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Method to establish the emission inventory of anthropogenic volatile organic compounds in China and its application in the period 2008–2012

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HIGHLIGHTS

- A recognized method to establish the VOC emission inventory in China was proposed.
- A four-level source classification and corresponding EFs database were developed.
- China's VOC emissions had increased from 22.45 Tg in 2008 to 29.85 Tg in 2012.
- Industrial process, transportation and solvent utilization are key VOC sources.
- Highly polluted areas have expanded in 2008–2012, forming polluted city clusters.

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ABSTRACT

A method was developed to establish a comprehensive anthropogenic VOC emission inventory in China, in which a four-level source categorization was proposed, and an emission factor determination system together with a reference database were developed. And this was applied to establish VOC emission inventories for the period 2008–2012. Results show China's anthropogenic VOC emissions increased from 22.45 Tg in 2008 to 29.85 Tg in 2012 at an annual average rate of 7.38%, with Shandong, Guangdong, Jiangsu, Zhejiang and Hebei provinces being the largest emitters. Industrial processes, transportation and solvent utilization were the key sources, accounting for 39.3%, 25.6%, and 14.9% of the total emissions in 2012, respectively. Passenger cars, biofuel combustion, coke production, field burning of biomass, and raw chemical manufacturing were the primary VOC sources nationwide. The key sources for each province were different because of the disparate industry and energy structure. China's VOC emissions displayed remarkable spatial variation, with emissions in the east and southeast regions being much larger than in the northwest, and the high emission areas mainly centered in the Bohai Economic Rim, the Yangtze River Delta, the Pearl River Delta and the Sichuan Basin. The size of high emission areas expanded over the period 2008–2012, with heavily polluted city clusters gradually emerging.

1. Introduction

Due to its rapid economic development and urbanization, China has experienced increasingly severe air pollution. High concentrations of ground-level ozone and fine particulate matter ($PM_{2.5}$) are observed in many cities in China. The 2013 annual average $PM_{2.5}$ concentration and 8-h ozone (O_3) maximum in Beijing were 89.5 and 183.4 μg m^{-3}, respectively, exceeding the second class $PM_{2.5}$ and O_3 standards in China's Ambient Air Quality Standard (GB 3095-2012) by 155.7% and 14.6%, respectively (BMEPB, 2014). Ozone and $PM_{2.5}$ have been the key air pollutants in China (Shao et al., 2009a, 2009b; Guo et al., 2014; Zhang et al., 2015).

Volatile organic compounds (VOCs) are crucial precursors of tropospheric ozone and secondary organic aerosol (SOA), which is a significant contributor to $PM_{2.5}$ in China (Toro et al., 2006; Shao et al., 2009a; Ling and Guo, 2014; Yuan et al., 2013). Ozone formation in China is under the control of VOCs (Shao et al., 2009a, 2009b; Zhang et al., 2007). Shao et al. (2009b) found it was more

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effective to control VOCs than NOx for reducing ground-level O_3 in Beijing. Zhang et al. (2007) and Shao et al. (2009a) reached a similar conclusion in the Pearl River Delta. Recent studies have revealed that VOCs play a significant role in SOA formation, contributing 35–51% of the measured SOA (Yuan et al., 2013). These studies have indicated that anthropogenic VOCs are of great significance to air pollution in China (Barlett et al., 2005; Yuan et al., 2013).

VOC emission inventories provides information about emission sources and their characteristics, assists air quality modeling, and guides policy makers in the formulation of control measures(Streets et al., 2003a,b; Zheng et al., 2009). China lags far behind foreign countries in developing emission inventories. There is neither an official emission factor (EF) database, nor a systematized emission inventory guidebook, and the characteristics of anthropogenic VOC emissions in China are still ambiguous.

Although several studies have considered China's VOC emissions (Klimont et al., 2002; Streets et al., 2003a,b; Bo et al., 2008; Wei et al., 2011; Zhang et al., 2009), there are many problems with these inventories. First, there are remarkable differences among the different studies. China's VOC emissions in 2000 estimated by Streets et al. (2003a,b) were 2.7 Tg and 6.4 Tg higher than those reported by Ohara et al. (2007) and Bo et al. (2008), respectively, due to overestimated energy data. Klimont et al. (2002) used vehicular EFs adopted by Europe in the mid-1980s to estimate vehicular emissions in China in 2000, which resulted in an overestimation of 5.1 Tg compared to Bo et al. (2008). Second, many local EF measurements have been conducted in recent years (Hsu et al., 2007; Tsai et al., 2008, 2014; Wang et al., 2009, 2013), but they have not been used in most inventories. Most previous studies used EFs established by developed countries. Wei et al. (2011) used industry and fossil fuel combustion EFs selected from the EPA's Compilation of Air Pollution Emission Factors (AP-42). However, the actual EFs were higher than those reported in AP-42 due to the less-advanced combustion and post-treatment technology in China. Third, there is no unified and systemized VOC source categorization in China. In previous studies, source classification mostly referred to those in developed countries, which are not suitable for China. Additionally, with the rapid economic growth and energy consumption in China, VOC emissions have increased markedly and the emission characteristics have changed considerably (Bo et al., 2008). Previous estimates do not reflect the current situation in China, and therefore cause high uncertainty when used in modeling and policy-making.

