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CO emissions in China: Uncertainties and implications of improved energy efficiency and emission control

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ABSTRACT

A bottom-up methodology and an improved database of emission factors combining the latest domestic field measurements are developed to estimate the emissions of anthropogenic CO from China at national and provincial levels. The CO emission factors for major economic sectors declined to varying degrees from 2005 to 2009, attributed to improved energy efficiency and/or emission control regulations. Total national CO emissions are estimated at 173 Tg for 2005 and have been relatively stable for subsequent years, despite fast growth of energy consumption and industrial production. While industry and transportation sources dominated CO emissions in developed eastern and north-central China, residential combustion played a much greater role in the less developed western provinces. The uncertainties of national Chinese CO emissions are quantified using Monte Carlo simulation at −20% to +45% (95% confidence interval). Due to poor understanding of emission factors and activity levels for combustion of solid fuels, the largest uncertainties are found for emissions from the residential sector. The trends of bottom-up emissions compare reasonably to satellite observation of CO columns and to ground observations of CO_2–CO correlation slopes. The increase in the ratio for emissions of CO_2 relative to CO suggests that China has successfully improved combustion efficiencies across its economy in recent years, consistent with national policies to improve energy efficiency and to control criteria air pollutants.

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1. Introduction

Atmospheric carbon monoxide (CO) results mainly from incomplete combustion of fossil fuels and biofuels in industrial boilers and kilns, vehicles, and residential stoves, and from open burning of biomass, notably crop waste. With an atmospheric lifetime varying from a few weeks to months, depending on seasonal meteorology and geography, CO has been used as a combustion tracer to identify regional transport of atmospheric pollution.

It is difficult to quantify CO emissions precisely with bottom-up methods, particularly in countries like China that have a complex mixture of emission sources with different technologies and different levels of combustion efficiencies (Streets et al., 2006). Using observational data from ground-, aircraft-, or satellite-based instruments, atmospheric modeling studies applying inverse and/or forward methods have estimated CO emissions in China over a range of 140–230 Tg yr^{-1} for the early 2000s (especially 2001), as summarized in Streets et al. (2006) and Tanimoto et al. (2008). These results are considerably higher than the bottom-up emission inventories of the TRACE-P (Transport and Chemical Evolution over the Pacific) and REAS (Regional Emission inventory in ASia) missions, presented respectively by Streets et al. (2003) and Ohara et al. (2007). Streets et al. (2006) reanalyzed CO emissions for TRACE-P with a revised, technology-based methodology, resulting in estimated annual anthropogenic emissions in China of 151 Tg in 2001 (excluding emissions from forest and grassland fires). With the same methods but updated emission factors (EFs), Zhang et al. (2009) estimated China's CO emissions at 167 Tg in 2006, to support the INTEX-B (the Intercontinental Chemical Transport Experiment-Phase B) mission. These two recent studies generated results closer to those of the observation-based modeling, albeit still lower.

The uncertainties of CO emissions have not been rigorously quantified to date. Streets et al. (2003) and Zhang et al. (2009) relied in part on expert judgment to assume coefficients of variation (standard deviation divided by the mean, CV) of the activity levels, i.e. energy consumption or industrial/agricultural production, based on measures of economic development and perceived

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statistical quality. They used CVs of emission factors based largely on the reliability ratings of U.S. emission factors (USEPA, 2002). They estimated the uncertainties of Chinese CO emissions for 2000 and 2006, respectively, at −61% to +156% and −41% to +70% (expressed as the lower and upper bounds of a 95% confidence interval, CI, around a central estimate). More quantitative analysis is needed to investigate the uncertainties of activity levels by source and those of emission factors by technology. Based on such information, the uncertainties of emissions and the parameters contributing most to the uncertainties by sector can help identify priorities for improving the accuracy of future CO emission inventories.

Since 2006, China has implemented a comprehensive national policy to improve energy conservation and to reduce emissions. Small and old plants and boilers have gradually been replaced by larger and more energy efficient alternatives incorporating advanced emission control devices, particularly in the power sector and heavy industrial sectors like cement and iron & steel production (Zhao et al., 2008; Lei et al., 2011). Although implemented foremost for the purpose of SO_2 control, those measures can also influence CO emissions since they significantly improve the national average combustion efficiency. Thus better quantitative characterization of trends of CO emissions in recent years is important for both atmospheric modeling and for the design and implementation of policies to control multiple pollutants effectively and efficiently.

This study mainly focuses on the following topics: uncertainties in China's anthropogenic CO emissions; recent inter-annual trends in national and regional emissions; and comparisons of bottom-up emission inventories to observations of concentrations. Incorporating the latest information from domestic field measurements, a database of CO emission factors with probability distributions for China is developed by sector and technology. Based on bottom-up emission inventory methods, provincial and national CO emissions are estimated for 2005–2009, indicating the effectiveness of improved energy efficiency and emission control efforts in recent years. The uncertainties of emissions are analyzed statistically by sector using Monte Carlo simulations for the typical year of 2005. The inter-annual emissions generated with bottom-up methods are compared with satellite observations and limited ground measurements in China.

2. Methods

2.1. Method of emission inventory

The bottom-up emissions of Chinese anthropogenic CO for 2005–2009 are estimated by province and sector and then aggregated to the national level. The emissions of a given source are calculated as the product of an activity level and an emission factor (expressed as the mass of emitted pollutant per unit activity). Four main sectors are included: coal-fired power plants (CPP), all other industry (IND), transportation (TRA, including on-road and non-road sources separately), and the residential sector (RES, including combustion of fossil fuel and biomass separately). Industry is further divided into cement production (CEM), iron & steel plants (ISP), other industrial boilers (OIB), and other non-combustion processes (PRO). The detailed structure of the emission inventory is presented in Fig. S1 in the Supplementary data. To be applicable for atmospheric modeling, annual emissions at provincial level are allocated into a 0.5° (latitude) × 0.667° (longitude) grid, matched to the nested GEOS-Chem model for East Asia (Chen et al., 2009). Detailed method is provided in the Supplementary data.

To evaluate the uncertainty of CO emissions for a typical year, 2005, all of the input parameters of activity levels and emission factors, with corresponding statistical distributions (described subsequently), are placed in a Monte Carlo framework (Zhao et al., 2011a). Ten thousands simulations are then performed to estimate the uncertainties of emissions and to identify the crucial parameters that significantly contribute to the uncertainties by sector.

2.2. Activity levels

The activity levels with uncertainties of China's air pollutant emissions for a typical year of 2005 were analyzed in a previous study (Zhao et al., 2011a). Following that work, activity levels for CO emissions during 2005–2009 are compiled annually by sector and briefly described below (see Table S1–S3 and Section 2 in the Supplementary data for details).

The fossil fuel consumption and industrial production from 2005 to 2009 at provincial level are obtained from Chinese official energy (NBS, 2010a) and industrial economy statistics (NBS, 2010b). 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 (see detailed methods in Supplementary data). The accuracy of Chinese energy statistics has been discussed by previous studies and large uncertainties are believed to exist (Akimoto et al., 2006; Zhang et al., 2007). For the power sector (CPP), however, the uncertainty is small by comparisons of official statistics and the "unit-level" coal consumptions compiled by the authors (Zhao et al., 2008). A normal distribution with coefficient of variation (CV) of 5% (i.e., the 95% CI is ±9.8% around the central estimate) is assumed for the probability distribution of CPP. For other sectors, larger inconsistencies (~20%) have been found between official statistics at provincial and national levels (Zhang et al., 2007). In this study, we follow the guidelines of the Intergovernmental Panel on Climate Change (IPCC) for countries with less developed statistical systems (IPCC, 2006) and assume CVs of 10% and 20%, respectively, for the energy use of industrial and residential sectors.

