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The effects of recent control policies on trends in emissions of anthropogenic atmospheric pollutants and CO_2 in China

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Abstract

To examine the effects of China's national policies of energy conservation and emission control during 2005–2010, inter-annual emission trends of gaseous pollutants, primary aerosols, and CO_2 are estimated with a bottom-up framework. The control measures led to improved energy efficiency and/or increased penetration of emission control devices at power plants and other important industrial sources, yielding reduced emission factors for all evaluated species except NO_x. The national emissions of anthropogenic SO_2, CO, and total primary PM (particulate matter) in 2010 are estimated to have been 89 %, 108 %, and 86 % of those in 2005, respectively, suggesting successful emission control of those species despite fast growth of the economy and energy consumption during the period. The emissions of NO_x and CO_2, however, are estimated to have increased by 48 % and 43 %, respectively, indicating that they remain largely determined by the growth of energy use, industrial production, and vehicle populations. Based on application of a Monte-Carlo framework, estimated uncertainties of SO_2 and PM emissions increased from 2005 to 2010, resulting mainly from weakly understood average SO_2 removal efficiency in flue gas desulfurization (FGD) systems in the power sector, and unclear changes in the penetration levels of dust collectors at industrial sources, respectively. While emission trends determined by bottom-up methods can be generally verified by observations from both ground stations and satellites, clear discrepancies exist for given regions and seasons, indicating a need for more accurate spatial and time distributions of emissions. Limitations of current emission control polices are analyzed based on the estimated emission trends. Compared with control of total PM, there are fewer gains in control of fine particles and carbonaceous aerosols, the PM forms most responsible for damages to public health and effects on radiative forcing. A decrease of alkaline base cations as primary PM that is much faster than that of SO_2 may have raised the acidification risks to ecosystems, indicating further control of acid precursors is required. Moreover, with relatively strict controls in developed urban areas, air pollution challenges have been expanding to less-developed neighboring

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regions. There is a great need in the future for multi-pollutant control strategies that combine recognition of diverse environmental impacts both in urban and rural areas with emission abatement of multiple species in concert.

1 Introduction

China suffers highly degraded air quality and related environmental impacts, due mainly to intensive fossil fuel consumption and rapid growth of the vehicle population. Based on satellite observations and chemical transport models, Eastern China has been found to have the highest concentrations of airborne fine particulate matter ($PM_{2.5}$) (van Donkelaar et al., 2010) and vertical column densities (VCD) of tropospheric NO_2 (Richter et al., 2005) in the world. Serious air pollution has caused huge public health damages particularly in mega cities (Parrish and Zhu, 2009) and has also threatened ecosystems. The highest acidity of precipitation in the world has been observed in south and southwest China (Larssen et al., 2006). A number of analysts have estimated swift increases in anthropogenic emissions, the main cause of China's severe air pollution, during the early-2000s (Ohara et al., 2007; Zhang et al., 2007; Lu et al., 2010; Lei et al., 2011a). According to the GAINS model developed by the International Institute for Applied Systems Analysis (IIASA), China accounted for 24 %, 14 %, 25 %, and 27 % of global emissions of SO_2, NO_x, black carbon (BC), and organic carbon (OC) in 2000, respectively (Cofala et al., 2007; Klimont et al., 2009). Although considerable uncertainties exist (Zhao et al., 2011a), estimates of China's total SO_2 and PM emissions are much more than those of the US or Europe (Zhao et al., 2009, 2011b).

Under heavy pressure to improve urban air quality, reduce regional air pollution, and limit carbon emissions, China's government has implemented a comprehensive national policy strategy of energy conservation and emission reduction since 2005. Its goal is to shift the country's development mode from one dependent on intense fossil energy inputs with consequent high emissions to a more resource-efficient and

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environment-friendly alternative. Stringent, compulsory measures to improve energy efficiency and control emissions have been required at many major source types, targeting a range of atmospheric pollutants. These measures include: replacement of small and inefficient plants or boilers with larger, energy-efficient ones in the power sector and certain heavy industrial sectors including cement production; installation of flue gas desulfurization (FGD) systems at all newly built thermal power units; application of more stringent emission standards in cement production; and staged implementation of tighter emission standards on vehicles. Evidence of success of these measures since 2005 has been confirmed in different ways. For example, improved combustion efficiency (and thus energy efficiency) is indicated in an increasing inter-annual trend in the ratio of CO_2 to CO, because CO_2 results from complete combustion and CO from incomplete combustion of carbon fuels. The trend has been observed instrumentally in air masses representative of north China emissions at a rural site north of Beijing (Wang et al., 2010a) and also indicated in bottom-up emission inventory studies (Zhao et al., 2012a, b). Reductions in regional and national SO_2 are similarly indicated both by observations, from satellites (Li et al., 2010), and by bottom-up emission trends based on fast-track energy statistics for recent years (Lu et al., 2010, 2011). Some studies also assess the effects of policies on other species including NO_x (Lin et al., 2011; Wang et al., 2012) and primary and secondary aerosols (Lu et al., 2011; Lin et al., 2010).

While studies that focus on individual species and/or source types are essential to building fundamental knowledge of atmospheric processes in China, they contribute piecemeal to understanding of China's atmospheric environment as an integrated system of sources and sinks of diverse reactive species. Interaction of emission trends, however, is often as significant to environmental outcomes of interest as the trend in any one species or source category taken alone. This is critical not only to understanding physical, chemical, and biological cycles but also to evaluating and informing the development of broadly effective air quality and climate protection policy strategies. The current study meets a need for comprehensive consideration of emission trends

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of different atmospheric pollutants and analyses of the main drivers of these trends. It focuses on 2005 through the end of China's 11th Five Year Plan in 2010, a discrete period of sharply heightened regulatory action in emission control.

The study analyzes the effects of recently implemented control measures on the inter-annual trends, sector and spatial distributions, and uncertainties of China's anthropogenic emissions. Incorporating the latest information from domestic field measurements and investigations, the trends of emission factors (i.e. the emission levels per unit consumption of energy or industrial production) for different kinds of pollutants from 2005 to 2010 are developed by sector and technology. Based on a bottom-up framework, provincial and national emissions are estimated for 2005–2010, indicating the effectiveness of improved energy efficiency and emission control efforts during those years. The uncertainties of emissions in 2010 are quantified statistically using Monte-Carlo simulation, which was developed and applied previously to emissions for 2005 (Zhao et al., 2011a). The causes of discrepancies between the uncertainty results for the two years are evaluated. To understand the effects of varied emissions on urban and regional air quality, available observations from ground measurements and satellites are reviewed and compared with the bottom-up emissions for corresponding time periods and locations in China. Limitations of current controls on diverse environmental impacts are analyzed based on the estimated emission trends for different species and regions, recommending a more comprehensive, multi-pollutant scope as China develops its future strategies in control of atmospheric pollutants.

2 Methods

2.1 The framework of the emission inventory

The methods of developing a bottom-up emission inventory are detailed in previous studies (Zhao et al., 2011a, 2012a). Figure S1 in the Supplement shows the categorical structure of sources of China's anthropogenic atmospheric emissions. At the

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largest scale, sources fall into four main sector categories: coal-fired power plants (CPP), all other industry (IND), transportation (TRA, including on-road and non-road subcategories), and the residential and commercial sector (RES, including fossil fuel and biomass combustion subcategories). IND is further divided into cement production (CEM), iron and steel plants (ISP), other industrial boilers (OIB), and other non-combustion processes (PRO), reflecting the structure of available data. Species considered in this work include gaseous pollutants (SO_2, NO_x, and CO), PM according to different size classes and chemical species (Total Suspended Particles (TSP), PM_{10}, $PM_{2.5}$, BC, Ca, and Mg), and the greenhouse gas CO_2. Annual anthropogenic emissions of these pollutants for 2005–2010 are estimated both by province and sector and then aggregated to the national level, using Eq. (1):

$$E_{i,j,t} = \sum_{k} \sum_{m} \sum_{n} \mathrm{AL}_{j,k,m,n,t} \times \mathrm{EF}_{i,j,k,m,t} \times R_{j,k,m,n,t} \times (1 - \eta_{i,n,t}) \quad (1)$$

where i, j, k, m, n and t stand for species, province, sector, fuel type, emission control technology and year, respectively; AL is the activity level, either energy consumption or industrial production; EF is the unabated emission factor; R is the penetration rate of emission control technology; and η is the removal efficiency.

2.2 Activity levels

Activity levels for 2005–2010 are compiled annually by sector from various data sources. The fossil fuel consumption and industrial production at provincial level are obtained from Chinese official energy (NBS, 2011a) and industrial economy statistics (NBS, 2011b). To avoid double counting, the fuel consumption by OIB is estimated by subtracting the fuel consumed by CEM, ISP and PRO from fuel consumed by total industry (Zhao et al., 2012a). The activity levels of biofuel use are taken from official statistics (NBS, 2011a). The biomass combusted in open fields is calculated as a product of grain production, waste-to-grain ratio, and the percentage of residual material burned in the field. Details are described in previous studies (Zhao et al., 2011a,

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2012a). From 2006 to 2010, the share of primary energy consumption by coal decreased from 74 % to 70 %, indicating a shift towards cleaner energy sources.

It should be noted that China's provincial and national energy statistics are often inconsistent. As shown in Fig. 1, the annual coal consumption levels reported officially for the entire country range 13–16 % lower than the sum of provincial consumption in each year from 2005 to 2010. On a sector basis, the differences reach 20–30 % for industry and exceed 30 % for the residential and commercial sector, while the difference for the power sector is relatively small (∼ 2 %). Akimoto et al. (2006) found China's provincial-level statistics to be within the uncertainty bounds of the satellite record of NO_2 over the country while the national-level statistics were not, and advised against use of the latter for emission inventories in China. Although this conclusion was drawn for 1996–2002, the differences between the national and aggregated provincial statistics have not diminished in following years and subsequent studies of China's emissions, many including comparisons to observations by ground stations, aircraft, or satellites, have held to the same conclusion (Streets et al. 2006; Zhang et al., 2007; Zhao et al., 2012b). We likewise believe that the provincial statistics are more accurate than the national ones.

For transportation, Chinese official statistics reflect only fuel used in commercial activities, and thus cannot be applied directly. In this work, on-road vehicles are classified into light-duty gasoline vehicles (LDGV), light-duty gasoline trucks (LDGT), light-duty diesel trucks (LDDT), heavy-duty gasoline vehicles (HDGV), heavy-duty diesel vehicles (HDDV), and motorcycles (MC). The oil consumption by each vehicle type is calculated as the product of the vehicle population, annual average mileage traveled per vehicle, and average fuel economy of the corresponding type (He et al., 2005). Detailed methods and data sources are provided in Zhao et al. (2012a). For non-road sources including railway, waterway, rural vehicles, and construction equipment, the fuel consumption in 2005 is taken from Zhang et al. (2008), while those for 2006–2010 are scaled by province according to the growth of passenger and freight traffic by rail and

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shipping, and the total growth of agricultural and construction equipment. All those data are obtained from official statistics (NBS, 2011c).

