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A high-resolution emission inventory for eastern China in 2000 and three scenarios for 2020

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Abstract

We develop a source-specific high-resolution emission inventory for the Shandong region of eastern China for 2000 and 2020. Our emission estimates for year 2000 are higher than other studies for most pollutants, due to our inclusion of rural coal consumption, which is significant but often underestimated. Still, our inventory evaluation suggests that we likely underestimate actual emissions. We project that emissions will increase greatly from 2000 to 2020 if no additional emission controls are implemented. As a result, $PM_{2.5}$ concentrations will increase; however O_3 concentrations will decrease in most areas due to increased NO_x emissions and VOC-limited O_3 chemistry. Taking Zaozhuang Municipality in this region as a case study, we examine possible changes in emissions in 2020 given projected growth in energy consumption with no additional controls utilized (BAU), with adoption of best available end-of-pipe controls (BACT), and with advanced, low-emission coal gasification technologies (ACGT) which are capable of gasifying the high-sulfur coal that is abundant in China. Emissions of NH_3 are projected to be 20% higher, NMVOC 50% higher, and all other species 130–250% higher in 2020 BAU than in 2000. Both alternative 2020 emission scenarios would reduce emissions relative to BAU. Adoption of ACGT, which meets only 24% of energy service demand in Zaozhuang in 2020 would reduce emissions more than BACT with 100% penetration. In addition, coal gasification technologies create an opportunity to reduce greenhouse gas emissions by capturing and sequestering CO_2 emissions below ground.

Keyword: Air pollution; Particulate matter; CO_2; Energy technology; Coal

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1. Introduction

We develop the high-resolution emission inventory described here in order to examine the link between energy consumption and technologies, air pollution and resulting impacts on public health in eastern China in 2000 and under different energy technology and environmental control scenarios in 2020. The health impact analysis is reported in Wang and Mauzerall (2005) and required the emission inventory to meet the following criteria. First, it needed to explicitly relate emissions in the region to source characteristics such as fuel type, economic sector, energy technology, and end-of-pipe controls, if any. Second, it needed to include major primary pollutants in the region, including gaseous species and particulate matter (PM) that significantly impact human health. Third, it needed to have as high a spatial and temporal resolution as possible in order to accurately represent population exposure. Fourth, the smallest administrative unit (such as county) for which the input data (such as population and fuel consumption) are available needed to be used.

Several studies have developed emission inventories of air pollutants for China for the present (Hao et al., 2002; Kato and Akimoto, 1992; Klimont et al., 2002; Streets et al., 2003) and the future (e.g. (Streets and Waldhoff, 2000; van Aardenne et al., 1999)). These emission inventories were compiled on a provincial basis and included one to several gaseous species and black carbon. Streets et al. (2003) is the most recent and comprehensive emission inventory available for China. However, it does not include primary PM emissions and the emissions of gaseous species are aggregated by province and major economic sectors and therefore did not meet our requirements.

In this paper, we develop a source-specific, high-resolution regional inventory of anthropogenic and biogenic emissions for the region of eastern China shown as the smaller solid rectangle in Fig. 1. Year 2000 is chosen as the base year, as it is the latest year for

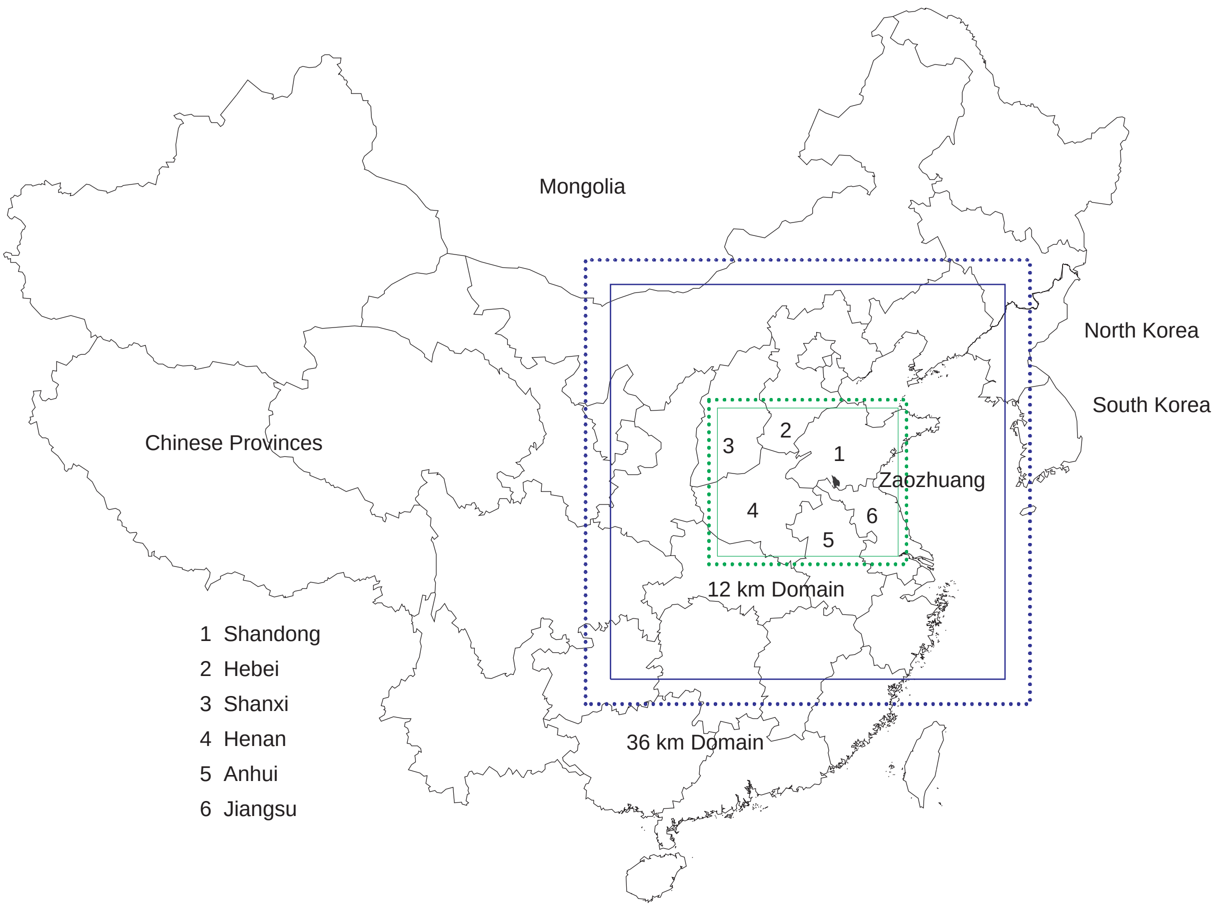

Fig. 1. MM5 and CMAQ model domains. The solid rectangles demarcate the 12 and 36 km CMAQ domains. The dashed rectangles represent the MM5 model domains.

which Chinese government statistics on energy consumption and socioeconomic indicators were available at the inception of this study. We select Zaozhuang in Shandong Province as a case study and estimate emissions in 2020 under three different scenarios.

Section 2 of this paper describes the methods used to estimate annual anthropogenic emissions from source categories, to conduct spatial and temporal allocations and chemical speciation of emissions and to evaluate the inventory. Section 3 presents the results, Section 4 analyzes the uncertainties involved in the emission estimates and Section 5 concludes.

2. Methodology

2.1. Estimating annual emissions in 2000

Like the emission inventories mentioned above, we estimate annual anthropogenic emissions as annual rates of emission-related activities multiplied by respective emission factors. The emissions are categorized by chemical species, municipality, economic sector or sub-sector, fuel or activity type and abatement technology (Eq. (1)). The resulting emissions are in the form of annual emissions for a specific chemical species, a municipality and a sector or sub-sector.

$$E_{j,k,l} = \sum_{m} \sum_{n} A_{j,k,l,m,n} \mathrm{EF}_{j,k,l,m,n}, \tag{1}$$

where E is emissions, A is activity rate, and EF is after-abatement emission factor. j, k, l, m, and n indicate the species, municipality, sector or sub-sector, fuel or activity, and type of abatement technology. EF values are estimated considering varying coal and oil quality with respect to ash and sulfur content as well as retention of ash and sulfur in boilers or other combustion installations during combustion.

As most energy and activity data in China is available only at a municipal or more aggregated level, the inventory is compiled based on municipality and includes 87 municipalities in 6 provinces (Shandong, Hebei, Shanxi, Henan, Anhui, and Jiangsu). The inventory includes 8 species: carbon monoxide (CO), ammonia (NH_3), nitrogen oxides ($NO_x = NO + NO_2$), $PM_{2.5}$ and PM_{10} (PM 2.5 or 10 μm in diameter or smaller, respectively), sulfur dioxide (SO_2), non-methane volatile organic compounds (NMVOC) and carbon dioxide (CO_2). CO_2 emissions are estimated to illustrate the potential co-benefits of greenhouse gas reduction resulting from local air pollution control.