The activity levels of biofuel use are taken from official statistics (NBS, 2010a), with assumed CVs of 30% in accordance with IPCC (2006). The biomass combusted in open field is calculated as a product of grain production, waste-to-grain ratio, and the percentage of residual material burned in the field. Detailed data with uncertainties are presented in Supplementary data.

For transportation, Chinese official statistics reflect only fuel used in commercial activities, and cannot thus be applied directly. Instead, the oil consumption by on-road vehicles is calculated as the product of the population of each vehicle type, annual average mileage traveled per vehicle by type, and average fuel economy of vehicle types. A normal distribution with a CV of 16% has been previously estimated by the authors and is applied in this work for the uncertainty of oil consumption in China's transportation (Zhao et al., 2011a). Detailed methods and data sources are provided in Supplementary data. For non-road sources including railway, inland shipping, rural and construction machines, the fuel consumptions in 2005 are taken from Zhang et al. (2008), while those for 2006–2009 are scaled by province according to the growth of passenger and freight traffic through railway and shipping, and the total power growth of agricultural and construction machines. All those data are obtained from official statistics (NBS, 2010c). With little information available for uncertainty analysis of non-road traffic fuel consumption, the probability distribution is assumed to be same as that for on-road vehicles.

The fractions of different technologies together with uncertainties for 2005 were discussed in Zhao et al. (2011a), and the results are applied in this work. From 2005 to 2009, the technology fractions for certain sectors changed considerably and those changes significantly affect CO emission factors. This is discussed in the next section.

3. Evolution of emission factors

3.1. Coal-fired power plants

Coal-fired power boilers include pulverized combustion (PC), grate stoker, and circulating fluidized bed (CFB) boilers. As the most widely applied technology, PC is considered to have the highest coal combustion efficiency and thus lowest CO emissions. The emission factors (EFs) for PC presented in AP-42 (the EF database maintained by the U.S. Environmental Protection Agency) is 0.25 kg per metric ton of coal (kg/t) (USEPA, 2002), while such values for China obtained through the field measurements conducted before 2005 were 0.5–4.0 kg/t, as summarized in Streets et al. (2006). After 2005, PC units less than 200 MW were gradually replaced with larger ones under China's national policy of energy conservation and emission control. Emission characteristics for units ≥200 MW were tested in the field by the authors and other researchers (Yi et al., 2006). In this study, the CO emission factor for PC units (<200 MW) is taken from Streets et al. (2006), i.e., 2.0 kg/t with a uniform distribution of 0.5–4.0 kg/t. Based on the results from recent field measurements, the emission factor for PC units (≥200 MW) is estimated at 0.66 kg/t with a beta distribution through a bootstrap simulation. Details are given in Section 3 and Fig. S2 of Supplementary data.

Due to lack of recent measurements, the CO emission factor of CFB boilers is adopted as 2.1 kg/t from SEPA (1996), and a uniform distribution of 0.5–4.0 kg/t, i.e., the same as that of PC units (<200 MW) is tentatively applied. The emission factor of grate stoker boilers in power units is assumed the same as that of industrial grate boilers, i.e., 2.6 kg/t with lognormal distribution, as described in Section 3.2. The emission factors with probability distributions for different boiler types are summarized in Table 1.

As a result of the replacement of small and old power units with larger and more advanced alternatives from 2005 to 2009, the penetration rates of PC units (≥200 MW) increased from 58% to 81% while those of PC units (<200 MW) and grate stokers decreased from 27% to 11% and from 8% to 1%, respectively, according to the compiled unit-based information for the power sector updated from Zhao et al. (2008). These improvements in the power sector reduced the average CO emission factor at the sector level from 1.3 to 0.9 kg/t, as shown in Fig. 1(a).

3.2. Industrial boilers

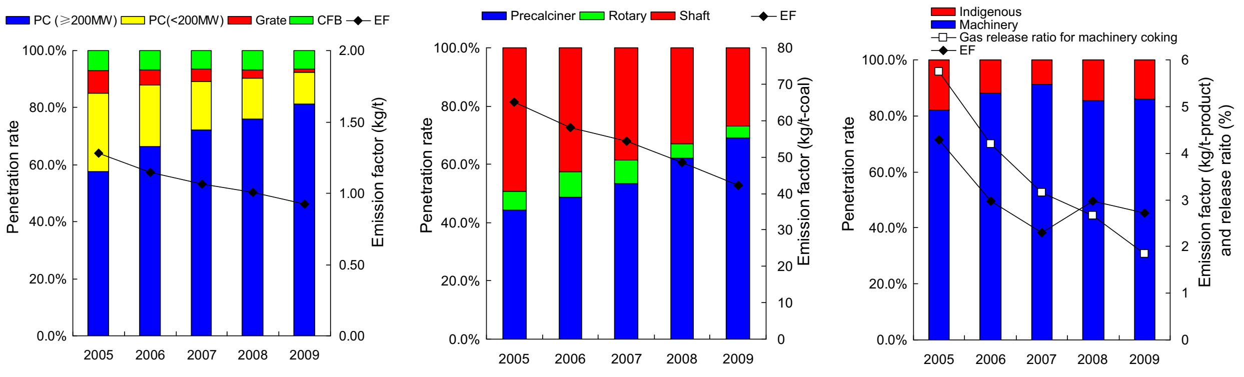
For automatic grate stokers burning coal, Streets et al. (2006) applied an emission factor of 15 kg/t, as measured for a stoker/chain watertube boiler by Ge et al. (2001). However, this result differs significantly from emission factors reported in other studies: SEPA (1996) and Wang et al. (2009a) reported values of 0.1–21 kg/t and 0.7–11 kg/t, respectively. Based on those data, the emission factor is fitted using Crystal Ball, a statistical software package, and the Kolmogorov–Smirnov test for the goodness-of-fit ($p = 0.05$). The mean value is 2.6 kg/t, much lower than that used in Streets et al. (2006), with a lognormal distribution (95% CI: 0.1–15 kg/t). The long tail of the distribution implies a relatively large uncertainty for this parameter.

Due to a lack of domestic tests, the emission factors for hand-fed grate stokers burning coal, oil, and gas are taken from USEPA (2002), i.e., 138, 0.6, and 1.9 kg/t-fuel, respectively. Given the large associated uncertainties, a normal distribution with CV of 25% (i.e., the 95% CI is approximately −50% to +50% around the central estimate) is tentatively applied for these parameters.