3 Evolution of emission factors

Driven mainly by official Chinese policies, the penetration levels of different energy efficiency technologies and emission control devices shifted considerably at the national level from 2005 to 2010, leading to strong changes in emission factors as clearly illustrated in Fig. 2. Details by source are discussed as follows. During the period of interest, the mass fractions of chemical species (BC, OC, Ca and Mg) in PM emissions are assumed unchanged, taken directly from Zhao et al. (2011a) and Zhu et al. (2004) by source.

3.1 Coal-fired power plants

Coal-fired power plants were targeted for the most stringent emission controls during 2005–2010, particularly for SO_2. According to a unit-based dataset of coal-fired power plants over the country (Zhao et al., 2008), the FGD penetration rate increased from 13 % in 2005 to 86 % in 2010, and the share of the units equal to or larger than 300MW rose from 51 % in 2005 to 78 % in 2010, as shown in Fig. 2a. Based on an unpublished official survey, the national average removal efficiency of FGD is set at 75 % in this study, resulting in a 61 % reduction in the SO_2 emission factor for the entire coal-fired power sector over 2005–2010. Fast growth of large power units with higher energy efficiency and advanced PM control devices like electrostatic precipitators (ESP) or fabric filters (FF) reduced the emission factors of PM and CO as well. Based on an emission factor database reported by Zhao et al. (2010, 2012a), the emission factors of TSP, PM_{10}, $PM_{2.5}$, and CO are, respectively estimated to have decreased 60 %, 55 %, 46 %, and 31 % from 2005 to 2010. (Note that increased wet-FGD also helped to reduce PM emissions due to its ancillary benefit on PM removal, Zhao et al., 2010).

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On the other hand, however, the emission factors of NO_x and CO_2 for power plants varied little from 2005 to 2010, due to the limited addition of selective catalytic/non-catalytic reduction (SCR/SNCR) and the experimental state of carbon capture and storage technologies. It should be noted that the emission factors here are expressed as the mass of emitted pollutants per unit consumption of coal. If evaluated as pollutants per unit of generated electricity, the emission factors of NO_x and CO_2 declined by 12 % and 19 %, respectively, resulting mainly from the improved energy efficiency of coal-fired power generation during the years of analysis.

3.2 Cement production

Cement kiln technologies in China include shaft, precalciner, and other rotary kilns. As shown in Fig. 2b, the penetration of precalciner kilns, the most energy-efficient technology, increased from 44 % to 81 % from 2005 to 2010, while that of shaft kilns declined from 49 % to 16 %, according to official statistics. Emission factors of cement production by technology have been compiled by Lei et al. (2011b). Newly built precalciner kilns with ESP or FF lead to reduced emission factors for PM and CO. However, such benefits are accompanied by an increased NO_x emission factor, because the higher operational temperatures and more automated air-flow systems of precalciner kilns increase emissions of NO_x compared to shaft kilns. Combining the emission factors by Lei et al. (2011b) and changes in penetration of different kiln types, the emission factors (expressed as pollutants per unit of cement production) for SO_2, TSP, PM_{10}, $PM_{2.5}$, and CO are estimated to have declined by 32 %, 72 %, 69 %, 64 %, and 55 %, respectively, during 2005–2010, while that for NO_x increased by 31 %. Since CO_2 is generated mainly from the non-combustion process of carbonate calcination (Zhao et al., 2012b), the technology changes in cement production yielded little mitigation of CO_2 emissions.

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3.3 Iron and steel production

The iron and steel industry employs the following processes: coking, sintering, pig iron making (in blast furnaces), steel making (nearly 90 % of which is in basic oxygen furnaces), and casting. SO_2 and NO_x come mainly from the sintering process, and those emission factors are assumed unchanged during 2005–2010 given no new control requirements.

Based on national statistics (CISA, 2011), the share of coke produced in machinery coking ovens (versus modified indigenous ovens with poor technology and manual operation) increased from 82 % in 2005 to 86 % in 2010, reaching a peak of 91 % in 2007. Due to improved use of waste heat, the release ratios of machinery coke oven gas declined from 5.7 % to 1.4 % between 2005 and 2010. The release ratio of blast furnace flue gas in the making of pig iron dropped from 8.4 % to 5.0 % in the same period, and the recycled flue gas in basic oxygen furnaces increased from 60 to 79 $N m^3 t^{-1}$-steel (CISA, 2011). Those improvements made the emission factors of PM, CO, and CO_2 for the combined processes (expressed as pollutants per unit of steel production) decline by 39 %, 44 %, and 18 %, respectively, as shown in Fig. 2c.

3.4 Transportation

Since 1999, staged emission standards (Stage I–IV, equivalent to Euro I–IV) for new on-road vehicles have been implemented nationwide, with earlier implementation in Beijing than in other provinces. The fleet compositions by control stages for 2005–2010 are determined based on reported annual newvehicle registrations (NBS, 2011c) and the retirement of old vehicles based on assumed vehicle lifetimes in China by type. The average lifetimes of light-duty vehicles, light-duty trucks, and heavy-duty trucks are assumed to be 15, 8, and 10 yr, respectively, based on previous studies (He et al., 2005; Lei, 2008; Huo et al., 2012)

As summarized in Tables S1 and S2 in the Supplement, prior measurements of emission factors of NO_x and $PM_{2.5}$ for vehicles in China by type and control stage were

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thoroughly investigated in this work including on-road tests, engine tests, carbon balance calculations, and remote sensing (see also the database for CO by Zhao et al., 2012a). Results of on-road tests with advanced measurement technologies (e.g., He et al. (2010) using SEMTECH-D and Oliver (2008) using OBS-2200) are given preference to calculated vehicle emissions. If two or more studies consider the same combination of vehicle type and control stage, the emission factors used here are calculated as the average of the original data weighted by the sampling size. Due to few on-road tests of LDGT, data from roadside remote sensing (Guo et al., 2007) are applied for this vehicle type. With almost no measurements for HDGV or MC by control stage, the standard limits of stage I-II are relied upon, and typical fuel economies (2.7 l-fuel/100 km for MC, He et al. (2005), and 250 g-fuel kW h^{-1} for heavy-duty engines, Chen et al. (2008); MIIT, (2010)) are applied to convert the standard limits to fuel-based values. The same assumption is also applied for most non-road sources, except rural vehicles (RV), for which emission factors are taken from on-vehicle tests using SEMTECH-D by Yao et al. (2011). As shown in Fig. 2d, the nationwide emission factor levels of NO_x, $PM_{2.5}$, and CO are estimated to have declined by 44 %, 41 %, and 52 %, respectively for LDGV, attributed to implementation of the staged regulations. The control effects for MC, HDDV, and rural vehicles (Fig. 2e–g) are less, particularly for NO_x, and indeed NO_x emission factors for rural vehicles are estimated to have increased. The reasons include (1) slower penetrations of new, lower-emission MC and rural vehicle technologies than that of LDGV, and (2) higher NO_x emission rates of diesel engines under recently implemented regulations.

3.5 Other industrial boilers, processes, and residential and commercial combustion

For industrial boilers and processes, the primary concern is changes of emission factors for PM, resulting from varied penetration levels of different dust collector technologies. Such information is available for 2005 from investigation by Lei et al. (2011a), but very few data can be found for subsequent years. In this work, we assume that new

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emission sources (reflected by the annual net growth of energy consumption or industrial production) applied the most advanced dust collectors that have already been deployed in the sector, reflecting the effect of national policies to foster energy conservation and emission reduction during 2006–2010. For example, the PM emission factors are estimated to have decreased by over 40 % in nonferrous metallurgy, attributed to increased application of FF (Fig. 2h). For brick making, the share of solid clay bricks in China is estimated to have declined from 86 % to 40 % from 2005 to 2010 (Xu and Wang, 2007; Zhou, 2009; Zhao et al., 2012a). The emission factors (expressed as pollutants per unit of brick production) declined by 43 %, 28 %, 16 % and 27 % for TSP, PM_{10}, $PM_{2.5}$ and CO, respectively (Fig. 2i). To examine the sensitivity of PM emissions to the estimated trends of dust collector penetration, we also make an additional case, in which the application rates of dust collectors for industrial boilers and processes are kept unchanged from 2005 to 2010. Comparison of emissions between the two cases is given in Sect. 4.3.

For the residential and commercial sector, very little progress in emission control is believed to have occurred during 2005–2010 in either fossil fuel consumption or biofuel/biomass burning. Emission factors for different species are taken from the database compiled by Zhao et al. (2011a, 2012a, b).

4 Results

4.1 Inter-annual emission trends by sector and province

The national emissions from 2005 to 2010 for different species are illustrated by sector in Fig. 3. The SO_2 emissions are estimated to have decreased from 31.1 million metric tons (Mt) in 2005 to 27.7 Mt in 2010, with a peak value of 32.1 Mt in 2006 (Fig. 3a). It is particularly notable that the emissions from coal-fired power plants declined from 16.3 to 9.2 Mt, and the sector share of total emissions from 52 % to 33 %, resulting mainly from the swift increase of FGD systems during 2005–2010. The SO_2 emissions

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of other industries, however, are estimated to have increased by 32 %, accounting for over half of total national emissions in 2010; this is attributed to the dramatic growth of industrial production and energy consumption. This result spotlights the industry sector as a key target for future SO_2 emission controls, not just power plants. In contrast to SO_2, national NO_x emissions increased dramatically, by 48 % to 29.0 Mt in 2010, attributed to swift growth of energy consumption and limited control measures (Fig. 3b). Specifically, emissions from cement and on-road vehicles are estimated to have increased 131 % and 68 %, respectively. Expanded application of precalciner kilns, while helping to control emissions of other pollutants, actually worsened the NO_x problem. The implementation of staged emission regulation of on-road vehicles could not keep pace with rapid growth of vehicle populations in recent years, and thus failed to prevent emissions from rising overall in this subsector. Moreover, NO_x emissions from residential combustion of fossil fuels also went up despite decreased coal consumption of that sector, although the share remained small. This resulted mainly from rising use of liquid and natural gas fuels, which emit much less SO_2 and particles than coal combustion but relatively more NO_x. All of these facts suggest that NO_x emission control will be a huge challenge for China in upcoming years. Regarding CO emissions, a much reduced growth rate was found for 2005–2010, reflecting the benefits of improved energy efficiency in recent years (Fig. 3c).

As shown in Fig. 3d–j, the emissions of PM of different sizes and chemical species are estimated to have declined to varied extent and Ca is the species with biggest emission abatement, by 25 % from 2005 to 2010. Attributed to the penetration of improved production technologies and dust collectors, national emissions of TSP are estimated to have decreased from 33.2 Mt in 2005 to 28.7 Mt in 2010, of PM_{10} from 18.9 to 16.9 Mt, and of $PM_{2.5}$ from 13.0 to 12.3 Mt. Emission control in cement production is found to have been highly effective, with PM emissions of different sizes and chemical species reduced around 50 %. The cement-making share of total emissions decreased from 22 % to 13 % for TSP and from 52 % to 34 % for Ca. In contrast, emissions of different PM categories from iron and steel plants are estimated to have increased

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24 %–39 % from 2005 to 2010, attributed mainly to huge growth of steel production. The annual variations of national emissions of primary carbonaceous aerosols ranged less than 10 %, exhibiting slight declines over the six-year period, and the source contributions were relatively stable. The combustion of fossil fuel and biofuel/biomass in residential and commercial activities accounted, respectively for around 20 % and 30 % for BC emissions, and 30 % and 50 % for OC emissions.