The major sectors included in the inventory are residential, power generation, industrial/commercial, transportation and agriculture. The inventory does not include emissions from off-road traffic (e.g. tractors and inland shipping), construction or forest fires and hence may underestimate total emissions. Anthropogenic emission sources are divided into three major categories: large point sources (LPS), area, and mobile sources, which are described below. Biogenic emissions for the study region are generated using a preliminary version of the Model of Emissions of Gases and Aerosols from Nature (MEGAN) described by (Guenther et al., 2005).

2.1.1. Large point sources

Each large point source is located by latitude and longitude. There are 24 LPS in the 6 provinces of interest, of which 20 are power plants, and the others are iron and steel manufacturers. Other LPS, which cannot be identified individually due to lack of data are included in the area source category.

2.1.2. Area sources

This category includes all stationary sources that are not included in the LPS category. Each area source is defined by a municipality code and a sector or sub-sector code. There are 134 sectors or sub-sectors for area sources.

The data on activity levels are taken from China's official statistics at the national, provincial and municipal level, the governmental agencies in charge of overseeing industries, or sources that reference China's official statistics (see Appendix A of Supplemental Materials for references). Where the requisite municipal data are not available, we prorate the provincial data for each category using appropriate socio-economic statistics such as population, area, industrial output or GDP, as shown in Table 1. We primarily rely on the statistics published by various government agencies of China in order to have consistent data sources. However, data from different government agencies does not always agree. When faced with discrepancies we use our judgment to choose a data source. For example, rural domestic coal consumption in 1999 provided in the most recent China Energy Statistical Yearbook 1997–1999 (National Bureau of Statistics of China, 1999) is significantly lower than that provided by China Rural Statistical Information 2000 (Ministry of Agriculture of China, 2001), by from 34% for Shanxi to 93% for Shandong. This discrepancy cannot be explained by inter-annual variation between 1999 and 2000. Rather, it is likely further evidence of the lack of reliability in China's energy data [e.g. (Sinton, 2001)]. Evaluation of the regional emission estimates by Streets et al. (2003) using the TRACE-P observations suggests an under-estimation of emissions from the domestic sector (Carmichael et al., 2003). Since Streets et al. (2003) estimates are based on the National Bureau of Statistics' energy data, we believe the higher figures from the Ministry of Agriculture of China better reflect reality, and thus use them in our subsequent analysis. To be

Table 1
Proxies used for disaggregating provincial activity levels and emissions to municipalities and counties

Emission source category	Type of activity data	Proxy
Residential	Fuel consumption	Population
Power generation	Fuel consumption	Industrial output or industrial GDP
Industrial	Fuel consumption	Industrial GDP
Commercial/institutional	Fuel consumption	GDP of the tertiary industry
Industrial process	Industrial product output	Industrial output
Solvent utilization	Number of plants concerned, product output or population	GDP
Agriculture	Amount of municipal waste and agricultural residues openly burnt, fertilizer use, animals	Area or population
Transport	Vehicle population and/or fuel consumption	GDP

consistent, we take all other data for rural domestic energy consumption from the same source.

To estimate the emissions from agricultural waste burning, the amount of crop residue burnt in open fields is needed but is unavailable in government statistics. We estimate it as a fraction of the total amount of crop residues generated which is derived as a product of the output of different grain crops and the residue to crop ratio (RCR). RCR is the ratio of the portion of residues above ground to grain weight and varies by crop species. We obtain the RCRs for different types of crops from a review of relevant literature by (Wang and Mendelsohn, 2003). An estimated 15% of the total crop residues generated are burned in the field during the harvest seasons (Gu and Duan, 1998).

We rely on published studies in the context of China for emission factors, if available. Otherwise, for PM_{10} and $PM_{2.5}$ emissions from coal and oil fired industrial boilers, we use emission factors for similar activities from the US Air Pollution (AP) −42 database (USEPA, 1995). Emission factors and ash and sulfur contents of coal used in our analysis are listed in Appendix A of Supplemental Materials.

For NMVOC, we use the provincial NMVOC emissions estimated by (Klimont et al., 2002) with sector and sub-sector updates provided by Z. Klimont (personal communication, 2003). We regroup the sectoral NMVOC emissions into sub-sectors that match those in this study and allocate the provincial NMVOC emissions to municipalities using the proxies listed in Table 1.

2.1.3. Mobile sources

For CO, NO_x, and $PM_{2.5}$ emissions from mobile sources, activities are calculated as the number of each type of passenger and freight vehicle multiplied by the annual average mileage of each vehicle. Zheng et al. (2003) provides annual mileage by vehicle type in Zaozhuang. No data are available for other municipalities in our modeling region. As Zaozhuang is well connected to other municipalities by national and provincial highways, we assume the annual mileage for vehicles in Zaozhuang is representative of other municipalities in our region.

The EFs for mobile CO and NO_x emissions are taken from Fu et al. (2001). They include EFs at two different average driving speeds: 24 km h^{-1} and 45 km h^{-1} which are believed to represent in-city and highway driving speeds, respectively, in China. We assume that, on average, 20% of vehicle miles traveled (VMT) is accumulated on city roads and 80% is on highways and weight the EF accordingly. For $PM_{2.5}$ emissions, we include black carbon (BC) but neither organic nor inorganic carbon emissions in our estimates due to lack of information on these EFs for China. Since BC represents about 50–65% of $PM_{2.5}$ emissions from automobiles, we potentially underestimate $PM_{2.5}$ emissions from mobile sources. However, as we will show in Section 3 $PM_{2.5}$ emissions from mobile sources account for less than 2% of total anthropogenic $PM_{2.5}$ emissions in 2000 in the study region (see Fig. 4); thus the impact on total $PM_{2.5}$ emission estimates is negligible.

SO_2 emissions from mobile sources are included in the category of area emissions and are estimated based on the quantity of gasoline and diesel fuels consumed.

2.2. Emission projections for 2020

We estimate anthropogenic emissions in 2020 under three possible future energy technology scenarios which all meet the same level of projected energy service demand but result in sizeable differences in emissions of air pollutants. The first scenario applies only current conventional energy and environmental technologies to satisfy future energy demand (called 'business-as-usual' or 'BAU'), the second uses conventional energy technologies with best available end-of-pipe environmental controls (called 'best-available-control-technology or 'BACT'), and the third adopts advanced, low-emission coal-gasification technologies (called 'ACGT'). Both BACT and ACGT are applied to Zaozhuang only.

With ACGT, coal is gasified to produce synthesis gas ('syngas'). After removal of fine particles, H_2S and sometimes CO_2, syngas primarily consists of H_2 and CO. Syngas can be directly used as fuel, used to co-produce electricity or used to produce liquid fuels such as dimethyl ether (DME) and methanol (thus also called "polygeneration", see (Larson and Ren, 2003) for details). Burning syngas is more energy efficient than directly burning coal in industrial boilers (Wang, 2004). ACGT can use high-sulfur coal, which is abundant in the region. It also makes possible capture and underground storage of CO_2 from flue gas rather than the usual direct release to the atmosphere. Secure, long-term underground storage of CO_2 is being widely explored today (Rubin et al., 2004). Although China is not presently obliged to control greenhouse gas emissions, ACGT is of strategic future importance because it could facilitate continued use of China's abundant carbon and sulfur intensive coal resources, while simultaneously reducing emissions of SO_2 and CO_2 to the atmosphere thus facilitating compliance with possible future carbon emission constraints. There are no apparent technological barriers to this occurring: the key enabling technologies for a polygeneration system, including large-scale gasifiers, advanced gas turbines and liquid phase synthesis reactors, are either commercially available (gasifiers and gas turbines) or will be commercially available in the coming few years (Larson and Ren, 2003; Zheng et al., 2003). However, concerted policy efforts are needed to encourage such developments.

The general methods for estimating the 2020 emissions under these energy technology scenarios are the same as for the year 2000. We adjust activity levels and emission factors to reflect projected changes from 2000 to 2020. For BAU, we use the growth rates for population, GDP and energy consumption by sector in Zaozhuang from Zheng et al. (2003) (Table 2). Vehicle population in Zaozhuang is projected to increase from 203,000 in 2000 to 606,000 in 2020 (Zheng et al., 2003). Most vehicles in China were company-owned and the VMT in 2000 was already quite high. For example, the annual VMT for small passenger cars was 20,000 km, according to the Zaozhuang Department of Motor Vehicle record (Zheng et al., 2003). As a result, we assume the average VMT in 2020 is the same as in 2000. In addition, vehicle fuel economy is also assumed to remain unchanged due to the offsetting effects expected from engine and comfort improvements (Zheng et al., 2003).

