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A highly resolved temporal and spatial air pollutant emission inventory for the Pearl River Delta region, China and its uncertainty assessment

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ABSTRACT

A highly resolved temporal and spatial Pearl River Delta (PRD) regional emission inventory for the year 2006 was developed with the use of best available domestic emission factors and activity data. The inventory covers major emission sources in the region and a bottom–up approach was adopted to compile the inventory for those sources where possible. The results show that the estimates for SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$ and VOC emissions in the PRD region for the year 2006 are 711.4 kt, 891.9 kt, 3840.6 kt, 418.4 kt, 204.6 kt, and 1180.1 kt, respectively. About 91.4% of SO_2 emissions were from power plant and industrial sources, and 87.2% of NO_x emissions were from power plant and mobile sources. The industrial, mobile and power plant sources are major contributors to PM_{10} and $PM_{2.5}$ emissions, accounting for 97.7% of the total PM_{10} and 97.2% of $PM_{2.5}$ emissions, respectively. Mobile, biogenic and VOC product-related sources are responsible for 90.5% of the total VOC emissions. The emissions are spatially allocated onto grid cells with a resolution of 3 km × 3 km, showing that anthropogenic air pollutant emissions are mainly distributed over PRD central-southern city cluster areas. The preliminary temporal profiles were established for the power plant, industrial and on-road mobile sources. There is relatively low uncertainty in SO_2 emission estimates with a range of −16% to +21% from power plant sources, medium to high uncertainty for the NO_x emissions, and high uncertainties in the VOC, $PM_{2.5}$, PM_{10} and CO emissions.

1. Introduction

The recent two decades' rapid economic development has already led to complex and regional air pollution problems such as acid rain, photochemical smog and haze over the Pearl River Delta (PRD) region of China (e.g., Wang et al., 2003, 2005a; Zhang et al., 2008a). Air quality monitoring data indicates that the highest ozone concentration observed is 0.45 mg m^{-3} and the average number of haze days is over 100 in the region (GDEMC and HKEPD, 2005–2008; Wu et al., 2007). Policy-makers and researchers are facing severe challenges in forming effective air quality management strategies in order to address the increasing air pollution issues in the region.

Air pollutant emission inventories are fundamental information for understanding regional air pollution formation and transport, guiding regional air quality management, and for air quality forecasting (Frey et al., 1999; Streets et al., 2003). Emission inventories with highly resolved temporal and spatial information are urgently needed in China in order to combat the increasing urban and regional air pollutions (Zhang, 2005). In recent decades, several air pollutant emission inventories have been established in China at different scales. These inventories covered areas as large as the national levels (Kilmont et al., 2002; Streets et al., 2003; Zhang et al., 2007; Wei et al., 2008) and down to regional (Wang et al., 2005b) and urban scales (Zhang et al., 2008b). However, most of these inventories were either developed using a top–down approach or not temporally and spatially resolved, or resolved with low resolutions. Furthermore, uncertainty in these inventories was either not discussed or assessed in a qualitative way. In recent decade, the quantitative approaches for characterizing uncertainty in emission inventories have been widely recommended by governmental agencies and academic communities in North America and Europe (NARSTO, 2005; NRC, 2000) and have been applied in real-world emission inventory development, since it can provide quantitative information to guide how future emission inventories are improved (Frey et al., 1999; Zheng, 2002).

The PRD region was one of the earliest regions in China to start compiling regional air pollutant emission inventories (EIs) including EIs for the base years of 1997, 2001 and 2003. The latest 2003-based EIs were jointly developed by the Guangdong Provincial Environmental Monitoring Center (GDEMC) and the Hong Kong Environmental Protection Department (HKEPD), and were publicly released in January 2008 (HG-JWGSDEP, 2008). However, these emission

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inventories were annual emission estimates on a city scale developed mostly with the use of foreign emission factors and missed some important emission sources (e.g., biogenic sources, biomass burning). No spatial and temporal emission variation information are available and thus they cannot be used for the inputs to air quality models such as the CMAQ or CAMx models to assess regional air pollution strategies and to forecast ambient air quality (Byun and Ching, 1999; ENVIRON, 2003). In addition, the PRD region is among the fastest developing areas in China or even in the world, and pollutant emission characteristics change substantially over the years. Therefore, a timely updated emission inventory with temporal and spatial variations is critically important for this region.

The main objectives of this paper are to develop a highly resolved temporal and spatial PRD regional emission inventory with a base year of 2006 and to assess associated uncertainty using a quantitative approach where possible. The pollutant types estimated include SO_2, NO_x, VOC, CO, PM_{10} and $PM_{2.5}$, and local or domestic emission factors were given high priority. High resolutions of population census, road network, land cover data, local traffic flow, and temporal information were used in this study to help establish 3 km × 3 km gridded emission inventories and emission temporal profiles.

Section 2 of this paper describes the data sources and associated methods used in this study. The PRD regional emission inventory, spatial and temporal emission characteristics, and the assessment of associated uncertainty are presented and discussed in Section 3. Findings and recommendations are given in Section 4.

2. Data and methodology

2.1. Study domain and source categorization

The study domain under the Lambert conformal projection for the PRD regional emission inventory is set between the latitudes of 21°27′47″ N and 23°56′13″ N, and between the longitudes of 111°59′52″ E and 115°24′48″ E. The domain is divided into 10 825 grid cells with 3 km × 3 km spatial resolution, and covers nine cities located in the PRD region as shown in Fig. 1 (exclusive of Hong Kong and Macau).

Similar to the source categorization methods used in the Guangdong-Hong Kong 2003-based air pollutant emission inventory (hereafter referred to as "HK-GD 2003-based EI" in this paper) (HG-JWGSDEP, 2008), the emission sources in this study were divided into six major categories including power plant, industry, mobile sources, VOC product-related sources, biogenic sources and others, as shown in Table 1.

2.2. Development of PRD regional emission inventory

A bottom–up approach was adopted in this study for those categories where possible, together with the use of a top–down approach for other categories. In the bottom–up approach, one estimates emissions for individual sources and sums all sources to obtain city, regional or country level estimates, which are typically used in point sources and generally require more information on emission sources. The top–down approach is typically used for area sources and other categories where city or regional level-statistical data are generally used (Francois et al., 2005).

The methods, activity data sources and emission factors used for compiling the regional emission inventory are briefly introduced below, category by category:

- Power plant and industrial sources

The detailed activity data of power plant and industrial sources are collected from the Guangdong provincial pollutant statistical reports. Information for each plant includes location (latitude and

Fig. 1. The PRD region and its location.

