



Emission inventory of primary pollutants and chemical speciation in 2010 for the Yangtze River Delta region, China



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HIGHLIGHTS

- An updated high-resolution emission inventory of 2010 for YRD is established.
- Facility-based emissions are calculated for large point sources.
- PM_{2.5} speciation database is established based on Chinese local data.
- Emissions of NMVOC species are estimated based on Chinese local data.

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ABSTRACT

We developed a high-resolution emission inventory of primary air pollutants for Yangtze River Delta (YRD) region, which included Shanghai plus 24 cities in the provinces of Jiangsu and Zhejiang. The emissions of SO₂, NO_x, PM₁₀, PM_{2.5}, NMVOCs and NH₃ in the year of 2010 were estimated as 2147 kt, 2776 kt, 1006 kt, 643 kt, 3822 kt and 1439 kt, respectively. Power plants are the largest emission sources for SO₂ and NO_x, which contributes 44.1% and 37.3% of total SO₂ and NO_x emissions. Emissions from industrial process accounted for 26.9%, 28.9% and 33.7% of the total PM₁₀, PM_{2.5} and NMVOCs respectively. Besides, 37.3% of NMVOCs emissions were contributed by solvent use. Livestock and fertilizer application contribute over 90% of NH₃ emissions. High emission densities are visible in Shanghai and the area around Tai Lake. This emission inventory includes the speciation of PM_{2.5} for the YRD region for the first time, which is important to source apportionment and secondary-pollution analysis. In 2010, emissions of three major PM_{2.5} species, namely OC, EC and sulfate, are 136.9 kt, 75.0 kt and 76.2 kt, respectively. Aromatics and alkanes are the main NMVOC species, accounting for 30.4% and 20.3% of total VOCs. Non-road transportation and biomass burning were main uncertain sources because of a lack of proper activity and emission factor data. Compared with other pollutants, NMVOCs and NH₃ have higher uncertainty. From 2000 to 2010, emissions of all pollutants have changed significantly, suggesting that the newly updated and high-resolution emission inventory will be useful for the identification of air pollution sources in YRD.

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1. Introduction

With rapid economic development as well as the dramatic growth of energy consumption and number of motor vehicles in China, emissions of air pollutants have significantly increased in recent years, leading to complex and regional air pollution issues such as haze and photochemical smog. The Yangtze River Delta (YRD) is located in the eastern part of China, and includes the city of Shanghai, plus the provinces of Jiangsu and Zhejiang. The region is one of China's most developed and heavy-polluted regions.

A significant decrease in visibility was seen from historical data (Chang et al., 2009; Gao et al., 2011). High concentrations of fine particles and ozone have been monitored, indicating the severe air pollution situation (Xiao et al., 2011) which has adverse impacts on human health (Ge et al., 2011). From 2010, the YRD region has been identified as a key area for joint prevention and control of air pollution, which is an important air pollution control plan for China (Wang and Hao, 2012). It has been a challenge for policy makers and researchers to identify emission sources, understand the contamination processes, and develop effective air pollution control strategies.

Emission inventories are the foundation for air quality modeling and analysis, in order to understand the formation and transport of

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pollutants and guide the air pollution control (Che et al., 2011; Li et al., 2011). Up to now, multi-scale emission inventories have been established, from urban (Zhang et al., 2008) and regional (Zheng et al., 2009b; Zhao et al., 2012), to national (Cao et al., 2011) or even continental (Ohara et al., 2007; Streets et al., 2003). Some studies also focused on individual pollutants (Su et al., 2011; Yu et al., 2011) or one specific sector (Li et al., 2012). For the YRD region, Li et al. (2011) established an emission inventory of 2004 for 16 major cities and Huang et al. (2011) estimated the emissions of 16 major cities in 2007. In addition, Zhang et al. (2008) prepared the inventory for the city of Hangzhou in the YRD.

Most previous studies focused on the conventional pollutants, such as SO₂, NO_x and PM₁₀. But with the worsening secondary air pollution situation, more and more researchers began to pay attention to VOC and PM_{2.5}. Each of VOC species has different ozone formation potential. PM_{2.5} species such as sulfate and nitrate are important to haze formation and radiative forcing. In the United States, the SPECIATE database (<http://www.epa.gov/ttnchie1/software/speciate/>) gave a detailed VOC and PM profile based on an extensive review of the literatures. Reff et al. (2009) presented the US National Emissions Inventory of PM_{2.5} with 37 trace elements for the first time. In China, some emission inventories (Wei et al., 2008; Zheng et al., 2009a) have quantified VOC species based on Chinese local measurements and the SPECIATE database, but lacked detail speciation of PM_{2.5}.

As one of the fastest developing regions in China, control status and the characteristics of emissions may change substantially over the years. Therefore, an updated and high-resolution emission inventory is critical to the study on atmospheric chemistry and development of control policies in YRD. In addition to 16 major cities, surrounding cities such as Xuzhou and Wenzhou should be included for more comprehensive prevention and control of air pollution in YRD. Knowledge of PM_{2.5} speciation is also important for the study on haze formation and radiative forcing in this region.

In this paper, we established a bottom-up emission inventory with high spatial resolution for total 25 cities of the YRD in 2010, including SO₂, NO_x, PM₁₀, PM_{2.5}, NMVOCs and NH₃. The speciation of PM_{2.5} and VOC has been further refined. Section 2 describes the relative methodology and data sources. The results about sectoral emissions, spatial distributions, as well as the PM_{2.5} and VOC speciation are presented in Section 3. Besides, the comparison with other studies and the uncertainties are also discussed.

2. Data and methods

2.1. Study domain

The YRD traditionally included only 16 cities, but with dramatic economic development and increasing environmental concerns in this region, 5 more cities in Jiangsu and 4 cities in Zhejiang are included in the larger YRD, for a total of 25 cities (see Fig. 1). The study domain is from 115.45°E to 124.07°E, and 27.26°N to 35.62°N. This region covers 213340 km², only about 2.22% of China. However, it represents 11.65% of population, 21.51% of GDP, 16.57% of energy consumption and 16.26% of vehicle population in 2010 (National Bureau of Statistics of China, 2011a, 2011b).

2.2. Methodology

An emission factor approach was used to estimate emissions, which was implemented by the Equation (1):

$$E_{ij} = \sum_{m,n} A_{i,j,m,n} EF'_{i,j,m,n} \left(1 - \sum_k C_{i,j,m,n,k} \eta_{i,j,m,n,k} \right) \quad (1)$$

where A is the activity data; EF' is uncontrolled emission factor; C is application rate of each control technology; η is removal efficiency; i, j, m, n and k represent the region, pollutants, sectors, fuel/activity/technology type and control technology, respectively.

We grouped the emission sources into 4 levels, including 10 major sectors (i.e. power plants, industry processes, on-road transportation, etc.) and each sector had subsectors based on different fuel, product, use and technology (see Table 1). Emissions from large point sources including 425 power plants, 704 cement factories, and 20 large iron–steel factories have been estimated based on detailed investigation data collected from relative associations. The pollutants including SO₂, NO_x, PM₁₀, PM_{2.5}, NMVOCs and NH₃ have been considered in this paper. Speciation of VOC and PM_{2.5} species has been conducted in order to better support the analysis on the studies of photochemical smog, haze and radiative forcing. The estimated inventories were gridded into 4 km × 4 km spatial grids based on high-resolution information of point sources, population, road network and land cover, using same method as documented in Wang et al. (2011).