Zheng et al. (2003) socio-economic projection for Zaozhuang was based on Zaozhuang's government planning and development strategies such as the Tenth Five-Year Plan, GDP-Doubling Plan and Long-Term Objectives in 2020, and the Municipal Integrated Urban Planning report. The general assumption in Zheng et al. (2003) energy projection is that the per capita energy consumption of Zaozhuang in 2020 will be close to or equal to that of Beijing in 2000. We assume other municipalities in the region will experience the same rates of socio-economic and energy growth as Zaozhuang under BAU. For other activities unrelated to energy use such as fertilizer application and livestock production which are the major sources of NH_3 emissions, we assume their growth rates from 2000 to 2020 are 17%, the same as that projected for population growth in this region (Zheng et al., 2003). Compared to a growth rate of 24% for fertilizer application and 52% for livestock production between 2000–2020 in East Asia projected by Bruinsma (2003), this is a conservative assumption, leading to a possible underestimate of the 2020 NH_3 emissions.

Table 2
Projected 2020–2000 ratio for population, GDP and energy consumption of major fuels in Zaozhuang under BAU Zheng et al. (2003)

	2020–2000 ratio
Population	1.17
GDP	5.6
Primary (agricultural) sector	2
Secondary (industry) sector	5.2
Tertiary (service) sector	8
Urban residential final energy demand	
Raw coal	2.14
LPG	6.73
Town gas	1.49
Rural residential final energy demand	
Raw coal	10.3
Biomass	1.22
LPG	6.9
Biogas	2.75
Power	
Raw coal	3.18
Washed coal	1.90
Transport	
Gasoline	2.62
Diesel	2.32
Commercial final energy demand	
Raw coal	0.9
LPG	38.13
Town gas	1.49

Under BACT, final energy service demand in Zaozhuang is projected to remain at the BAU levels, but power plants will be equipped with desulfurization with an average sulfur removal rate of 90% (Zheng et al., 2003). Under BACT, the transport sector is projected to see a 75% decrease of all pollutants by 2020 due to enforcement of stricter exhaust standards and technological advances in catalytic converters (Zheng et al., 2003). The vehicle population and VMT in BACT are assumed to be the same as in BAU. Coal is a major fuel in the industrial sector and coal and biomass are major fuels in the residential sector of China. The emissions from these two sectors are generally dispersed. As massive, dispersed coal and biomass use is unique to China, there is little pollution control experience in developed countries from which to draw. Thus, it is difficult to assess the best available control technologies for the industrial and residential sectors in China in 2020. We believe that in addition to SO_2 the emission rates for other species will also decline over time due to regulation and technological improvements, but the size of the emission reduction from 2000 to 2020 is highly uncertain. Somewhat arbitrarily, we assume emissions from sectors other than power generation and transport in BACT are 20% lower than in BAU.

Under ACGT, final energy service demand in Zaozhuang is again assumed to remain at the BAU levels but we assume that seven polygeneration plants will be operating in Zaozhuang in 2020 to meet 24% of energy service demands within Zaozhuang as well as 10% of the energy needs in the residential and commercial sectors in three surrounding municipalities—Jining and Linyi in Shandong Province and Xuzhou in Jiangsu Province. Specifically, these plants will replace 26% of energy service demand in electricity generation, 34% in industry, 50–60% in residential and commercial sectors, and 50% of transport gasoline demand and 30% of transport diesel demand (Wang, 2004; Zheng et al., 2003). We assume the rest of the energy service demand in the modeling region under ACGT will be met by the same energy carriers as under BAU.

Emissions of air pollutants are significantly reduced with coal gasification based polygeneration processes and with the use of the coal gasification-based energy products. In the polygeneration system, essentially all sulfur in coal is removed from syngas by physical or chemical processes (Larson and Ren, 2003). The sulfur content of coal used for polygeneration is assumed to be a relatively high 3.7% (Zheng et al., 2003). Emissions of PM and other air pollutants from ACGT are negligible. Emissions from the use of DME in the residential and commercial sectors are low and are equivalent to liquefied petroleum gas (LPG) on a GJ fuel basis. Emissions from the use of LPG and thus DME are significantly lower than from raw coal in terms of SO_2, CO, and PM but slightly higher for NO_x (Zhang et al., 2000). M5 (a mixture of 5% methanol and 95% gasoline by molar fraction), M100 (transportation fuel grade methanol) and DME powered vehicles emit 10–70% less than gasoline or diesel powered vehicles (Table 3) (Zheng et al., 2003). Due to lack of data, we make the conservative assumption that there is no reduction in emission factors for $PM_{2.5}$ using alternative fuels.

Table 3
Ratios of emission factors of alternative vehicle fuels to conventional vehicle fuels. The ratios for HC, CO and NO_x are reproduced from Zheng et al. (2003)

Alternative over conventional fuels	HC	CO	NO_x	$PM_{2.5}$
M5/gasoline	0.9	0.9	0.9	1.0
M100/gasoline	0.8	0.8	0.8	1.0
DME/diesel oil	0.3	0.3	0.3	1.0

M5 = a mixture of 5% methanol and 95% gasoline in molar fraction; M100 = transportation fuel grade methanol.

2.3. Emission inventory evaluation through air quality model simulations

To evaluate our emission inventory for year 2000, we use the Community Multiscale Air Quality (CMAQ) model version 4.3 (Byun and Ching, 1999) with the year 2000 emissions estimated above to simulate surface concentrations of PM_{10} and SO_2. We then compare our simulated concentrations with measurements of SO_2 and PM_{10} from China's daily air quality reports obtained for 37 municipalities within the 12 km domain (see Appendix A for details). Results of the comparison will be described in Section 3; below we describe the configuration of CMAQ, SMOKE and MM5.

2.3.1. CMAQ configuration

CMAQ simulations require emissions and meteorology as inputs to generate tropospheric concentrations of more than 70 gaseous and aerosol species across the three-dimensional model domain. We use the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System (UNC Carolina Environmental Program, 2005) to spatially and temporally distribute our emission inventory and the fifth-generation Penn State/NCAR Mesoscale Model (MM5) (Grell et al., 1994) to generate meteorology to drive CMAQ.

We include two nested modeling domains in CMAQ (see Fig. 1). The smaller domain, for which we built the emission inventory, is defined such that Zaozhuang in Shandong Province, which is selected as a case study for examining the health impacts of air pollution resulting from energy use, is situated near the center. The domain is much larger than Zaozhuang in order to capture the effect of air pollution transport. The choice of the model domain size balances the availability of input data needed for the emission inventory, the scope of air pollution transport, and the computational resources available. The smaller domain is 648 km north–south and 792 km east–west with southwestern (SW) and northeastern (NE) corner coordinates of (32°N, 112°E) and (38°N, 120.6°E). It has a horizontal resolution of 12 × 12 km (hereafter called 'the 12 km domain'). A larger CMAQ domain with 36 km resolution (called 'the 36 km domain') is included to generate concentration boundary conditions for the 12 km domain. The SW and NE coordinates for the 36 km domain are (26.8°N, 107.3°E) and (42.5°N, 126.7°E), and the domain is 1728 km north–south and east–west. CMAQ has 13 vertical layers extending from the surface to 100 mb.

CMAQ initial conditions are set uniformly across the domains at a low value as predefined in CMAQ (Byun and Ching, 1999). We obtain boundary conditions for the 36 km domain based on monthly mean concentrations of a 1990 simulation of the global Model of Ozone and Related Tracers, Version 2 (MOZART-2) (Horowitz et al., 2003). We extract concentrations from MOZART-2 for both gas species (i.e. NO_2, NO, O_3, NO_3, OH, N_2O_5, HNO_3, H_2O_2, CO, SO_2, PAN, formaldehyde, high molecular weight aldehydes, isoprene, ethene, and NH_3) and sulfate aerosol and organic carbon.

To represent the four seasons of the year, CMAQ is run at both 12 km and 36 km resolution for 3–18 of January, April, July and October for the years 2000 and 2020. Our sensitivity analyses show that after the first 4 days the simulated concentrations converge with different initial conditions (Wang, 2004). Thus, we discard the first 4 days of CMAQ simulations.

2.3.2. *SMOKE configuration*

We use SMOKE version 1.4 to process the spatial and temporal allocation and chemical speciation of the emission inventory for the 12 km domain for use in CMAQ. SMOKE takes land use, meteorology and emission inventories as inputs and processes biogenic, mobile, area and large point sources separately. Gridded biogenic emissions generated using the biogenic emission model MEGAN are then merged with mobile, area and large point source emissions. SMOKE was originally developed for use in the US, and takes county-based emissions as input by design. Therefore, we use various proxies (see Table 1) to disaggregate the municipality-based emissions in the model region into county-based emissions.

Spatial allocation of emissions in SMOKE vary by emission source. For area sources, we use area as a surrogate to allocate county-total emissions to relevant grid cells. For mobile sources, county-based mobile emissions are apportioned based on the fraction of highway length of a county in a grid cell. Emissions from point sources are located in the grid cell in which the source is found.

