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The study of emission inventory on anthropogenic air pollutants and VOC species in the Yangtze River Delta region, China

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Abstract

The purpose of this study is to develop an emission inventory for major anthropogenic air pollutants and VOC species in the Yangtze River Delta (YRD) region for the year 2007. A "bottom-up" methodology was adopted to compile the inventory based on major emission sources in the sixteen cities of this region. Results show that the emissions of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, and NH_3 in the YRD region for the year 2007 are 2391.8 kt, 2292.9 kt, 6697.1 kt, 3115.7 kt, 1510.8 kt, 2767.4 kt, and 458.9 kt, respectively. Ethylene, mp-xylene, o-xylene, toluene, and 1,2,4-trimethylbenzene, 2,4-dimethylpentane, ethyl benzene, propylene, 1-pentene, and isoprene are the key species contributing 77% to the total OFPs. The spatial distribution of the emissions shows the emissions and OFPs are mainly concentrated in the urban and industrial areas along the Yangtze River and around the Hangzhou Bay. The industrial sources including power plant, other fuel combustion facilities, and non-combustion processes contribute about 97%, 86%, 89%, 91%, and 69% of the total SO_2, NO_x, PM_{10}, $PM_{2.5}$, and VOC emissions. Vehicles take up 12.3% and 12.4% of the NO_x and VOC emissions, respectively. Regarding OFPs, chemical industry, domestic use of paint and printing, and gasoline vehicle contribute 38.2%, 23.9%, and 11.6% to the ozone formation in the YRD region.

1 Introduction

The complex and regional air pollution issues such as acid deposition, photochemical smog and haze caused by fine particles have been the most crucial topic on atmospheric environment research in the Yangtze River Delta (YRD) region in China. High ozone concentration and fine particle pollution were monitored to demonstrate the deteriorated regional air pollution situation in this region (Gao et al., 2009; Geng et al., 2009; Tie et al., 2009). Rapid growth of transportation, industries, and urbanization in the YRD region comes to be the major environmental driving force to the deterioration

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of urban and regional air quality. Therefore, the quantification of anthropogenic air pollutant emissions will be fundamental to the understanding of air pollutant emission levels, forecasting air quality status, and guiding air pollutant control strategies.

As the key to air pollutant emission control and management, emission inventories are paid an increasing attention by policy-makers and researchers. In the last decade, some emission inventories have been developed on multi-scales in China, and most of them were compiled on national scale, even covering the whole of East Asia (Streets et al., 2003; Zhang, 2005; Ohara et al., 2007; Zhang et al., 2009). The objectives of prior studies were mainly focused on the primary air pollutants from fuel combustion sources, like SO_2, NO_x, and CO, etc. (Akimoto and Narita, 1994; Hao et al., 2002; Wang et al., 2005). After that, more studies began to involve other specific anthropogenic sources. Song and Xie (2006) estimated a vehicular emission inventories for the whole of China. Lei et al. (2008) and Wei et al. (2008) introduced PM and VOCs emission inventories based on the surveys of cement industries and other non-combustion industrial sources. Biomass burning emissions were also quantified as an important air pollution sources in some studies (Cao et al., 2005; Yan et al., 2006). Besides of primary air pollutants, researchers are gradually emphasizing the key precursors of secondary air pollution in order to mitigate the complex air pollution of ozone and haze in the regions of China. The species in the VOCs and PM emissions were quantified for various emission sources in recent studies (Streets et al., 2001; Cao et al., 2006; Bo et al., 2008).

From the view of method, most of the macro- or meso-scale emission inventories in China were established based on a top-down approach with low resolution of emission allocation. Low-resolution inventories were thought to cause under-estimation of air pollution simulation in recent modeling studies (Liu et al., 2010). In order to support regional air pollution study and management in the city clusters of China, some studies introduced highly resolved regional air pollutant emission inventories by bottom-up approach (Zheng et al., 2009a,b). Relatively, most of the studies on air pollutant emission inventories in the YRD region were still on the city-scale. Chen et al. (2006) and Zhang

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et al. (2008) have published the emission inventories for Shanghai and Hangzhou city, respectively. However, with city-scale emission inventory, it is hard to draw a whole picture of emission level and emission distribution for the YRD region.

Therefore, this paper aims to create an anthropogenic air pollutant emission inventory of the YRD region based on the year 2007. The pollutant types include SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs and NH_3. VOCs species emissions were further developed to evaluate the ozone formation potential in the YRD region. After calculation, the emissions were distributed into 4 km×4 km resolved grids to describe the spatial characteristics of air pollutant emissions in the YRD region.

2 Materials and methods

2.1 Domain of the study

The YRD region is located on China's eastern coast and covers eight city scale administrative regions in Jiangsu Province, which include Nanjing, Yangzhou, Tai'zhou, Nantong, Zhenjiang, Changzhou, Wuxi, and Suzhou, seven city scale administrative regions of Zhejiang Province, namely Hangzhou, Huzhou, Jiaxing, Shaoxing, Ningbo, Zhoushan, and Taizhou, and lastly Shanghai municipality itself. Each administrative region is about 7000 km^2 on the average. Most of the regions consist of an urban area and several counties, dozens of towns and villages, and extensive rural areas. Figure 1 shows the domain for the YRD regional emission inventory. The area is from 118.25° E to 122.42° E, and 28.90° N to 33.30° N, and divided into 11 979 grid cells with 4 km×4 km resolution.

The total area of the YRD region is about 110 000 km^2, representing 2% of China's total area. However, the GDP (Gross Domestic Product) of the YRD region reached 6.55 trillion yuan, about 20% of total national GDP in 2007 (National Bureau of Statistics of China, 2008b). Correspondingly, the energy consumption in the YRD region reached 440 million tce, about 17% of the national total by the end of 2007. Coal was

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still the major energy category of the region, which contributed over 60% of the total energy consumed (National Bureau of Statistics of China, 2008a). The automobile population grew to 8 million units in 2007, which accounts for 18% of the national whole. Huge energy consumption and vehicle population induced a great amount of the primary emissions and precursors in the region. The satellite observed the tropospheric NO_2 showed a sharp increase of 100% over the YRD region from the year 2000 to 2008. Figure 1 shows the satellite observed NO_2 concentration over the eastern China in 2007.

