected to have changed the emission pathway significantly. According to the Ministry of Environmental Protection (MEP), the emissions of total suspended particulates (TSP) and SO_2 decreased by 39.0% and 14.3% respectively from 2005 to 2010 (MEP 2012). Nitrogen oxide (NO_X) emissions were estimated to have increased by about 30% during the same period in East Central China (Zhang *et al* 2012a, 2012b). The quantification of the changes of pollutant emissions and the driving force underlying these changes calls for a systematic multi-pollutant emission inventory, on which there were only limited studies for the studied period.

The environmental effects of changes of air pollutant emissions can be complex, and even appear to conflict. Lin *et al* (2010) reported a general downward trend of the concentrations of particulate matter less than or equal to 10 μm (PM_{10}), in comparison with an upward trend of aerosol optical depth (AOD) in east central China during October 2004–September 2008. Guo *et al* (2011) illustrated that AOD increased slowly by 0.0004 per year in China during 2000–2008, and the upward tendency was observed in most parts of China. Hao (2012) analyzed the long-term trend of visibility in six major cities of the Yangtze River Delta, and concluded that the visibility have been fluctuating around a constant value since 2000, in contrast with the decreasing trend of PM_{10} concentrations in most cities. As for soil acidification, Zhao *et al* (2009) suggested that the benefits of SO_2 reductions during 2005–2010 might be negated by enhanced NO_X and NH_3 emissions on the basis of critical load exceedance. Previous studies on environmental effects of emission changes focused either on the national level or on a specific region. In particular, the environmental effects of the national and regional emission trends during China's 11th Five Year Plan have not been evaluated systematically, which would be beneficial for air pollution study and future policy making.

This paper aims to quantify the emission changes in China and the driving force underlying these changes during 2005–2010, and to understand the complex environmental effects of the emission changes. We developed a new multiple pollutant emission inventory for the years 2005–2010 and evaluated the impacts of emission changes on different particulate matter species and soil acidification over both national and regional scales. We focused on how the different emission trends and chemistry mechanisms in various seasons and regions affect the environmental quality, which had important policy implications but were not addressed in previous studies.

2. Methodology and data sources

2.1. Emission inventory

We developed an emission inventory of SO_2, NO_X, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC, and NH_3 for China for the years 2005–2010. An 'emission factor method' was used to calculate air pollutant emissions, as described in detail in Wang *et al* (2011a). A unit based method was applied to estimate the emissions from large point sources including coal-fired power plants, iron and steel plants, and cement plants (Lei *et al* 2011, Wang *et al* 2011a, Zhao *et al* 2008). The data sources for emission estimation and parameters for spatial and temporal distribution are described in detail in the supplementary materials (available at stacks.iop.org/ERL/8/024031/mmedia).

2.2. Modeling system configuration and evaluation

The Models-3 community multi-scale air quality (CMAQ) modeling system version 4.7.1 was used in this study. The modeling domain covered most of China and part of East Asia with a 36 km × 36 km grid resolution, as shown in figure 1. The six typical regions for region specific analysis are also shown in figure 1, including Eastern China, the North China Plain, the Yangtze River Delta, the Pearl River Delta, the Sichuan Basin, and Eastern Hubei. These regions are chosen because of their high emission intensities and elevated pollutant concentrations. The configurations of geophysical projection, vertical resolution, scientific options, initial conditions, and boundary conditions for CMAQ were consistent with our previous paper (Wang *et al* 2010). The Weather Research and Forecasting Model (WRF, version 3.3) was used to generate the meteorological fields. The WRF data sources, and the major physics options of WRF are described in the supplementary materials (available at stacks.iop.org/ERL/8/024031/mmedia). The simulation periods include the whole year of 2005 and 2010. The changes of particulate matter concentrations from 2005 to 2010 are driven by both emission changes and meteorological conditions. Previous studies (Xing *et al* 2011, Wang *et al* 2009) have conducted sensitivity simulations with the same meteorology and different emissions compared with the baseline case to evaluate the contributions of emission changes in isolation. Similarly, we developed a sensitivity case (2005SENS) with meteorology of 2010 and the emissions of 2005 to quantify the environmental effects solely of emission changes.

The comparison of the meteorological predictions and air quality simulations to surface and satellite observations