We transform annual total emissions to hourly emissions based on monthly, weekly and daily allocation profiles. The temporal profiles are source specific. In 2000, residents in Zaozhuang consumed about four times more final energy for heating than for all other energy needs (such as lighting, cooking and hot water) combined (Zheng, 2003). We take Zaozhuang as representative of the study region, and assume that residential energy needs other than heating are constant throughout the year. Consequently, we obtain a monthly residential emission profile in which residential energy consumption during the winter heating months (November to April) is eight times higher than during other months of the year. We also assume that on weekends it is 20% higher than on weekdays since people in China spend more time at home on weekends. The daily residential emissions vary by hour of day for all pollutants concerned, as shown in Fig. 2. Residential emissions are highest during breakfast and dinner hours and lowest from late night to early morning. The daily profile is based on the author's (X. Wang) living experience in both urban and rural areas of this region as no survey data exists. Despite its lack of scientific rigor, we believe this is an improvement over the common practice of holding emissions constant throughout the day. Power sector (mainly coal fired) emissions from May to October are assumed to be 25% higher than between November and April because of the higher electricity demand in warm months, and are assumed constant throughout the day and week. Industrial sector emissions are nearly constant on a daily, weekly and monthly basis. Vehicle emissions have no month-to-month variation, but are assumed to be 20% higher on weekdays than weekends and about five times higher from 7 a.m. to 7 p.m. than other hours of the day.

Chemical speciation uses the Carbon Bond 4 (CB-4) chemical mechanism and source-specific speciation factors from (Hu, 2000) and (USEPA, 1995). NMVOCs are disaggregated into high molecular weight aldehydes, ethene, formaldehyde, isoprene, olefinic carbon bond, paraffin carbon bond, toluene, xylene and monoterpene.

Spatially distributed (but not temporally resolved) emissions for the 36 km domain were obtained from Streets et al. (2003). We then disaggregated annual

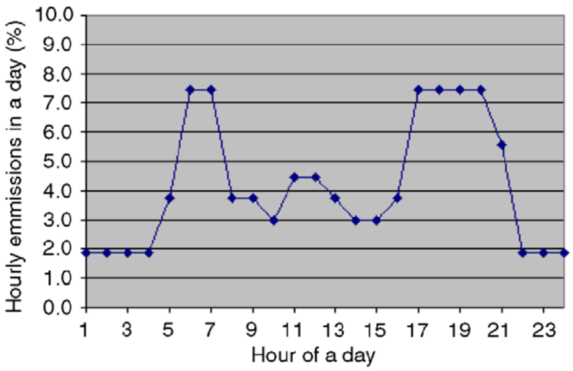

Fig. 2. Daily profile of residential emissions (author's estimate).

emissions into hourly emissions using the temporal profiles used in the 12 km domain.

2.3.3. MM5 configuration

The MM5 domains (dashed rectangles in Fig. 1) have the same horizontal resolution as CMAQ and are 3 grid cells larger on each side than the corresponding CMAQ domains. MM5 has 34 vertical layers extending from the surface to 100 mb.

In the study region, in January northerly winds prevail, in April easterly winds prevail in the south and westerly and southwesterly winds prevail in the north of the model domain, in July southeasterly and southerly winds prevail, and northwesterly and northeasterly winds prevail in October. The prevailing seasonal surface wind directions simulated by MM5 is similar to simulations of the NCEP Global Data Assimilation System (GDAS) (at a resolution of 2.5° latitude by longitude). Surface wind speeds are highest in January and lowest in April. Average air temperature was between 262 K and 278 K in January, 277 K and 289 K in April, 290 K and 302 K in July, and 279 K and 283 K in October. Identical meteorology is used for CMAQ 2000 and 2020 simulations.

3. Results

3.1. Emission inventory

3.1.1. Year 2000 emission estimates

Our year 2000 emission estimates by province are shown in Table 4. In 2000, the largest sector contributors to CO, NH_3, NO_x, PM_{10}, $PM_{2.5}$ and SO_2 emissions in the region are residences (53%), agriculture (99%), power generation (35%), industry (80%), industry (64%) and power generation (46%), respectively. In general, our regional estimates are close to Streets et al. (2003) except for NO_x for which ours are 70% higher and for SO_2 in Anhui province for which our estimates are more than twice as large. We believe the discrepancies mainly result from our more complete accounting of rural coal consumption and a higher NO_x emission factor for mobile sources. Our NMVOC emissions are slightly lower than Streets et al. (2003) because ours do not include emissions from savanna and forest burning.

We also compare our estimates with other available studies on specific pollutants (Table 5). Our estimated SO_2 emissions are several percent lower for Shandong and Shanxi, but 40–50% higher for Hebei and Jiangsu

Table 4
Comparison of year 2000 emission estimates (this study) with Streets et al. (2003). Units are in kton

Province	Data source	CO	NH_3	NO_x	PM_{10}	$PM_{2.5}$	SO_2	NMVOC	CO_2
Shandong	Wang et al.	6831	1645	1174	9539	2647	1756	1070	264,924
	Streets et al.	7339	1093	812			1967	1197	256,850
	Wang/Streets	0.9	1.5	1.4			0.9	0.9	1.0
Shanxi	Wang et al.	2806	288	807	5346	1532	1087	353	183,852
	Streets et al.	3254	208	558			1481	401	162,960
	Wang/Streets	0.9	1.4	1.4			0.7	0.9	1.1
Henan	Wang et al.	5702	1267	993	7486	2125	1775	704	231,550
	Streets et al.	5970	1135	534			1205	855	193,950
	Wang/Streets	1.0	1.1	1.9			1.5	0.8	1.2
Hebei	Wang et al.	6589	1173	1337	12,906	3591	1928	759	326,546
	Streets et al.	6806	840	686			1351	855	235,280
	Wang/Streets	1.0	1.4	1.9			1.4	0.9	1.4
Anhui	Wang et al.	4523	562	610	9419	2409	1044	419	148,393
	Streets et al.	4042	671	347			446	537	127,210
	Wang/Streets.	1.1	0.8	1.8			2.3	0.8	1.2
Jiangsu	Wang et al.	5166	1002	1079	10,535	3085	1817	862	246,991
	Streets et al.	6083	1007	693			1191	861	226,850
	Wang/Streets.	0.8	1.0	1.6			1.5	1.0	1.1
Regional Total	Wang et al.	31,617	5938	5999	55,231	15,390	9408	4167	1,402,256
	Streets et al.	33,494	4954	3631			7641	4707	1,203,100
	Wang/Streets	0.9	1.2	1.7			1.2	0.9	1.2

Notes: (1) Streets et al refers to Streets et al. (2003). Wang et al. refers to this study. Wang/Streets indicates the ratio of emission estimates by this study to Streets et al. (2003).
(2) NMVOC emissions in this study are taken from Klimont et al. (2002).

and over 100% higher for Anhui and Henan than SO_2 emissions reported by State Environmental Protection Agency (SEPA) of China (2001). As it is unclear how the SEPA data is collected, we cannot explain the difference. Our NH_3 estimates are about 10% (for Shanxi)–80% (for Hebei) higher than the 1992 emissions estimated by Sun et al. (1997) which we attribute to the growth of agricultural production between 1992 and 2000. Our NO_x estimates are 40–70% higher than the 1998 emissions estimated by Hao et al. (2002) due to our inclusion of NO_x emissions from both commercial and non-commercial energy use while Hao et al. (2002) included only emissions from commercial energy consumption.

Table 5
NH_3, NO_x and SO_2 estimates by other studies (unit: kton)

Province	NH_3 (Sun et al. 1997)	NO_x (Hao et al. 2002)	SO_2 (SEPA, 2001)
Shandong	985	755	1796
Shanxi	270	571	1202
Henan	1001	564	877
Hebei	647	7567	1321
Anhui	461	432	395
Jiangsu	593	749	1202
Total	3957	3828	6793

3.1.2. Year 2020 emission estimates

With no additional controls, the 2020 BAU emissions of NH_3 are 20% higher, NMVOC emissions 50% higher, and emissions of all other species are 130%–250% higher than in 2000 (Fig. 3 and Table 6). Emissions from the residential sector are projected to grow fastest because the highest rate of growth in energy demand is projected to be in residential energy consumption, particularly coal use. We believe growth in residential coal consumption can be attributed to both an increase in per capita coal consumption and a switch from biomass to coal. As a result, the relative contribution of residential emissions for all species is larger in 2020 BAU than in 2000. For all species, the 2020 ACGT emissions in Zaozhuang are close to 2000 emissions and much lower than both the 2020 BAU and 2020 BACT emissions (Fig. 3). The 2020 ACGT NO_x, $PM_{2.5}$, PM_{10}, SO_2 and CO emissions are 15%, 15%, 15%, 40% and 60% lower than the 2020 BACT emissions, respectively.

The sector contributions of emissions from Zaozhuang under emission scenarios for 2000 and 2020 vary (Fig. 4). Biogenic emissions in 2020 are assumed to be the same as in 2000.