CO_2 emissions are estimated to have increased from 7126 Mt in 2005 to 10,174 Mt in 2010, with an annual growth rate of 7.4 % (Fig. 3k). Note that these totals include emissions from biofuel/biomass burning, which are omitted in many CO_2 inventories. Regarding sector distributions, emissions from power plants, industry, and transportation increased by 44 %, 60 %, and 64 % during 2005–2010, and the three sectors accounted for 32 %, 41 %, and 8 % of national total emissions in 2010, respectively. Meanwhile emissions from residential and commercial activities declined slightly.

Provided in Table 1 are the emissions of different species at the provincial level in 2010. The developed regions of East, North-Central, and South-Central China (as defined in Table 1 and illustrated in Fig. S2 in the Supplement) are estimated to account together for around 70 % of total national emissions of all concerned species in 2010. Notably, the SO_2 emissions in North-Central and East China declined by 12 % and 20 %, respectively during 2005–2010, indicating considerable achievements of emission control in these heavily polluted regions. The analogous reductions for Northeast and Southwest China, however, are merely 2 %. In the northeast, where coals with low sulfur content (such as lignite) are widely used, the SO_2 emissions from coal combustion were relatively small and thus there is less need for FGD at existing power units than in other areas. In contrast, the sulfur content of coals in the southwest are extremely high and many power units had FGD already installed by 2005, leading to limited potential for further reduction of emissions after 2005. Among all the regions, the northwest had the fastest growth of NO_x and CO_2 emissions, indicating a relatively rapid increase of economic activity and energy consumption in that less-developed area, although its shares of total national emissions remained small.

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4.2 Comparisons with other studies

Figure 3 also shows national emission trends estimated by other studies for different species since 2000. Only studies with emissions for multiple years are selected for comparison with current results. Along with CO_2, CO and NO_x are the species with monotonic emission increases during 2000–2010. Ohara et al. (2007) estimated an average annual growth of 4.9 % for CO from 2000 to 2003, while the value for 2005–2010 indicated by this work is 1.6 %, reflecting improved energy efficiency and emission control of CO after 2005. For NO_x, Zhang et al. (2007) and Ohara et al. (2007) estimated average annual growth rates of 10.2 % and 8.9 % for 2000–2004 and 2000–2003, respectively, close to 8.1 % for 2005–2010 by this work, all suggesting limited overall effectiveness of NO_x abatement policies to date.

For SO_2 and PM of different size classes, combining results of other studies and this work shows that the growth of emissions in the early-2000s have been gradually reversed in recent years. Generally consistent trends of SO_2 emissions from 2005 to 2010 were indicated by Lu et al. (2011) and this work, although the current estimates are consistently lower. The discrepancy is likely attributable mainly to applications of different FGD removal efficiencies in the two studies. Lacking estimates in more recent years for comparison, the emissions of TSP, PM_{10}, and $PM_{2.5}$ for 2005 in Lei et al. (2011a) and this work are close. Regarding chemical species, however, the different studies are relatively inconsistent. For example, decreasing trends of carbonaceous aerosol emissions were given by Klimont et al. (2009) for 2000–2005, while increasing trends were suggested by other studies. Even with similar inter-annual trends, Zhang et al. (2009) estimated higher BC emissions from 2001 to 2006 than Lu et al. (2011), but much lower OC. The carbonaceous aerosol emissions estimated by this work are close to or higher than those of Klimont et al. (2009) and Lei et al. (2011a), but clearly lower than those of Lu et al. (2011). Although Lu et al. (2011) included emissions from burning of forest and savanna that is omitted by this work, the contribution of that source was very small and cannot fully explain the differences. The divergent results indicate

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large uncertainties for estimates of emissions of chemical species of PM, particularly from the burning of fossil fuel and biofuel/biomass in residential activities, helping to motivate the uncertainty analysis of bottom-up emissions described in Sect. 4.3.

Compared to gaseous and PM pollutants, the disparities in CO_2 emissions between different studies are smaller, as shown in Fig. 3k. Note the results of other studies and this work for total CO_2 emissions cannot be directly compared because the emissions estimated by the US Carbon Dioxide Information Analysis Center (CDIAC, http://cdiac.ornl.gov/ftp/trends/emissions/), the PBL Netherlands Environmental Assessment Agency (PBL, http://www.pbl.nl/sites/default/files/cms/publicaties/500212001.pdf), the US Energy Information Administration (USEIA, http://www.eia.gov/cfapps/ipdbproject/IEDIndex3.cfm?tid=90&pid=44&aid=8), and the International Energy Agency (IEA, http://www.iea.org/publications/freepublications/publication/name,4010,en.html) include only those from fossil fuel combustion and sometimes cement production, while this work also includes emissions from other non-combustion industrial processes and biofuel/biomass burning (see details in Zhao et al., 2012b). (Note that USEIA and IEA do not report cement process emissions, and the estimates by CDIAC are added to USEIA and IEA fossil fuel emissions in Fig. 3k to facilitate some comparison of the studies by equivalent inclusion of source types.) Even excluding the emissions from biomass/biofuel, discrepancy remains between this work and other studies. The CO_2 emissions for 2005–2010 estimated by us are generally higher than those by most of other studies, attributed mainly to the application of a domestic CO_2 emission factor database (Zhao et al., 2012b) and the use of provincial-level energy data in this work. Moreover, the non-combustion CO_2 emissions from industrial processes, such as the emissions from primary aluminum production due to the consumption of carbon anodes in the reaction to convert aluminum oxide to aluminum metal, are also included in this work, although the amount from those sources is relatively small. From 2000 to 2005, the annual growth rates of CO_2 emissions from fossil fuel combustion plus cement production are estimated to have ranged from 10.2 % by PBL to 13.9 % by USEIA, while growth in annual emissions

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declined to 8.5 % from 2005 to 2010 based on the current study. This suggests the effects both of slowed economic development and of improved energy efficiency for the country during 2005–2010.

4.3 Uncertainty analysis of emissions in 2010

The uncertainties of emissions for different species in 2010 are estimated using a Monte-Carlo framework developed by Zhao et al. (2011a). The principles of determining the uncertainties of all the parameters, expressed as the probability distribution function (PDF), were described in detail by Zhao et al. (2011a, 2012a, b). With updated PDFs for 2010, a total of 10 000 simulations are performed and the uncertainties of emissions, expressed as 95 % confidence intervals (CIs) around the central estimates, are generated by sector and species, as shown in Table 2. The parameters most significant in determining the uncertainties of emissions, judged by their contribution to the variance, are also identified by the Monte-Carlo simulations and are shown in Table 3.

The uncertainties of China's anthropogenic emissions of gaseous pollutants SO_2, NO_x, and CO in 2010 are estimated to be −15 % to +26 %, −15 % to +34 %, and −18 % to +42 %, respectively; those for primary aerosols TSP, PM_{10}, $PM_{2.5}$, BC, OC, Ca, and Mg are −21 % to +52 %, −14 % to +51 %, 14 % to +59 %, −25 % to +131 %, −39 % to +122 %, −75 % to +76 %, and −46 % to +151 %, respectively; and that of the greenhouse gas CO_2 is 10 % to +9 %. In general the results of the uncertainty analyses are similar to those for 2005 (Zhao et al., 2011a, 2012a, b) in that: (1) the uncertainties of emissions of gaseous pollutants are smaller than those of primary aerosols; (2) among sectors, the uncertainties associated with residential and commercial activities are the largest; and (3) in most cases, parameters related with emission factors contribute most to the uncertainties of atmospheric pollutants, while activity levels are more significant to uncertainties of CO_2, except for the industry sector (Table 3).

Some of the estimated uncertainties for given sectors and species change significantly from 2005 to 2010. First, the uncertainty of SO_2 emissions from power plants rises from −16 % to +21 % in 2005 to −27 % to +59 % in 2010. The uncertainty of

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total national SO_2 emissions is accordingly larger in 2010, at −15 % to +26 %, than that in 2005, at −14 % to +13 %. This results mainly from the swift penetration of FGD systems, of which the SO_2 removal efficiency may vary nationally and is poorly quantified. Although designed FGD removal efficiencies can reach 95 %, the installed FGD systems are not believed to have achieved such high values because they were not always operated fully (Xu et al., 2009) FGD systems operated less consistently than expected reduces the anticipated benefits of SO_2 emission control on power plants, and increases the uncertainty of the SO_2 emission inventory for recent years as well. As shown in Table 3, the removal efficiency of FGD contributed 72 % to the variance of SO_2 emissions from power plants, and more field investigations are thus necessary to better quantify typical removal efficiencies of FGD systems. Second, the uncertainties of NO_x and PM emissions from transportation declined from 2005 to 2010. As staged emissions regulations of vehicles have been implemented since 2005, more field measurements on the emission factors of on-road and rural vehicles designed for different control standards have been conducted and reported (e.g., He et al., 2010). The increased sampling sizes and improved measurement methods of those studies have helped considerably to reduce the uncertainties of vehicle emission factors. Third, the uncertainties of PM emissions of different particle size classes have increased, particularly for TSP. This is attributed mainly to penetration levels of dust collectors for given industrial processes during 2005–2010 that must be assumed, given insufficient field data. As shown in Table 3, the penetration level of dust collectors for lime production is the most significant parameter contributing to the uncertainty of TSP emissions from the industry sector for 2010, while the analogous parameters for 2005 are the emission factors for non-combustion cement processing and grate boiler combustion (Zhao et al., 2011a). Compared to fine particles, the uncertainty of TSP emissions increased more significantly, reaching −21 % to +52 % in 2010, exceeding those of PM_{10} or $PM_{2.5}$. If no technology improvement of dust collectors is assumed for industrial processes, as described in Sect. 3.5, the national TSP emissions are reestimated at 39.6 Mt, i.e. 38 % higher than the original estimate. The analogous values for PM_{10}

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and $PM_{2.5}$ would be 16 % and 12 % higher, respectively, much smaller than that of TSP; and those for BC and OC less than 5 %. These results indicate that the uncertainty of dust collector penetrations in industrial sources have fewer impacts on emission estimates for fine particles and carbonaceous aerosols than for TSP, since the benefits of those technologies on PM control are less for fine particles than coarse ones, and result particularly from certain industrial sources with relatively large emission fractions of coarse particles such as lime and brick production.

4.4 Comparisons with ground observation

The inter-annual trends of emissions are compared with those derived from observations. During the study period, SO_2, NO_2, and PM_{10} were criteria pollutants for which concentrations were reported for 113 "key" cities by the Ministry of Environmental Protection (MEP) of China, based on their large populations, developed economies, and/or heavy pollution levels. Figure 4 shows comparisons of relative inter-annual variations between the estimated emissions and officially reported concentrations of monitored cities by region from 2005 to 2010, normalized to 2005 levels. The maps illustrate the changes in emissions for provinces and concentrations for cities, while the panels around the maps illustrate the trends in emissions for broader regions (as defined in Table 1) and concentrations for cities in those regions.