2.2 Methodology

The study is focused on the major anthropogenic emission sources in the YRD region, so the main sources of the industry, transportation, residential and agriculture sectors are included in the inventory. The industry sectors include fuel combustion processes of power plants, boilers, furnaces, kilns, and non-combustion processes such as iron and steel manufacturing, oil refining, cement producing, etc. Transport emission sources mainly consist of vehicle exhaust and road dust emissions. The residential emission sources cover most of the emissions associated with daily life, such as residential fuel combustion emissions, domestic paint and solvent use, gas evaporation in the service station and so on. The agriculture emission sources mainly include the emissions from livestock feeding, fertilizer application and biomass burning. The pollutants included in this study are SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs and NH_3. As an important precursor of photochemical smog, VOCs emissions were decomposed into various species by each kind of emission sources.

A large amount of activity data on sources and emission factors of these pollutants was collected to compile the emission inventory of the YRD region in 2007. And then GIS technology was utilized to allocate the emissions based on the geographical information of the individual emission sources. Finally, the emissions in the YRD region were allocated to the 4 km×4 km grid cells.

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2.3 Determination of emission factors

2.3.1 Fuel-related combustion sources

Fuel-related combustion sources in the YRD region mainly include power plants, boilers, furnaces, kilns, and some residential stoves. A nationwide campaign of pollution source census was conducted to get the basic data of these facilities by each city for the year of 2007. The activity data of each individual facility was directly collected from the census in some of the cities. For the other cities whose census data were not available, the activity data were collected from statistical information of the national key pollution sources reported by the government every year. The information of the facilities collected in the study included geographical data, fuel type, fuel consumption, sulfur content, ash content, boiler types, capacity, and exhaust treatment efficiency, etc. For the fuel combustion emissions in commercial and civil sector, the top-down approach is used based on the statistical data of fuel consumption from the statistical yearbooks of the cities in the YRD region.

The air pollutant emissions of fuel-related combustion are estimated using emission factor methods by Eq. (1):

$$E_{i,j} = \sum_{j,k} A_{j,k} \mathrm{EF}_{i,j,k} \left(1 - \eta_{i,j}\right) \quad (1)$$

Where, A is the activity level of the sources (e.g., annual fuel consumption); EF is the average emission factor. η is the exhaust control efficiency; and i, j, k represent the pollutant type, source category, and technology type, respectively.

For SO_2 emissions, the annual inventory was compiled using the mass balance method by Eq. (2). The emission factors of NO_x, CO, PM_{10}, $PM_{2.5}$, and VOCs were taken directly from the latest literature. NO_x emission factors of coal combustion came from the study of Zhang et al. (2007). NO_x emission factors of other fuel combustion were cited from Tian et al. (2003). CO emission factors were cited from Wang et al. (2005). PM_{10} and $PM_{2.5}$ emission factors were cited from Zhang et al. (2005).

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VOCs emission factors came from the study of Bo et al. (2008). NH_3 emission factors were negligible in the fuel combustion sources. The emission factors of NO_x for coal combustion facilities, and CO, PM_{10}, $PM_{2.5}$ emission factors were based on local or domestic studies. The emission factors of other pollutants were mainly based on the European or American results due to the lack of corresponding measurement data in China. The emission factors from coal combustion sources with different technologies are listed in Table 1. The emission factors of other fuel types are given in Table 2.

$$EF_{SO_2} = 2 \times Cs \times P \times 1000 \quad (2)$$

Where, Cs is the percentage of fuel sulfur content; P is the conversion efficiency from sulfur to SO_2.

2.3.2 Non-combustion sources in industry

Production processes including cement processing, iron and steel manufacturing, oil refining, etc. are other important emission sources in the industrial sector. This study uses the emission factor method shown in Eq. (1) to calculate non-combustion emissions in industrial process. Owing to the lack of local studies in China, the emission factors were mainly cited from the research findings of the United States or Europe. Table 3 lists the emission factors and their references of non-combustion industrial process sources.

2.3.3 Road transport emission sources

The study surveyed the numbers of each vehicle type from the cities' statistical yearbooks and estimated the annual travel mileages by gasoline and diesel consumption in the transport sector. The IVE model (ISSRC, 2004) was used to calculate vehicle emissions during the phases of driving, cold start and VOCs evaporation. According to the model needs, we classified the vehicle fleet into light-duty car, light-duty truck, taxi, urban bus, heavy-duty bus, heavy-duty truck, and motorcycle. To prepare the fleet files

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of each city, detailed information about vehicle technology, fuel type, emission standard, and vehicle age was surveyed in the representative cities of Nanjing, Hangzhou, and Shanghai. Other input data on driving cycle, fuel quality, and environmental conditions in the study were obtained from existing studies in the representative cities of Shanghai and Hangzhou (Guo et al., 2007; Wang et al., 2008). Some real world test data of heavy-duty diesel vehicles were used to adjust the emissions factors calculated by the model (Chen et al., 2007).

Road dust emission is another important source of pollution from the transport sector. The approach to estimate road dust emission is adopted from USEPA (2002), mainly related to such parameters as average weight of the vehicles traveling the road, road surface silt loading, and a particle size multiplier.

2.3.4 Fugitive VOCs emission sources

The fugitive VOCs emission sources considered in this study mainly consist of domestic paint use for building and furniture, domestic solvent use, and oil/gas dissipation. The oil/gas dissipation includes the volatilization and leakage of oil/gas during storage and transport, liquid loading losses, tank breathing losses, and vehicle refueling operation losses in gas stations (Bo et al., 2008). The activity data of each source comes from the statistical yearbooks of the cities, and the emission factors and their references are listed in Table 4.

2.3.5 Anthropogenic ammonia sources

The emissions from the typical anthropogenic ammonia sources including livestock feeding, N-fertilizer application, sewage treatment, waste landfills, and human discharge are considered, based on the cities' annual statistical data. The emission factors of each source type are listed in Table 5.

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2.3.6 Biomass burning

Biomass burning emissions in this study come from household use and crop residue burning. The burning mass is estimated based on the statistical data of crop production, residue/crop ratio, and the percentage of burning. The emission factors for these sources are taken from Cao et al. (2005).

2.4 Speciation of VOC emissions

VOC emissions were broken down to the chemical compositions based on the species profiles of various emission sources. Sources were categorized to coal burning, petrochemical industry, coking industry, paint and solvent use, gasoline fueled vehicle, diesel fueled vehicle, gasoline evaporation, and biomass burning. Some local experimental results of coking industry were adopted to determine the source profiles. However, most of the source profile data were cited from the work of Liu et al. (2008), Wang et al. (2008) and Yuan et al. (2010).