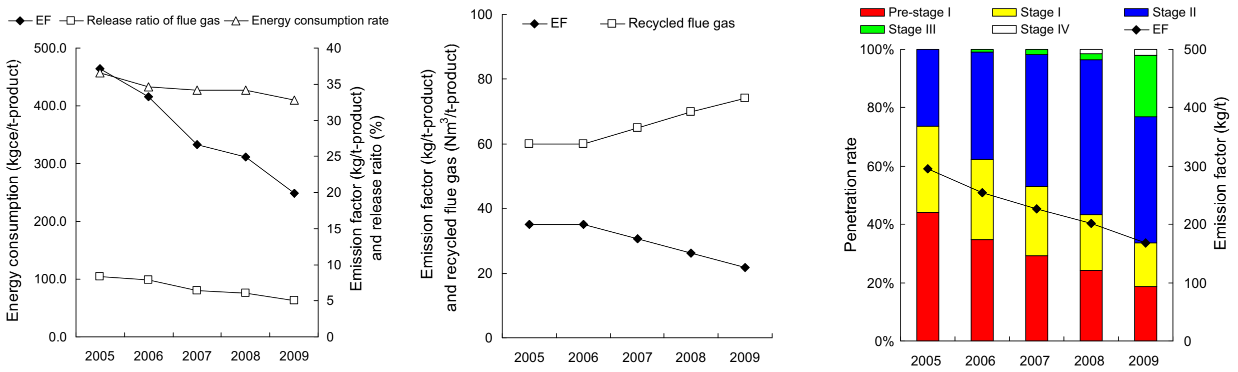
Table 1
CO emission factors with probability distributions by sector and fuel for Chinese stationary sources. Upper and lower bounds are given for uniform and triangular distributions, and 95% CIs are given for other distributions. Unit is kg/t-product for ISP, ammonia production and refinery, and kg/t-fuel for others.

Sector	Sub-sector/fuel	Emission factor	Distribution	Source
CPP	PC (≥200 MW)	0.66 (0.48, 0.86)	Beta	Tests by authors; Yi et al. (2006)
	PC (<200 MW)	2.0 (0.5, 4.0)	Uniform	Streets et al. (2006)
	Grate stoker	2.6 (0.1, 15)	Lognormal	The same as industrial automatic grate stoker
	CFB	2.1 (0.5, 4.0)	Uniform	SEPA (1996); Streets et al. (2006)
CEM	Precalciner	12 (9, 16)	Triangular	CRAES (unpublished)
	Rotary	18 (5, 32)	Triangular	Su et al. (1998); CRAES (unpublished)
	Shaft	115 (75, 175)	Triangular	Su et al. (1998); CRAES (unpublished)
ISP	Machinery coke	1.8 (1.4, 2.5)[a]	Gamma	Streets et al. (2006); CISA (2010)
	Indigenous coke	15.6 (0.8, 29)	Beta	Streets et al. (2006)
	Sintering	22 (11, 33)	Normal	USEPA (2002)
	Foundry	41 (0, 73)	Uniform	USEPA (2002)
	Iron production	37 (27, 55)[a]	Gamma	Streets et al. (2006); CISA (2010)
	BOF steel	35 (25, 48)[a]	Weibull	Streets et al. (2006); CISA (2010)
	EAF steel	9 (4.6, 13.4)	Normal	USEPA (2002)
OIB	Automatic grate stoker	2.6 (0.1, 15)	Lognormal	SEPA (1996); Ge et al. (2001); Wang et al. (2009a)
	Hand-feed stoker	138 (69, 207)	Normal	USEPA (2002)
	Oil boiler	0.6 (0.3, 0.9)	Normal	USEPA (2002)
	Gas boiler	1.9 (1.0, 2.9)	Normal	USEPA (2002)
PRO	Brick production	150 (75, 225)	Triangular	Tang et al. (1995)
	Lime production	115 (75, 175)	Triangular	The same as shaft kilns of cement
	Ammonia production	142 (6, 242)	Triangular	SEPA (1996)
	Refinery	10 (0, 49)	Uniform	USEPA (2002)
RES	Grate stoker	2.6 (0.1, 15)	Lognormal	The same as industrial automatic grate stoker
	Hot water system	12 (0.2, 74)	Lognormal	SEPA (1996)
	Small coal stove	73 (0, 150)	Normal	Zhang and Smith (2000)
	Oil boiler	0.6 (0.3, 0.9)	Normal	USEPA (2002)
	Gas boiler	1.9 (1.0, 2.9)	Normal	USEPA (2002)
	Straw waste stove	101 (28, 181)	Gamma	Zhang and Smith (2000); Li et al. (2007a); Wang et al. (2009b)
	Firewood stove	67 (25, 117)	Gamma	Zhang and Smith (2000); Li et al. (2007a); Wang et al. (2009b)
	Biomass open burning	56 (36, 83)	Uniform	Li et al. (2007b)

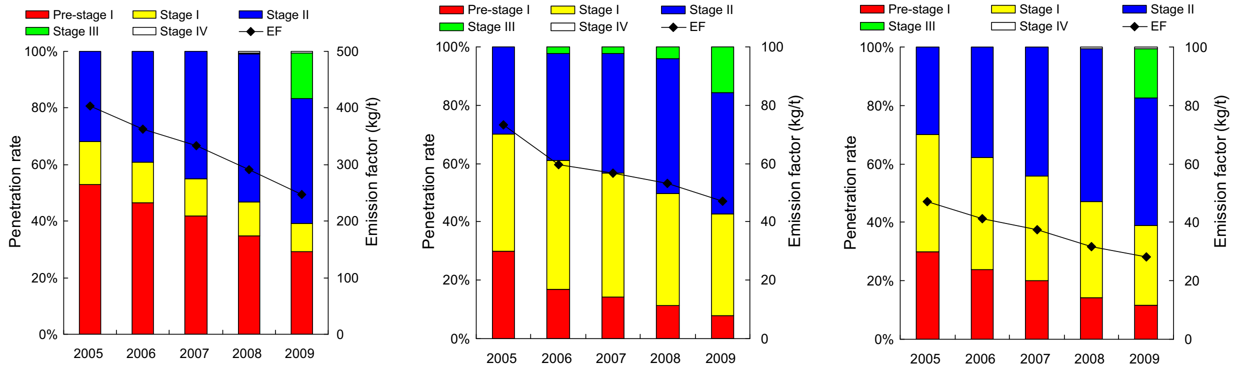
[a] Values for 2005 only.



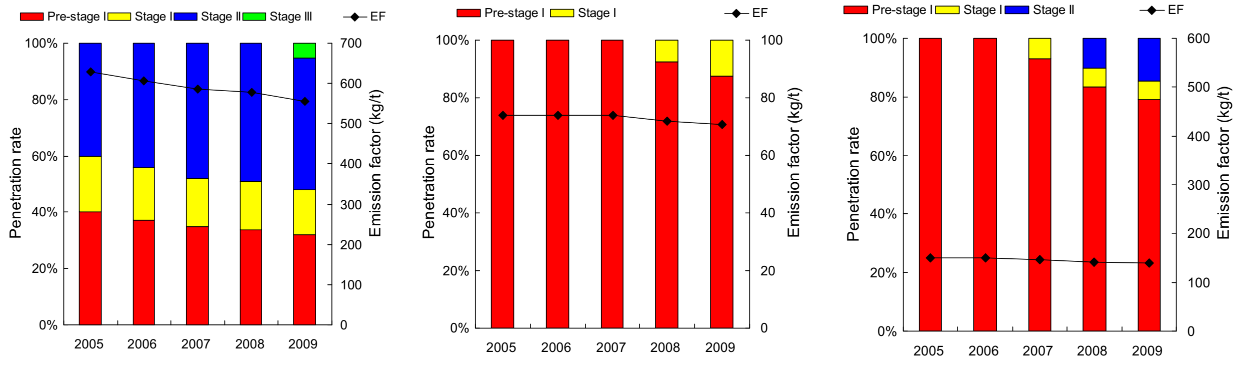
a Coal-fired power plants **b** Cement **c** Coke production



d Pig iron production **e** BOF steel production **f** Light-duty gasoline vehicles and trucks



g Heavy-duty gasoline vehicles **h** Light-duty diesel vehicles and trucks **i** Heavy-duty diesel vehicles



j Motorcycles **k** Construction and rural machines **l** Rural vehicles

Fig. 1. The average CO emission factors of given source types at the sector level for 2005–2009. (a) Coal-fired power plants; (b) cement; (c) coke production; (d) pig iron production; (e) BOF steel production; (f) light-duty gasoline vehicles and trucks; (g) heavy-duty gasoline vehicles; (h) light-duty diesel vehicles and trucks; (i) heavy-duty diesel vehicles; (j) motorcycles; (k) construction and rural diesel machinery; and (l) rural vehicles and tractors.

