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Vehicular emissions in China in 2006 and 2010

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Abstract

Vehicular emissions are one of the most important sources of pollution in China, and they can increase the ambient concentrations of air pollutants and degrade the air quality. Using data released by the National Bureau of Statistics, vehicular emissions in China in 2006 and 2010 were calculated at a high spatial resolution, by taking the emission standards into consideration. The results show that China's vehicular emissions of CO, NO_x, VOCs, NH_3, $PM_{2.5}$, PM_{10}, black carbon (BC), and organic carbon (OC) were 30113.9, 4593.7, 6838.0, 20.9, 400.2, 430.5, 285.6, and 105.1 Gg, respectively, in 2006 and 34175.2, 5167.5, 7029.4, 74.0, 386.4, 417.1, 270.9, and 106.2 Gg, respectively, in 2010. CO, VOCs, and NH_3 emissions were mainly from motorcycles and light-duty gasoline vehicles, whereas NO_x, $PM_{2.5}$, PM_{10}, and BC emissions were mainly from rural vehicles and heavy-duty diesel trucks. OC emissions were mainly from motorcycles and heavy-duty diesel trucks. Euro 0 and Euro I vehicles were the primary contributors to all of the pollutant emissions except NH_3, which was mainly from Euro III and Euro IV vehicles. The spatial distribution of vehicular emissions in China in 2006 and 2010 were developed at a high resolution of $0.25^\circ \times 0.25^\circ$, by using the road traffic density to characterize the busyness of a road. This method could overcome the problem of getting traffic flow information and make the spatial allocation more closed to the actual road emissions. The results showed that vehicular emissions presented significant regional spatial distribution, and emissions in the eastern and southern parts of China were much higher than those in western and northern China in both years. The North China Plain, Yangtze River Delta, and Pearl River Delta regions jointly accounted for nearly half of the emissions. NH_3 emissions increased greatly in big cities from 2006 to 2010. Emissions of CO, NO_x, and VOCs could increase 52 %, 9 %, and 68 %, if the emission standard and oil quality remained in the Euro I stage, so the policies on vehicular emissions implemented in China were demonstrated to be effective. Nevertheless, greater efforts are needed to improve the oil quality so that the new emission standard can implement timely and catch the international level quickly, especially

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the sulfur level because NO_x emission is very sensitive to it. By comparing with coal consumption and NO_2 column density observed by SCIA satellite, the increase of NO_x emission in China from 2006 to 2010 was mainly caused by coal consumption.

1 Introduction

As China's economy has developed, the total number of civilian motor vehicles has surged from 14.5 million in 1999 to 78.1 million in 2010, an increase of 437.0 % in 12 yr. Especially in recent years, the annual growth rate of civilian vehicles has been approximately 20 % (National Bureau of Statistics, 1999–2010). This increase in motor vehicles has caused vehicle use to be an important contributor to China's air pollution. In 2004, half of the NO_x in Chinese cities was emitted from motor vehicles, and vehicles were considered one of the main sources of China's air pollution by the State Environmental Protection Administration of China (2004). The northern and southeastern coasts of China, which represent a small share of the Chinese territory but include many of its vehicles, produce a substantial amount of pollution per unit area, aggravating the regional air pollution. Among the pollutants emitted by motor vehicles, black carbon (BC), organic carbon (OC), $PM_{2.5}$, and PM_{10} can lower visibility by the extinction process. CO, NH_3, VOCs, and NO_x are important precursors of secondary aerosols and O_3 because they can generate a series of photochemical reactions once they reach a certain concentration in the atmosphere. These reactions may have a great influence on regional visibility, accelerate the formation of haze, and exacerbate the photochemical pollution problem (Zhang et al., 1998). To satisfy the Euro III emission standards for motor vehicles, newly registered motors vehicles must install three-way catalytic converters (TWCs); however, these devices can lead to NH_3 emissions while reducing CO, NO_x, and VOCs through redox reactions (Heeb et al., 2006a; Kean et al., 2009). NH_3 in the atmosphere is highly reactive, and heterogeneous atmospheric reactions can readily generate nitrates and sulfates, which are the main components of $PM_{2.5}$ (Bouwman et al., 1997; Goebes et al., 2003). Strong hazy pollution affected the

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Beijing–Tianjin–Hebei (BTH) region five times in January 2013, and vehicular emissions made significant contributions to this pollution (Wang et al., 2013). Therefore, calculating vehicular emissions is a convenient way to describe the characteristics of regional vehicular pollution in China and can supply reliable data for regional air quality modeling, pollution prevention, and policy-making.

In the 1990s, a number of foreign scholars developed pollutant inventories for East Asia, with China, as a dominant country, as a primary focus. Vehicular emissions were considered as well. Van Aardenne et al. (1999) used the Regional Air Pollution Information and Simulation (RAINS-ASIA) methodology to calculate the energy consumption over the next 30 yr based on 1990 data and then calculated Asia's NO_x emissions from 1990 to 2020. The results showed that NO_x emissions from motor vehicles in China accounted for 7 % of the total NO_x emissions in 1990, and the share would increase to 33 % in 2020. The emissions profiles of SO_2, NO_x, CO, and BC for China for the years 1995 and 2020 were presented by Streets et al. (2000, 2001) as a component of NASA's (the National Aeronautics and Space Administration) China-MAP program, and the authors suggested that motor vehicles were the main source of NO_x and CO. In addition, during the studies of the Transport and Chemical Evolution over the Pacific (TRAC-P) mission funded by NASA and in the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) funded by the National Science Foundation (NSF) and the National Oceanic and Atmospheric Administration (NOAA), Streets et al. (2003) improved their method incrementally by dividing the motor vehicles into categories and taking total annual mileage driven into consideration. They then developed an inventory of air pollutant emissions in Asia in the year 2000. The spatial distribution of the inventory was showed at a resolution of $1^\circ \times 1^\circ$. The global emissions inventory EDGAR (Emission Database for Global Atmospheric Research) also included vehicular emissions (Olivier et al., 1999; Olivier et al., 1998). T. Ohara et al. (2007) developed a new Asian emissions inventory called REAS (Regional Emission inventory in Asia) that covered the 40 yr from 1980 to 2020, and vehicle classification, traffic volume, and fuel ratio information were taken into consideration in this work. For further

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study, gridded emissions in Asia were given at a resolution of 0.5° × 0.5°. In order to update the TRAC-P inventory to reflect the extremely rapid economic growth in Asia since 2000, the Intercontinental Chemical Transport Experiment-Phrase B (INTEX-B) was conducted by NASA. SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, and OC emissions from motor vehicles were estimated in detail for 2006 (Zhang et al., 2009). Compared with the previous studies, INTEX-B not only took the vehicle classifications and fuel ratio into consideration but also incorporated technological innovation when calculating vehicular emissions in China, and the results showed that vehicular emissions increased markedly from 2001 to 2006. This inventory has received widespread application because of its substantially improved accuracy compared to its predecessors.

Because vehicles play an important role in the air pollution of China, studies on vehicular emissions were carried out in China. Li et al. (2003) calculated the emission factors and emissions of ten vehicular pollutants in 1995, including VOCs, CH_4, CO, NO_x, CO_2, SO_2, Pb, PM_{10}, and N_2O, based on the MOBILE5 model and fuel consumption. The results showed that the vehicular emission factors in China were several times higher than in countries of the same economic development level, and vehicular emissions had climbed rapidly because of the sustained growth in the number of motor vehicles. Song and Xie (2006) established the Chinese inventories of vehicular emissions with a resolution of 40 km × 40 km using GIS in 2002, based on the statistical data from yearbooks and the emission factors of each vehicle category in each province calculated by the COPERT III program. Their results showed that motorcycles and light-duty gas vehicles contributed greatly to the emissions and that the emissions in the Beijing–Tianjin–Hebei (BTH) region, Yangtze River Delta (YRD), and Pearl River Delta (PRD) were much higher than in other regions of China. Multi-year inventories of vehicular emissions at a high spatial resolution of 40 km × 40 km in China were established by Cai and Xie (2007) using the same method for the period 1980–2005, and they analyzed the correlation coefficients between emissions and GDP. Yao et al. (2012b) calculated the vehicular emissions inventories from 1990 to 2009 for 12 typical cities in China and analyzed the vehicular emissions trends in each city. To address the de-

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fects in traditional methods of spatial allocation, Zheng et al. (2009b) proposed a new approach, named "standard road length", that makes use of GIS-based road network information and traffic flows to help allocate the vehicular emissions into grid cells. This approach was demonstrated to be practicable in the spatial allocation of the regional vehicular emissions in the PRD in 2004. Subsequently, another high-resolution emissions inventory for the PRD region in 2006 was developed by similar methods (Zheng et al., 2009a). Lang et al. (2012) developed multi-year emissions inventories for NO_x, CO, VOCs, and PM_{10} in the BTH region for the period 1999–2010 and discussed the effects of vehicle category, fuel ratio, and emission standards. Che et al. (2009) developed the vehicular emissions inventories of PRD in 2006 and thoroughly analyzed the emission contributions of different vehicle categories and fuel types. Fu et al. (2013) developed a high-resolution emission inventory of primary air pollutants for YRD region, including Shanghai and 24 cities in Zhejiang and Jiangsu. Cao et al. (2011) took the traffic source into account when calculating the emission of primary particles and pollutant gases in China in 2007, and the national emissions were gridded with resolution of $0.5^\circ \times 0.5^\circ$ for the air-quality models. Other scholars have also developed municipal-level vehicular emissions inventories (Fu et al., 2000; He, 2011; Li et al., 2010; Ma, 2008; Wang et al., 2012; Yao et al., 2012a; Yu, 2007; Zhang, 2005).

