Editor

I would suggest to make a few comparisons between satellite-retrieved NO_2 concentrations and NO_x emissions (2 to 3 power plants, especially those dominating the local emissions). Even if we see some differences, it will still add values to this manuscript for the orientation of future work.

Response: Thanks for the suggestion. We have added the comparison of bottom-up NO_x emissions from power plants with NO_2 columns derived from OMI in the revised manuscript.

High-resolution inventory of technologies, activities, and emissions of coal-fired power plants in China from 1990 to 2010

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22 Abstract

This paper, which focuses on emissions from China's coal-fired power plants during 23 1990–2010, is the second in a series of papers that aims to develop high-resolution 24 emission inventory for China. This is the first time that emissions from China's 25 coal-fired power plants were estimated at unit level for a 20-yr period. This inventory 26 27 is constructed from a unit-based database compiled in this study, named the China 28 coal-fired Power plant Emissions Database (CPED), which includes detailed information on the technologies, activity data, operation situation, emission factors, 29 30 and locations of individual units and supplements with aggregated data where unit-based information is not available. Between 1990 and 2010, compared to a 479% 31 growth in coal consumption, emissions from China's coal-fired power plants 32 increased by 56%, 335% and 442% for SO₂, NO_x and CO₂, respectively, and 33 decreased by 23% and 27% for PM_{2.5} and PM₁₀ respectively. Driven by the 34 accelerated economy growth, large power plants were constructed throughout the 35 36 country after 2000, resulting in dramatic growth in emissions. Growth trend of emissions has been effective curbed since 2005 due to strengthened emission control 37 measures including the installation of flue-gas desulfurization (FGD) systems and the 38 optimization of the generation fleet mix by promoting large units and 39 decommissioning small ones. Compared to previous emission inventories, CPED 40 41 significantly improved the spatial resolution and temporal profile of power plant emission inventory in China by extensive use of underlying data at unit level. The 42 43 new inventory developed in this study will enable a close examination for temporal and spatial variations of power plant emissions in China and will help to improve the 44 45 performances of chemical transport models by providing more accurate emission data.

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47 **1. Introduction**

Bottom-up emission inventories, which are compiled from activity rates and emission 48 49 factors, provide crucial information for understanding the variability of atmospheric compositions and for regulating climate and air quality policies. However, the current 50 understanding of anthropogenic emissions in China is insufficient because of a lack of 51 52 underlying data such as detailed activity rates and local measured emission factors 53 (Zhao et al., 2011). This paper is the second in a series that aims to reduce these uncertainties and to improve the spatial and temporal resolution of bottom-up 54 emission inventories in China. The first paper developed a high-resolution emission 55 map for on-road vehicles (Zheng et al., 2014), and this paper focuses on coal-fired 56 power plants. 57

Power plants consumed approximately half of the total coal production in China over 58 59 the past decade (China Energy Statistical Yearbook, National Bureau of Statistics (NBS), 1992–2011) and contributed significantly to the total national emissions of 60 greenhouse gases and air pollutants (32% of CO₂, 33% of SO₂, 33% of NO_x, and 6% 61 62 of PM_{2.5} in 2010, Y. Zhao et al. (2013)). Therefore, developing a coal-fired power 63 plant emission inventory with high spatial and temporal resolution can significantly improve the accuracy of the anthropogenic emission inventory in China. In the 64 meanwhile, because the power plant sector plays a key role in energy and 65 environmental policies, a well-developed power plant database with accurate energy 66 consumption and emission data could help to guide future policies and evaluate the 67 dynamic changes in emissions induced by those policies. 68

As one of the major anthropogenic emitting sources, coal-fired power plant emissions in China have been estimated in many national, regional, and global inventories. Early studies (Kato and Akimoto, 1992; Klimont et al., 2001; Hao et al., 2002; Ohara et al., 2007) used yearly activity data with fixed emission factors to estimate emissions, which ignored the fact that the net emission rates were changing rapidly with the emergence of new technologies into the market. In recent studies, technology-based methodologies and locally measured emission factors were used to represent the dynamic changes in emissions, which improved the estimates of the magnitudes of
and trends in power plant emissions throughout China (e.g., Zhang et al., 2007, 2009a;
Klimont et al., 2009; Lei et al., 2011; Klimont et al., 2013; Tian et al., 2013; Y. Zhao
et al., 2013).

In addition to the accuracy of the magnitudes, accurate information for each 80 81 generation unit (i.e., location, emission) is also critical for a power plant inventory because power plant emissions are typically large, and improper treatment may lead 82 to significant bias in the spatial distribution of emissions. Owing to the difficulties in 83 84 acquiring information for all of the power plants in China, many bottom-up 85 inventories only identified emissions from large power plants and allocated them according to their latitude and longitude coordinates, whereas emissions from other 86 small units were distributed as area sources (e.g., Streets et al., 2003; Ohara et al., 87 88 2007; Zhang et al., 2009a; Lu et al., 2011). For the first time, Zhao et al. (2008) used 89 unit-level coal consumptions to calculate emissions of individual electric generation units for the years of 2000 and 2005 and assigned them to each location. Subsequent 90 91 studies developed unit-based power plant emission inventories for NO_x for the period 92 of 2005–2007 (Wang et al., 2012) and for SO₂, NO_x, particulate matter and PM_{2.5} for 93 2011 (Chen et al., 2014). The Carbon Monitoring for Action (CARMA) database 94 (Wheeler and Ummel, 2008), a global power plant database at the factory level, has been widely used in bottom-up emission inventories to allocate power plant emissions 95 (EC-JRC/PBL, 2011; Oda and Maksyutov, 2011; Kurokawa et al., 2013; Wang et al., 96 97 2013). However, the accuracy of the emission strengths and locations in the CARMA database is questionable given that it is not a scientific-level dataset that has 98 99 undergone critical evaluation (Oda and Maksyutov, 2011; Gurney, 2012).

There are two major deficiencies in the current power plant inventories throughout China for revealing emissions at the unit level. First, owing to the lack of detailed information at the unit level, emissions from each plant are generally divided by the provincial totals according to capacity (e.g., Zhang et al., 2009a; Lu et al., 2011), which ignores the differences in the emission rates among units introduced by different technologies. Second, in a rapidly developing country such as China, emission factors for a given power plant may change over time as new combustion or emission control technologies are applied following the implementation of new emission standards. Therefore, these time-dependent parameters should be included dynamically when constructing an accurate emission trend for the power plants in China.

The purpose of this study was to develop a high-resolution inventory of the 111 technologies, activity rates, and emissions of coal-fired power plants in China for the 112 113 period of 1990–2010 using extensive underlying data at the unit level, supplemented 114 with aggregated data where unit-based information is not available. This is the first 115 time that coal-fired power plant emissions in China were estimated for each unit from the bottom-up for a two-decade period. We construct a unit-based database, called the 116 117 China coal-fired power plant emissions database (CPED), by collecting information regarding the technologies, activity data, emission factors, and locations of individual 118 electricity generating units. To improve the accuracy of the emission estimates at the 119 120 unit level, the database developed in this study includes not only the type and removal 121 efficiency of emission control equipment for each unit but also the operating 122 conditions of the equipment (i.e., when the equipment was commissioned).

123 Based on the unit-specific parameters from the CPED (e.g., unit capacity, boiler type, operation and phasing-out procedures, the sulfur content and ash content of coal, the 124 type of emission control equipment and the time at which the equipment was 125 commissioned, along with its removal efficiency), the SO₂, NO_x, fine particulate 126 matter (PM_{2.5}), PM₁₀ and CO₂ emissions were estimated on a monthly basis for each 127 coal-fired power generation unit over the period of 1990-2010. CO, VOC, BC and 128 OC emissions were not estimated in this work because coal-fired power plants 129 contributed very small fractions to national total emissions of these species (e.g., less 130 131 than 1% of total CO emissions in 2010 estimated by Y. Zhao et al. (2013)).

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133 **2.** Unit-based Methodology and Data

The CPED database developed in this study consists of 7657 coal-fired electric generating units in mainland China, including ~5700 units in use in 2010 and ~1900 units retired since 2005. The SO₂, NO_x, PM_{2.5}, PM₁₀ and CO₂ emissions from a specific unit in a given month from 1990 to 2010 were estimated using the following equation:

139
$$\operatorname{Emis}_{s,y,m} = U \times P \times (H_0 / H_y) \times T_y \times f_{m,y} \times \operatorname{EF}_{s,k,y} \times \prod_n (1 - \eta_{n,s} \times \tau_{n,m,y})$$
(1)

140 where s represents the emission species, k represents the boiler type, n represents the 141 emission abatement technology type, y represents the year, and m represents the 142 month. U is the unit capacity, in MW, P is the coal consumption rate presented in 143 grams coal equivalent per kWh supplied (gce/kWh), H is the heating value of coal used for each unit in kJ/g, H_0 is the heating value of standard coal, which is 29.27 144 kJ/gce, and the ratio of H_0 to H converts the coal equivalent (gce) to the physical 145 146 quantity of coal (gram). T is the annual operation in hours, the product of U and T is the annual electricity generation, f is the monthly fraction of annual electricity 147 generation, and EF is the unabated emission factor, in g/kg-coal. The parameter η is 148 149 the removal efficiency of the abatement equipment, and τ is the state factor for the abatement equipment; $\tau = 1$ when the equipment is present and running, otherwise $\tau =$ 150 151 0.

152 **2.1. Activity Rates**

153 Detailed activity data are available for each generation unit for the period of 2005–2010 from China's Ministry of Environmental Protection (MEP; unpublished 154 data, referred to hereafter as MEP-database). We used the MEP-database as the basis 155 of deriving the activity rates for each unit for the period of 1990-2010 from a 156 combination of different datasets. The capacity (U) and operational status (when the 157 unit was commissioned/decommissioned) for each unit were collected from the 158 159 MEP-database and the National Development and Reform Commission (NDRC, 160 2013). The annual coal use and power generation of each unit from 2005 to 2010 were

161 also obtained from the MEP-database and were used to calculate the coal 162 consumption rate (P) for each unit. The details about the generation unit fleet mix 163 according to capacity size and efficiency are presented in Sect. 3.1.

