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Asian emissions in 2006 for the NASA INTEX-B mission

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Abstract. A new inventory of air pollutant emissions in Asia in the year 2006 is developed to support the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) funded by the National Aeronautics and Space Administration (NASA). Emissions are estimated for all major anthropogenic sources, excluding biomass burning. We estimate total Asian anthropogenic emissions in the year 2006 as follows: 47.1 Tg SO_2, 36.7 Tg NO_x, 298.2 Tg CO, 54.6 Tg NMVOC, 29.2 Tg PM_{10}, 22.2 Tg $PM_{2.5}$, 2.97 Tg BC, and 6.57 Tg OC. We emphasize emissions from China because they dominate the Asia pollutant outflow to the Pacific and the increase of emissions from China since 2000 is of great concern. We have implemented a series of improved methodologies to gain a better understanding of emissions from China, including a detailed technology-based approach, a dynamic methodology representing rapid technology renewal, critical examination of energy statistics, and a new scheme of NMVOC speciation for model-ready emissions. We estimate China's anthropogenic emissions in the year 2006 to be as follows: 31.0 Tg SO_2, 20.8 Tg NO_x, 166.9 Tg CO, 23.2 Tg NMVOC, 18.2 Tg PM_{10}, 13.3 Tg $PM_{2.5}$, 1.8 Tg BC, and 3.2 Tg OC. We have also estimated 2001 emissions for China using the same methodology and found that all species show an increasing trend during 2001–2006: 36% increase for SO_2, 55% for NO_x, 18% for CO, 29% for VOC, 13% for PM_{10}, and 14% for $PM_{2.5}$, BC, and OC. Emissions are gridded at a resolution of 30 min × 30 min and can be accessed at our web site (http://mic.greenresource.cn/intex-b2006).

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1 Introduction

In 2006 the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) was conducted by the National Aeronautics and Space Administration (NASA). The INTEX-B mission was broadly designed to (a) improve our understanding of sources and sinks of environmentally important gases and aerosols through the constraints offered by atmospheric observations, and (b) understand the linkages between chemical source regions and the global atmosphere and the implications of human influence on climate and air quality (Singh et al., 2006). INTEX-B had a spectrum of measurement objectives for which individual aircraft flights were conducted in spring 2006. One of the specific objectives of INTEX-B was to quantify transport and evolution of Asian pollution to North America and assess its implications for regional air quality and climate (Singh et al., 2009). In this respect, INTEX-B had similar goals to a predecessor NASA mission in 2001, TRACE-P (Transport and Chemical Evolution over the Pacific) (Jacob et al., 2003), which studied outflow of pollution from the Asian continent and subsequent transport across the Pacific Ocean.

The processes of interest to INTEX-B operate at a variety of scales from local to global. To efficiently represent

these scales in a flight-planning context and in post-mission data analysis, multi-scale atmospheric models are used. One such modeling system used was developed at the University of Iowa (Carmichael et al., 2003a). This system includes global-scale inputs from the MOZART global chemical transport model (Horowitz et al., 2003), the intercontinental chemical tracer model CFORS (Uno et al., 2003a), and a nested regional chemical transport model, STEM-2K3 (Tang et al., 2004). In order to drive such a modeling system, emission inventories are necessary. For the TRACE-P mission a detailed emission inventory was prepared for the year 2000 (Streets et al., 2003a, b) that has received widespread application both within the TRACE-P mission and in subsequent Asian modeling studies. To support INTEX-B it has been necessary to update the TRACE-P inventory to reflect the extremely rapid economic growth in Asia since 2001. In addition, new work was necessary to refine the temporal and spatial resolution of the emission data and to add important new species, source types, and geographical regions. This new inventory for 2006 enables a more accurate representation of Asian outflow, cross-Pacific transport, and North American inflow to be provided for INTEX-B studies.

During the past several years, China's atmospheric emissions are known to have increased markedly, following the dramatic growth of its economy and energy use. The general methodology used to build the new Asian regional emission inventory has been described in Streets et al. (2003a, b). Using the same general approach, we have implemented an improved technology-based methodology, in order to be able to reflect the types of technology presently operating in China. We also implemented a new anthropogenic PM emission model (Zhang et al., 2006) to calculate primary PM emissions, including PM_{10} and $PM_{2.5}$, which the TRACE-P inventory did not address.

The key elements of the INTEX-B inventory are listed in Table 1. The domain covers 22 countries and regions in Asia and stretches from Pakistan in the West to Japan in the East and from Indonesia in the South to Mongolia in the North (Fig. 1). In this paper we emphasize emissions from China because they dominate the Asia pollutant outflow to the Pacific and the increase of emissions from China since 2000 is of great concern.

Emissions are estimated for eight major chemical species: SO_2, NO_x, CO, nonmethane volatile organic compounds (NMVOC), particulate matter with diameters less than or equal to $10\,\mu$m (PM_{10}), particulate matter with diameters less than or equal to $2.5\,\mu$m ($PM_{2.5}$), black carbon aerosol (BC), and organic carbon aerosol (OC). Emissions of methane (CH_4) and ammonia (NH_3) were not updated from TRACE-P in this work, because they have not changed much since 2000, as confirmed by the REAS inventory (Ohara et al., 2007). In addition, CH_4 and NH_3 were low priorities of the INTEX-B mission (Singh et al., 2006). NMVOC emissions are speciated into five categories corresponding to five different chemical mechanisms (CBIV, CB05, RADM2, SAPRC99, and SAPRC07); this aspect of the inventory is described in a separate paper (Zhang et al., 2009).

Fig. 1. Definition of the inventory domain.

Only anthropogenic emissions are estimated in this work. Biomass burning emissions were included in the TRACE-P inventory, but since then a number of new high-resolution biomass burning inventories have been developed using satellite observations of burning (e.g., Duncan et al., 2003; van der Werf et al., 2006; Randerson et al., 2007) that offer superior representation of emissions for specific years. TRACE-P biomass burning emissions can still be used by modelers interested in obtaining a typical representation of Asian biomass burning. The detailed emission calculations for the 2006 INTEX-B inventory are aggregated into four source categories: electricity generation, industry, residential, and transportation.

Emission estimates in this work are specifically for the year 2006, because this inventory was prepared for the INTEX-B field campaign undertaken in spring 2006, and it was intended to reflect the actual magnitude of emissions during that period as closely as possible. However, when construction of the inventory took place in 2006 and 2007, most of the necessary statistics for Asian countries were only available for 2004/2005 and very few for the year 2006. Thus this inventory is built on a mixture of trend extrapolations from 2004/2005 and actual 2006 data.

Section 2 documents the methodology used in this work. The estimation of emissions from such a wide variety of species and regions cannot be described in complete detail due to space limitations. However, we give a general overview of methods, data, and data sources for this inventory and highlight the major advances from the previous TRACE-P inventory.

Emission estimates for the year 2006 are presented in Sect. 3. It should be noted that this 2006 INTEX-B inventory (I06) and the 2000 TRACE-P inventory (T00) are not directly comparable, because several important methodological changes were made at the same time to improve the

Table 1. Summary of the INTEX-B Asia emission inventory dataset.

Item	Description
Domain	22 countries and regions in Asia, see Fig. 1
Species	SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC
VOC speciation	by mechanism: CB04, CB05, RADM2, SAPRC99, SAPRC07
Sectors	power plants, industry, residential, transportation
Representing Year	2006
Spatial resolution	30 min×30 min
Seasonality	monthly
Data availability	available online at http://mic.greenresource.cn/intex-b2006
	http://www.cgrer.uiowa.edu/EMISSION_DATA_new/index_16.html

representation of actual emissions. In Sect. 3.1, we revisit China's emissions for 2001 (R01), the year of the TRACE-P campaign, using our new methodology. Then the differences between R01 and T00 reflect the improvements and corrections made to the T00 inventory, and the changes between I06 and R01 represent actual growth in emissions in China between 2001 and 2006. Asian emission estimates by country are presented in Sect. 3.2.

Emissions are initially calculated by country (by province for China) on an annual basis. However, emissions from some species have strong seasonal variations associated with such activities as fossil-fuel and biofuel use for home heating in winter. The seasonality in emissions is important when comparing emissions with time-specific field measurements. For this reason, we have also developed monthly emissions using a variety of methods, which are discussed in Sect. 3.3. Atmospheric models also require gridded emissions as inputs, rather than regional emission totals. Section 3.4 presents the spatial distribution of emissions at a resolution of 30 min×30 min, using various spatial surrogates. All regional summaries and gridded data can be downloaded from several websites, as described in Sect. 3.5.

In the discussion section (Sect. 4), we compare our estimates with other inventory studies. Top-down constraints on emissions also provide valuable clues for verifying emission estimates, which have been successfully used in the revision and improvement of China's CO emission inventory after the TRACE-P campaign (Streets et al., 2006). Therefore, in Sect. 4 we also compare our inventory with various top-down constraints, e.g., forward modeling, inverse modeling, and constraints from satellite and in-situ observations, and try to explain any discrepancies between inventories and top-down studies. In Sect. 4.3, we discuss the major uncertainties in this inventory and the future efforts that are needed to develop an even better understanding of Asian emissions. Finally, in Sect. 4.4, we summarize the relevant studies that have used this inventory and the implications for the present inventory.

2 Methodology

Figure 2 shows the general methodology for this inventory. We assemble the new Asian emission inventory according to the following steps. First, we implement a series of improved, technology-based methodologies to develop a new emission inventory for China. The key aspects of these improvements are documented in Sect. 2.1. This same approach was used for the development of the improved CO inventory (Streets et al., 2006), the first primary particulate emission inventory (Zhang et al., 2006, 2007a), and a new NO_x emission trend for China (Zhang et al., 2007b). We update China's emissions to the year 2006 with these new methodologies. Second, we update emissions for other Asian countries to the year 2006 following the methodology of the TRACE-P inventory but using the most recent statistics available. Third, we incorporate the best available datasets for some selected regions, where good national inventories exist that are thought to be more accurate than the TRACE-P inventory, being built on local data sources and local knowledge. In this respect we have incorporated the following external data sources into the INTEX-B inventory: SO_2 and aerosol inventories for India (Reddy and Venkataraman, 2002a, b); a 1 km×1 km high-resolution emission inventory for Japan (Kannari et al., 2007); a South Korean inventory from the National Institute of Environmental Research of Korea (NIER, 2005, 2008), and a Taiwan inventory from the Taiwan Environmental Protection Administration (Fu et al., 2009). These override the TRACE-P updates. Finally, we check for consistency among the different datasets, choose the appropriate precision for the final product, and finally export the dataset over the whole of Asia with a uniform data format.

In this section, we focus on the new methodology for China's emission estimates, because it is the major revision of the inventory compared with the TRACE-P inventory and the change that has the single largest impact on total Asian emissions.

