Science of the Total Environment 573 (2016) 974–984

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Unregulated pollutant emissions from on-road vehicles in China, 1999–2014

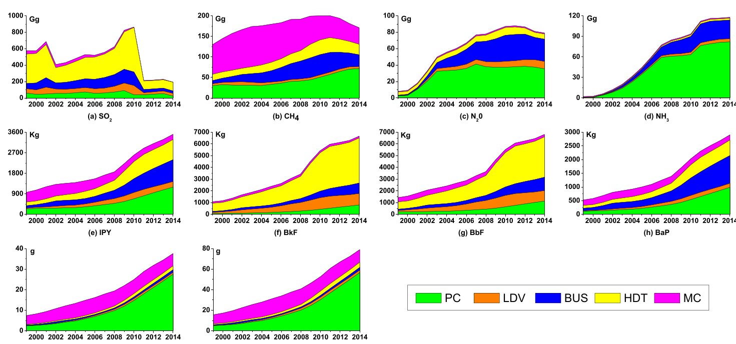
Jianlei Lang [a,b,*], Ying Zhou [a,b], Shuiyuan Cheng [a,b,c], Yanyun Zhang [a,b], Meng Dong [b], Shengyue Li [b], Gang Wang [a,b], Yonglin Zhang [a]

[a] Key Laboratory of Beijing on Regional Air Pollution Control, Beijing University of Technology, Beijing 100124, China
[b] College of Environmental & Energy Engineering, Beijing University of Technology, Beijing 100124, China
[c] Collaborative Innovation Center of Electric Vehicles, Beijing 100081, China

HIGHLIGHTS

- Long-term vehicular unregulated pollutant emissions in China were estimated.
- Sulfur content limit reduced SO_2 emission significantly.
- CH_4 and N_2O increased from 1999 to 2011 but decreased since 2012.
- NH_3 increased greatly in 1999–2014, but the growth rate slowed in recent years.
- PAHs, dioxins and furans increased sustainably in 1999–2014.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 31 May 2016
Received in revised form 3 August 2016
Accepted 25 August 2016
Available online xxxx

Editor: D. Barcelo

Keywords:
Vehicular emissions
Gaseous pollutants
PAHs
Dioxins and furans
China

ABSTRACT

Multi-year (1999–2014) vehicular unregulated pollutants emissions in China, including SO_2, CH_4, N_2O, NH_3, Indeno(1,2,3-*cd*)pyrene (IPY), Benzo(*k*)fluoranthene (BkF), Benzo(*b*)fluoranthene (BbF), Benzo(*a*)pyrene (BaP), dioxins and furans, were estimated based on emission factors calculated by COPERT. The inter-annual trends, correlation with GDP and population, spatial distribution characteristics, contributions from various vehicle types for the ten pollutants emissions were analyzed. Results showed that the emissions of the above ten pollutants changed from approximately 576.9 Gg, 130.0 Gg, 8.1 Gg, 2.1 Gg, 1.0 Mg, 1.1 Mg, 1.4 Mg, 0.5 Mg, 7.4 g and 15.6 g in 1999 to 193.8 Gg, 171.1 Gg, 79.1 Gg, 117.8 Gg, 3.5 Mg, 6.7 Mg, 6.8 Mg, 2.9 Mg, 37.6 g and 79.1 g in 2014, respectively. Implementation of stringent sulfur content limit during the past decade reduced approximately 94.4% of the SO_2 emission in 2014. CH_4 and N_2O increased from 1999 to 2011, but began to decrease since 2012; NH_3 emission had the highest annual average change rate (35.5%) from 1999 to 2014; PAHs, dioxins and furans increased continuously during the past decade. The vehicular emissions were higher in Guangdong, Shandong, Henan, Jiangsu, Zhejiang and Hebei. Good linear correlation between vehicular emissions and GDP was found (except SO_2); in the provinces/municipalities with higher population density, the emission density was also larger, indicating more significant vehicular emissions' potential damage on human health. HDT and PC, PC and MC, PC and BUS were the major contributors to SO_2, CH_4, N_2O emissions, respectively. In 2014, PC was the dominant contributor to NH_3 emission; PC, BUS and HDT had higher fraction in the total IPY and BaP emissions; HDT was the major contributor to BkF and BbF emissions. In addition, the uncertainties of estimated emissions were also analyzed based on Monte Carlo simulation.