3.3. Cement production

Cement kilns contains shaft, precalciner, and other rotary kiln technologies. Based on field tests, Su et al. (1998) reported CO emission factors for shaft and rotary kilns of 110–175 kg/t-coal and 5–32 kg/t-coal, respectively. The emission inventory of Streets et al. (2006) applied averages of these results, 156 and 18 kg/t, respectively. More recently, measurements by the Chinese Research Academy of Environmental Science (CRAES) indicated that emission factor for shaft kilns decreased to 73 kg/t, and that for precalciner kilns, the most energy-efficient technology, ranged from 9 to 16 kg/t (unpublished). These data imply that the emission level for Chinese precalciner kilns remains higher than that realized in US. USEPA (2002) reported values of 0.49–1.8 kg/t-clinker, or 2.8–10.4 kg/t-coal based on the assumption that 1 ton of cement is produced from 0.72 ton of clinker, and that production of 1 ton of cement requires 125 kg of coal in precalciner kilns (Lei et al., 2011). In this study, averages of the emission factors by Su et al. (1998) and CRAES are applied as the mean values, i.e., 12, 18, and 115 kg/t for precalciner, other rotary, and shaft kilns, respectively. Since current available data are insufficient for data fitting, the emission factor is assumed to have a triangular distribution, which accepts the minimum and maximum values from field measurements as the lower and upper bounds for the corresponding technology, as shown in Table 1.

From 2005 to 2009, the application rate of precalciner kilns increased from 44% to 69%, while that of shaft kilns declined from 49% to 27% according to official statistics. This technology improvement drove the emission factor at the sector level down from 65 to 42 kg/t, as shown in Fig. 1(b).

3.4. Iron & steel production

The iron & steel industry employs five processes as shown in Fig. S1 (although part of coke production occurs outside of the iron & steel industry in China, we include it in this sector for simplicity of classification). Following the methodology of Streets et al. (2006), the CO emission factor for each process is calculated based on the flue gas volume per unit production, CO concentration of flues gas, and the release ratio of flue gas (the fraction of flue gas that releases to the atmosphere).

For coke production, about 500 m^3 of flue gas ($\sim$5% is CO) is generated when 1 ton of coke is produced (Streets et al., 2006), and the release ratio for 2005 is estimated at 6% and 50% for machinery (CISA, 2010) and indigenous coking (Streets et al., 2006), respectively. Without further detailed information, a uniform distribution (±20% around the central estimate) and a normal distribution with a CV of 10% are assumed for the flue gas volume per unit production and for the release ratio for machinery coking, respectively. The release ratio for indigenous coking can vary over a wide range and a uniform distribution 6%–100% is tentatively assumed. Based on these data, the emission factors for machinery and indigenous coking are calculated as 1.8 and 15.6 kg/t-product, with a gamma and a beta distribution respectively, as shown in Table 1. Due to the retirement of indigenous coking, the national application rate of machinery coking increased from 82% to 91% during 2005–2007, but dropped again to 85% in 2008 (NBS, 2010b). The release ratio of coke oven gas for machinery coking decreased from 5.7% to 1.8% between 2005 and 2009, attributed to improved technologies (CISA, 2010). The improvement reduced the national average emission factor for coke-making from 4.3 to 2.7 kg/t-product, as shown in Fig. 1(c).

For iron production, the flue gas volume is 1000–1500 Nm^3/t-product, and the CO concentration is 25%–35% (Streets et al., 2006). Uniform distributions are assumed. According to CISA (2010), the release ratio of flue gas from blast furnaces is 8.4% for 2005, and we assume a normal distribution with CV of 10% is assumed. Based on these data, the CO emission factor is calculated at 37 kg/t-product, with a gamma distribution as shown in Table 1. Through the retirement of small plants and improved use of waste heat, the energy consumption rate per unit product and the release ratio of flue gases decreased from 457 to 411 kgce/t-product and from 8.4% to 5.0%, respectively, during 2005–2009 (CISA, 2010). Correspondingly, the average CO emission factor declined from 37 to 20 kg/t-product, as shown in Fig. 1(d).

Steel production furnace technologies include open-hearth, basic oxygen (BOF), and electric arc (EAF) furnaces. During the study period, the shares of BOF and EAF remained stable, i.e., 88% vs. 12%, while the fraction of open-hearth furnaces was insignificant. For BOF, the flue gas volume is about 100 Nm^3/t-product (±20% around the central estimate is assumed for the uncertainty range), and CO concentration is 65–75% (Streets et al., 2006), with assumed uniform distribution. The recycled flue gas from blast furnaces is 60 Nm^3/t for 2005 (CISA, 2010), and we adopt a normal distribution with CV of 10%. The emission factor is calculated then as 35 kg/t-product with a Weibull distribution, as shown in Table 1. Through the improved use of waste heat, the recycled flue gas increased from 60 to 74 Nm^3/t during 2005–2009 (CISA, 2010; Wang, 2011), and the average CO emission factor decreased from 35 to 22 kg/t-product, as shown in Fig. 1(e). For EAF, the emission factor of 9 kg/t-product is taken from AP-42 (USEPA, 2002), with assumed normal distribution (CV: 25%), due to lack of domestic information.

For sintering process, similarly, the CO emission factor of 22 kg/t-product is taken from AP-42 (USEPA, 2002), with assumed normal distribution (CV: 25%). For foundries, AP-42 provides the emission factors ranging 0–73 kg/t-product according to different levels of technologies. Without detailed investigation of Chinese technologies, we applied the average of these values with a uniform distribution in this study, as shown in Table 1.

3.5. Other industrial processes

Other industrial processes include brick production, lime production, ammonia production, and refining. There are limited domestic test data on those processes, and the emission factors are thus determined largely based on authors' assumptions.

For brick making, Tang et al. (1995) reported an emission factor of 150 kg/t-coal for Chinese tunnel kilns, which is used in this study. Given that the value is quite old, a large uncertainty is assumed and a triangular distribution (±50% around the central estimate) is applied. Under the national policy of energy conservation from 2005 to 2009, the share of solid clay bricks in China decreased from 86% to 52% (CBTIA, 2006; Xu and Wang, 2007; SBMIA, 2009), and the national average coal consumption per 10,000 bricks is estimated to have declined from 1.20 to 0.97 tce, based on coal consumption rates of 1.3 tce/10,000 solid clay bricks and 0.6 tce/10,000 hollow ones (Xu and Wang, 2007). The emission factor, expressed as kg CO/10,000 bricks, declined thus from 252 to 204 during the period.

For lime production, insufficient domestic measurements are available, and we have to apply the emission factor of shaft kilns for cement, i.e., 115 kg/t-coal.

CO emissions from ammonia production come mainly from the process using coal as a raw material, which has a market share of 71% (Zhou et al., 2010). SEPA (1996) reported a range of 6–242 kg/t-product for emission factors with a central value at 142 kg/t-product. These values are adopted in this study, assuming a triangular distribution.