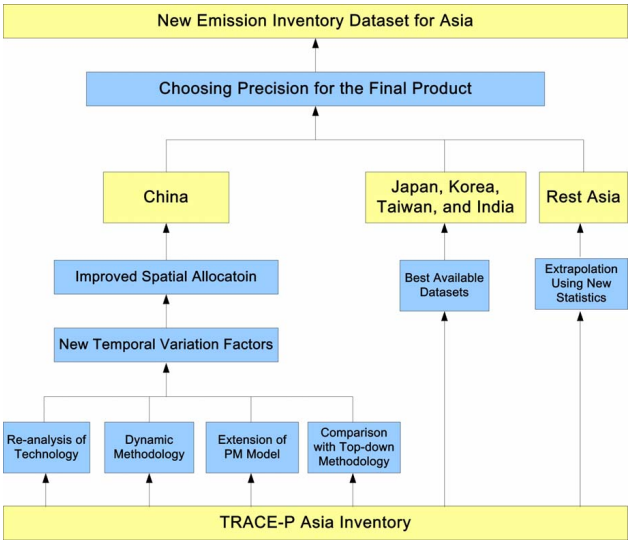


Fig. 2. Schematic methodology for the development of the INTEX-B Asia emission inventory.

The emissions of a particular species are estimated by the following equation:

$$E_i = \sum_j \sum_k A_{i,j,k} \left[\sum_m X_{i,j,k,m} EF_{j,k,m} \right] \quad (1)$$

For a given technology m, the net emission factor is estimated as follows:

$$EF = EF_{\mathrm{RAW}} \sum_n C_n (1 - \eta_n) \quad (2)$$

where i represents the province (municipality, autonomous region); j represents the economic sector; k represents the fuel or product type; m represents the technology type for combustion and industrial process; n represents a specific control technology; A represents the activity rate, such as fuel consumption or material production; X is the fraction of fuel or production for a sector that is consumed by a specific technology; EF is the net emission factor; EF_{RAW} is the unabated emission factor; C_n is the penetration of control technology n; and η_n, is the removal efficiency of control technology n.

2.1 Revision and improvement of the TRACE-P inventory for China

In this work, we have researched many aspects of the China part of the TRACE-P inventory for possible improvements by critical retrospective examination of how the original inventory was constructed and how well it performed in the various modeling and assessment projects in which it was used. We note the following major improvements thought to be necessary:

a) A detailed technology-based approach. The final release rates of pollutants greatly depend on combustion efficiency, control equipment, and operating conditions. Thus, a detailed source classification by technology level is critical for obtaining reliable emission estimates. In the TRACE-P inventory, emitting sources were usually classified at the economic sector level, say, power generation, industry, or residential, and an average emission factor was applied for the whole sector. However, in a rapidly developing country like China, both advanced and old-fashioned technologies coexist in the marketplace, which can have very different levels of emissions. For example, the CO emission factors of industrial combustion devices can vary from 2 g/kg for large, modern coal-fired boilers to 156 g/kg for old kilns, leading to an average emission factor of 85.7 g/kg for the industrial combustion sector as a whole, more than a factor of two greater than the value used in the TRACE-P estimates. We have successfully applied such a technology-based methodology to improve the CO emission inventory for China (Streets et al., 2006), and we expand the method to all species in this work.

b) Re-examination of energy statistics. Data inconsistency in Chinese energy statistics downgrades the accuracy of emission inventories that largely rely on statistics (Akimoto et al., 2006). In recent work on China's NO_x emission trend (Zhang et al., 2007b), we critically evaluated the quality and reliability of current Chinese energy statistics and used several approaches for better representation of the real-world situation in China when compiling activity data. These approaches include: using coal consumption data in the provincial energy balance tables of the China Energy Statistical Yearbooks (CESY) to reflect the actual coal production and consumption; using diesel consumption data in the national energy balance table of CESY to avoid the "lost diesel" from inter-province transportation; and a model approach for fuel consumption for each vehicle type, as these data are not available in statistics. We followed these procedures in this work. For more details, the reader is referred to Sects. 3.2, 3.3, and 4.5 of Zhang et al. (2007b).

c) A dynamic methodology representing rapid technology renewal. Potentially rapid changes of emission factors should be considered when evaluating emission trends in developing countries. In China, new technologies are constantly coming into the marketplace – sometimes to replace older technologies, sometimes not – causing rapid changes in net emission rates in just a few years. Therefore, it is necessary to develop a representation of the dynamic change in net emission factors driven by the technology renewal process, rather than simply to use year-by-year activity data with fixed emission factors. In this study, we use this strategy when revisiting the TRACE-P emissions and comparing with 2006 emissions, to get a more reliable picture of emission trends. In Sect. 2.3, we summarize the change of emission factors from 2001 to 2006 due to technology renewal and discuss the resulting emission changes in Sect. 3.1.2.

d) A size-fractioned primary PM emission inventory. The emissions of two aerosol species, BC and OC, were estimated in the TRACE-P inventory, but primary PM_{10} and $PM_{2.5}$ emissions were not reported. In this paper, we present a comprehensive estimation of primary particulate emissions in China by size distribution and major components, using a technology-based approach described in Zhang et al. (2006, 2007a). With this approach, we are able to classify particulate emissions into three size ranges, total suspended particulates (TSP), PM_{10}, and $PM_{2.5}$ (the latter two are reported in this paper), and also identify the contributions of BC and OC.

e) A new scheme of NMVOC speciation for model-ready emissions. NMVOCs differ significantly in their effects on ozone formation, and these differences need to be represented appropriately in the air quality models used to predict the effects of changes of emissions on formation of ozone. This requires appropriate methods to specify the chemical composition of the many types of NMVOCs that are emitted and appropriate methods to represent these compounds in the models. In the TRACE-P inventory, NMVOC emissions were speciated into 19 categories based on chemical reactivity and functional groups. However, these emissions are usually not ready for model use: atmospheric modelers have to map those 19 categories into the categories that their models use. This conversion process is not accurate and can introduce unpredictable uncertainties.

In this work, we improve the NMVOC speciation methodology toward an atmospheric-model-ready dataset by using a step-by-step VOC speciation assignment process. Emissions for individual VOC species are calculated by applying a state-of-the-art source profile database (e.g., Liu et al., 2008) to each source category. Then we lump individual NMVOC emissions to emitted species in different chemistry mechanisms. Up to now, we have developed model-ready emissions for five mechanisms: CBIV, CB05, RADM2, SAPRC99, and SAPRC07. The detailed description of this methodology and the results are presented in a separate paper (Zhang et al., 2009).

f) Comparison with top-down constraints. Last, but not least, top-down analytical tools applied to the interpretation of emissions provide valuable constraints to improve bottom-up emission inventories such as this one. Such techniques include forward modeling and inverse modeling using in situ and satellite observations, or even simply using observation data without models. In the years after the TRACE-P mission, these techniques have become widely used to constrain Asian emissions against a priori estimates. The results of these analyses sometimes support the inventory, while more often they raise questions about the accuracy of the inventory. In Sects. 4.1 and 4.2 of this paper, we present an intensive review of these analyses, discuss the existing discrepancies, and attempt to find a direction to reconcile the inventory in light of these findings.

2.2 Activity rates

2.2.1 China

We derive activity data for China for the years 2001 and 2006 from a wide variety of sources, with a critical examination of the data reliability. Fuel consumption in stationary combustion sources by sector and by province (A in Eq. 1) is derived from the provincial energy balance tables of the CESY (National Bureau of Statistics, 2004, 2007a), with the exception of diesel consumption. We use diesel consumption values in the national energy table of CESY and then derive shares from the provincial tables (see explanation in item *b* of Sect. 2.1). Industrial production by products and by province is derived from other governmental statistics (National Bureau of Statistics, 2002a, b, 2006, 2007b; AISIC, 2002, 2006). The methods for determining activity levels of non-energy sources for NMVOC are the same as in previous analyses (Klimont et al., 2002). The distributions of the combustion technology in each sector and the processing technology in each industrial product (X in Eq. 1) are generally not available from government statistics. In this work, these data were collected from a wide range of unpublished statistics by various industrial association and technology reports. The data sources of key emitting sources in China are summarized in Table 2.

When this inventory was developed in 2006 and 2007, most of the available statistics for Chinese provinces were for 2004/2005 and very few for the year 2006. We therefore extrapolated activity data to the year 2006 based on various fast-track statistics that are published monthly (Beijing Huatong Market Information Co. Ltd., various issues, 2006; China Statistical Information and Consultancy Center, various issues, 2006).

We classify vehicles into light-duty gasoline vehicles (LDGV), light-duty gasoline trucks up to 6000 lb gross vehicle weight (LDGT1), light-duty gasoline trucks with gross vehicle weight 6001–8500 lb (LDGT2), light-duty diesel trucks (LDDT), heavy-duty gasoline vehicles (HDGV), heavy-duty diesel vehicles (HDDV), and motorcycles, corresponding to the classification method in the US EPA's MOBILE emission factor model. It is not possible to derive the fuel consumption for each vehicle type from CESY. As an alternative approach we estimate fuel consumption from vehicle population, annual average vehicle mileage traveled, and fuel economy for each vehicle type. This method has been documented in our previous work (Streets et al., 2006), and the full details of the model used and the methodological approach are described elsewhere (He et al., 2005).

2.2.2 Other Asian countries

We follow the approach of the TRACE-P inventory for activity rates for other Asian countries but use International Energy Agency (IEA) energy statistics (IEA, 2006) for energy

Table 2. The data sources of key emitting sources in China.

Emitting Sources	Activity Data	Technology Distribution
Power plants	China Energy Statistical Yearbook	Ministry of Environmental Protection of China, unpublished data
Industry boilers	China Energy Statistical Yearbook	China Mechanical Industry Yearbook
Residential combustion	China Energy Statistical Yearbook	N/A
Coke production	China Energy Statistical Yearbook	National Bureau of Statistics, unpublished data
Cement production	China Statistical Yearbook	Chinese Cement Association, unpublished data
Iron & Steel production	China Statistical Yearbook	China Iron and Steel Statistics
Vehicles	China Automotive Industry Yearbook	Using a modeling approach documented in He et al. (2005)

use by fuel type, sector, and country instead of the RAINS-ASIA database. Activity data for the year 2006 are extrapolated from 2000–2004 IEA energy data using the average growth rate during 2000–2004. Technology distributions within each sector were obtained from the IMAGE 2.2 database (RIVM, 2001). Industrial production by product and country is derived from United States Geological Survey statistics (USGS, 2006) and also extrapolated to the year 2006. The methods for determining activity levels of non-energy sources of NMVOC are the same as in previous work (Klimont et al., 2001).