The Environmental Protection Ministry of China therefore proposed a national program called "*the Research of Characteristics and Controlling Measures of VOCs Emissions from Typical Anthropogenic Sources*" to develop an approach to estimate VOC emissions in China. As a core component of the program, this study developed a systemized method to establish the VOC emission inventory in China. A comprehensive four-level source categorization was proposed, and an EF determination system together with a reference EF database was developed. Based on this method, the anthropogenic VOC emissions in China for the period 2008–2012 were studied. In the calculation, the most recent activity data and local EFs were preferentially selected, and vehicular EFs were calculated taking consideration of deterioration. Additionally, a technical guidebook for establishing the VOC emission inventory has been developed and used nationwide.

2. Method for estimating VOC emissions in China

2.1. Source classification

Source classification is crucial for establishing an emission inventory. Generally, VOC source categorization is based on industrial classification standards. Source categorization in the US is determined according to Standard Industry Classifications, and in Europe is based on the Statistical Classification of Economic Activities in the European Community. The industrial classification system in China differs from those used elsewhere, so applying a foreign classification directly to China would be unreasonable. Additionally, many important industrial sources were omitted in previous inventories (Wei et al., 2011). Therefore, a more reasonable and comprehensive source classification was proposed in this study. In total, over 70 different key sources were included in the industrial processes according to China's National Industries Classification (GB/T4754-2011).

In developed countries, biofuel is rarely used in daily life, and crop residues are not burnt in the field on a large scale. However, biomass is commonly used as a fuel in China, especially in rural areas. According to statistical data, over 300 Tg crop residues and 170 Tg firewood are consumed annually in China (China Energy Statistics Yearbooks, 2000–2008), of which about 100 Tg crop residues are burnt in the field (Cao et al., 2007; Wang and Zhang, 2008). These activities produce massive amounts of VOCs (Bo et al., 2008; Wei et al., 2011). Consequently, biomass burning is an important VOC source in China, and was considered as a major sector in this study.

Taking the above into consideration, we proposed a detailed four-level source categorization, including biomass burning, stationary fossil fuel combustion, solvent utilization, industrial processes and transportation in Level 1. Level 1 sources were further divided, with biomass burning classified into biofuel combustion and the field burning of crop resides in Level 2. Biofuel combustion and field burning of crop residues were further classified according to the fuel types as listed in Table 1. Industrial processes were divided into petroleum & related industries and other industrial processes in Level 2. Level 2 sources were further classified into oil and natural gas extraction, petroleum refining, manufacturing of raw chemicals, and synthetic materials, etc. in Level 3 (see Table 1). Thereafter, Level 3 sources were sorted into Level 4 by products. Unlike other studies, transport and storage, as well as waste disposal, were incorporated into the source category of industrial processes because they usually occurred in it. This has been accepted by relevant experts. Details of the classification are listed in Table 1. In total, 152 different VOC sources were covered in the classification.

2.2. Emission estimation and allocation

Anthropogenic VOC emissions were estimated using the "emission factor method". Eq. (1) was formulated to calculate the total emissions for a certain year in China.

$$Et = \sum_{p=1}^{n} \left(\sum P_{i,j} \times VMT_{i,j} \times EF_{i,j} + \sum (1 - R_k) \times EF_{s,k} \times A_{s,k} \right) \tag{1}$$

where E_t is the total VOC emissions (Tg) in a certain year; $P_{i,j}$ is the population of vehicle fleet *i* under emission standard *j* in province *p* (p = 1,2,3, … …, n; n = 31 provinces, autonomous regions and municipalities in the China mainland excluding Hong Kong, Macau and Taiwan); $VMT_{i,j}$ is the annual average mileage traveled by vehicle fleet *i* under emission standard *j* (km); $EF_{i,j}$ is the corresponding EF (g/km); R_k is the removal efficiency under the technological condition of *k*; $EF_{s,k}$ is the EF for source *s* (except on-road vehicles) under the technological condition of k; and $A_{s,k,}$ is the corresponding activity data.

Based on the provincial emissions calculated by Eq. (1), county