* Corresponding author at: College of Environmental & Energy Engineering, Key Laboratory of Beijing on Regional Air Pollution Control, Beijing University of Technology, Beijing 100124, China.
E-mail address: jllang@bjut.edu.cn (J. Lang).

http://dx.doi.org/10.1016/j.scitotenv.2016.08.171

1. Introduction

Motor vehicle had been recognized as an important contributor to the regulated pollutants, such as CO, NO_X, NMVOCs and PM, due to the rapid growth and large population of vehicle in China (Fan et al., 2015a; Ohara et al., 2007; Wu et al., 2012; Zhang et al., 2009). In addition to the above regulated pollutants, motor vehicles could also exhaust other unregulated pollutants in the courses of oil combustion and ternary catalytic process, such as SO_2, CH_4, N_2O, NH_3, PAHs (e.g., benzo[*a*]pyrene (BaP)), dioxins and furans (Bakeas et al., 2011; Karlsson, 2004; Macor et al., 2011). These unregulated species could bring significant impact on human health, air quality and climate change. SO_2 is an important atmospheric $PM_{2.5}$ precursor (Zhang et al., 2015); CH_4 and N_2O are typical greenhouse gases (Tian et al., 2016); NH_3 is an atmospheric alkaline gas, it could be converted to ammonium and also promote the formation of nitrate and sulfate (Behera and Sharma, 2010); PAHs, dioxins and furans are typical toxic substances with significant adverse impact on human health (Becher and Flesch-Janys, 1998; Garrido et al., 2014; Polder et al., 2016; Rouleau et al., 2013). In addition, motor vehicle emissions occur mostly in urban areas where population densities are much higher than the regional average. Consequently, the contribution of pollutants from motor vehicles to human exposure risk is much higher than its contribution to emission itself (Shen et al., 2011). As a result, it is of great importance to estimate the vehicular unregulated pollutant emissions in order to provide basic data for further studies on the vehicular emission impact on human health, air quality, climate change, and also for effective mitigation measures development.

A few studies have been carried out to investigate the vehicular unregulated emissions characteristics. These studies mainly focused on the emission factors of the unregulated vehicular pollutants. For example, Liu et al. (2015) measured the PAHs concentrations in a road tunnel of Shanghai and quantified the emission factors through fuel burned model and vehicle kilometer travelled model. Kean et al. (2009) measured the motor vehicle ammonia emissions at a California highway tunnel and found that the emission factors of ammonia had decreased by 38% from 1999 to 2006. Bermudez et al. (2011) assessed the impact of low pressure exhaust gas recirculation (EGR) on the N_2O emissions in a light-duty diesel engine and indicated that the use of low pressure EGR could decrease the N_2O emissions. For inventory development, vehicular emissions are generally calculated based on the emissions factors (EFs) and activity data (i.e., vehicle population and kilometer travelled) (Song et al., 2012). Although a few studies have been carried out to estimate the local EFs in China for SO_2, N_2O, CH_4, NH_3, PAHs, dioxins and furans in China (Zhu et al., 2011; Yang, 2011), however, the results of these studies could not meet the requirement of estimating unregulated emissions for multi-year, multi-vehicle, multi-fuel types and multi-emission standards (i.e., State 0 – State V). As an alternative, the EU's COPERT model could be used to calculate the EFs for China, because the vehicle emission system in China is similar with that in Europe. In fact, many studies have applied the COPERT model for estimating the regulate pollutant emissions from motor vehicles in China (Cai and Xie, 2007; Fan et al., 2015b; Lang et al., 2014; Qiu et al., 2014).