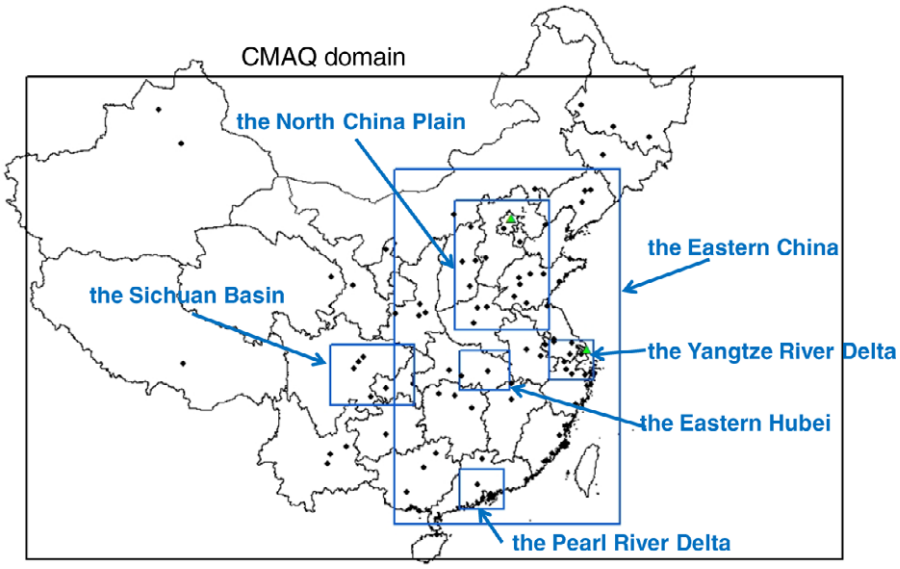

Figure 1. The Models-3/CMAQ modeling domain at a horizontal grid resolution of 36 km (164 × 97 cells), and the locations of the six studied regions, i.e. Eastern China, the North China Plain, the Yangtze River Delta, the Pearl River Delta, the Sichuan Basin, and Eastern Hubei. The black dots indicate the 86 major cities monitored by MEP (58 of them are located in Eastern China). The two green triangles indicate the locations of Miyun (north) and Chongming (south) site.

in 2005 and 2010 is described in detail in the supplementary materials. In general, the model can capture the meteorological conditions and pollutant concentrations. For example, the simulated monthly average $PM_{2.5}$ concentrations are comparable with observations in Miyun and Chongming sites (see figure 1), with NMBs ranging from −15% to 1% for Miyun, and from −23% to 4% for Chongming (see figure S4 available at stacks.iop.org/ERL/8/024031/mmedia). We also evaluated the simulated trends of air quality during 2005–2010. MEP reported the daily air pollution index (API) of 58 major cities in Eastern China (the major contaminative region of China) on its official website (http://datacenter.mep.gov.cn). The average observed PM_{10} concentrations of these cities decreased by 7.6% during the studied period, which agrees well with the declining rate of the average simulated PM_{10} concentrations (7.3%). The comparison of simulated AOD changes with the observation of moderate resolution imaging spectroradiometer (MODIS) could verify the simulated changes of fine particulate matter pollution in the whole domain. As shown in figures S8(a) and (b) (available at stacks.iop.org/ERL/8/024031/mmedia), the modeling results can reproduce the spatial pattern of AOD changes fairly well, especially the notable increase in the Sichuan Basin and the southern part of the North China Plain, and the decline along the southeastern coast.

2.3. Critical load for soil acidification

The effect of air pollutant emissions on soil acidification is estimated by comparing the simulated deposition with critical load (CL), which has been used as the basis of emission control and acid rain mitigation in Europe (UBA 2004) as well as China (Hao *et al* 2000, Zhao *et al* 2011a, 2009, 2011b). The maximum critical loads of S deposition, i.e. $CL_{max}(S)$, were quantified with an extended steady state mass balance (SSMB) method (Zhao *et al* 2009). A critical load for nutrient nitrogen, $CL_{nut}(N)$, was derived independently from the mass balance for N (UBA 2004). When combining the effects of S and N, the critical load of S deposition might be lower than $CL_{max}(S)$, therefore, the critical load of S is recalculated with (1):

$$CL(S) = \begin{cases} CL_{max}(S) & (N_D \le N_I + N_U) \\ CL_{max}(S) - (1 - f_{DE}) & \\ \quad \times [N_D - (N_I + N_U)] & (N_D > N_I + N_U) \end{cases} \quad (1)$$

where CL(S) is the critical load of S considering the effect of N deposition; f_{DE} is the nitrate lost ratio by denitrification; N_D is the deposition of N; N_I is the net N immobilization rate in the soil; and N_U is the net uptake of N by vegetation. This equation lays the basis for integrated analyses of S and N in this assessment. CL maps across China were generated with a resolution of 36 km × 36 km (the same as CMAQ). More details of the method and the values of major parameters are described in Zhao *et al* (2009).

3. Results and discussions

3.1. Air pollutant emissions during 2005–2010

In 2010, the anthropogenic emissions of SO_2, NO_X, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC, and NH_3 in China were estimated at 24.42 Mt, 26.06 Mt, 15.81 Mt, 11.79 Mt, 1.93 Mt, 3.51 Mt, 22.86 Mt, and 18.31 Mt respectively. The sectoral emissions of SO_2, NO_X, $PM_{2.5}$, and NH_3 during 2005–2010 are shown in figure 2, and those of PM_{10}, BC, OC, and NMVOC are shown in figure S1 (available at stacks.iop.org/ERL/8/