In summary, the target years of the existing vehicular emissions inventories are almost from 1999 to 2007. However, there have been dramatic changes in China in recent years, particularly in vehicle ownership and emissions standards. Therefore, much of the published research is not applicable to the current situation in China. In addition, the difference in emissions factors among vehicles with different emission standards (from Euro 0 to II in 2006 and to Euro IV in 2010) was often overlooked in previous studies. As a result, the emissions from the vehicles registered earlier were underestimated. In addition, some vehicular pollutants were not considered, especially the NH_3 emissions, which have been a focus of many foreign researchers (Durbin et al., 2002; Heeb et al., 2006a, b, 2008; Kean et al., 2009; Perrino et al., 2002) but few in China (Yang, 2011; Yao et al., 2011b; Yin et al., 2010). Finally, most of the foreign studies were

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for all of Asia, which may lead to great uncertainty for the lack of detailed information of China. However, studies within China have often examined small regions, such as the BTH or PRD regions, or cities; studies at these scales cannot describe China's vehicular emissions systematically or satisfy the requirements for numerical simulations because of their low resolution. For these reasons, developing a new, complete, and high-resolution vehicular emissions inventory is necessary.

To study the characteristics of vehicular emissions, meet the needs of the regional air-quality simulation, and provide a reference for policy-making, the vehicular emissions of CO, NO_x, VOCs, NH_3, $PM_{2.5}$, PM_{10}, BC, and OC in China in 2006 and 2010 were estimated in the present study. The vehicle emissions standards, categories, and fuel types were incorporated to improve the accuracy and reliability of the findings. The spatial distributions of vehicular emissions for the whole China were examined at a resolution of $0.25^\circ \times 0.25^\circ$. For further study, the emissions calculated in this work were compared with the existing inventories, and the results of the spatial distributions were compared with the fuel consumption and NO_2 column density observed by satellite. The effects of emission standards and oil quality improvement were analyzed. The Monte Carlo method was used to calculate the uncertainties associated with the emissions.

2 Data and methodology

2.1 Emissions calculation method

2.1.1 Calculation formula

Vehicular emissions of CO, NO_x, VOCs, NH_3, $PM_{2.5}$, PM_{10}, BC, and OC in different provinces of China were calculated based on emission factors, average annual vehicle traveled mileages (VTM), and vehicle population of various vehicle types and emissions

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standards using the following formula:

$$E_{m,n} = \sum_{j} \sum_{i} (P_{m,i,j} \times \mathrm{VTM}_{i,j} \times \mathrm{EF}_{i,j,n}) \times 10^{-6} \quad (1)$$

where m represents the province; n represents the pollutants considered in this study (i.e., CO, NO_x, VOCs, NH_3, $PM_{2.5}$, PM_{10}, BC, and OC); i represents the type of vehicle; j represents the national vehicular emission standards (from Euro 0 to Euro IV); $E_{m,n}$ represents emissions (tons) of pollutant n in province m; $P_{m,i,j}$ represents the population of type i vehicle with emissions standard j in province m; $\mathrm{VTM}_{i,j}$ represents the average annual traveled mileage (km) of type i vehicle with emission standard j in province m; and $\mathrm{EF}_{i,j,n}$ represents the emission factor ($\mathrm{g\,km^{-1}}$) of pollutant n emitted from type i vehicles with emission standard j.

2.1.2 Activity data research

(1) Vehicle population

We classified vehicles into light-duty gasoline vehicles (LDGVs), heavy-duty gasoline vehicles (HDGVs), light-duty gasoline trucks (LDGTs), light-duty diesel vehicles (LDDVs), heavy-duty diesel vehicles (HDDVs), light-duty diesel trucks (LDDTs), heavy-duty diesel trucks (HDDTs), rural vehicles (RVs), and motorcycles (MCs). The original data for each vehicle class in a province were obtained from the official statistical yearbooks (National Bureau of Statistics, 2006, 2010). However, there is no information about the vehicle population classified by fuel types, which have great influence on emission factors. To address this problem, an investigation of fuel ratios was conducted based on the production of vehicles by fuel type for the period 2002–2010 from the Automotive Industry Yearbook (China Automobile Industry Association, 2002–2006). We then took the average annual fuel ratio as the fuel ratio for this study, as shown in Table S1. Because there were no data on the RVs and MCs population, we assumed. Because there were no data on the RVs and MCs population, we assumed that the RVs pop-

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ulation in 2010 was approximately the same as in 2006, and the MCs population was calculated based on the MCs ownership per 100 households and the household data from the official statistical yearbook.

To mitigate vehicular emissions, several emissions standards (from Euro I to Euro IV) have been implemented over the past decade in China, so the emission factors of vehicles with different emission standards differ from one another. For this reason, it is necessary to estimate the population of vehicles with various emission standards. We assumed that after a new emission standard was introduced, newly registered vehicles must be in compliance with that standard. Therefore, the vehicle population by emission standards can be calculated by knowing the total population and the population of newly registered vehicles. Details are shown in the following formula:

$$P_{m,i,j} = \begin{cases} \sum_{\text{ys}} N_{m,i,j,\text{ys}} & j \neq \text{Euro0} \\ P_{m,i,\text{total}} - \sum_{j=1}^{4} \sum_{\text{ys}} N_{m,i,j,\text{ys}} & j = \text{Euro0} \end{cases} \tag{2}$$

where ys represents the years when the emission standard j was implemented. $N_{m,i,j,\text{ys}}$ represents the population of newly registered vehicles of type i and emission standard j in year ys in province m. $P_{m,i,\text{total}}$ represents the total population of vehicle type i in province m for the target calculation year.

It should be noted that if the calculated population of Euro 0 is negative, and then the value will be set to 0, and the population of Euro I will be modified accordingly. The total numbers of vehicles with each emission standard in 2006 and 2010 were listed in Tables S2 and S3.

(2) Average annual vehicle traveled mileages

The average annual vehicle traveled mileages are required for estimating the final emissions. These data were not directly available through official statistical records, so we

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adopted the results from a previous study (Liu et al., 2008). Details are shown in Table 1.

2.1.3 Emission factors

Systematic studies of vehicular emission factors are rare in China, and most studies have been limited to a city area; thus, there is no complete and suitable emission factor database for all of China. We obtained the emission factors needed in our study through a comprehensive literature search and a thorough analysis.

CO, NO_x, VOCs, and PM_{10} were obtained from the results of Beijing Municipal Science and Technology Project (2012), which were calculated by the COPERT IV module recently, and we used the emission factors published by the Vehicular Emission Control Center of Chinese State Environmental Protection Administration (SEPA) as a complement (2005). However, the Euro 0 vehicle emission factors for these two results were substantially different; thus, the CO, VOCs, and NO_x emission factors of Euro 0 vehicles were from Street et al. (2006) and Fu et al. (2001). For the NH_3, $PM_{2.5}$, BC, and OC emissions, little research has been conducted in China. NH_3 emission factors were obtained from the Emission Inventory Improvement Program of the US EPA (Environmental Protection Agency of US) (2005); $PM_{2.5}$, BC, and OC emission factors were calculated according to the fraction of these three particulate matters in relation to the total PM_{10} (Streets et al., 2001). The RVs in our study were all assigned the same emission factors because few research was conducted on it (Yao et al., 2011a). All of the emission factors are listed in Table 2.

2.2 Spatial distribution method

Zheng et al. (2009b) proposed a GIS-based method for the spatial allocation of mobile source emissions named “standard road length”, which is based on the road network and traffic flow information. However, traffic flow information is unavailable. To use this method successfully, we used the passenger and cargo turnover volume and the length

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of transport routes from the China Statistical Yearbook (National Bureau of Statistics, 2006, 2010). to calculate the road traffic density, which was used to characterize the busyness of a road. The specific methods are as follows:

(1) Develop the county-level vehicular emission inventory

The city-level inventory should be calculated first, combined with the proportions of vehicles for each city from the China Statistical Yearbook (National Bureau of Statistics, 2006, 2010). after the vehicular emissions inventory of each province is calculated using Eq. (1). Then, the sum of the values of secondary and tertiary industries of each county, which was from the China County Statistical Yearbook were used to complete the county-level vehicular emissions inventory (Investigeion General Team of Rural Economic Society in State Statistical Bureau, 2006, 2010).

(2) Calculate the road traffic density for each grade of road

The road traffic density is used to replace the traffic flow to characterize the busyness of the roads and can be calculated using Eq. (3):

$$F_m = \frac{T_m}{\mathrm{TL}_{m,\mathrm{total}}}; F_{m,k} = F_m \times P_{m,k} \tag{3}$$

where k represents the grade of the road; F_m represents the average road traffic density in province m; T_m represents the passenger and cargo turnover volume in province m; $\mathrm{TL}_{m,\mathrm{total}}$ represents the length of transport routes in province m; $F_{m,k}$ represents the road traffic density of grade k road in province m; and $P_{m,k}$ represents the proportion of grade k roads' standard average daily value of traffic flow (Ministry of Transport of the People's Republic of China, 2004).

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(3) Calculate the standard length commutation factor of each road grade

Equation (4) describes this relation.

$$A_{m,k} = \frac{F_{m,k}}{\mathrm{SF}} \tag{4}$$

where $A_{m,k}$ represents the standard length commutation factor of grade k road in province m and SF represents the standard road traffic density (custom value).

To obtain the standard length commutation factor for each county, we assumed that the standard length commutation factor of each province is applicable to each county located in this province, that is to say $A_{m,k} = A_{c,k}$, where c represents the county name.

