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Ozone and secondary organic aerosol formation potential from anthropogenic volatile organic compounds emissions in China

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ABSTRACT

Volatile organic compounds (VOCs) are major precursors for ozone and secondary organic aerosol (SOA), both of which greatly harm human health and significantly affect the Earth's climate. We simultaneously estimated ozone and SOA formation from anthropogenic VOCs emissions in China by employing photochemical ozone creation potential (POCP) values and SOA yields. We gave special attention to large molecular species and adopted the SOA yield curves from latest smog chamber experiments. The estimation shows that alkylbenzenes are greatest contributors to both ozone and SOA formation (36.0% and 51.6%, respectively), while toluene and xylenes are largest contributing individual VOCs. Industry solvent use, industry process and domestic combustion are three sectors with the largest contributions to both ozone (24.7%, 23.0% and 17.8%, respectively) and SOA (22.9%, 34.6% and 19.6%, respectively) formation. In terms of the formation potential per unit VOCs emission, ozone is sensitive to open biomass burning, transportation, and domestic solvent use, and SOA is sensitive to industry process, domestic solvent use, and domestic combustion. Biomass stoves, paint application in industrial protection and buildings, adhesives application are key individual sources to ozone and SOA formation, whether measured by total contribution or contribution per unit VOCs emission. The results imply that current VOCs control policies should be extended to cover most important industrial sources, and the control measures for biomass stoves should be tightened. Finally, discrepant VOCs control policies should be implemented in different regions based on their ozone/aerosol concentration levels and dominant emission sources for ozone and SOA formation potential.

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Introduction

Volatile organic compounds (VOCs) play a crucial role in tropospheric chemistry. They are major precursors for ozone and secondary organic aerosol (SOA), both of which are hazardous to human health (Nel, 2005) and affect the Earth's climate significantly as they are short-lived climate forcers (Stocker et al., 2013). In addition, SOA obscures visibility (Zhang et al., 2012) and ozone has adverse effects on the ecosystem (Amann et al., 2008). Globally, China has the largest VOCs emissions, as most of the VOCs sources remain uncontrolled except for a few metropolitan regions. Therefore, it is necessary to estimate the ozone and SOA formation potential from anthropogenic VOCs emissions in China, and based on the estimation, develop effective VOCs control strategies.

The formation mechanism of ozone has been extensively studied and is well understood. Chemical mechanism modeling is the most frequently used approach to estimate ozone formation potential (OFP) from VOCs. Chemical mechanisms such as SAPRC07, CB05, RADM2 have been widely used in chemical transport models (CTMs) (Appel et al., 2007; Yarwood et al., 2008; Lin et al., 2009; Hirtl et al., 2011; Herwehe et al., 2011; Heo et al., 2012; Zhao et al., 2013b). They can estimate OFP with good accuracy but requires heavy computation. A rapid method to estimate OFP involves multiplying the concentration/emission of each precursor and index such as maximum incremental reactivity (MIR) and photochemical ozone creation potential (POCP). MIR is derived from chamber experiments and thus confined to specific atmosphere conditions (Carter et al., 1995). In contrast, POCP is calculated from photochemical trajectory model that takes long range transport into consideration (Derwent et al., 2007), so it is more suitable for OFP estimation for China.

SOA formation potential (SOAFP) can be calculated by explicit chemical models (e.g. Master Chemical Mechanism (MCM; Saunders et al., 2003; Bloss et al., 2005)) and SOA yield models. The former approach has limited application due to its severe underestimation of SOA yields and high computational expense. The latter approach obtains yield parameters by empirical fits to smog chamber data. The SOA yield model, especially the two-product model (Odum et al., 1996), is widely applied in three-dimensional CTMs such as CMAQ and PMCAMx, but it still shows universal underestimation in SOA concentrations (Karydis et al., 2007; Appel et al., 2008; Lane et al., 2008b; Murphy and Pandis, 2009; Carlton et al., 2010; Tsimpidi et al., 2011; Fountoukis et al., 2011; Zhao et al., 2013b; Li et al., 2015a). Recent studies have made great strides toward solving this problem. On one hand, SOA mass yield is affected by several factors including experimental condition, measuring method, and wall loss effect (Hildebrandt et al., 2009), resulting in underestimation in yields of known precursors. Therefore, some researchers reestimated SOA mass yield under different experimental conditions. For example, Hildebrandt et al. (2009) reported higher yield than previous study after examining SOA mass yield from toluene under different OH, NO_x and aerosol concentration levels in smog chamber. On the other hand, the underestimation also

ascribed from omission of some SOA precursors, e.g., the intermediate-volatility organic compounds (IVOCs), which has smaller emission rates but much higher SOA yields than traditional precursors. Researchers have examined those ignored species so that the range of SOA precursors is extended. For example, the SOA mass yields have recently been obtained for alkylnaphthalene (Chan et al., 2009) and long-chain alkanes (Presto et al., 2010; Tkacik et al., 2012; Aumont et al., 2012). Nevertheless, these new results are scarcely applied in SOAFP estimation.

Detailed VOCs source profiles are required for both OFP and SOAFP estimation. Previous VOCs source profiles databases in China were established mainly for the estimation of ozone formation, resulting in more attention paid to species with low molecule weight. Consequently, species with high molecule weight which have considerable influence for SOA formation are roughly estimated, leading to uncertainty for SOAFP estimation.

Given the adverse environment effects of ozone and SOA, and the common links between each other, VOCs control strategies should be developed in light of both issues. However, most research on VOCs emission control strategy solely focused on ozone control (Avery, 2006; Luecken and Mebust, 2008; Louie et al., 2013; Ling and Guo, 2014; Ou et al., 2015). Dechapanaya et al. (2004) and Lv et al. (2009) estimated SOA formation from VOCs. Only a few studies have simultaneously estimated the ozone and SOA formation potential from VOCs (Shin et al., 2013; Cui, 2013; Han et al., 2013; Lin et al., 2015). Shin et al. (2013) presented some discussion on VOCs control strategy in Seoul, Korea based on the calculation results. Nevertheless, most estimation was based on ambient VOCs concentration rather than VOCs emission rates, making it difficult to link the ozone and SOA formation potential to individual sources. Also, the latest SOA yield results obtained from recent laboratory experiments were not incorporated in these studies. In addition, studies carried out in China only focused on specific city and seasons. Therefore, a detailed and comprehensive examination of VOCs control strategy simultaneously considering ozone and SOA formation is urgently needed for the whole country.

This study rapidly and simultaneously estimates the total and source-specific ozone and SOA formation potential from anthropogenic VOCs in China. Firstly, we establish a VOCs source profiles database in China according to latest results of sources measurements, with special attention given to large molecular species. Then, a database for OFP and SOAFP parameters is established using the latest SOA yield curves from smog chamber experiments, and the OFP and SOAFP for China in 2010 are estimated and analyzed. Finally, we propose recommendations for future VOCs control policies based on the evaluation results.

1. Methodology and data sources

1.1. VOCs species, emission sectors and source profile

In this study, all VOCs species are classified into 115 lumped species (Table 1). Compared to previous VOCs classification, species with high molecule weight including aliphatic alkanes

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