In order to evaluate the impact of the VOC species to ozone formation, ozone formation potentials (OFP) of each VOC species was calculated by multiplying the emission by its corresponding maximum increment reactivity (MIR) factor (Cater, 1994). The total OFP of each source can be calculated by summarizing all of the OFPs of individual VOC species, as shown in Eq. (3).

$$\mathrm{OFP}_i = \sum_{j=1} E_{ij} \times \mathrm{MIR}_j \quad (3)$$

Where, OFP_i is the ozone formation potential of source i, E_{ij} is the VOC emission of source i, MIR_j is the maximum increment reactivity for the jth chemical species.

2.5 Methodology for emission allocation

The method for the regional emission allocation in the study depends on the source type and its spatial characteristics. For the power plants, boilers, furnaces or the other

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stationary fuel combustion facilities, the emissions were spatially allocated into the appropriate grid cells based on their longitude and latitude. Other small distributed industrial sources and non-combustion emissions were treated as area sources. The emissions were firstly estimated on the county-scale, and then allocated to grid cells according to 1 km×1 km resolution level GDP distribution data in China (RESDC, 2003). The other anthropogenic emissions are deemed to be related to the distribution of population density. Therefore, these kinds of emissions were spatially allocated into grid cells based on 30″×30″ resolution gridded population densities from 2006 LandScan Asia Population data (ORNL, 2007).

3 Results and discussion

3.1 Air pollutant emissions and distributions

The anthropogenic emission inventories in the sixteen cities of the YRD region for the year 2007 are summarized in Table 6. The total emissions of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, and NH_3 were 2391.8 kt, 2292.9 kt, 6697.1 kt, 3115.7 kt, 1510.8 kt, 2767.4 kt, and 458.9 kt, respectively. Shanghai, Suzhou, Wuxi, Ningbo, Hangzhou, and Nanjing are the six largest contributors, accounting for 65%, 62%, 56%, 57%, 71%, and 38% of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, and NH_3 emission. It is worthwhile to mention that flue-gas desulfurization (FGD) projects are being implemented to the power plants in the YRD region during the national 11th Five-Year Plan (2006–2010). By the end of 2007, about 40% of power plants had accomplished the installation of FGD units. The units achieved a SO_2 emission reduction of approximately 153 kt, assuming their removal efficiencies reached 90%. Another 79 kt SO_2 emission could be expected when the rest FGD installation were finished before the end of 2010.

Figure 2 illustrates the spatial allocation of emissions and relative concentrations of SO_2, NO_2, and PM_{10} (concentrations of Shanghai are equivalent to be 1) in the cities of YRD region. The annual average concentrations of SO_2, NO_2, and PM_{10} came

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from the annual statistical data of the monitoring network in each city (JSEPB, 2008; ZJEPB, 2008; SEPB, 2008). Figure 2a–e show that the highest emission intensities of SO_2, NO_x, PM_{10}, $PM_{2.5}$, and VOCs are located along the banks of the Yangtze River and the Hangzhou Bay. The area along the southern banks of the Yangtze River has a lot of industrial zones for manufactory, steel production, and chemical production, etc. The major emission sources of the Hangzhou Bay are oil refining and its downstream industry. High emission intensities in the city center of Shanghai, Hangzhou, Nanjing, and Wuxi mainly come from on-road vehicles. As NH_3 emission mainly comes from the agriculture sector, high NH_3 emission intensity is distributed in the cities of Nantong, Jiaxing, Yangzhou, Taizhou, and the suburban area of Shanghai, as shown in Fig. 2f. The emission distributions in the YRD region are reasonably correlated with SO_2, NO_2, and PM_{10} annual concentrations as shown in Fig. 2g–i. Relatively, PM_{10} concentration seems more homogeneous in the YRD region, which implies that regional aerosol pollution is mainly induced by secondary reaction of multi-pollutants in the whole region.

3.2 Emission contributions by source categories

Figure 3 shows the emission contributions of more than 20 source categories in the YRD region. It is shown that power plant is the major pollution source of the primary pollutants, which occupies 46%, 59%, 21%, and 28% of the total SO_2, NO_x, PM_{10}, and $PM_{2.5}$ emissions. The SO_2 emission contribution of power plants will be reduced to about 19% when the FGD projects are accomplished in 2010. The non-combustion industrial process including iron and steel process, refinery process, paint and solvent use is the major source of fugitive dust and VOC, taking up 58%, 54%, 68% of PM_{10}, $PM_{2.5}$, and VOC emissions. Other VOC emission mainly comes from domestic use of paint and solvent, gasoline evaporation, and biomass burning, contributing about 20%, 2%, and 3% of the total. On-road vehicle contributes 12.3% and 12.4% of the total NO_x and VOC emissions. The NH_3 emissions from livestock feeding and N-fertilizer application take up 48% and 40% of the total, respectively. According to this inventory, the contributions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, VOC, and NH_3 emission for biomass

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burning are only 0.1%, 0.7%, 1.6%, 2.6%, 3.5%, and 2.2%. But it is necessary to emphasize that the crop residue burning emissions are usually concentrated in the spring harvest period during May to June and the autumn harvest period during September to October, which and frequently result in the heavy pollution episodes in the YRD region.

To better understand the emission level in the YRD region, we select another developed city cluster in southern China named the Pearl River Delta (PRD) region (including Guangzhou, Hong Kong, etc.) to compare their emission intensities of SO_2, NO_x, PM_{10}, $PM_{2.5}$, VOCs, NH_3, and source contributions as shown in Fig. 4. The emission data come from the 2006 PRD emission inventories (Zheng et al., 2009b; Yin et al., 2010). The emission intensities of SO_2, NO_x and VOCs in the YRD and PRD regions are all relatively high, reached (2.2–2.5) t km^{-2} and (1.8–2.2) t km^{-2}, respectively. However, the PM_{10} and $PM_{2.5}$ emission intensities in the YRD region are more than 2.5 times the values in the PRD region. Primary particulate matters from power plant and other industry sectors could be the major contributor which needs to be focused on the air pollution control strategies in the YRD region. In contrast, road transport in the PRD region contributes more (about 36% and 53% of NO_x and VOCs emissions) than that of YRD region (about 12.3% and 12.4% of NO_x and VOCs emissions).