Table 1
Emission source categorization in the PRD region.

Category	Sub-category	Category	Sub-category
Power plant		Industry	Pulp and paper industries
Industry	Alcoholic beverage production	Mobile sources	On-road mobile sources
	Chemicals/rubber/plastic		Marine
	Oil refinery		Agricultural machinery
	Petrol distribution & handling		Airport
	Printing	VOC products-related	Personal domestic product
	Electronic manufacture		Paint
	Food and beverage		Coating
	Manufacture – light	Biogenic sources	
	Manufacture – heavy	Others	Residential fuel
	Nonmetallic mineral products		Waste incineration
	Mining/mineral extraction		Biomass burning

longitude), fuel type, fuel consumption, sulfur content, boiler types and capacity, stack parameters, control devices, removal efficiency, product output and others. A bottom–up approach was used to calculate emissions for the two categories. Emissions are estimated using either mass balance or emission factor methods, depending on the type of pollutants. For SO_2 emissions, the annual inventory was compiled using the mass balance method by the following Equation (1):

$$E = \sum_{i,k} C_k A_{i,k} S_{i,k} (1 - \eta_i) \quad (1)$$

Where, i represents the ith plant; k represents fuel type; E is annual total emission of SO_2; C is the fuel-based coefficient, where $C = 16$ for coal and $C = 20$ for oil; A is the activity data (e.g., annual fuel consumption); S is the sulfur content of fuel; η is the removal efficiency.

For NO_x, CO, PM_{10}, $PM_{2.5}$ and VOC, emissions from fuel combustion or manufacturing process were calculated by using Equation (2):

$$E_p = \sum_{i,k,l,m} A_{i,k,l} \times X_{i,k,l,m} \times EF_{k,l,m} \quad (2)$$

Where, p, i, k, l, m represent the pollutant type, the ith plant, fuel type, source category, technology type, respectively; E is annual emissions of pollutant p; A is the activity level (e.g., fuel consumption or product output); X is the application rate of technology m at the l source category; EF is the average emission factor.

In order to best reflect current emission levels of air pollutants in the PRD region, the emission factors for these two categories are chosen mainly from the latest research results based upon local or domestic measurements where possible, and these emission factors are given in Tables 2 and 3. Although these emission factors seem to be generally higher than those measured in Europe and America, they might be better representatives of emission levels under the current processes and control levels in China.

- On-road mobile sources

A modified MOBILE 5b model with the use of domestic base emission rates, vehicle emission standards, local ambient conditions and other local data is adopted in this study to estimate fleet-based vehicle emission factors for the year 2006 in the PRD region. The MOBILE model, developed by USEPA, has the ability to estimate current and future emissions from on-road motor vehicles with series models dating back to 1978 (USEPA, 2003). In this study, the MOBILE 5b was chosen because its motor vehicle classification is more similar to that used in China. MOBILE 5b has already been used for estimating mobile source emissions in Beijing and Shenzhen (Hao et al., 2001; SESRI, 2007). Seven vehicle classes were used in this study including light duty gasoline vehicle (LDGV), light duty gasoline truck (LDGT), light duty diesel truck (LDDT), heavy duty diesel vehicle (HDDV), heavy duty gasoline vehicle (HDGV), heavy duty diesel truck (HDDT), and motorcycle (MC). Details for estimating PRD regional on-road mobile source emissions are referred to Che et al. (2009).

Table 2
NO_x, PM_{10}, $PM_{2.5}$, CO and VOC emission factors used in power plant category.

Pollutant	Fuel type	Technology type	Emission factor	Reference
NO_x	Coal	<100 MW, without LNB	10.5 g kg^{-1}	Zhang et al. (2007)
		≥100 MW, but <300 MW, without LNB	8.85 g kg^{-1}	Zhang et al. (2007)
		≥100 MW, but <300 MW, with LNB	5.85 g kg^{-1}	Zhang et al. (2007)
		≥300 MW, with LNB	5.55 g kg^{-1}	Zhang et al. (2007)
	Fuel oil		10.06 g kg^{-1}	Tian (2003)
	Diesel		7.4 g kg^{-1}	Tian (2003)
	Natural gas		1.76 g m^{-3}	Zhao and Ma (2008)
$PM_{10}/PM_{2.5}$	Coal	Pulverized coal boiler	46/12 g kg^{-1}	Zhang (2005)
		Stoker furnace	13.88/5.25 g kg^{-1}	Zhang (2005)
	Fuel oil		0.85/0.62 g kg^{-1}	Zhang (2005)
	Diesel		0.5/0.5 g kg^{-1}	Zhang (2005)
	Natural gas		0.24/0.17 g m^{-3}	Zhao and Ma (2008)
CO	Coal	Pulverized coal boiler	2 g kg^{-1}	Wang et al. (2005c)
		Stoker furnace	8 g kg^{-1}	Wang et al. (2005c)
	Oil		0.6 g kg^{-1}	Wang et al. (2005c)
	Natural gas		1.3 g m^{-3}	Wang et al. (2005c)
VOC	Coal		0.15 g kg^{-1}	Bo et al. (2008)
	Oil		0.13 g kg^{-1}	Bo et al. (2008)
	Natural gas		0.18 g m^{-3}	Bo et al. (2008)

Table 3
NO_x, PM_{10}, $PM_{2.5}$, CO and VOC emission factors used in industry sources.