CO_2 emissions in 2020 BAU are estimated to be 2.3 times higher than in 2000. As we did not consider additional energy needed for operating end-of-pipe controls under 2020 BACT, CO_2 emissions in 2020 BACT are the same as in 2020 BAU. With its market share as currently designed, the ACGT scenario with no CO_2 capture is estimated to emit 9% more CO_2 than

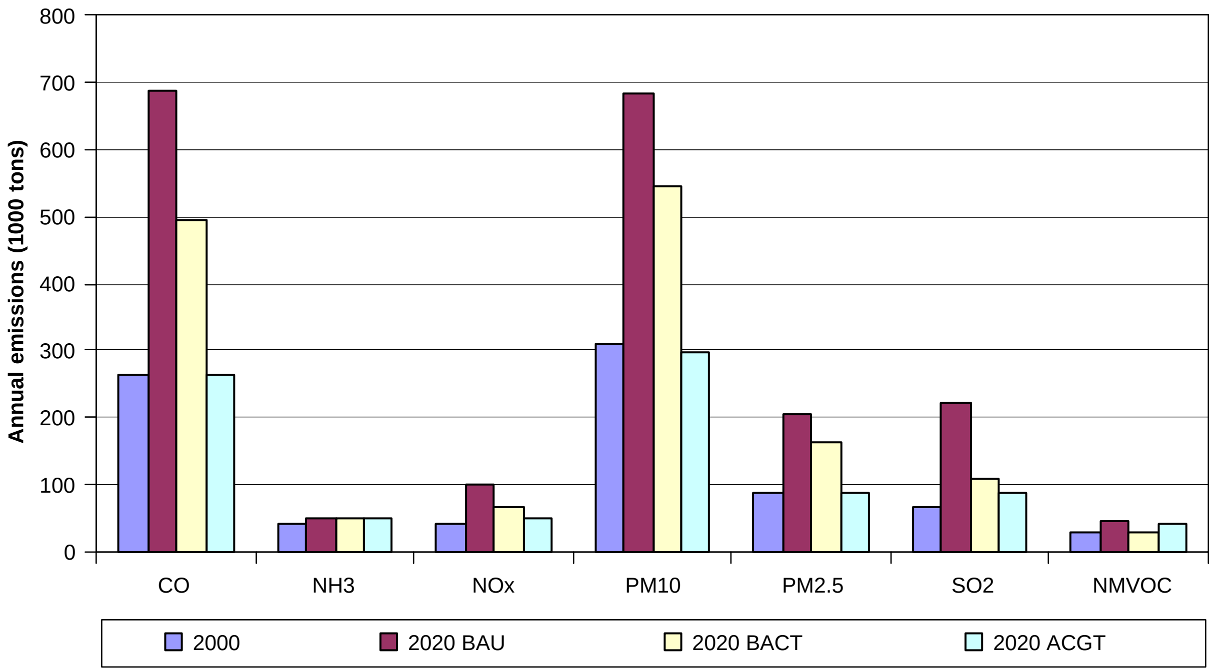

Fig. 3. Anthropogenic emissions of air pollutants from Zaozhuang in 2000 and under BAU, BACT and ACGT scenarios in 2020.

Table 6
Ratio of 2020 BAU to year 2000 emissions by sector and species (6-province average)

Sector	CO	NH_3	NO_x	PM_{10}	$PM_{2.5}$	SO_2	NMVOC[a]
Residential	3.9	1.4	5.7	6.6	6.6	8.5	0.8
Power			3.1	3.2	3.2	3.1	0.5
Industry	1.9		2.1	1.9	1.9	2.1	1.8
Transport	2.2		1.5	1.5	1.5		4.9
Agri/others	1.0	1.2	1.0	1.0		1.0	
All sectors	2.9	1.2	2.5	2.3	2.6	3.5	1.5

[a]Based on the estimates of NMVOC emissions Klimont et al. (2002) with updates from Z. Klimont.

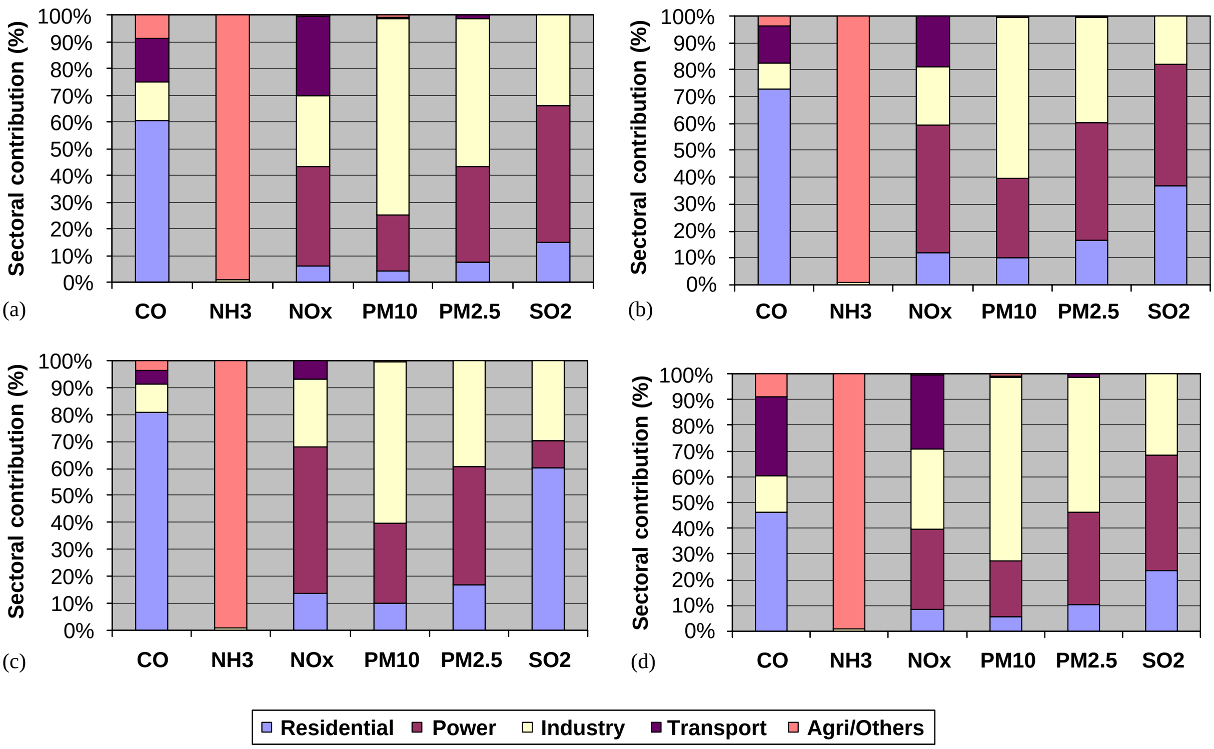

Fig. 4. Sector contributions of emissions from Zaozhuang in 2000 and under different 2020 emission scenarios. (a) 2000; (b) 2020 BAU; (c) 2020 BACT and (d) 2020 ACGT.

BAU or BACT. On a per unit energy service basis, polygeneration plants emit more CO_2 than coal-steam electric plants and substituting DME or methanol in the transport sector for gasoline or diesel emits less CO_2 (Larson and Ren, 2003; Zheng et al., 2003). As the increase outweighs the decrease, the net effect is an increase in CO_2 emissions. The ACGT scenario with CO_2 capture results in 13% less CO_2 from Zaozhuang than the ACGT scenario without CO_2 capture. All emissions under 2020 ACGT include emissions from coal polygeneration systems used to produce energy services exported to areas outside of Zaozhuang. These export-associated emissions are small.

Emission reductions in the three municipalities resulting from import of DME from Zaozhuang are barely noticeable (less than 4% for all species). However, export of energy services improves the economics of polygeneration in Zaozhuang by enabling economy of scale gains (Larson and Ren, 2003; Zheng et al., 2003) without adding any air pollution (except CO_2) to Zaozhuang.