3.3 VOCs species emissions and ozone formation potentials

The percentages of alkanes, alkenes, and aromatics in the total VOC emission are 44%, 11%, and 36%, respectively. However, an aircraft measurement shows the proportion of ambient VOC concentration is 52%, 16%, and 17% in the YRD region (Geng et al., 2009). Our results seem to overestimate the content of aromatic. The main reason for the difference is believed to be that the aircraft measurement was mainly conducted in the rural region and most of the aromatics were consumed for the ozone formation during transportation in the downwind. Figure 5 shows the largest 10 species of ozone formation potential and their relative emission in the YRD region in 2007. The figure illustrates that ethylene, mp-xylene, o-xylene, toluene, and 1,2,4-trimethylbenzene, 2,4-dimethylpentane, ethyl benzene, propylene, 1-pentene, and isoprene are the key

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species to the formation of secondary air pollution. These 10 species contribute 77% to the total OFP and 49% to total VOC emission in the region.

Figure 6 shows the source contributions of the largest 10 species and total OFP in the YRD region. The emissions of ethylene, toluene, and 1-pentene mainly come from chemical industry, contributing about 92.8%, 42.2%, and 42.0%, respectively. Paint and printing is the major sources of mp-xylene, o-xylene, and ethyl benzene, accounting for 42.5%, 61.1%, and 41.9%, respectively. Biomass burning takes up 45.8% and 54.3% of propylene and isoprene emissions. Gasoline vehicle contribute 20.0%, 36.3%, 11.5%, and 11.2% of mp-xylene, 1,2,4-trimethylbenzene, propylene, and isoprene emission.

In the terms of OFP, chemical industry, domestic paint and printing, gasoline vehicle, industrial coating, and oil refinery are the five major sources to the ozone formation, accounting for 38.2%, 23.9%, 11.6%, 9.6%, and 4.2%, respectively. The VOC emission contributions of these sources are about 34.5%, 21.7%, 9.7%, 18.8%, and 6.0%, respectively. The results shows the ozone formation contributions of the sources are not always correlated with their emission values in the YRD region. It is worthwhile to mention that gasoline vehicles are responsible for 9.7% of VOC emissions, as well as 11.6% for ozone formation.

3.4 Spatial characteristics of total OFPs

Figure 7 shows the spatial allocation of the OFPs in the YRD region. It is shown from the figure that OFPs are mainly concentrated in the well developed urban and industrial areas along the Yangtze River and around the Hangzhou Bay, which is similar with the distribution of NO_x emission intensity and other pollutants in Fig. 2a–e. Nevertheless, according to the real world observation studies in the YRD region (Cheung et al., 2001; Geng et al., 2009), high ozone concentration are not expected to occur in these urban areas with high OFP value. The NO_x emission, topographic feature, and meteorological natures in various seasons are all important factors to the actual ozone formation in the region. Further photochemical modeling studies are necessary to be conducted to

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reveal the mechanism of ozone formation and the contributions of precursor emissions in the YRD region. In this case, the study provides a high resolved emission inventories covering the primary air pollutants and VOC species to the detailed photochemical modeling studies, which will be helpful to design efficient emission control strategies for the improvement of the regional ozone and secondary aerosol pollution in the YRD region.

3.5 Assessment of uncertainty in the emission inventory

The YRD regional emission inventory was compiled by bottom-up approach based on the local surveys of major air pollution sources in the region. Most of the activity data including fuel consumption, sulfur content, ash content, exhaust treatment efficiency and geographic location of industrial sources were collected from the local pollution source census in the cities of the YRD region. However, most of the emission factors cited in this study are still considerable uncertainties to the inventory due to the lack of local measurement data. Relatively, the SO_2 and NO_x emission factors are relatively convincing because of their solid researches in China (Zhang et al., 2007). Vehicle emissions in this study are based on local research in the YRD region, which has proved to be comparable with the real world measurement data (Wang et al., 2008). However, the emission factors of PM_{10}, $PM_{2.5}$, VOCs and NH_3 were mainly taken from the research findings of the United States and Europe, which posed high uncertainties with respect to the YRD region's real situation.

There are still some sources not considered in this study, which are off-road emission sources, including locomotive, shipping, aircraft, and construction machinery, dust emissions from yards and construction sites. The lack of these sources will cause a slight underestimate in the emission inventory. In addition, many studies have confirmed the impact of biogenic source to the air quality. Our study did not include the biogenic emissions in the YRD region, while it can be estimated that the biogenic VOC emission almost occupies 23% of the total in the summer and 3% of the total VOC emission in the winter if we use the GEIA natural VOC emission database in 1990.

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The 2006 INTEX-B Asian emission inventory (Zhang et al., 2009) is compared to the emission inventory after compilation. The result shows that the difference of SO_2, NO_x, PM_{10} and VOCs emissions between the relative grids of both studies are about −3.8%, 7.0%, 58.1% and 23.8%, respectively. SO_2 and NO_x emission have relatively high agreement, while the uncertainties of PM_{10} and VOCs emission in both inventories are all higher than the others. The correlation coefficients of the grids are about 0.81, 0.80, 0.78 and 0.79, implying the similar spatial distribution patterns of emissions for the two studies.

4 Conclusions and recommendations

A regional emission inventory for the YRD region for the year 2007 was compiled based on local source information, urban energy consumption, vehicle composition, as well as other information on the industry, commercial and agriculture sectors. It is estimated that the emissions of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs and NH_3 in the YRD region account for 2391.8 kt, 2292.9 kt, 6697.1 kt, 3115.7 kt, 1510.8 kt, 2767.4 kt, and 458.9 kt, respectively. The percentages of alkanes, alkenes, and aromatics in the total VOC emission are 44%, 11%, and 36%, respectively. Ethylene, mp-xylene, o-xylene, toluene, and 1,2,4-trimethylbenzene, 2,4-dimethylpentane, ethyl benzene, propylene, 1-pentene, and isoprene are the key species which contribute 77% to the total OFP and 49% to the VOC emission in the whole region. The air pollutant emissions and OFPs in the YRD region were mainly concentrated in the urban and industrial areas along the Yangtze River and around the Hangzhou Bay, where the conventional air pollutant concentrations are relatively high.