For refining, AP-42 reported emission factors in the range 0–49 kg/t-product according to different levels of technologies (USEPA, 2002). Lacking detailed domestic information in China, the average of those values is applied in this study with a uniform distribution, as shown in Table 1.

3.6. Residential boilers

Residential coal boilers include grate stokers, hot water systems, and small stoves. For grate stokers, we assume that the emission factor is the same as that for industrial applications. For hot water systems, SEPA (1996) reported emission factors in the range 0.2–42 kg/t, and the mean value is fitted at 12.1 kg/t with a lognormal distribution (95% CI: 0.1–15 kg/t). For small stoves, Zhang and Smith (2000) conducted field measurements of 28 fuel/stove combinations (including other fuels like biofuel) in China, yielding CO emission factors for coal stoves in the range 9–166 kg/t. The mean value is fitted at 73 kg/t with a normal distribution (CV: 55%). To avoid minus values sampled during the Monte Carlo simulation, the distribution curve was truncated at zero.

The emission factors for residential oil and gas boilers are taken from AP-42 (USEPA, 2002), and normal distributions with CV of 25% are assumed.

Domestic measurements were conducted by Zhang and Smith (2000), Li et al. (2007a), and Wang et al. (2009b) for biofuel stoves, and 19 and 21 data points are available for the burning of firewood and agricultural wastes, respectively. Based on this data, gamma distributions are fitted for both with mean values of 67 and 101 kg/t, respectively. For biomass open burning, uniform distribution is assumed, of which the ranges (36–83 kg/t) are taken from the results of field measurements by Li et al. (2007b).

3.7. Transportation

For on-road sources, seven vehicle types are assessed: light-duty gasoline vehicles (LDGV), light-duty diesel vehicles (LDDV), light-duty gasoline trucks (LDGT), light-duty diesel trucks (LDDT), heavy-duty gasoline vehicles (HDGV), heavy-duty diesel vehicles (HDDV), and motorcycles (MC). Since 1999, staged emission standards (Stage I–IV, equivalent to Euro I–IV) for new vehicles have been implemented nationwide. In addition, they were implemented in Beijing earlier than in other provinces. The fleet compositions of control stages for 2005–2009 were deduced based on the annually reported new vehicle registrations (NBS, 2010c) and the retirement of old ones estimated with assumptions of Chinese vehicle lifetimes by type. The average lifetimes of light-duty vehicles, light-duty trucks, and heavy-duty trucks are assumed at 15, 8, and 10 years, respectively, based on previous studies (He et al., 2005; Lei, 2008; Huo et al., in press).

Prior measurements of CO emission factors for vehicles in China by type and control stage were thoroughly investigated in this work, as summarized in Table S5 in the Supplementary data. As shown in Table S5, the measurements include on-road tests, engine tests, carbon balance calculations, and remote sensing. Results of on-road tests with advanced technologies (e.g., He et al. (2010) with SEMTECH-D and Oliver (2008) with OBS-2200) are given preference to estimate emissions from vehicles. Since more than one study is applicable for pre-Stage I and Stage I LDGV, the emission factors are calculated as the averages of original data weighted by the sampling size. For vehicle types without on-road tests, data from roadside remote sensing (Guo et al., 2007) are applied. There is currently no test on HDGV or MC by control stage, thus the results from Streets et al. (2006) (considered as pre-Stage I) and standard limits of stage I-II have to be relied upon. The same assumption is also applied for most non-road sources, except rural vehicles (RV), of which the emission factors are taken from tests by Yao et al. (2011) using SEMTECH-D. Typical fuel economies, 2.7 L/100 km for MC (He et al., 2005) and 250 g kWh^{-1} for heavy-duty engines (Chen et al., 2008; MIIT, 2010) are applied to convert the standard limits to fuel-based values.

Lognormal distributions are assumed for emission factors of mobile sources, as suggested by Kioutsioukis et al. (2004). CVs for on-road vehicles (except HDGV and MC) and rural vehicles are based on uncertainties derived by measurements, as listed in Table S5. For HDGV, MC, and other non-road sources with few domestic tests in China, the uncertainties are expected to be larger, and we tentatively adopt a conservative CV of 100%. All of the compiled emission factors, with uncertainties by vehicle type and control stage, are summarized in Table 2. From 2005 to 2009, the increasing proportion of vehicles meeting stricter standards resulted in a significant reduction in the average emission factors for all vehicle types, as shown in Fig. 1(f–l).

4. Results and discussions

4.1. Annual national emissions 2005–2009

As shown in Fig. 2(a), the total anthropogenic CO emissions in China are estimated at 173 Tg in 2005 using bottom-up methods. The value matches well with the result derived from an inverse modeling approach, 170 Tg, based on surface observations in East Asia for the same year (Tanimoto et al., 2008). Compared to other pollutants such as SO_2 and NO_X (Zhao et al., 2009), the inter-annual changes in CO emissions were relatively small even with rapid increases in energy consumption and industrial production during

Table 2
CO emission factors for Chinese mobile sources. Unit is kg/t-fuel unless noted. Lognormal distributions are assumed for the emission factors and the CVs are indicated in the parentheses.

	Pre-stage I	Stage I	Stage II	Stage III	Stage IV
On-road					
LDGV	550 (24%)	140 (93%)	55 (73%)	28 (145%)	5 (100%)
LDGT	384 (10%)	293 (14%)	225 (20%)	225 (20%)	–
LDDV/LDDT	145 (100%)[a]	47 (36%)	37 (73%)	19 (21%)	–
HDGV[a]	682 (100%)	136 (100%)	70 (100%)	22 (100%)	–
HDDV	104 (100%)[a]	30 (80%)	13 (37%)	12 (35%)	6 (100%)[a]
MC[a]	855 (100%)	834 (100%)	299 (100%)	150 (100%)	–
Non-road					
Railway[a]	8 (100%)	–	–	–	–
Inland Shipping[a]	8 (100%)	–	–	–	–
Small engine (g/kWh)[a]	734 (100%)	–	–	–	–
Machine[a]	74 (100%)	49 (100%)	–	–	–
RV-3w	178 (100%)	118 (87%)	118 (87%)	–	–
RV-4w	41 (54%)	31 (35%)	31 (35%)		

[a] The CVs are conservative estimates based on the authors' judgment, lacking alternative sources.