The purpose of this study is to develop the long-term (1999–2014) emission inventory of unregulated pollutants from on-road vehicles in China. The unregulated pollutants in this study refer to SO_2, CH_4, N_2O, NH_3, dioxins, furans and four kinds of PAHs (Indeno(1,2,3-*cd*)pyrene (IPY), Benzo(*k*)fluoranthene (BkF), Benzo(*b*)fluoranthene (BbF) and Benzo(*a*)pyrene (BaP)). This is the first time that multi-year vehicular SO_2, NH_3, dioxins, furans and four kinds of PAHs emission inventory was developed considering different vehicle types, emission standards (State 0 – State V) and fuel oil sulfur contents in China. The inter-annual trends, correlation with GDP, spatial distribution characteristics of the ten vehicular emissions were analyzed. The contribution of different vehicle types was also investigated. In addition, the uncertainty of the estimated emissions was analyzed based on the Monte Carlo simulation.

2. Methodology

2.1. Calculation of emissions

Vehicular emissions of unregulated pollutants in 31 municipalities/provinces of China were estimated based on emission factors, vehicle population, and vehicle kilometers travelled (VKT) using Eq. (1):

$$Q_{m,n} = \sum_i \sum_j (P_{m,i,j} \times VKT_{m,i} \times EF_{i,j,n}) \tag{1}$$

Where m means the 31 municipalities/provinces in China (except Hong Kong, Macao and Taiwan, as shown in Fig. 1); n represents the pollutants, ten pollutants were considered in this study, including SO_2, CH_4, N_2O, NH_3, four kinds of PAHs (Indeno(1,2,3-*cd*)pyrene (IPY), Benzo(*k*)fluoranthene (BkF), Benzo(*b*)fluoranthene (BbF) and Benzo(*a*)pyrene (BaP)), dioxins and furans; i is the vehicle type, five types were included: passenger cars (PC), light duty vehicles (LDV), heavy duty trucks (HDT), buses (BUS), and motorcycles (MC); j means the national vehicular emission standards (from State 0 to State V); $Q_{m,n}$ means emission (g) of pollutant n in area m; $P_{m,i,j}$ is the number of type i vehicle with emission standard j in area m; $VKT_{m,i}$ is the annual average travel mileage (km) of type i vehicles in area m; $EF_{i,j,n}$ is the emission factor (g/km) of pollutant n exhausted from type i vehicles with emission standard j.

2.2. Vehicle population

The annual vehicle population and new registration number in different municipalities/provinces were required in this study. They were obtained from the official statistical yearbooks (NBSC, 1999–2015). Fig. 2 shows the inter-annual trends of the vehicle population in China from 1999 to 2014. Sharp vehicle pollution growth could be found during the past decade. The total number increased from 45.9 million in 1999 to 236.0 million in 2014. The annual average growth rate (AAGR) was 11.5%. This rate was close to that for the GDP in China (~14.1%). Among various vehicle types, MC was the dominant contributor before 2012. However, the contribution ratio decreased gradually from 72.1% in 2003 to 38.8% in 2014. In addition, the MC number has begun to decrease since 2012. PC had the fast growth, with an AAGR of 21.8%. Since 2013, PC has become the dominant vehicle type in China. By the end of 2014, the number of PC had reached 120.8 million, accounting for more than half (51.2%) of the total vehicle population. The numbers of LDV, BUS and HDT in 2014 were 14.0, 2.5 and 7.2 million, the AAGR from 1999 to 2014 was 9.3%, 5.7% and 6.2%, respectively.

The total population and the registration numbers of new vehicles were then processed based on Eq. (2) (Lang et al., 2012) in order to get the numbers of motor vehicles with different emission standards.

$$P_{m,i,j} = \begin{cases} \sum_y N_{m,i,j,y} & i \neq State0 \\ P_{m,i,total} - \sum_{j=StateI}^{StateIV} \sum_y N_{m,i,j,y} & i = State0 \end{cases} \tag{2}$$

Where y represents the year in which vehicular emission standard j was being implemented. $N_{m,i,j,y}$ is the new registration number of vehicle i with emission standard j in year y in area m. $P_{m,i,total}$ is the total population of vehicle i in area m in the target calculation year. It is noted that if the number of vehicle with State i (e.g., State I) was negative, it should be set to 0, and the number of vehicle with Sate (i + 1) (e.g., State II) would be modified accordingly. The calculation method based on the total vehicle population and the new registration numbers could reflect the lifespan of vehicles (or the elimination of old vehicles) inherently.