The industrial sources including power plants, other fuel combustion facilities, and non-combustion processes are the major contributors to in the YRD region, generally account for 97%, 86%, 89%, 91%, and 69% of the total SO_2, NO_x, PM_{10}, $PM_{2.5}$, and VOC emissions. On-road vehicles in the YRD region contribute 12.3% of the NO_x emission and 12.4% of the VOC emission. The livestock feeding and N-fertilizer application

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source take up 48% and 40% of the total NH_3 emission, respectively. In addition, chemical industry, domestic paint and printing, gasoline vehicle, industrial coating, and oil refinery are the five major sources to the ozone formation, accounting for 38.2%, 23.9%, 11.6%, 9.6%, and 4.2%, respectively.

NO_x and VOC emissions are the key precursors required to be reduced to improve the regional ozone and secondary aerosol pollution. However, neither of the pollutants is regulated currently in China. Therefore, more targeted air pollution control measures on reducing NO_x and VOC emissions are needed to be put forward in the national or regional planning. Based on the emission inventory in the YRD region, power plants and vehicles should be taken into accounts to reduce the NO_x emission. In this case, some control measures such as denitrification of power plants, universally upgrading new vehicle emission standard and fuel quality (Shanghai has implemented Euro IV standard in 2009), should be considered in the next stage for the cities of the YRD region. VOC emissions are more complicated to be controlled. The control measures of supervising the fugitive VOC emission in the refining, chemical, and other industries, gasoline vapor recovery, and the use of water-based paint are expected to reduce the total VOC emissions in the YRD region. In addition, considering most of the gasoline vehicle emissions are concentrated in urban areas and have strong reactivity to ozone formation, some corresponding measures such as accelerating the phase out of old vehicles and strengthening vehicle inspection and maintenance would be more effective to the mitigation of ozone concentration.

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Table 1. Emission factors of coal combustion sources.

Source	Technology	Emission factor ($kg\,t^{-1}$) NO_x (Zhang et al., 2007)	CO (Wang et al., 2005)	PM_{10} (Zhang, 2005)	$PM_{2.5}$ (Zhang, 2005)	VOCs (Bo et al., 2008)
Power plant	< 100 MW, without LNB	10.50				
	100~300 MW, without LNB	8.85				
	100~300 MW, with LNB	5.85				
	> 300 MW, with LNB	5.55				
	stoker furnace		8.00	13.88	5.25	
	pulverized coal boiler		2.00	46.00	12.00	
	other					0.15
Boiler	hand-feed stoker furnace	3.80	124.00	5.40	1.89	
	automatic stoker furnace	4.00	15.00	5.40	1.89	
	fluidized-bed furnace	7.50	2.00	28.08	5.40	
	other					0.18
Kiln	shaft kiln	1.70	155.70			
	New-dry process	15.30	17.80			
	other rotary kiln	18.50	17.80			
	lime kiln	1.70	155.70			
	brick kiln	4.70	150.00			
	other			5.40	1.89	0.18
Commercial use		3.75	75.00	3.50	2.00	0.60
Residential use		1.88	75.00	8.82	6.86	0.60

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Table 2. Emission factors of other fuel combustion sources in industry.

Pollutant	Source	Coal gas ($g\,m^{-3}$)	Natural gas ($g\,m^{-3}$)	LPG ($kg\,t^{-1}$)	Diesel ($kg\,t^{-1}$)	Fuel oil ($kg\,t^{-1}$)	Reference
NO_x	power plant		4.10		7.40	10.06	Tian (2003)
	Other industry	1.35	2.09	2.63	9.62	5.84	Tian (2003)
	residential use	0.74	1.46	1.58/0.88	5.77	3.50	Tian (2003)
CO	–	1.30	1.30	0.36	0.60	0.60	Wang et al. (2005)
PM_{10}	power plant		0.12		0.50	0.85	Zhang (2005)
	Other industry	0.20	0.12	0.08	0.50	1.03	Zhang (2005)
	residential use	0.20	0.12	0.08	0.50	1.20	Zhang (2005)
$PM_{2.5}$	power plant		0.12		0.50	0.62	Zhang (2005)
	Other source	0.20	0.12	0.08	0.50	0.67	Zhang (2005)
	residential use	0.20	0.12	0.08	0.50	0.90	Zhang (2005)
VOCs	power plant		0.18		0.13	0.13	Bo et al. (2008)
	Other source	0.00044	0.18	0.19	0.15	0.15	Bo et al. (2008)
	residential use	0.00044	0.18	0.19	0.15	0.15	Bo et al. (2008)

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Table 3. Emission factors of industrial process sources.

Process	Activity rate	Unit	SO_2	NO_x	CO	PM_{10}	$PM_{2.5}$	VOCs	NH_3	Refence
Vegetable oil processing	production	$kg\,t^{-1}$						2.45		USEPA (2002)
Beer manufacturing	production	$kg\,m^{-1}$						0.20		Klimont et al. (2002)
Spirits manufacturing	production	$kg\,m^{-1}$						20.00		Klimont et al. (2002)
Oil refining	crude oil feed	$kg\,t^{-1}$	1.58	0.22				2.65		Klimont et al. (2002)
Coke production	coal charged	$kg\,t^{-1}$	2.01	0.02	0.64	2.75	1.75	2.10	0.09	USEPA (2002)
Synthetic Ammonia	production	$kg\,t^{-1}$	0.03		7.90			4.72	2.10	USEPA (2002)
Sulfuric Acid	production	$kg\,t^{-1}$	7.00							USEPA (2002)
Urea	production	$kg\,t^{-1}$				1.74	1.50		9.69	USEPA (2002)
Paint production	production	$kg\,t^{-1}$						15.00		Klimont et al. (2002)
Ink production	production	$kg\,t^{-1}$						30.00		Klimont et al. (2002)
Plastic processing	production	$kg\,t^{-1}$						2.20		Bo et al. (2008)
Synthetic fibre	production	$kg\,t^{-1}$						73.40		Bo et al. (2008)
Tyres production	production	$kg\,tyre^{-1}$						0.29		Klimont et al. (2002)
Synthetic rubber	production	$kg\,t^{-1}$						15.00		Klimont et al. (2002)
Cement manufacturing	production	$kg\,t^{-1}$				8.05	3.40			Lei et al. (2008)
Bricks production	production	$kg\,t^{-1}$				0.13	0.04			Zhang et al. (2009)
Iron manufacturing	production	$kg\,t^{-1}$	0.30	0.09	40.50	0.72	0.45			USEPA (2002)
Steel manufacturing	Production	$kg\,t^{-1}$			9.00	11.78	8.62	0.06		USEPA (2002)
Steel foundries	Production	$kg\,t^{-1}$				3.58	1.70			USEPA (2002)
Ferroalloy	production	$kg\,t^{-1}$				3.61	2.79			USEPA (2002)
Aluminum products	production	$kg\,t^{-1}$				2.63	2.08			USEPA (2002)
Paint application	paint used	$kg\,t^{-1}$						730.00		Klimont et al. (2002)
Print	Ink used	$kg\,t^{-1}$						300.00		Klimont et al. (2002)
Degreasing operations	solvent used	$kg\,t^{-1}$						900.00		Klimont et al. (2002)
Solvent use	solvent used	$kg\,t^{-1}$						300.00		USEPA (2002)