2.3 Emission factors

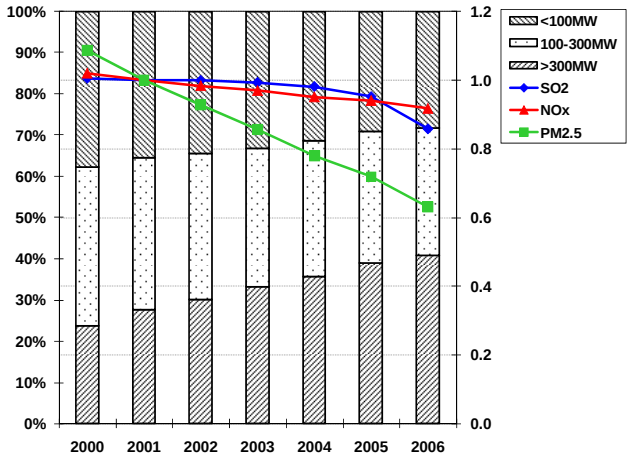
Emission factors for the years 2001 and 2006 for China are developed using our dynamic, technology-based methodology. We assume that the emission rate is fixed over the years for a given combustion/process technology (m in Eq. 1) and control technology (n in Eq. 2). Development of emission factors by technology has been documented in our previous work (Klimont et al., 2002; Streets et al., 2006; Zhang et al., 2006, 2007a, b). However, for a fast developing country like China, new technologies are constantly coming into the marketplace, causing rapid changes in the penetration of technologies (X in Eq. 1 and C_n in Eq. 2) and therefore rapid changes in net emission factors for a fuel/product in a specific sector. We estimate year-by-year changes in X and C_n, where possible.

In some cases, we use the same emission factors for the years 2001 and 2006. For example, VOC emission factors of various industrial processes are the same for the years 2001 and 2006, because we are not aware of any VOC capture technologies being used for those processes. We also use fixed emission factors for many small combustion devices like coal and biofuel stoves because there is no efficient way to control their emissions. But for most sectors, net emission factors were fundamentally changed to reflect the dramatic economic growth and dynamic technology penetration. Table 3 summarizes the significant changes of emission factors between 2006 and 2001 in China.

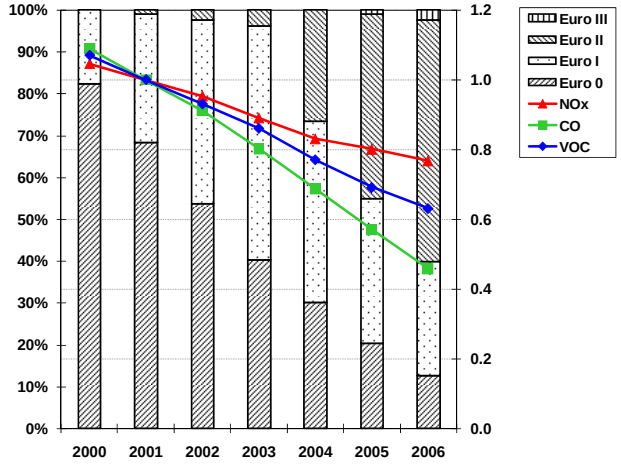
Environmental legislation is always an important determinant of emission factors. For example, the Chinese government has announced an ambitious plan to reduce national SO_2 emissions by 10% in 2010 compared with 2005. To achieve this goal, flue-gas desulfurization (FGD) devices are now being widely installed in coal-fired power plants. From 2001 to 2006, FGD penetration increased from 3% to 30%, causing a 15% decrease in the average SO_2 emission factor for coal-fired power plants (see Fig. 3a). Likewise, during the same period, net $PM_{2.5}$ emission factors in power plants declined from 2.0 g/kg coal to 1.2 g/kg coal, a reduction of 40%. This reduction is largely attributed to a new, strengthened PM emission standard for power plants published in 2003 (SEPA, 2003).

A series of emission standards was implemented for new vehicles in 1999, as shown in Table 4. Since then, new vehicles with advanced emission-control technologies began to join the fleet and replace old ones. In 2006, 60% of on-road gasoline vehicles could meet EURO II or EURO III emission standards, increased from 1% in 2001. As a result, from 2001 to 2006, the average emission factors of gasoline vehicles decreased by 23% for NO_x, 54% for CO, and 36% for VOC (Fig. 3b).

Technology improvement is another important driving force. Since the year 2000, the market share of large boilers (capacity >300 MW) has increased rapidly in power plants. Those boilers were usually equipped with low-NO_x burner technology (LNB) and hence have lower NO_x emission factors than the older, smaller plants. This transition has cut the average NO_x emission factor of the ensemble of plants by about 10% from 2001 to 2006 (Fig. 3a). Technology renewal in China's cement plants has also caused a significant change in the net emission factor. There are two main types of kilns in China's cement plants: shaft kilns and rotary kilns. Shaft kilns have higher CO emission factors but lower NO_x emission factors than rotary kilns, because the high concentration of CO in the combustion gas produces a reducing atmosphere that restrains the formation of NO_x. In recent years, shaft kilns have been largely replaced by rotary kilns. From 2001 to 2006, the market share of rotary kilns increased from 29% to 50% (Chinese Cement Association,



a) coal-fired power plants



b) gasoline vehicles

Fig. 3. Technology renewal and average emission factors for China. Bars represent the percentage of each technology: **(a)** share of power units with different boiler size; **(b)** share of gasoline vehicles with different stages of control technologies. Line: trends of average emission factors. All data are normalized to the year 2001.

unpublished data, 2007), leading to a 25% decrease in the average CO emission factor but a 35% increase in the average NO_x emission factor of China's cement plants.

For other Asian countries, we have generally used the same emission factors as in the TRACE-P inventory. An exception is for vehicle emissions. Emission factors for vehicles were derived using the MOBILE model, by integrating the varying stages of emission restrictions in recent years, to reflect the changes of emission factors due to implementation of emission standards.

3 Results

3.1 China emissions

3.1.1 Revisiting 2001 emissions: learning from methodology improvements

With the improved methodology described above, we estimate China's anthropogenic emissions in the year 2001 as follows: 22.9 Tg SO_2, 13.4 Tg NO_x, 141.6 Tg CO, 18.1 Tg NMVOC, 16.1 Tg PM_{10}, 11.7 Tg $PM_{2.5}$, 1.6 Tg BC, and 2.8 Tg OC. Table 5 summarizes the 2001 emission estimates by species and by sector and presents the difference between this 2001 inventory (R01) and the TRACE-P inventory for the year 2000 (T00). R01 estimates generally show a significant increase compared with T00, ranging from a 6% increase for OC to 70% for BC. Because the actual emission increases from 2000 to 2001 were not so significant (e.g., 5% increase for NO_x), these differences between R01 and T00 can be mainly attributed to the improvements of methodology.

The reasons for these differences vary among sectors and species. The most important reason is that R01 uses a technology-based approach that can identify emissions from specific types of technology. For example, industrial CO emissions in R01 are higher than in T00 by a factor of three, contributing significantly to the difference of CO emissions between T00 and R01. Compared with T00, R01 has a much more detailed categorization of sources in the industrial sector, which allows the identification of important CO emitting sources from specific industries such as cement kilns and brick kilns, which were missing in T00 (Streets et al., 2006). The situation is similar for other species. In R01, traditional brick kilns and coking production are identified as two important individual sources for BC and OC emissions. Using emission factors from Bond et al. (2004), BC emissions from traditional brick kilns and coking processes for the year 2001 are estimated to be 241 Gg and 183 Gg, respectively, accounting for 15.1% and 11.5% of total anthropogenic emissions. These two important carbonaceous aerosol sources were missing in T00.

Another important reason is that R01 includes more emitting sources than T00. For example, we include off-road diesel emissions in R01 in the transportation sector. As a result, although emissions for on-road vehicles are similar in R01 and T00, NO_x, VOC, BC and OC emissions from the transportation sector as a whole in R01 are higher by 37%, 23%, 140%, and 35%, respectively, than in T00 due to the inclusion of off-road vehicle emissions. R01 also has higher VOC, BC, and OC emissions for power plants than T00, which can be attributed to the inclusion of oil-fired power plant emissions in R01, but not in T00. And, NO_x emissions from the residential sector in R01 are 37% higher than in T00, because R01 includes emissions from liquid fuels.

Table 3. Changes of emission factors between 2006 and 2001.

Sector	Fuel/product	Species	Unit	Emission Factor		Reason for Change
				2006	2001	
Power plants	coal	SO_2	g/kg-coal	15.4	18.1	Installation of FGD required by government
	coal	NO_x	g/kg-coal	7.1	7.8	Increased market share of large units
	coal	$PM_{2.5}$	g/kg-coal	1.2	2.0	Implementation of new emission standard[a]
Cement kilns	coal/cement	NO_x	g/kg-coal	8.1	6.0	Shift from shaft kilns to rotary kilns,
	coal/cement	CO	g/kg-coal	90.4	121.0	which release less CO but more NO_x
	coal/cement	$PM_{2.5}$	g/kg-cement	3.3	6.5	Implementation of new emission standard[a]
Brick kilns	coal/brick	CO	g/kg-coal	128.0	150.0	Phase out of old beehive kilns
	coal/brick	BC	g/kg-coal	2.6	3.3	
Coke production	coke	CO	g/kg-coke	4.1	5.6	Phase out of old indigenous process
	coke	BC	g/kg-coke	0.77	1.8	
Iron & steel production	sinter, pig iron[b]	CO	g/kg-iron	39.6	59.0	Increased ratio of by-pass gas recycle in
	steel	CO	g/kg-steel	24.0	37.0	new factories
	sinter, pig iron[b]	$PM_{2.5}$	g/kg-iron	0.55	0.85	New factories with lower emissions come
	steel	$PM_{2.5}$	g/kg-steel	0.32	0.51	into the marketplace
Gasoline vehicles [c,d]	gasoline	NO_x	g/kg-fuel	21.5	28.0	New emission standards were implemented
	gasoline	CO	g/kg-fuel	294.2	633.3	in 1999; new vehicles rapidly replace
	gasoline	VOC	g/kg-fuel	88.4	139.1	old vehicles
Diesel vehicles [c,e]	diesel	NO_x	g/kg-fuel	55.2	65.0	Same as gasoline vehicles
	diesel	CO	g/kg-fuel	59.1	116.1	
	diesel	VOC	g/kg-fuel	15.7	31.1	
	diesel	$PM_{2.5}$	g/kg-fuel	3.1	4.2	

[a] SEPA (2003, 2004); [b] emission factors are given for the sum of sintering processes and pig iron production; [c] emission factors were calculated by MOBILE model in g/km, then converted to g/kg-fuel according to the fleet average fuel economy data of He et al. (2005); [d] emission factors were calculated for LDGV (car), LDGT1, LDGT2, and HDGV separately. Here average emission factors are presented; [e] emission factors were calculated for LDDT and HDDT separately. Here average emission factors are presented.

Table 4. Implementation schedule of new vehicle emission standards in China.