The heating value of the coal (H) used for each unit in 2010 was obtained from the 164 MEP-database. In other years for which the unit-level data are not available, the 165 average heating values of the coal used in power plants were derived by year and by 166 province from the energy statistics (NBS, 1992–2011) and were then adopted to scale 167 the 2010 value of each unit to the corresponding years. The heating values of coal 168 169 decreased remarkably since 2007 (from 20.0 kJ/g-coal in 2007 to 18.8 kJ/g-coal in 170 2010 as the national average), indicating the downgraded coal quality in the power sector due to a shortage of coal induced by a surge of electricity demand in recent 171 years (Liu 2007; Shen and Song, 2010). Table S3 of the supplemental information 172 summarizes the provincial average of coal consumption rate and heating value for the 173 year 2010. 174

The annual operating hours (T) for each unit from 2005-2010 were obtained from the 175 MEP-database. In other years for which the unit-based data are not available, 176 operating hours were scaled from the 2005 data according to the ratio of the 177 provincial average operating hours in 2005 and the corresponding year. The provincial 178 179 average operating hours before 2005 were estimated from the provincial total coal consumptions (NBS, 1992–2011) and the product of the corresponding unit capacity 180 and the coal consumption rate obtained from our database. It should be noted that 181 emissions estimates priori to 2005 are more uncertain because the extrapolated 182 parameters were used. 183

The monthly fraction of annual electricity generation (f) is quantified by province, due to the lack of data at unit level. For 2003–2010, f was derived from the statistics (NBS, 2013) and was applied to the units with adjustments if the unit was commissioned or decommissioned within that year, following Eq.(2). For the years prior to 2003, a monthly climatological profile of the 2003–2007 average was used.

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$$f_m = \frac{\gamma_m F_m}{\sum_{m=1}^{12} \gamma_m F_m}$$
(2)

190 where m represents the month. f and F is the monthly fraction of annual electricity

191 generation at unit and province level respectively. γ is the state factor for the unit; $\gamma = 1$

192 when the unit has been commissioned and in operation, otherwise $\gamma=0$.

193 Coordinates of each unit (latitude and longitude) were obtained from the MEP-dataset 194 and then individually validated using Google Earth to ensure that the accurate 195 locations are presented in the CPED.

196 **2.2. Emission Factors**

197 **2.2.1. SO₂**

198 The unabated SO_2 emission factors for a specific unit were estimated via the sulfur 199 mass balance approach using the following equation:

$$200 EF_{SO_2, y} = 2 \times SCC_y \times (1 - Sr) (3)$$

where *y* represents the year, EF_{SO2} is the unabated SO₂ emission factor in g/kg, SCC is the sulfur content of coal, and Sr is the fraction of sulfur retention in ash.

203 The SCC for each unit from 2005–2010 was obtained from the MEP-database. The 204 SCC ranges widely with a mean value of 0.95%. The SCC in the northeast power 205 plants is lowest, whereas the SCC in the central and south power plants is significantly higher than that of plants in other regions, reflecting the different sulfur 206 content in coal production in the various regions (Tang, et al., 2008). For the years 207 208 before 2005, the SCC for each unit was scaled from 2005 data using the ratio of the provincial average SCC in 2005 and the corresponding year. The provincial average 209 SCC before 2005 was calculated from the sulfur contents of coal production in each 210 211 province using the coal transportation matrix approach (Zhang et al., 2012). The sulfur retention ratio was assumed to be 15% for all of the units (Zhang et al., 2009a; 212 Lu et al., 2010) because of the lack of unit-specific data. 213

Flue-gas desulfurization (FGD) systems have been widely installed in coal-fired power plants in China since 2005. This is the most important step for the emission reduction plan to reduce national SO_2 emissions by 10% during the 11th five-year 217 period (2005–2010). In this study, the operating conditions of FGD for each unit were obtained from the MEP-database. The actual SO₂ removal efficiencies for each unit in 218 2010 were also obtained from the MEP-database and were applied to every year 219 because no data are available for the other years. The coal-consumption weighted 220 mean SO₂ removal efficiency of all FGD facilities in 2010 is 78%. Surveys and 221 satellite observations confirmed that some of the early installed FGD facilities were 222 not actually in operation prior to 2008 as the factories reported (Xu et al., 2009; Li et 223 224 al., 2010; Xu, 2011), implying that our assumption may underestimate the SO₂ 225 emissions from 2005 to 2007 for some units. SO₂ emissions can also be removed from wet scrubbers as a co-benefit of particulate matter removal. In this study, we assumed 226 that the removal efficiency of wet scrubbers for SO₂ is 20% (Yao, 1989; Xie, 1995). 227

228 2.2.2. NO_x

229 NO_x emission rates from coal-fired power plants vary significantly by boiler size, combustion technology, and coal type. In this study, we classified the units into three 230 231 categories by size: large units (\geq 300 MW), medium units (\geq 100 MW and <300 MW), and small units (<100 MW). We also classified the units into three categories by 232 233 combustion technology (traditional low-NO_x burner technology (traditional LNB), 234 advanced LNB, and without LNB (Non-LNB)) and into two categories by coal type 235 (bituminous and anthracite). Table 1 summarizes the measured NO_x emission factors in China's coal-fired power plants from each category. 236

Selective catalytic reduction (SCR) and selective noncatalytic reduction (SNCR) are 237 two major de-NO_x technologies used in coal-fired power plants. In 2010, 194 238 coal-fired electric generation units (13% of the national total capacity) with a total 239 capacity of 84 GW were equipped with SCR or SNCR. However, the actual operating 240 conditions of the installed de-NO_x devices are questionable due to the lack of 241 242 inspections by local environmental protection bureaus before 2010. Our recent study 243 also found that satellite-recorded tropospheric NO₂ columns around the power plants with de-NO_x devices were stable before 2010, indicating the poor operating 244 conditions of these devices (Wang et al., 2015). In this study, we assumed that the 245

de-NO_x devices were not in operation until 2010 by setting the state factor in Eq. (1)
to zero.

Prior to 2010, LNB technology was the only widely used technology in China's power 248 plants to reduce NO_x emissions. Beginning in 1997, the use of LNB technologies in 249 China's power plants increased, following the strengthened emission standards for 250 251 thermal power plants (State Environmental Protection Administration of China (SEPA), 1996, 2003) in China. Since approximately 2005, newly established large 252 generation units have been widely equipped with advanced LNB technologies, i.e., 253 254 the stereo-staged combustion technology (Zhang et al., 2009b) and the so-called 255 "double-scale" combustion technology, which can significantly reduce the emission 256 rates of NO_x. Recent measurements of China's coal-fired power plants confirmed that NO_x emission rates from large units with advanced LNB technologies are remarkably 257 lower than units with traditional LNB technologies (e.g., Zhu et al., 2009; Zhu, 2011; 258 259 Cao and Liu, 2011; see Table 1).

260 Based on the discussion above, we assigned the appropriate LNB technology to each generation unit according to the following assumptions, given that the LNB 261 information was absent from the MEP-database: (1) All large units constructed before 262 2006 are equipped with traditional LNB, and units constructed after 2006 are 263 264 equipped with advanced LNB; (2) medium units constructed after 1997 are equipped with traditional LNB to meet the emission standards (SEPA, 1996), whereas units 265 constructed before 1997 are not equipped with LNB; and (3) no small units are 266 equipped with LNB during the study period. We then used the emission factors 267 268 presented in Table 1 to calculate the NO_x emissions for each unit.

269 **2.2.3. PM**

270 PM emissions were estimated for two size fractions: $PM_{2.5}$ and $PM_{2.5-10}$ (PM with 271 diameter more than 2.5 µm but less than 10 µm, coarse particles). The unabated 272 emission factor of PM was calculated using the following equation: 273 $EF_{k,d} = AC \times (1 - ar_k) \times f_{k,d}$ (4) where *k* represents the boiler type, *d* represents the diameter range of PM; EF_d is the emission factor of PM in diameter *d*, AC is the ash content of coal, *ar* is the mass fraction of retention ash, and f_d is the mass fraction of PM in diameter *d* to the total particulate matter in fly ash.

When calculating PM emissions, coal-fired generation units are classified into three 278 279 boiler types: pulverized coal boilers, circulating fluidized beds, and grate furnaces. 280 The boiler type information for each unit was obtained from the MEP-database. For each boiler type, the fraction of retention ash was derived from the Greenhouse Gas 281 282 and Air Pollution Interactions and Synergies (GAINS) database (Klimont, et al., 2002; 283 Amann et al., 2011), with values of 20%, 44% and 85% for pulverized coal boilers, circulating fluidized beds, and grate furnaces, respectively. The mass fraction of PM 284 in diameter d to total particulate matter in fly ash was derived from the GAINS 285 286 (Klimont, et al., 2002; Amann et al., 2011) and local databases (Zhao et al., 2010), as presented in Table 2. The ash content of coal for each unit in 2010 was obtained from 287 the MEP-database and was applied to every year. Table S3 presents the provincial 288 average of coal sulfur content and ash content for the year 2010. 289

290 The four types of technologies used in power plants to remove particulate matter are cyclones, wet scrubbers, electrostatic precipitators, and bag filters. The technology 291 292 type for each unit was obtained from the MEP-database. The removal efficiencies of 293 each technology were obtained from our previous study (Lei et al., 2011) and are shown in Table 3. Particulate matter can also be removed via wet FGD as a co-benefit 294 of SO₂ removal. In this study, we assume the same PM_{2.5} removal efficiency for wet 295 FGD equipment as that for wet scrubbers (Zhao et al., 2010). The uncertainty of the 296 effect of the assumption on PM emissions was discussed in Sect.4.1. 297

298 **2.2.4. CO₂**

299 The emission factor for CO_2 was calculated using guidelines from the 300 Intergovernmental Panel on Climate Change (IPCC, 2006), as follows:

$$301 \qquad EF_{CO_2,y} = A \times O \times 44/12 \times H_y \tag{5}$$

where *y* is the year, EF_{CO2} is the CO₂ emission factor in g/kg, *A* is the carbon content in kg-C/GJ, *O* is the oxidization rate, and *H* is the heating value in kJ/g-coal. In this study, we used 25.8 and 26.7 kg-C/GJ for the carbon contents of bituminous and anthracite coal, respectively, and 100% for the oxidization rate; these values were obtained from the IPCC guidelines (IPCC, 2006). The data source of the coal heating value is presented in Sect. 2.1.

