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Ambient volatile organic compounds pollution in China

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ABSTRACT

Owing to rapid economic and industrial development, China has been suffering from degraded air quality and visibility. Volatile organic compounds (VOCs) are important precursors to the formation of ground-level ozone and hence photochemical smog. Some VOCs adversely affect human health. Therefore, VOCs have recently elicited public concern and given new impetus to scientific interest. China is now implementing a series of policies to control VOCs pollution. The key to formulating policy is understanding the ambient VOCs pollution status. This paper mainly analyzes the species, levels, sources, and spatial distributions of VOCs in ambient air. The results show that the concentrations of ambient VOCs in China are much higher than those of developed countries such as the United States and Japan, especial benzene, which exceeds available standards. At the same time, the ozone formation potential (OFPP) and secondary organic aerosol formation potential (SOAFP) of various VOCs are calculated. Aromatics and alkenes have much higher OFPPs, while aromatics have higher SOAFP. The OFPPs of ambient VOCs in the cities of Beijing, Guangzhou and Changchun are very high, and the SOAFP of ambient VOCs in the cities of Hangzhou, Guangzhou and Changchun are higher.

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Introduction

Volatile organic compounds (VOCs) are a class of organic compounds, but they do not have a uniform worldwide definition. The term VOCs usually refers to all organic compounds whose boiling temperatures are less than 50–260°C at standard atmospheric pressure and have melting points below room temperature. This includes alkanes, alkenes, alkynes, aromatics, alcohols, aldehydes, ethers, ketones, esters, halogenated hydrocarbons and others. VOCs are not only

important precursors to the formation of ground-level ozone (Solomon et al., 2007), but are also harmful to human health. For example, 1,3-butadiene and benzene are known carcinogens (USEPA, 2009, 2012).

Recently, VOCs have received increased attention because of heavy haze pollution in China (Zhang et al., 2014; Yang et al., 2014). The Chinese central government has implemented a system of countermeasures to improve ambient air quality and protect human health. VOCs were officially controlled during “the 12th Five-year” period. In October, 2012, the “12th Five-year Plan”

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on Air Pollution Prevention and Control in Key Regions (<http://www.mep.gov.cn/gkml/hbb/bwj/201212/t20121205243271.htm>) was issued by the Ministry of Environment Protection of the People's Republic of China, which regards VOCs pollution control as an important index for assessing the influence of construction projects on the environment; and since then, a VOCs monitoring program has been launched, and VOCs treatment has come into the view of governments. In May 2013, the Ministry of Environment Protection of the People's Republic of China issued *Technologies and Policies on Volatile Organic Compounds (VOCs) Pollution Control* (http://kjs.mep.gov.cn/hjbhzb/bzwb/wrfzjszc/201306/t20130603_253125.htm), in which control technologies for various VOC-emitting industries were recommended. In September 2014, the Chinese State Council issued the State Council Air Pollution Prevention and Control Action Plan (http://zfs.mep.gov.cn/fg/gwyw/201309/t20130912_260045.htm). This action plan was put forward to improve VOC pollution control. In January 2015, the Technical Guide on Atmospheric Volatile Organic Compounds Source Inventory (http://www.mep.gov.cn/gkml/hbb/bgg/201408/t20140828_288364.htm) was issued. This is an effective guide to building VOC inventories in China. However, almost all the emission factors used were cited from the U.S. EPA, so there remains much work to be done. In August 2015, the Air Control Law (revised) was issued, with four new legal items on VOC control of raw materials, processing and product use. In June 2015, the Ministry of Finance of the People's Republic of China, the National Development and Reform Commission and the Ministry of Environmental Protection jointly issued *Measures for the Pilot Project of Collecting VOCs Pollution Discharge Fees* (Finance & Tax (2015), No. 71) (http://www.mep.gov.cn/gkml/hbb/bgth/201601/t20160125_326889.htm), and started to impose VOCs discharge fees on the petrochemical industry and packaging and printing industries on October 1, 2015. Thereafter the Beijing and Shanghai Municipal governments also issued fee details for VOCs emission; on the basis of national pilot programs, more industries, such as coating and ink manufacturing, automobile making and ship building industries, have been extended as VOCs pollution charge pilots. In total, 5 categories and 13 small and medium-size industries have been involved. In addition, VOCs monitoring and treatment policies have been also issued in areas such as Tianjin, Guangdong, Zhejiang and Jiangsu.