Pollutant	Activity	Technology type	Emission factor	Reference
NO_x	Coal	Fluidized-bed furnace	7.5 g kg^{-1}	Zhang et al. (2007)
		Automatic stoker	4.0 g kg^{-1}	Zhang et al. (2007)
	Fuel oil		5.84 g kg^{-1}	Tian (2003)
	Diesel		9.62 g kg^{-1}	Tian (2003)
	Natural gas		1.76 g m^{-3}	Zhao and Ma (2008)
	Coal/cement industry	Shaft kiln	1.7 g kg^{-1}	Zhang et al. (2007)
		New-dry process	15.3 g kg^{-1}	Zhang et al. (2007)
		Other rotary kiln	18.5 g kg^{-1}	Zhang et al. (2007)
	Synthetic ammonia		0.9 g kg^{-1}	Zhao and Ma (2008)
$PM_{10}/PM_{2.5}$	Coal	Fluidized-bed furnace	28.08/5.4 g kg^{-1}	Zhang (2005)
		Stoker furnace	5.4/1.89 g kg^{-1}	Zhang (2005)
	Fuel oil/Industry		1.03/0.67 g kg^{-1}	Zhang (2005)
	Diesel		0.5/0.5 g kg^{-1}	Zhang (2005)
	Gas		0.24/0.17 g m^{-3}	Zhao and Ma (2008)
	Cement production		54.6/23.4 g kg^{-1}	Zhang (2005)
	Iron making		0.24/0.15 g kg^{-1}	Zhang (2005)
	Steel making	Converter	23.45/22.7 g kg^{-1}	Zhang (2005)
CO	Coal/industry	Fluidized-bed furnace	2 g kg^{-1}	Wang et al. (2005c)
		Automatic stoker	15 g kg^{-1}	Wang et al. (2005c)
		Hand-feed stoker	124 g kg^{-1}	Wang et al. (2005c)
	Oil		0.6 g kg^{-1}	Wang et al. (2005c)
	Gas		1.3 g m^{-3}	Wang et al. (2005c)
	Cement production	Shaft kiln	155.7 g kg^{-1}	Wang et al. (2005c)
		Rotary kiln	17.8 g kg^{-1}	Wang et al. (2005c)
VOC	Coal		0.18 g kg^{-1}	Bo et al. (2008)
	Fuel oil		0.15 g kg^{-1}	Bo et al. (2008)
	Natural gas		0.18 g m^{-3}	Bo et al. (2008)
	Liquor producing		25.35 g kg^{-1}	Wang (2006)
	Beer producing		0.44 g kg^{-1}	Wang (2006)
	Paint/vehicle coating		475 g kg^{-1}	Wang (2006)
	Paint/others		730 g kg^{-1}	Wang (2006)
	Ink use	Gravure printing	800 g kg^{-1}	Wei et al. (2008)
		Relief printing	850 g kg^{-1}	Wei et al. (2008)
	Crude oil refining		2.34 kg m^{-3}	Wang (2006)

The vehicle population data, vehicle kilometers traveled (VKT), base emission rate measurements and other associated data and information used in this study were collected from Guangdong governmental annual statistical reports (National Bureau of Statistical of China, 2007a,b), field surveys conducted in the PRD region, and previous studies (Che et al., 2009; Ma, 2007).

- Non-road mobile sources

The non-road mobile sources considered in this study included marine, agricultural machinery, and airport sources. The associated activity data for these sources are collected from the 2007 urban statistical books (National Bureau of Statistical of China, 2007a,b).The fuel-based top–down approach was used for estimating emissions from these sources except airport source. The emission factors for these sources are referred to Fu et al. (2005).

A full Landing and Take-off (LTO) cycle was used as activity data for estimating emissions from airport plane sources. A standard LTO cycle consists of take-off, climb-out, approach-landing and taxi. The detailed methods and emission factors for estimating airport emissions used in this study are given in the handbook released by Guangdong and Hong Kong governments (HG-JWGSDEP, 2005).

- Biogenic sources

The Global Biosphere Emissions and Interactions System (GloBEIS) model was used in this study to estimate the VOC emissions from the biogenic sources in the PRD region. The GloBEIS was developed at the National Center for Atmospheric Research (NCAR) to allow users to estimate biogenic emissions of VOC, carbon monoxide, and soil NO_x over any scales and domain. The data sources used in this study include the remote-sensing image interpretation data of PRD land use and vegetation for the year 2006 and observed hourly meteorological data during 1/1/2006–12/31/2006. The details and determination of model input parameters for estimating biogenic VOC emissions can be found in Zheng et al. (2009a).

- VOC product-related sources

The VOC product-related sources considered in this study mainly include paint, coating and personal domestic products (e.g., detergents, shampoos). The VOC emissions from these sources are generated when these products are used. For paints and coatings, the current governmental statistical books only record the yields from manufacturers in the PRD region, and the city-based consumption data for these products are not available except for in the city of Guangzhou (Guangzhou Statistics Bureau, 2007). In this study, the yield and consumption information of paints and coatings in Guangzhou were used as basis to infer the amounts of these products consumed in other cities of the PRD region. A top–down approach with the use of "emission factor methods" was adopted to estimate the annual VOC emissions from these sources. For personal domestic products, population data including immigrant workers in the PRD region were used as activity data to estimate VOC emissions. The emission factors used for the sources are given in Table 4.

- Other sources

The other sources considered in this study include residential fuel consumption, waste burning, and biomass burning. The

Table 4
NO_x, PM_{10}, $PM_{2.5}$, CO and VOC emission factors used in VOC product-related category and other sources.

Pollutant	Activity	Emission factor	Reference
NO_x	Coal/residential	1.88 g kg^{-1}	Tian (2003)
	Natural gas/residential	1.76 g m^{-3}	Zhao and Ma (2008)
	Coal gas/residential	0.8 g m^{-3}	Zhao and Ma (2008)
	LPG/residential	2.1 g kg^{-1}	Zhao and Ma (2008)
	Waste	2.5 g kg^{-1}	HG-JWGSDEP, 2005
	Crop straw	2.5 g kg^{-1}	Wang et al. (2008)
$PM_{10}/PM_{2.5}$	Coal/residential	1.62/0.77 g kg^{-1}	Zhao and Ma (2008)
	Gas/residential	0.24/0.17 g m^{-3}	Zhao and Ma (2008)
	LPG/residential	0.22/0.15 g kg^{-1}	Zhao and Ma (2008)
	Waste	0.35/0.15 g kg^{-1}	HG-JWGSDEP, 2005
	Crop straw	5.77/2.5 g kg^{-1}	Wang et al. (2008)
CO	Coal/residential	52.3 g kg^{-1}	Zhao and Ma (2008)
	Gas/residential	1.3 g m^{-3}	Wang et al. (2005c)
	LPG/residential	0.36 g kg^{-1}	Wang et al. (2005c)
	Crop straw	56.4 g kg^{-1}	Wang et al. (2008)
	Waste	5 g kg^{-1}	HG-JWGSDEP, 2005
VOC	Coal/residential	0.6 g kg^{-1}	Bo et al. (2008)
	Coal gas/residential	0.088 g m^{-3}	Zhao and Ma (2008)
	LPG/residential	66 g m^{-3}	Bo et al. (2008)
	Natural gas/residential	0.18 g m^{-3}	Bo et al. (2008)
	Waste	0.02 g kg^{-1}	HG-JWGSDEP, 2005
	Crop straw	15.7 g kg^{-1}	Wang et al. (2008)
	Paint	730 g kg^{-1}	Wang (2006)
	Coating/construction use	360 g kg^{-1}	Wang (2006)
	Domestic solvent use	0.5 kg $people^{-1}$	Wang (2006)

activity data for these sources are collected from annual statistical books (National Bureau of Statistical of China, 2007a,b). The emission factors for these sources are given in Table 4.