308 **2.3. Uncertainty Analysis**

An uncertainty analysis was performed for our estimates using a Monte Carlo 309 approach. The term "uncertainty" in this study refers to the lower and upper bounds of 310 311 a 95% confidence interval (CI) around a central estimate. The Monte Carlo simulation uses specified probability distributions for each input parameter (e.g., activity data, 312 emission factors) to generate random variables. The probability distribution of 313 emissions is estimated according to a set of runs (10,000 runs in this study) in a 314 Monte Carlo framework with probability distributions of the input parameters (Lu et 315 316 al., 2011; Zhao et al., 2011). Table S1 in the supplementary information summarizes the probability distributions of all of the input parameters used to estimate the 317 uncertainties of the national total emission estimates. For parameters with adequately 318 319 measured data (e.g., NO_x emission factors), distribution functions were fitted from the distributions of those data. Probability distributions of other parameters were obtained 320 321 from previous studies (Zhao et al., 2010, 2011; Lu et al., 2011) or were based on our own discretion. 322

Uncertainties associated with emission estimates could vary with time. The uncertainties for a unit in 1990 can be considered larger than the uncertainties in 2010, for which all of the specific information is available in the CPED. In this study, we also calculated the emission uncertainties of one selected generation unit for 2000 and 2010 to demonstrate the uncertainties at the unit level. The probability distributions of the unit-level parameters are presented in Table S2 in the supplementary information. In contrast to uncertainty analyses for national total emissions, we used discrete distributions (i.e., "Yes/No" distributions) to represent the probability distributions of the technologies, which represent situations in which our assumptions about the technology for a specific unit are correct/incorrect.

333

334 3. Results

335 3.1. Evolution of technologies in coal-fired power plants

The energy efficiency of power plants in China has improved significantly over the 336 337 past two decades. As shown in Fig. 1, the average coal consumption per unit electricity supplied decreased from 407 gce/kWh in 1990 to 327 gce/kWh in 2010, 338 representing an improvement of 20% in energy efficiency over 20 years. This 339 340 significant change could be attributed to the measures imposed by the Chinese government to encourage large-scale power units and to decommission small units. 341 Figure 1 also presents the variation trend in the share of units of different sizes from 342 343 1990 to 2010. The share of the unit capacity of large units (\geq 300 MW) increased sharply from 18% in 1990 to 74% in 2010, whereas the share for small units (<100 344 MW) dropped to 9%. In particular, the construction rate of large units equal to or 345 346 larger than 600 MW began to accelerate after 2005. The capacity of units equal to or larger than 600 MW was only 46 GW in 2005 but increased to 262 GW by 2010, 347 accounting for 39% of the national total capacity. 348

Figure 2 further examines the measures taken to drive the rapid change from 349 350 2005-2010. To fulfill the increasing demand for electricity, China constructed 417 GW capacities from 2005–2010, of which 83% were large units. Fig. 2(a) shows the 351 352 growth of new power units since 2005. During this time, large units began to account for a greater share of new units. For all of the newly constructed units, the percentage 353 of large units increased significantly from 29% to 49% from 2006 to 2010, whereas 354 the percent of small units decreased from 57% to 41%. In addition, the construction of 355 new power generation capacity decreased from 86 GW in 2006 to 66 GW in 2010. In 356 the meanwhile, China has taken measures to phase out low-efficient power plants. 357

Figure 2(b) illustrates that small units, especially those smaller than 25 MW, constitute the largest component of retired units, accounting for 89% of the number of retired units in 2006. However, this ratio dropped to 62% in 2010 because the phase-out strategy gradually pursued larger units once the majority of units smaller than 25 MW had been phased out. The average capacity of the units retired in 2010 was 40 MW, three times the value in 2006 (13 MW).

The great effort from 2005 to 2010 to construct large units and phase out small units 364 significantly improved China's power plant energy efficiency, which is indicated by 365 366 the shift of the coal consumption rate shown in Fig. 3. Figure 3 compares the number 367 of plants by coal consumption rate (gce/kWh) in 2005 and 2010. In 2005, 62% of power plants in China had a coal consumption rate of 400-700 gce/kWh, and 20% of 368 power plants had a consumption rate greater than 700 gce/kWh. In 2010, 57% of 369 370 power plants in China had a coal consumption rate of 400 gce/kWh or lower. Generally, large units consume less coal than small units for the same amount of 371 electricity generated because of the more advanced combustion technology used in 372 373 larger units such as supercritical and ultra-supercritical. From 2005 to 2010, with the 374 increase in the number of large units, the average coal consumption rate decreased 375 from 356 gce/kWh to 327 gce/kWh, representing an 8% total efficiency improvement 376 from 2005–2010.

377 3.2. Inter-annual Emissions

Figure 4 and Table 4 summarizes the emissions of each species from China's 378 coal-fired power plants during 1990-2010. The total coal consumptions in China's 379 coal-fired power plants increased significantly by 479% in China from 1990 to 2010, 380 whereas SO₂ emissions from the power plants increased by 56%, NO_x emissions 381 increased by 335%, CO₂ emissions increased by 442%, PM_{2.5} emissions decreased by 382 23%, and PM_{10} emissions decreased by 27% during the same period, indicating that 383 384 significant technological changes occurred in the power sector. Table 4 also presents 385 the variation in technology penetration rates and emission factors of coal-fired power 386 plants from 1990 to 2010.

387 3.2.1. SO₂

Figure 4 shows the SO₂ emissions from power plants estimated in this study. From 388 1990 to 2005, SO₂ emissions increased at an annual rate of 8%, driven by the 389 390 ever-increasing demand for electricity, at a growth rate of 10%. The improved energy 391 efficiency and co-benefit of wet scrubbers on SO_2 removal slightly mitigated the emission growth trend. In 2005, to control emissions, China began to require the 392 393 installation of FGD in power plants (Table 4). Therefore, the SO₂ emissions peaked at 394 16.7 Tg in 2006 and began to decrease sharply. By 2010, 84% of the total unit capacity in our database was equipped with FGD, which was estimated to reduce SO₂ 395 emissions to 7.7 Tg, 54% lower than the 2006 emission level. 396

397 Figure 5 presents the FGD installation process. As shown in Fig. 5, in 2006, FGD was primarily installed for new units, and the share of unit capacity installed with FGD 398 was 69% for new units, whereas it was only 15% for those over 10 years old. 399 400 Influenced by the premium price for desulfurized electricity and the penalties incurred for non-desulfurized electricity since 2007 (Xu et al., 2009), the deployment of FGD 401 sharply increased for new and aged units. As Fig. 5 shows, there was no difference in 402 403 the FGD installation ratio between new and aged units younger than 20 years old in 2010, and the share of the unit capacity with FGD reached 63% for units over 10 404 years old. 405

However, the SO₂ removal efficiencies vary among the different units. As presented in 406 Fig. 6, FGD equipped on larger units exhibited better SO₂ removal efficiencies than 407 408 that on small units. In 2010, the average SO₂ removal efficiencies were approximately 80% for large units but only 60% for small units. Figure 7 presents the cumulative 409 ratio of SO₂ emissions by unit size for 2005 and 2010. The cumulative ratio of the 410 411 unit capacity was comparable to that of the SO₂ emissions in 2005 (Fig. 7a), but they 412 differed significantly in 2010 (Fig. 7b). The capacity share of small units decreased 413 from 20% in 2005 to 9% in 2010, but the contribution to the total SO₂ emissions 414 remained unchanged at ~20%. Before 2005, the emission contribution to SO_2 of a power unit was largely dependent on its capacity because desulfurization devices were 415 416 seldom employed at that time. Thus, the cumulative ratios of the unit capacity and SO₂ emissions could be similar. However, in 2010, 92% of large units were equipped 417 with FGD, which is considerably higher than the number of small units (52%). In 418 addition, large units tend to have higher SO₂ removal efficiencies. In 2010, large units 419 contributed to 55% of the total SO₂ emissions in 2010 while comprising 76% of the 420 421 total capacity.

422 3.2.2. NO_x

423 As shown in Fig. 4, NO_x emissions from power plants continued to increase from 1990 to 2010, except for the period of 2007–2009. NO_x emissions from power plants 424 increased by a factor of 3.4 from 1990 to 2010, from 1.9 Tg (all of the values herein 425 426 are calculated as NO₂) in 1990 to 8.3 Tg in 2010. This dramatic growth was largely 427 driven by the increasing electricity demand and was partially offset by the installation 428 of LNB. Our study suggests that the average NO_x emission factor (in g/kg of coal) slightly decreased at an annual rate of 1% from 1990 to 2005 with increasing LNB 429 430 penetrations (Table 4). From 1990 to 2005, NO_x emissions increased at an annual 431 growth rate of 8.6%, comparable to the 9.4% annual growth rate of coal consumption 432 during the same period. After 2005, the decreased rate of average NO_x emissions accelerated (at 3% per year) because of the higher NO_x removal efficiencies of 433 434 advanced LNB technologies compared with traditional LNB. From 2005 to 2010, NO_x emissions increased by 126%, which is remarkably lower than the 150% increase 435 in coal consumption. Owing to the decline in emission factors and the reduction in 436 437 electricity demand led by the global economic crisis, NO_x emissions decreased in 438 2008 and 2009 but increased again in 2010 at a growth rate of 9% after recovery from 439 the economic crisis.