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Table 4. Emission factors of fugitive VOCs emission sources.

Source	Activity rate	Unit	Emission factor	Reference
Domestic paint use	paint used	kg t^{-1}	0.36	Klimont et al. (2002)
Domestic solvent use	solvent used	kg capita^{-1}	0.20	Klimont et al. (2002)
Vehicle treatment	cars registered	kg vehicle^{-1}	1.11	Klimont et al. (2002)
Waste landfills	amount of waste	kg t^{-1}	0.23	Klimont et al. (2002)
Oil storage				
Crude oil	storage rate	kg t^{-1}	0.12	Bo et al. (2008)
Gasoline	storage rate	kg t^{-1}	0.16	Bo et al. (2008)
Underground tank filling	gasoline consumed	kg t^{-1}	1.16	Bo et al. (2008)
Tank breathing and emptying	gasoline consumed	kg t^{-1}	0.16	Bo et al. (2008)
Vehicle refueling operations				
displacement losses (uncontrolled)	gasoline consumed	kg t^{-1}	1.74	Bo et al. (2008)
Spillage	gasoline consumed	kg t^{-1}	0.11	Bo et al. (2008)

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Table 5. Emission factors of anthropogenic ammonia emission sources.

Source	Activity rate	Unit	Emission factor	Reference
Livestock				
Dairy cattle	livestock	kg N head^{-1}	21.93	Zhang et al. (2010)
Other cattle	livestock	kg N head^{-1}	7.31	Zhang et al. (2010)
Pig	livestock	kg N head^{-1}	4.61	Zhang et al. (2010)
Sheep and goat	livestock	kg N head^{-1}	2.24	Zhang et al. (2010)
Horse	livestock	kg N head^{-1}	5.66	Zhang et al. (2010)
Poultry	livestock	kg N head^{-1}	0.41	Zhang et al. (2010)
N-fertilizer application	fertilizer applied	kg kg^{-1}	16.81	Dong et al. (2009)
Sewage treatment	amount of sewage	kg m^{-3}	0.0032	Yin et al. (2010)
Waste landfills	amount of waste	kg t^{-1}	0.56	Yin et al. (2010)
Human discharge	capita	kg capita^{-1}	0.05	EEA (2006)

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Table 6. Air pollutant emission inventory of the cities in the YRD region in 2007.

Province	City	Distance to Shanghai /km	Annual air pollutant emissions/kt						
			SO_2	NO_x	CO	PM_{10}	$PM_{2.5}$	VOCs	NH_3
Jiangsu	Suzhou	85	281.7	368.6	1205.1	389.2	208.1	483.3	26.3
	Nantong	98	139.4	90.5	308.9	111.6	56.8	87.7	71.1
	Wuxi	117	261.4	183.7	488.0	347.5	167.8	191.7	26.8
	Changzhou	158	145.4	90.7	455.4	211.6	98.8	73.2	16.3
	Tai'zhou	200	18.2	52.5	148.7	64.9	31.2	62.8	32.9
	Zhenjiang	220	131.6	103.3	128.5	208.3	96.4	45.9	14.3
	Yangzhou	234	43.9	110.4	164.8	81.8	48.3	58.1	50.7
	Nanjing	272	221.0	144.2	739.7	255.9	129.7	153.2	24.1
Zhejiang	Jiaxing	85	120.9	114.3	203.3	219.7	97.4	58.0	36.7
	Huzhou	137	81.3	71.5	144.2	190.0	83.2	53.7	20.1
	Ningbo	150	254.8	211.0	284.0	211.6	111.6	150.3	25.4
	Zhoushan	157	4.2	15.7	18.6	10.4	6.1	17.9	1.7
	Shaoxing	163	135.0	105.0	213.5	196.8	88.2	287.3	21.3
	Hangzhou	164	126.2	134.5	482.5	282.0	129.3	391.6	29.6
	Taizhou	267	28.1	117.1	156.5	69.1	38.5	66.0	19.0
Shanghai		0	398.8	380.0	1555.5	265.5	119.2	586.9	42.6
Total		–	2391.8	2292.9	6697.1	3115.7	1510.8	2767.4	458.9

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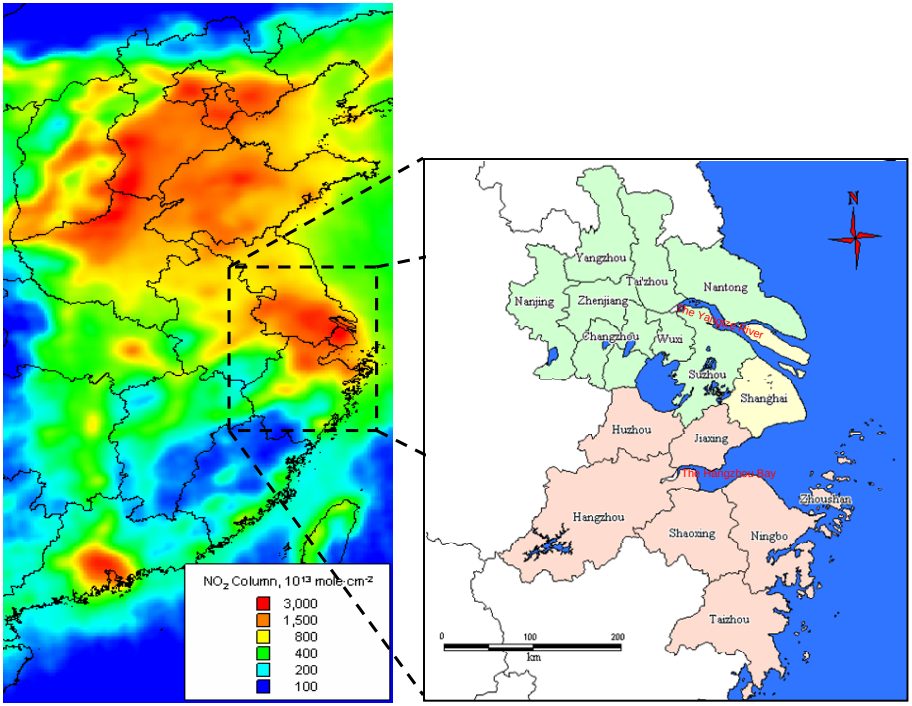