The inter-annual trends of emissions and observed concentrations are generally consistent with each other, although there are also some discrepancies for given species and regions. For SO_2, as shown in Fig. 4a, similar declining trends for emissions and concentrations are found for North-Central Northeast, East, and South-Central China between 2005 and 2010, confirming the effects of national policy on SO_2 control. However, increased SO_2 concentrations are found for several cities in East China while decreased provincial emissions are estimated. This is possibly because (1) some local sources like small coal stoves which are missed in the energy statistics and thus omitted in the emission inventory framework grew in recent years; and/or (2) the operation of FGD was actually poorer than expected in some specific areas. In contrast,

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decreased concentrations were observed with increased emissions between 2005 and 2010 for some provinces in Southwest China. As discussed in Sect. 4.1, the limited potential expansion of FGD for power plants in the region could not significantly reduce SO_2. The declining concentrations thus imply that some local measures of SO_2 control, such as coal washing may not be well characterized by this work. The disagreements of emission and concentration trends in certain areas indicate the pressing needs of more detailed investigations on emission characteristics and control measures at local scales.

As shown in Fig. 4b, NO_x emissions are estimated to have increased much faster than NO_2 concentrations from 2005 to 2010 in all regions. Indeed decreased urban average NO_2 concentrations were reported in North-Central and Northeast China, shown in the line plots for those regions. The relatively large gaps between regional emissions and observed urban concentrations likely reflect that NO_x (the estimated emission species) includes NO as well as NO_2 (the observed concentration species). Complex local NO_x sources contributed higher levels of NO in urban areas than rural or remote ones, though it is omitted in measurements of urban NO_2 concentrations. Satellite observation at larger spatial scales will be used to further examine the inter-annual trends of estimated emissions, as described in Sect. 4.5.

The inter-annual trends of emissions and concentrations for PM_{10} match well for most regions, as shown in Fig. 4c. In North-Central China, faster reduction of urban PM_{10} concentrations is found than that of estimated provincial emissions. This is probably attributable to gradually implemented control measures (e.g., road paving and afforestation) on fugitive dust from construction sites, unpaved roads, or natural sources. These are not included in the current emission inventory. Similar to SO_2, there are some cities in East China with increased observed PM_{10} concentrations despite estimated declines in regional emissions from 2005 to 2010, suggesting that some local small industrial and/or residential and commercial sources, which generate PM emissions from coal or biomass combustion, probably increased though they are not well accounted for in recent years.

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The government has not systematically reported urban concentrations of other species including BC, OC, CO, or CO_2. As summarized in Table 4, however, a number of independent studies have been conducted in different periods and locations in China to observe ambient BC, OC, CO, and CO_2 levels. Correlation slopes between some of these observed species, e.g., dBC/dCO, dOC/dBC, and dCO_2/dCO, have been estimated to approximate emissions. To test the accuracy of the bottom-up emissions in this work, the ratios of BC to CO emissions ($\mu g\,m^{-3}\,ppbv^{-1}$, note that $1\,\mu g\,m^{-3}\,ppbv^{-1} = 1.25\,kt\,kt^{-1}$) and those of OC to BC ($\mu g\,m^{-3}/\mu g\,m^{-3}$) are calculated for the corresponding periods and locations in which the observations were conducted (see also an analysis of CO_2/CO ratios in Zhao et al., 2012a). Monthly variations of emissions are generated following the methods of Zhang et al. (2009), and are used in the calculation of emission ratios for periods matching the observations. In one case, Andreae et al. (2008), the observations were conducted outside of the study period of this work, in October-November 2004, so emissions for the corresponding months in 2005 are instead applied here for comparison. The ratios of emissions are determined at two spatial scales: a local scale, based on provincial emissions, and a multi-province regional scale, as defined in Table 1. One exception to this definition is for the comparison with Kondo et al. (2011), which reported the results of dBC/dCO observed at Cape Hedo on Okinawa Island, Japan, representing emissions exported from East China. In this case the local emission ratio is calculated based on emissions in East China while the regional one is based on those for the whole country.

As shown in Table 4, in most cases the BC/CO ratios from estimated emissions are higher than the correlation slopes from observations in China. However, once the influence of wet deposition and atmospheric chemistry processes are excluded (Kondo et al., 2011; Wang et al., 2011), the ratios are much closer to each other, indicating consistency of observations and bottom-up emissions. In all cases, the BC/CO emission ratios at regional levels are larger than those at local levels. Since the observations were mainly conducted in or close to developed mega cities, on-road gasoline vehicles which generate elevated CO contribute more to local emissions than they do at regional

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scale. The regional emissions include more combustion of residential solid fuels in less developed rural areas, which generate higher BC emissions. Zhou et al. (2009) conducted observations in Beijing (in North-Central China) and Taicang (in Jiangsu province, East China) during the same period and found a much higher dBC/dCO in Taicang than Beijing. However, this big diversity is not well indicated by the bottom-up emission inventory, with just a slightly higher local BC/CO emission ratio for Jiangsu province than Beijing. Although the large population of gasoline vehicles in Beijing and the diesel use by shipping in East China helped generate higher BC/CO emission ratio for Jiangsu than Beijing, the more consumption of residential coals with elevated BC/CO in Beijing than Jiangsu partly compensated the difference. Further studies are thus recommended on differentiated emission characteristics by region and sector

The comparisons of OC/BC ratios are shown in Table 4 as well. The minimum OC/BC slopes from observation, if available, are used in the comparisons to eliminate the effects of secondary organic aerosols as much as possible. The ratios from estimated emissions are generally close to observed slopes, enhancing confidence in the bottom-up emission inventory for primary carbonaceous aerosols. However, the estimated emissions fail to capture the very high OC/BC ratios in rural Inner Mongolia in fall and winter (Han et al., 2008), suggesting that the current inventory might miss or underestimate emissions from some sources that generate large amounts of OC, such as biofuel use and biomass open burning. Besides short-term observations, relatively long-term observations (2005–2008) of BC and OC have also been conducted in both urban and rural Beijing (Yang et al., 2011). That the ratios from regional (North-Central China) emissions are closer to the observed slopes than local (Beijing) emissions indicates that sources outside Beijing contribute substantially to carbonaceous aerosol levels. Although BC emissions in Beijing are estimated to have declined by 22 % from 2005 to 2008, BC concentrations in urban Beijing did not decline much, and observed BC levels in rural Beijing in fact increased by 21 % (Yang et al., 2011), resulting mainly from increased BC emissions in nearby provinces.

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4.5 Comparisons with satellite observation

SO_2 and NO_2 retrievals from satellite observations are used for comparisons to the primary emissions estimated in this work. Data for SO_2 VCDs of are from the Scanning Imaging Absorption Spectrometer for Atmospheric CHartographY (SCIAMACHY), and the monthly level-3 product with spatial resolution of 0.25° × 0.25° from Support to Aviation Control Service (SACS) is used (data source: http://sacs.aeronomie.be/archive/month/index_VCD_month.php). To approximate the effects of anthropogenic activities, it is assumed that the SO_2 is in the lowest 2 km above the surface, i.e. that SO_2 is found only in the planetary boundary layer. The VCDs of tropospheric NO_2 are from the Ozone Monitoring Instrument (OMI), retrieved by the Royal Netherlands Meteorological Institute (Boersma et al., 2007), and the monthly data with spatial resolution of 0.125° × 0.125° are used (data source: http://www.temis.nl/airpollution/no2col/no2regioomimonth_v2.php). The annual means of SO_2 and NO_2 VCDs over mainland China for 2005–2010 are calculated based on the monthly data and are shown in Figs. S3 and S4 in the Supplement.

Figure 5 illustrates the trends of monthly VCDs of tropospheric SO_2 and NO_2 from satellite observations and of annual emissions of SO_2 and NO_x, from 2005 to 2010 by region. To eliminate seasonal variations, the satellite observations are presented as 12-month moving averages, calculated as the means of the data for the previous and subsequent six months. All values are normalized to 2005 levels, to reflect relative changes during the study period. Generally, the growth of tropospheric NO_2 is consistent with the trends of bottom-up annual emissions, confirming increasing NO_x pollution in mainland China. Specifically, the emissions and observations match well for developed regions including North-Central, East, and South-Central China. In the west of the country, however, bigger discrepancy is found between the growth trends of emissions and VCDs (Fig. 5e, f). This is partly because the random retrieval errors of satellite observation can be significant over western regions with relatively clean environment and low NO_2 values, and natural sources such as lighting could contribute

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more than they do in the developed eastern regions (Lin et al., 2010b). Moreover, the estimated emissions fail to fully capture the drop of NO_2 during late 2008–early 2009, attributed to limits on economic activities and energy consumption to improve air quality for the Beijing Olympics (Wang et al., 2010b) and/or economic downturn in the country (Lin et al., 2011). This discrepancy reveals the limits of emission inventories at annual temporal resolution to reflect responses to short-term variations of economic activity and control policies at local or regional scales.

The comparisons for SO_2 are perplexing. As shown in Fig. 5, the SO_2 VCDs are observed to increase first with a subsequent abatement during 2005–2009, consistent with the trends of estimated emissions. The trends are confirmed as well by Fig. S4a–e in the Supplement, showing that the areas with relatively high SO_2 VCDs in mainland China expanded from 2005 to 2007 but then shrank in the following two years. At the national level, the SO_2 VCDs from satellite observation declined by 9 % from 2005 to 2009, close to the value of 11 % for emissions estimated by this work. Nevertheless, the observed SO_2 rebounded again from late 2009 or early 2010 for all regions, while little increase in bottom-up emissions is estimated. The disagreement can come from either or both the uncertainties of emission estimation and satellite observations. As analyzed in Sect. 4.3, the variable and unclear operation levels of FGD systems in China's power plants contribute significantly to the uncertainty of national SO_2 emissions, particularly for the most recent years when greater penetration of FGD in power sector occurred. A much higher upper limit to the 95 % CI (+26 % higher than the central value for 2010) is thus estimated than that of previous years (e.g., +13 % for 2005). On the other hand, as shown in Fig. S4f, the spatial pattern of retrieved SO_2 VCDs for 2010 changed dramatically from those in the previous years of 2005–2009, with significant growth throughout country including the less developed west, regions with sparse anthropogenic SO_2 sources. This exaggerated growth of SO_2 columns implies possible increased emissions from non-anthropogenic sources, and/or large uncertainties in satellite data retrieval due to variations of atmospheric conditions including the varied aerosol absorption and conversion efficiency of SO_2 to sulfate, as suggested by Lu

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et al. (2011). The discrepancy between emissions and observation has been realized by SACS and reduced uncertainty of data retrieval is expected through reprocessing of SO_2 VCDs from SCIAMACHY (personal communication with SACS).

5 Discussion

5.1 The effects of policy on emission abatement

During 2005–2010, substantial efforts were undertaken in China to achieve national targets in both energy conservation and emission reduction, particularly in sectors with high energy intensities and large emissions of air pollutants and CO_2. These included power generation, cement production, and iron and steel production. Those efforts are estimated to have had varied effects by sector and species. Figure 6 illustrates the inter-annual trends of production, coal consumption, and emissions for the three sectors from 2005 to 2010.