440 **3.2.3. PM**

441 $PM_{2.5}$ and PM_{10} emissions from power plants decreased from 1.08 and 1.79 Tg in

442 1990 to 0.83 and 1.32 Tg in 2010 respectively, with two fluctuating peaks occurring in 1996 and 2005, which were due to the combined effect of electricity demand and 443 environmental regulations. Our estimates for the period of 1990–2005 are generally 444 consistent with our previous estimates (Lei et al., 2011). The decline of emissions 445 after the first peak was driven by the technology renewal progress following the 446 implementation of the first emission standards for power plants in 1996 (SEPA, 1996), 447 and the deceleration of the Chinese economy. PM emissions rebounded after the 1998 448 449 financial crisis but decreased again after 2005, in compliance with the implementation of stricter emission standards for power plants (SEPA, 2003). PM_{2.5} and PM₁₀ 450 emissions decreased by 40% and 47% from 2005-2010 respectively, which may be 451 due to the following reasons. First, small units with poorly efficient PM emission 452 control facilities were phased out from the unit fleet. Second, electrostatic 453 precipitators and bag filters with high removal efficiencies were widely equipped in 454 generation units under the requirement of the new emission standards. In addition, 455 FGD installation further removed PM emissions from the end-pipe. Due to the 456 combination of these three factors, the average $PM_{2.5}$ and PM_{10} emission factors 457 decreased by 60% and 65% from 2005-2010 respectively, completely offsetting the 458 effect of the 50% increase in coal consumption. 459

460 **3.2.4.** CO₂

Of the examined species emitted from power plants, CO2 emissions increased most 461 rapidly from 1990 to 2010 because, in contrast to SO₂, NO_x, and PM_{2.5}, no control 462 measures were implemented to remove CO₂. We estimated that China's coal-fired 463 power plants emitted 2.8 Pg CO₂ in 2010, an increase of 442% compared with 464 465 emissions in 1990. The increase is in line with the 574% growth in electricity generation (China Energy Statistical Yearbook, NBS, 1992–2011) but is slightly offset 466 by the improved energy efficiency resulting from the spread of large and efficient 467 468 units. Due to the improvement in energy efficiency, CO_2 emissions per unit of electricity supplied were reduced by 20% from 1990 to 2010, which is a great 469 470 achievement, although far from constraining the growth of CO₂ emissions.

471 **3.3. Evaluation of Major Policies for Emission Mitigation**

This section evaluates the effects of the major emission control measures on reducing 472 SO₂, NO_x, PM_{2.5} and CO₂ emissions during the 11th five-year period (2005–2010). As 473 described in Sect. 3.1, China primarily implemented two policies for power plants 474 during this period, including the installation of FGD and the optimization of the mix 475 of generation unit fleets by promoting large power plants and decommissioning small 476 plants. We developed two hypothetical scenarios to evaluate the effects of these two 477 policies on emission mitigation, as follows: (1) Scenario I: we assumed that China did 478 479 not adjust its fleet mix, i.e., its distribution of capacity size. In this scenario, the 480 amount of newly-built capacity is the same as the actual case, but the fleet mix was assumed unchanged during 2005 and 2010. (2) Scenario II: based on Scenario I, we 481 further assumed that no new FGD installations were performed after 2005. 482

Figure 8 compares the emission differences between the hypothetical Scenarios I and II and the actual cases during 2005 and 2010. Restructuring the unit fleet resulted in coal savings by improving efficiency, which contributed to emission abatement for all of the species. In 2010, the restructuring aided in the reduction of 83.7 Tg of coal use, 4.3 Tg SO₂, 1.8 Tg NO_x, 0.4 Tg PM_{2.5} and 238.6 Tg CO₂ emissions compared with the hypothetical Scenario I.

489 The differences between the hypothetical Scenario I and Scenario II represent the 490 effects of FGD installations. As shown in Fig. 8, FGD installation was a significant 491 contributor to emission mitigation of SO₂ and PM_{2.5}. During the 6-year period from 2005 to 2010, FGD installation was estimated to reduce 51.6 Tg of cumulative SO₂ 492 493 emissions or 36% of the cumulative SO₂ emissions from power plants compared with the hypothetical Scenario II. In 2010, FGD installation prevented 16.3 Tg of SO₂ 494 495 emissions, a value that is 2.1 times higher than the total actual emissions. In addition, FGD facilities aided in reducing PM_{2.5} by 0.54 Tg in 2010, owing to the co-benefit of 496 wet-FGD on particulate matter removal. 497

498 **3.4. Spatial Distribution of Emissions**

499 Table 5 summarizes the unit fleet mix by capacity size and technology penetration rates, as well as the emission factors of China's six large interprovincial power grids, 500 501 which are named according to the regions they serve, as follows: Northeast China, 502 North China, Central China, East China, Northwest China, and South China. A significant decrease in the emission factors of each of the five species can be observed 503 504 for all of the power grids from 2005 to 2010, especially for SO₂ and PM, which is consistent with the national trend. The emission factors are different among the grids 505 506 due to their different mix of unit fleets, fuel qualities, and penetration of emission 507 control technologies. Of the six grids, the east and central grids exhibited the lowest CO₂ emission factors in 2010, primarily due to their high percentage of large units in 508 the generation mix (the capacity share of units larger than 300 MW was more than 509 510 75% in 2010) and the higher combustion efficiency of large units. The variations of SO₂ emission factors among the grids represent the differences in FGD penetration 511 512 and the sulfur content of coal. The SO₂ emission factors for the south and central grids 513 are higher than the other grids due to the high sulfur content of coal. The FGD 514 penetration rate of the northeast grid was significantly lower than that of the south 515 grid in 2010 (60.1% in the northeast versus 92.7% in the south). However, the 516 northeast grids had a lower SO₂ emission factor (2.23 g/kWh in the northeast versus 3.41 g/kWh in the south) due to the differences in the sulfur content of coal between 517 518 the two regions. The PM_{2.5} emission factors varied remarkably due to the regional 519 differences in the penetration rates of efficient PM_{2.5} removal facilities (electrostatic precipitators and bag filters). In 2010, the average PM_{2.5} emission factor in the 520 521 northeast grid was more than two times higher than that of the east grid due to its lower penetration rates of electrostatic precipitators (89.0% versus 96.2%). Because 522 an electrostatic precipitator has very high removal efficiency for PM_{2.5} (93%) 523 compared with wet scrubbers (50%) and cyclones (10%), small differences in 524 technology penetration among regions could result in significant disparities in the 525 final emission factors. 526

Figure 9 depicts the yearly evolution of the SO₂ emissions from China's coal-fired 527 power plants from 1990 to 2010 at the unit level (only eastern China is shown on the 528 map). New power plants were constructed throughout the country after 2000. 529 Particularly, large units were rapidly constructed in the north regions, where large coal 530 mines are located, and along the eastern coastal regions, where economies are most 531 active. In addition, SO₂ emissions from large units have declined significantly since 532 2005, and many small units were terminated. Figure 10 shows NO_x emissions by unit 533 534 for the years 1990, 2000, 2005, and 2010. In contrast to SO₂, NO_x emissions continuously increased over the entire study period given that no effective NO_x 535 emission control facilities (e.g., SCR) were operated after the generation units were 536 537 commissioned.

3.5. Monthly Variation of Emissions

Figure 11 presents the monthly profiles of power generation, CO_2 emissions, and SO_2 emissions from 2005–2010, which were aggregated from the monthly profiles of each unit. Power generations and emissions typically peaked in December of each year due to high year-end industrial activities, with the exception of 2008 during the financial crisis. The second emission peak occurs in July and August, which is driven by the electricity demand of air conditioners. The low point of emissions occurs in January or February of each year, depending on the time of the Spring Festival.

546 As shown in Fig. 11, monthly variations in CO₂ emissions generally follows the 547 variation in power generation, whereas the monthly variation of SO₂ emissions differs from that of the power generation after 2007 when FGD installations were widespread. 548 After 2007, the monthly fraction of SO_2 emissions was typically higher than the 549 fraction of power generation during the first half of the year but reversed during the 550 second half of the year, reflecting that many FGD facilities were installed by the 551 year-end to meet the government requirements of that year. In this case, the monthly 552 emission profiles developed in this study differ from previous inventories for which 553 554 temporal variations in power plant emissions were derived from the monthly

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electricity generation of each province (e.g., Streets et al., 2003; Zhang et al., 2007).

556 **3.6. Data availability**

The early version of CPED has been integrated into the MEIC (Multi-resolution Emission Inventory for China) database (both MEIC 1.0 and 1.2), which is available at the following website: http://www.meicmodel.org/. MIEC 1.0 was incorporated to the MIX Asian emission inventory (Li et al., in prep.). The most recent version of CPED (documented in this work) will be incorporated to the next version of MEIC.