2.3. Activity data

We used detailed point source information, including latitude/longitude coordinates, annual product, technology/process, and pollution control facilities. The location information of these point sources is shown in Fig. 1. Activity data of other sectors were collected at the city or even county level based on the statistical yearbooks of the 25 cities, covering energy consumption, production, vehicle population and so on. However, activity data for some solvent use sources such as adhesives and printing were difficult to collect directly at the city level, so we used activity data at the provincial or national level (Gong, 2008; China Printing Yearbook Press, 2011) and then allocated them into cities according to construction areas, vehicle production, industrial GDP and so on. Table 2 shows the activity data for major sectors in 2010.

2.4. Emission factor

2.4.1. Power plant

The calculation methods for emission factors of SO₂, NO_x, PM were based on the studies of Zhao et al. (2008) and Zhao et al. (2010). The parameters such as the sulfur content and the ash content in each type of fuel were at provincial level and taken from Zhao et al. (2008). But the application rates of emission control technology have been further investigated. In 2010, the installation rates of FGD in Shanghai, Jiangsu and Zhejiang have reached 81.3%, 85.3% and 86.9%, respectively (http://www.mep.gov.cn/gkml/hbb/bgg/201104/t20110420_209449.htm). With regard to NO_x control, 29.0%, 21.9% and 32.4%, respectively, of power plants have installed SCR/SNCR (http://www.mep.gov.cn/gkml/hbb/bgg/201104/t20110420_209449.htm). For PM control, over 95% of power plants use ESP (China Electricity Council, 2011).

2.4.2. Other fossil fuel combustion in industrial and domestic sector

Except for power plants, other fossil fuel combustion sources are also important for most pollutants. The emissions of SO₂ were calculated in the same way as for the power plant sector. During past decade, Tsinghua University has collected some data for emission factors based on field measurements and the literature, covering NO_x (Tian et al., 2001; Zhang et al., 2007), PM (Lei et al., 2011; Wang et al., 2009b; Li et al., 2009a) and VOCs (Wei et al., 2008). The emission factors of different fuel types in industrial and domestic sectors are summarized in Table 3.

2.4.3. Industrial process

In addition to fuel combustion sector, industrial processes also emit much pollution. The YRD is one of the largest cement

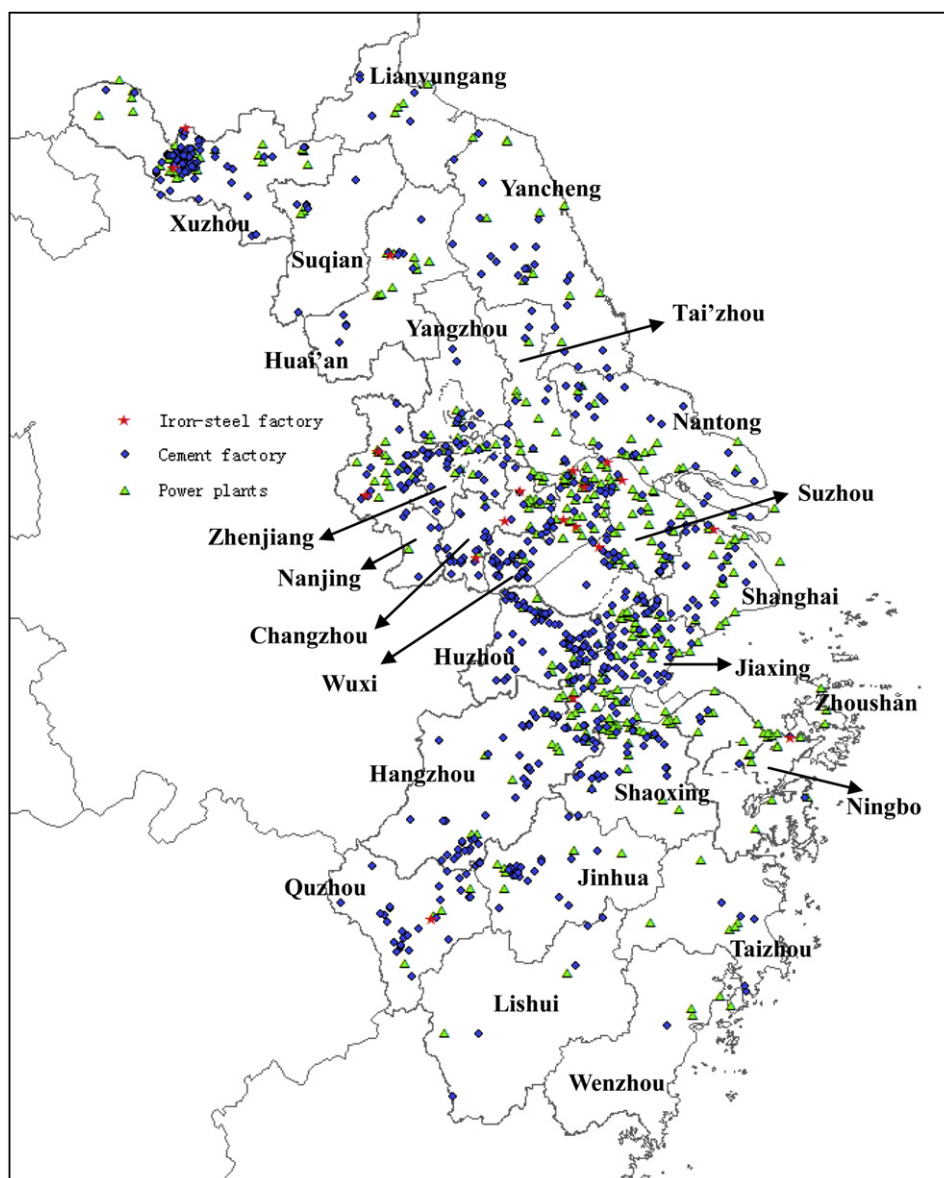


Fig. 1. Study domain and location of point sources in the larger YRD region.

production areas. By surveys, we obtained the information about cement kilns type from the China Cement Association (CCA) and Lei et al. (2011), including precalciner kilns, rotary kilns and shaft kilns. Zhang (2008) and Lei et al. (2011) reported the emission factors of SO_2 , NO_x and PM for different types of kilns. Control measurements were considered based on local information. Emission factors of other subsectors were from the results of Zhang (2008) and Zhao et al. (2012). VOC emission factors were referred to the study of Wei (2009). We followed the method provided by Dong et al. (2010) to calculate NH_3 emissions. The summary of emission factors used in industrial process is given in Table 4.

2.4.4. Transportation sector

The newest result about emissions estimation for transportation sector was used in this study (Shu et al., 2012). The emission factors were calculated by a new developed model called EMBEV, which can provide detailed estimates of emission factors for an on-road vehicle fleet combining vehicle population, annual VKT, and fuel type share of each technology group. The sources of relevant data

including fuel quality, technology distribution, driving conditions and I/M program were described in Shu et al. (2012).

2.4.5. Solvent use and fossil fuel distribution sector

For solvent use, some kinds of products are limited on solvent content by Chinese national standards, such as wood paint by GB18581-2009, interior wall paints by GB18582-2008, decorative adhesives by HBC12-2003 and GB18583-2008, adhesives used in bags and shoes industry by HBC12-2003, ink in offset-printing by HJT370-2007, and ink in flexography and rotogravure by HJT371-2007. For some other subsectors, such as domestic solvent use, decorative solvent-based paints, emission factors came from the study of Wei et al. (2008).