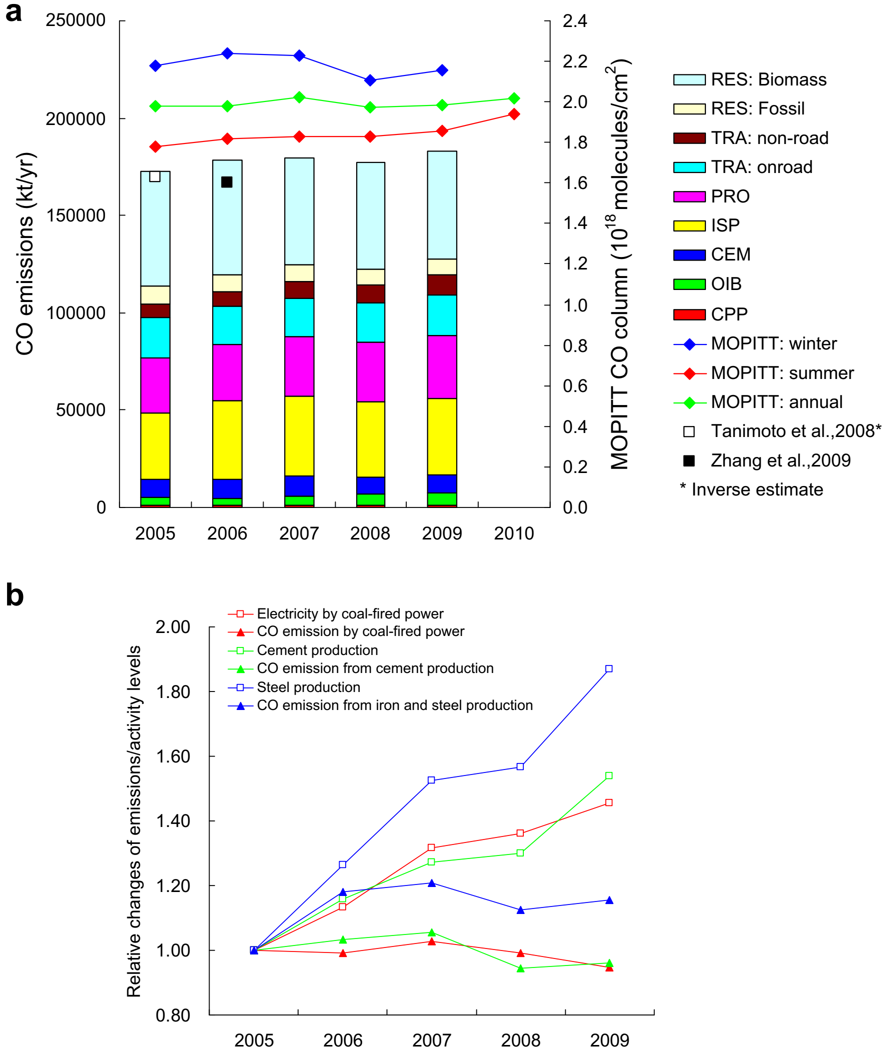


Fig. 2. Chinese CO emissions 2005–2009. (a) Emissions by sector and comparisons with other studies and satellite CO; (b) Emission and activity level trends for given sectors (all the values are normalized to the levels in 2005).

2005–2009. The estimated emissions of 2006 and 2007 are extremely close at 179 Tg, and those for 2008 decreased slightly to 177 Tg. The reasons may include (1) the improved efficiency of energy use, and the resulting reduced emission factors for major sectors, as described in Section 3; (2) reduction in the use of biofuel in recent years, an important source of anthropogenic CO for China; and (3) the relatively slow rise of activity levels in 2008, attributed largely to the strategies adopted to limit air pollution for the Beijing Olympics. In 2009, another increase is indicated and the annual emission is estimated to have reached 183 Tg, 6% higher than that for 2005.

Besides this work, Ohara et al. (2007) estimated China's CO emissions rising from 137 Tg in 2000 to 158 Tg in 2003. Although the research period is different from this work and thus the results of the two studies cannot be directly compared, our study implied a much slower growth rate of China's CO emissions in the late 2000s than that in the early 2000s indicated by Ohara et al. (2007), indicating the benefits of China's energy-saving and emission-control polices on restraining CO emissions after 2005. Another bottom-up study by Zhang et al. (2009), an improvement of Streets et al. (2003) and Streets et al. (2006), reported an annual emission of 167 Tg for 2006, 7% lower than the value derived in this study for the same year (shown in Fig. 2(a) as well). The difference is within the estimated uncertainty range of this study (described below in Section 4.4), and can be explained by (1) inclusion of open burning of biomass in this work, which is omitted by Zhang et al. (2009); and (2) the more conservative emission factors used in this work for certain industrial sub-sectors, including iron & steel production and brick making.

Interannual trends for CO emissions are compared with the results by satellite observations during the MOPITT (Measurements Of Pollution In The Troposphere) mission. The MOPITT data used here are "Level-3" monthly averaged daytime (10:00–12:00 local time) CO columns at 1° × 1° resolution (http://www.acd.ucar.edu/mopitt/products.shtml). The CO columns over mainland China are averaged for annual, summer (from June to August), and winter (from November to February) time periods during 2005–2010. As shown in Fig. 2(a), similar trends are found between the bottom-up emission estimates and MOPITT data, i.e., a slight increase from 2005 to 2007, followed by a slight decrease in 2008 and an increase again in 2009. Due to the small inter-annual variabilities and the relatively long lifetime of CO in the atmosphere, however, uncertainties in both data series could affect the comparison and lead to inconsistencies. For example, the MOPITT data in 2009 are not higher than those in 2005 except for the summer average, while the largest annual emission estimates are derived using the bottom-up method.

4.2. Emissions by sector

In 2005, industry contributed most to national emissions (43%), followed by residential combustion (40%), and transportation (16%). Due to high combustion efficiency among all sectors, coal-fired power plants emitted less than 1% of national CO. Regarding sub-sectors, biomass burning, including biofuel use and open burning, emitted 59 Tg CO (i.e., 34% of the total anthropogenic emissions) and are recognized thus as the largest sector source of CO in China. Among industrial sub-sectors, iron & steel production, brick making, and cement plants contributed 34, 18, and 9 Tg respectively to CO emissions, collectively accounting for 81% of industrial CO. From 2005 to 2009, the shares of emissions by sector changed slightly. Due to the large growth of industry, the share of industrial emissions increased from 43% to 48%, while that for residential combustion decreased from 40% to 34%, attributed to the reduced use of solid fuels including coal and biomass.

The national policy of energy conservation and emission control played an important role in abatement of CO emissions from the main sectors. For example, the total number of on-road vehicles (not including motorcycles) almost doubled from 2005 to 2009, while the corresponding CO emissions are estimated to have slightly declined by 3%. This is attributed to the application of strict emission regulations on new vehicles and to accelerated forced retirement of old vehicles with high emission levels. Fig. 2(b) shows the production outputs and emission trends for power, cement, and iron & steel plants from 2005 to 2009. During the five years, coal-fired electricity generation, cement production, and steel production increased by 46%, 54%, and 87%, respectively. For cement and steel production, there was a leveling off in 2008, attributable probably to the production constraints imposed for the Beijing Olympics and to the economic recession starting at the end of 2008. However, production increased sharply again under a major economic stimulus policy to respond to the recession, centered on a major investment in construction of new infrastructure. In contrast to the large growth of production outputs, CO emissions from power and cement plants decreased by 5% and 4% from 2005 to 2009, respectively, due mainly to the replacement of old and small plants with those with advanced technologies and high combustion efficiencies. In this study, CO emission factors for the cement sector as a whole and for the most advanced precalciner kilns are estimated respectively at 42 and 12 kg/t-coal, both of which are higher than the U.S. emission factors 3–10 kg/t-coal (USEPA, 2002). Further reduction of CO emissions can be expected with additional penetration of precalciner kilns in the future. For iron & steel plants, although the technology fractions of steel making remained stable in recent years, large benefits in CO emission control were achieved through improved use of energy (such as the capture of waste heat) and reduction of flue gas release. From 2005 to 2009, the increase in CO emission from iron & steel plants was only 19%, far less than that in steel production.

4.3. Provincial and gridded emissions

Provincial CO emissions are presented for 2005 in Fig. 3(a). Population density and concentrated economic activity largely determine the emission distribution over the country. Eastern, north-central, and south-central China, which cover only 35% of the country's territory but encompass 69% of the national population and 78% of GDP, are estimated to account for 71% of the national total emissions of anthropogenic CO in 2005. The largest emissions are found for Shandong, Hebei, and Jiangsu provinces, all of which are located in the most developed regions in eastern and north-central China. Sichuan province, which is in the southwestern China and consumes a significant quantity of biofuel, is also estimated to have high CO emissions. The shares of emissions by sector are clearly different by region. In eastern and north-central provinces, emissions from industry and transportation accounted for 69% of total emissions, while those of residential combustion were only 30%. In contrast, the shares of emissions produced by the residential sector reached nearly 50% in southwestern and northwestern provinces, attributed to greater household use of solid fuels as compared to the more economically developed provinces.