562 **4.** Discussion

563 **4.1. Uncertainty in Emission Estimates**

564 The uncertainty ranges of emissions estimated in this study are presented in Fig. 12. The average uncertainties of emissions from coal-fired power plants in China in 2010 565 are estimated as -22% to 23% for SO₂, -15% to 15% for NO_x, -31% to 38% for 566 $PM_{2.5}$, -26% to 30% for PM_{10} and -15% to 16% for CO_2 . The higher uncertainty 567 range of the PM emission estimates is dominated by the uncertainties in the unabated 568 emission factors and the efficiencies of PM removal facilities. The development of a 569 local database of the actual removal efficiencies for emission control in the future will 570 help to reduce the uncertainties. The uncertainty ranges narrowed gradually from 571 1990 to 2010, representing the improved knowledge of the underlying data over time. 572 The uncertainty ranges declined from -36~38%, -24~26%, -43~55%, -32~39% and 573 -24~27% in 1990 to -22~23%, -15~15%, -31~38%, -26~30% and -15~16% in 574 2010 for SO₂, NO_x, PM_{2.5}, PM₁₀ and CO₂ respectively. As discussed in Sect. 2, many 575 576 of the input data in the CPED in 1990 were determined by extrapolations and assumptions associated with high uncertainties, whereas the uncertainty ranges for the 577 578 2010 emission estimates are significantly reduced because of the extensive use of 579 unit-specific data. The unit-specific annual coal use in 2010 contributed to the 580 improved accuracy for all five species. In addition, a better understanding of sulfur 581 content and removal efficiency of FGD, coal type, ash content and heating value of 582 coal for each unit in 2010, on which the accuracy of SO₂, NO_x, PM and CO₂ emission factors depend respectively, is the primary reason for the narrowed uncertainties for 583 584 corresponding species.

585 We further demonstrated how the emission uncertainties changed over time at the unit level. For the selected generation unit (600 MW, pulverized boiler, equipped with 586 FGD, LNB, and an electrostatic precipitator), the uncertainty ranges of the emission 587 estimates for 2000 and 2010 are presented in Table 6. The uncertainty ranges for the 588 2010 estimates are significantly reduced compared with the uncertainties for 2000 589 because more unit-specific information became available in 2010. For 2010, the 590 591 uncertainties at the unit level are comparable with the national average, given that all 592 of the available unit-specific input data correspond to low uncertainties. However, in 593 2000, the uncertainties at the unit level are significantly higher than the national average because several key parameters (e.g., annual operating hours, sulfur content 594 and heating value of coal) were derived from extrapolations and assumptions. 595

596 In addition, we quantified uncertainties of other potential sources by sensitivity 597 analysis. The assumption of SO₂ removal efficiencies for FGD prior to 2008 may 598 have underestimated SO₂ emissions, as some of the early installed FGD facilities were 599 not actually in operation then. Assuming 20% of FGD did not operate properly, national total emissions could increase by 2%, 4% and 9% for 2005, 2006 and 2007 600 601 respectively. The assumption of PM_{2.5} removal efficiency for wet FGD may have underestimated PM_{2.5} emissions for wet limestone-gypsum FGD. Particulate matters 602 in desulfurizers of the spray slurry from scrubbers of wet FGD are likely to exhaust 603 from stacks along with plumes. These particulate matters would offset PM_{2.5} 604 emissions absorbed by scrubbers of wet FGD (Meij and te Winkel, 2004). By 605 assuming 10% changes of PM_{2.5} emissions are induced by gypsum spray (Meij and 606 607 te Winkel, 2004), PM_{2.5} emissions could be increased by 0.3% in 2005 and 6.4% in 2010, depending on the penetrations of wet FGD. We further quantified the 608 609 uncertainties induced by the assumption that de-NO_x devices were not in operation until 2010. By assuming that de-NO_x devices were put into operation in Beijing, 610 611 Shanghai and Guangdong in 2010, NO_x emission estimates could be reduced by 67 Gg (1% of total), indicating our assumptions have small impacts on national total NO_x 612 emission estimates. Overall, the sensitivity studies indicate that our assumptions have 613 614 relatively small impacts on national total emission estimates.

4.2. Comparison with Previous Estimates of Emission Trends 615

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In this section, we compared our new inventory with other bottom-up emission

inventories, as shown in Fig. 12, in which multi-year estimates are provided (more 617 than five data points from 1990 to 2010). The discussion is focused on inventories 618 that are available for multiple species and are widely used in the community, i.e., 619 Emission Database for Global Atmospheric Research version 4.2 (EDGAR 4.2, 620 EC-JRC/PBL, 2011) and Regional Emission inventory in Asia version 2 (REAS 2, 621 Kurokawa et al., 2013). We initially compared the CO₂ emission estimates among the 622 different emission inventories. Our estimate is consistent with Guan et al. (2012) but 623 624 is approximately 16%–25% lower than the estimates by three other studies (EDGAR 4.2, REAS 2, and Y. Zhao et al. (2013)). Our estimates are similar to those of Guan et 625 al. (2012) because both estimates used a lower coal heating value (an average of ~ 20 626 kJ/g) derived from energy statistics, which was approximately 20% lower than the 627 IPCC's recommended value (25.8 kJ/g) used in other studies. The lower estimate in 628 this study compared with EDGAR 4.2 might also be because the public electricity and 629 heat production sector in EDGAR 4.2 include emissions from heating plants. 630

For SO₂ emissions, EDGAR 4.2 and the official estimates by the MEP (China 631 Statistical Yearbook, NBS, 1997–2011) exceed the boundary of the 95% CI calculated 632 in this study. EDGAR 4.2 estimated a positive trend until 2008, which differs from 633 other studies, likely because EDGAR 4.2 failed to characterize the SO2 emission 634 control progress in China's power plants after 2005. Three other inventories (REAS 2, 635 Lu et al., 2011, and this study) provided consistent trajectories for SO₂ emissions and 636 are higher than the official estimates for the period of 1998 to 2008, likely due to 637 underreported emissions by the MEP. All of the studies presented a similar growth 638 trend for NO_x emissions over the last two decades, whereas EDGAR 4.2 and REAS 2 639 are slightly higher than the upper bound of the 95% CI calculated in this study. By 640 revisiting the local emission factor measurements (Table 1), our new estimates for 641 642 NO_x emissions are 15%–24% lower than previous estimates (Zhang et al., 2007) for the period of 1995–2004. REAS 2 used emission factors from Zhang et al. (2007) and 643 then derived higher emissions than those in this study (Kurokawa et al., 2013). REAS 644 2 concluded that NO_x emissions from China's power plants increased by 136% from 645

646 2000 to 2008, higher than the value of 125% of growth estimated in this study during 647 the same period due to different assumptions in the evolution of combustion 648 technologies. The PM emission trends presented in this study generally agree well 649 with previous studies (Lei et al., 2011; Y. Zhao et al., 2013) but significantly differ 650 from REAS 2. REAS 2 presented a 36% increase in PM_{2.5} emissions from 2005 to 651 2008, whereas we estimated a 24% decrease during the same period, most likely due 652 to different assumptions regarding the penetration of PM_{2.5} removal devices.

4.3. Comparison with the CARMA Database

The CARMA database (Wheeler and Ummel, 2008; Ummel, 2012) has been widely 654 used to allocate power plant emissions in different global and regional emission 655 inventories (e.g., EDGAR 4.2 and REAS 2). In this section, we compared the 656 657 magnitude and spatial distribution of CO₂ emissions between this study and the CARMA database throughout China for 2009. The total magnitude of CO₂ emissions 658 659 for the two inventories is comparable, with a large discrepancy in the numbers of power plants. In this study, we estimated 2.51 Pg CO₂ emissions from 2320 power 660 plants, whereas CARMA estimated 2.47 Pg CO₂ emissions from 945 plants. 661

Figures 13a and b show the spatial distributions of CO₂ emissions for CPED and the 662 CARMA database, which illustrate that CARMA neglects many small power plants. 663 Figure 13c depicts the cumulative curves of the power plant numbers sorted by CO₂ 664 665 emissions from low to high. In this study, power plants with annual CO₂ emissions less than 1 Tg accounted for 76% of the total plants, whereas the share of these plants 666 was only 44% in CARMA. In summary, CARMA omitted ~1300 small power plants 667 throughout China (annual CO₂ emissions less than 1 Tg) in 2009. In addition, for 668 power plants consisting of several generating units, CARMA may omit information 669 on partial units. For example, the Tuoketuo Power Plant located in Inner Mongolia is 670 composed of 10 generating units with a total capacity of 5400 MW. Its CO₂ emission 671 estimated by CARMA is 15.1 Tg, which is only 56% of the value estimated in this 672 673 study, indicating CARMA's significant underestimation of coal consumption for the 674 plant, which is most likely caused by missing information on some units.

Another major difference between the two inventories is the emission locations from 675 the power plants. Figure 13d shows a magnified comparison of the spatial 676 distributions of CO₂ emissions between the two inventories over the southwest region 677 of China, which illustrates the plant-specific emissions and locations. The power plant 678 locations in CARMA deviate from those in our inventory due to the different 679 geographical allocation methods used in the two datasets. In this study, the location of 680 each power plant was obtained from the MEP database and was manually verified 681 682 using Google Earth, which allowed for a high accuracy in the geographical 683 distribution of emissions. CARMA generally treats the city-center latitudes and longitudes as the approximate coordinates of the power plants in China (Wheeler and 684 Ummel, 2008). Ummel (2012) reported that the precise coordinates are only available 685 for 10% of the plants worldwide in CARMA, and the reported emissions are within 686 20% of the actual values for only 75% of plants. For 46 power plants included in both 687 CARMA and in CPED over the southwest region, the average distance between the 688 locations reported in CARMA and in CPED is approximately 50 km, indicating that 689 690 the CARMA database may be insufficient to support air quality modeling on regional 691 and urban scales.

Figure 14 further presents the relative differences in the CO_2 emission flux (g/m²) at 692 693 various spatial resolutions (0.1°, 0.5°, 1°, and 2°) in 2009 for the two datasets. The degree of differences between the two datasets is highly correlated to the spatial 694 resolution. The differences are diminished as the spatial resolution decreases. The 695 average differences between the two datasets are within 10% at a 2° resolution and 696 20-30% at a 1° resolution, indicating that CARMA has an acceptable accuracy to 697 support modeling studies at the global scale. However, at a 0.1° resolution, the 698 relative differences between the two inventories are as high as 70%, suggesting that 699 700 CARMA is not appropriate for high-resolution modeling.

