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## Vehicular emissions in China in 2006 and 2010

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### ARTICLE INFO

#### Article history:

Received 31 August 2015

Revised 30 December 2015

Accepted 5 January 2016

Available online 26 May 2016

#### Keywords:

Vehicular emissions  
Gridded emissions  
Control policies  
China

### ABSTRACT

Vehicular emissions in China in 2006 and 2010 were calculated at a high spatial resolution based on the data released by the National Bureau of Statistics, by taking the emission standards into consideration. China's vehicular emissions of carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>), volatile organic compounds (VOCs), ammonia (NH<sub>3</sub>), fine particulate matters (PM<sub>2.5</sub>), inhalable particulate matters (PM<sub>10</sub>), black carbon (BC), and organic carbon (OC) were 30,113.9, 4593.7, 6838.0, 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<sub>3</sub> emissions were mainly from motorcycles and light-duty gasoline vehicles, whereas NO<sub>x</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>, and BC emissions were mainly from rural vehicles and heavy-duty diesel trucks. OC emissions were mainly from motorcycles and heavy-duty diesel trucks. Vehicles of pre-China I (vehicular emission standard of China before phase I) and China I (vehicular emission standard of China in phase I) were the primary contributors to all of the pollutant emissions except NH<sub>3</sub>, which was mainly from China III and China IV gasoline vehicles. The total emissions of all the pollutants except NH<sub>3</sub> changed little from 2006 to 2010. This finding can be attributed to the implementation of strict emission standards and to improvements in oil quality.

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### Introduction

As China's economy has developed rapidly, the total number of civilian motor vehicles (motorcycles and rural vehicles are not included) has surged from 14.5 million in 1999 to 78.1 million in 2010, an increase of 437.0% within 12 years. Especially in recent years, the annual growth rate of civilian vehicles has been approximately 20% (NBS, 2011). This increase in motor vehicles has caused vehicles to be an important contributor to China's air pollution. Among the pollutants emitted by motor vehicles, fine particulate matters (PM<sub>2.5</sub>), and inhalable particulate matters (PM<sub>10</sub>) can lower

visibility by the extinction process. Ammonia (NH<sub>3</sub>), volatile organic compounds (VOCs), and nitrogen oxides (NO<sub>x</sub>) are important precursors of secondary aerosols and ozone (O<sub>3</sub>) 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; Tang et al., 2009, 2012, 2015a).

To satisfy the China III (vehicular emission standard of China in phase III) emission standards, newly registered light-duty gasoline vehicles must install three-way catalytic

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converters (TWCs). However, these devices can lead to  $\text{NH}_3$  emissions while reducing CO,  $\text{NO}_x$ , and VOCs through redox reactions (Heeb et al., 2006a, 2008).  $\text{NH}_3$  in the atmosphere is highly reactive, and heterogeneous atmospheric reactions can readily generate nitrates and sulfates, which are the main components of  $\text{PM}_{2.5}$  (Bouwman et al., 1997; Goebes et al., 2003). At both urban and rural sites in the eastern region, the sum of sulfate, nitrate and ammonia in  $\text{PM}_{2.5}$  typically constituted much higher fractions (40–57%) than the western region (Yang et al., 2011). In January 2013, five times of heavy hazy pollution broke out in the Beijing–Tianjin–Hebei (BTH) region, and vehicular emissions made significant contributions to this pollution (Wang et al., 2013).