All these actions show that the Chinese Central Government has the determination to control ambient VOCs pollution. However, the country has paid little attention to VOCs for a long time; the following embarrassing realities are evident: (1) the current ambient VOCs pollution status is unknown due to the lack of systematic monitoring data, and the evaluation of VOCs pollution in China is not easy because of the lack of ambient environmental standards; (2) The available VOCs emission standards only limit non-methane hydrocarbons (NMHCs) and a few species, so they are in need of revision; (3) VOCs pollution control is urgent, but technologies are inadequate. Therefore, study of the VOCs pollution status is the key to fully understanding ambient VOCs pollution in China. The results are very important for establishing VOCs standard and countermeasures in the country. Studies on the characteristics and sources of ambient VOCs in China have been conducted around Beijing–Tianjin–Hebei, the Pearl River and Yangtze River deltas and other areas (Yuan et al., 2012,

2013; Geng et al., 2009; Liu et al., 2008). The present work presents the levels of atmospheric VOCs pollution using a literature study and data reanalysis, and investigates the ozone formation potential (OFP) and secondary organic aerosol formation potential (SOAFP) of VOCs in cities.

1. Data sources

There were two data sources for the study. The first are monitoring data gathered by the authors. Sampling and analysis of ambient VOCs were done continuously and automatically at temporal resolution 30 minutes, using a GC955 series online gas chromatography instrument (GC-PID/FID) (Syntech Spectras Instrument Co., Ltd., The Netherlands). This two-channel system uses dual columns (PLOT and ATTM-1) and detectors (photoionization detector PID, and flame ionization detector FID) to simultaneously analyze C_2 – C_{12} VOCs. After drying under ambient air pressure, tube samples were directly input to the analysis system. Using these instruments, 31 types of organic compounds could be detected, including 19 species of alkanes, alkenes and alkynes and 12 types of aromatic hydrocarbons and alkanes. The limits of quantification were from 0.1×10^{-9} to 300×10^{-9} , and the maximum range is around 1.0×10^{-4} . The detection limits of these substances are shown in Table 1. To check instrument stability, we monitored zero gas every day at 0:00 o'clock. Calibration was done weekly, using a standard gas prepared by the gravimetric method (Spectra Gases Inc., Branchburg, New Jersey, USA), containing 35 target species with mixing ratios in the range 3×10^{-9} – 15×10^{-9} . Results indicate that sensitivity variation of all measured compounds was 10% (Zhang et al., 2012). We monitored ambient VOCs (31 species) in Tianjin (11 August to 16 September 2006 and 18 June 2007 to present), Jiaying (25 September to 7 December 2006), Guangzhou (4 November to 24 December 2009) and Beijing (8 June 2006 to present) using the GC955.

Other data were taken from the literatures. Informations from scientific journals, university journals and governmental releases are complicated, focusing mainly on alkanes, alkenes, aromatics, and carbonyl compounds. Three online databases were used: (1) Science Citation Index Expanded (1900 to present, ISI Web of Knowledge); (2) ScienceDirect; (3) China National Knowledge Infrastructure (CNKI). The selection criteria for the literature data were as follows: (1) For offline monitoring, the sample number should be more than 20. (2) For online monitoring, the sampling period should be longer than one week. (3) The analysis method should be GC, GC/MS and HPLC. An Endnote database was assembled because there are dozens of literature sources for cities such as Beijing, Guangzhou and Shanghai. All data were converted into the same units. Some data shown in figures were not included. In this paper, the VOC concentrations are 1-hour averages in one day. The Chinese ambient VOC concentration is the mean concentration in more than three cities for ambient alkanes, aromatics and alkenes; and ambient carbonyl compound concentrations are their means in more than two cities because of a lack of data.

At present, more than 1000 VOC species can be detected by instruments. Only 300 species, including alkanes, aromatics,

Table 1 – VOC detection limit of GC955 instrument.

Alkane ($\times 10^{-9}$)		Alkene ($\times 10^{-9}$)		Aromatic ($\times 10^{-9}$)		Alkyn ($\times 10^{-9}$)	
Ethane	0.10	Ethene	0.05	Benzene	0.03	Acetylene	0.10
Propane	0.10	Propylene	0.05	Toluene	0.03	Propyne	0.10
Iso-butane	0.05	Butene	0.03	Ethylbenzene	0.03		
Butane	0.05	Iso-butene	0.03	m,p-Xylene	0.03		
Iso-pentane	0.05	Trans-2-butene	0.03	o-Xylene	0.03		
Pentane	0.05	Cis-2-butene	0.03	1,3,5-Trimethybenzene	0.05		
2-Methylpentane	0.05	1,3-Butadiene	0.03	1,2,4-Trimethybenzene	0.05		
3-Methylpentane	0.05	Trans-2-pentene	0.03				
Hexane	0.05	Cis-2-pentene	0.03				
Cyclopentane	0.05	Isoprene	0.03				
Heptane	0.05	1-hexene	0.03				

VOCs: volatile organic compounds.

alkenes, alkynes and carbonyl compounds, could be analyzed. In China today, there are many data available on ambient aromatics but a lack of data on carbonyl compounds.