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702 5. Concluding Remarks

703 This is the first study to develop a unit-based inventory of technologies, activities, and emissions for China's coal-fired power plants for the period of 1990-2010. The CPED 704 database developed in this study includes ~5700 in-use electricity generating units in 705 2010 and ~1900 retired units since 2005. From the high-resolution CPED database, 706 707 spatial and temporal variations of China's power plant emissions were presented from 1990 to 2010. In 2010, SO₂, NO_x, PM_{2.5}, PM₁₀ and CO₂ emissions from China's 708 coal-fired power plants are estimated to be 7.7 Tg, 8.3 Tg, 0.83 Tg, 1.32 Tg and 2.8 709 Pg respectively. From 1990 to 2010, SO₂, NO_x, and CO₂ emissions from power plants 710 711 increased by 56%, 335%, and 442%, respectively, and PM_{2.5} and PM₁₀ emissions decreased by 23% and 27% respectively during the same period. Energy efficiency of 712 coal-fired power plants in China has been improved by approximately 20% in 20 713 years owing to measures imposed by the Chinese government to encourage 714 715 large-scale power units and to decommission small units.

716 The most significant changes in power plant emissions occurred during 2005–2010, driven by the dramatic economy growth and offset by the strengthened emission 717 control measures. Large units were rapidly constructed in the north regions and 718 eastern coastal regions to meet the high electricity demand, while growth trend of 719 720 emissions has been effectively curbed since 2005 by installation of FGD and the optimization of the generation fleet mix. 84% of the total unit capacities were 721 equipped with FGD in 2010, which helped reducing SO₂ emissions to half of the 2006 722 723 emission level. The increasing penetration of advanced LNB after 2006 has reduced the average NO_x emission factor by 16%, but still did not constrain the growth of NO_x 724 725 emissions. New environmental regulations, including the phase-out of small units with inefficient PM emission control facilities, the widespread use of electrostatic 726 727 precipitators and bag filters, and FGD installations that has a co-benefit to PM removal, have led to the 40% decrease of PM_{2.5} emissions from 2005 to 2010. 728

Great emission reduction potentials from coal-fired power plants are expected in the near future by implementation of new policies including promotion of ultra-low emission units, decommission of flue gas bypass system, and strengthening supervision and management, etc. The removal efficiencies of existing FGD and de-NO_x devices are expected to be improved with decommission of flue gas bypass system. More efficient emission control technologies are expected to continuously come into the marketplace, with the implementation of the government plan (NDRC,
2014) which requires reducing emissions from coal-fired plants down to the level of
gas-fired plants.

The new inventory developed in this work has several advantages against previous 738 studies. First, to our best knowledge, it is the most complete coal-fired power plant 739 740 database for China with inclusion of more than 7657 in-use and retired units, enabling more accurate emission estimates at unit level. Second, CPED has dynamic 741 information for a given unit including commission/decommission time of units, 742 changes in technologies, and operating condition of emission control facilities. The 743 above information further improved the accuracy of emission estimates for every time 744 step. Third, exact locations of each unit were obtained from MEP and crosschecked 745 by Google Earth manually, which could benefit to chemical transport modelling at 746 high spatial resolution. The improved accuracy of CPED has been validated by 747 748 another recent study using satellite-recorded tropospheric NO₂ columns around the 749 power plants (Liu et al., 2015). We also compared the NOx emission trends of two isolated power plants in CPED (Tuoketuo and Yangcheng) with OMI NO₂ column 750 751 trend (Fig. S1). Good agreement between NO₂ column trend and NO_x emission trend 752 were found, indicating the reasonable accuracy of emission trend estimates in CPED. 753 Detailed information for the comparison is presented in the supplementary information. 754

Although we believe that the accuracy of CPED has been substantially improved, it 755 still has some uncertainties. Emission estimates for 1990s are thought to be more 756 uncertain than 2000s because a few parameters during 1990s were determined by 757 758 extrapolations and assumptions rather than using unit-specific data. Units retired 759 before 2005 were not included in our database. However, we believe that omitting those units would have minor impacts to the accuracy of CPED as large scale 760 retirement of coal-fired power plants were only occurred after 2005. Local 761 762 measurements for PM emission factors are still rare compared to SO₂ and NO_x, 763 leading to higher uncertainties in PM emission estimates. In recent years, continuous emission monitoring systems (CEMS) were gradually equipped in electricity 764 765 generating units, offering the opportunities of using real-time emission data. Applying CEMS data in the future will further improve the accuracy of emission estimates in 766 CPED. 767

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977	Table 1. Summary of NO _x emission factors for different types of coal-fired power plants	

Unit Size	Combustion Technology	Bituminous Coal, g/kg ^a	Average Emission Factor, g/kg ^a	Anthracite Coal, g/kg ^a	Average Emission Factor, g/kg ^a
	Advanced LNB ^b	2.88 ¹ ,3.05 ² ,3.28 ³ ,3.55 ⁴ ,4.13 ⁵ , 4.17 ⁶ ,4.64 ⁷	4.06	6.14 ⁷ ,6.58 ⁴ ,6.99 ⁸	6.50
Large (≥300 MW)	Traditional LNB	4.40 ⁹ ,4.98 ¹⁰ ,5.23 ¹¹ ,5.06 ¹² , 5.65 ⁸ , 7.78 ⁴	5.08	4.61 ¹¹ ,4.99 ¹² ,7.77 ⁷ ,7.94 ⁸ ,8.05 ¹⁰ ,8.73 ⁹	8.04
Medium (≥100 MW	Traditional LNB	4.34 ¹⁰ ,5.52 ¹¹ ,6.97 ¹³	6.78	7.07 ¹¹ ,7.56 ¹⁰	7.29
and <300 MW)	Non-LNB	5.46 ¹⁴ ,8.12 ¹¹	7.63	8.25 ¹⁰ ,12.11 ¹¹	10.46
Small (<100 MW)	Non-LNB	6.55 ¹⁵ ,6.88 ¹¹	6.66	10.01 ¹⁵ ,11.50 ¹¹	10.50

^aSample weighted mean

^bLNB: Low-NO_x Burners

Data sources: ¹Qian,2010, ²Cao and Liu,2011, ³Zhu,2009, ⁴Wang et al.,2008, ⁵Yi et al.,2006, ⁶Zhu et al.,2009, ⁷Xie et al.,2008, ⁸Wang et al.,2007, ⁹Bi and Chen,2004, ¹⁰Tian,2003, ¹¹Zhu, 2011, ¹²Zhu, et al.,2004, ¹³Feng and Yan,2007, ¹⁴Zhao et al., 2010, ¹⁵Zhao et al., 2008

978	Table 2. Summar	y of the mas	ss fractions of	of particulate	matter of	different si	ze fractions
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to the total particulate matter in fly ash for different types of boilers ^a; values are given

980 as percentages (%)

	Boiler Type				
Size Fraction	Pulverized	Circulating	Grate Furnaces		
_	Boilers	Fluidized Beds			
$PM_{>10}$	77	71	63		
PM _{2.5-10}	17	22	23		
PM _{2.5}	6	7	14		

^aData sources: Klimont et al. (2002) and Zhao et al. (2010).

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Table 3. Removal efficiencies of different control technologies for SO₂ and particulate

985 matters; values are given as percentages (%)

Technology	SO ₂	PM _{2.5}	PM _{2.5-10}	PM _{>10}
Cyclones		10	70	90
Wet scrubbers	20	50	90	99
Electrostatic Precipitators		93	98	99.5
FGD ^a	78 ^b	50	90	99
Bag Filters		99	99.5	99.9

^aFGD: Flue-gas Desulfurization

^bTime dependent parameter, 78% is the coal-consumption weighted mean efficeicney in 2010.

986	Table 4. Capacity sizes, technology penetrations	, fuel qualities, em	ission factors and emissions	of coal-fired power plants in China from 1990 to
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Category	Subcategory	1990	1995	2000	2005	2006	2007	2008	2009	2010
	<100 MW	39.3%	34.0%	29.1%	25.5%	23.1%	19.1%	15.3%	13.1%	11.5%
Capacity	[100,300) MW	48.7%	44.0%	35.7%	31.1%	29.1%	26.7%	23.9%	21.4%	18.7%
Size ^a	[300,600) MW	10.9%	19.9%	30.1%	33.4%	34.0%	33.2%	33.3%	34.3%	35.4%
	≥600 MW	1.2%	2.2%	5.0%	9.9%	13.8%	21.0%	27.5%	31.2%	34.4%
	Traditional LNB	12.0%	22.1%	38.7%	53.7%	51.8%	46.6%	44.2%	42.1%	39.4%
	Advanced LNB	0.0%	0.0%	0.0%	0.0%	7.4%	19.8%	29.2%	35.9%	42.0%
Technology	FGD	0.1%	1.0%	2.1%	12.2%	29.5%	49.9%	70.2%	81.9%	85.6%
Penetration ^a	Cyclones	7.6%	7.4%	5.2%	3.6%	3.0%	2.3%	1.6%	0.7%	0.3%
reneuration	Wet Scrubbers	46.3%	40.4%	19.0%	6.9%	6.1%	5.0%	3.9%	3.1%	2.5%
	Electrostatic Precipitators	44.3%	49.8%	72.5%	86.1%	87.5%	89.1%	90.8%	92.0%	92.8%
	Bag Filters	1.7%	2.3%	3.3%	3.4%	3.4%	3.5%	3.7%	4.2%	4.4%
	Heating Value (kJ/g-coal)	20.1	20.2	21.0	19.0	19.3	20.0	19.3	18.9	18.8
Fuel Quality	Coal Consumption Rate (gce/kWh)	406.7	389.0	374.3	356.4	351.8	343.5	335.3	330.5	327.1
	Sulfur Content (%)	1.07	1.12	1.10	1.04	1.03	1.00	0.97	0.95	0.95
	SO ₂ (g/kWh)	10.73	9.82	9.15	8.69	7.47	5.34	4.06	3.00	2.48
	NO _x (g/kWh)	4.14	3.82	3.37	3.41	3.23	2.92	2.84	2.78	2.67
Emission	PM _{2.5} (g/kWh)	2.34	1.84	1.12	0.73	0.62	0.48	0.39	0.31	0.27
Factor	PM ₁₀ (g/kWh)	3.89	3.11	1.92	1.29	1.09	0.83	0.66	0.50	0.42
	$CO_2(g/kWh)$	1126.1	1077.1	1036.5	986.9	974.1	951.2	928.4	915.1	905.6
	SO ₂ (g/kg-coal)	18.12	17.42	17.52	15.85	14.03	10.62	7.98	5.84	4.89