The fossil fuel distribution sector is another important source of VOC. Based on the work of Wei et al. (2008), we considered the limitation of some new Chinese national regulations including GB11085-89, GB20950-2007, GB20951-2007 and GB20952-2007. From 2010, vapor recovery systems should be installed for oil storage, tankers and oil stations. Table 5 shows the major emission factors in solvent use and fossil fuel distribution sector.

Table 1
Emission source categorization in the larger YRD region.

Sector	Subsector	Fuel/product/use	Technology/process
Power plant	—	Coal/oil/gas fuel	Stoker furnace/pulverized coal boiler/circulating fluidized bed boiler
Industrial combustion	—	Coal/oil/gas fuel	Stoker furnace/pulverized coal boiler/circulating fluidized bed boiler
Domestic combustion	Fossil fuel	Coal/oil/gas fuel	Stoker furnace/stove
	Biomass	Rice straw/wheat residue/maize residue	—
Industrial process	Cement plant	Clinker production	Shaft kilns/precaciner kilns/other rotary kilns
		Other process	Crushing/grinding/blending/loading
	Iron–steel plant	Sintering/iron/steel	Open–hearth furnace/electric furnace/converter
	Other industry	Refining-plant/coking-plant/brick-plant/food-industry/textile-industry/paper-production/chemistry-industry/pharmaceutical-industry	—
On-road transportation	Truck/bus/car	Gasoline/diesel	Heavy/light
Off-road transportation	Boat/construction machine/train/agriculture machine	Gasoline/diesel/coal	—
Solvent use	Paint use	Building/vehicle/wood/industry protection	—
	Adhesive use	Wood-processing/shoes/clothing/packaging/building	—
	Printing	Packaging/publication	Offset/flexography/rotogravure/screen printing
Fossil-fuel distribution	—	Crude oil/gasoline/diesel	Storage/handling/transportation/sale
Biomass open burning	—	Rice straw/wheat residue/maize residue	—
Agriculture sources	Livestock	Cow/pig/goat/fowl/rabbit	—
	Fertilizer application	Nitrogen fertilizer/compound fertilizer	—

2.4.6. Biomass burning

Biomass burning, which includes household combustion and opening burning, is an important source of PM and gaseous pollutants. In this paper, we chose the emission factors from local field measurements. Li et al. (2007a) and Wang et al. (2009c) measured emission characteristics of particulate matter and gaseous pollutants from rural household bio-fuel combustion. Li et al. (2007b, 2009b) did the same measurements for open burning of wheat straw and corn stover. We also referred to the research of Lu et al. (2011) and Tian et al. (2002). The data used in this paper are listed in Table 6.

2.4.7. Agriculture sources

The livestock and fertilizer application are the largest emission sources for NH₃. We estimated the NH₃ emissions of livestock

including cattle, pigs, horses, and chickens. The NH₃ emission factors for fertilizer application were estimated based on fertilizer types and their application rate. All data came from some Chinese local studies (Yin et al., 2010; Dong et al., 2010).

2.5. Speciation of PM_{2.5} and NMVOC

In this paper, 18 species of PM_{2.5}, including OC, EC, sulfate, nitrate, H₂O, Na, Cl, NH₄⁺, non-carbon organic matter (NCOM), Al, Ca, Fe, Si, Ti, Mg, K, Mn and others, have been estimated. The Chinese local measurements data were first used for PM_{2.5} speciation in each emission source, including power plants (Yi, 2006; Wang, 2007), industrial combustion (Wang et al., 2009b; Li et al., 2009a), domestic coal combustion (Zhi et al., 2008; Chen et al., 2006), biomass burning (Li et al., 2007b), vehicles (He et al., 2008; Cheng et al., 2010), cement production (Ma, 2010), iron–steel production (Ma, 2009) and coking (Li, 2009). Some other

Table 2
Activity data for major emission sources in 2010.

	Shanghai	Jiangsu	Zhejiang
Power plant			
Coal (10 ⁶ t)	34.21	126.13	82.54
Industry combustion			
Coal (10 ⁶ t)	7.56	45.69	36.99
Oil (10 ⁶ t)	5.00	4.45	6.64
Natural gas (10 ⁹ m ³)	1.96	3.57	0.79
Domestic combustion			
Coal (10 ⁶ t)	1.48	1.31	0.49
Natural gas (10 ⁹ m ³)	1.18	0.96	0.64
LPG (10 ⁶ t)	0.47	0.95	2.73
Industry process			
Cement production (10 ⁶ t)	6.71	156.75	112.75
Crude steel production (10 ⁶ t)	22.14	62.43	12.29
Transportation			
Goods vehicle population (10 ³ Vehicles)	238	725	873
Passenger vehicle population (10 ³ Vehicles)	1462	4728	4508
Motorcycle (10 ³ Vehicles)	1291	8075	5584
Solvent use			
Adhesive (10 ³ t)	92.41	575.22	585.21
Ink (10 ³ t)	14.31	22.25	26.57
Agriculture sources			
Nitrogenous fertilizer application (10 ³ t)	62	1795	525

Table 3
Emission factors for fossil fuel combustion sources.

		NO _x	PM ₁₀	PM _{2.5}	VOC
Industrial combustion					
Coal	g kg ^{−1}	7.5/4.0/3.8 ^{a,*}	0.90 ^{b,c}	0.59 ^{b,c}	0.04 ^f
Fuel oil	g kg ^{−1}	5.84 ^d	1.03 ^e	0.67 ^e	0.12 ^f
Diesel	g kg ^{−1}	9.62 ^d	0.50 ^e	0.50 ^e	0.12 ^f
LPG	g kg ^{−1}	2.63 ^d	0.17 ^e	0.17 ^e	0.10 ^f
Natural gas	g m ^{−3}	2.09 ^d	0.17 ^e	0.17 ^e	0.10 ^f
Domestic combustion					
Coal	g kg ^{−1}	4.0/3.8/0.91 ^{a,**}	5.4/3.5/8.8 ^{e,**}	1.89/2/6.86 ^{e,**}	4.5 ^f
Fuel oil	g kg ^{−1}	1.95 ^d	0.75 ^e	0.28 ^e	0.12 ^f
Diesel	g kg ^{−1}	3.21 ^d	0.50 ^e	0.50 ^e	0.12 ^f
LPG	g kg ^{−1}	0.88 ^d	0.17 ^e	0.17 ^e	6.51 ^f
Natural gas	g m ^{−3}	1.46 ^d	0.17 ^e	0.17 ^e	0.15 ^f

* For fluidized-bed furnace, automatic stoker and hand-feed stoker.

** For automatic stoker, hand-feed stoker and coal stove.

^a Zhang et al. (2007).

^b Wang et al. (2009b).

^c Li et al. (2009a).

^d Tian et al. (2001).

^e Lei et al. (2011).

^f Wei et al. (2008).

Table 4

Emission factors for main subsectors of industrial process.