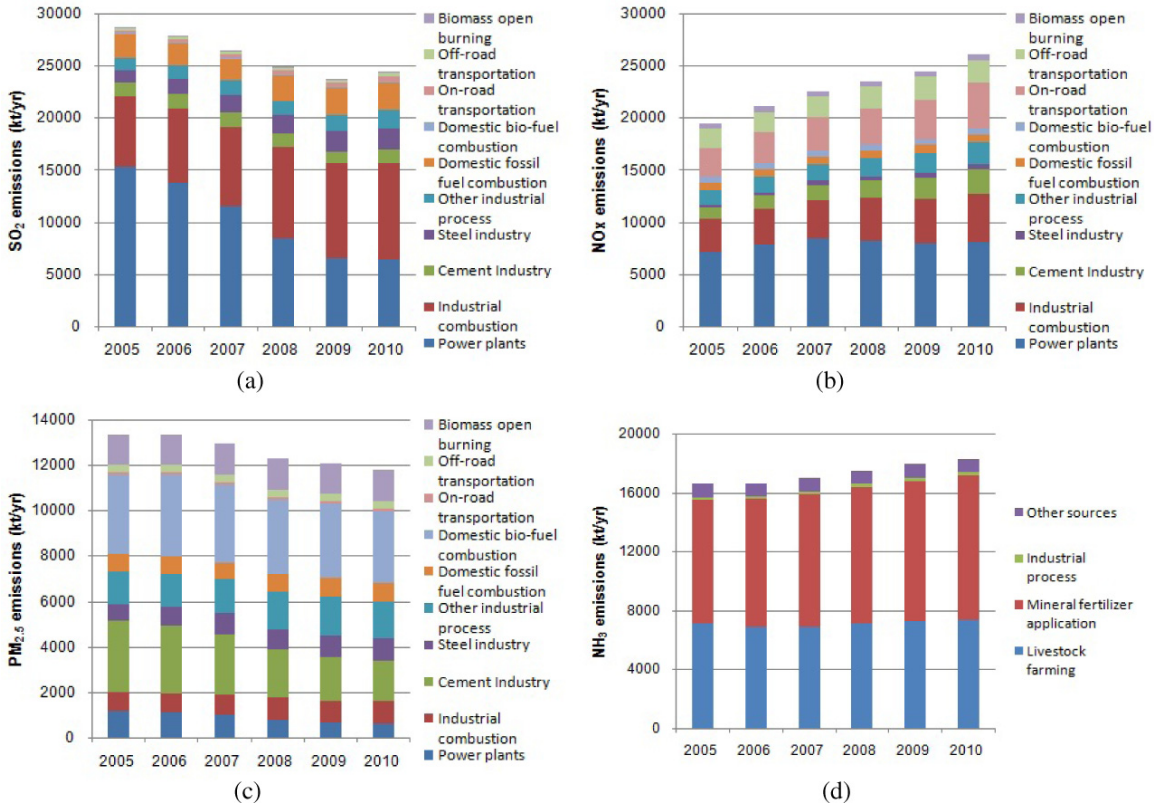

Figure 2. The emissions of SO_2, NO_X, $PM_{2.5}$, and NH_3 by sector from 2005 to 2010. (a) SO_2. (b) NO_X. (c) $PM_{2.5}$. (d) NH_3.

024031/mmedia). Total SO_2 emissions decreased by 14.9% from 2005 to 2010, mainly attributable to the large scale installation of flue gas desulfurization (FGD) for power plants (see section 1). Total NO_X emissions increased by 33.8% from 2005 to 2010, driven by the rapid increase of emissions from industry and transportation. The PM_{10} and $PM_{2.5}$ emissions decreased by 15.1% and 11.7% respectively from 2005 to 2010. The emissions of power plants and cement industry experienced the fastest decrease (43%–47% reduction from 2005 to 2010), as a result of the rapid installation of end-of-pipe removal equipment. Total NH_3 emissions grew by 10.4% during the 5 years, mainly attributed to the increase of fertilizer application.

Trends of emissions during the five years differed notably in different regions (figure 3). While the national emissions of SO_2, PM_{10}, and $PM_{2.5}$ decreased significantly, the Sichuan Basin experienced a 26% increase of SO_2 emissions, and the Eastern Hubei experienced growth rates of 14% and 0.4% for SO_2 and $PM_{2.5}$ emissions, respectively. In comparison, SO_2 and $PM_{2.5}$ emissions decreased by 31% and 30% respectively in the Yangtze River Delta, much faster than the national rate of decline. The growth rates of NO_X emissions varied from 66% in the Sichuan Basin, and 46% in Eastern Hubei, to only 17% in the Yangtze River Delta. The temporal evolution of energy consumption is an importance driving force of emission changes. According to the Chinese Energy Statistical Yearbook (CESY), industrial coal consumption increased by 131%, 70%, and 12% in Sichuan and Chongqing provinces (representative of the Sichuan Basin), Hubei province (representative of Eastern Hubei), and Shanghai, Jiangsu, and Zhejiang provinces (representative of the Yangtze River Delta) respectively, leading to different emission trends. In addition, control equipment is usually more widespread in large city clusters, because of more aggressive environmental policies. For example, Shanghai, Jiangsu, and Zhejiang provinces (representative of the Yangtze River Delta) possessed 17% of the total coal-fired power generation capacity, while they possessed 35% of the total flue gas denitrification facilities by 2010, leading to a slower increase in NO_X emissions.

The differences in emission estimates between different studies may affect the modeled environmental effects to some extent. Zhao *et al* (2013) estimated the SO_2 and primary $PM_{2.5}$ emissions to decrease by 10.8% and 5.9% respectively during 2005–2010, with slower declining rates than our estimates (14.9% and 11.7% decline respectively). In contrast, they estimated a larger growth rate (46.7%) in NO_X emissions than our work (33.8%). Despite the discrepancy in growth rates, the temporal trends are generally consistent in these two studies, therefore it would not change the major conclusions and policy implications of this study.

3.2. Effects on particulate matter pollution

Figure 4 illustrates the annual mean $PM_{2.5}$ concentrations in 2010, and the changes from 2005 to 2010. The difference between the concentrations in 2010 and the 2005SENS case (see section 2.2), representing the effects solely of