(4) Calculate the emissions intensity per unit standard road length in each county

First, the total standard road length in each county should be calculated, and then the target value can be obtained. Equations (5) and (6) describe this relation.

$$\mathrm{SL}_{c,\mathrm{total}} = \sum_{k=1}^{4} A_{c,k} \times L_{c,k} \tag{5}$$

$$\mathrm{EI}_c = \frac{E_c}{\mathrm{SL}_{c,\mathrm{total}}} \tag{6}$$

where $\mathrm{SL}_{c,\mathrm{total}}$ represents the total standard road length (km) in county c; $L_{c,k}$ represents the actual length (km) of the grade k road; EI_c represents the emissions intensity per unit standard road length in each county ($\mathrm{t\,km^{-1}}$); and E_c represents the total emissions in county c (t).

(5) Calculate the emissions in each cell

For the cells located in county c, the emissions can be calculated based on the standard road length of the target cell and the emissions intensity per unit standard road

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length, as shown in Eqs. (7) and (8):

$$GSL_{c,x} = \sum_{k=1}^{4} A_{c,k} \times GL_{c,x,k} \quad (7)$$

$$GE_x = GSL_{c,x} \times EI_c \quad (8)$$

where $GSL_{c,x}$ represents the standard road length of the cell x in county c (km); $GL_{c,x,k}$ represents the actual length of the grade k road of the cell x in county c (km); and GE_x represents the emissions in cell x (tons).

For cells located on the boundary of multiple counties, the standard road length of each county included in the target cell is calculated first, followed by the sum of the emissions of these counties, as shown in Eqs. (9) and (10):

$$BGSL_{c,x} = \sum_{k=1}^{4} A_{c,k} \times BGL_{c,x,k} \quad (9)$$

$$BGE_x = \sum_{c=1}^{a} BGSL_{c,x} \times EI_c \quad (10)$$

where $BGSL_{c,x}$ represents the standard road length of county c included in cell x (km); $BGL_{c,x,k}$ represents the actual road length of the grade k road of county c included in cell x (km); BGF_x represents the emissions of the cell x (tons); and a represents the number of counties located in cell x.

3 Results

3.1 Total vehicular emissions for China in 2006 and 2010

We estimate China's vehicular emissions of CO, NO_x, VOCs, NH_3, $PM_{2.5}$, PM_{10}, BC, and OC to be 30 113.9, 4593.7, 6838.0, 20.9, 400.2, 430.5, 285.6, and 105.1 Gg,

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respectively in 2006 and 34 175.2, 5167.5, 7029.4, 74.0, 386.4, 417.1, 270.9, and 106.2 Gg, respectively, in 2010. Table 3 presents the emissions of these two target years.

Compared with 2006, CO, NO_x, VOCs, NH_3, and OC emissions increased in 2010: 254.8 % for NH_3 because of the comprehensive use of the TWC, 13.5 % for CO, 12.5 % for NO_x, 2.8 % for VOCs, and 1.1 % for OC. In contrast, $PM_{2.5}$, PM_{10}, and BC emissions decreased by 3.4 %, 3.2 % and 5.2 %, respectively. These emission changes can be attributed to the populations and distributions in each vehicular category between the two years.

On the national scale, the contributions of each vehicle category to the emissions of the eight pollutants in 2006 and 2010 are shown in Fig. 1. According to Fig. 1a and b, the contribution of each category of vehicle in 2006 was the same as that of 2010 on the whole. MCs and LDGVs were the main contributors to CO, VOCs, and NH_3 emissions, which in total accounted for approximately 66.2 %, 80.3 %, and 68.8 % of the emissions for these three pollutants in 2006, respectively, and approximately 70.9 %, 81.7 %, and 87.7 %, respectively, in 2010. RVs and HDDTs were the main contributors to NO_x, $PM_{2.5}$, PM_{10}, and BC emissions, which accounted for approximately 68.1 %, 53.9 %, 52.7 %, and 60.9 % in 2006, and approximately 69.6 %, 53.7 %, 52.4 %, and 60.7 % in 2010, respectively. The main contributors to OC were MCs and HDDTs, which accounted for approximately 61.5 % of the OC emissions in 2006 and 58.7 % in 2010.

Among fuel types, gasoline-fueled vehicles were the major contributors to the CO, VOCs, and NH_3 emissions and made up 72.8–90.2 % of the contributions of these three pollutants in both years. NO_x, $PM_{2.5}$, PM_{10}, and BC were mainly from diesel-fueled vehicles, which accounted for 76.5–90.4 % of the contributions of these four pollutants in both years. OC emissions from gasoline-fueled and diesel-fueled vehicles were similar, and the differences were all within 6.5 % in 2006 and 2010.

The contributions of vehicles with each emission standard to the emissions of these eight pollutants in 2006 and 2010 are shown in Fig. 2. For the lack of the population and emission factors of RVs with different emission standards, contribution of

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RVs is not considered in this part. In 2006, the Euro II emissions standard was implemented throughout China, and the Euro III emission standard was implemented in most provinces in 2010, except in Beijing, where the Euro IV emission standard was implemented. The results show that in 2006, Euro 0 and I vehicles were the major contributors to emissions, accounting for approximately 69.1–91.1 % of the emissions of each pollutant. The contributions of Euro I vehicles were the most significant. In 2010, Euro 0 to II vehicles made great contributions to the emissions of most pollutants (except the NH_3), accounting for approximately 72.7–88.8 % of the emissions, and Euro I vehicles were again the greatest contributors. Euro III vehicles were the major contributors to the emissions of NH_3, and the contribution was as high as 68.0 % because of the use of TWC. Therefore, eliminating the vehicles with old emission standards appropriately could effectively reduce the vehicular emissions.

3.2 The contribution of each vehicle category and emissions standard to emissions by province

Total emissions in each province in both years were listed in the Tables S4 and S5. Figures 3 and 4 illustrate the provincial differences in the contributions of vehicle categories to emissions in 2006 and 2010. The contributions of each vehicle category to emissions in each province varied from one another, due to the variety of vehicle compositions in each province. In 2006, LDGVs were the major contributors to CO, VOCs, and NH_3 emissions, and HDDVs were the major contributors to NO_x, $PM_{2.5}$, PM_{10}, BC, and OC emissions in Beijing, Tianjin, and Shanghai. In other provinces, MCs were the major contributors to CO, VOCs, NH_3, and OC emissions; RVs were the major contributor to NO_x emission; and HDDTs were the major contributors to $PM_{2.5}$, PM_{10}, and BC emissions (except Hebei, Anhui, Shandong, and Henan, where $PM_{2.5}$, PM_{10}, and BC emissions were primarily from RVs). These differences could be due to the large population densities, the sharp growth in the number of vehicles, and the great demand for public transportation in Beijing, Tianjin, and Shanghai. In addition, MCs emissions in these three cities were not significant because strict regulations to restrict the growth of

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motorcycles have been implemented. In 2010, LDGVs were the major contributors to CO, VOCs, NH_3, and OC emissions, and HDDVs were the major contributors to NO_x, $PM_{2.5}$, PM_{10}, and BC emissions in Beijing. In other provinces, LDGVs were the major contributors to CO and NH_3 emissions, MCs were the major contributors to VOCs and OC emissions, RVs were the major contributors to NO_x emissions, and HDDTs were the major contributors to $PM_{2.5}$, PM_{10}, and BC emissions (except in Hebei, Anhui, Shandong, and Henan, where these emissions were primarily from RVs). Therefore, measures to reduce provincial emissions should be taken individually, according to the specific major contributors to emissions in each province.

The contributions of vehicles with different emission standards in each province in 2006 and 2010 were the same as the characteristics of the whole nation generally, as shown in Figs. S1 and S2. In 2006, the Euro I vehicles were the primary contributors to pollutant emissions (except NH_3), followed by vehicles with Euro 0 emissions standard in most provinces. In 2010, the contributions of Euro 0 to Euro II vehicles to CO emissions both hovered 27 % approximately in most provinces. Euro III vehicles contributed more to NO_x emissions; thus, emissions from Euro 0 vehicles were reduced accordingly from 2006 to 2010. VOCs emissions from Euro I vehicles were comparable to Euro II vehicles and higher than Euro 0. NH_3 emissions were mainly from Euro IV vehicles in Beijing, which accounted for approximately 60.3 %, as shown in Fig. 5. The total contribution of Euro III and Euro IV vehicles was as high as 88.1 %. In other provinces, the NH_3 emissions were mainly from Euro III vehicles, which accounted for 54.7 % to 82.5 %. Tianjin had the greatest contribution from Euro III vehicles (82.5 %), followed by Zhejiang (79.6 %), Shaanxi, Chongqing, Shanghai, Jiangsu, Guizhou, and Sichuan, where were all above 75.0 %.

3.3 Spatial distribution of vehicular emissions

Gridded emissions were calculated with a resolution of $0.25^\circ \times 0.25^\circ$ based on the county-level emissions drawn by ARCGIS. All eight pollutants showed consistent char-

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acteristics in their spatial distribution; thus, CO and NH_3 emissions in 2006 and 2010 are shown as examples in Fig. 6.

Figure 6 clearly illustrates that vehicular emissions in China presented significant regional spatial distributions. Emissions in the eastern and southern parts of China were much higher than those in western and northern parts in 2006 and 2010. The NCP (the North China Plain, including Beijing, Tianjin, Hebei, Shandong, and Henan), YRD, and PRD regions, which cover only 8.4 % of Chinese territory, generated approximately 50 % of the total emissions in 2006 and 2010. The details are shown in Table 4. The high emissions in these regions were mainly concentrated in the large capital cities with large numbers of motor vehicles and dense networks of busy roads. The three provinces of northeastern China, the Sichuan Basin, Wuhan and Urumqi regions, also had high emissions. It is worth noting that NH_3 emissions increased significantly from 2006 to 2010, as shown in Fig. 6a-2 and b-2, and the intensity in Beijing was significantly higher than in other cities because of the use of three-way catalytic converters on Euro III and IV vehicles, but few studies have focused on this issue domestically.