Activity		SO ₂	NO _x	PM ₁₀	PM _{2.5}	VOC	NH ₃
Cement	g kg ⁻¹	0.23/1.46/0.64 ^{c,*}	1.10/2.75/0.66 ^{c,*}	0.77/1.25/0.60 ^{a,*}	0.46/0.80/0.35 ^{a,*}	0.12 ^f	
Sintering	g kg ⁻¹	3.00 ^c	0.14 ^c	0.20 ^b	0.15 ^b	0.03 ^f	
Iron	g kg ⁻¹	0.30 ^d	0.09 ^d	0.10 ^b	0.08 ^b		
Steel	g kg ⁻¹			0.16/0.13/0.07 ^{b,**}	0.14/0.10/0.06 ^{b,**}	0.12 ^f	
Coke	g kg ⁻¹	2.01 ^d	0.02 ^d	0.45 ^b	0.44 ^b	2.40 ^f	0.01 ^g
Brick	g kg ⁻¹		0.32 ^c	0.71 ^b	0.24 ^b	0.20 ^f	
Crude oil	g kg ⁻¹	3.62 ^e	0.56 ^e	0.12 ^b	0.09 ^b	2.10 ^f	0.16 ^g
Sulfuric acid	g kg ⁻¹	13.46 ^c		0.38 ^e	0.38 ^e		
Nitric acid	g kg ⁻¹		1.38 ^c				
Fertilizer	g kg ⁻¹			0.40 ^e	0.04 ^e		2.50 ^g
Paint	g kg ⁻¹					15.00 ^f	
Vegetable oil	g kg ⁻¹					3.69 ^f	
Beer	g kg ⁻¹					0.35 ^f	
Spirits	g kg ⁻¹					24.99 ^f	
Synthetic rubber	g kg ⁻¹					4.69 ^f	

* The data for cement plant represent the emission factors for precalciner kilns, rotary kilns shaft kilns, respectively. Control measurements have been considered.

** The data for steel represent the emission factors for open-hearth furnace, electric furnace and converter. control measurements have been considered.

^a Lei et al. (2011).^b Lei (2008).^c Zhang (2008).^d Huang et al. (2011).^e Zhao et al. (2012).^f Wei (2009).^g Dong et al. (2010).

data lacking local measurements came from the research of Reff et al. (2009). Using the methodology described by Reff et al. (2009), we generated the final profile, particularly the mass fractions determination of unmeasured species and mass balance. The profile is shown as Fig. 2. BC, OC and SO₄²⁻ are mainly emitted from fossil fuel combustion, but the shares in different sources varied. For example, SO₄²⁻ accounts for 28% of PM_{2.5} for industrial coal combustion, while only 2% for residential coal combustion. The sulfur content is an important influencing factor. For power plants, the fuel combustion efficiency is high, so the emissions of OC and

EC are low. For sintering and iron production, the emissions of Fe, Si, Ca and Al are large because they are major components in the iron ore. For cement production, Ca, Si, K and SO₄²⁻ which come from calcium carbonate, clay and coal account for a large proportion. Besides BC and OC, biomass burning also emits a considerable amount of Cl and K. The profiles have some differences with the US SPECIATE database, because of different fuel quality, different combustion types, different industrial materials, or different technology. For example, for sintering furnace, the emission factors of Na and Ca in the US account for 13.0% and 0.6%. But in China, they account for 3.4% and 9.1%.

We also performed a detailed analysis of VOC speciation, which was broken down into 40 categories as described by Wei (2009). Most data of VOC chemical profile came from Chinese local measurements, including biomass burning (Wei et al., 2008; Li et al., 2009b), solvent use (Yuan et al., 2010; Liu et al., 2008), vehicle exhaust and evaporation (Liu et al., 2008; Fu et al., 2008; Wang et al., 2008; Cai and Xie, 2009), residential fuel burning (Liu et al.,

Table 5

The major VOC emission factors for solvent use and fossil fuel distribution sector.

Activity	VOC emission factors
Solvent use ^a	
Paint_interior wall (g kg ⁻¹)	120
Paint_external wall (g kg ⁻¹)	580
Paint_manufacture of vehicle (g kg ⁻¹)	460
Paint_wood coating (g kg ⁻¹)	637
Adhesive_wood processing (g kg ⁻¹)	88
Adhesive_manufacturing of shoes (g kg ⁻¹)	664
Ink_offset-printing (g kg ⁻¹)	658
Ink_flexography and rotogravure in the packaging (g kg ⁻¹)	515
Ink_rotogravure in publication (g kg ⁻¹)	668
Ink_screen printing (g kg ⁻¹)	668
Solvent_leather production (g kg ⁻¹)	224
Crude oil distribution ^b	
Storage_oilfield/port/transfer station/refinery (g kg ⁻¹)	0.02/0.02/0.02/0.02
Loading_oilfield/port/transfer station (g kg ⁻¹)	0.24/0.08/0.24
Unloading_port/transfer station/refinery (g kg ⁻¹)	0.05/0.03/0.05
Gasoline distribution ^b	
Storage_transfer station/refinery (g kg ⁻¹)	0.03/0.03
Loading_transfer station/refinery (g kg ⁻¹)	0.87/0.87
Unloading_transfer station (g kg ⁻¹)	0.1
Sale_service/others (g kg ⁻¹)	2.44/3.97
Diesel distribution ^b	
Storage_transfer station/refinery (g kg ⁻¹)	0.10/0.10
Loading_transfer station/refinery (g kg ⁻¹)	0.10/0.10
Unloading_transfer station (g kg ⁻¹)	0.03
Sale_service/others (g kg ⁻¹)	0.07/0.10

^a Chinese national regulations or standards listed in the paper.^b Based on the study of Wei et al. (2008), considering about the new regulation.**Table 6**

Emission factors for biomass burning.

	SO ₂	NO _x	PM ₁₀	PM _{2.5}	VOC	NH ₃
Household burning						
Rice	0.53 ^a	0.42 ^c	3.32	1.66 ^d	7.36 ^c	1.30 ^e
Wheat	0.53 ^a	0.86 ^c	11.22	5.61 ^d	13.74 ^c	1.30 ^e
Maize	0.53 ^a	0.76 ^c	4.9	2.45 ^d	10.59 ^c	1.30 ^e
Cotton	0.53 ^a	1.29 ^a	12.08	6.04 ^d	10.56 ^e	1.30 ^e
Sorghum	0.53 ^a	0.90 ^c	12.54	6.27 ^d	0.23 ^c	1.30 ^e
Other	0.53 ^a	1.29 ^a	7.88	3.94 ^c	10.56 ^e	1.30 ^e
Open burning						
Rice	0.53 ^a	1.29 ^a	19.3	9.65 ^c	15.70 ^e	0.52 ^e
Wheat	0.85 ^b	3.30 ^b	15.2	7.60 ^b	7.50 ^b	0.37 ^b
Maize	0.44 ^b	4.30 ^b	23.4	11.7 ^b	10.00 ^b	0.68 ^b
Cotton	0.53 ^a	1.29 ^a	19.3	9.65 ^c	15.70 ^e	0.52 ^e
Sorghum	0.53 ^a	1.29 ^a	19.3	9.65 ^c	15.70 ^e	0.52 ^e
Other	0.53 ^a	1.29 ^a	19.3	9.65 ^c	15.70 ^e	0.52 ^e

* As no relevant literatures for PM₁₀, assume PM₁₀/PM_{2.5} = 2 (Zhao et al., 2012).^a Tian et al. (2002).^b Li et al. (2007b).^c Wang et al. (2009c).^d Li et al. (2007a).^e Lu et al. (2011).