	Euro I	Euro II	Euro III	Euro IV
Gasoline (Beijing)	1999	2003	2006	2008
Gasoline (national)	2001	2004	2007	2010
Diesel (national)	2001	2004	2008[a]	2010

[a] initially scheduled for 2007, but postponed to 2008.

Different data sources can also lead to different results. For the power plant sector, SO_2 and NO_x emissions in R01 are both 22% higher than in T00. This is mainly because the coal consumption data used in R01 are actual statistical data from CESY, while the data used in T00 were extrapolated from 1995 data, which was lower than in the actual reported statistics. SO_2 emissions for transportation in R01 are 82% lower than in T00 because we use lower sulfur contents for transportation fuels – though the contribution of the transportation sector to total SO_2 emissions is small.

3.1.2 2006 emissions: emission growth and driving forces

We estimate China's anthropogenic emissions in the year 2006 to be as follows: 31.0 Tg SO_2, 20.8 Tg NO_x, 166.9 Tg CO, 23.2 Tg NMVOC, 18.2 Tg PM_{10}, 13.3 Tg $PM_{2.5}$, 1.8 Tg BC, and 3.2 Tg OC. Table 6 presents the 2006 emission estimates and Fig. 4 shows the emission increase from 2001, by species and by sector. Compared with the decreasing or flat emission trend during 1995–2000 (Hao et al., 2002; Streets et al., 2001), all species show an increasing trend during 2001–2006: 36% increase for SO_2, 55% for NO_x, 18% for CO, 29% for VOC, 13% for PM_{10}, and 14% for $PM_{2.5}$, BC, and OC. These emission increases can be viewed in the context of 92% growth of GDP and 72% increase of total energy consumption in the same period.

Table 5. Anthropogenic emissions in China in the year 2001 (units: Gg/year).*

Species	Power	Industry	Residential	Transportation	Total
SO_2	12 270 (1.22)	7946 (1.08)	2599 (1.03)	75 (0.18)	22 891 (1.13)
NO_x	5390 (1.22)	3405 (1.22)	997 (1.42)	3604 (1.37)	13 397 (1.27)
CO	1861 (n/a)	53 526 (2.97)	48 254 (1.10)	37 930 (1.00)	141 571 (1.42)
NMVOC	547 (6.02)	4982 (1.34)	5996 (1.07)	6547 (1.23)	18 072 (1.23)
PM_{10}	1873 (n/a)	9647 (n/a)	4258 (n/a)	292 (n/a)	16 070 (n/a)
$PM_{2.5}$	1152 (n/a)	6398 (n/a)	3853 (n/a)	284 (n/a)	11 687 (n/a)
BC	38 (5.59)	545 (6.13)	868 (1.11)	143 (2.40)	1595 (1.70)
OC	8 (1.52)	496 (17.95)	2254 (0.88)	70 (1.35)	2827 (1.06)

* Numbers in parentheses represent the emission ratio between this inventory for the year 2001 (R01) and the TRACE-P inventory for the year 2000 (T00).

Table 6. Anthropogenic emissions in China in the year 2006 (units: Gg/year).*

Species	Power	Industry	Residential	Transportation	Total
SO_2	18 333 (1.49)	9725 (1.22)	2838 (1.09)	123 (1.64)	31 020 (1.36)
NO_x	9197 (1.71)	5371 (1.58)	1166 (1.17)	5096 (1.41)	20 830 (1.55)
CO	2362 (1.27)	74 936 (1.40)	55 883 (1.16)	33 709 (0.89)	166 889 (1.18)
NMVOC	961 (1.76)	8056 (1.62)	7601 (1.27)	6630 (1.01)	23 247 (1.29)
PM_{10}	2476 (1.32)	10 436 (1.08)	4884 (1.15)	427 (1.46)	18 223 (1.13)
$PM_{2.5}$	1474 (1.28)	6932 (1.08)	4461 (1.16)	398 (1.40)	13 266 (1.14)
BC	36 (0.94)	575 (1.06)	1002 (1.15)	198 (1.38)	1811 (1.14)
OC	6 (0.72)	505 (1.02)	2606 (1.16)	101 (1.45)	3217 (1.14)

* Numbers in parentheses represent the emission ratio between the year 2006 (I06) and the year 2001 (R01).

It is quite clear that the dramatic emission increases in China after 2001 were driven by the economic boom and growing infrastructure investments. Figure 5 shows how China's power plants grew during 2001–2006 compared with the previous five years. Many energy-consuming activities doubled in just a few years in China, resulting in a significant increase in relevant emissions. For example, total thermal based electricity generation increased from 1.17 trillion kWh in 2001 to 2.37 trillion kWh in 2006, and total vehicle numbers increased from 18 million to 37 million during the same period.

On the other hand, China has made substantial efforts on technology improvement and emission control during this period. These measures have offset the emission growth significantly. We note several developments that have had important impacts on emissions in the following areas:

a) New technologies with improved energy intensity and/or lower emissions. These technologies include: replacement of small power generation boilers by large ones that have better combustion efficiencies; use of power generation boilers with LNB technologies to reduce NO_x emissions; replacement of indigenous processes by modern processes for coke production, resulting in a significant reduction of emissions; transition from shaft kilns to new-dry kilns in the cement industry, which reduces CO emissions (but increases NO_x emissions);

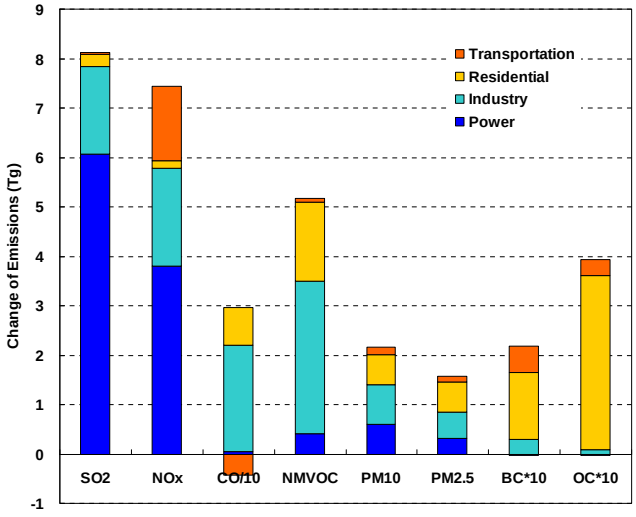


Fig. 4. Change of China's emissions between 2006 and 2001 by sector (units: Tg).

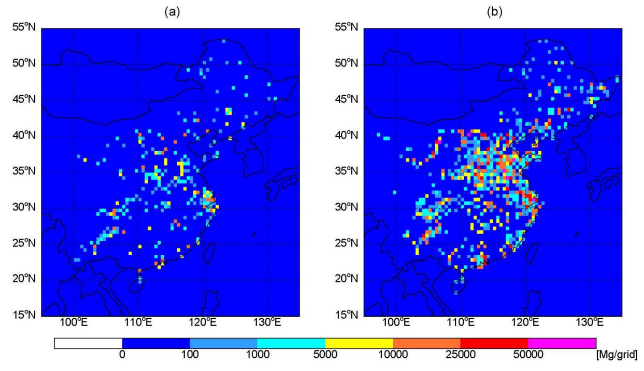


Fig. 5. NO_x emission increase from China's power plants. Left panel: NO_x emission change from China's power plants between 2001 and 1996. Right panel: NO_x emission change from China's power plants between 2006 and 2001 (units: Mg per grid).

and advanced technologies to capture by-pass gas during iron and steel production, to avoid the CO releases from by-pass gas.

b) FGD installation on coal-fired power plants. As discussed in Sect. 2.3, FGD has been widely installed in power plants in recent years under new requirements of central and local government. By the end of 2006, 30% of coal-fired power plants were equipped with FGD, which is estimated to eliminate about 6 Tg of SO_2 emissions in that year. FGD penetration in power plants further increased to 50% at the end of 2007, leading to a 4.7% reduction of national SO_2 emissions in 2007, which is the first decrease in national SO_2 emissions since the year 2002 (MEP, 2008).

c) Strengthened PM emission standards for cement plants and coal-fired power plants. Cement plants and coal-fired power plants contributed 37% and 10% of national $PM_{2.5}$ emissions, respectively in 2001. In 2003 and 2004, China implemented new emission standards for these two sectors, which strengthened the limits for TSP emissions from 150–600 mg/Nm^3 to 50–100 mg/Nm^3 for all cement plants, and from 200–600 mg/Nm^3 to 50 mg/Nm^3 for new coal-fired power plants (SEPA, 2003, 2004; CRAES, 2003). To meet these standards, high-efficiency PM removal equipment was widely installed, and some small, dirty factories were closed. As a result, $PM_{2.5}$ emissions from cement plants and coal-fired power plants decreased by 7% and increased by 23% during 2001–2006, respectively, in contrast to the doubled activity rates in each sector.

d) Emission standards for new vehicles. Table 4 lists the emission standards for new vehicles in China in recent years, and Fig. 3b shows the decreasing trend of emission factors when new vehicles join the fleet and replace old ones. CO emissions from the transportation sector decreased by 11% during 2001–2006 during a period when the total number of vehicles doubled, providing an excellent illustration of effective control measures. NMVOC emissions in 2006 were almost the same as in 2001, while NO_x emissions increased by 41%, but still showing a much lower growth than the growth in the vehicle population.

Table 7 presents China's emissions by province for the year 2006. Emissions vary considerably from province to province, with the highest emissions mainly located in the eastern and central regions of China. Hebei, Henan, Jiangsu, Shandong, and Sichuan Provinces are the five largest contributors for most species. Shandong is the largest contributor for SO_2, NO_x, NMVOC, PM_{10}, and $PM_{2.5}$ and the second largest contributor for CO and OC. Emissions from western provinces, e.g., Qinghai and Xizang, were much less than from eastern ones. The regional differences of emissions are mainly caused by differences of economic development, industry structure, and population.

3.2 Total Asian emissions

We estimate total Asian anthropogenic emissions in the year 2006 as follows: 47.0 Tg SO_2, 36.8 Tg NO_x, 298.1 Tg CO, 54.6 Tg NMVOC, 28.9 Tg PM_{10}, 22.0 Tg $PM_{2.5}$, 2.91 Tg BC, and 6.54 Tg OC. These values are not directly comparable with the TRACE-P inventory due to the fundamental changes in methodologies discussed previously. However, most impacts of methodology improvement can be removed by replacing the China part of the TRACE-P inventory with the R01 inventory of this work. Then we can compare the revised TRACE-P Asian emissions with our new estimates, to explore the actual emission changes during the intervening years. Asian emissions continue the significant increasing trends that have been reported in the last two decades (van Aardenne et al., 1999; Streets et al., 2001; Ohara et al., 2007). From the beginning of the 21st century, Asian anthropogenic emissions increased by 33% for SO_2, 44% for NO_x, 18% for CO, 25% for NMVOC, and 9% for BC in just 5 years. The most significant growth was found for NO_x emissions, which is driven by both industrial and vehicular emissions. In contrast, BC emissions, which are dominated by the residential sector, show a relatively small increase. OC emissions decreased by 9%, but this cannot be viewed as a real emission decrease, because in this 2006 inventory we used lower estimates of emissions from Reddy and Venkataraman (2002a, b) than the TRACE-P estimates. Reddy and Venkataraman (2002a, b) estimated that the OC emissions in India were 1.0 Tg in 1999, much lower than TRACE-P estimates of 2.2 Tg in 2000. The main reason for this difference is that Reddy and Venkataraman (2002b) used lower OC emission factors for biofuel combustion.