Fig. 1. Satellite observed tropospheric NO_2 concentration in 2007 (left) and research domain of the YRD region (right).

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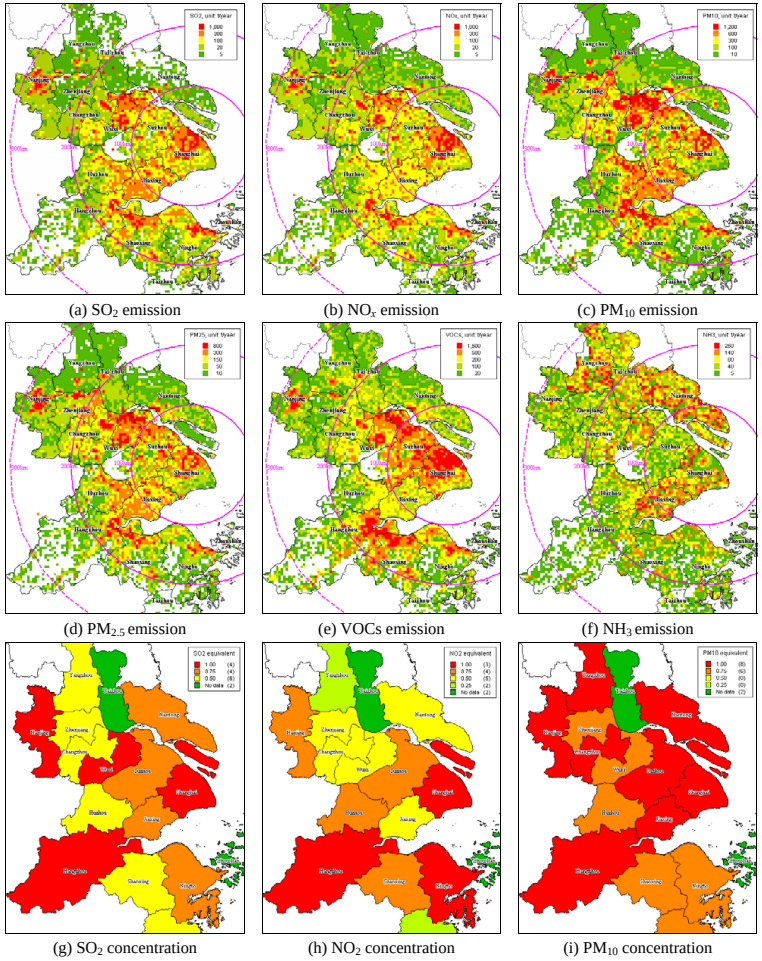

(a) SO_2 emission (b) NO_x emission (c) PM_{10} emission

(d) $PM_{2.5}$ emission (e) VOCs emission (f) NH_3 emission

(g) SO_2 concentration (h) NO_2 concentration (i) PM_{10} concentration

Fig. 2. Spatial allocation of air pollutant emissions and annual concentrations in the YRD region in 2007.

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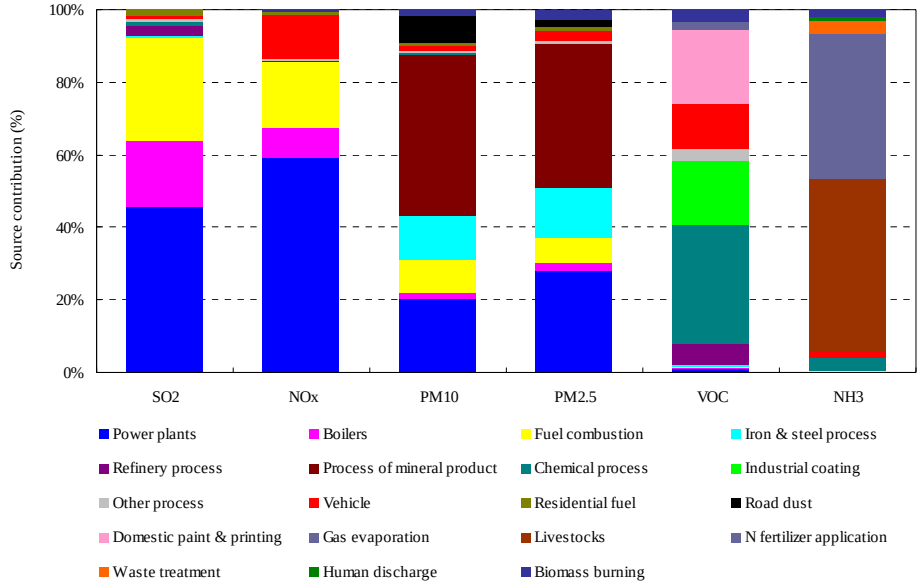

Fig. 3. Emission contribution of source categories in the YRD region in 2007.

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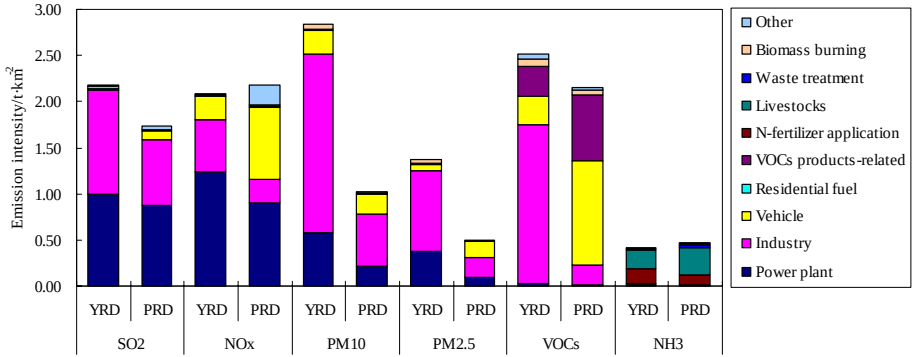

Fig. 4. Comparison of air pollutant emissions and source contributions in the YRD and PRD regions.