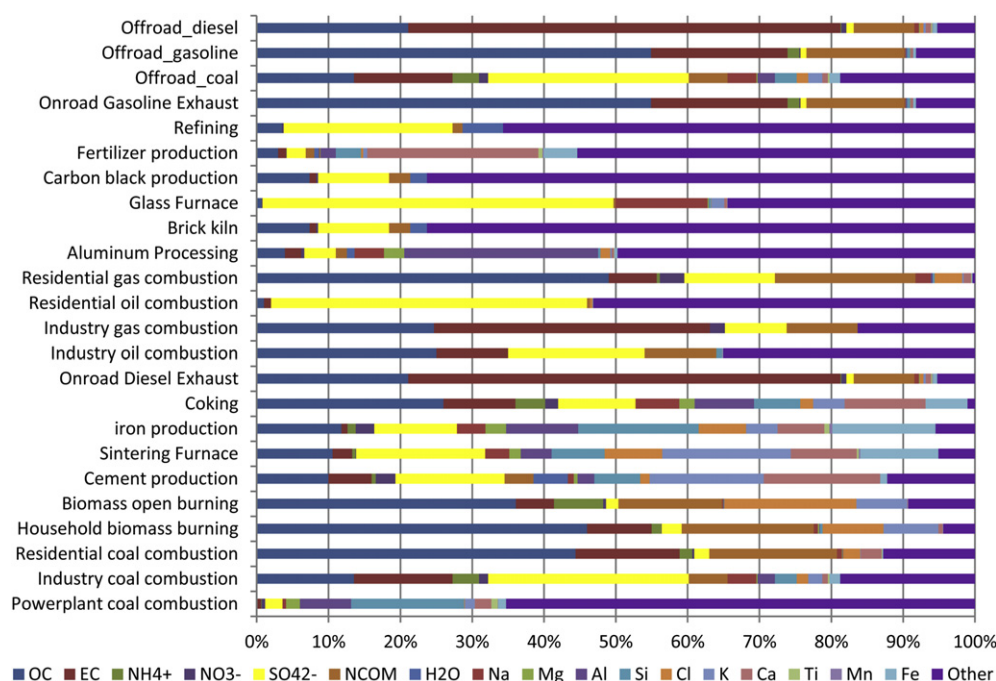


Fig. 2. The speciation profile of PM_{2.5} for different emission sources.

2008; Zhang et al., 2000; Wei, 2009), petrochemical industry (Liu et al., 2008; Wang et al., 2008), coking (He et al., 2005; Jia et al., 2009), and synthetic leather production (Wang et al., 2009a). Due to the lack of Chinese local information for some other emission sources, data in the SPECIATE database were used.

3. Results and discussion

3.1. Pollutant emissions by city and sector

In 2010, the total emissions of SO₂, NO_x, PM₁₀, PM_{2.5}, NMVOCs and NH₃ were 2147 kt, 2776 kt, 1006 kt, 643 kt, 3822 kt and 1439 kt, respectively. The city-level emissions are shown in Table 7. Shanghai, Nanjing, Suzhou, Wuxi, Xuzhou, Hangzhou and Ningbo are the top 7 cities with largest emissions in the YRD, sum of which accounts for 56%, 57%, 47%, 48%, 40% and 31% of total SO₂, NO_x, PM₁₀, PM_{2.5}, NMVOCs and NH₃.

Fig. 3 shows the sectoral contributions to total emissions for 6 pollutants. For SO₂, power plants, industrial combustion and industrial process are the largest sources, contributing 44.1%, 25.5% and 17.9%, respectively. Besides power plants, SO₂ emission from other sources should also be controlled, especially industrial boilers and sinter machines. Application of low sulfur coal is a good control option for domestic combustion.

NO_x emissions largely come from power plants (37.3%) and on-road transportation (24.9%). Power plants are the primary objects for NO_x control in the twelfth Five-Year period. The installation rates of SCR/SNCR in Shanghai, Jiangsu and Zhejiang are only 29.0%, 21.9% and 32.4% in 2010, so there is still a large emission reduction potential. On the other hand, the vehicle population has grown rapidly in recent years. From 2000 to 2010, emission standards for vehicles has developed from Euro I to Euro III. In 2010, Shanghai began to implement Euro IV for light-duty gasoline passenger cars and public fleets.

Industrial processes are the major contributors for PM₁₀ and PM_{2.5}, accounting for 26.9% and 28.9%, respectively. For industrial subsectors, cement plants contribute 52% of PM₁₀ and 43% of PM_{2.5}.

The YRD region is the largest producer of cement in China. In recent years, the Chinese government has taken some measures to promote technical transformation. Compared with 2005, the percent of cement produced by shaft kilns and other rotary kilns has decreased from 28% to 2%. Besides, bag filters are now being widely used in the cement industry.

The sources from industrial process and solvent use contribute 33.7% and 37.3% of NMVOC emissions. So far, VOC hasn't gained as much attention as SO₂, NO_x or PM. From 2010, the YRD region is scheduled to implement national regulations including GB20950-2007, GB20951-2007 and GB20952-2007 to control VOC

Table 7
Emission inventory of primary air pollutants for 25 cities in the YRD in 2010 (kt).

City	SO ₂	NO _x	PM ₁₀	PM _{2.5}	VOC	NH ₃
Shanghai	259.5	452.7	86.1	59.3	421.7	64.5
Nanjing	131.8	133.6	51.6	35.3	167.7	38.5
Suzhou	275.6	236.2	89.6	60.1	229.0	37.4
Wuxi	141.7	151.9	52.0	33.1	147.5	25.7
Yangzhou	55.5	65.9	33.4	20.9	106.0	45.8
Tai'zhou	49.0	58.1	32.0	20.4	111.7	50.6
Changzhou	72.4	67.5	32.4	21.1	126.0	29.3
Zhenjiang	68.7	75.6	31.1	18.9	72.8	18.6
Nantong	50.1	79.0	44.9	28.0	240.1	112.0
Lianyungang	32.5	43.3	33.1	21.9	68.9	75.9
Xuzhou	151.8	179.0	83.0	52.4	125.0	186.5
Huai'an	54.6	59.2	42.4	28.0	75.3	83.7
Yancheng	26.9	65.8	61.0	38.6	154.4	189.1
Suqian	15.3	41.7	32.2	21.8	134.1	82.7
Hangzhou	98.3	155.4	44.2	28.2	278.1	51.2
Jiaxing	120.8	89.4	37.5	20.9	158.9	66.2
Zhoushan	17.4	13.3	3.3	1.8	131.0	3.9
Shaoxing	53.3	70.2	30.3	19.1	284.3	37.5
Ningbo	140.4	262.2	54.5	33.8	176.6	43.0
Taizhou	92.8	107.1	24.7	15.0	98.5	28.4
Huzhou	50.0	93.6	33.3	19.6	156.0	36.4
Wenzhou	29.3	86.9	16.0	10.5	197.7	33.0
Jinhua	77.6	126.9	30.9	18.6	85.4	40.0
Quzhou	51.6	38.1	21.2	12.9	38.8	41.2
Lishui	30.5	23.9	5.4	3.3	36.1	17.5