For the power sector, as shown in Fig. 6a, the electricity generated from coal-fired plants increased 62 % during 2005–2010, while coal consumption by the power sector increased 46 %, reflecting the progress of energy conservation in the sector. This is mainly due to the replacement of small and old power units with more energy-efficient large units (e.g., super-critical and ultra super-critical units), reducing the coal consumption per unit electricity generation by 10 %, from 370 to 335 g of coal equivalent per unit kilowatt-hour (gce kWh^{-1}) (updated from previous work by Zhao et al., 2008). The higher penetration of large units raised as well the application of advanced emission control technologies for PM (e.g., ESP and fabric filter) and SO_2 (e.g., wet-FGD systems), resulting in strong emission abatement of the two pollutants, by 46 % and 42 %, respectively from 2005 to 2010. Even without dedicated control measures growth in CO emissions from power plants was also successfully restrained, attributed to the improved combustion efficiency of boilers. The annual emissions of NO_x and CO_2, however, had similar growth trends as that of coal consumption, indicating that current

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control of these two species depends significantly on growth of energy consumption. Although the penetration of SCR technology reached 10 % in the power sector in 2010, the actual effects on NO_x control cannot yet be verified (Zhao et al., 2010), in the same manner that FGD has not taken full control effect on SO_2 in recent years. Since coal will continue to dominate the energy structure of China's electricity generation in the near future, the improvement of SCR use, in terms not only of penetration in the sector but also of operational performance and removal efficiency, is likely the most effective way to constrain growth of NO_x emissions from coal-fired power plants.

As shown in Fig. 6b, cement production has increased by 76 % from 2005 to 2010, and the actual production of 1.9 billion metric tons in 2010 already exceeded the prediction for 2020 of an analyses using a computable general equilibrium (CGE) economic model (Lei et al., 2011b). The dramatic growth of cement production, together with that of steel (described later), reflects the unexpectedly swift development of infrastructure facilities in the past few years in China. Attributed to increased penetration of precalciner kilns, with higher energy efficiency than other kilns (Lei et al., 2011b), the growth of coal consumption in the sector increased 67 % during 2005–2010, slower than that of cement production. The growth of precalciner kilns has improved sector-wide combustion efficiency and increased the use of emission control devices like fabric filters, leading to considerable reduction of CO and PM. However, NO_x emissions increased by 130 % during the study period, resulting both from the swift growth of cement production and the higher NO_x emission levels produced by precalciner kilns compared to other kilns. This tension between improved technology and increased NO_x emissions in the cement industry indicates that current policies are far from sufficient to reduce associated NO_x emissions, and suggests that SCR/SNCR systems need to be promoted in the sector in the future.

During 2005–2010, iron and steel production increased by 116 %, even faster than cement production, while the coal consumption of smelting and pressing of ferrous metals increased by only 45 % (Fig. 6c). This big achievement in energy saving resulted mainly from the retirement of small steel production plants and the increased

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use of recycled gas in coke ovens, blast furnaces, and basic oxygen furnaces (Zhao et al., 2012a). However, those improvements had very limited effects on the sintering process, leading to continued growth of SO_2 and NO_x emissions. Moreover, PM emissions from the iron and steel industry are estimated to have increased as well, resulting mainly from the fugitive emissions from coking, pig iron production, and casting processes, which have not been subject to significant controls in recent years. Among all the species, CO_2 emissions are estimated to have increased fastest, by 87 % from 2005 to 2010, although this growth rate is still lower than that of production. The comparisons of the inter-annual trends between production and emissions confirm that technology improvement in the iron and steel industry have had some emission control effect but not to the extent of reversing growth of emissions of any species. A way to further abate emissions would be to increase the penetration of electric arc furnaces in steel making, which employs a short flow process (i.e. reuses waste steel in the material flow) and thus has much higher energy efficiency and lower emission factors than basic oxygen furnaces.

Although polices of emission control were implemented in some key sectors, the emission trends of most species (particularly NO_x and CO_2) are still largely driven by the underlying activity levels, i.e. energy consumption or industrial production. Because of the economic downturn from late 2008 to early 2009, there was a clear leveling off of energy and industrial production at that time, attended by temporarily slowed growth in NO_x and CO_2 emissions. However, starting in 2009, emissions accelerated again corresponding to the national policy to stimulate the economy including enormous investments in infrastructure construction. This strong dependence of emissions on the economy and energy implies that there are still major challenges in emission abatement in China as the economy continuously develops, and stronger control measures are needed not only in the power and heavy industrial sectors but likely also should be extended to other emission sources.

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5.2 Implications of emission trends of different aerosol species

From 2005 to 2010, China's TSP emissions are estimated to have decreased by 14 % from 33.2 to 28.7 Mt, implying progress in control of PM in these years. The emissions of finer particles, PM_{10} and $PM_{2.5}$, are estimated to have declined by 10 % and 5 %, respectively. The lesser abatement indicates more difficulty in emission control of fine particles than the coarse fraction. In recent years, penetration of dust collectors into industrial process sources has grown, leading to improved emission reduction of TSP. However, many such dust collectors are cyclones or wet scrubbers, with much lower removal efficiencies for fine particles than TSP. Application of fabric filter systems, which are considerably more effective at $PM_{2.5}$ control, is still limited at major PM sources including power, cement, and iron and steel plants. As shown in Fig. 2, the reduction of emission factors for $PM_{2.5}$ is estimated to have been smaller than that of PM_{10} or TSP in all sectors during 2005–2010, resulting in less benefit in emission abatement for finer particles. Moreover, the emissions of BC and OC were even less reduced, since the main sources of those species are residential small stoves burning solid fuels and open biomass burning, for which very few technology improvements have been developed or successfully deployed during recent years. Since fine particles and carbonaceous primary aerosols are much more closely associated with public health and radiative forcing than TSP, there is an urgent need for control measures targeting those aerosol species, particularly for industrial processes and residential fuel combustion.

Reduced emissions of PM and thereby alkaline base cations with acid-neutralizing effects may increase the ecosystem acidification risks in China. From 2005 to 2010, Ca emissions are estimated to have declined by 24 %, while SO_2 emissions by only 11 %, as shown in Fig. 7. The emission reduction rates of 2005–2010 are faster than those of a longer-term trend projected by the authors on the basis of current policy commitments, which suggest that China's base cation and SO_2 emissions will decline 49 % and 22 % from 2005 to 2020, respectively (Zhao et al., 2011b). In contrast, the US national emissions of SO_2 and PM_{10} (as a surrogate for base cations, since no emission

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of base cation was reported) are officially reported to have declined 36 % and 24 %, respectively, from 1990 to 2005, the 15 yr following enactment of the 1990 amendments to the Clean Air Act (USEPA, 2010). Even with this aggressive level of SO_2 abatement in the US, it was estimated that the amendments would not be adequate to protect surface waters and forest soils of the Northeastern US against further anthropogenic acidification based on long-term observation of a forest catchment (Likens et al., 1996) Comparing the situations of the two countries, the much smaller percentage decline of SO_2 and much larger decline of base cations in China indicate that recovery of acidification in the country may be more difficult under current control policies than the US experienced in 1990–2005. Recently, a long-term monitoring study found an association between increased acidity of precipitation and decreased PM concentrations at many sites across China, which cannot be explained by changes in natural sources (Tang et al., 2009). The observation confirmed increased acidification risks due to decreased emissions of anthropogenic base cations over the country. Since PM control efforts will doubtlessly continue in China to achieve important benefits of reduced aerosol pollution and avoided damages to public health, little other choice is available to alleviate acidification but to pursue even more stringent SO_2 controls.

5.3 The spread of air pollution challenges from urban centers to less developed areas

While China's mega cities have been suffering from poor air quality for a long time (Parrish and Zhu, 2009), satellite observations suggest that even faster growth of air pollutants such as NO_2 is now seen in less densely developed regions compared to mega cities (Zhang et al., 2012). As shown in Fig. S3 in the Supplement, very limited increase in tropospheric NO_2 VCDs was found in the developed Yangtze River Delta from 2005 to 2010; indeed there was even a small reduction in NO_2 VCD in the mega city of Shanghai. In contrast, much larger growth of NO_2 (exceeding 20 % during 2005–2010) was retrieved for the less developed areas adjacent to Shanghai and the Delta, such as north Jiangsu and Anhui provinces. Similarly, the NO_2 increase during the five

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years in the mega city of Beijing (around 20 %) was much smaller than that in nearby provinces including Tianjin and Hebei (over 40 %), and the NO_2 columns in Guangdong province containing the developed Pearl River Delta region showed a decline of 13 % during 2005–2010. All of these trends indicate that China's air pollution challenges have been expanding from developed urban areas to nearby regions, explained partly by changes in anthropogenic activities and thereby emissions. In recent years, China's rapid urbanization of relatively small cities and development policies targeting interior regions have spread economic growth to less developed areas, which now often have increased industrial production and more emission sources. Furthermore, gradually tightened emission controls in the most highly developed, heavily polluted urban areas has lead to relocation of major industrial operations from urban to rural regions. Based on county-level economic data provided by the China Data Center, University of Michigan (http://chinadataonline.org/), the fraction of national GDP generated by "secondary industry" (including mining, manufacturing, and construction) in China's urban areas is estimated to have declined from 54 % to 48 % during 2005–2010. As a more specific example, the fraction of capacity of coal-fired power plants in urban areas decreased from 56 % to 47 %, according to the updated unit-level database of power plants by Zhao et al. (2008). Moreover, the emission factors of on-road transportation, which has contributed an increasing share of urban pollution, have at the same time been gradually constrained by the implementation of staged emission standards, while small industrial boilers and residential stoves, which contribute significantly to rural emissions, have not been effectively targeted by control policies. At the provincial level, Table 5 provides the fractions of the sum of China's most developed provinces to the national total in typical activity levels and emissions of different species for 2005 and 2010. The provinces include Beijing and Tianjin in the North China Plain; Shanghai, Jiangsu, and Zhejiang, encompassing the Yangtze River Delta; and Guangdong, which includes the Pearl River Delta. The lower values for 2010 compared to 2005 in all the entries in Table 5 confirm that China's economic activities, energy consumption, and atmospheric pollutant emissions have shifted proportionately to relatively less developed

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areas under current emission control and economic development polices. National air pollution control strategies will increasingly need also to address conditions in these areas in the future.

6 Conclusions

Under pressures of enormous energy consumption and severe atmospheric pollution, China has been implementing a series of policies in energy conservation and emission reduction in recent years. These include the retirement of small and inefficient power and industrial plants, deployment and operation of FGD and SCR systems in the power sector, and implementation of staged emission control regulations for on-road vehicles. The measures have had varied impacts on the inter-annual trends of emissions of different atmospheric species. The emissions of SO_2 and primary PM have been gradually reduced, although uncertainties around these emission estimates have increased from 2005 to 2010, mainly because of the weakly understood operational conditions of the swiftly increased FGD systems and the unclear penetration levels of dust collectors in key industrial sectors. Emissions of NO_x and CO_2 are estimated to have continued increasing, with average annual growth rates of 8.2 % and 7.4 % during 2005–2010, respectively, indicating the limited progress of current measures and ongoing major challenges in emission control of these two species. Although emission control policies greatly reduced TSP, fewer benefits were achieved for primary fine particles and carbonaceous aerosols, which contribute more to human health damages and climate forcing. Moreover, the estimated swift decline of alkaline base cations in primary PM compared to SO_2 suggests rising acidification risks to ecosystems, as also indicated by long-term observations at multiple sites across the country. There is therefore a great future need for a comprehensive, multi-pollutant control strategy, consisting not of separately developed piecemeal policies targeting single atmospheric species but rather conceived to redress a complex of emissions and diverse environmental impacts.