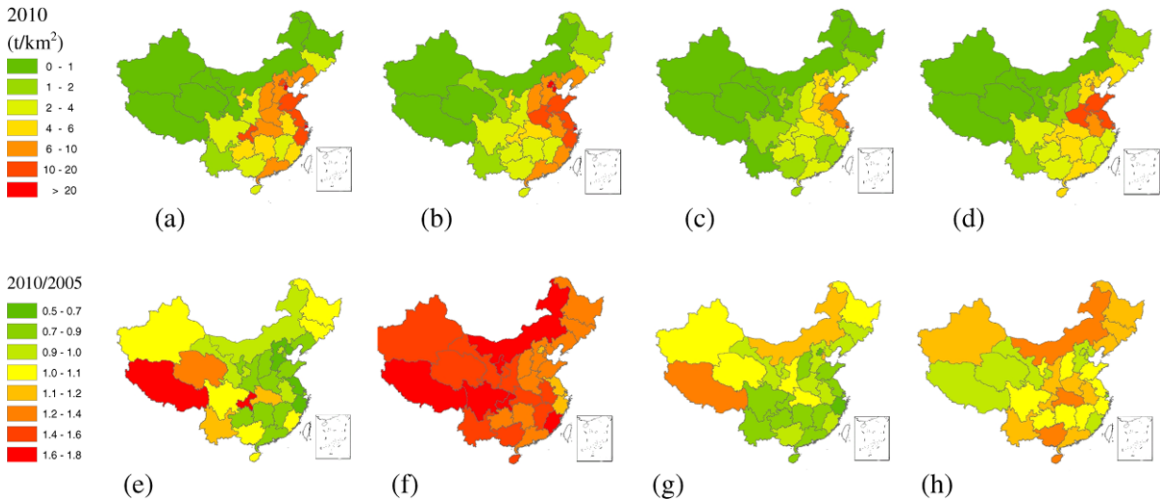

Figure 3. Emission intensities of SO_2, NO_X, $PM_{2.5}$ and NH_3 in 2010 and the changes from 2005 to 2010. Data not available for Taiwan. (a) SO_2, 2010. (b) NO_X, 2010. (c) $PM_{2.5}$, 2010. (d) NH_3, 2010. (e) SO_2, 2010/2005. (f) NO_X, 2010/2005. (g) $PM_{2.5}$, 2010/2005. (h) NH_3, 2010/2005.

emission changes, is also shown in figure 4. It can be seen that $PM_{2.5}$ concentrations in Eastern China are significantly higher than other regions due to higher emission intensity; the most polluted regions include the North China Plain, the Yangtze River Delta, Eastern Hubei, the Sichuan Basin, and the Pearl River Delta. The effects of emission changes on $PM_{2.5}$ concentrations differ greatly in different regions. During 2005–2010, the annual average $PM_{2.5}$ concentration decreased by 2–17 $\mu g\ m^{-3}$ in most parts of the North China Plain, the Yangtze River Delta, and the Pearl River Delta, contrasted by an increase of 4.5–16 $\mu g\ m^{-3}$ in most parts of the Sichuan Basin and Eastern Hubei, as an effect solely of emission changes. The changes of annual average $PM_{2.5}$ concentrations due to emission changes are $-0.9\ \mu g\ m^{-3}$, $-6.1\ \mu g\ m^{-3}$, $-2.1\ \mu g\ m^{-3}$, $5.2\ \mu g\ m^{-3}$, and $5.6\ \mu g\ m^{-3}$ in the North China Plain, the Yangtze River Delta, the Pearl River Delta, the Sichuan Basin, and Eastern Hubei, respectively, while the absolute changes of $PM_{2.5}$ concentrations are estimated to be $-0.6\ \mu g\ m^{-3}$, $-6.5\ \mu g\ m^{-3}$, $-3.2\ \mu g\ m^{-3}$, $10.4\ \mu g\ m^{-3}$, and $6.0\ \mu g\ m^{-3}$ respectively during the five years. The emission changes dominated the changes of particulate matter concentrations in four of the five regions. In the Sichuan Basin, the meteorological conditions made approximately equal contribution to the enhancement of particulate matter concentrations.

Concentrations and changes of different species of $PM_{2.5}$ are also illustrated in figure 4. The changes of primary $PM_{2.5}$ emissions led to the notable reduction of primary $PM_{2.5}$ concentrations in most parts of Eastern China, except for the slight increase in Eastern Hubei. The reduction was especially effective in urban areas with high emissions. Primary PM emissions from industrial combustion, steel industry, domestic fossil-fuel and biomass combustion continued to increase or remained constant during this period, which still have substantial reduction potential (see figure 2). The controls of primary PM should be strengthened in the future to lower $PM_{2.5}$ concentrations.

Secondary inorganic aerosols (SIA) account for 58%–60% of the simulated annual mean $PM_{2.5}$ concentrations in Eastern China. The concentrations and spatial distributions of SIA have large discrepancies in different seasons. Therefore, the seasonal average concentrations of sulfate, nitrate, and SIA, and their changes during the five years are shown in table 1 and figures S5–S7 (available at stacks.iop.org/ERL/8/024031/mmedia). Similar to $PM_{2.5}$ concentrations, we will focus on the effects solely of emission changes, and discuss the impact of meteorology briefly at the end. Nitrate concentrations increased almost universally in China during all the seasons, driven by the increase of NO_X and NH_3 emissions, and thermodynamic effects of the sulfate reduction. The most remarkable increase occurred in Eastern Hubei and the Sichuan Basin, resulting from the notable increase in winter and fall in these two regions. In winter and fall, average nitrate concentrations are evidently higher than in spring and summer, as high temperature favors the evaporation of nitrate. In addition, the largest increase in winter and fall occurs in Eastern Hubei and the Sichuan Basin, attributable to favorable meteorological conditions for nitrate formation, and rapid NO_X emission growth in the two regions (see section 3.1).