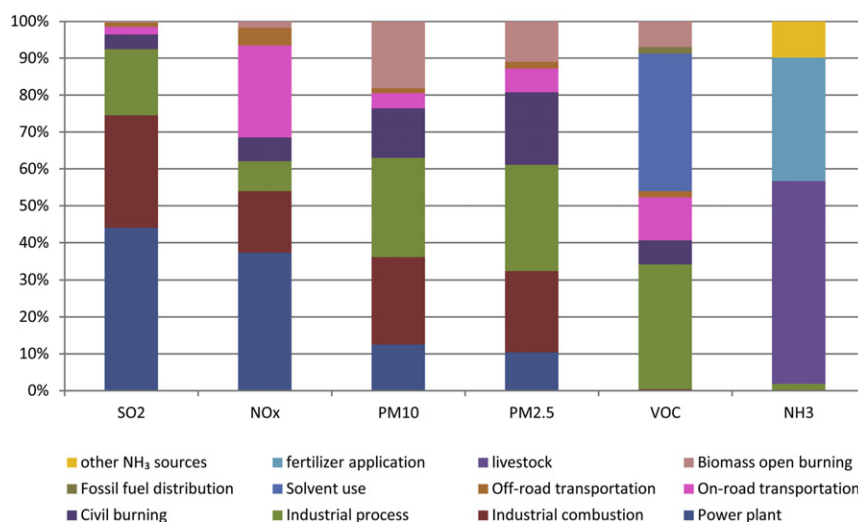


Fig. 3. Sectoral contributions to emissions in the YRD region for the year of 2010.

emissions from fuel depots, tankers and filling stations. As for industrial process and solvent use, the Chinese government only requires some solvent content limits which are still not strict compared with developed countries. Few end treatment facilities for VOC have been installed. Unlike other pollutants, the NH_3 emissions from non-industrial sources are far more important than other sources, with 54.9% from livestock and 33.4% from fertilizer application.

3.2. Spatial distribution and emission intensity

For use in air quality model, the emission inventories were gridded into $4 \text{ km} \times 4 \text{ km}$ grids (see Fig. 4). Heavy emission densities are shown in Shanghai and the area around Tai Lake, which is higher by 1–2 orders of magnitude than most area in northern part of Jiangsu and southern part of Zhejiang. Even though the spatial distribution of different pollutants is generally similar, some differences can be seen. For example, the road network is clearly visible for the spatial distribution of NO_x , indicating the large contribution of the transportation sector.

The average emission intensity in the YRD for SO_2 , NO_x , PM_{10} , $\text{PM}_{2.5}$, NMVOCs and NH_3 are 10.06, 13.01, 4.72, 3.01, 17.91 and 6.74 t km^{-2} , respectively, which are 3–8 times of the average level of China. For different cities in this region, the range of the emission intensities are 1.58–40.93 t km^{-2} for SO_2 , 1.38–71.40 t km^{-2} for NO_x , 0.31–13.58 t km^{-2} for PM_{10} , 0.19–9.35 t km^{-2} for $\text{PM}_{2.5}$, 2.09–90.97 t km^{-2} for VOC and 1.01–16.91 t km^{-2} for NH_3 . This huge variation range comes from different level of development, different industrial structure, area and so on. Such cities as Shanghai and Suzhou are in a high level of emission intensity, but such cities as Quzhou and Lishui are in relatively low level. For NH_3 , Such cities with developed agriculture as Xuzhou and Nantong, have relatively high emission intensities.

3.3. Emissions of $\text{PM}_{2.5}$ and NMVOC species

Detailed speciation profiles for 23 subsectors have been collected in this study. More than 70% of the data are from Chinese local measurements. Based on the profiles, emissions of 18 $\text{PM}_{2.5}$ species were further calculated. For the YRD region in 2010, the major $\text{PM}_{2.5}$ species are OC, EC and sulfate, with 136.9 kt, 75.0 kt and 76.2 kt, respectively. Fig. 5(a) illustrates the speciation for each sector. For power plants, fuel combustion efficiency is high, so

emissions of OC and EC are low. In domestic combustion sector, household biomass burning emits most $\text{PM}_{2.5}$, so the speciation result of domestic combustion sector is similar to biomass open burning, predominated by OC, NCOM, CI and K. OC and EC are the largest-emitted species for transportation sector. Sulfate emission mainly comes from industrial combustion and industrial process. Fig. 5(b) shows the speciation for different cities and this result depends on species profile and emission by sector in each city.

Fig. 6 shows the NMVOC speciation by sectors and cities. The main NMVOC species are aromatics (30.4%) and alkanes (20.3%). For the largest emission sources, i.e. solvent use sector, ketones, aromatics and esters are the predominant species, accounting for 41.4%, 28.1% and 27.6%, respectively. And the species emitted by industrial processes are relatively numerous and at a similar level. Alkanes and aromatics are also the major emission species for the on-road transportation sector, contributing 51.2% and 34.1%, respectively. The NMVOCs profile for the cities are somewhat similar, mainly aromatics and alkanes. However, some differences can be seen. For example, aromatics, accounting for 78.5%, are predominant in Zhoushan, but in Wenzhou the contributions of ketones and esters are nearly equal, indicating the different VOC source in different cities.

3.4. Comparison with other inventories

Some regional emission inventories for the YRD are available. For example, Huang et al. (2011) developed a 2007 inventory, but only for 16 cities in the YRD. In addition, some Asian or China emission inventories are available from 2000, which included all 25 cities in the YRD region by province level. We chose the research of Streets et al. (2003), Wei et al. (2008), Lei et al. (2011) and Wang et al. (2011), and compared the air pollutant emission inventories in different year for the YRD region, shown as Fig. 7. The emissions have changed significantly from 2000 to 2010. Emissions of SO_2 have declined 49% from 2005 to 2010, which reflects the impact of SO_2 control policies, especially the FGD implementation at power plants, in the period of China's Eleventh Five-Year Plan. At the same time, Chinese government has also focused on PM control, so the PM_{10} and $\text{PM}_{2.5}$ emissions continued to decline from 2000 to 2010, which corresponds to the changing trend of ambient PM_{10} concentration (Cheng et al., 2013). NO_x emissions only increased 1.1% from 2005 to 2010. VOC emissions have an increase of 110% from 2000 to 2010, because currently