3.2. Spatial and temporal distributions of emissions

The spatial distribution of population and selected pollutant emissions in 2000 is shown in Fig. 5. High anthropogenic emissions of CO, NO_x and SO_2

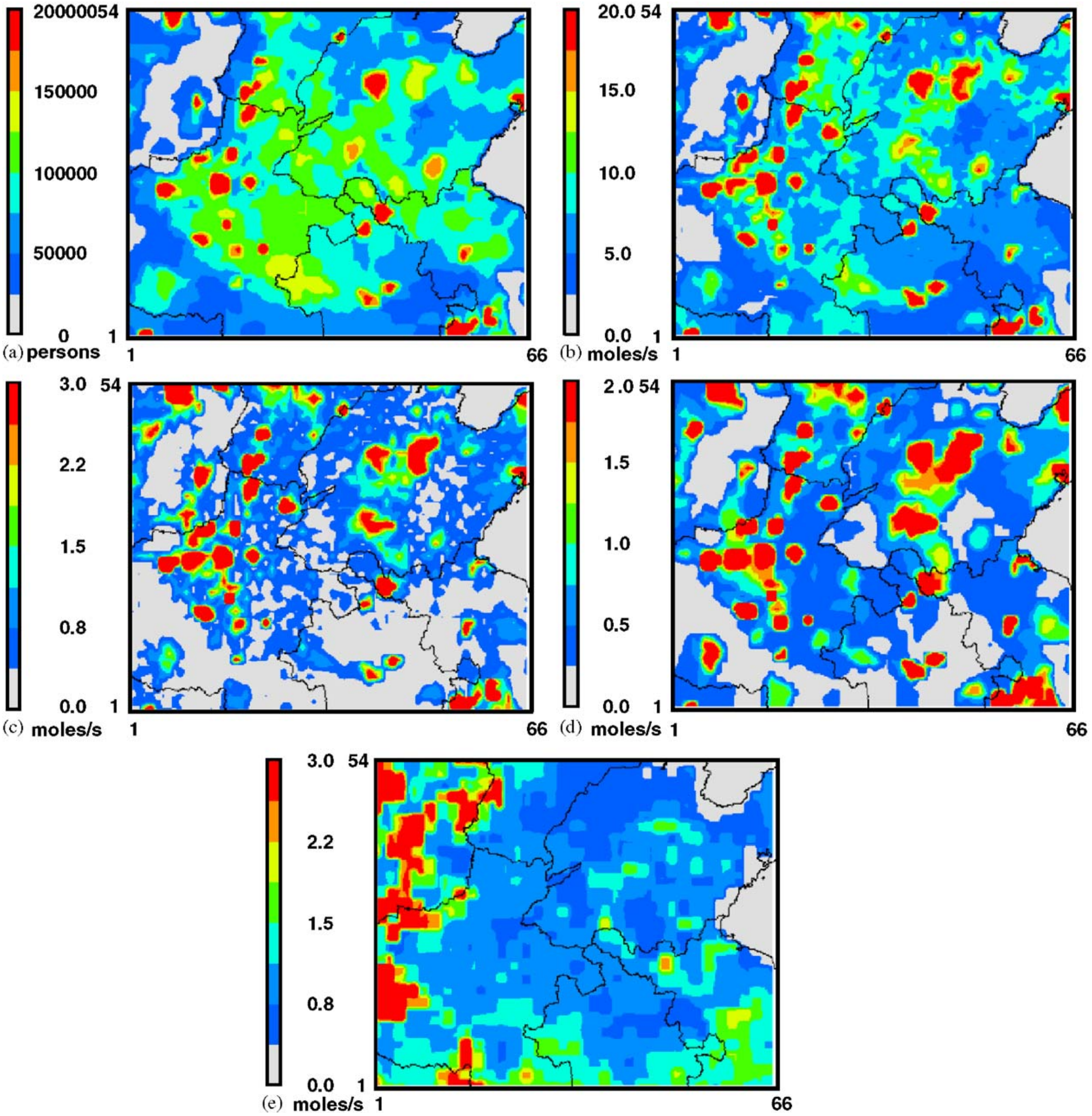

Fig. 5. Spatial distributions of population anthropogenic CO, NO_x, SO_2 and biogenic VOC surface emissions in the 12 km domain. The black lines in the background are provincial boundaries. (a) Population; (b) CO; (c) SO_2; (d) NO_x and (e) biogenic VOC.

correspond to urban areas with high population density (Fig. 5a) or places where industry concentrates. High biogenic emissions correspond to areas covered with vegetation and with low population density.

Surface CO, SO_2 and NO_x emissions show great variation between cold months (November–April) and warm months (May–October) due to the use of coal for space heating in the residential and commercial (included in the industrial sector) sectors. Biogenic VOC emissions are lowest in January and highest in July due to higher temperature, solar radiation and leaf area in summer than in winter.

3.3. *Inventory evaluation*

3.3.1. *Comparison of simulated and observed surface concentrations*

As shown in Fig. 6 (6 out of 37 municipalities exhibiting the range of agreement with available observations are shown, for others see Wang (2004)),

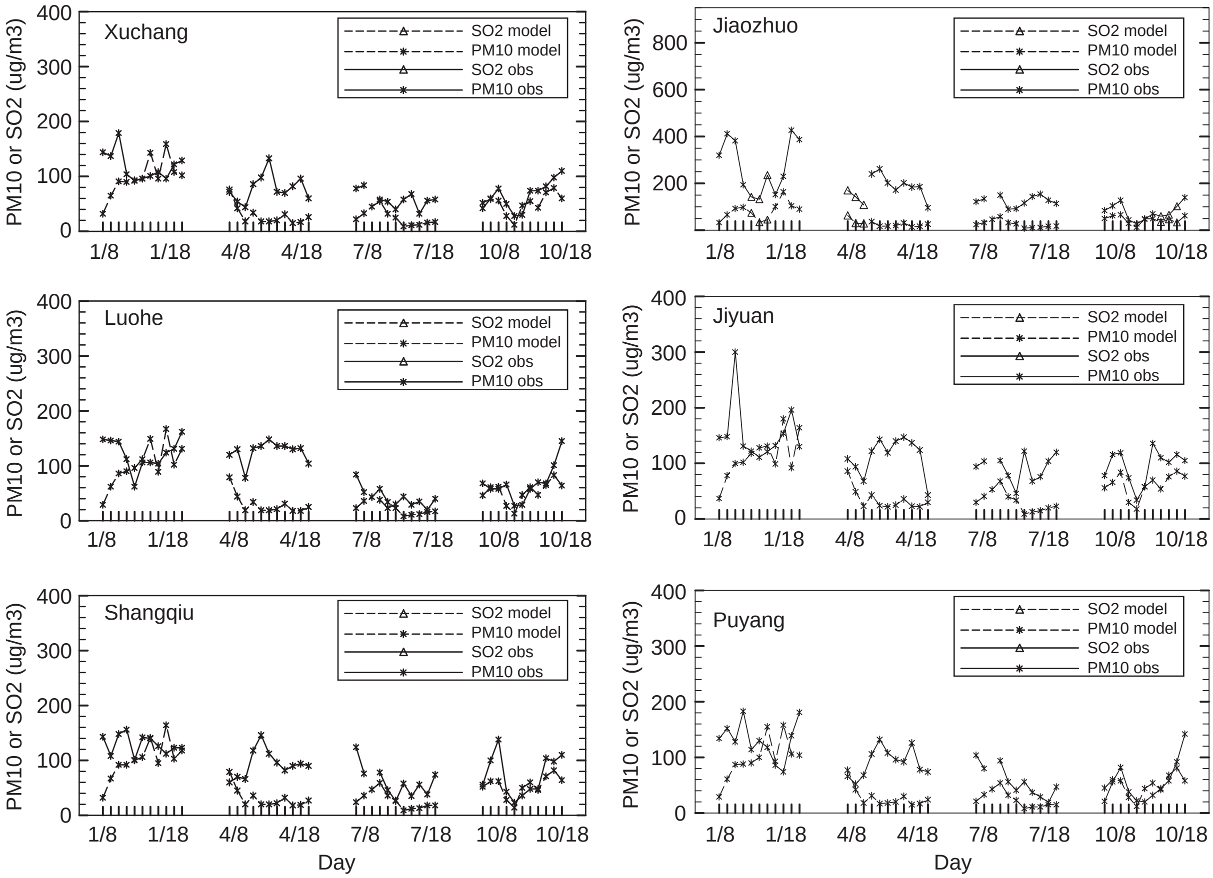

Fig. 6. Comparison of CMAQ-simulated ambient concentrations (dashed lines) in 2000 with observations (solid lines) of principal pollutants in different municipalities during 2002–2004. For a given day, the principal pollutant was either PM_{10} (asterisks) or SO_2 (triangles). Note that the *y*-axis scale is from 0 to 950 $\mu g\,m^{-3}$ for Jiaozhuo and from 0 to 400 $\mu g\,m^{-3}$ for other municipalities.

the simulated concentrations agree reasonably well with observations in October, but the model frequently under-predicts surface concentrations in April, and, to a lesser extent, in July. The model under-predicts annual average PM_{10} concentrations for the region by 50%, under predicting least for January (by 30%) and most for April (by 76%) with all 37 municipalities considered. The discrepancy between observations and model results can be attributed to several factors. First, the reported observations are averaged over the measurements of individual stations in a municipality while the simulated concentrations are averaged over an entire municipality. It is impossible for us to disentangle background and highly polluted areas in a municipality. Second, our emission inventory does not include desert dust, off-road machinery emissions, construction, etc. Third, there is a lack of specific Chinese emission characteristics for a number of sources and pollutants such as ammonia from animal production, PM from industrial combustion and processes, actual effectiveness of PM abatement equipment, VOC emission factors for a number of industrial processes, etc. As a result, we have to rely on those reported in the literature for other countries, if available. Fourth, the observations are from 2002–2004, but our model simulations use 2000 emissions and do not reflect potential recent increases in emissions. Fifth, the uncertainties inherent in measurements, our emission inventory and the air quality and meteorological models could contribute to the discrepancy. In sum, the comparison suggests that we are more likely to underestimate than to overestimate actual emissions particularly in spring.