In comparison to nitrate, annual average sulfate concentrations decreased in most parts of Eastern China, with the largest reduction in the North China Plain, while they increased in the Sichuan Basin, and over small regions around the city centers of the Yangtze River Delta and Eastern Hubei. Wang *et al* (2011b) indicated that most parts of Eastern China, in particular the regions with high SIA concentrations, are located in NH_3-rich conditions. Therefore, the sulfate concentration decreased in most parts of Eastern China with the reduction of SO_2 emissions and the growth of NH_3 emissions. The fact that the largest reduction happened in the North China Plain results from the prominent reduction in that region during summer and fall. Two factors underlie this notable reduction: higher temperature and stronger

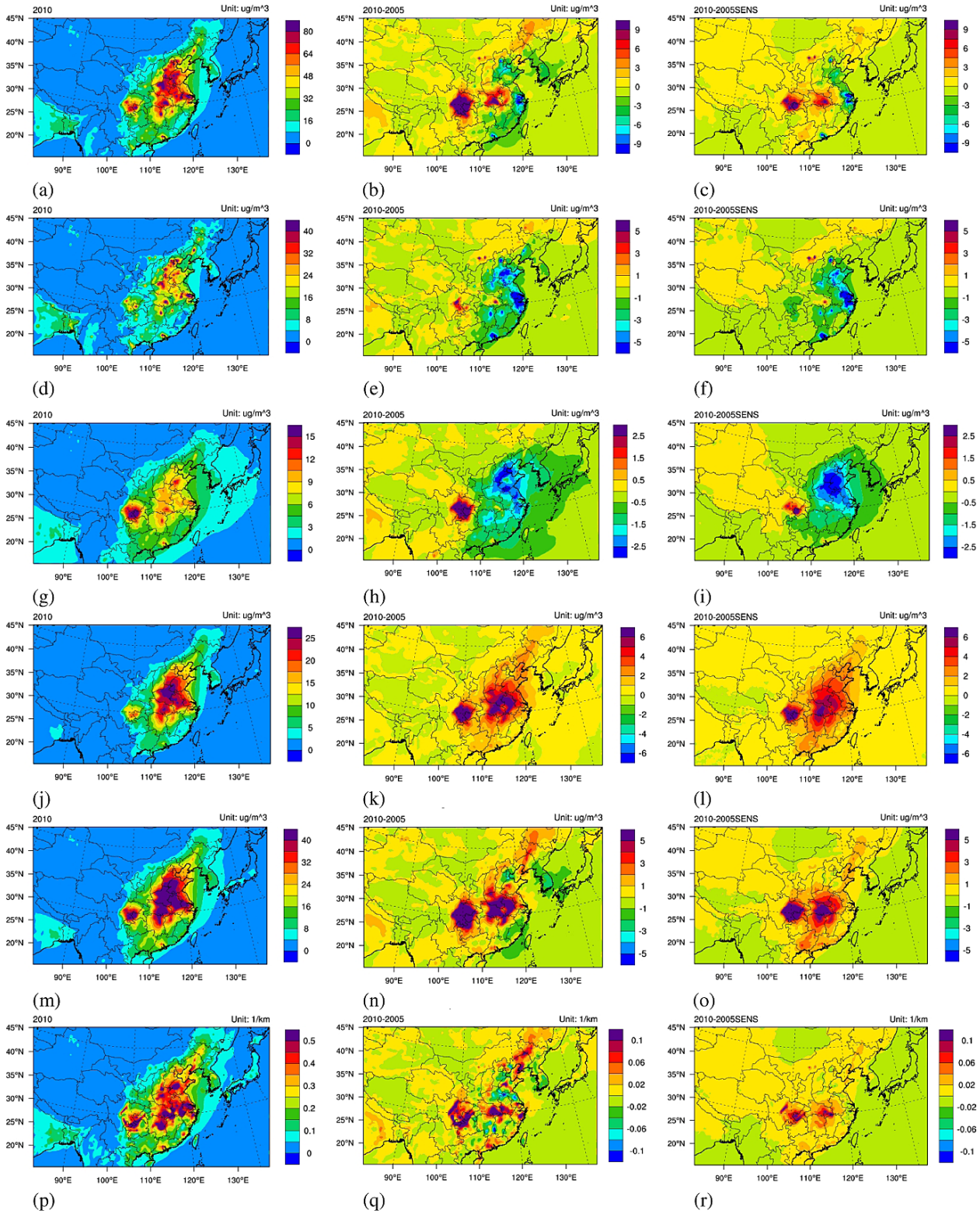

Figure 4. Spatial distribution of the annual mean concentrations (values) of $PM_{2.5}$, primary $PM_{2.5}$, SO_4^{2-}, NO_3^-, SIA, and extinction coefficient in 2010 (left column), the changes during 2005–2010 (2010 minus 2005, middle column), and the changes as an effect solely of emission changes (2010 minus 2005SENS, right column): (a)–(c) $PM_{2.5}$; (d)–(f) primary $PM_{2.5}$; (g)–(i) SO_4^{2-}; (j)–(l) NO_3^-; (m)–(o) SIA; (p)–(r) extinction coefficient.

oxidation capacity lead to larger reduction in summer and fall than in winter; transport of sulfate and its precursors from southern China (driven by southerly monsoon) results in larger reduction in the North China Plain than in south. The increase of sulfate concentrations in the Sichuan Basin results from the increase of SO_2 (see section 3.1) and NH_3 emissions

Table 1. The seasonal and annual mean simulated concentrations of $PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, and SIA in the studied regions in 2010, and the changes from 2005 to 2010 as an effect solely of emission changes (the values in brackets).