We analyzed 25 alkanes. These are ethane, propane, butane, iso-butane, pentane, iso-pentane, cyclopentane, 2,2-dimethyl butane, 2,3-dimethyl butane, 2-methyl pentane, 3-methyl pentane, hexane, cyclohexane, methyl cyclopentane, 2,3-dimethyl pentane, 2,4-dimethyl pentane, methyl cyclohexane, 2-methyl hexane, 3-methyl hexane, heptane, 2,2,4-trimethyl pentane, 2,2,4-trimethyl pentane, 2-methyl heptane, 3-methyl heptane, octane and nonane.

Fourteen ambient aromatics had data available: benzene, toluene, ethyl benzene, m, p-xylene, o-xylene, p-xylene, m-xylene, xylene, styrene, n-propyl benzene, cumene, mesitylene, 2,4-trimethyl benzene and 1,2,3-trimethyl benzene.

Thirteen alkenes had data: ethylene, propylene, butene, isobutene, 1,3-butadiene, trans-2-butene, cis-2-butene, pentene, trans-2-pentene, cis-2-pentene, isoprene, α -pinene and β -pinene. For alkynes, only acetylene data were available, so alkynes were analyzed together with alkenes.

Nine carbonyl compounds had data available: acetone, formaldehyde, benzaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, valeraldehyde, hexanal, and ethyl acetate.

2. Results and discussion

2.1. Characteristics of ambient VOCs

All concentrations of ambient VOCs in 23 major Chinese cities during the last 20 years are listed in Appendix A Table S1. The average concentration of ambient alkane is $32.00 \times 10^{-9} \pm 21.80 \times 10^{-9}$. C_2 – C_5 alkanes have higher concentrations; among these, the highest is propane ($7.10 \times 10^{-9} \pm 6.08 \times 10^{-9}$), followed by ethane ($4.50 \times 10^{-9} \pm 2.09 \times 10^{-9}$), iso-pentane ($4.19 \times 10^{-9} \pm 2.62 \times 10^{-9}$) and butane ($3.62 \times 10^{-9} \pm 2.79 \times 10^{-9}$).

The concentration of ambient aromatics is $34.08 \times 10^{-9} \pm 19.77 \times 10^{-9}$ in China (Appendix A Table S1), among which toluene was the highest ($10.83 \times 10^{-9} \pm 7.06 \times 10^{-9}$), followed by xylene ($7.37 \times 10^{-9} \pm 6.54 \times 10^{-9}$) and benzene ($6.81 \times 10^{-9} \pm 6.04 \times 10^{-9}$). Benzene is carcinogenic, but there is no limit for it in either the current Chinese national ambient air quality standard (NAAQS; GB3095-1996) or new standard (GB3095-2012). The United States has set the limit of benzene in ambient air

to 2.87×10^{-9} (Barbara et al., 2008). By this standard, the ambient benzene concentration in 15 cities is in exceedance: Beijing (4.42×10^{-9}), Guangzhou (14.16×10^{-9}), Shanghai (6.95×10^{-9}), Jiaxing (3.94×10^{-9}), Nanjing (3.76×10^{-9}), Hangzhou (6.30×10^{-9}), Macau (6.61×10^{-9}), Changchun (13.09×10^{-9}), Changzhou (20.10×10^{-9}), Ji'nan (20.47×10^{-9}), Lianyungang (11.36×10^{-9}), Nanning (4.71×10^{-9}), Zhengzhou (6.47×10^{-9}) and Dongguan (6.45×10^{-9}). Six cities are below the standard: Tianjin (0.92×10^{-9}), Anshan (1.89×10^{-9}), Shenyang (1.61×10^{-9}), Shaoxing (1.42×10^{-9}), Tai'an (0.94×10^{-9}) and Hong Kong (0.73×10^{-9}). This is in good agreement with a study by Duan et al. (2013). In addition, the national average of ambient benzene is $6.81 \times 10^{-9} \pm 6.04 \times 10^{-9}$, about five times the benzene limit in the European Union and England standard (1.43×10^{-9}), and about seven times the Japanese standard benzene limit (0.93×10^{-9}). This indicates that ambient benzene pollution is heavy in China.