Most existing spatial distribution method show that pollutants are mainly centralized in areas with large populations and developed economies because they use the economic indices and demographic information to allocate information on the grid. Our methodology was able to assign emissions to remote or sparsely populated but busy regions with a high resolution by using the busyness of the roads and road network information, and the results have been demonstrated to approximate the actual road emissions.

4 Discussion

4.1 Comparison with previous studies

Vehicular emissions of previous studies, both domestic and international, are summarized and compared with the results of this study in Table 5. As seen from the table,

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China's total vehicular emissions for each pollutant in 2006 in our study were highly consistent with the inventory developed by Zhang et al., which was one of important achievements of the INTEX-B program executed by NASA and which has been widely acknowledged for its reliability. Specifically, the differences in CO and NO_x emissions were within 10.7 % and 10.9 %, respectively; VOCs and OC emissions were within 4 %; and $PM_{2.5}$ and PM_{10} were within 1 %. In contrast, there was a large difference in BC emissions, up to 30.7 %, which may have been caused by the uncertainties in emission factors. A glaring discrepancy has emerged between the results of our study and RAINS, which calculated the vehicular emissions based on fuel consumption as estimated by the RAINS-ASIA module. That method used the fuel consumption in 1990 to predict the corresponding data in 2010, which inevitably led to uncertainties. The emissions in 2010 estimated in our study did not agree well with results calculated by Zhao et al. (2013) (except the NO_x emission) either, because emission factors given in their paper were different from ours and there were some differences in vehicle classifications.

Vehicles emissions in BTH, YRD, and PRD regions had been calculated by some scholars and were also showed in Table 5. Compared with these studies, differences could be found. These might have been caused by the sources of fuel ratio and the VTM. It should be noted that emissions from RVs were significant but were neglected in Lang's and Huang's study, and the emission standards were out of consideration in Zheng's study. Considering the old technology of vehicles registered earlier, emission factors of Euro 0 vehicles in our study were revised by referring previous studies, as discussed in Sect. 2.1.3. Therefore, big differences could be found in CO, NO_x, and VOCs emissions, compared with these three studies. Emissions on the city scale were also compared in this study. For example, emissions in Hangzhou in our study were close to Wang's results, whereas emissions in Guangzhou and Xi'an were not. NH_3 emission was just involved in Zhao' study, but it was much lower than our result, and this mainly caused by the differences in emission factors. At present, few researches have

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been conducted in vehicular emission of NH_3, and this may lead to the underestimation of total NH_3 emissions, therefore, more researches should be done on it.

4.2 Analysis of vehicular emissions and oil consumption

Theoretically speaking, oil consumption can generally characterize vehicular emissions, but it was not involved in the calculations in the present work. Therefore, the correlation between oil consumption and vehicular emissions was analyzed here to analyze the accuracy of the emissions results.

As shown in Fig. 7, the spatial distribution of oil consumption, which is available from the China Energy Statistical Yearbook compiled by the National Bureau of Statistics (2006, 2010), agreed closely with vehicular emissions on the whole, with a correlation coefficient of 0.83 in 2010. As shown in Fig. 8a, in 2010, oil consumption in the coastal areas of China was much higher than in the western region, and provinces with high oil consumption shared much contribution of emissions, such as Guangdong, Shandong, Jiangsu, and Zhejiang. But in Hebei and Henan the high emissions did not accompanied with very significant oil consumption. It was because that the numbers of RVs and MCs were large in these provinces and emission factors of these two kinds of vehicles were high. From 2006 to 2010, the oil consumption and vehicular emissions all increased in most provinces, especially in Shanxi, Liaoning, Guizhou, and Qinghai, where oil consumption increased by more than 60 % and CO emissions increased by more than 33 %. However, in Beijing, Shanghai, and Jiangsu, oil consumption increased by approximately 40 % from 2006 to 2010, as shown in Fig. 8b, but CO emissions declined slightly. This difference could be attributed to the tighter emission standards in these cities and provinces. Beijing has played a leading role in implementing emission standards, and good results have been achieved. Shanghai and Jiangsu also actively implement the emission standards on newly registered vehicles to control vehicular emissions. Therefore, stricter emission standards should be introduced comprehensively to control the vehicular emissions in China.

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4.3 Comparison with satellite data

Because both the life expectancy and transmission of NO_2 in the atmosphere are short, the NO_2 concentration observed by satellite can establish a direct link with the NO_x emissions on the ground. Fossil energy use is one of the important anthropogenic sources of NO_x emissions, particularly fuel consumption by motor vehicles, whose emissions are one of the main driving forces of NO_x in the atmosphere. Therefore, the spatial distribution of NO_2 observed by satellites can partially reflect the spatial distribution of vehicular emissions in polluted areas. We took NO_x emissions from vehicles in 2010 and the variations from 2006 to 2010 as examples to compare with the NO_2 column densities observed by the SCIA satellite, as shown in Fig. 9.

From Fig. 9c-1, it seems that high NO_2 emissions are mainly distributed in the NCP, YRD, and PRD regions in 2010, followed by the three Northern Provinces, Sichuan Basin, Wuhan, and Urumqi regions. This pattern agrees well with the vehicular emissions calculated in this study, as shown in Fig. 9d-1. But there are small differences, for example, the vehicular emissions in Tianjin, south of Hebei, and north of Shandong were less significant in 2010, as shown in Fig. 9d-1 compared with Fig. 9c-1. To understand these differences further, we collected the coal consumption, which is also one of the most important contributors of fossil energy of NO_x emissions. The coal consumption was obtained from the China Energy Statistical Yearbooks (2006, 2010), and then we used it to calculate the coal consumption per area in each province. As shown in Fig. 10a, Tianjin, Shandong, and Hebei were among the top five provinces in coal consumption in 2010, which could have led to the high NO_x emissions in these regions. Therefore, high NO_2 emission could be observed by satellite when vehicular emissions were not significant. Furthermore, from 2006 to 2010, the variations of the NO_2 column density were not highly consistent with the vehicular NO_x emissions, e.g., in Tianjin, south of Shandong, Jiangsu, south of Hebei, and southeast of Henan, as shown in Fig. 9c-2 and d-2. This also could be attributed to the coal used in these provinces. As shown in Fig. 10b, coal consumption in 2010 was clearly higher than in 2006, especially

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in Tianjin, Shandong, Jiangsu, Hebei, and Henan. Therefore, the variations of the NO_2 levels observed by satellite in these regions were also high, as shown in Fig. 9c-2 but not reflected in Fig. 9d-2. By this analysis, a conclusion can be draw that the increase of NO_x in China from 2006 to 2010 can be attributed to the heavy utilization of energy, and the coal consumption shares more contribution.

4.4 Policy impacts on vehicular emissions

Compared with 2006, the total fuel consumption of China in 2010 increased by 46.6 % on average, and the total number of Euro 0 to I vehicles decreased by 6.5 %, whereas the total number of other emission standard vehicles rose by 385.6 %. However, a small increase was shown in vehicular emissions of CO, NO_x, VOCs, and OC: 13.5 % for CO, 12.5 % for NO_x, 2.8 % for VOCs, and 1.1 % for OC. $PM_{2.5}$, PM_{10}, and BC emissions declined, as discussed in Sect. 3.1. Therefore, an interesting conclusion was draw in our study that the number of vehicle increased dramatically from 2006 to 2010, while the vehicular emissions calculated in this study did not show significant increase trend. Driven by curiosity, further work was conducted.

This modest increase in vehicular emissions can be attributed to the implementation of vehicle emissions standards. Table 6 illustrates the schedule of emission standard implementation for different vehicle types in China. For gasoline vehicles, the timing of implementation of the emission standards in most provinces is the same as at the national level, whereas that of Beijing was much earlier. Specifically, the implementation time of Euro I to III emission standards was one year earlier in Beijing than in other provinces, and the implementation of Euro IV was much earlier. Diesel vehicle standards were implemented at the same time across the nation. In the target years of this study, the Euro II emission standard was implemented in 2006, and the Euro III emission standard was implemented in 2010 (except in Beijing, where Euro III and IV were implemented, respectively). When the Euro III emission standard was implemented, the emissions limits of VOCs and PM from diesel vehicles became much stricter than before; this may have mitigated the emissions when the number of vehicles in the

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country increased rapidly. In addition, vehicular emissions in Beijing did not increase significantly between 2006 and 2010 because of the implementation of the Euro IV emission standard, as discussed in Sect. 4.2. Therefore, the appropriate restriction of vehicular emissions can effectively reduce pollutants.

As the new emissions standards have been implemented, the oil quality has improved correspondingly. The key parameters for gasoline and diesel with each standard are summarized in Tables 7 and 8. As shown in these tables, the sulfur content in the oil decreased incrementally, which means that catalyst pollution was reduced. Olefin in gasoline is also decreasing. As a result, oil burning can be more efficient and less PM is produced. In addition, the cetane level in diesel was increased appropriately as the new standard was implemented, which can improve the efficiency of oil combustion. All of these improvements can lead to a reduction in vehicular emissions.

To analyze the effects of emission standards and oil quality (sulfur level) deeply, we proposed four scenarios (S1–S4) to calculate the vehicular emissions in 2010 by refereeing the research of Liu et al. (2008). The scenario designs and results are listed in Table 9. In S1 both the emission standard and sulfur level can only meet the Euro I standard; In S2 the emission standard remains the same as S1, but sulfur level can meet Euro III standard; In S3 the emission standard can meet the Euro III standard, but sulfur level is not improve compared with S1; In S4 both of the emission standard and sulfur level are all improved to meet the Euro III standard (Euro IV for Beijing), which was implemented currently.