Unit: $\mu g\ m^{-3}$	Eastern China	The North China Plain	The Yangtze River Delta	The Pearl River Delta	The Sichuan Basin	Eastern Hubei
Winter						
$PM_{2.5}$	42.36 (1.47)	63.20 (−0.15)	69.98 (−6.60)	34.38 (−2.44)	57.65 (7.00)	85.02 (9.33)
SO_4^{2-}	4.60 (−0.49)	4.53 (−0.68)	6.63 (−1.31)	5.52 (−0.27)	10.51 (1.09)	7.85 (−0.67)
NO_3^-	13.57 (2.67)	19.10 (2.13)	22.81 (3.23)	8.49 (1.73)	15.80 (6.10)	30.80 (6.80)
NH_4^+	5.68 (0.60)	7.27 (0.36)	9.24 (0.47)	4.59 (0.42)	8.51 (2.17)	11.89 (1.72)
SIA	23.85 (2.78)	30.92 (1.82)	38.70 (2.40)	18.61 (1.89)	34.83 (9.37)	50.55 (7.86)
Spring						
$PM_{2.5}$	25.41 (0.39)	40.94 (0.11)	47.44 (−5.21)	24.87 (−1.82)	28.82 (3.99)	50.13 (4.81)
SO_4^{2-}	4.62 (−0.72)	5.31 (−1.34)	7.43 (−1.49)	5.60 (−0.19)	8.53 (0.86)	8.69 (−0.92)
NO_3^-	7.02 (1.81)	13.17 (2.70)	12.40 (2.35)	4.73 (1.12)	5.79 (2.70)	15.76 (4.72)
NH_4^+	3.78 (0.27)	5.83 (0.28)	6.48 (0.14)	3.46 (0.28)	4.86 (1.11)	7.84 (1.03)
SIA	15.43 (1.37)	24.33 (1.65)	26.31 (1.01)	13.81 (1.23)	19.18 (4.67)	32.31 (4.84)
Summer						
$PM_{2.5}$	24.53 (−1.02)	45.93 (−2.89)	45.95 (−6.04)	23.42 (−1.52)	27.15 (3.31)	40.63 (2.58)
SO_4^{2-}	6.56 (−1.55)	10.52 (−3.86)	10.10 (−2.36)	6.11 (0.03)	9.78 (0.78)	10.53 (−1.25)
NO_3^-	5.97 (1.60)	13.94 (3.20)	11.23 (2.28)	3.99 (1.06)	4.45 (2.24)	10.12 (3.38)
NH_4^+	4.20 (−0.12)	8.01 (−0.52)	7.08 (−0.22)	3.47 (0.32)	4.96 (0.94)	6.89 (0.51)
SIA	16.75 (−0.05)	32.48 (−1.17)	28.43 (−0.28)	13.58 (1.42)	19.19 (3.97)	27.55 (2.66)
Fall						
$PM_{2.5}$	43.30 (0.70)	64.52 (−0.66)	69.06 (−6.59)	43.04 (−2.52)	42.64 (6.47)	76.81 (5.86)
SO_4^{2-}	7.43 (−1.55)	8.44 (−2.55)	11.36 (−2.48)	8.36 (−0.99)	11.6 (0.83)	12.03 (−2.31)
NO_3^-	14.13 (3.35)	22.47 (3.91)	20.75 (3.38)	10.33 (2.7)	10.95 (4.83)	28.14 (7.35)
NH_4^+	6.91 (0.40)	9.71 (0.18)	10.42 (0.07)	6.19 (0.42)	7.52 (1.71)	12.72 (1.27)
SIA	28.48 (2.21)	40.63 (1.54)	42.53 (0.98)	24.88 (2.14)	30.08 (7.39)	52.90 (6.32)
Annual						
$PM_{2.5}$	33.90 (0.38)	53.65 (−0.90)	58.11 (−6.11)	31.43 (−2.08)	39.06 (5.20)	63.15 (5.64)
SO_4^{2-}	5.80 (−1.08)	7.20 (−2.11)	8.88 (−1.91)	6.40 (−0.36)	10.10 (0.89)	9.77 (−1.29)
NO_3^-	10.17 (2.36)	17.17 (2.99)	16.80 (2.81)	6.89 (1.65)	9.25 (3.97)	21.20 (5.56)
NH_4^+	5.14 (0.29)	7.71 (0.08)	8.31 (0.12)	4.43 (0.36)	6.46 (1.48)	9.84 (1.13)
SIA	21.13 (1.58)	32.09 (0.96)	33.99 (1.03)	17.72 (1.67)	25.82 (6.35)	40.82 (5.42)

in that region; the increase over small regions around the city centers of the Pearl River Delta and Eastern Hubei can be attributed to the increase of ground-level SO_2 emissions (although total SO_2 emissions might have decreased), which favors local sulfate formation.

The concentrations of SIA increased in most of China, especially in the Sichuan Basin and Eastern Hubei. The largest increase in the Sichuan Basin results from simultaneous increase of sulfate and nitrate. The temporal trends elsewhere are determined by the competition of the nitrate increase and sulfate reduction. The increase of nitrate usually plays a dominating role. The annual average SIA concentrations increased by 1.58 $\mu g\ m^{-3}$, 0.97 $\mu g\ m^{-3}$, 1.03 $\mu g\ m^{-3}$, 1.67 $\mu g\ m^{-3}$, 6.36 $\mu g\ m^{-3}$, and 5.43 $\mu g\ m^{-3}$ in Eastern China, the North China Plain, the Yangtze River Delta, the Pearl River Delta, the Sichuan Basin, and Eastern Hubei respectively, as an effect solely of emission changes. The increase of SIA in most of China implies that the NO_X and NH_3 emissions shall be reduced simultaneously to mitigate the severe haze pollution in China. When considering the impact of meteorological conditions, the total changes of SIA are 1.38, 1.65, 1.26, 0.77, 9.29, and 5.90 $\mu g\ m^{-3}$, respectively, in the six regions above. The emission changes usually dominate the temporal trends of SIA concentrations. But in some regions the meteorological conditions also have considerable effects. The meteorological condition, in particular the changes of wind speed, enhanced the SIA concentrations in the Sichuan Basin and North China Plain, and mitigated the SIA concentration in the Pearl River Delta (see details in section 4 of the supplementary materials, available at stacks.iop.org/ERL/8/024031/mmedia).