2.3. Methods for spatial allocation of PRD regional emission inventory

The choice of method for spatially allocating regional emission inventories into grid cells depends on the source characteristics and resolution size. In this paper, different spatial allocation methods were used based on source characteristics. For power plant and large industrial sources (chimney height higher than 30 m), emissions from these sources are directly allocated into grid cells where the plants are located based on its latitude and longitude information.

The emissions from small industrial plants, VOC product-related sources and other emission sources except biomass burning are treated as area sources for spatial allocation in this study. The spatial surrogates used for these area sources are typically population density, GDP and others (Zhang, 2005). In this study, the 30″ × 30″ resolution grid cell-based population densities from 2006 LandScan Asia Population were used as surrogates to aid in the spatial allocation of area sources (ORNL, 2007). This approach has been widely used in spatial allocation of regional or national emission inventories, especially for area source emissions (Zhang, 2005; Streets et al., 2003). The detailed description for the allocation process is referred to Zheng et al. (2009b).

A new approach using GIS (Geographic Information System)-based road network information and road types-based traffic flows as spatial surrogates was adopted for allocating the PRD regional on-road mobile source emissions. This approach has been shown to provide more reasonable spatial allocations of regional mobile source emissions, especially at higher resolutions (Zheng et al., 2009c). Detailed procedures for allocating on-road mobile sources can be found in Zheng et al. (2009c).

The spatial allocation of biogenic sources is derived directly from the GloBEIS model outputs because the GloBEIS model can provide gridded estimates of emissions from biogenic sources. The spatial allocations of other sources such as biomass burning are conducted based upon land use and associated surrogate data. With the gridded emissions from individual sources, gridded PRD regional emission inventories including all emission sources considered in this study were established by using GIS technology (Zhang et al., 2008b).

2.4. Methods for uncertainty analysis

There are various approaches for characterizing uncertainty in emission inventories including qualitative, semi-quantitative and quantitative approaches (NARSTO, 2005). In this paper, both semi-quantitative and quantitative approaches were used depending on the data availability. For those source categories or pollutants in which there are enough data available, the quantitative approach was used. The quantitative approach features the use of bootstrap simulation and expert elicitation for quantifying uncertainty in emission factors or activity data, and the use of Monte Carlo simulation for propagating uncertainty (Zheng, 2002; Frey et al., 1999). For those categories or pollutants in which quantitative uncertainty analysis cannot be conducted, the uncertainty ranges of emission estimates were judged by assessing the reliability and accuracy of data sources, estimation methods used, and uncertainty in emission factors.

3. Results and discussion

3.1. PRD 2006 regional emission inventory

The source-specific emission inventories of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$ and VOC in the PRD region for the year 2006 are summarized in Table 5. The total emissions of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$ and VOC were 711.4 kt, 891.9 kt, 3840.6 kt, 418.4 kt, 204.6 kt, and 1180.1 kt, respectively. Fig. 2 shows the pollutant-based contributions of major source categories to the total emissions in the PRD region. Power plant share the largest contribution of SO_2, about 50.6% of the total emission for the regional SO_2 emissions. The power plant source is also a major source of NO_x, PM_{10} and $PM_{2.5}$ emissions, and responsible for 41.3%, 21.3% and 18.7% of total emissions, respectively; while its contributions to the CO and VOC emission are only 2.9% and 0.6%. The mobile sources (including on-road and non-road sources) are the largest categories for NO_x, CO and VOC emissions,

Table 5
Emission inventory in the PRD region for the year of 2006 (unit: kilo-ton).

Category or sub-category	SO_2	NO_x	CO	PM_{10}	$PM_{2.5}$	VOC
Power plant	360.3	368.6	111.1	89.1	38.2	7.2
Industry	289.6	104.1	1041.5	228.9	88.2	85.6
Mobile sources						
On-road mobile sources	37.7	321.9	2582.3	92.5	74.2	464.7
Marine	19.6	94.0	17.5	4.6	3.7	9.3
Agricultural machinery		0.5	0.3			0.1
Airport	0.3	4.7	2.6			0.3
VOC products-related						
Personal domestic product	–	–	–	–	–	21.9
Paint	–	–	–	–	–	182.5
Coating	–	–	–	–	–	88.4
Biogenic sources	–	–	–	–	–	295.8
Others						
Residential fuel	4.9	6.8	15.0	2.4	2.5	0.1
Waste incineration	0.1	0.1	0.3	0.2	0.1	
Biomass burning	0.5	3.1	70.0	7.2	3.1	19.5
Total	711.4	891.9	3840.6	418.4	204.6	1180.1

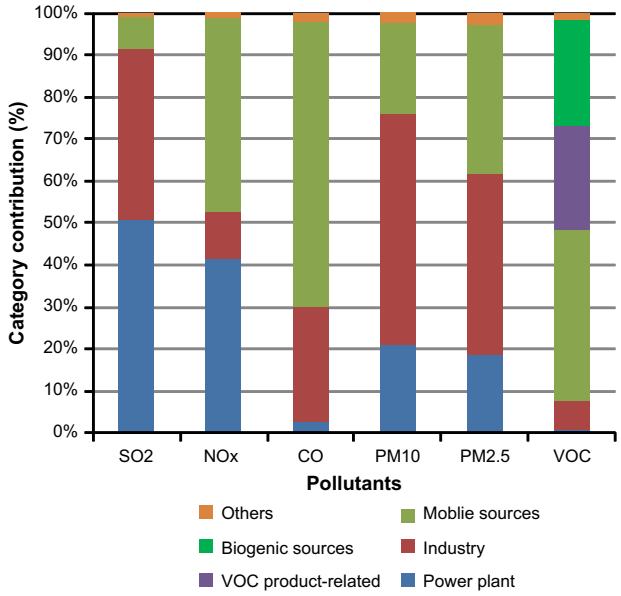

Fig. 2. Emission contributions by categories in the PRD region for the year of 2006.