	NO _x (g/kg-coal)	7.00	6.78	6.46	6.23	6.07	5.81	5.58	5.41	5.26
	PM _{2.5} (g/kg-coal)	3.95	3.26	2.14	1.33	1.17	0.96	0.77	0.61	0.53
	PM ₁₀ (g/kg-coal)	6.58	5.51	3.67	2.35	2.05	1.66	1.29	0.97	0.83
	CO ₂ (g/kg-coal)	1902.9	1910.8	1984.8	1801.2	1828.1	1890.6	1822.9	1784.3	1781.9
	SO ₂ (Tg/year)	4.94	7.74	9.27	16.70	16.73	14.15	10.96	8.22	7.71
	NO _x (Tg/year)	1.91	3.01	3.42	6.56	7.24	7.75	7.67	7.62	8.29
Emissions	PM _{2.5} (Tg/year)	1.08	1.45	1.13	1.40	1.39	1.28	1.06	0.85	0.83
	PM ₁₀ (Tg/year)	1.79	2.45	1.94	2.48	2.44	2.21	1.77	1.37	1.32
	CO ₂ (Pg/year)	0.52	0.85	1.05	1.90	2.18	2.52	2.51	2.51	2.81

^aShares of coal consumption for each capacity size/technology

Table 5 Capacity sizes, technology penetrations, and emission factors of coal-fired power plants in China's six interprovincial power grids in

989	2005	and	2010
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Year Grid		Capacity Size (MW) ^a			Techno	Technology Penetration ^a				Emission Factor (g/kWh)				
		(0,100]	[100,300)	[300,600)	≥600	FGD	LNB	ESP	Content (%)	SO_2	NO _x	PM _{2.5}	PM ₁₀	CO_2
	North	20.0%	29.6%	36.4%	14.0%	14.5%	62.6%	89.1%	1.05	8.74	3.36	0.66	1.20	954.6
	Northeast	23.9%	41.2%	24.5%	10.4%	2.1%	43.7%	79.6%	0.41	4.01	3.99	1.18	2.01	1094.0
2005	East	17.6%	18.4%	39.7%	24.3%	27.1%	70.5%	92.2%	0.74	5.28	2.96	0.45	0.78	958.9
2003	Central	22.3%	30.6%	45.0%	2.1%	9.5%	57.6%	87.1%	1.45	12.56	3.76	0.96	1.75	971.4
	Northwest	23.3%	29.4%	40.4%	6.9%	0.2%	59.9%	92.0%	1.21	11.09	3.40	0.69	1.23	1001.5
	South	18.4%	30.7%	39.1%	11.8%	18.3%	66.9%	88.6%	1.45	12.66	3.52	0.74	1.28	1038.2
	North	11.1%	21.4%	38.4%	29.2%	88.0%	81.4%	91.0%	1.00	2.45	2.79	0.26	0.41	914.7
	Northeast	12.9%	24.5%	31.1%	31.5%	60.1%	73.6%	89.0%	0.51	2.23	3.32	0.55	0.88	1042.9
2010	East	10.0%	7.1%	25.7%	57.2%	94.3%	87.2%	96.2%	0.69	1.26	2.28	0.16	0.25	877.3
2010	Central	6.2%	17.5%	36.1%	40.2%	78.7%	86.6%	92.6%	1.18	3.27	2.73	0.34	0.55	821.7
	Northwest	10.1%	20.2%	39.8%	29.9%	77.0%	83.8%	95.7%	0.98	3.44	2.78	0.27	0.43	956.7
	South	4.4%	14.4%	39.8%	41.4%	92.7%	91.4%	98.0%	1.32	3.41	2.56	0.20	0.31	904.4

⁹⁹⁰ ^aShares of coal consumption for each capacity size/technology

Table 6 Uncertainty ranges of emission estimates for a large coal-fired generation unit

992	(600	MW,	pulverized	boiler,	equipped	with	FGD,	LNB,	and	an	electrostatic

	Year	2000	2010		
Species		2000	2010		
SO ₂		-58%~56%	-21%~14%		
NO _x		-100%~179%	-28%~47%		
PM _{2.5}		-61%~95%	-38%~49%		
PM_{10}		-81%~112%	-39%~44%		
CO_2		-28%~33%	-16%~18%		

993 precipitator) in China; the values represent the 95% CI around the mean



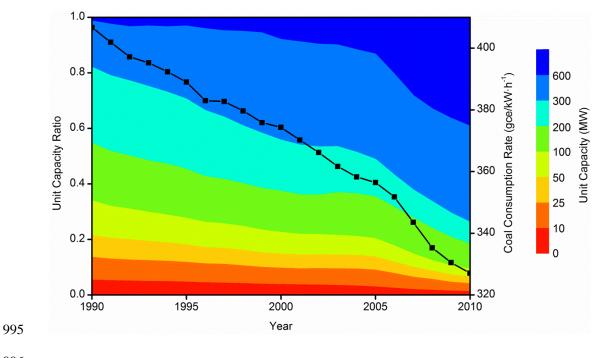
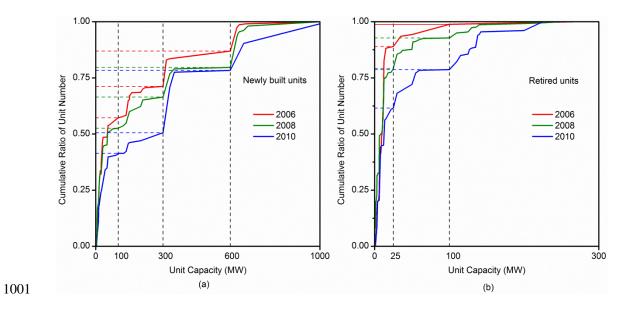


Figure 1. Trends in generation mix by capacity and the average coal consumptionrates (black line) during 1990-2010.



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Figure 2. Cumulative ratio of unit number for (a) newly constructed and (b) retired electric generating units for 2006, 2008 and 2010. The units are sorted according to ascending capacity along the x-axis.



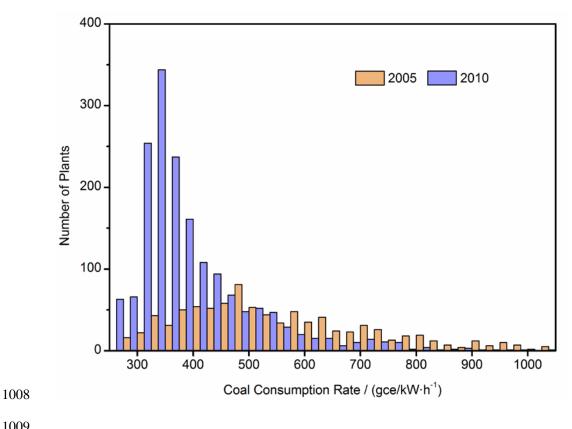


Figure 3. Distribution of coal consumption rates in coal-fired power plants in 2005 and 2010.

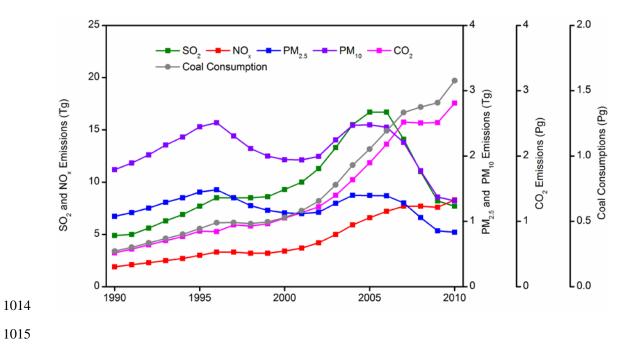


Figure 4. Coal consumptions and emissions of SO₂, NO_x, $PM_{2.5}$, PM_{10} and CO₂ of coal-fired power plants in China from 1990 to 2010.

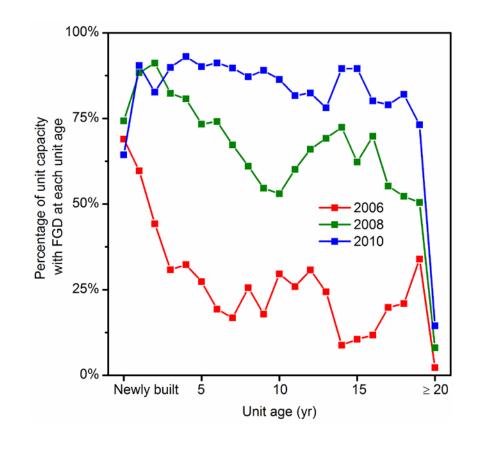
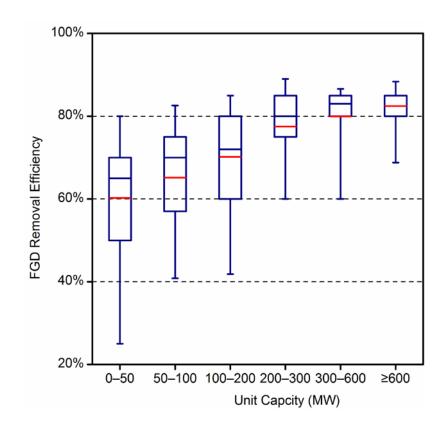


Figure 5. Distributions of FGD penetration for electric generating units of variousages in 2006, 2008 and 2010.