Table 8 summarizes the emissions of each species in each country in 2006, and Fig. 6 shows the shares of emissions among different sub-regions of Asia. China and India are the two most important individual contributors to Asian emissions. China's contributions to total Asian emissions are: 66% for SO_2, 57% for NO_x, 56% for CO, 43% for NMVOC, 63% for PM_{10}, 60% for $PM_{2.5}$, 62% for BC, and 49% for

Table 7. Anthropogenic emissions in China by province in 2006 (units: Gg/year).

Province	SO_2	NO_x	CO	VOC	PM_{10}	$PM_{2.5}$	BC	OC
Anhui	693	715	7986	958	757	574	84	173
Beijing	248	327	2591	497	123	90	19	19
Chongqing	1211	326	2928	343	340	257	34	75
Fujian	460	547	3895	701	435	337	44	127
Gansu	338	323	2688	303	296	222	35	55
Guangdong	1175	1493	8693	1780	942	680	55	120
Guangxi	880	435	4258	640	468	348	40	94
Guizhou	1952	485	4409	481	571	435	90	162
Hainan	76	83	724	117	67	53	7	18
Hebei	2281	1308	15 505	1521	1371	981	137	200
Heilongjiang	242	839	4967	771	579	440	72	144
Henan	1591	1197	10 957	1289	1193	834	133	197
Hong Kong	118	148	127	109	27	18	1	1
Hubei	2200	930	7482	875	772	559	73	137
Hunan	915	563	5124	641	576	424	54	105
Jiangsu	1697	1486	11 326	1814	1200	881	87	186
Jiangxi	533	390	3963	463	586	400	39	76
Jilin	357	473	3794	523	395	293	45	82
Liaoning	1027	955	8105	989	710	512	64	111
Nei Mongol	1171	860	5253	575	574	420	71	113
Ningxia	380	175	961	131	134	98	11	18
Qinghai	18	46	616	74	70	54	8	11
Shaanxi	907	352	3528	491	474	328	49	81
Shandong	3102	1759	14 970	2093	1702	1212	132	213
Shanghai	618	631	1958	594	138	91	10	8
Shanxi	1804	934	5787	627	969	669	139	158
Sichuan	2555	873	10 945	1312	1068	845	133	318
Tianjin	336	365	1860	381	161	109	15	18
Xinjiang	210	356	2775	391	257	194	37	55
Xizang	0	5	94	14	9	6	1	1
Yunnan	489	344	3765	515	454	343	56	97
Zhejiang	1434	1106	4857	1233	806	556	36	45
China Total	31 020	20 830	166 889	23 247	18 223	13 266	1811	3217

OC. India follows China as the second largest contributor with the following shares: 12% for SO_2, 13% for NO_x, 20% for CO, 20% for NMVOC, 14% for PM_{10}, 14% for $PM_{2.5}$, 12% for BC, and 14% for OC. Other countries contribute much smaller individual shares. China's contribution to Asian emissions has increased since the year 2000, reflecting faster economic development and industrialization than other Asian developing countries. South Asia and Southeast Asia contribute significantly to emissions of CO, NMVOC, and OC, due to the large amount of residential biofuel use.

Figure 7 compares the sectoral contributions of Asian emissions in 2000 and 2006. The sectoral distribution of emissions is similar between the two years, with some small but meaningful changes. The contribution from power plants to SO_2 and NO_x emissions has increased, driven by the industrialization progress and spread of electrification in the past years. Although the vehicle stocks in Asia increased

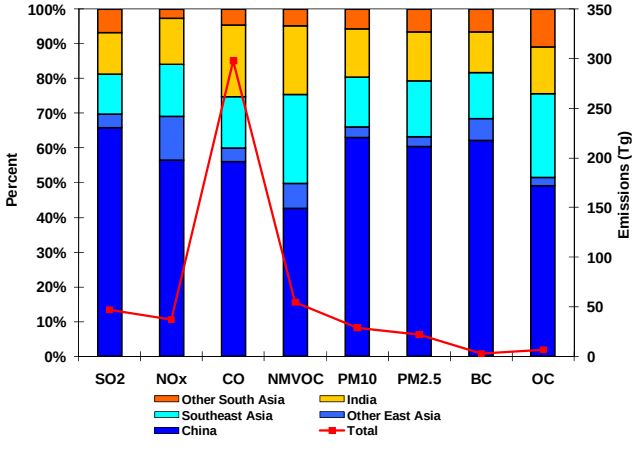


Fig. 6. Species emissions by Asian region and absolute values of emissions.

Table 8. Summary of national emissions in Asia in 2006 (units: Gg/year).

Country	SO_2	NO_x	CO	NMVOC	PM_{10}	$PM_{2.5}$	BC	OC
China	31 020	20 830	166 889	23 247	18 223	13 266	1811	3217
Japan[a]	871	2404	5314	2033	195	141	51	21
Korea, Rep of	408[b]	1307[b]	789[b]	796[b]	67[b]	54	17	11
Korea, DPR	233	270	3583	212	301	245	21	95
Mongolia	84	38	351	23	27	22	2	9
Taiwan, China[c]	189	642	1672	864	337	175	91	9
Brunei	7	23	15	46	4	3	0	0
Cambodia	34	27	570	113	68	61	7	32
Indonesia	1451	1583	17 742	6617	1838	1610	170	803
Laos	8	18	195	61	22	20	2	11
Malaysia	1137	1664	4286	1267	471	322	14	40
Myanmar	51	85	2568	641	271	244	29	130
Philippines	943	310	1998	1157	231	199	23	96
Singapore	163	193	149	101	32	22	2	1
Thailand	1299	1278	7189	2638	475	388	49	141
Vietnam	385	330	9843	1441	737	657	90	326
Bangladesh	148	182	3218	550	438	390	43	205
Bhutan	5	5	97	21	12	11	1	6
India	5596[d]	4861	61 106	10 767	4002	3111	344[d]	888[d]
Nepal	31	27	1659	251	205	186	21	103
Pakistan	2882	681	7378	1405	873	752	115	349
Sri Lanka	98	72	1501	371	100	88	11	44
Asia 2006 Total	47 045	36 829	298 112	54 620	28 930	21 967	2914	6537
Asia 2000 Total[e]	35 450	25 540	252 891	43 538	n/a	n/a	2679	7209
2006/2000	1.33	1.44	1.18	1.25	n/a	n/a	1.09	0.91

[a] From Kannari et al. (2007); [b] from NIER (2008); [c] from Fu et al. (2009); [d] based on Reddy and Venkataraman (2002a, b) and scaled to the year 2006; [e] here Asia 2000 emissions consist of R01 inventory for China and TRACE-P inventory for other Asian countries, to better represent the real-world Asia emissions around the year 2000.

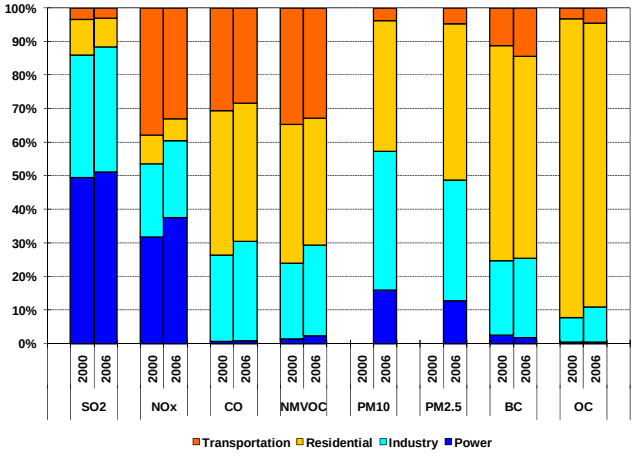


Fig. 7. Share of emissions by sector in Asia in 2000 and 2006.

dramatically during the past few years, the relative contributions from the transportation sector decreased for NO_x, CO, and NMVOC, indicating the effectiveness of control measures on gasoline vehicles. However, the increasing contribution of transportation emissions to carbonaceous aerosols indicates the expanding diesel vehicle fleet and slow progress on control measures for diesel particles.

3.3 Seasonality of emissions

In the TRACE-P inventory, Streets et al. (2003a) developed seasonal variations of residential energy consumption, assuming a dependence of stove operation on regional monthly mean temperatures. However, no seasonal variation was considered for the power generation and industrial sectors in the TRACE-P inventory. In this work, we follow the same methodology to generate monthly emissions for the residential sector for the whole of Asia, but also develop monthly emissions for power generation and industry for China according to the monthly activity data on power generation, cement production, and industrial GDP at the provincial level (Beijing Huatong Market Information Co. Ltd, 2006; China Statistical Information and Consultancy Center, 2006).

Table 9. Monthly anthropogenic emissions in China in 2006 (units: Gg/month).