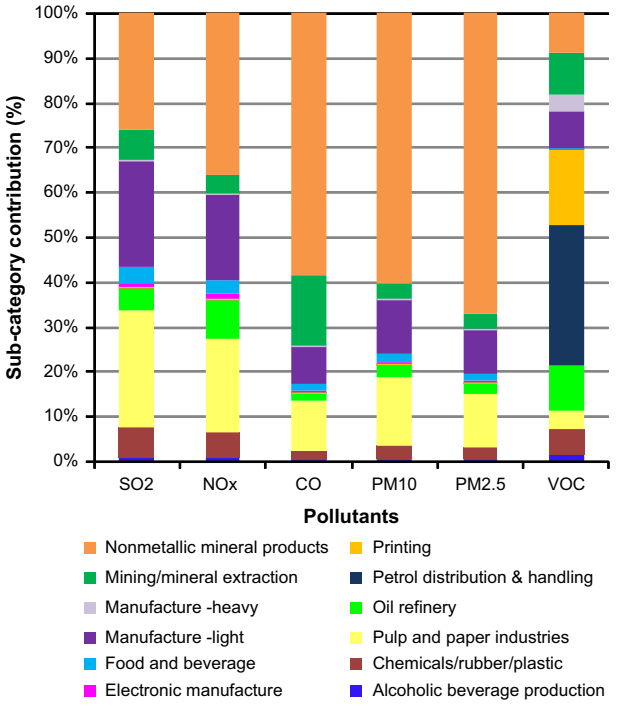

Fig. 3. Contributions by the sub-categories to the total emissions under industrial source in the PRD region.

accounting for 45.9%, 67.8%, and 40.6% of the total NO_x, VOC and CO emissions, respectively. The PM_{10} and $PM_{2.5}$ emissions from mobile sources are also very high, about 21.7% and 35.5% of the total PM_{10} and $PM_{2.5}$ emissions. The industrial categories are the highest emission sources of PM_{10} and $PM_{2.5}$, and account for 54.7% and 43.1% of the total emissions, while their contributions to SO_2, NO_x, CO and VOC emissions are 40.7%, 11.7%, 27.1%, and 7.3%, respectively. For the VOC emissions, it is worthwhile to note that the biogenic and VOC product-related sources are important VOC sources, accounting for 25.1% and 24.8% of the total VOC emissions in the region, respectively. Less than 3% of the total SO_2, NO_x, CO, PM_{10} and $PM_{2.5}$ and VOC emissions are contributed by other sources.

Fig. 3 presents the contributions of different pollutants from the 12 sub-sectors under the industrial category. The nonmetallic mineral products sector is a major contributor, especially for the PM_{10} and $PM_{2.5}$ pollutants. The SO_2, NO_x, CO, PM_{10} and $PM_{2.5}$ emissions from this sector are about 26.3%, 36.3%, 58.5%, 60.3% and 67.1% of the total industrial emissions, respectively. Pulp and paper, and light-manufacturing sectors are also important emission emitters within the industrial category, contributing about 25.8% and 23.9% of total SO_2 emissions, 21.0% and 19.3% of NO_x emissions, 11.1% and 8.8% of CO emissions, 15.3% and 12.3% of PM_{10} emissions, and 12.0% and 9.8% of total $PM_{2.5}$ emissions from the industrial category, respectively. For the VOC emissions, most are emitted from industrial production processes, solvent usage-related manufacturers, and transportation and storage industries. For example, petrol transportation & storage, printing, and oil refinery industries account for about 31.1%, 17.1% and 10.2% of the total VOC emissions from the industrial category, respectively.

A preliminary comparison of emission estimates from anthropogenic sources was made here for the inventory developed in this study to other similar inventories including the HK-GD 2003-based EI (JWGSDEP, 2008) and 2006 INTREX-B by Zhang and Streets (2008). Due to using similar source categorization, a relatively detailed comparison was made to the HK-GD 2003-based EI. Overall, considering inherent uncertainties in compiling emission inventories, the estimates from the two inventories are comparable with the exception of some categories. For example, estimates of SO_2 emissions are in good agreement except that the estimate for the power plant category (360.3 kt) in this study is lower than that from the HK-GD 2003-based EI (435.4 kt). This is probably reasonable because the installation of desulphurization measures has been required for power plants in the PRD region starting since 2003. While for NO_x emissions, the estimate from this study for the power plant category (368.6 kt) is much higher than the one from the HK-GD 2003-based EI (144 kt). This discrepancy might have arisen from the use of different estimation methods and different emission factors in the two inventories. In this study, the state-of-science domestic emission factors were used. Also, the estimate of NO_x emissions in this study for the sector are very comparable with the estimate (397.4 kt) from 2006 INTEX-B emission inventory (Zhang and Streets, 2008).

The PM_{10} emissions in the two inventories agree well except for the industry category in which the estimate (228.9 kt) from this study is lower than the one (352.6 kt) by the HK-GD 2003-based EI (HG-JWGSDEP, 2008). One possible reason is that enhancement of dust removal measures have been mandated by governments for major industrial sources in recent years, especially for the nonmetallic mineral products industry. The anthropogenic VOC emission estimates in this study are much higher than those from the HK-GD 2003-based EI (HG-JWGSDEP, 2008) especially for mobile and VOC product-related source categories, which are major contributors to the VOC emissions in the PRD region. The differences are most likely caused by increasing activity data (e.g., vehicle population and paint and coating consumption), the estimation methods, choice of emission factors, and various uncertainties. In this study, we have relatively more reasonable estimates from mobile source emissions and more comprehensive collection of VOC product-related sources activity data than the HK-GD 2003-based EI.

3.2. Spatial characteristics of PRD emissions

The PRD regional emission inventory was spatially allocated to the 3 × 3 km grid cells with the use of the methods introduced in Section 2.3. Fig. 4(a)–(f) shows the 3 km × 3 km spatial distributions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, CO and VOC emissions over the PRD region, respectively. The annually averaged primary pollutant (SO_2, NO_x, and PM_{10}) concentrations from the PRD regional air quality monitoring network (GDEMC-HKEPD, 2007) are shown together in the corresponding figures in order to help illustrate whether or not the spatial distributions are reasonable.