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In this work, emissions estimated bottom-up are compared with available observations from ground sites and satellites for different species, locations, and periods. While observations generally reflect inter-annual trends of estimated emissions in most cases, clear discrepancies exist for given regions, seasons, or years. These discrepancies result from uncertainties both of observations and of bottom-up emission estimations. Limits of data prevented full realization of regional and seasonal differentiations in activity levels, technology distributions, and emission factors for some sectors, particularly industrial processes and residential fuel combustion. Considering the rapidly changing complex of emission sources across China, further investigation of spatial and time distributions of contributing factors are needed to reduce uncertainties and to generate more accurate trends of emissions.

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Table 1. Emissions in China 2010 by province (unit: Mt for CO_2 and kilo metric tons (kt) for other species).

Province	Region	Gaseous pollutants			Primary aerosols							GHG
		SO_2	NO_X	CO	TSP	PM_{10}	$PM_{2.5}$	BC	OC	Ca	Mg	CO_2
Beijing	North-central	187	312	2267	242	118	83	15	18	36	3	98
Tianjin	North-central	351	594	3003	292	181	137	17	24	24	6	186
Hebei	North-central	1942	2009	16 730	2353	1395	1021	125	204	263	44	782
Shanxi	North-central	1660	1243	6639	1185	656	473	71	129	139	16	443
Inner Mongol	North-central	1304	1248	5273	968	697	534	88	183	76	15	470
Liaoning	Northeast	1188	1339	9421	1199	724	525	65	97	136	20	456
Jilin	Northeast	356	586	4168	634	410	298	41	84	52	7	212
Heilongjiang	Northeast	309	764	5258	651	460	356	47	113	41	5	260
Shanghai	East	691	914	4020	360	212	154	17	15	39	8	194
Jiangsu	East	1341	1889	11 500	1662	1019	749	77	160	222	22	710
Zhejiang	East	909	1335	5263	809	446	299	32	47	145	8	413
Anhui	East	803	1184	9702	1236	782	617	74	178	116	9	402
Fujian	East	486	766	3414	521	321	219	29	40	76	4	249
Jiangxi	East	633	576	4643	1008	442	288	33	52	262	11	225
Shandong	East	3199	2610	17 234	2961	1704	1182	153	258	490	44	905
Henan	South-central	1402	1874	12 418	2406	1237	859	98	181	451	29	683
Hubei	South-central	1241	1107	8869	1260	741	539	75	138	161	13	412
Hunan	South-central	1036	963	7423	1295	769	571	66	144	181	12	336
Guangdong	South-central	1112	1836	8834	1319	737	492	57	90	292	11	607
Guangxi	South-central	738	710	7384	1018	618	483	56	134	189	8	269
Hainan	South-central	38	127	674	78	50	38	5	10	17	0	38
Chongqing	Southwest	1148	487	3088	509	303	214	30	58	90	4	179
Sichuan	Southwest	1813	1083	10 276	1245	768	573	74	168	212	9	409
Guizhou	Southwest	1075	752	3896	715	409	305	60	107	133	6	259
Yunnan	Southwest	616	735	4440	851	550	404	77	98	108	16	232
Tibet	Southwest	1	23	136	9	8	7	1	2	1	0	4
Shaanxi	Northwest	926	703	4794	744	400	289	45	86	142	6	276
Gansu	Northwest	409	380	2708	386	256	200	27	56	61	7	149
Qinghai	Northwest	36	93	534	114	78	61	8	14	19	2	38
Ningxia	Northwest	303	277	842	204	136	94	15	20	27	5	103
Xinjiang	Northwest	460	458	3047	448	306	230	39	65	58	7	176
Total		27 714	28 980	187 900	28 680	16 931	12 293	1619	2973	4262	358	10 176

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Table 2. Uncertainties of Chinese emissions by sector in 2010. The emissions are expressed as Mt for CO_2 and kt for other species. The percentages in the parentheses indicate the 95 % CI around the central estimate.

	Power plants	Total industry	Transportation	Residential and commercial	Total
SO_2	9199 (−27 %, 59 %)	15 254 (−22 %, 27 %)	374 (−21 %, 41 %)	2888 (−46 %, 51 %)	27 714 (−15 %, 26 %)
NO_x	9629 (−19 %, 15 %)	9541 (−32 %, 90 %)	7207 (−30 %, 54 %)	2604 (−37 %, 101 %)	28 981 (−15 %, 34 %)
CO	1400 (−27 %, 38 %)	90 058 (−11 %, 31 %)	32 676 (−44 %, 74 %)	63 765 (−49 %, 101 %)	187 900 (−18 %, 42 %)
TSP	1592 (−22 %, 37 %)	21 141 (−28 %, 65 %)	716 (−29 %, 47 %)	5231 (−48 %, 81 %)	28 680 (−21 %, 52 %)
PM_{10}	1233 (−25 %, 43 %)	10 254 (−16 %, 69 %)	697 (−29 %, 48 %)	4747 (−50 %, 87 %)	16 932 (−14 %, 51 %)
$PM_{2.5}$	717 (−34 %, 62 %)	6394 (−15 %, 85 %)	661 (−29 %, 50 %)	4522 (−50 %, 88 %)	12 293 (−14 %, 59 %)
BC	5 (−68 %, 574 %)	574 (−49 %, 117 %)	274 (−72 %, 75 %)	766 (−45 %, 251 %)	1619 (−25 %, 131 %)
OC	0 (−76 %, 2373 %)	493 (−41 %, 141 %)	122 (−67 %, 87 %)	2366 (−53 %, 146 %)	2982 (−39 %, 122 %)
Ca	69 (−28 %, 45 %)	4119 (−78 %, 77 %)		74 (−68 %, 171 %)	4262 (−75 %, 76 %)
Mg	17 (−26 %, 37 %)	325 (−52 %, 37 %)		16 (−69 %, 157 %)	358 (−46 %, 151 %)
CO_2	3253 (−13 %, 14 %)	4635 (−14 %, 17 %)	834 (−12 %, 16 %)	1454 (−37 %, 20 %)	10 176 (−10 %, 9 %)

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Table 3. The top two parameters contributing most to emission uncertainties by sector for 2010. The percentages in the parentheses indicate the contributions of the parameters to the variances of emissions (see Eq. 1 for the abbreviations of parameters).

	Coal-fired power plants	Total industry	Transportation	Residential and commercial activity
SO_2	$\eta_{SO2,FGD}$ (72 %) AL_{coal} (5 %)	$SR^{1}_{grate\ boiler}$ (22 %) AL_{coal} (19 %)	$SR_{non\text{-}road\ coal\ combustion}$ (43 %) $AL_{non\text{-}road\ source,\ coal}$ (29 %)	$EF_{SO2,\ hot\ water\ system}$ (38 %) AL_{coal} (37 %)
NO_X	$EF_{NOx,\ tangential\ bituminous\ boiler}$(28 %) AL_{coal} (27 %)	$EF_{NOx,\ grate\ boiler}$(74 %) $EF_{NOx,\ precalciner\ cement\ kiln}$ (9 %)	$EF_{NOx,\ inland\ ship}$ (49 %) $AL_{HDDV,\ Diesel}$ (8 %)	$EF_{NOx,\ oil}$ (45 %) $EF_{NOx,\ biomass\ open\ burning}$(14 %)
CO	$EF_{CO,\ pulverized\ boilers\ (<\ 200\ MW)}$ (41 %) $EF_{CO,\ pulverized\ boilers\ (\geq\ 200\ MW)}$ (28 %)	$EF_{CO,\ refinery}$ (36 %) $EF_{CO,\ brick\ making}$ (14 %)	$EF_{CO,\ small\ gasoline\ engine}$ (24 %) $R_{LDGV,\ Stage\ III}$ (13 %)	$EF_{CO,\ straw\ as\ biofuel}$ (64 %) $AL_{biofuel,\ straw}$ (19 %)
TSP	$f^{2}_{PM2.5\ pulverized\ boiler}$ (28 %) AL_{coal} (11 %)	$R_{lime\ production,\ cyclone}$ (26 %) $R_{lime\ production,\ ESP}$ (15 %)	$EF_{PM2.5,\ rural\ machine}$ (34 %) $EF_{PM2.5,\ construction\ machine}$ (12 %)	$EF_{PM2.5,\ straw\ as\ biofuel}$(30 %) $EF_{TSP,\ coal\ stove}$(20 %)
PM_{10}	$f_{PM2.5\ pulverized\ boiler}$ (37 %) $\eta_{PM2.5\ ESP}$ (13 %)	$AR^{3}_{grate\ boiler}$ (12 %) $EF_{TSP,\ precalciner\ cement\ kiln}$(10 %)	$EF_{PM2.5,\ rural\ machine}$ (36 %) $EF_{PM2.5,\ construction\ machine}$ (12 %)	$EF_{PM2.5,\ straw\ as\ biofuel}$(30 %) $EF_{TSP,\ coal\ stove}$(21 %)
PM_{25}	$f_{PM2.5\ pulverized\ boiler}$ (55 %) $\eta_{PM2.5\ ESP}$ (19 %)	$f_{PM2.5\ grate\ boiler}$ (17 %) $AR_{grate\ boiler}$ (12 %)	$EF_{PM2.5,\ rural\ machine}$ (38 %) $EF_{PM2.5,\ construction\ machine}$ (13 %)	$EF_{PM2.5,\ straw\ as\ biofuel}$(30 %) $EF_{TSP,\ coal\ stove}$(21 %)
BC	$F^{4}_{BC,\ pulverized\ boiler}$ (79 %) $f_{PM2.5\ pulverized\ boiler}$ (5 %)	$AL_{coke\ production}$ (26 %) $F_{BC,\ grate\ boiler}$ (15 %)	$F_{BC,\ non\text{-}road\ coal\ combustion}$ (70 %) $EF_{PM2.5,\ rural\ machine}$ (8 %)	$EF_{BC,\ coal\ stove}$ (50 %) $EF_{BC,\ wood\ as\ biofuel}$ (22 %)
OC	$F_{OC,\ grate\ boiler}$ (42 %) $f_{PM2.5\ grate\ boiler}$ (24 %)	$AL_{coke\ production}$ (24 %) $F_{OC,\ grate\ boiler}$ (17 %)	$F_{OC,\ non\text{-}road\ coal\ combustion}$ (52 %) $F_{OC,\ on\text{-}road\ diesel\ vehicle}$ (21 %)	$EF_{OC,\ straw\ as\ biofuel}$ (38 %) $EF_{OC,\ coal\ stove}$ (25 %)
Ca	$f_{PM2.5\ pulverized\ boiler}$ (18 %) $\eta_{PM2.5\ ESP}$ (7 %)	$R_{lime\ production,\ cyclone}$ (40 %) $R_{lime\ production,\ ESP}$ (23 %)		$EF_{TSP,\ coal\ stove}$ (68 %) AL_{coal} (13 %)
Mg	$f_{PM2.5\ pulverized\ boiler}$ (24 %) AL_{coal} (9 %)	$F_{Mg,\ iron\ and\ steel\ production}$ (60 %) $R_{iron\ and\ steel\ production\ cyclone}$(12 %)		$EF_{TSP,\ coal\ stove}$ (70 %) AL_{coal} (14 %)
CO_2	AL_{coal} (53 %) $EF_{CO2,\ bituminous\ boiler}$ (43 %)	$EF_{CO2,\ bituminous\ boiler}$ (42 %) $EF_{CO2,\ iron\ and\ steel\ production}$ (31 %)	$AL_{LDGV,\ gasoline}$ (30 %) $AL_{HDDV,\ diesel}$ (18 %)	$AL_{biofuel,\ straw}$ (38 %) $EF_{CO2,\ straw\ as\ biofuel}$ (21 %)

[1] SR, the release ratio of sulfur content during combustion; [2] f, the mass fraction of particles with specific size to TSP; [3] AR, the release ratio of ash content during combustion; [4] F, the mass fraction of chemical species to $PM_{2.5}$ (for carbonaceous aerosols) or TSP (for base cations).