Species	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
SO_2	2853	2416	2628	2368	2389	2430	2472	2485	2460	2503	2794	3220
NO_x	1839	1666	1770	1627	1631	1654	1667	1675	1667	1697	1866	2071
CO	18 051	15 123	14 677	12 194	12 131	12 382	11 714	11 911	12 302	12 806	15 041	18 552
NMVOC	2528	2154	2034	1707	1702	1727	1663	1684	1720	1778	2037	2513
PM_{10}	1808	1516	1575	1356	1361	1401	1320	1346	1395	1445	1673	2026
$PM_{2.5}$	1416	1166	1163	963	962	986	934	951	982	1023	1211	1507
BC	240	189	168	120	117	118	114	116	118	126	164	221
OC	511	385	310	193	185	183	182	183	182	200	283	419

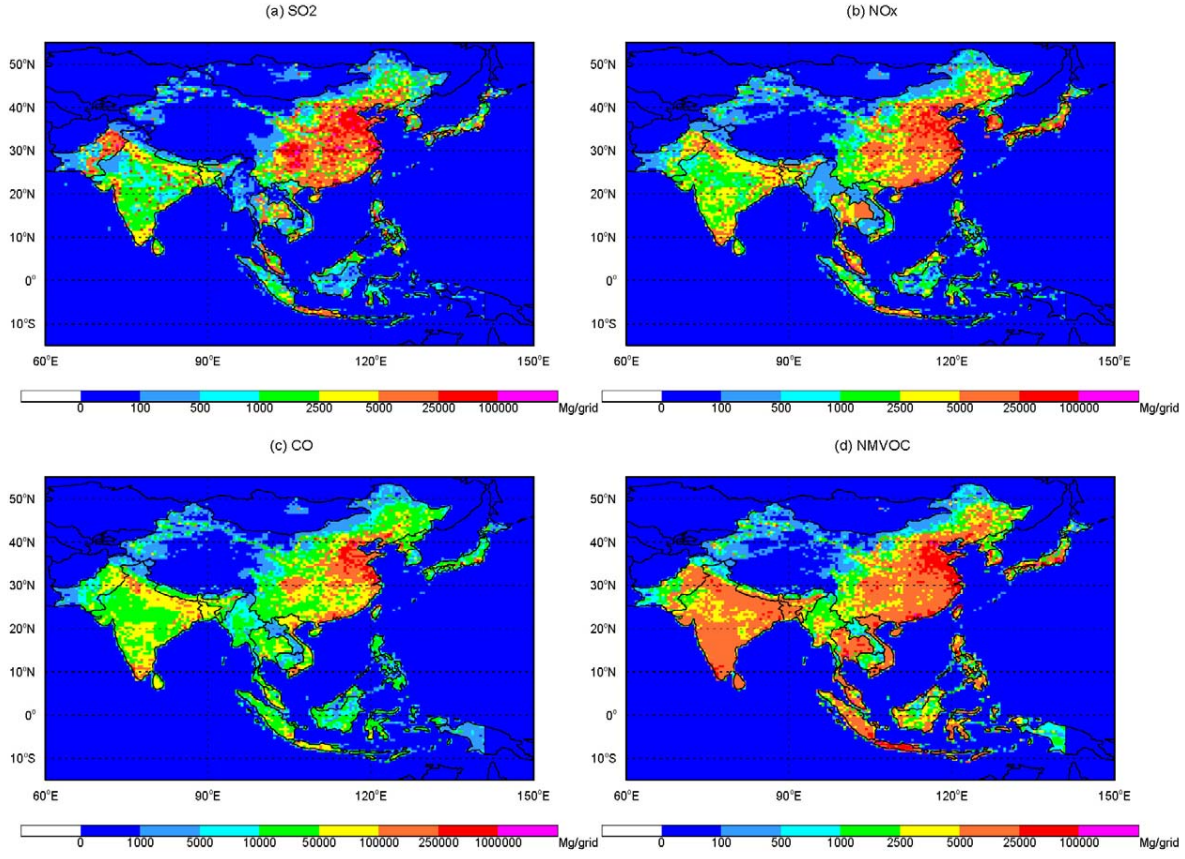


Fig. 8. Emission distributions at 30 min × 30 min resolution of gaseous species (units: Mg/year per grid).

Table 9 presents monthly emissions in China in 2006 by species. Strong seasonal variations are observed for CO, BC, and OC, where the residential sector contributes the largest portion of emissions. The ratios of monthly CO, BC, and OC emissions between maxima and minima are 1.6, 2.1, and 2.8, respectively. In contrast, SO_2 and NO_x emissions have weaker seasonal variations, with ratios of 1.4 and 1.3 between maxima and minima, because they mainly come from industrial and transportation emissions that have less of a seasonal cycle. We also find that SO_2 and NO_x emissions in February are lower than in neighboring months, because of reduced industrial activity during the Chinese Spring Festival holiday.

3.4 Gridded emissions

Figures 8 and 9 show the spatial distribution of gaseous pollutants and aerosol emissions in Asia in 2006 at a resolution of 30 min × 30 min. Emissions are distributed using various spatial proxies at 1 km × 1 km resolution (Streets et al., 2003a; Woo et al., 2003). For Japan, Korea, Taiwan,

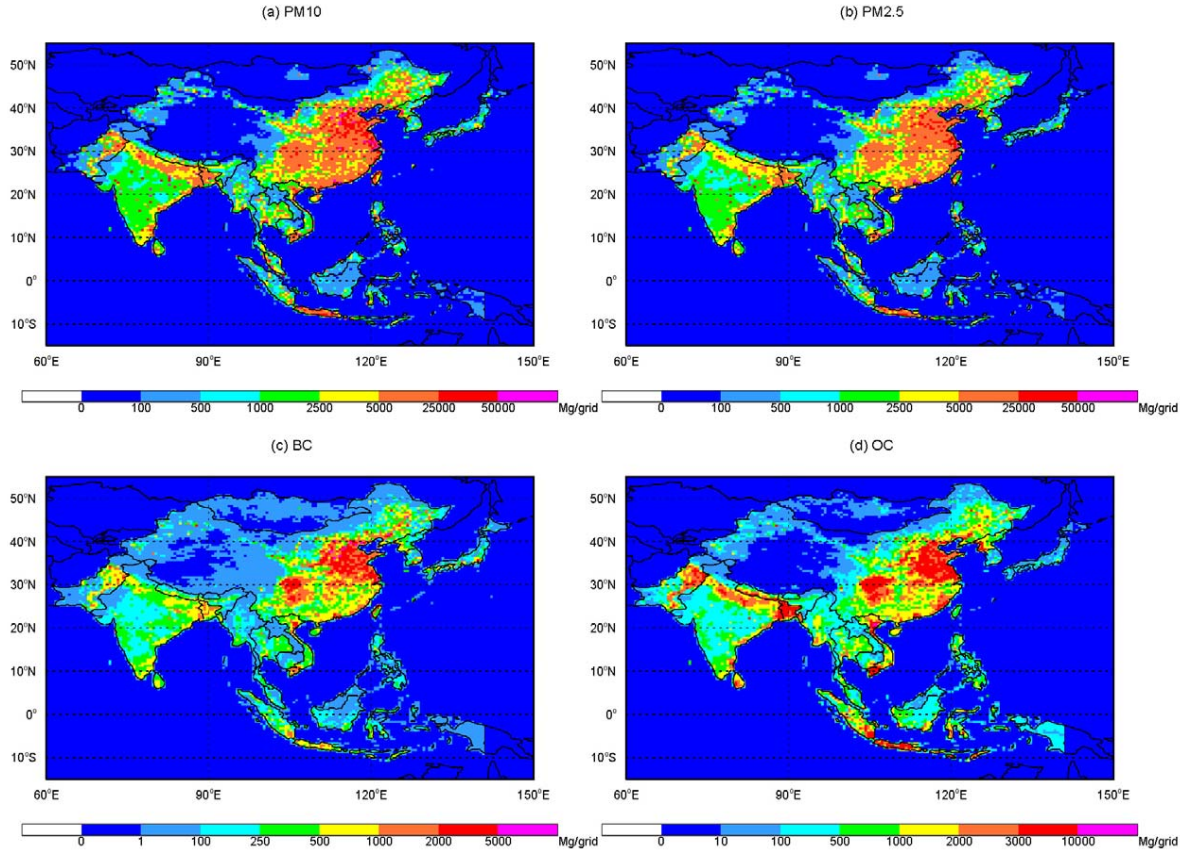


Fig. 9. Emission distributions at 30 min×30 min resolution of aerosol species (units: Mg/year per grid).

and India, where the emissions were obtained from national inventories, we keep the spatial distribution characteristics of the original inventories and simply re-grid them to 30 min×30 min resolution. All power generation units with capacity larger than 300 MW (∼400 units) in China are identified as large point sources, while other plants are treated as area sources.

3.5 Data access

All regional and gridded emission data sets can be downloaded from our web site (http://mic.greenresource.cn/intex-b2006). Users can examine emissions by country and by sector from the summary tables. Gridded data include the emissions of all species by sector (power, industry, residential, and transportation) at 30 min×30 min resolution. At the time this paper was submitted, NMVOC emissions speciated according to the SAPRC-99 mechanism are available by sector (power, industry, residential biofuel, residential fossil fuel, residential non-combustion, and transportation) for download at 30 min×30 min resolution, but we will add speciated VOC emissions for other mechanisms later. These emission data are also downloadable from the website at the University of Iowa (http://www.cgrer.uiowa.edu/EMISSION_DATA_new/index_16.html).

4 Discussion

4.1 Magnitude of China's emissions in inventories and top-down constraints

Ohara et al. (2007) conducted a comprehensive comparison of different emission inventories for Asia, China, and India during 1995–2000 and discussed the reasons for the differences. In this section, we will not repeat that comparison, but focus instead on a comparison of the magnitude of China's emissions in inventories and from top-down constraints for years after 2000 (Table 10), in order to highlight the implications for emission inventory development.

a) Sulfur Dioxide. SO_2 is one of the few pollutants for which China's government reports national emissions annually. Our estimate for the year 2006 is 20% higher than SEPA reported; however, the two estimates generally agree well at the sector level, considering that SEPA's estimate does not include emissions from rural industries and residential biofuels. The increasing trends of the two estimates are also similar: we estimate a 35% increase of SO_2 emissions in 2001–2006, while SEPA presented a 33% increase during the same period. Our estimates for the year 2001 are close to the value of the TRACE-P inventory, but significantly lower than the

Table 10. Estimates of China's and East Asian annual emissions after the year 2000.[a]