The relative changes of emissions between 2005 and 2009 by province are also illustrated in Fig. 3(a). While increasing in most provinces, the emissions for metropolitan Beijing and Shanghai and some provinces in southwestern and south-central China are estimated to have declined during these five years. However, the drivers for these reductions are different. During 2005–2009, strong measures to control emissions were implemented in Beijing and Shanghai to improve air quality for the Olympics and World Exposition, respectively. With new limits on industrial and transportation activities, the CO emissions of all sectors in the two big cities are estimated to have declined. For the southwestern provinces (Tibet is excluded due to tiny emissions), the emissions from industry and transportation are estimated to have increased by 19% and 16%, respectively, reflecting the influence of economic growth in this less developed region. The abatement of CO emissions for those provinces is attributed thus to significant reduction in solid fuel use and thereby the emissions from residential combustion.

Shown in Fig. 3(b) and (c) are the gridded emissions of China's anthropogenic CO in 2005 and 2009, respectively. Those gridded emissions can be applied for GEOS-Chem modeling.

4.4. Uncertainty analysis

The uncertainties of China's CO emissions in 2005 are summarized by sector in Table 3, expressed as the 95% CIs around the central estimates. With Monte Carlo simulations, the uncertainty of total anthropogenic CO emissions is estimated at −20% to +45% (95% CI). This result is much smaller than that based on an expert judgment reported in Zhang et al. (2009) (−41% to +70%), since random errors are significantly reduced in the Monte Carlo simulation through the "compensation-of-error" mechanism (Zhao et al., 2011a). Among all sub-sectors, the uncertainties of emissions from non-road mobile sources and residential fossil and biomass combustion are the greatest, attributed to the large variation of technologies and energy combustion efficiencies in those sources. The uncertainty of emissions from the power sector is also considerable, attributed to the conservatively assumed ranges of emission factors for small units. The large uncertainty of power sector emissions, however, has limited impact on the estimate of total emissions due to its tiny share of CO emissions. The uncertainty of the industry sector is the smallest. This result, however, does not imply that CO emission characteristics of industry are well understood, because it simply aggregates the uncertainties of all industrial sub-sectors and does not thus reflect the larger uncertainties associated with individual sources. For example, industrial boilers are poorly classified due to lack of detailed information on technologies, and the uncertainty of their emissions reaches −49% to +75%, much higher than that of total industry.

Shown in Table 3 as well are the parameters contributing to the emission uncertainty by sector. For all the sources, emission factors are identified as the parameters contributing most significantly to the uncertainty. Specifically, emission factors of CEM shaft kilns, hand-feed stokers, pre-Stage I MCs, small coal stoves, and biofuel (straw) combustion, are estimated to contribute over 50% to the variance of emissions for the corresponding sources. Moreover, due to lack of domestic information, probability distributions of certain

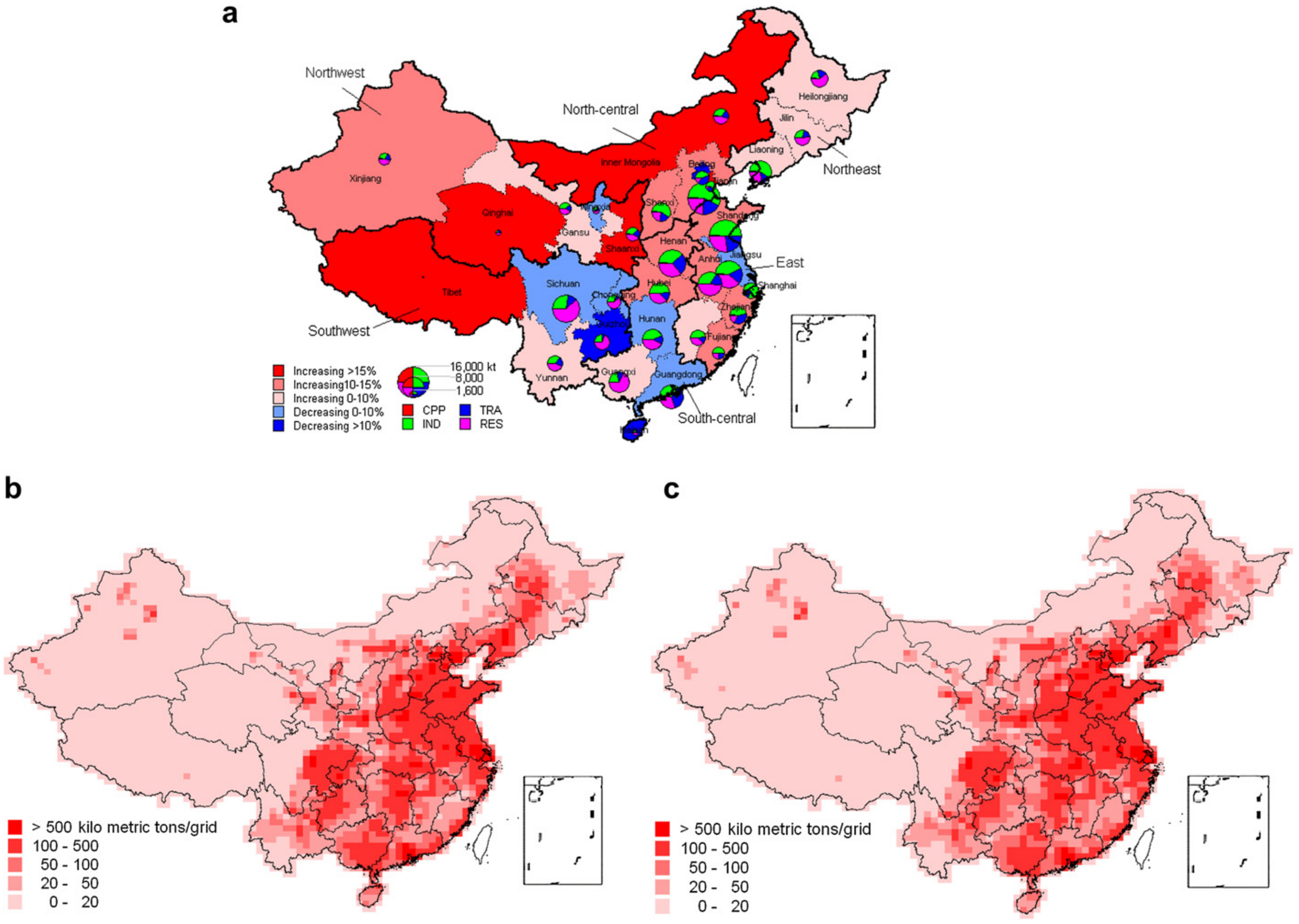


Fig. 3. Spatial distribution of China's CO emissions. (a) The provincial emissions in 2005 and the relative changes between 2005 and 2009 (the sizes of the pie graphs indicate absolute emissions by sector in 2005), and the gridded emissions in 2005 (b) and 2009 (c) at the resolution of 0.5° (latitude) × 0.667° (longitude).

parameters have to be assumed based either on foreign studies (e.g., emission factors of sintering and refinery process), or on authors' judgment (e.g., release ratios of flue gas for iron & steel production). With conservative assumptions (e.g., relatively large CVs), those parameters are also important contributors to the uncertainties of emissions. Given both large contributions to uncertainties and major shares of total emissions, more field measurements for iron & steel production, transportation, and residential combustion are suggested to narrow the CIs of emission factors and to improve the accuracies of CO emissions in China.