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Table 4. The ratios of BC to CO ($\mu g\,m^{-3}\,ppbv^{-1}$) and OC to BC ($\mu g\,m^{-3}/\mu g\,m^{-3}$) from observations and bottom-up emissions in China.

Sources	Location	Period and seasons	Observed slopes	Emission ratios Local	Emission ratios Regional
BC/CO					
Kondo et al. (2011)	Okinawa Island, Japan	Feb 2008–May 2009	0.0039[1] 0.0075[2]	0.0095	0.0103
Wang et al. (2011)	Miyun, Beijing (rural)	Apr–Oct 2010	0.0046[1] 0.0095[3]	0.0080	0.0113
Han et al. (2009)	PKU Beijing (urban)	Nov 2005–Jan 2006	0.0035	0.0062	0.0125
		Mar–May 2006	0.0034	0.0092	0.0115
		Aug–Sep 2006	0.0048	0.0091	0.0112
		Sep–Oct 2006	0.0058	0.0090	0.0112
Zhou et al. (2009)	Changping Beijing (town)	Summer 2005	0.0046	0.0063	0.0127
	Taicang, Jiangsu (suburban)	Summer 2005	0.0126	0.0077	0.0095
Verma et al. (2010)	Guangzhou, Guangdong (urban)	Jul 2006	0.0054	0.0092	0.0102
Li et al. (2007)	Xianghe, Hebei (rural)	Mar 2005	0.0101	0.0104	0.0129
OC/BC					
Yang et al. (2011)	Miyun, Beijing (rural)	2005–2008	1.8	1.3	1.7
	THU, Beijing (urban)	2005-2008	1.7	1.3	1.7
Gu et al. (2010)	Tianjin (urban)	Jan 2008	2.0[4]	2.0	2.1
		Apr 2008	2.1[4]	1.6	1.6
		Jul 2008	1.4[4]	1.3	1.8
		Oct 2008	1.7[4]	1.6	1.6
Han et al. (2008)	Daihai, Inner Mongol (rural)	Fall 2005	4.1	2.4	1.9
		Winter 2006	6.1	2.4	2.1
		Summer 2006	2.5	2.4	1.8
		Spring 2007	1.8	2.0	1.6
Andreae et al. (2008)	Guangzhou Guangdong (urban)	Oct–Nov 2004	1.4	1.5[5]	1.5[5]

[1] Original from observation; [2] Excluding influence of wet deposition and representing mainly the effects of emissions from China; [3] Excluding influence of biomass burning, wet deposition and atmospheric processes, and representing mainly the effects of emissions from North China Plain; [4] Minimum values during the observation to approximate the emission ratios of OC/BC; [5] Values for Oct–Nov 2005.

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Table 5. The fractions of the sum of developed provinces (including Beijing, Tianjin, Shanghai, Jiangsu, Zhejiang, and Guangdong) to total country in activity levels and emissions for 2005 and 2010.

	The fractions of developed provinces to total country	
	2005	2010
Activity levels		
Capacity of coal-fired power plants	28 %	24 %
Cement production	28 %	22 %
Steel production	30 %	28 %
Coal consumption	18 %	17 %
On-road vehicle population	36 %	34 %
Emissions		
SO_2	22 %	17 %
NO_x	28 %	24 %
CO	21 %	18 %
PM	19 %	16 %
CO_2	23 %	21 %

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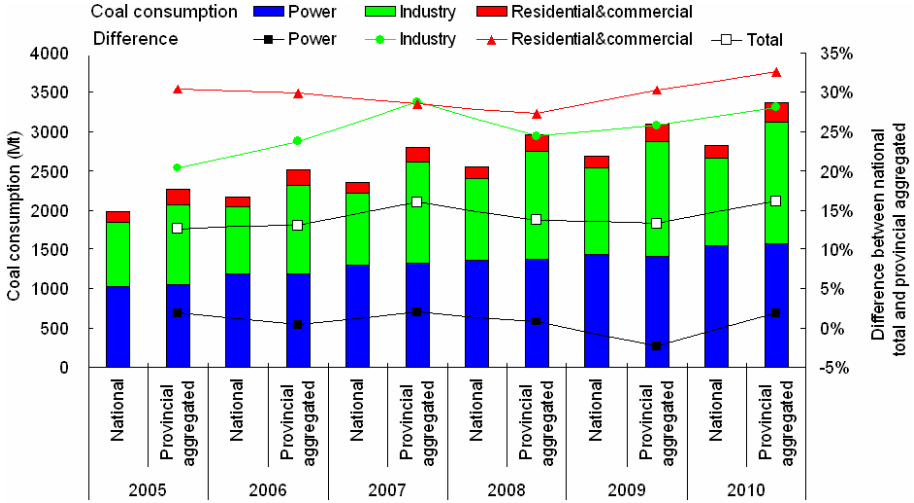

Fig. 1. China's coal consumption by sector and the relative difference between the national total statistics and aggregation of provincial statistics from 2005 to 2010.

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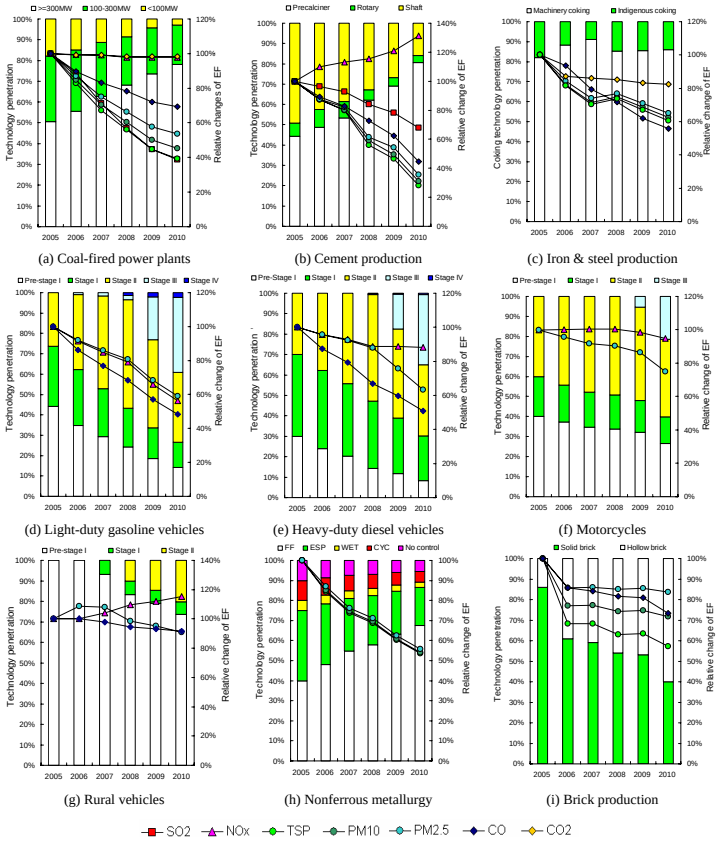

Fig. 2. The penetrations of technologies and inter-annual trends of emission factors for typical sources in China from 2005 to 2010. In each panel, left-hand vertical axis indicates the percentages of various technologies and right-hand vertical axis indicates the relative changes of emission factors for various species.

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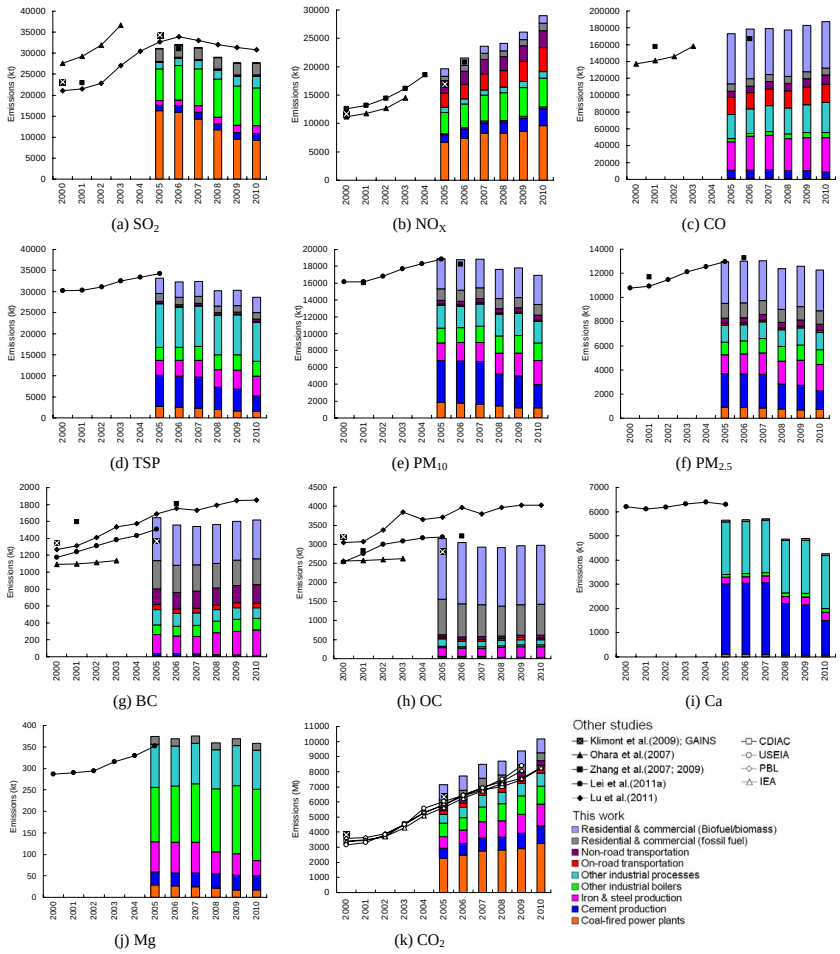

Fig. 3. The annual emissions by sector in China from 2005 to 2010 as well as total emissions from other studies 2000–2010.