Compared with S4, if both of the emission standard and sulfur level were not improved, as described in S1, vehicular emissions of CO, NO_x, and VOCs could increase 52 %, 9 %, and 68 %, respectively; if only the sulfur level was improved, as described in S2, emissions of CO, NO_x, and VOCs could increase 35 %, 1 %, and 49 %, respectively; if only the emission standard was improved, as described in S3, CO, NO_x and VOCs emissions decreased 20 %, 9 % and 18 %, respectively. Here we can draw a conclusion that policies implemented in China are really effective to control the vehicular emissions, besides, NO_x emission is more sensitive to sulfur level than CO and NO_x.

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Therefore, stricter emission standard should be introduced accompanied by oil quality improvement.

Although China has make great efforts to improve its oil quality, the implementation of each oil standard was nearly ten years later than in the European Union, especially the IV standard, which cannot be implemented widely in China until the year of 2014 for gasoline and 2015 for diesel. This is one reason that the Euro IV emission standard cannot be implemented sooner. Not only are the implementation times of each standard later than Europe's, but the level of sulfur in the oil also cannot reach the world standards level, and the technology for controlling the content of aromatics and olefin in gasoline and the cetane number and aromatics in diesel is still behind the European Union and world standards. Therefore, technological improvements should be continued to reduce the vehicular emissions so that China can reach the world standards sooner.

4.5 Uncertainty analysis

Uncertainty analysis is an important component of a complete inventory. This process comprehensively embodies the uncertainties originating from the emission factors and the corresponding activity data. There are many ways to characterize the uncertainty in emissions inventories, including qualitative, semi-quantitative, and quantitative approaches (Wei et al., 2011). In this paper, the uncertainties of vehicular emission inventory in China were quantitatively evaluated with Monte Carlo simulations, depending on the data availability. Lognormal distribution was used to describe the uncertainties of activity data and emission factors, and this distribution can avoid negative values occurring during the simulation (Wei et al., 2011; Zhao et al., 2011; Fu et al., 2013). The reliability of date sources and estimation methods were used to estimate the standard deviations of the distribution based on the expert judgment. After 10 000 times repeated calculations, the uncertainties of the vehicular emissions in 2006 and 2010 estimated in this study were obtained, as shown in Table 10.

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In 2006, the overall uncertainties for CO, NO_x, VOCs, NH_3, $PM_{2.5}$, PM_{10}, BC, and OC are −32 % to 46 %, −35 % to 53 %, −34 % to 53 %, −51 % to 103 %, −38 % to 63 %, −36 % to 58 %, −47 % to 86 %, and −41 % to 69 %, respectively. These uncertainties mainly caused by the fuel ratio, VTM, and emission factors. As discussed in Sect. 2.1.2, the fuel ratios and VTM are not available from official statistics directly. Therefore, the uncertainties were aroused inevitably. Science the emission standards were taken into consideration in this study, the emission factors would be various from one standard to another. However, there was not enough deep research on emission factors of vehicles with each standard, especially on the vehicles registered early, which share much contribution to the emissions. Uncertainties of BC, and OC were high than CO, NO_x, VOCs, $PM_{2.5}$, and PM_{10} because the emission factors of them are calculated according to the fraction of these three particulate matters in relation to the total PM_{10}. Few researched were conducted on vehicular NH_3 emission, so NH_3 emission factors used in this study were all from the results of EPA, but these might not be suitable to China. Therefore, uncertainties of NH_3 emissions were much higher than other pollutants.

Compared with 2006, uncertainties of emissions in 2010 were a little higher generally. Except the major contributors mentioned before, the uncertainties might be caused for the lack of population of rural vehicles in 2010 as well. Because the rural vehicles in 2010 were not available from the statistic book, we amused that little change has taken place in the population of rural vehicles from 2006 to 2010. Uncertainties of NH_3 emissions increased a lot in 2010 because Euro III vehicles shared much contribution to NH3 emissions, while the uncertainties of NH_3 emission factors were high.

5 Conclusions

Vehicular emissions in China in 2006 and 2010 were calculated based on statistical data released by the State Statistics Bureau and additional relevant emission factors collected from domestic and international research. China's vehicular emissions of CO, NO_x, VOCs, NH_3, $PM_{2.5}$, PM_{10}, BC, and OC were 30 113.9, 4593.7, 6838.0,

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20.9, 400.2, 430.5, 285.6, and 105.1 Gg, respectively, in 2006 and 34 175.2, 5167.5, 7029.4, 74.0, 386.4, 417.1, 270.9, and 106.2 Gg, respectively, in 2010. CO, VOCs, and NH_3 emissions were mainly from motorcycles and light-duty gasoline vehicles, whereas NO_x, $PM_{2.5}$, PM_{10}, and BC were mainly from rural vehicles and heavy-duty diesel trucks. OC was mainly from motorcycles and heavy-duty diesel trucks. Euro 0 and Euro I vehicles were the main contributors to all emissions, except NH_3, which was mainly from Euro III and Euro IV vehicles.

The spatial distribution of emissions with a resolution of $0.25^\circ \times 0.25^\circ$ was developed using GIS. Emissions in the eastern and southern parts of China were much higher than those in western and northern parts in both years. The NCP, YRD, and PRD regions generated nearly 50 % of the total emissions of each pollutant in both years, followed by the three provinces of Northeastern China, the Sichuan Basin, and the Urumqi region. These results show good agreement with satellite data and the distribution of fuel consumption, and the spatial distribution method used in this study was demonstrated to be very practical. The total emissions estimated in this study are roughly equal to most previous studies.

Although the number of vehicles has increased dramatically, the total emissions of all the pollutants except NH_3 changed little from 2006 to 2010. This finding can be attributed to the implementation of strict emission standards and to improvements in oil quality. Therefore, restricting the vehicular emissions appropriately and rapidly improving oil quality are effective measures for controlling vehicular emissions. However, the implementation of the Euro IV emission standards has been postponed repeatedly because the oil quality still cannot satisfy the needs of the new emission standard. Therefore, the responsible departments should speed the improvement of oil quality. In addition, further research on NH_3 emissions should be conducted.

During the past several years, great efforts have been exerted to improve China's air quality, and restricting vehicular emissions has been an effective measure. Nevertheless, the haze in China is still severe. The vehicular emission inventory developed in this work is useful for describing the characteristics of regional vehicular pollution

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in China and can provide reliable data for regional air quality modeling, policy-making, and air-quality improvement.

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Table 1. Average annual traveled mileages of vehicles in 2006 and 2010 ($\times 10^3$ km yr^{-1}).

Year	LDGVs	LDDVs	HDGVs	HDDVs	LDGTs	LDDTs	HDDTs	MCs	RVs
2006	28	28	35	35	25	30	35	15	20
2010	25	25	30	30	20	25	30	10	20

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Table 2. Emission factors for vehicles ($g\,km^{-1}$).

Vehicle type	Fuel type	Emission standard	CO	NO_x	VOCs	NH_3	$PM_{2.5}$	PM_{10}	BC	OC
LDVs	gasoline	Euro 0	56.00[a]	1.80[e]	6.90[e]	0.0043[c]	0.013[d]	0.015[b]	0.00477[d]	0.00778[d]
		Euro I	11.88[b]	0.40[b]	1.80[b]	0.0094[c]	0.013[d]	0.015[b]	0.00477[d]	0.00778[d]
		Euro II	5.80[b]	0.09[b]	1.59[b]	0.0094[c]	0.013[d]	0.015[b]	0.00477[d]	0.00778[d]
		Euro III	4.60[b]	0.058[b]	0.64[b]	0.0632[c]	0.013[d]	0.015[b]	0.00477[d]	0.00778[d]
		Euro IV	1.40[b]	0.10[b]	0.32[b]	0.0632[c]	0.013[d]	0.015[b]	0.00477[d]	0.00778[d]
	diesel	Euro 0	1.50[a]	1.10[e]	0.44[e]	0.0042[c]	0.475[d]	0.500[f]	0.37665[d]	0.08835[d]
		Euro I	0.90[f]	1.10[f]	0.12[f]	0.0042[c]	0.095[d]	0.100[f]	0.07533[d]	0.01767[d]
		Euro II	1.00[f]	0.80[f]	0.11[f]	0.0042[c]	0.095[d]	0.100[f]	0.07533[d]	0.01767[d]
		Euro III	0.20[f]	0.60[f]	0.06[f]	0.0042[c]	0.076[d]	0.080[f]	0.06026[d]	0.01414[d]
		Euro IV	0.30[f]	0.29[f]	0.04[f]	0.0042[c]	0.076[d]	0.080[f]	0.06026[d]	0.01414[d]
HDVs	gasoline	Euro 0	156[a]	11.90[e]	13.30[e]	0.0280[c]	0.085[d]	0.100[f]	0.03116[d]	0.05084[d]
		Euro I	106[f]	1.10[f]	13.00[f]	0.0280[c]	0.085[d]	0.100[f]	0.03116[d]	0.05084[d]
		Euro II	9.50[f]	0.80[f]	1.20[f]	0.0280[c]	0.085[d]	0.100[f]	0.03116[d]	0.05084[d]
		Euro III	4.80[f]	0.60[f]	0.60[f]	0.0280[c]	0.085[d]	0.100[f]	0.03116[d]	0.05084[d]
		Euro IV	3.96[f]	0.29[f]	0.50[f]	0.0280[c]	0.085[d]	0.100[f]	0.0.3116[d]	0.05084[d]
	diesel	Euro 0	24.60[a]	24.9[e]	7.90[e]	0.0168[c]	0.725[d]	0.763[b]	0.57477[d]	0.13482[d]
		Euro I	7.84[b]	9.30[b]	1.81[b]	0.0168[c]	1.191[d]	1.254[b]	0.94464[d]	0.22158[d]
		Euro II	5.54[b]	8.57[b]	1.53[b]	0.0168[c]	0.441[d]	0.464[b]	0.34953[d]	0.08199[d]
		Euro III	3.15[b]	6.08[b]	0.20[b]	0.0168[c]	0.238[d]	0.251[b]	0.18908[d]	0.04435[d]
		Euro IV	0.30[b]	6.40[b]	0.035[b]	0.0168[c]	0.083[d]	0.087[b]	0.06554[d]	0.01537[d]

LDVs: light-duty vehicles; HDVs: heavy-duty vehicles; LDTs: light-duty trucks; HDTs: heavy-duty trucks.
[a] Streets et al. (2006).
[b] Beijing Municipal Science and Technology Project (2012).
[c] US EPA (2005).
[d] Streets et al. (2001).
[e] Fu et al. (2001).
[f] VECCSEPA (2005).
[g] Yao et al. (2011a).