4.5. Comparison with surface observation of CO_2/CO

In this section, the molar ratios of CO_2 to CO of the estimated bottom-up emissions and surface observations of concentrations are compared. Wang et al. (2010) reported the inter-annual trends

Table 3
Uncertainties of Chinese CO emissions in 2005 and the parameters contributing significantly to the uncertainties. The estimated emissions are expressed as kt, followed by the 95% CIs around the central estimates. Significance of parameters to uncertainties is identified with the percentage contributions of parameters to the variance of corresponding emissions.

		Emission (95% CI)	Parameter (percentage contribution to variance)	
			1st	2nd
CPP		1382 (−43%, +95%)	EF (PC, <200 MW) (47%)	EF (grate stoker) (23%)
IND		75487 (−12%, +29%)		
	CEM	9486 (−29%, +50%)	EF (shaft kiln) (56%)	Cement production (19%)
	ISP	33938 (−20%, +28%)	EF (iron production) (26%)	EF (sintering) (16%)
	OIB	3814 (−49%, +75%)	EF (hand-feed stoker) (52%)	EF (automatic grate stoker) (15%)
	PRO	28249 (−26%, +58%)	EF (refinery) (35%)	EF (brick making) (29%)
TRA		27641 (−34%, +56%)		
	On-road	20491 (−35%, +65%)	EF (MC, pre-Stage I) (39%)	EF (LDGV, pre-Stage I) (10%)
	Non-road	7149 (−65%, +140%)	EF (rural vehicle, pre-Stage I) (34%)	EF (machine) (16%)
RES		68362 (−50%, +102%)		
	Fossil	8952 (−78%, +130%)	EF (small stove) (64%)	Coal consumption (12%)
	Biomass	59410 (−56%, +116%)	EF (straw as biofuel) (55%)	Straw production as biofuel (16%)
Total		172871 (−20%, +45%)		

Table 4
The molar ratios of CO_2 to CO of observations and bottom-up emissions in China.

	2005	2006	2007	2008
Observations				
Miyun, rural Beijing (Wang et al., 2010)	23.8 ± 1.8[a]	26.0 ± 2.0[a]	28.6 ± 2.2[a]	26.6 ± 1.8[a]
PKU, urban Beijing (Han et al., 2009)	22.8[a]	33.1[b]		
Bottom-up emissions by region				
China (MCN)	26.1	27.3	30.0	31.2
North China (NCN)[c]	27.9	27.6	30.4	31.9
Beijing	27.3	27.9	30.9	32.8
Bottom-up emissions by sector				
Coal-fired power plants (CPP)	1040	1153	1243	1306
Cement (CEM)	45	51	54	62
Iron & steel plants (ISP)	14	14	16	18
Other industry	29	29	31	33
Transportation (TRA): on-road	9	10	11	12
Transportation (TRA): non-road	21	21	22	21
Residential (RES): fossil	33	42	41	41
Residential (RES): biomass	11	10	11	11

[a] Mean values for winter, with 95% CIs for Miyun observations.
[b] mean value for fall.
[c] including Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia, Liaoning, and Shandong.

of CO_2 and CO concentrations observed at Miyun, a rural site near urban Beijing, and found that the correlations of CO_2 and CO were strongest in winter, when photosynthesis is weak and the biospheric influence on CO_2 is limited. By cluster analysis of back trajectories, an "NCN" (North China) air mass group was identified, the winter correlation slopes (dCO_2/dCO) of which were considered most representative of regional anthropogenic emissions in northern China. As summarized in Table 4, the dCO_2/dCO of NCN in winter increased from 23.8 to 28.6 during 2005–2007, followed by a decrease in 2008. Aside from the Miyun results, Han et al. (2009) reported dCO_2/dCO during 2005–2006 at an urban Beijing site at Peking University (PKU) to range from 22.8 in winter to 33.1 in fall.

To test the accuracy of the bottom-up emissions, the molar ratios of CO_2 to CO emissions (CO_2/CO) for 2005–2008 were calculated based on the annual CO emissions estimated in this study and CO_2 emissions derived using the same methodology by the authors (Zhao et al., submitted for publication). The emission ratios are determined at three spatial scales: all of mainland China (MCN), North China (NCN), and Beijing. As shown in Table 4, the national CO_2/CO increased from 26.1 to 31.2 during 2005–2008. The ratios of NCN are close to those of MCN, while those of Beijing are somewhat higher for most years. The calculated CO_2/CO ratios of MCN and NCN are also close to the observed dCO_2/dCO values in winter, particularly for 2006 and 2007, and the MCN emission ratios are within the 95% CIs of the observed dCO_2/dCO for the two years. However, the emission ratios are generally larger than the observed correlation slopes, possible explanations for which follow. The emission ratios are estimated annually, while the correlation slopes are observed for winter (or fall) only. The CO emissions for combustion processes are likely higher in winter than the annual average, due particularly to a lower combustion efficiency for the transportation sector (e.g., colder starts of vehicles) and greater residential use of solid fuels (which generally has low combustion efficiency). While open burning of biomass, which has a relatively low CO_2/CO emission ratio (the same as that of biofuel in this study) compared to other activities (see Table 4), occurs in summer and fall, its fraction of total biomass burning emissions is relatively small (13% and 19% for CO and CO_2 in 2005, respectively). That summer CO_2/CO emission ratios are believed to be higher than those in winter implies that annual emission ratios would be higher, too. Moreover, no single measurement site is able to observe a perfectly representative sample of emission sources within a region given influences of local geography and prevailing meteorology, and some differences with ratios of bottom-up emissions should be expected. Taking this and the noted disparity in annual versus winter coverage into account, the considerable consistency of the CO_2-to-CO ratios of emissions and observations should strengthen confidence both in the bottom-up estimates and in the capacity of the Miyun site to measure a reasonably representative sample of regional and even national emissions.

As shown in Table 4, almost all sectors have exhibited increasing CO_2/CO except for non-road transportation and biomass burning. Notably, the CO_2/CO of cement, iron & steel plants, and on-road vehicles increased respectively by 36%, 25%, and 37% during 2005–2009. This rise of CO_2/CO indicates successful improvement of combustion efficiencies across the Chinese economy, consistent with national policies implemented in recent years to improve energy efficiency and to control criteria pollutants.

5. Conclusions

The national and provincial emissions of anthropogenic CO in China are estimated from 2005 to 2009, with a bottom-up methodology and an improved database of emission factors combining latest domestic field measurements. Despite the rapid increase in energy consumption and industrial economy, total national CO emissions are estimated to have been relatively stable due to improvements in energy efficiency and emission control regulations. These conclusions are confirmed by satellite observations of CO columns and ground observations of CO_2–CO correlations. Field measurements on emission factors for iron & steel production, non-road transportation, and residential combustion could lead to an important reduction in uncertainties for the CO emission inventories.

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Appendix. Supplementary material

Included in Supplementary data are the source categories of China's CO emissions, the activity level data with uncertainties by sector, CO emission factor of China's coal-fired power plants ($\geq$200 MW) through field measurements, CO emission factor database of China's transportation sector by vehicle type and control stage, and the method of developing gridded emissions.

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.atmosenv.2011.12.015.

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