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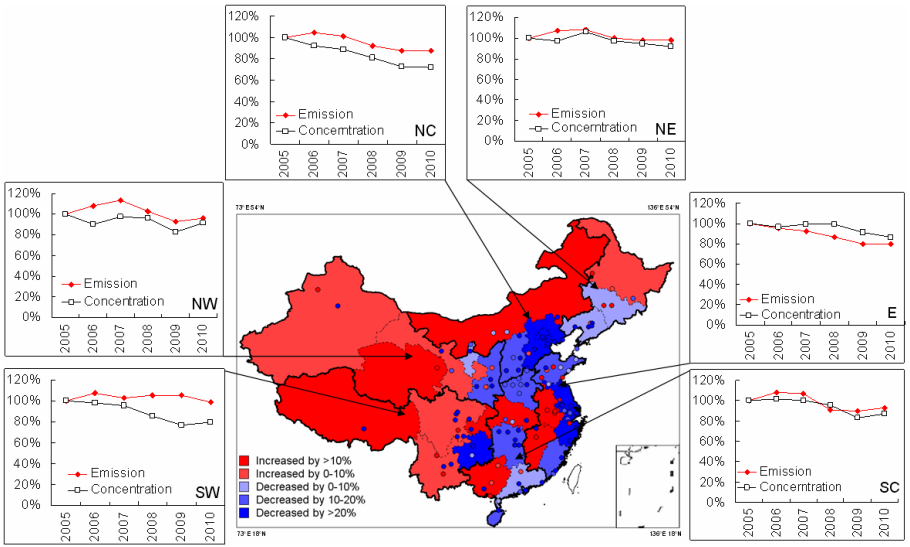

(a) SO_2

Fig. 4. Inter-annual trends of the average ambient concentrations for 113 key cities reported by MEP and emissions estimated by this work. The maps illustrate the changes in emissions by province and concentrations by city between 2005 and 2010. The panels around the maps illustrate the relative changes in emissions by region and concentrations by city from 2005 to 2010. Thick lines in the maps indicate borders of the six regions: North-Central China (NC), Northeast China (NE), East China (E), South-Central China (SC), Southwest China (SW), and Northwest China (NW).

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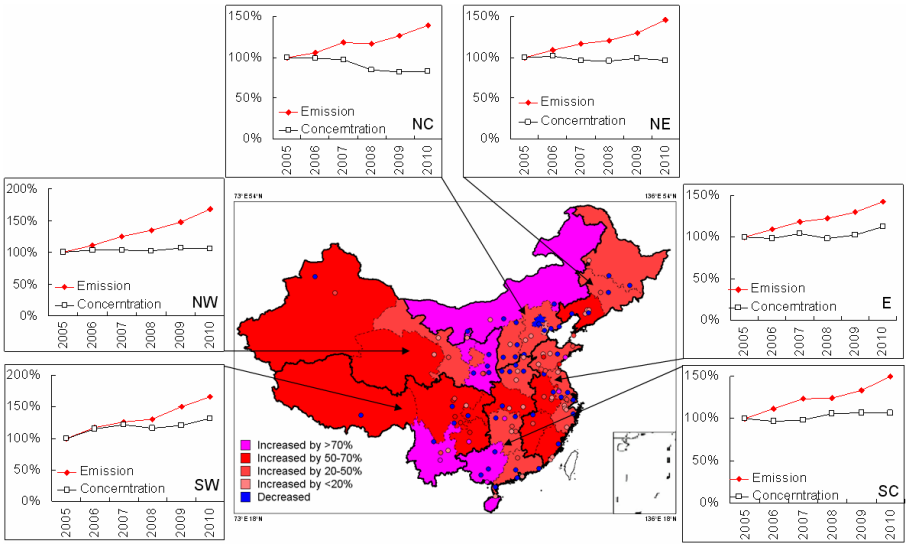

(b) NO_2 (for concentration)/NO_X (for emission)

Fig. 4. Continued.

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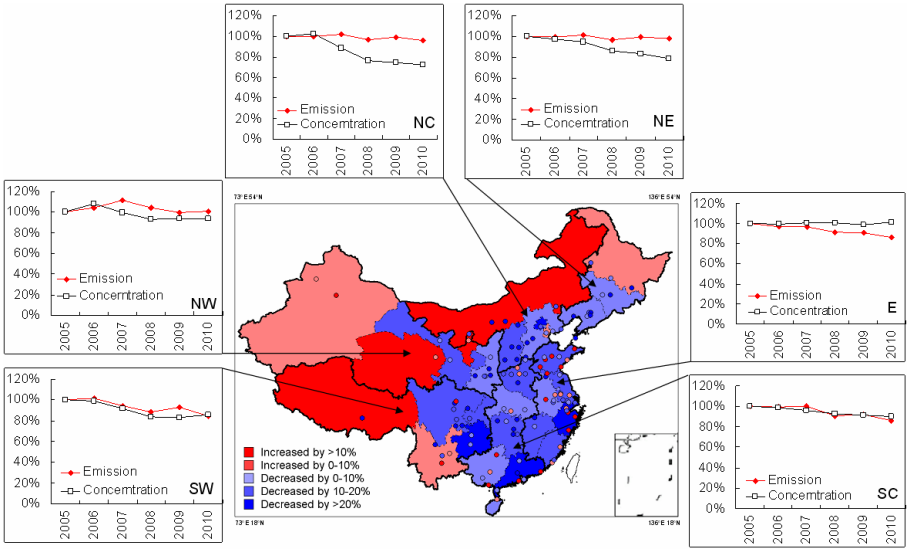

(c) PM_{10}

Fig. 4. Continued.

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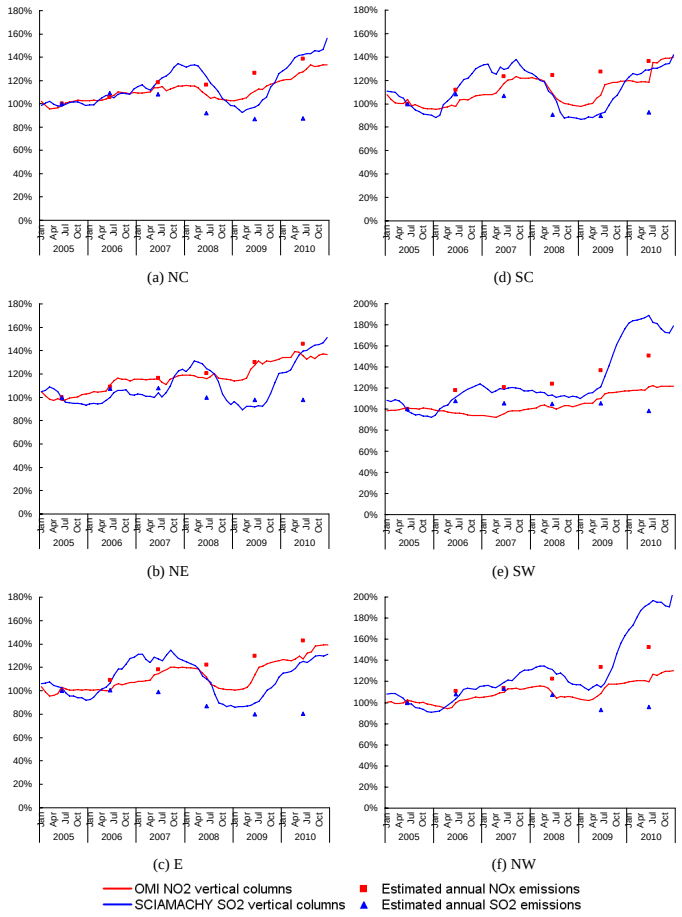

Fig. 5. The comparisons of inter-annual trends between satellite observation and bottom-up emissions from 2005 to 2010. All the data are normalized to 2005 levels.

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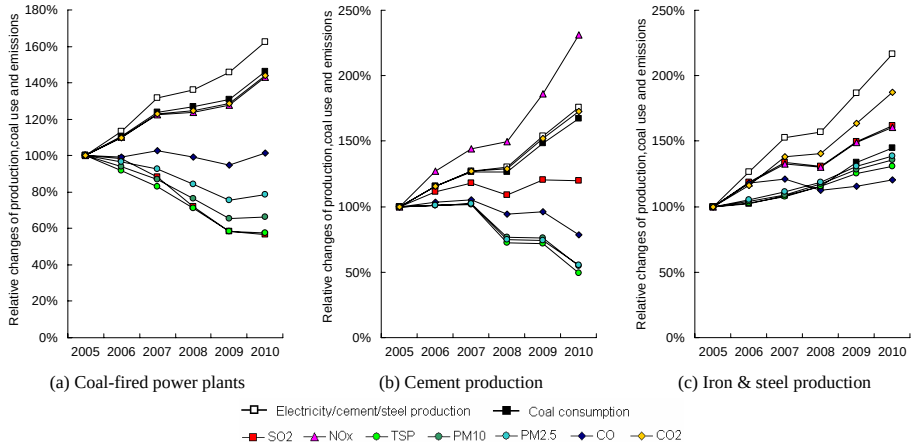

Fig. 6. Relative changes of production, coal consumption and emissions for coal-fired power plants **(a)**, cement plants **(b)** and iron and steel plants **(c)** from 2005 to 2010. Note the scales are different for the three panels.

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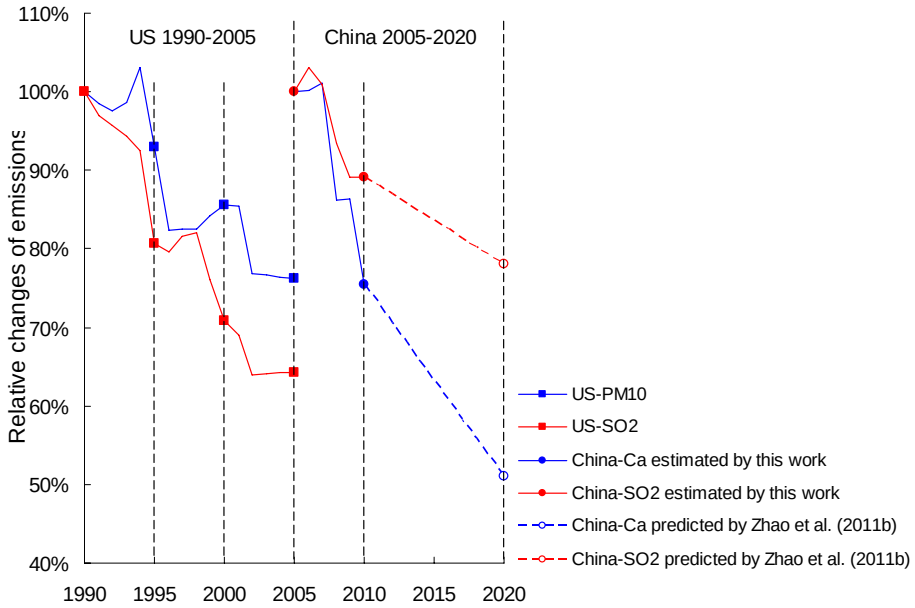

Fig. 7. Relative changes of SO_2 and PM_{10}/Ca emissions for USA 1990–2005 and China 2005–2020.

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