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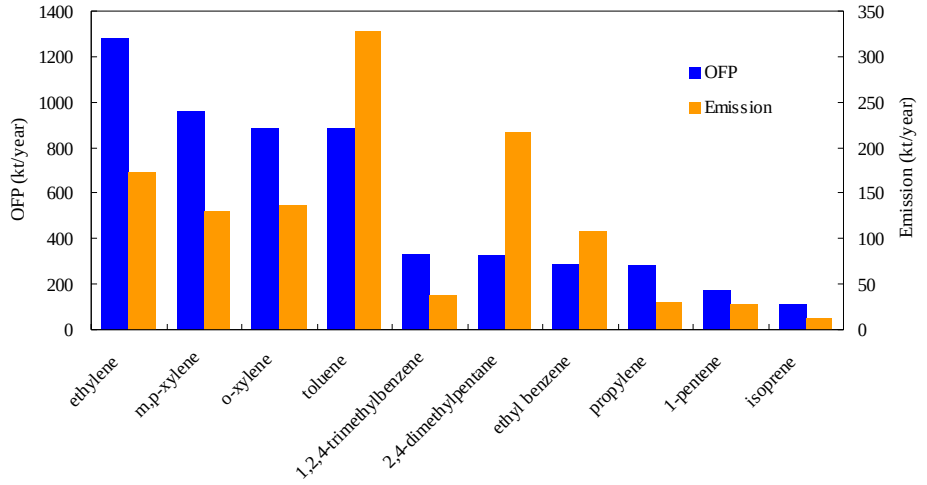

Fig. 5. Largest 10 VOC species of OFP contribution and their emissions in the YRD in 2007.

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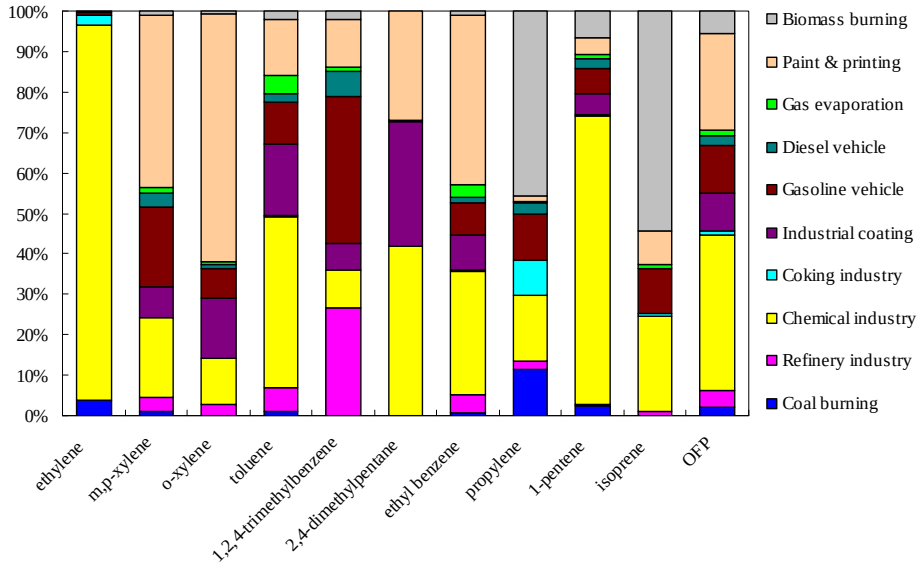

Fig. 6. Source contributions of the largest 10 VOC species and OFP in the YRD region.

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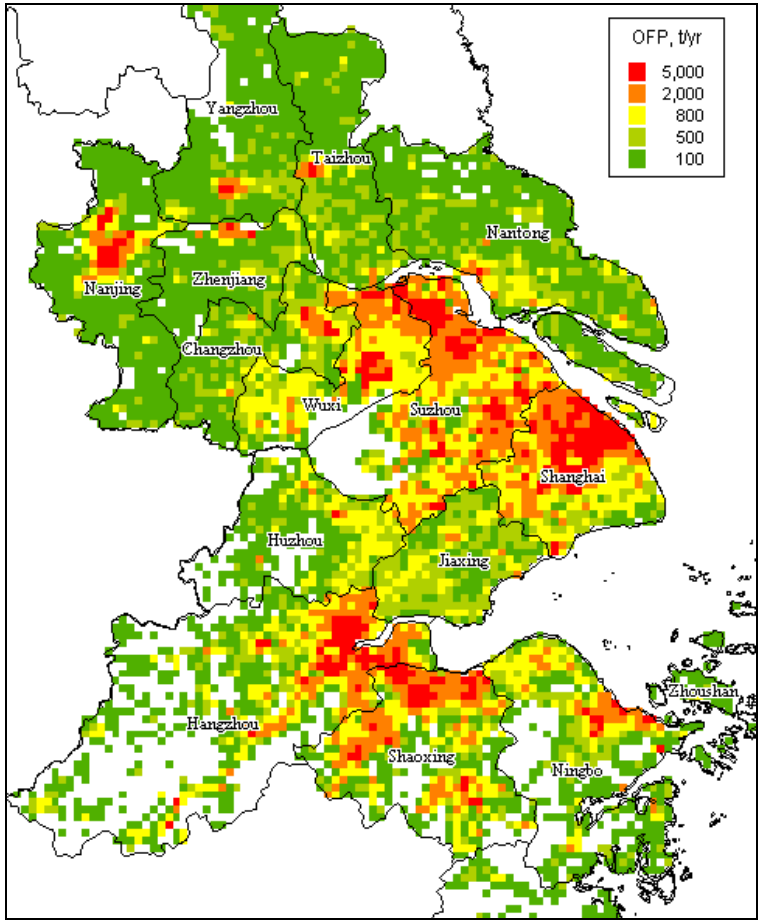

Fig. 7. Spatial distribution of the OFPs in the YRD region.

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