Pollutant and Study	Method[b]	Sources[c]	Region	2000	2001	2002	2003	2004	2005	2006
SO_2										
Streets et al. (2003a)	EI	FF+BF	China	20.3						
Olivier et al. (2005)	EI	FF+BF	China	34.2						
Ohara et al. (2007)	EI	FF+BF	China	27.6	29.3	31.9	36.6			
SEPA	EI	FF[d]	China	20.0	19.5	19.3	21.6	22.5	25.5	25.9
This work	EI	FF+BF	China		22.9					31.0
NO_x										
Streets et al. (2003a)	EI	FF+BF	China	10.5						
Olivier et al. (2005)	EI	FF+BF	China	13.7						
Ohara et al. (2007)	EI	FF+BF	China	11.2	11.8	12.7	14.5			
Zhang et al. (2007b) and this work	EI	FF+BF	China	12.6	13.2	14.4	16.2	18.6	19.8	20.8
Wang et al. (2004)	IM	FF+BF+BB+SL	China		16.5					
Jaegle et al. (2005)	IM	FF+BF	China	14.5						
Martin et al. (2006)	IM	FF+BF+BB+SL	East Asia					32.2		
CO										
Streets et al. (2003a)	EI	FF+BF	China	100						
Olivier et al. (2005)	EI	FF+BF	China	87						
Streets et al. (2006) and this work	EI	FF+BF	China		142					167
Ohara et al. (2007)	EI	FF+BF	China	137	141	146	158			
Palmer et al. (2003a)	IM	FF+BF	China		168					
Arellano et al. (2004)	IM	FF+BF+BB	East Asia	196–214						
Heald et al. (2004)	IM	FF+BF+BB	East Asia		192					
Wang et al. (2004)	IM	FF+BF+BB	China		166					
Petron et al. (2004)	IM	FF+BF+BB	East Asia	186						
Yumimoto and Uno (2006)	IM	FF+BF+BB	China		147					
Tanimoto et al. (2008)	IM	FF+BF+BB	China						170	
Kopacz et al. (2009)	IM	FF+BF+BB	China		141.5					
Carmichael et al. (2003b)	FM	FF+BF+BB	China		163–210					
Heald et al. (2003)	FM	FF+BF+BB	China		181					
Allen et al. (2004)	FM	FF+BF	China		145					
Tan et al. (2004)	FM	FF+BF+BB	China		174					
VOC										
Streets et al. (2003a)	EI	FF+BF	China	14.7						
Olivier et al. (2005)	EI	FF+BF	China	11.5						
Wei et al. (2008)	EI	FF+BF	China						20.1	
Bo et al. (2008)	EI	FF+BF+BB	China	11.0					16.5	
This work	EI	FF+BF	China		18.1					23.2
Fu et al. (2007)	IM	FF+BF+BB	China	21.7[e]						
BC/OC										
Streets et al. (2003a)	EI	FF+BF	China	0.94/2.66						
Bond et al. (2004)	EI	FF+BF	China	1.36/2.11						
Cao et al. (2006)	EI	FF+BF	China	1.40/3.81						
Ohara et al. (2007)	EI	FF+BF	China	1.09/2.56	1.10/2.58	1.11/2.60	1.14/2.62			
This work	EI	FF+BF	China		1.60/2.83					1.81/3.22

[a] Units are Tg/year. Units for NO_x are Tg-NO_2/year. [b] EI = emission inventory; IM = inverse modeling; FM = forward modeling. [c] FF = fossil fuel; BF = biofuel combustion; BB = open biomass burning; and SL = soil emissions. [d] Only includes fossil-fuel emissions from power plants and industry. [e] Fu et al. (2007), required a 25% increase in reactive VOC emissions from the TRACE-P inventory (including BB emissions) to agree with inverse modeling.

values estimated by REAS (Ohara et al., 2007) and EDGAR (Olivier et al., 2005).

It appears that the magnitude of China's SO_2 emissions in the TRACE-P inventory is reasonable on the basis of CTM model simulations and comparison with in situ measurements (Carmichael et al., 2003b; Russo et al., 2003; Tan et al., 2004). The accuracy of our SO_2 estimate for the year 2006 is of more concern, because we have no guarantees that the recently installed FGD equipment is run continuously, as we assume. This could impact China's SO_2 emission trend significantly. Recent developments on satellite-based SO_2 column observations show good potential for constraining

surface emissions (Richter et al., 2006; Krotkov et al., 2008). Further trend analysis of satellite SO_2 columns over China may be able to provide valuable information for verifying SO_2 emissions.

b) Nitrogen Oxides. NO_x emission estimates for China are all quite close (see Table 10). Analysis from modeling and measurements during the TRACE-P campaign also indicated that the estimates of China's NO_x emissions in the TRACE-P inventory are reasonably accurate (Carmichael et al., 2003b). However, several inverse modeling analyses constrained by satellite-based data concluded that China's NO_x emission inventory was significantly underestimated (Martin et al., 2003, 2006; Jaeglé et al., 2005; Wang et al., 2007). In the meantime, forward modeling studies also under-predicted NO_2 columns compared to satellite retrievals by a factor of two over East China, which is usually attributed to underestimation of NO_x emissions (Ma et al., 2006; Uno et al., 2007). China's NO_x emissions are mainly contributed by power plants and vehicles, and there is no clear evidence to suggest such a remarkable underestimation of emissions from those two sectors from the perspective of inventory development. One plausible reason is that current estimates of soil NO_x emissions are too low (Wang et al., 2007), and further investigations are required to reconcile NO_x emission estimates over China.

c) Carbon Monoxide. Analysis of CO observations using chemical transport models in inverse and forward modes suggested that previous China's CO inventories were underestimated by about 50% at the time of the TRACE-P mission (Carmichael et al., 2003b; Heald et al., 2003, 2004; Palmer et al., 2003a; Allen et al., 2004; Arellano et al., 2004; Petron et al., 2004; Tan et al., 2004; Wang et al., 2004), as shown in Table 10. Motivated by those top-down constraints, we reexamined the source characteristics and concluded that emissions from cement kilns, brick kilns, and the iron and steel industry were underestimated (Streets et al., 2006). Bottom-up and modeled emission estimates are now in good agreement (Yumimoto and Uno, 2006; Tanimoto et al., 2008; Kopacz et al., 2009), which represents a major success story for the TRACE-P mission. This was the first study in which bottom-up and top-down approaches for quantifying China's emissions were truly integrated. Inadequacies in the bottom-up approach were identified by top-down studies, and the findings from the top-down studies were able to be used to improve our understanding of emissions and guide improvement of the bottom-up inventory.

d) Nonmethane Volatile Organic Compounds. Wei et al. (2008) and Bo et al. (2008) recently estimated China's NMVOC emissions for the year 2005. Our estimates for the year 2006 are 15% higher than Wei et al. (2008). Considering the emission growth from 2005 to 2006, these two estimates may actually be in reasonable agreement. Bo et al. (2008) presented lower NMVOC emissions than both Wei et al. (2008) and this work, which seems to be due to their low estimates for stationary combustion emissions.

Satellite observations of formaldehyde columns offer top-down constraints on reactive NMVOC emissions (Palmer et al., 2003b, 2006; Millet et al., 2006). Fu et al. (2007) found that wintertime GOME observations can diagnose anthropogenic reactive NMVOC emissions from China, leading to an estimate 25% higher than the TRACE-P inventory, which is in good agreement with our new estimates for the year 2001 (23% higher than the TRACE-P inventory). Modeling evaluations of the TRACE-P inventory using field measurements from the TRACE-P campaign concluded that the inventory performed well for the light alkanes and ethyne, but estimates for other speciated NMVOCs are highly uncertain (Carmichael et al., 2003b). Speciated NMVOC emissions are highly dependent on the source profiles used. In this work, we applied both local source profiles and international profiles when developing speciated VOC emissions, to investigate the impact of different source profiles on emissions. This work is documented in Zhang et al. (2009).

e) Black Carbon. The range of 2000/2001 estimates for China's anthropogenic BC emissions varies from 0.94 Tg to 1.60 Tg (see Table 10). This is not surprising because of the high uncertainties in emission estimates. Analysis of forward and inverse model calculations using TRACE-P and ACE-Asia measurement data concluded that the TRACE-P estimates of BC are qualitatively correct (Carmichael et al., 2003b; Uno et al., 2003b; Clarke et al., 2004; Hakami et al., 2005), but Tan et al. (2004) suggested a 60–90% increase of TRACE-P BC emissions was necessary to bring the model-predicted BC concentrations into agreement with in situ measurements. However, Carmichael et al. (2003b) pointed out a systematic problem in under-predicting BC levels at low altitudes in the Yellow Sea; Clarke et al. (2004) found that BC emissions from combustion sources north of 25° N were underestimated by a factor of ∼3, and Hakami et al. (2005) concluded that anthropogenic BC emissions over Southeastern China were overestimated while those in Northeast China were underestimated. These findings indicated that the regional distributions of the current BC inventory are questionable, although emission estimates of the TRACE-P inventory may be correct at the national level. One possible reason for this situation is discussed below (Sect. 4.3.2).

4.2 Constraining the trajectory of China's emission trends

Recent developments in satellite observations allow for the development of long-term emission trends, which is very helpful for constraining emission inventories. Satellite-observed tropospheric NO_2 columns have been widely used for evaluating the recent dramatic increase in NO_x emissions over China (e.g., Richter et al., 2005; van de A et al., 2006, 2008; Stavrakou et al., 2008). We have developed a 10-year trend of NO_x emissions in China during 1995–2004 and compared it with the satellite observations (Zhang et al., 2007b). The growth rate from the emission inventory is lower

than that from the satellite observations. We found quantitative agreement during summertime but a large discrepancy during winter time. Additional analysis is needed to find the reason of the discrepancy, but the consistency between the summertime trends suggests that the bias cannot be associated with systematic error of the basic inventory data. In this work, we estimate that NO_x emissions in China increased by 55% during 2001–2006, at a 9.2% annual growth rate. This is comparable with the satellite-inferred trend of NO_2 columns over China for recent years (Table 11).

Satellite-inferred trends also show good agreement with our inventory for other species. Tanimoto et al. (2008) obtained a 3.8% annual growth rate of Chinese CO emissions during 2000–2005, by using MOPPIT satellite observations and inverse modeling methods. This is in good agreement with the 3.4% annual growth rate during 2001–2006 from this work. van Donkelaar et al. (2008) analyzed aerosol data from MISR and MODIS for 2000–2006 with the GEOS-Chem model to estimate annual growth in Chinese sulfur emissions of 6.2% and 9.6%, respectively, which is comparable with the 6.3% annual growth rate of Chinese SO_2 emissions during 2001–2006 in this work.

4.3 Main uncertainties in this inventory

Compared with the TRACE-P inventory, we believe that emission estimates for China in this work are significantly improved as a result of our detailed technology approach and other methodological improvement. We also believe that the emission estimates for Japan, Korea, and Taiwan in this work are improved from the TRACE-P inventory, as they rely on thorough inventory compilation at the local level with local knowledge. For other Asian countries (mainly in Southeast Asia), the accuracy of the estimates is less than in other regions, as activity data were extrapolated from the year 2004 and few local emission factors were applied. So users should be cautioned if using this inventory for the Southeast Asia region specifically. The uncertainty of this inventory in Southeast Asia is at about the same level as the TRACE-P inventory, because the same methodology was used.

For the TRACE-P inventory, a detailed uncertainty analysis was performed for each species by combining emission measurement uncertainties with uncertainties in activity levels (Streets et al., 2003a). The overall uncertainty in TRACE-P emissions for all of Asia was calculated as follows, ranked in increasing order of uncertainty and measured as 95% confidence intervals: $\pm$16% (SO_2), $\pm$37% (NO_x), $\pm$130% (NMVOC), $\pm$185% (CO), $\pm$360% (BC), and $\pm$450% (OC). It is impossible for us to conduct such an analysis for the whole of Asia in this work, since we use several local emission inventories and their uncertainties are not known. However, we have repeated the TRACE-P uncertainty analysis for China, by taking into account the new emission factor data and the improved estimates of activity levels by sector, fuel, and technology type. The uncertainties for China's emissions in the year 2006 are estimated as follows: $\pm$12% (SO_2), $\pm$31% (NO_x), $\pm$68% (NMVOC), $\pm$70% (CO), $\pm$132% (PM_{10}), $\pm$130% ($PM_{2.5}$), $\pm$208% (BC), and $\pm$258% (OC). Compared with the TRACE-P inventory, the uncertainties in SO_2 and NO_x emissions are similar, while other species show significant improvements in accuracy. These improvements are mainly gained from the technology-based methodology.