SO_2 emissions are mainly distributed over the grid cells of central and southern PRD areas where large power plants and industrial plants are heavily located (see Fig. 4(a)), indicating that these large point sources are major contributors to SO_2 emissions. A large amount of NO_x emissions in the PRD region are contributed from Guangzhou, Fohsan, Dongguan and Shenzhen areas where there are dense populations, heavy traffic flows and numerous

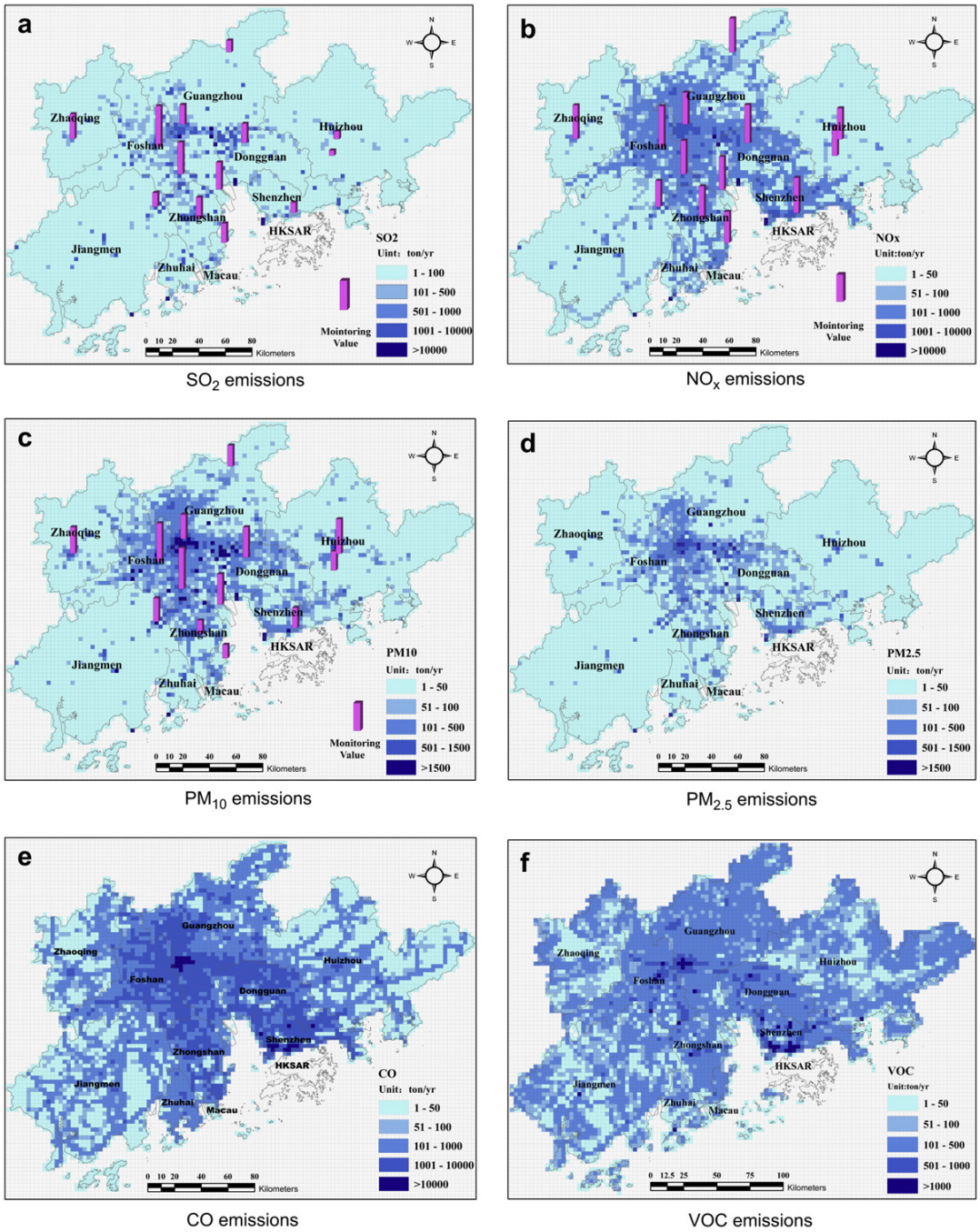

Fig. 4. Spatial distributions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, CO and VOC emissions in PRD region for the year of 2006.

major point sources (see Fig. 4(b)). Foshan, Guangzhou, and Dongguan have larger PM_{10} and $PM_{2.5}$ emissions (see Fig. 4(c) and (d)), since they have high motor vehicle ownership and industrial sources, especially nonmetal mineral plants.

The VOC emissions exhibit a relatively uniform distribution over the PRD region as shown in Fig. 4(f) due to the contributions from biogenic sources in the boundary cities (Huizhou, Jiangmen, and Zhaoqing) of the PRD region. However, the peak of VOC emission areas is still distributed over the central and southern economically developed areas, suggesting that anthropogenic sources are major contributors to the VOC emissions in the region. The spatial distribution of CO emissions is very consistent with the patterns of transportation networks in the PRD region (see Fig. 4(e)), as expected, since mobile sources contribute 67.8% to the CO emissions in the region.

Comparing observed ambient SO_2, NO_x and PM_{10} concentrations from the PRD air quality monitoring network (see Fig. 4(a)–(c)), it was found that there are good agreements between the monitored ambient air pollutant concentrations and spatial distributions of corresponding pollutant emissions. This implies that the spatial allocations of regional emission inventories made in this study are reasonable.

3.3. Temporal characteristics of emissions

Temporal emission variations are investigated for power plant, industrial and on-road mobile sources in this study. Since emissions in power plants are generally proportional to energy consumptions or electricity production, in this study, energy consumptions of typical power plants from Guangdong Electricity Group were collected to reflect the temporal characteristics of emission variations on a monthly basis for the PRD power plant sources (see Fig. 5). Overall, there are relatively higher energy consumptions during July–November and lower consumptions during December–February, energy consumption during April through July is unexpectedly lower than other calendar months. One reason may be that these calendar months are typically within the wet seasons in the PRD region, and another reason may be that the PRD region generally purchases electric power from the western provinces of China in summer. Both reasons reduce the needs for local power generation during this period.

Fig. 6 shows the monthly profiles of emission variations of industrial sub-categories based on the analysis of monthly product yields of different sub-categories. The industrial sector-based monthly product yields are derived from the statistical reports of Guangdong province (Guangdong Provincial Bureau of Statistics, 2007). A common feature is that almost all industrial sectors in 2006 have the lowest monthly yields in February because of Chinese New Year occurring in this month. There are significant differences in the monthly profile of the alcoholic beverage production industry when compared with other industries, it increases gradually from January and reaches a peak in August and then decreases. This profile is reasonable because alcoholic beverages, especially beer, are consumed in vastly larger quantities in summer, and consumed relatively less in the colder months. Also, it is interesting to see that the light-manufacturing industry has higher emissions in January, April, May and December, and lower emissions in March. The Christmas, New Year, Chinese New Year (February in 2006), and Chinese Children's Day (June 1) typically occur in these months. The variations of emissions from the light-manufacturing industry show obvious holiday and seasonal characteristics because it is typically involved in toy and other personal domestic products.