The extinction coefficient (inversely proportional to visual range) is a measure of absorption and scattering of light.

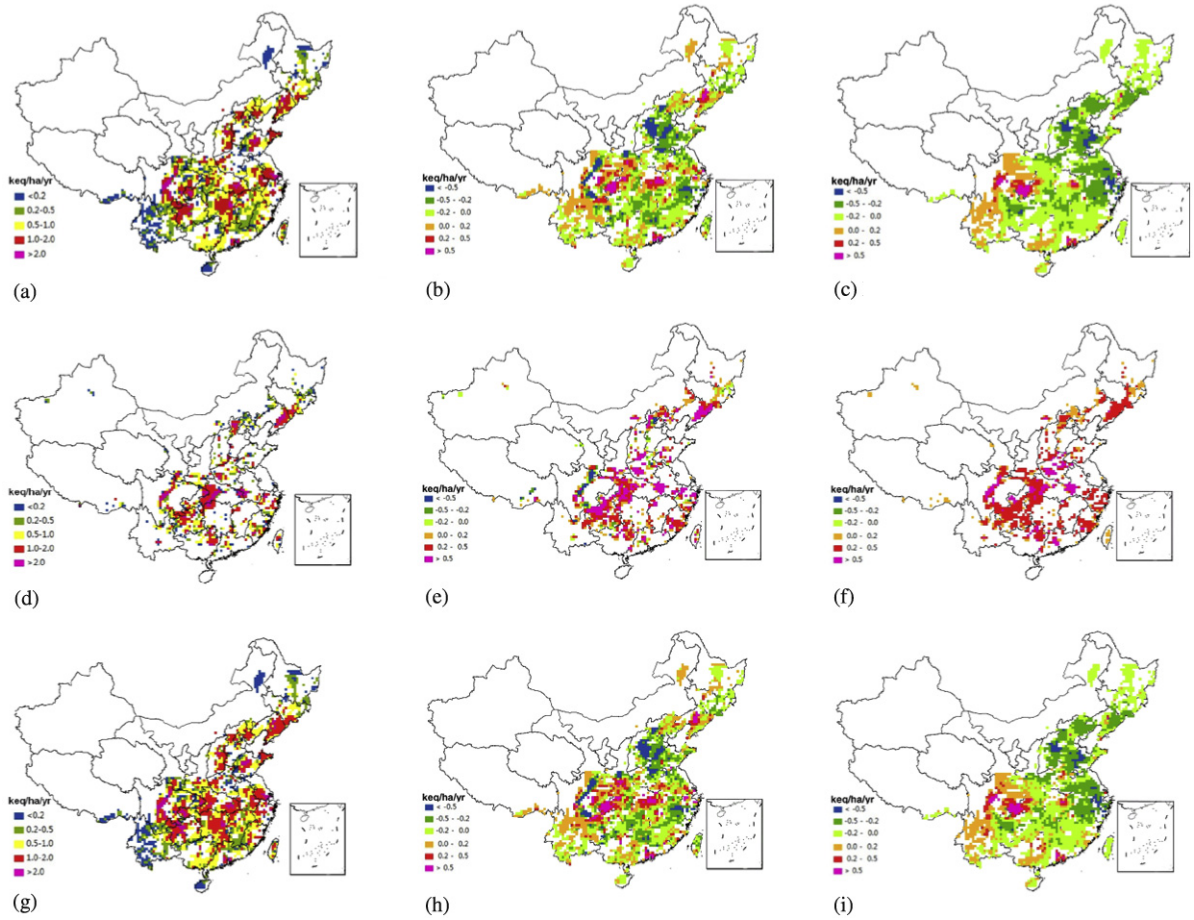

Figure 5. Exceedances of critical loads for acidification and nutrient nitrogen in 2010 (left column), the changes during 2005–2010 (2010 minus 2005, middle column), and the changes as an effect solely of emission changes (2010 minus 2005SENS, right column). Depositions simulated by CMAQ were compared with selected criteria, including $CL_{max}(S)$, $CL_{nut}(N)$, and CL(S). (a)–(c) Exceedance of $CL_{max}(S)$; (d)–(f) exceedance of $CL_{nut}(N)$; (g)–(i) exceedance of CL(S).

Therefore, it is frequently used as a comprehensive index of fine particulate matter pollution. As shown in figure 4, the extinction coefficient presented an upward tendency in most parts of China, especially in the Sichuan Basin and Eastern Hubei, as an effect solely of emission changes. The notable increase in the Sichuan Basin and Eastern Hubei is mainly attributed to the increase of SIA concentrations (see figure 4). The extinction coefficient declined slightly in the Yangtze River Delta and urban Beijing despite the slight increase of SIA concentration, attributable to the decrease of primary $PM_{2.5}$ (in particular BC/OC emissions, see figure S1 available at stacks.iop.org/ERL/8/024031/mmedia).