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Table 2. Continued.

Vehicle type	Fuel type	Emission standard	CO	NO_x	VOCs	NH_3	$PM_{2.5}$	PM_{10}	BC	OC
LDTs	gasoline	Euro 0	95.30[a]	4.80[e]	12.60[e]	0.0044[c]	0.029[d]	0.034[b]	0.01059[d]	0.01729[d]
		Euro I	26.30[b]	0.60[b]	1.60[b]	0.0094[c]	0.029[d]	0.034[b]	0.01059[d]	0.01729[d]
		Euro II	17.90[b]	0.40[b]	1.40[b]	0.0094[c]	0.029[d]	0.034[b]	0.01059[d]	0.01729[d]
		Euro III	15.40[b]	0.20[b]	1.30[b]	0.0632[c]	0.028[d]	0.033[b]	0.01028[d]	0.01678[d]
		Euro IV	5.80[b]	0.10[b]	1.20[b]	0.0632[c]	0.028[d]	0.033[b]	0.01028[d]	0.01678[d]
	diesel	Euro 0	10.00[a]	3.40[e]	0.24[e]	0.0042[c]	0.369[d]	0.388[b]	0.29228[d]	0.06856[d]
		Euro I	2.43[b]	1.60[b]	0.32[b]	0.0042[c]	0.333[d]	0.350[b]	0.26366[d]	0.06185[d]
		Euro II	2.47[b]	1.60[b]	0.31[b]	0.0042[c]	0.233[d]	0.235[b]	0.17703[d]	0.04152[d]
		Euro III	1.71[b]	1.40[b]	0.21[b]	0.0042[c]	0.124[d]	0.130[b]	0.09793[d]	0.02297[d]
		Euro IV	0.60[b]	1.10[b]	0.10[b]	0.0042[c]	0.079[d]	0.083[b]	0.06252[d]	0.01467[d]
HDTs	diesel	Euro 0	24.60[a]	24.90[e]	7.90[e]	0.0168[c]	2.236[d]	2.354[b]	1.77327[d]	0.41595[d]
		Euro I	2.33[b]	5.29[b]	2.11[b]	0.0168[c]	1.061[d]	1.117[b]	0.84144[d]	0.19737[d]
		Euro II	1.90[b]	10.00[b]	0.70[b]	0.0168[c]	0.128[d]	0.135[b]	0.1017[d]	0.02385[d]
		Euro III	2.60[b]	8.40[b]	0.60[b]	0.0168[c]	0.138[d]	0.145[b]	0.1092[d]	0.02562[d]
		Euro IV	0.20[b]	4.90[b]	0.035[b]	0.0168[c]	0.067[d]	0.071[b]	0.0535[d]	0.01255[d]
MCs	gasoline	Euro 0	20.00[a]	0.10[e]	6.30[e]	0.007[c]	0.102[d]	0.120[b]	0.0373[d]	0.06101[d]
		Euro I	8.10[b]	0.11[b]	3.40[b]	0.007[c]	0.051[d]	0.060[b]	0.0187[d]	0.03050[d]
		Euro II	4.95[b]	0.15[b]	1.415[b]	0.007[c]	0.027[d]	0.032[b]	0.0100[d]	0.01627[d]
		Euro III	2.75[b]	0.15[b]	0.855[b]	0.007[c]	0.015[d]	0.018[b]	0.0056[d]	0.00915[d]
RVs	diesel	–	15.00[g]	4.50[g]	1.40[g]	0.0042[g]	0.171[g]	0.180[g]	0.1356[g]	0.03181[g]

LDVs: light-duty vehicles; HDVs: heavy-duty vehicles; LDTs: light-duty trucks; HDTs: heavy-duty trucks.
[a] Streets et al. (2006).
[b] Beijing Municipal Science and Technology Project (2012).
[c] US EPA (2005).
[d] Streets et al. (2001).
[e] Fu et al. (2001).
[f] VECCSEPA (2005).
[g] Yao et al. (2011a).

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Table 3. Vehicular emissions of China in 2006 and 2010 (unit: $Gg\,yr^{-1}$).

Year	CO	NO_x	VOCs	NH_3	$PM_{2.5}$	PM_{10}	BC	OC
2006	30 113.9	4593.7	6838.0	20.9	400.2	430.5	285.6	105.1
2010	34 175.2	5167.5	7029.4	74.0	386.4	417.1	270.9	106.2

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Table 4. Contributions of high-emissions regions in China (%).

Year	Region	CO	NO_x	VOCs	NH_3	$PM_{2.5}$	PM_{10}	BC	OC
2006	NCP	29.5	31.3	26.8	27.1	25.9	25.9	26.0	25.6
	YRD	9.9	8.2	10.9	10.8	9.4	9.4	9.1	10.0
	PRD	10.2	6.7	11.5	11.0	9.3	9.3	8.9	10.3
	Total	49.6	46.2	49.2	48.9	44.6	44.6	44.0	45.9
2010	NCP	28.9	31.1	26.4	27.6	27.8	27.7	28.3	26.6
	YRD	11.6	9.7	11.8	14.8	10.6	10.6	10.4	11.1
	PRD	9.6	6.0	10.8	9.8	8.4	8.5	7.9	9.6
	Total	50.1	46.8	49.0	52.2	46.8	46.8	46.6	47.3

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Table 5. Comparison of vehicular emissions inventories ($Gg\,yr^{-1}$).

Study	Study area	Base year	CO	NO_x	VOCs	NH_3	$PM_{2.5}$	PM_{10}	BC	OC
EDGAR (2005)	China	2000	14 958	1366	1963	–	–	–	–	–
Cai et al. (2007)	China	2005	36 197	4539	6288	–	–	983	–	–
Zhang et al. (2009)	China	2006	33 709	5096	6630	–	398	427	198	101
Cofala (2005)	China	2010	12 101	1934	–	–	–	–	–	–
Hu et al. (2002)	China	2010	32 440	5140	5370	–	–	–	–	–
Zhao et al. (2013)	China	2010	–	4382	2466	2	126	133	55	40
This study	China	2006	30 114	4594	6838	21	400	431	286	105
This study	China	2010	34 175	5167	7030	74	387	417	271	106
Lang et al. (2012)	BTH region	2008	2641	788	631	–	–	48	–	–
This study	BTH region	2010	3833	562	709	9	41	44	29	11
Huang et al. (2011)	YRD region	2007	–	306	332	–	40	43	–	–
This study	YRD region	2006	2557	302	654	2	31	34	22	9
Zheng et al. (2009a)	PRD region	2006	2583	322	465	–	74	93	–	–
This study	PRD region	2006	2514	254	646	2	30	33	21	9
Yao et al. (2012a)	Guangzhou	2010	332	61	59	–	–	3	–	–
This study	Guangzhou	2010	676	63	155	2	7	7	4	2
Wang et al. (2012)	Hangzhou	2010	441	44	23	–	–	7	–	–
This study	Hangzhou	2010	396	39	81	1	3	4	2	1
He (2011)	Xi'an	2010	446	80	64	–	–	–	–	–
This study	Xi'an	2010	321	54	64	0.8	3	4	2	0.9

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Table 6. Implementation schedule for new vehicle emission standards in China.

	Euro I	Euro II	Euro III	Euro IV
Gasoline (Beijing)[c, d]	1999	2003	2006	2008
Gasoline (national)[c, d]	2000	2004	2007	2013[b]
Diesel (national)[c, d]	2000	2004	2008[a]	2013[b]
Gasoline (Europe)[e]	1992	1996	2000	2005
Diesel (Europe)[e]	1992	1996	2000	2005

[a] initially scheduled for 2007 but postponed to 2008.
[b] initially scheduled for 2011 but postponed to 2013.
[c] Zhang et al. (2009).
[d] Lang et al. (2012).
[e] Vestreng et al. (2009).

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Table 7. Key parameters of gasoline with each standard.

Content	World				European Union				China			
	I	II	III	IV	I	II	III	IV	I	II	III	IV
	/	/	/	/	1993	1998	2000	2005	2003	2005	2010	2014
Sulfur/ppm ≯	1000	150	30	10	1000	500	150	50	1000	500	150	50
Aromatic/%(v/v) ≯	50	40	35	35	/	/	42	35	40	40	40	40
Olefin/%(v/v) ≯	/	18	10	10	/	/	18	18	35	35	30	28

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Table 8. Key parameters of diesel with each standard.

Content	World				European Union				China			
	I	II	III	IV	I	II	III	IV	I	II	III	IV
	/	/	/	/	/	/	/	/	2002	2003	2010	2015
Sulfur/ppm ≯	5000	300	30	10	2000	500	350	50	2000	500	350	50
Cetane number/%(m/m) ≯	48	53	55	55	49	49	51	51	45	49	49	51
Total aromatic/%(m/m) ≯	/	25	15	15	/	/	/	/	/	/	/	/

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Table 9. CO, NO_x, and VOCs emissions in four scenarios, units: Gg.