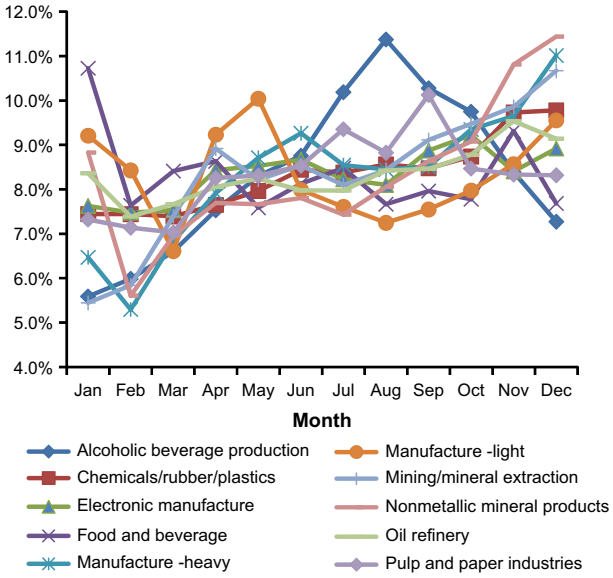

Fig. 6. Monthly variation profiles for industrial sectors.

Fig. 7 illustrates the day-of-week variations of on-road mobile source emissions based on traffic flows in the PRD region. The traffic flow data were collected from the Guangzhou-Shenzhen highway toll stations, field investigations in Guangzhou urban area conducted in this study, and other studies (Che et al., 2009; Zheng et al., 2009c). Basically, there are no obvious variations of traffic flows during the weekdays except that Monday and Friday have slightly higher traffic flows. Significant decreases of traffic flows are observed in the weekend, as expected. Fig. 8 shows diurnal

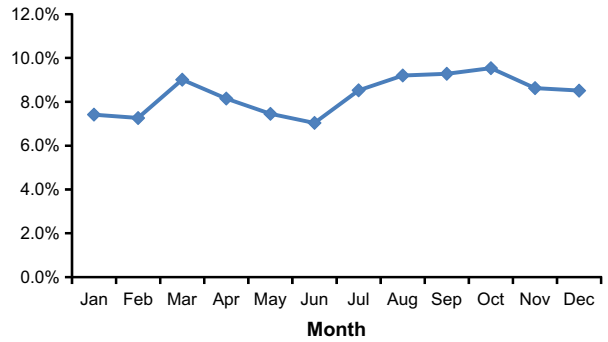

Fig. 5. Monthly variation profile for power plant.

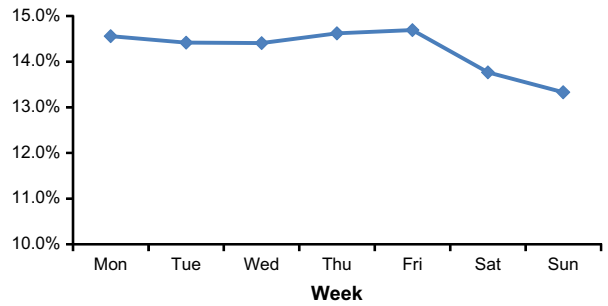

Fig. 7. Day-of-week variation profiles for on-road mobile sources.

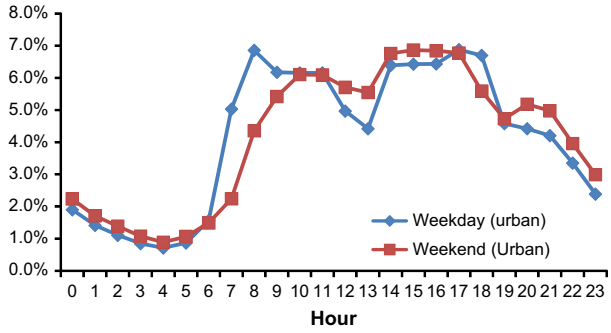

Fig. 8. Dinural variations for on-road mobile sources at urban areas.

variations of traffic flows in the urban areas of the PRD region for both weekdays and weekends. Obvious commuting traffic peaks are observed on the weekdays with a morning peak at 7:00–9:00 and an evening peak at 17:00–18:00. For the weekends, the overall profile is similar to that during the weekdays except that there are no obvious commuting traffic peaks and higher traffic flows in the afternoon. However, both weekdays and weekends demonstrate similar troughs in the traffic flows around the meal times (13:00 for lunch and 19:00 for dinner). Fig. 9 illustrates diurnal variations in traffic flows on typical highways in the PRD region for both weekdays and weekends. Because the highways are typically used for inter-city transportation in the PRD regions, the diurnal profiles during weekdays and weekends are very similar. Also, there are no commuting peaks but an obvious trough of traffic flows at lunch time is observed.

3.4. Uncertainty assessment

A preliminary uncertainty assessment of the 2006 PRD regional emission inventory was made in this study. Table 6 shows the source category-based uncertainty assessment results. As shown in Table 6, due to lack of data support, for most source categories and pollutants, quantitative uncertainty assessment cannot be made.

As shown in Table 6, estimates from power plant categories have less uncertainty compared to other categories due to using a bottom–up approach and having detailed fuel consumption activity data. SO_2 emission estimates from power plant category have the lowest uncertainty, which is about within −16% to +21%. However, for the CO and VOC emissions, there is relatively higher uncertainty compared to other pollutants in the power plant category because less domestic measurements regarding emission factors are available for the two pollutants.

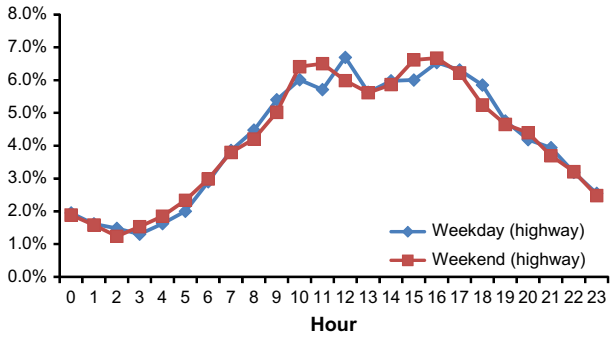

Fig. 9. Dinural variations for on-road mobile sources at highway.

For the industrial source categories, though a bottom–up approach was used for estimating emissions in this study, there are higher uncertainties in emission estimates compared to power plant category because industrial categories involve a large number of manufacturers with various industries and product processes. The uncertainty sources for this category are mainly attributed to lack of detailed information such as product processes and facility parameters for combustion and emission controls, localized emissions factors for some sectors, especially for $PM_{2.5}$, CO, and VOC emission factors, and human or statistical errors in activity data collection.