In the 1990s, a number of foreign researchers developed pollutant inventories for East Asia, which included China. Vehicular emissions were considered as well, the detailed characteristics of different vehicular emission inventories shown in Table S1. 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 years based on data in 1990 and then calculated Asia's  $\text{NO}_x$  emissions from 1990 to 2020. Streets and Waldhoff (2000) and Streets et al. (2001) suggested that motor vehicles in China were the main source of  $\text{NO}_x$  and carbon monoxide (CO) based on the NASA's (the National Aeronautics and Space Administration) China-MAP program. In addition, during the studies of the Transport and Chemical Evolution over the Pacific (TRACE-P) mission, Streets et al. (2003) improved their method incrementally by dividing the motor vehicles into categories and taking total annual mileage driven into consideration. The global emissions inventory EDGAR (Emission Database for Global Atmospheric Research) also included vehicular emissions (Olivier et al., 1998, 1999). Ohara et al. (2007) developed a new Asian emissions inventory called REAS (Regional Emission inventory in Asia) that covered the 40 years from 1980 to 2020 at a resolution of  $0.5^\circ \times 0.5^\circ$ , and vehicle classification, traffic volume, and fuel ratio information were taken into consideration in this work. The Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) was conducted by NASA to update the TRACE-P inventory (Zhang et al., 2009). Compared with the previous studies, INTEX-B took the technological innovation of vehicles into consideration, 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, methane ( $\text{CH}_4$ ), CO,  $\text{NO}_x$ , carbon dioxide ( $\text{CO}_2$ ), sulfur dioxide ( $\text{SO}_2$ ), Pb,  $\text{PM}_{10}$ , and nitrous oxide ( $\text{N}_2\text{O}$ ), based on the MOBILE5 model and fuel consumption. Song and Xie (2006) established the Chinese inventories of vehicular emissions with a resolution of  $40 \text{ km} \times 40 \text{ km}$  using GIS in 2002. Multi-year inventories of vehicular emissions at a high spatial resolution of  $40 \text{ km} \times 40 \text{ km}$  in China were established by Cai and Xie (2007). Yao et al. (2012b) calculated the vehicular emission inventories from 1990 to 2009 for 12 typical cities in China and analyzed the vehicular emissions trends in each city.

To address the defects in traditional methods of spatial allocation, Zheng et al. (2009a) and Che et al. (2009) developed the vehicular emissions in the Pearl River Delta (PRD) region. Lang et al. (2012) developed multi-year emissions inventories for  $\text{NO}_x$ , CO, VOCs, and  $\text{PM}_{10}$  in the Beijing–Tianjin–Hebei (BTH) region for the period 1999–2010 and discussed the effects of vehicle category, fuel ratio, and emission standards. Fu et al. (2013) developed a high-resolution emission inventory of primary air pollutants for Yangtze River Delta (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).

However, there have been dramatic changes in China in recent years, particularly in vehicle ownership and emission standards, and much of the published research is not applicable to the current situation in China. The effects of emission standards (from pre-China I to China II in 2006 and to China III/IV in 2010) were often overlooked in previous studies, and the emissions from the vehicles registered earlier were underestimated. Besides, some vehicular pollutants were not considered, especially the  $\text{NH}_3$  emissions, which have been a focus of many foreign researchers (Durbin et al., 2002; Heeb et al., 2006a, 2006b, 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 for all of Asia, which may lead to great uncertainties because of the lack of detailed information of China. Besides, 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 small scale of area.

To provide a reference for policy-making, and meet the needs of the regional air-quality simulation, the vehicular emissions of CO,  $\text{NO}_x$ , VOCs,  $\text{NH}_3$ ,  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ , black carbon (BC), and organic carbon (OC) in China in 2006 and 2010 were estimated in the present study at a resolution of  $0.25^\circ \times 0.25^\circ$ . The vehicle emission standards, categories, and fuel types were incorporated to improve the accuracy and reliability of the findings. For further study, the emissions calculated in this work were compared with the existing inventories, fuel consumption and  $\text{NO}_2$  column density observed by satellite. The effects of emission standards and oil quality improvement were analyzed.

## 1. Data and methodology

### 1.1. Emission calculation method

#### 1.1.1. Calculation formula

Vehicular emissions of CO,  $\text{NO}_x$ , VOCs,  $\text{NH}_3$ ,  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ , BC, and OC in different provinces of China were calculated based on emission factors, average annual vehicle traveled mileages

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