Scenario	Emission standard	Sulfur level (ppm)	CO	NO_x	VOCs
S1	Euro I	1000(G)/2000(D)	52 017.0	5614.3	11 816.6
S2	Euro I	150(G)/350(D)	46 265.9	5229.2	10 503.8
S3	Euro III(IV)	1000(G)/2000(D)	41 011.5	5662.6	8267.0
S4	Euro III(IV)	150(G)/350(D)	34 175.5	5167.5	7029.4

G: gasoline; D: diesel.

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Table 10. Uncertainties of vehicular emission in China in 2006 and 2010.

	2006	2010
CO	−32 % to 46 %	−33 % to 53 %
NO_x	−35 % to 53 %	−38 % to 80 %
VOCs	−34 % to 53 %	−31 % to 43 %
NH_3	−51 % to 103 %	−62 % to 165 %
$PM_{2.5}$	−38 % to 63 %	−37 % to 63 %
PM_{10}	−36 % to 58 %	−35 % to 59 %
BC	−47 % to 86 %	−45 % to 88 %
OC	−41 % to 69 %	−38 % to 62 %

Uncertainty analysis is at the 95 % confidence interval for both years.

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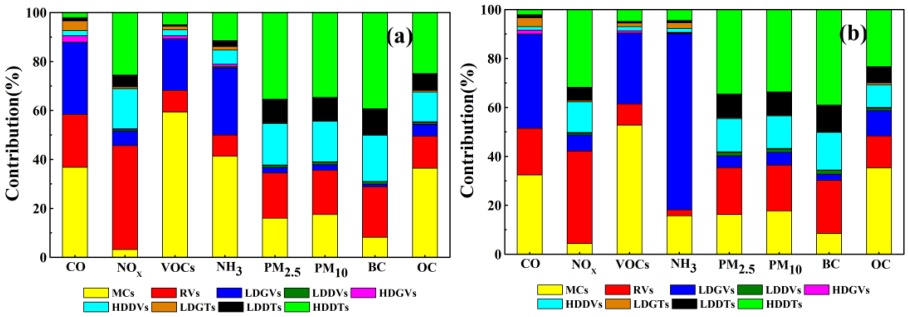

Fig. 1. Emissions contributions of different vehicle categories in China in 2006 **(a)** and 2010 **(b)**.

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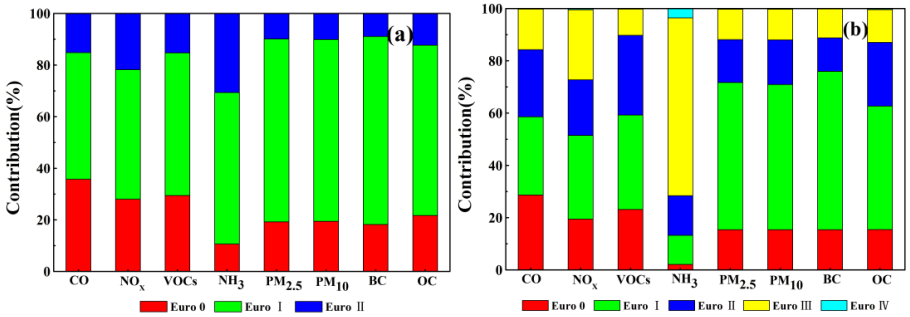

Fig. 2. Emissions contributions of vehicles with different emission standards in China in 2006 **(a)** and 2010 **(b)**.

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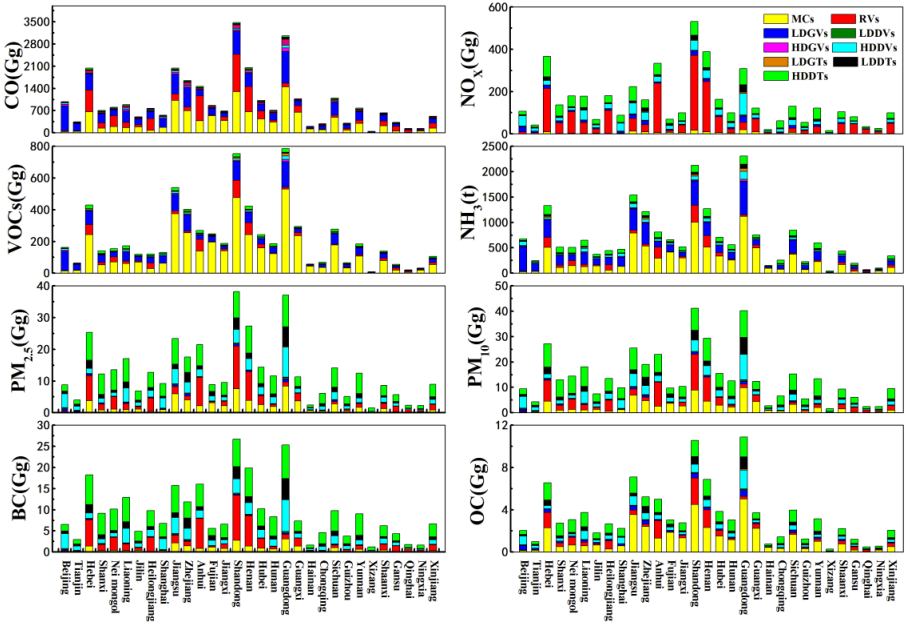

Fig. 3. The contributions of vehicle categories to 2006 emissions of eight pollutants by province.

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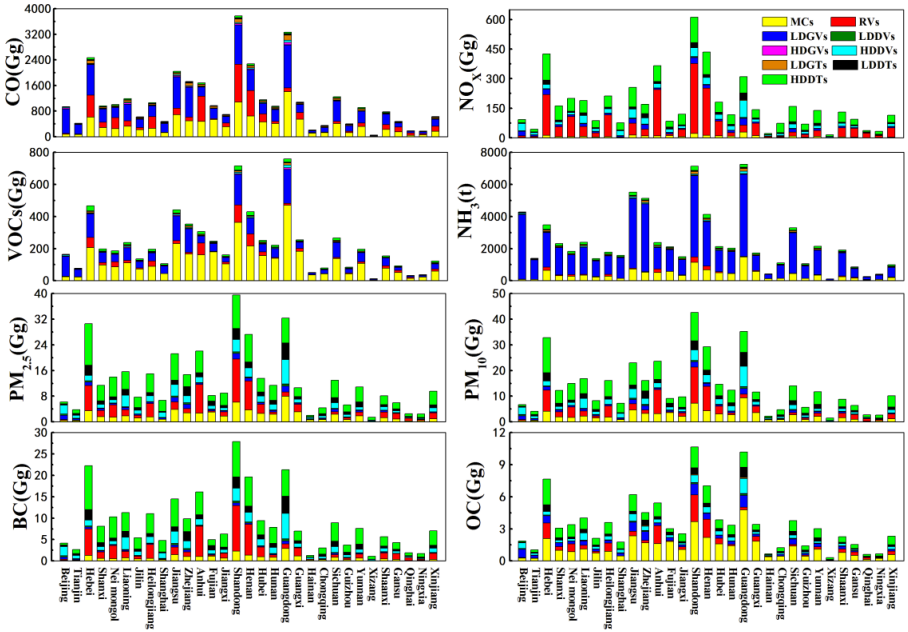

Fig. 4. Contributions of vehicle categories to 2010 emissions of eight pollutants by province.

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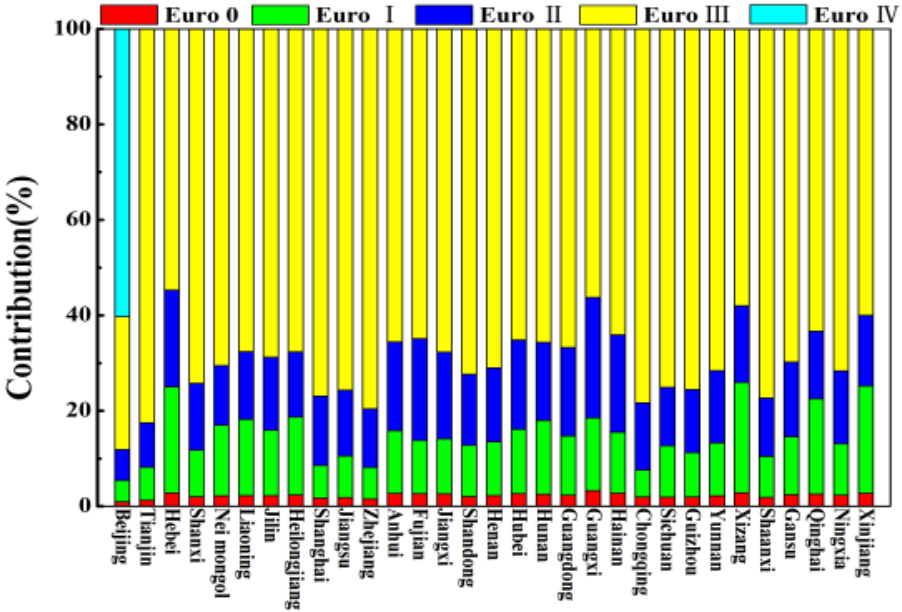

Fig. 5. Contributions of vehicles with different emission standards to NH_3 emissions in 2010 by province.