3.3.2. *Simulated surface concentrations in 2000 and 2020 BAU in the model region*

In general, total $PM_{2.5}$ concentrations are highest in January and lowest in July as a result of higher emissions of $PM_{2.5}$ and its precursors such as SO_2 and NO_x in January (Fig. 7). High $PM_{2.5}$ concentrations occur in areas where emissions are high due to high

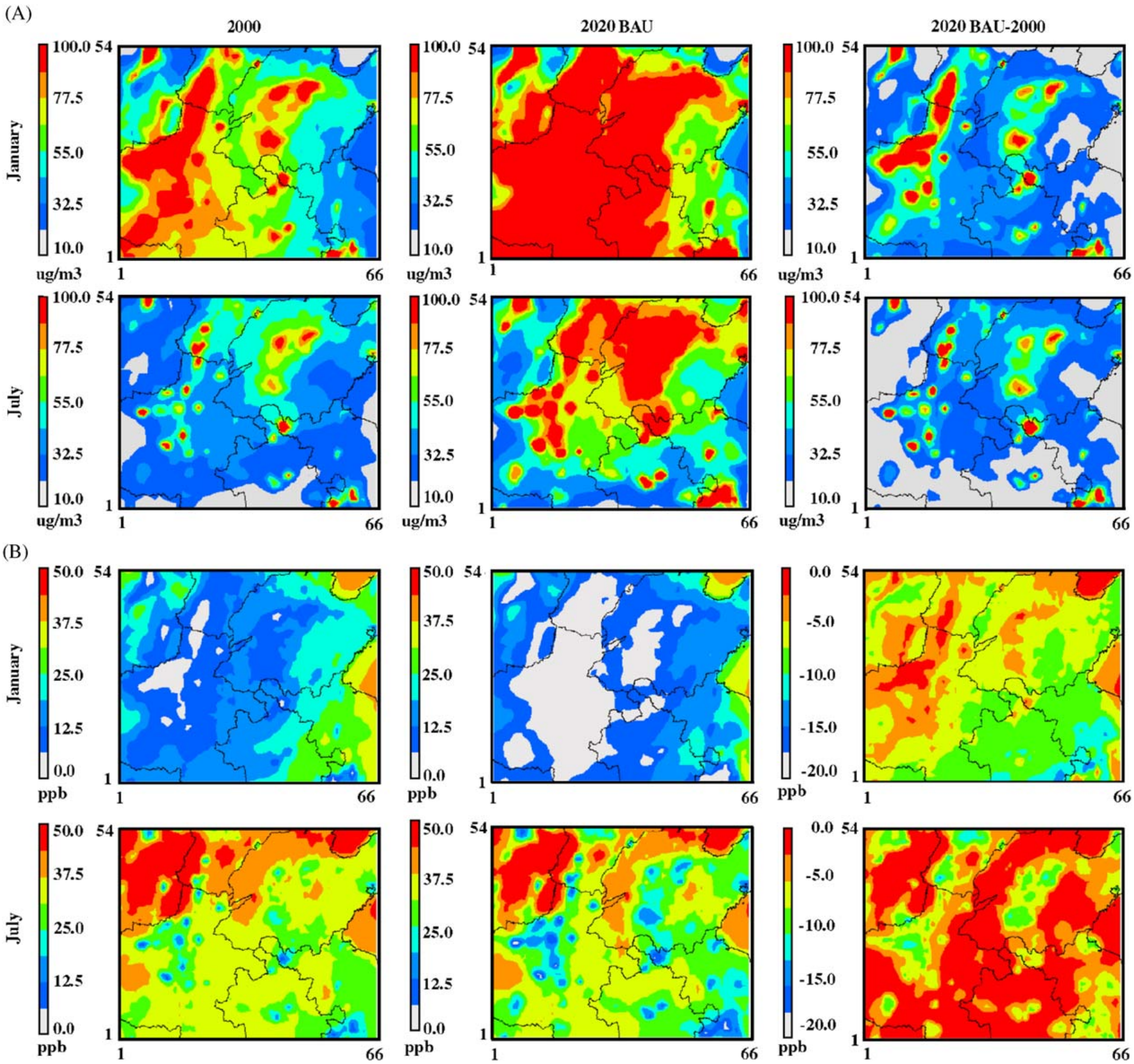

Fig. 7. Seasonal variations of surface $PM_{2.5}$ and O_3 concentrations in 2000 and 2020 BAU. (A) $PM_{2.5}$ and (B) O_3.

population density and/or industry. The 2020 BAU $PM_{2.5}$ concentrations are projected to be much higher than in 2000 in all four seasons.

Secondary $PM_{2.5}$, including sulfates (SO_4), ammonium (NH_4), nitrates (NO_3), and secondary anthropogenic and biogenic organic carbon aerosols, accounted for 12–84% (by weight) of total $PM_{2.5}$ in 2000 in the surface layer over the model region. Higher percentages occurred over the ocean and in areas with high forest coverage due to little primary PM emissions in these areas, and lower percentages occurred in areas with dense population and industrial activities where coal burning produced significant primary PM (Fig. 8). In 2020 BAU, we project the fraction of secondary PM to be lower than in 2000 because primary PM emissions will grow faster than emissions of secondary PM precursors primarily because of increasing use of coal in the residential sector. Concentrations of secondary $PM_{2.5}$ are quite evenly distributed throughout the model region largely because $PM_{2.5}$ is small in size and is transported long distances.

Simulated O_3 concentrations are highest in April and lowest in January. Areas with relatively high NO_x concentrations had relatively low O_3 concentrations.

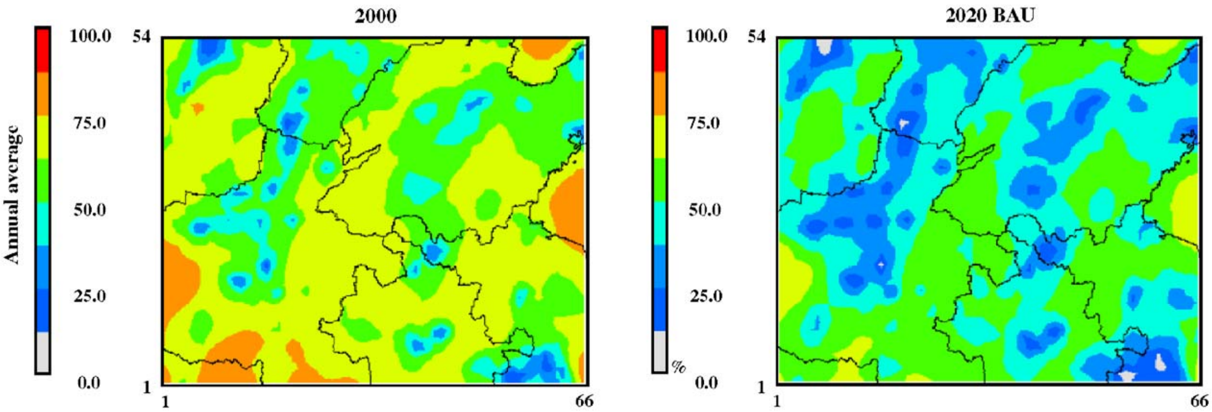

Fig. 8. Mass fraction (%) of secondary $PM_{2.5}$ in total $PM_{2.5}$ at the surface in 2000 and 2020 BAU.

Lowest O_3 concentrations occurred in January in areas of low VOC to NO_x concentration ratios (VOC/NO_x), which were generally less than 3 (Fig. 9). This resulted in VOC limited O_3 formation (Sillman, 1999) leading to lower O_3 concentrations at higher NO_x concentrations. O_3 production was VOC-limited throughout the year except July. In July, in areas where biogenic VOC emissions are high, the VOC/NO_x ratios increase significantly, ranging from single digits to 140 (Fig. 9). In urban areas with high population density the VOC/NO_x ratios remained less than 4 in July making O_3 production again VOC limited.

O_3 concentrations in 2020 BAU are projected to be lower than in 2000 because NO_x emissions are projected to grow faster than VOC emissions, and consequently the VOC/NO_x ratios will decrease in 2020. Simulations with the global tropospheric chemistry model MOZART-2 also projected that O_3 concentrations in this region will decrease between 1990 and 2020 in all seasons except summer due to projected increases in NO_x emissions (Wang and Mauzerall, 2004).

4. Uncertainties in emission estimates

The quality of our emission inventory is affected both by the quality of the input data we have collected and by use of extrapolation and our own judgment to fill the remaining data gaps. The reliability and accuracy of China's official statistics on which we rely have been heavily debated. First, coal production and, to a lesser degree, coal and oil consumption are believed to be under-reported largely due to illegal operation of the officially banned small coal mines, oil smuggling and technical lapses gathering energy consumption data from small enterprises and rural households (Sinton, 2001). Second, for the transportation sector, the statistics for ton–kilometers and passenger–kilometers for the 1990s is believed to miss a significant portion of the total traffic volume (Huenemann, 2001).