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Figure 6. FGD removal efficiencies for electric generating units of various sizes in China in 2010. The blue horizontal line represents the median of the removal efficiencies, the red horizontal line represents the mean removal efficiencies, the box denotes the 25% and 75% percentiles, and the whiskers denote the 5% and 95% percentiles.

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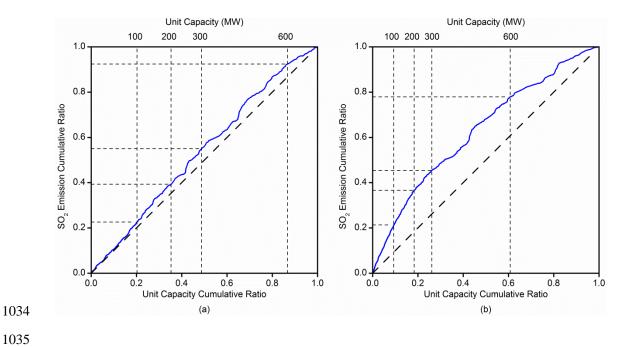
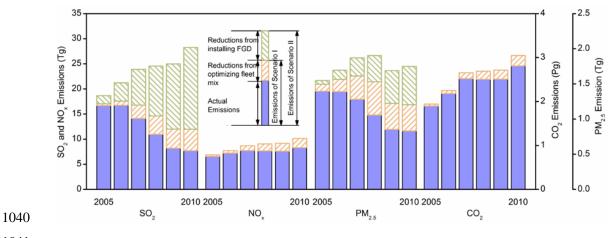


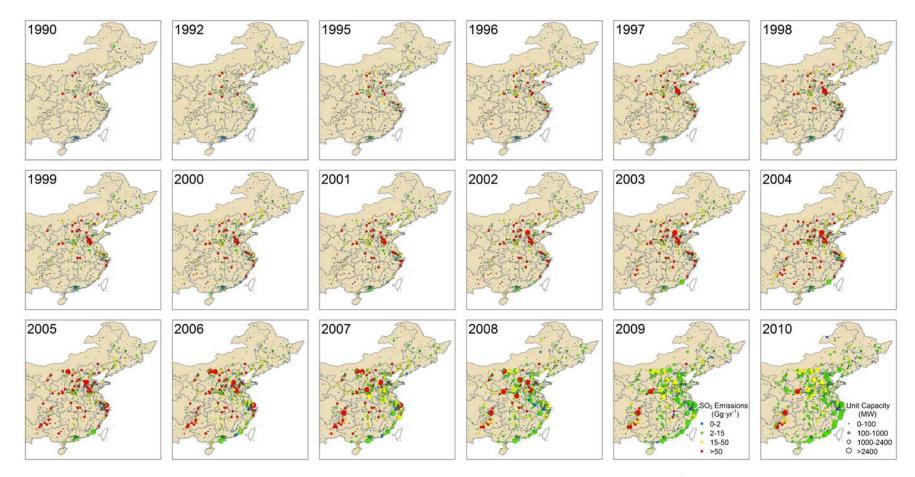


Figure 7. Cumulative ratio of SO_2 emissions by unit capacity for the years (a) 2005 and (b) 2010. The units are sorted according to ascending capacity along the x-axis.



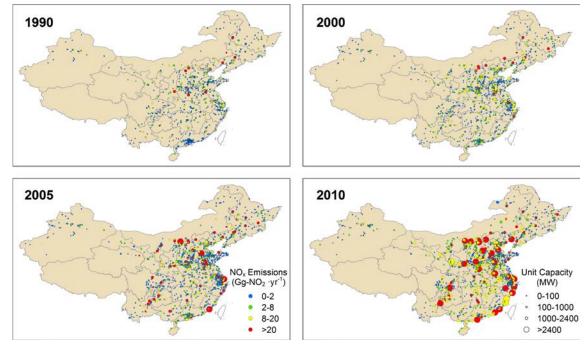
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Figure 8. Reductions in SO₂, NO_x, PM_{2.5} and CO₂ emissions from major emission control measures during the 11^{th} five-year period (2005–2010). The solid blue bar denotes our estimates of inter-annual power plant emissions. The green and yellow bars illustrate the reduction in emissions due to FGD installations and optimization of the generation unit fleet mix, respectively.



1049 Figure 9. Evolution of SO₂ emissions from coal-fired power plants in China, 1990-2010. Units: $Gg \cdot yr^{-1}$.





1052 Figure 10. Spatial distribution of NO_x emissions from China's coal-fired power plants

1053 in 1990, 2000, 2005 and 2010. Units: Gg-NO₂· yr⁻¹.

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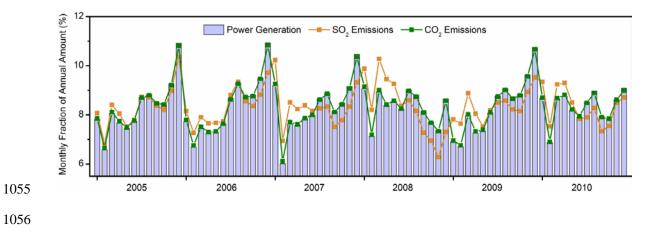


Figure 11. Monthly profiles of thermal power generation and coal-fired power plant SO_2 and CO_2 emissions in China. The y-axis values represent the fraction of the monthly emissions to annual emissions.

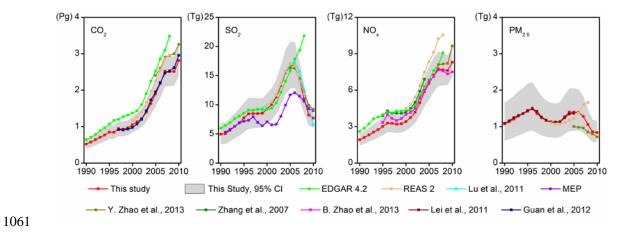
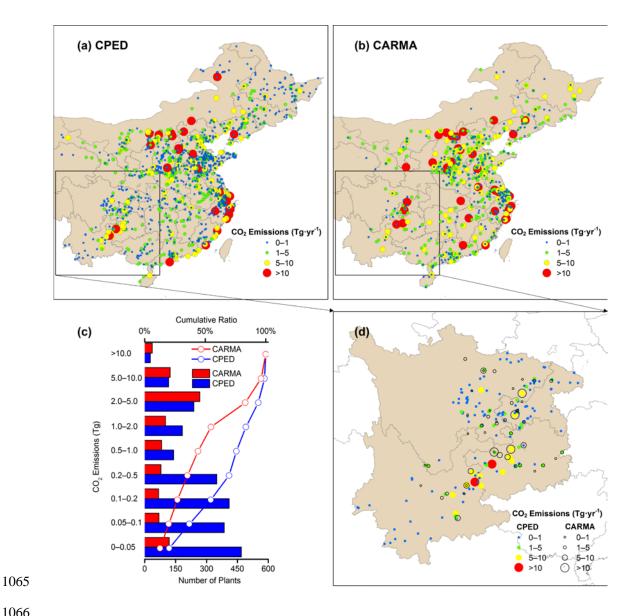
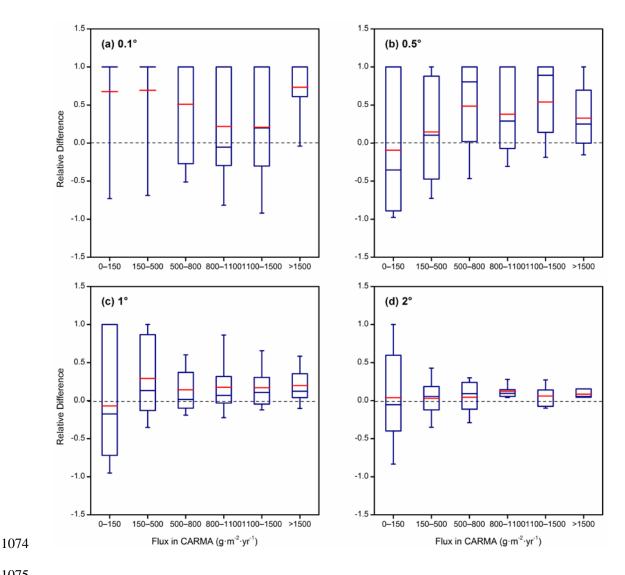




Figure 12. Comparisons of SO_2 , NO_x , $PM_{2.5}$ and CO_2 emissions from China's coal-fired power plants during 1990 and 2010.



1067 Figure 13. (a) Spatial distribution of CO₂ emissions in CPED in 2009. (b) Spatial distribution of CO₂ emissions in CARMA in 2009. (c) Comparisons of CO₂ emissions 1068 1069 between CARMA and CPED by plant numbers in 2009. The plants are sorted according to ascending CO₂ emissions along the y-axis. The red and blue lines denote 1070 the plant number cumulative ratio for CARMA and CPED, respectively. (d) 1071 Comparisons of the spatial distribution of CO₂ emissions in southwest China between 1072 CARMA and CPED in 2009. 1073



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Figure 14. Comparisons of CO₂ emissions between CARMA and CPED for various 1076 1077 spatial resolutions (from 0.1° to 2°) in 2009. The box plots show the binned relative differences (a-b)/(a+b), where a is the CARMA estimate, and b is the CPED estimate. 1078 1079 The blue horizontal line is the median of the relative differences, the red horizontal line is the mean of the relative differences, the box denotes the 25% and 75% 1080 percentiles, and the whiskers denote the 10% and 90% percentiles. A perfect 1081 agreement would correspond to a median and mean equal to zero. 1082