3.3. Effects on soil acidification

Soil acidification as well as soil eutrophication by S and N was evaluated by comparing the simulated S and N deposition and critical load maps. Figure 5 shows the critical load exceedance in 2010, and the changes during 2005–2010; table S9 (available at stacks.iop.org/ERL/8/024031/mmedia) summarizes the area percentage and amount of critical load exceedance in the studied regions. As an effect solely of emission changes, the areas exceeding $CL_{max}(S)$ and the total exceedance decreased by 5.5% and 13.8% respectively in China, mainly attributable to SO_2 emission reduction. Driven by the nation-wide increase of NO_X and NH_3, the areas exceeding CL(N) and the total exceedance increased by 21.8% and 45.8%, respectively.

When combining the acidification effect of N and S, the areas exceeding CL(S) and the total exceedance decreased by 4.1% and 10.3% respectively, as an effect solely of emission changes. As for the studied regions, the change rates of total exceedance of $CL_{max}(S)$ were −18.5%, −31.3%, −24.1%, 17.9%, 32.4%, and 0% in Eastern China, the North China Plain, the Yangtze River Delta, the Pearl River Delta, the Sichuan Basin, and Eastern Hubei respectively, while the corresponding change rates of the exceedance of CL(S) were −14.8%, −28.6%, −21.8%, 24.1%, 36.0%, and 2.5%, as an effect solely of emission changes. This indicates that the reduction of S deposition has played a dominating role in altering the critical load exceedance over the increase of N deposition. The increasing acidification effects of N slowed down the decline, or accelerated the increase of the exceedance of critical loads. It is also noted that the exceedance of critical loads decreased across the country, but increased in large parts of southern China, especially the

Sichuan Basin, the Pearl River Delta, and Eastern Hubei, where the soil acidification was the most serious. Therefore the SO_2 emission controls should be strengthened in southern China. In addition, a stringent multi-pollutant control strategy should be implemented in the Sichuan Basin and Eastern Hubei.

4. Conclusions

The change rates of China's anthropogenic emissions of SO_2, NO_X, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC and NH_3 were estimated to be −14.9%, 33.8%, −15.1%, −11.7%, −0.9%, −5.3%, 21.0%, and 10.4%, respectively during 2005–2010. The major driving forces of the decrease of SO_2 and particulate matter are large scale installation of FGD for power plants, and rapid evolution of dust removal equipment for power plants and the cement industry, respectively. The trends of emissions during the five years differed notably between regions, attributed to distinct growth rate of energy consumption, and different penetration of removal equipment.

During 2005–2010, the $PM_{2.5}$ concentration decreased by 2–17 $\mu g\ m^{-3}$ in most parts of the North China Plain, the Yangtze River Delta, and the Pearl River Delta, and increased by 4.5–16 $\mu g\ m^{-3}$ in most parts of the Sichuan Basin and Eastern Hubei, as an effect solely of emission changes. Different species of $PM_{2.5}$ presented diverse temporal trends. The changes of $PM_{2.5}$ emissions led to the decline of primary $PM_{2.5}$ concentrations in most parts of eastern China, except for the increase in Eastern Hubei. As an effect solely of emission changes, nitrate increased to some extent in most parts of China, and most evidently in Eastern Hubei and the Sichuan Basin. Sulfate concentrations decreased in most parts of Eastern China, with the largest reduction in the North China Plain, while they increased in the Sichuan Basin, and over small regions around the city centers of the Pearl River Delta and Eastern Hubei. The increase of nitrate is usually dominant over the reduction of sulfate, therefore SIA concentrations increased in most parts of China, with most evident increase in the Sichuan Basin and Eastern Hubei. The extinction coefficient presented an upward tendency in most parts of China, especially in the Sichuan Basin and Eastern Hubei. Combining the acidification effects of S and N, the areas exceeding CL(S) and the total exceedance decreased by 4.1% and 10.3% respectively, as an effect solely of emission changes. The reduction of S deposition played a dominant role in altering the critical load exceedance over the increase of N deposition.

The results of this study provide important implications for policy making. First, reductions of primary PM emissions have been the most effective control measures to mitigate particulate matter pollution during 2005–2010, especially for urban areas with large intensive emissions. We propose the controls of primary PM be strengthened in the future as substantial reduction potential remains. Second, SIA increased in most parts of China despite reductions in SO_2 emissions, leading to the deterioration of visibility, a major index of haze pollution. The NO_X and NH_3 emissions should be reduced simultaneously to mitigate the severe haze pollution in China. Third, this study demonstrated the need to implement different control policies in different regions. The reduction of SO_2 led to large reduction of sulfate in the North China Plain and the Yangtze River Delta. However, sulfate increased in at least part of the Sichuan Basin, Eastern Hubei, and the Pearl River Delta. In addition, the increase of NO_X and NH_3 induced the largest increase of nitrate in the Sichuan Basin and Eastern Hubei. Although the total exceedance of critical loads decreased across the country, the acidification effect was worse in large parts of the southern China, especially the Sichuan Basin, the Pearl River Delta, and Eastern Hubei, where the soil acidification was the most serious. The reduction of SO_2 emissions, especially ground-level SO_2 emissions favoring local sulfate formation, should be specifically strengthened in southern China (in particular the Sichuan Basin, Eastern Hubei, and the Pearl River Delta). NO_X emission reductions should be strengthened significantly in most of eastern China. A stringent multi-pollutant control strategy should be implemented in the Sichuan Basin and Eastern Hubei, which were characterized by increasing SIA concentrations, deteriorative visibility and soil acidification conditions.

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