For missing data, we have to make assumptions and use indirect estimates, inevitably introducing uncertainty. We rely on socio-economic data to extrapolate information on activity levels, use EFs from another country (e.g. PM emission factors for the industrial sector from the US) or the global average EF (e.g. NH_3 emission factors from the agricultural sector) for the study region and use uniform coal quality data across municipalities in a province. In other cases, we have to make crude assumptions based on our best judgment, such as PM reduction potential under the 2020 BACT scenario in Zaozhuang, diurnal variation in residential emissions in the study region and projected agricultural production in 2020. There are also several missing sources in our inventory as mentioned earlier, as our emphasis is on energy related activities and the health effects attributable to them.

It is difficult to quantify the uncertainties in input variables such as emission factors and activity levels because there is little specific information on the subject and expert judgment is often used to assign estimates of uncertainties to emissions (e.g. (Hana et al., 2001; Streets et al., 2003)). Hana et al. (2001) show that the range of uncertainty in area emissions in the eastern US is $\pm 100\%$ and that of major point emissions is $\pm 50\%$ (95% confidence level) around the central estimates. Streets et al. (2003) estimate that the uncertainty of their year 2000 Asian emission inventory ranges from $\pm 13\%$ for SO_2 to $\pm 185\%$ for CO (95% confidence level).

To characterize the uncertainties involved in China's official statistics on energy use and socio-economic activity is beyond our reach. Nonetheless, we believe the uncertainty of our emission estimates is likely greater than that for the US inventory and thus propose a range of $\pm 100\%$ around the central estimates as the uncertainty in our emission inventory. As indicated by the comparison of

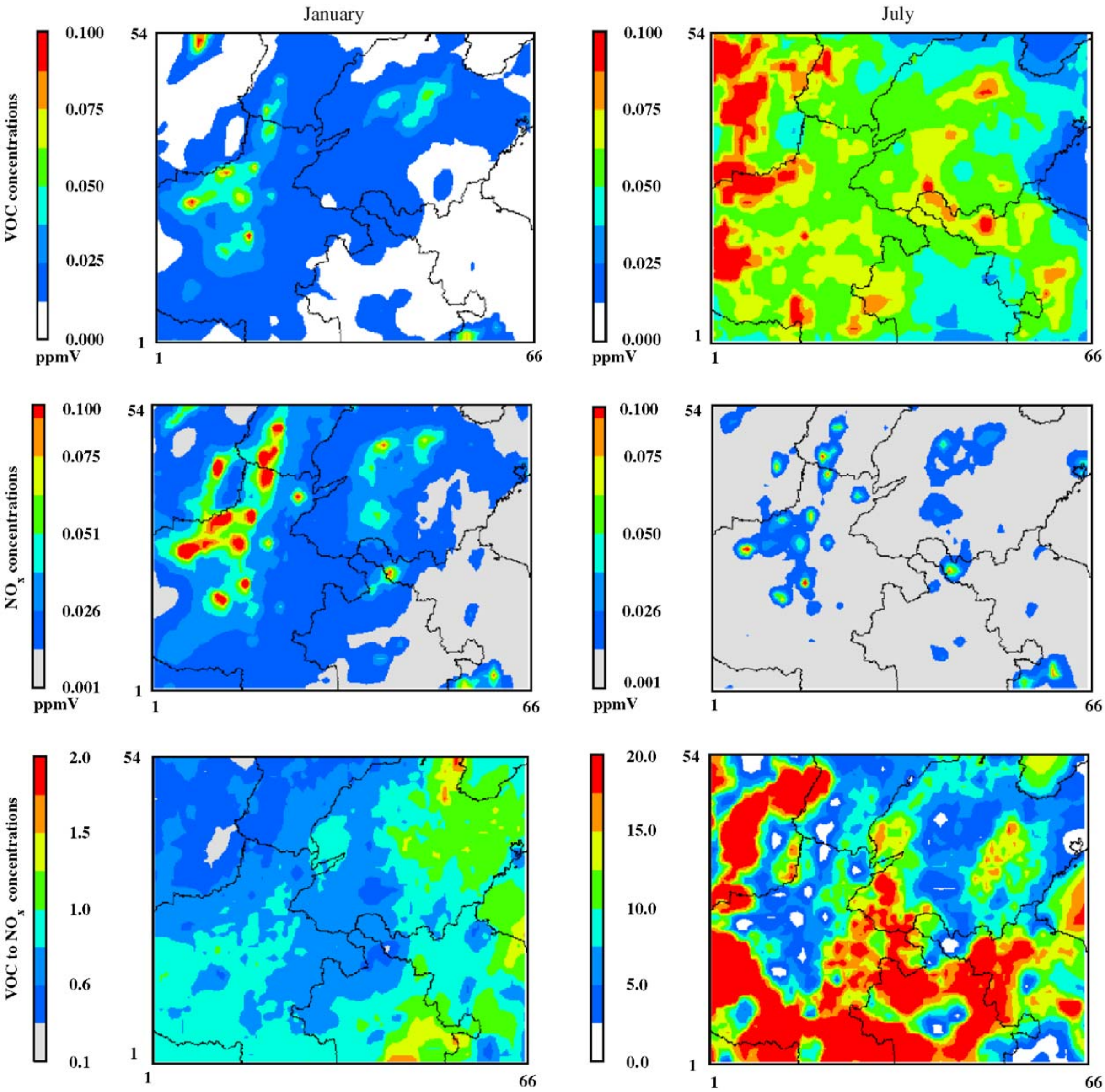

Fig. 9. Average VOC and NO$_x$ concentrations and VOC to NO$_x$ concentration ratios in January and July 2000. Note that the scale for the VOC to NO$_x$ concentration ratio is 10 times higher for July than January.

simulated surface concentrations based on this inventory with observations, we are more likely to underestimate the actual emissions than to overestimate them.

The uncertainties of biogenic VOC emissions are associated with each of the three components of biogenic emission models: emission factors, source densities and emission activity algorithms. Biogenic VOC emission factors and emission activity algorithms for vegetation in China are similar to those observed in other regions (e.g., Xiaoshan et al., 2000, Klinger et al., 2002, Loreto et al., 2002). The major uncertainty with China's biogenic VOC emission estimates is associated with land cover distributions and the process of assigning emission factors. Our estimates tend to be within 35% of the emissions estimated by Klinger et al. (2002) and Guenther et al. (1995). For a region around Beijing that falls within our domain, Wang et al. (2003) estimates emissions for isoprene and monoterpenes that are approximately 50% and for other VOC 80% lower than our estimates. The difference appears to be primarily associated with their lower estimates of tree cover. While earlier tree coverage estimates (e.g. Guenther et al. (1995)) were based on generic global values that were highly uncertain for specific regions, our estimates are based on satellite derived estimates of tree coverage and type with 1 km resolution (DeFries et al., 2000).

A significant conclusion of this paper that O_3 will decrease from 2000 to 2020 is based on the assumption of constant biogenic VOC emissions. It is likely that biogenic VOC will increase from 2000 to 2020 due to reforestation and tree plantation in China (Shi et al., 1997). Any warming possibly occurring during this period will further increase biogenic VOC emissions. Higher biogenic VOC emissions would potentially increase O_3 concentrations.

5. Conclusions

We have developed a high-resolution emission inventory for the Shandong region of China. Our emission estimates for year 2000 are higher than estimates from other studies for most pollutants largely because we have accounted for rural coal consumption, which was significant but was usually missing or underestimated in the data published by China's National Bureau of Statistics. We evaluate our 2000 inventory by comparing observed PM_{10} and SO_2 concentrations with simulated concentrations from MM5/SMOKE/CMAQ using this inventory as input. The model and inventory together under predict observations, partly due to missing sources such as fugitive dust and construction, partly due to the significant uncertainty associated with the government statistics in China on which we rely, and partly due to conservative assumptions we make to fill in missing data.

Our CMAQ simulations using the 2000 and 2020BAU inventories show that the 2020 BAU $PM_{2.5}$ concentrations are projected to be much higher than in 2000 in all four seasons, but O_3 concentrations in 2020 BAU are projected to be lower than in 2000 in most areas because of projected increases in NO_x emissions and the dominate VOC-limited O_3 production regime.

Taking Zaozhuang Municipality of China as a case study, we find with no additional pollution controls implemented between 2000 and 2020, emissions of NH_3 are projected to increase by 20%, NMVOC by 50%, and all other species by 130–250%. Emissions from the residential sector are projected to grow fastest due to the highest demand growth occurring in the residential energy sector. Both BACT and ACGT would contribute to reducing emissions, but with just 24% of the market share of the final energy service demand in Zaozhuang ACGT would reduce emissions to levels similar to 2000—a larger reduction than we project will occur with BACT. In addition, ACGT can use high-sulfur coal (which is abundant in the region) and offers an opportunity to reduce CO_2 emissions by potentially capturing and sequestering them underground. We will examine the implications of the 2020 BAU, BACT and ACGT scenarios for public health, air pollution and energy policies in China in Wang and Mauzerall (2005).

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Appendix A. Supplementary Materials

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.atmosenv.2005.06.051.

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