Uncertainty in the PRD on-road mobile source emission inventory arises not only from lack of local vehicle emission measurements and fleet characteristics, but also from the discrepancy in vehicle classifications between annual statistical reports and the model use. Because the VKT numbers are estimated based upon surveys not available through official channels, it inevitably leads to systematic bias in estimating PRD on-road mobile sources. Therefore, although the emissions from on-road mobile source in this study are estimated using the best available data and knowledge, the overall uncertainty is assessed as high in this study due to various uncertainty sources mentioned above.

Little studies on non-road mobile source emission estimation including emission factor measurements and proper statistics of activity data were done in China at present. Thus, there is higher uncertainty in emission estimates for the category. The VOC product-related source might have the highest uncertainty due to lack of city-based official statistics data regarding the consumption of these products and high uncertainty of emission factors for different coating and painting products. There is also high uncertainty for the biogenic VOC emissions.

Overall, there is low uncertainty for the overall SO_2 inventory in the PRD region. For the NO_x emissions, on-road and non-road mobile sources are major contributors to the uncertainty. Industrial and on-road mobile source categories are key sources of uncertainty for PM_{10} and $PM_{2.5}$ emissions, and biomass burning and other sources have high uncertainty for the $PM_{2.5}$ emissions. The on-road mobile, VOC product-related and biogenic sources contribute the most to the uncertainty in the VOC emissions in the PRD region.

4. Conclusions and recommendations

A highly resolved temporal and spatial PRD regional emission inventory for the year of 2006 was developed with the use of best available data and knowledge in this study. The inventory was prepared for the use in air quality models such as CAMx or CMAQ. The estimates from the inventory show that the total emissions of SO_2, NO_x, CO, PM_{10} and $PM_{2.5}$ and VOC in the PRD region for the year of 2006 were 711.4 kt, 891.9 kt, 3840.6 kt, 418.4 kt, 204.6 kt, and 1180.1 kt, respectively.

The estimates show that about 50.7% of SO_2 emissions in the PRD region came from the power plant, 40.7% from industrial sources, and 8.6% from other sources. 41.3% of NO_x emissions were contributed by the power plant, 45.9% from mobile sources, and 12.8% from others. The industrial, mobile and power plant source categories are also major contributors for PM_{10} and $PM_{2.5}$ emissions, accounting for 54.7%, 21.7%, and 21.3% of the total PM_{10} emissions and 43.1%, 35.5%, and 18.7% of the $PM_{2.5}$ in the PRD region, respectively. For VOC emissions, mobile sources are the largest contributor, responsible for 40.6%, and biogenic and VOC product-related sources account for 25.1% and 24.8% of the total VOC emissions in the region. Mobile and industrial sources shared 67.8% and 27.1% of the emissions of CO, while 5.1% was contributed by other sources.

Table 6
Uncertainty assessment of major source emission estimates in the PRD region.

Source	Uncertainty range (%)					
	SO_2	NO_x	CO	PM_{10}	$PM_{2.5}$	VOC
Power plant	−16% to 21%[a]	−23% to 33%[a]	−30% to 40%	−35% to 40%	−45% to 70%	−60% to 80%
Industry	−21% to 25%[a]	−31% to 47%[a]	−70% to 90%	−50% to 70%	−60% to 80%	−50% to 100%
On-road mobile sources	−50% to 60%	−55% to 70%	−40% to 70%	−60% to 70%	−60% to 70%	−50% to 70%
Marine	−70% to 80%	−40% to 65%	−60% to 100%	−70% to 110%	−75% to 150%	−70% to 120%
Agricultural machinery	NA	−30% to 120%	−70% to 120%	NA	NA	NA
Airport	−28% to 31 %[a]	−27% to 31%[a]	−45% to 47%[a]	NA	NA	−35% to 33%[a]
VOC product-related	NA	NA	NA	NA	NA	−65% to 120%
Biogenic sources	NA	NA	NA	NA	NA	−60% to 100%
Residential fuel	−25% to 35%	−50% to 70%	−70% to 100%	−60% to 70%	−60% to 80%	−50% to 80%
Biomass burning	−70% to 120%	−50% to 150%	−75% to 120%	−70% to 120%		−75% to 150%

[a] Uncertainty ranges are quantitatively characterized on the 95% confidence interval.

The spatial distribution patterns of emissions show that air pollutant emissions in the PRD region are mainly distributed over central-southern city cluster areas. The analysis of temporal variations of emissions shows that there are relatively large emissions from power plant during July–November and lower emissions during December–February, emissions during April through July is unexpectedly lower than other calendar months mainly due to purchasing electricity power from western provinces. The temporal variations from industrial sources exhibit obvious holiday and seasonal characteristics, especially for light-manufacturing and alcoholic beverage industries. Lower traffic flows are found on the weekend, and obvious commuting traffic peaks are found in the urban areas for weekdays, while traffic flow troughs at meal times are observed on both weekends and weekdays. Similar temporal profiles for the highways are found during weekend and weekdays for the highways.

Assessment of uncertainty in the 2006-based PRD emission inventory shows that there is relatively low uncertainty in SO_2 emission estimates; while there is medium to high uncertainty for the NO_x emissions. High uncertainties for the NO_x emissions are mainly caused by on-road and non-road mobile sources. VOC, $PM_{2.5}$, PM_{10} and CO emissions have high uncertainties due to lack of key representative emission factors and large uncertainties in activity data. On-road mobile, VOC product-related and biogenic sources are key contributors to the high uncertainty in VOC emissions; while industrial and on-road mobile sources are main uncertainty sources leading to high uncertainty in PM_{10} and $PM_{2.5}$ emission estimates.

Potential future improvements for the PRD regional source inventory include the enhancement of local emission factor development, proper classification of emission sources, and comprehensive collection of activity data, and temporal and spatial surrogate data. In particular, improving estimates of emissions from mobile sources will greatly increase the accuracy of NO_x, VOC, CO, PM_{10} and $PM_{2.5}$ estimates in the PRD region. In industrial sources, especially for the nonmetallic mineral product industry, improving the estimates for $PM_{2.5}$ and PM_{10} emissions should be prioritized. Enhancement of collection of activity data for the VOC product-related sources will greatly improve VOC emission estimates. Other important sources including fugitive sources, road dusts and biomass burning either not considered or considered to a lesser extent in this study should further be examined. Additionally, an open data sharing policy among different parties would be another important step to improving the PRD regional emission inventory.

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