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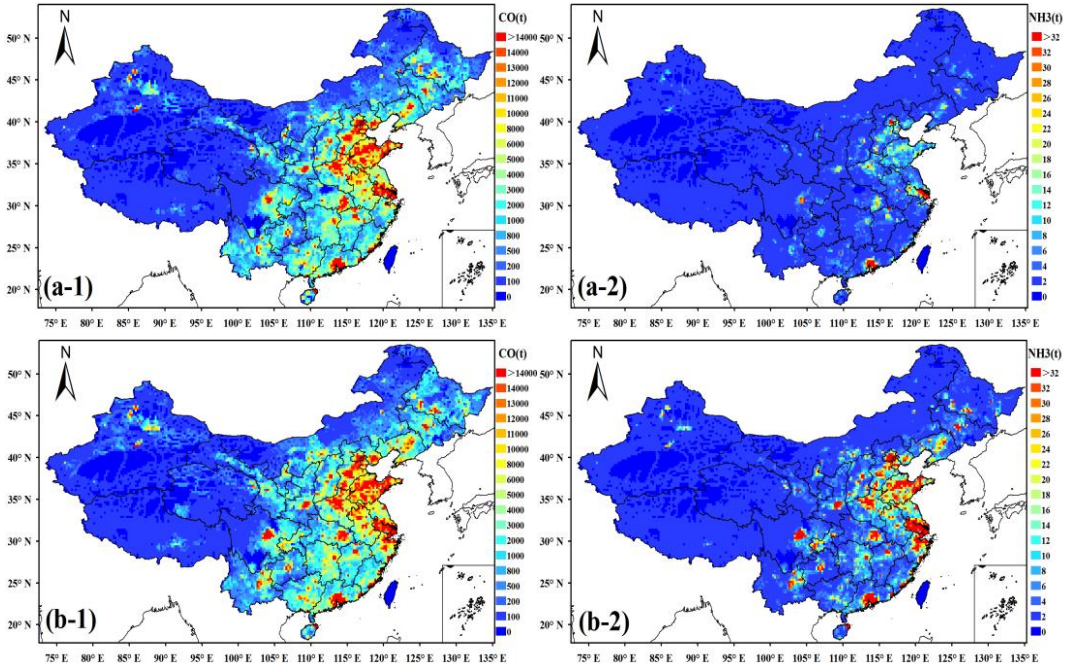

Fig. 6. Spatial distributions of CO and NH_3 emissions in China in 2006 and 2010 ($0.25^{\circ} \times 0.25^{\circ}$). (**a** and **b** represent the year of 2006 and 2010, respectively; **1** and **2** represent CO and NH_3 emissions, respectively).
* Emissions in Taiwan, Macao, and Hong Kong were set to 0 because they lacked relevant data.

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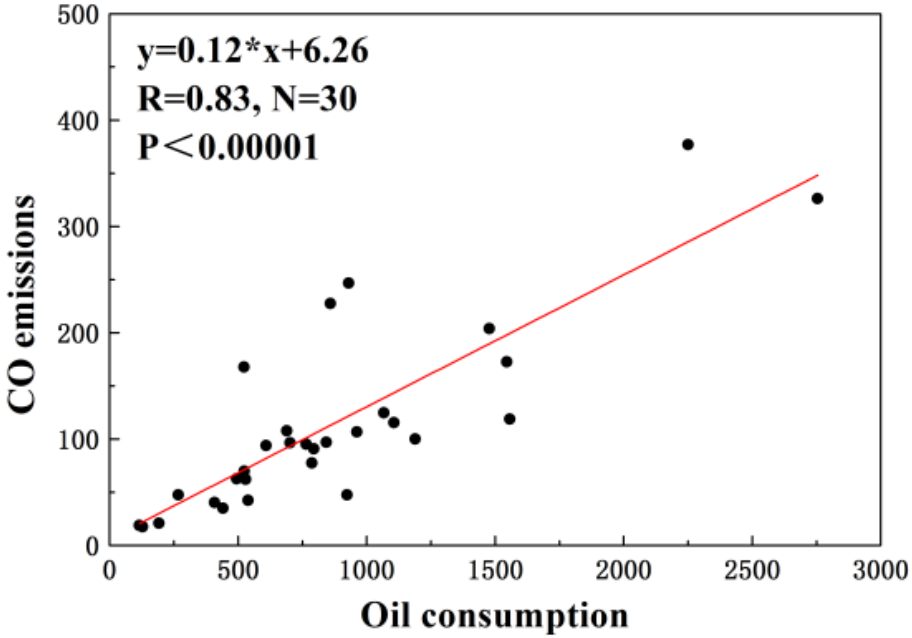

Fig. 7. Correlation between oil consumption and CO emissions in each province of China in 2010, units: 10^4 t.
* Tibet, Taiwan, Macao, and Hong Kong are not considered because of the lack of fuel consumption.

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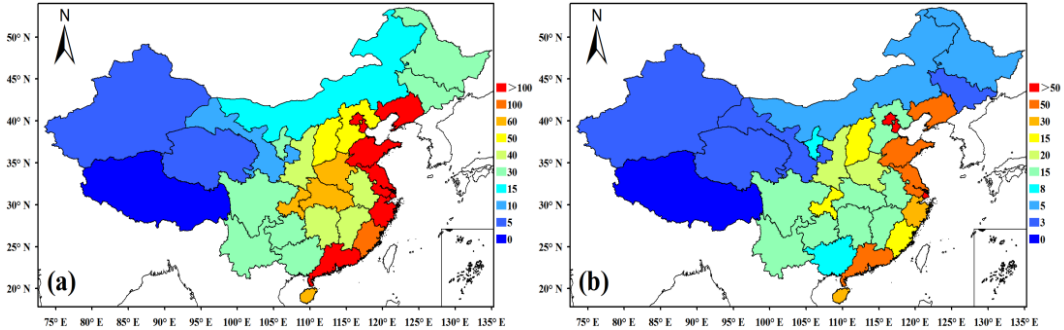

Fig. 8. Spatial distribution of oil consumption in each province of China in 2010 **(a)** and differences between 2006 and 2010 **(b)**, units: $t\,m^{-2}$.
* Tibet, Taiwan, Macao, and Hong Kong are not considered because of the lack of fuel consumption.

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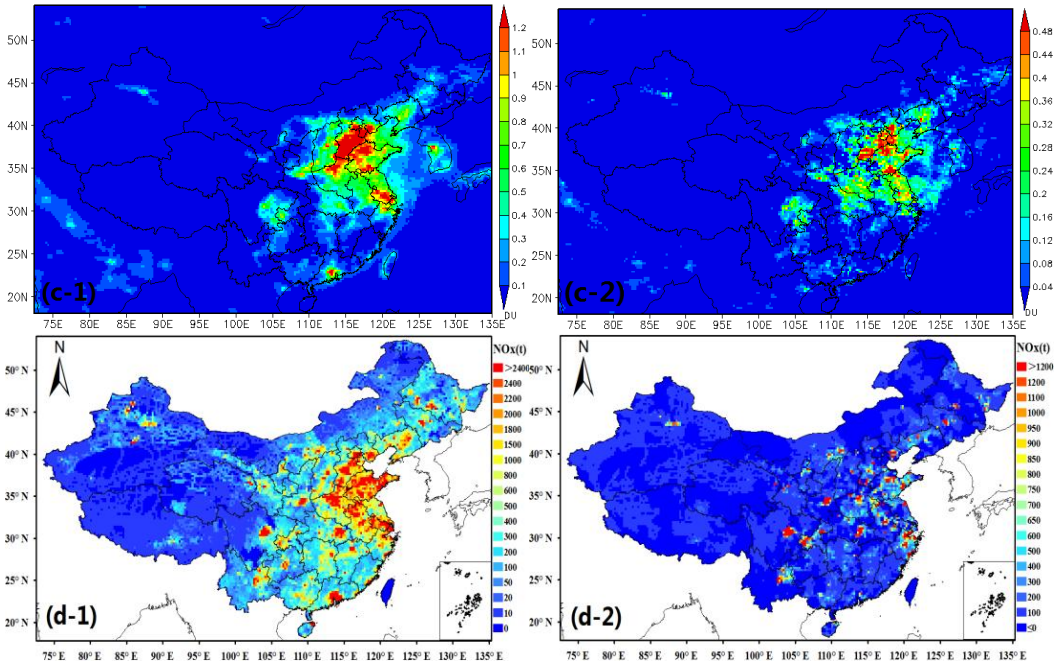

Fig. 9. NO_2 column density observed by SCIA satellite and vehicular NO_x emission in 2010 and variations from 2006 to 2010 (0.25° × 0.25°). (**c** and **d** represent satellite data and vehicular emission calculated in this study, respectively; **1** and **2** represent the year of 2010 and the variations from 2006 to 2010, respectively.)
* Emissions in Taiwan, Macao, and Hong Kong were set to 0 because they lacked relevant data.

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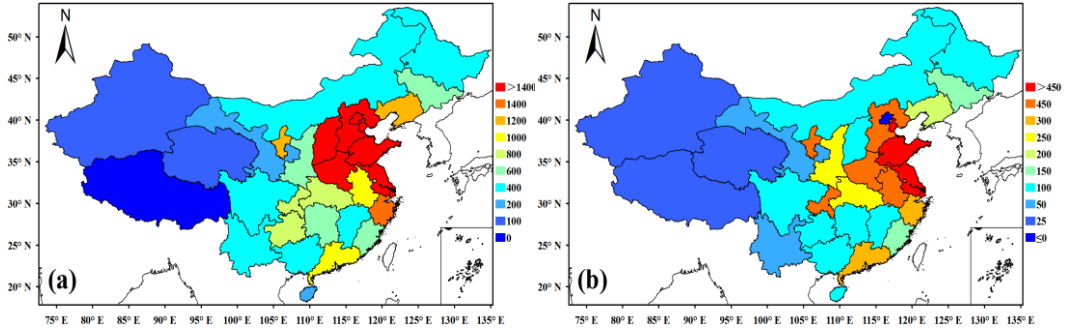

Fig. 10. The spatial distribution of coal consumption in each province of China in 2010 **(a)** and differences between 2006 and 2010 **(b)**, units: $t\,m^{-2}$.
* Tibet, Taiwan, Macao, and Hong Kong are not considered because of the lack of coal consumption.

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