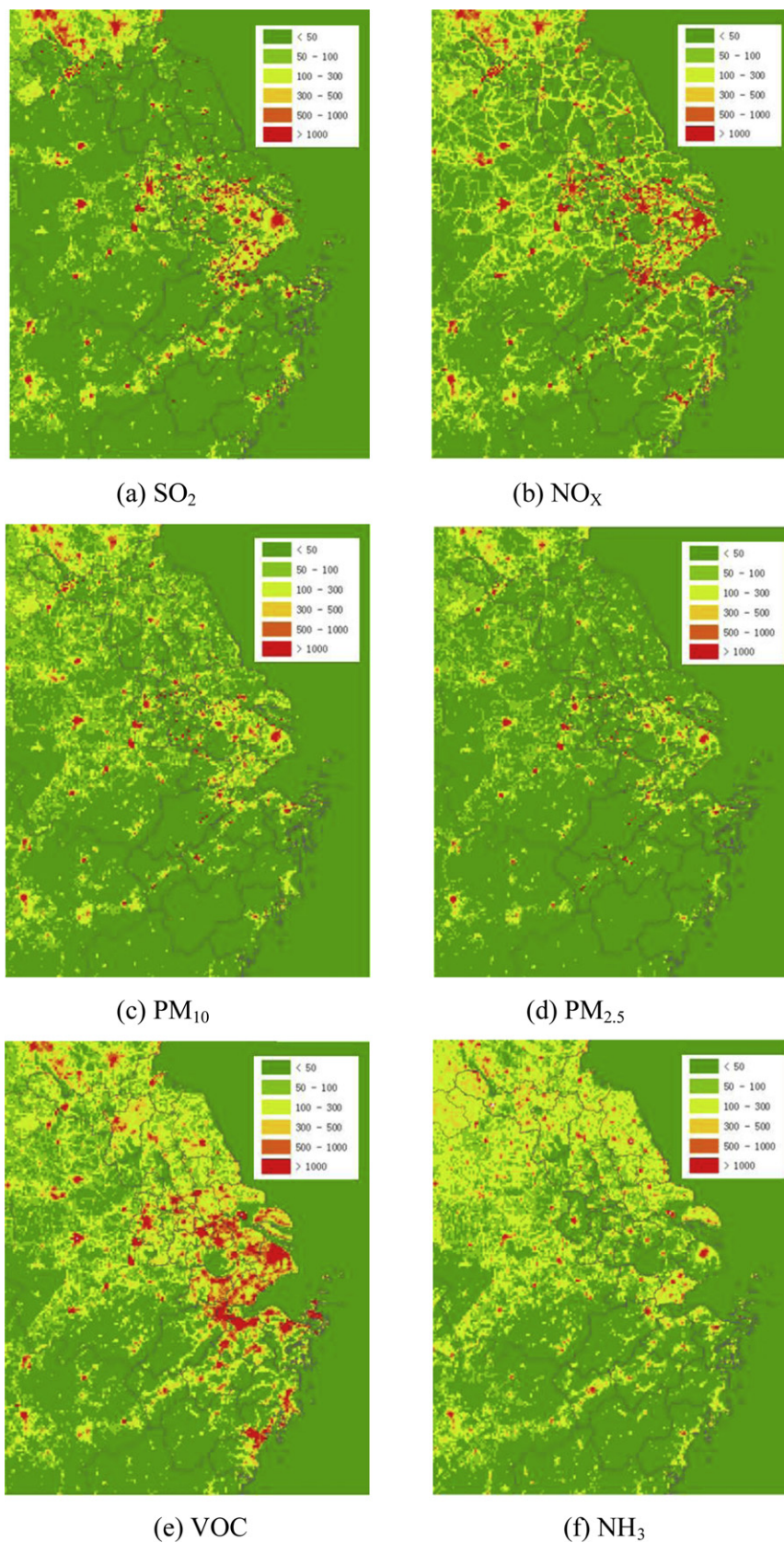


Fig. 4. Spatial distribution of air pollutant in the YRD region in 2010 (t 16 km⁻² per year).

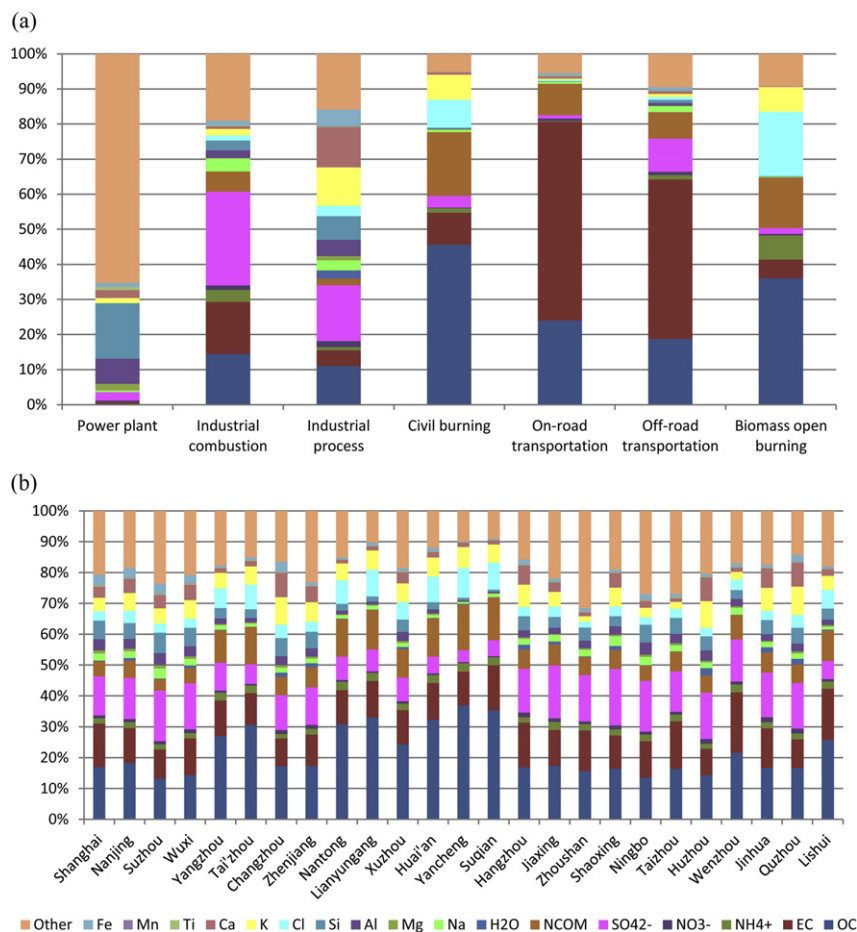


Fig. 5. $PM_{2.5}$ speciation. (a) $PM_{2.5}$ speciation by sectors; (b) $PM_{2.5}$ speciation by cities.

there are few control measures for VOC emissions in China. As the important precursors of secondary pollution, NO_x and VOC should be addressed for control in future.

3.5. Uncertainty analysis

The Monte Carlo method was used to estimate the uncertainty of this emission inventory (Wei et al., 2008; Zheng et al., 2009b). We assumed that the uncertainties of the activity data can be described by a normal distribution and emission factors can be described by a lognormal distribution (Zhao et al., 2011). The standard deviations for these distributions were estimated based on expert judgment, considering the reliability of data sources and estimation methods used. The total uncertainty for the emissions of SO_2 , NO_x , PM_{10} , $PM_{2.5}$, VOC and NH_3 are −12% to 16%, −16% to 21%, −27% to 40%, −31% to 46%, −52% to 105% and −36% to 77% respectively at the 95% confidence interval.

For PM, non-road mobile sources and biomass open burning are major contributors to the uncertainties. For biomass burning, the activity levels are estimated based on some parameters with large uncertainties, such as the application rate of open burning. The emission factors also have large uncertainties. Based on the measurement results of Li et al. (2009b), the $PM_{2.5}$ emission factor for open burning of wheat straw varies from 3.5 to 11.7 g kg^{-1} . The uncertainty ranges of emissions from biomass burning are −74% to 151% and −82% to 202% for PM_{10} and $PM_{2.5}$, respectively. Another large uncertainty contributor is the non-road transportation sector due to the lack of activity data and emission factor measurements.

For example, many factors can influence emission of boat, such as boat type, driving conditions, etc. However, we use one average emission factor for boat emission, which has high uncertainty. Compared with traditional pollutants, VOC and NH_3 have higher uncertainties. As the largest VOC sources, industrial process and solvent use have the uncertainty ranges of −57% to 152% and −60% to 147%. Most of the NH_3 emissions come from livestock and fertilizer application, but there are many influencing factors and related measurements are not enough. For example, the emission factors for fertilizer application can be affected by many factors, such as weather, soil moisture, soil pH etc. So the uncertainty for NH_3 is relatively large.