The uncertainties in emission estimates for China vary considerably across sectors and source types. Uncertainties in power-plant emissions are less than for other sectors, because the activity data are well known and local emission factors are available. This is confirmed by a recent unit-based power-plant emission inventory for China (Zhao et al., 2008): our estimates are in good agreement with the unit-based estimates. We also feel comfortable with the estimates for the cement industry and the iron and steel industry, because factory-level information was applied when building this inventory. The larger remaining uncertainties are now confined to small industries, residential combustion, and the transportation sector.

4.3.1 Small industries

Bond et al. (2004) concluded that coke production and brick production are important emitting sources for China, which were omitted in previous inventory studies. Cao et al. (2006) also identified rural industry in China as an important contributor of carbonaceous aerosol emissions. These small industries are thought to be highly polluting, because the low level of technology inhibits the use of efficient control devices. In this work, we estimate that industrial processes contributed 0.45 Tg BC and 0.50 Tg OC in 2006, mostly from coke production and brick production, by using the same emission factors as in Bond et al. (2004). However, we are not confident about these numbers, because those emission factors are based on very few measurements. Emission factors for coke ovens were determined using assumptions about actual operation conditions that have 100% uncertainty (Bond et al., 2004, Sect. 5.2.3). There is no information available on measured emission factors of traditional brick production. Emission factors were interpolated between values for home-heating stoves and stoker-fired boilers with high uncertainty (Bond et al., 2004). Local measurements on those sources are required to remedy this situation and narrow the range of uncertainty.

4.3.2 Residential coal combustion

The residential sector has been identified as a major cause of uncertainty in estimates of CO, NMVOC, and carbonaceous aerosol emissions in Asia (Streets et al., 2003a), because of the lack of reliable statistics and local emission factor measurements. In this work, we follow the method used in the TRACE-P inventory for the residential sector, by applying a

Table 11. Comparison of emission trends over China with top-down inferred methods.

Species	Study	Method	Period	AGR (%/yr)[a]
SO_2	van Donkelaar et al. (2008)	modeling and satellite	2000–2006	6.2, 9.6[b]
	This work	inventory	2001–2006	6.3
NO_x	Richter et al. (2005)	satellite	1996–2004	8.7[c]
	Uno et al. (2007)	forward model	2000–2002	8–9
	Zhang et al. (2007b)	inventory	1996–2004	6.1
	L. Zhang et al. (2008)	inverse modeling	2000–2006	12.2[d]
	Stavrakou et al. (2008)	satellite	1997–2006	7.3
	This work	inventory	2001–2006	9.2
CO	Tanimoto et al. (2008)	inverse modeling	2001–2005	3.8
	This work	inventory	2001–2006	3.4

[a] AGR = annual growth rate; [b] 6.2% AGR was inferred by MODIS satellite data, and 9.6% AGR was inferred by MISR; [c] Richter et al. (2005), presented trends for 1996–2002, here extended to 1996–2004 using the same dataset; [d] L. Zhang et al. (2008), argued that NO_x emissions in China increased by a factor of two during 2000–2006, constrained by GEOS-Chem Model and OMI observations, equal to a 12.2% AGR.

uniform emission factor for a given fuel for the whole sector. However, large variations of BC emission factors have been observed in residential coal combustion in recent measurements (Chen et al., 2005, 2006; Y. Zhang et al., 2008; Zhi et al., 2008), depending on coal type (bituminous or anthracite) and combustion type (raw coal or briquette). In this case, the average BC emission factor in residential combustion is dominated by the share of bituminous coal use and the share of raw coal use, because BC emissions from bituminous raw coal combustion are 30–500 times higher than others (Table 12). However, we don't know the real-world fraction of coal briquette use in China. The China Energy Statistical Yearbook only reports a small portion of residential coal use (<10%) as coal briquettes, but actually coal briquettes are widely used in the residential sector. This results in difficulty in determining BC emission factors accurately. Figure 10 illustrates the dependence of BC emission factors on bituminous coal use and coal briquette use in China's residential coal combustion. We were able to identify BC emission factors for a few provinces such as Beijing, Fujian, and Shaanxi, but we do not know the situations in other provinces. This may result in significant errors in the regional distribution of BC emissions in China when applying a uniform emission factor for the whole of China. Remedying this problem will require a reassessment of China's official statistics for the residential sector.

Table 12. BC emission factors for residential coal combustion.

	Bituminous coal	Anthracite coal
Raw coal	3.81[a]; 2.75[c]; 3.32[d]	0.007[a]; 0.028[c]; 0.004[d]
Coal briquette	0.082[a]; 0.28[b]; 0.095[c]	0.004[a,b]

[a] Zhi et al. (2008); [b] Chen et al. (2005); [c] Y. Zhang et al. (2008); [d] Chen et al. (2006).

4.3.3 Vehicle emissions

Recent estimates of China's vehicle emissions are surprisingly close (see Table 13), but this does not mean that the accuracy of the estimates is high. Even for developed countries, estimating vehicle emissions is still difficult. For example, Parrish (2005) argued that CO emissions from on-road vehicles in the US were overestimated by about a factor of two in USEPA's National Emissions Inventory. In China, the quality of the vehicle emission inventory is downgraded by a couple of issues. First, transportation-related activity data are not well reported through the national or provincial statistical systems. Vehicle population data are usually available at provincial level only with simple classifications; transportation energy consumption is not properly reported in statistics; and systemic surveys of vehicle travel mileage are also absent. We have to use modeling methods to obtain some of the necessary activity data (He et al., 2005). On the other hand, there is no transportation emission model specifically designed for the China situation available for national emission inventory development. Emission models from developed countries have usually been applied in the past, using base emission factors from their own databases with assumptions and adjustments to match China's fleet structure (Streets et al., 2003a; Cai and Xie, 2007).

Recent efforts to develop high-resolution vehicle emission inventories for China are encouraging (Liu et al., 2007; Huo et al., 2009; Wang et al., 2009). Bottom-up methods were used in these studies, by collecting data on vehicle fleet composition, daily travel mileage, and driving patterns at city level, which can reduce estimation uncertainties significantly. Extending these activities to more cities would be very beneficial for national emission inventory development. Vehicle tests are also ongoing, to begin the development of emission factors specific to China. Continued

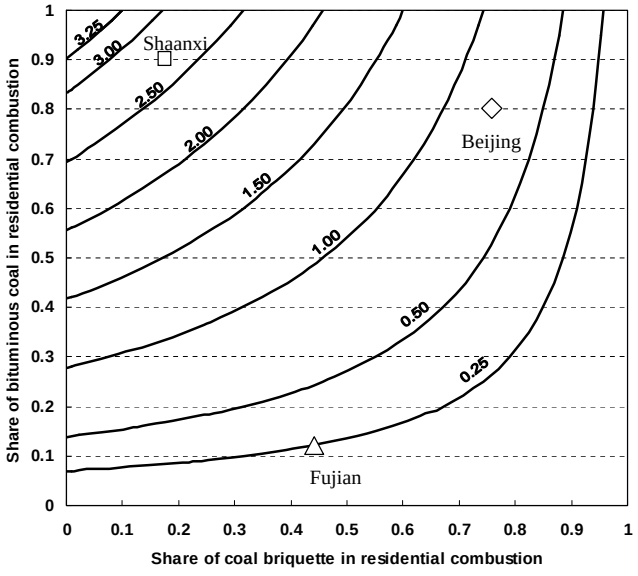


Fig. 10. Dependence of BC emission factors on bituminous coal use and coal briquette use in China's residential coal combustion. Contour lines represent the average BC emission factor (unit: g/kg-coal) in residential coal combustion. Diamond, square, and triangle dots represent the average BC emission factors in Beijing, Shaanxi, and Fujian, respectively.

updates and improvements in this field are required to gain a better understanding of emissions in China.

4.4 Applications and evaluations of the inventory

This inventory has been used in the modeling community during and after the INTEX-B mission. Initial results from the INTEX-B experiments are encouraging. By using this inventory and the GEOS-Chem model, van Donkelaar et al. (2008) and L. Zhang et al. (2008) examined the characteristics of Asian outflow and found that the model provides a good simulation of the SO_x, SO_4^{2-}, ozone, NO_x, and PAN mean vertical profiles observed from INTEX-B aircraft. By comparing with the trend in satellite retrievals, they also found that this inventory well represented the SO_2 and NO_x emission increases in Asia between 2000 and 2006.

The NO_x budget over East Asia has been investigated by integrating atmospheric models, satellite observations, and this inventory. Good agreement was found. Han et al. (2009) suggested that our estimates over North China, coupled with another Korean inventory, appear to provide a better estimation of the real world situation compared to the TRACE-P inventory. In two most recent inversion modeling studies using OMI and GOME2 satellite observations, the a posteriori estimates suggest that the NO_x budget was close to (Lin et al., 2009) or slightly lower than (Zhao and Wang, 2009) the bottom-up estimates.

Table 13. China's vehicle emissions from recent inventory estimates (units: Tg/year).

	Year	NO_x	CO	VOC
Cai and Xie (2007)	2005	4.5	36.2	5.9
Bo et al. (2008)	2005	–	–	5.5
Wei et al. (2008)	2005	–	–	5.6
This work	2006	5.1	33.7	6.6

Several regional modeling studies have used this inventory and compared their model predictions against surface observations in China. Both consistencies and inconsistencies between model and observation were both found in those studies. Zhao et al. (2009) simulated ozone pollution over East China in 2004 by using a regional chemical transport model (REAM) and this inventory. The model errors for ozone simulation are within the recommend ranges of the USEPA, but the model significantly underestimates some ozone peaks at Mt. Tai. Chen et al. (2009) used a nested GEOS-Chem model and this inventory to investigate regional CO pollution over China. They found that the model predicted surface CO concentrations well at a suburban site near Beijing but underestimated CO concentrations significantly in the Beijing urban area during summer 2005. Finally, Matsui et al. (2009) found that CMAQ model calculations using this inventory cannot agree with all of the observations made in downtown and rural Beijing sites in summer 2006. Systematically large SO_2/CO and BC/CO ratios as compared with observations were found, indicating there are some inconsistencies in the emission data. Although emission estimates for the whole of China are thought to be more accurate than previous studies, there can still be large uncertainties remaining in individual locations. This is because emissions are estimated at the provincial level, while emission patterns may vary within a province due to local differences in economic and industrial structures. The next step of our work will be to better present the spatial pattern of the inventory at local level, by using more region-specific activity data/emission factors, and high-resolution GIS databases.

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