The detailed $PM_{2.5}$ profile for China is newly developed, but still has high uncertainty due to the lack of enough measurements. For example, in the test results of Wang et al. (2009b) and Li et al. (2009a), the ratio of SO_4^{2-} for different furnaces in industrial sector varies from 1.5% to 55.2%, which is affected by coal type, furnace type, and emission control technology, etc. For power plant and residential sector, most researchers focus on the emission of total $PM_{2.5}$, BC or OC. For SO_4^{2-} , there are only a few studies and the results of 1–2 samples were used, so the uncertainty is considerably high. For vehicle emissions, the testing methods, such as tunnel, on-board or roadside measurements, might affect the emission profile. Besides, vehicle type, oil quality and emission standards also affect the chemical composition of PM emission. In addition, different regions may exhibit variations in the profile. Therefore, more measurements are necessary in future to improve the speciation profile for source analysis or air quality modeling.

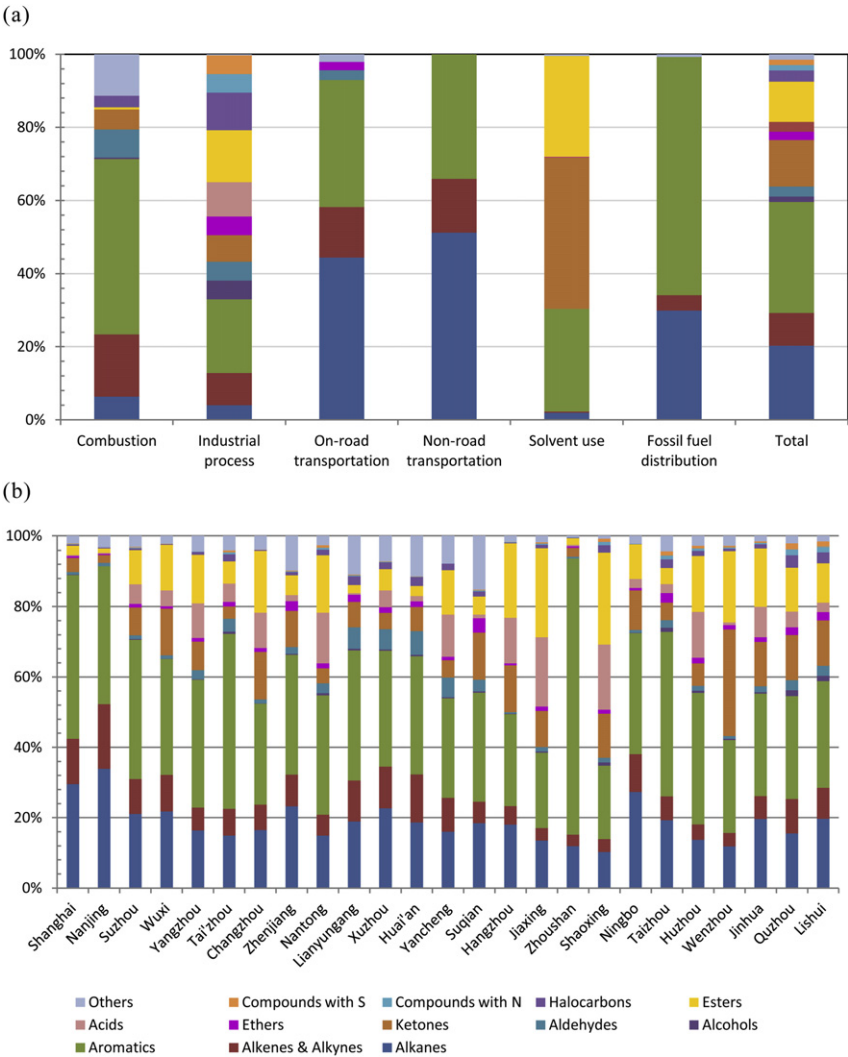


Fig. 6. NMVOC speciation. (a) NMVOC speciation by sectors; (b) NMVOC speciation by cities.

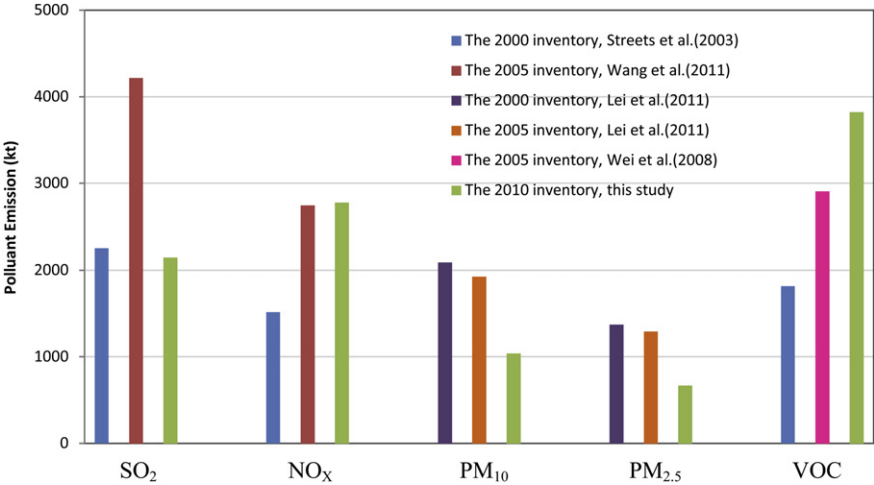


Fig. 7. Comparison of air pollutant emission inventory established by different studies.

4. Conclusion

In this paper, a highly resolved YRD region emission inventory for the year 2010 was established. The estimated emissions of SO₂, NO_x, PM₁₀, PM_{2.5}, NMVOCs and NH₃ were 2147 kt, 2776 kt, 1006 kt, 643 kt, 3822 kt and 1439 kt, respectively. Shanghai, Nanjing, Suzhou, Wuxi, Xuzhou, Hangzhou and Ningbo are the major contributors, accounting for 56%, 57%, 47%, 48%, 40% and 31% of SO₂, NO_x, PM₁₀, PM_{2.5}, NMVOCs and NH₃. Power plants contributed 44.1% of SO₂ and 37.3% NO_x. Emissions from industrial process accounted for 26.9% of PM₁₀, 28.9% of PM_{2.5} and 33.7% of VOC. Besides, 37.3% of NMVOC emission came from solvent use. Livestock and fertilizer application are major NH₃ sources.

A detail speciation of PM_{2.5} for the YRD region was developed for the first time. Composite speciation profiles for more than 20 subsectors have been accessed from numerous publications, of which more than 70% are from Chinese local measurements. For the YRD region in 2010, the major PM_{2.5} species were OC, EC and sulfate, with 136.9 kt, 75.0 kt and 76.2 kt, respectively. Sulfate emissions come mainly from industrial combustion and industrial process. Except for PM_{2.5}, detail NMVOC species were also estimated. Aromatics and alkanes were the main species, accounting for 30.4% and 20.3%, respectively.

The comparison of this inventory with previous estimates indicates that the emission of air pollutants in YRD has changes significantly in the past decade. Therefore, regularly updates of emission inventory are very crucial to reflect the sources of air pollution in fast changing areas such as China.

The uncertainties of PM are mainly from non-road transportation and biomass burning because of limited activity data statistics and few emission factor measurements. Among all air pollutants, NMVOCs and NH₃ are most uncertain. To further improve the emission inventory, more measurements of emission factors and PM_{2.5} speciation should be conducted for the high uncertainty sectors and pollutants. In addition, in order to apply the emission inventory from this study to air quality models, some more information needs to be incorporated in future work, such as emissions of biogenic VOC, CO and so on.

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