Fewer species of alkynes are monitored in China today, and there are only some data for acetylene. Therefore, we analyzed acetylene together with alkenes. Based on the available studies, the concentration of ambient alkenes and alkynes in China is $22.40 \times 10^{-9} \pm 19.63 \times 10^{-9}$ (Appendix A Table S1); the most abundant is ethylene, with concentration $7.40 \times 10^{-9} \pm 4.62 \times 10^{-9}$, followed by acetylene ($6.27 \times 10^{-9} \pm 6.48 \times 10^{-9}$) and butene ($3.21 \times 10^{-9} \pm 3.73 \times 10^{-9}$). Concentrations of α -pinene ($1.17 \times 10^{-9} \pm 1.34 \times 10^{-9}$), isoprene ($2.26 \times 10^{-9} \pm 1.83 \times 10^{-9}$) and β -pinene ($0.98 \times 10^{-9} \pm 1.59 \times 10^{-9}$) emissions from plants are also high. Carbonyl compounds are a very important part of VOCs, and play a key role in atmospheric chemistry. However, studies on these compounds are few in number. The concentration of ambient carbonyl compounds in China is $14.16 \times 10^{-9} \pm 20.38 \times 10^{-9}$ (Appendix A Table S1); formaldehyde has the highest concentration ($11.24 \times 10^{-9} \pm 16.47 \times 10^{-9}$), followed by acetaldehyde ($5.83 \times 10^{-9} \pm 3.61 \times 10^{-9}$), and acetone ($4.02 \times 10^{-9} \pm 3.53 \times 10^{-9}$). This result is consistent with most previous studies (Chi et al., 2008; Feng et al., 2007; Xu et al., 2006; Lu et al., 2006; Pang and Mu, 2006).

Globally, VOC emissions from anthropogenic activities are lower than those from natural sources, but in China the emissions are equal, both 20 Mt/year (Yan et al., 2005; Klimont et al., 2002; Tonooka et al., 2001). In urban and intensive industrial regions, the amount of anthropogenic VOCs is much higher than that of natural VOCs. There are ambient

VOC data available from 23 Chinese cities. To analyze VOCs' spatial distribution, those cities were divided into north and south by a boundary running through the Qinling Mountains and Huaihe River. There are 11 northern cities: Beijing, Tianjin, Changchun, Qingdao, Ji'nan, Lianyungang, Shenyang, Zhengzhou, Tai'an, Anshan and Dongying. Twelve southern cities: Guangzhou, Shanghai, Jiaxing, Nanjing, Hongkong, Hangzhou, Nanning, Changzhou, Shaoxing, Dongguan, Panyu and Macao. Spatial distributions of alkanes, aromatics, alkenyl alkynes and carbonyl compounds are shown in Fig. 1. From this figure, the following conclusions can be drawn. For the various VOC species, alkanes have the highest concentration, followed by aromatics, alkenes, alkynes and carbonyl compounds. Based on the available literature, concentrations of ambient aromatics, alkanes, alkenes and alkynes in southern cities were slightly higher than those in the north, but concentrations of carbonyl compounds in the south were slightly lower than those in the north. As more studies become available, the spatial distribution of ambient VOCs in China may change.

2.2. Emission source identification

Alkanes with low numbers of carbon atoms are usually used to evaluate fossil fuel combustion emission. In Chinese cities, gasoline and liquefied petroleum gas (LPG) are the main fossil fuels for vehicles. Studies have shown that in gasoline exhaust gas, iso-pentane, pentane and hexane are very abundant. In diesel exhaust gas there are more alkanes with higher numbers of carbons. In LPG exhaust gas propane, butane and i-butane account for >90% of the total (Shi, 2010; Kwangsam et al., 2004; Sawyer et al., 2000; Nelson et al., 1983). Thus, one can conclude that ambient alkanes are mainly emitted by gasoline and LPG combustion. Most publications focus on urban atmospheric VOCs, and less on rural ones. Methylcyclohexane and heptane concentrations are higher, which means that diesel emission control needs more attention. Propane is relatively stable in the atmosphere and its life cycle exceeds one week, so it usually has high concentrations (Zhang, 2007).

Acetone is an organic or analysis solvent commonly used in industry, especially in painting and manufacturing factories. Because of its chemical stability, acetone has a long life in the atmosphere. Therefore, acetone emissions from a variety of sources accumulate in the atmosphere and reach high concentrations. The ratio of formaldehyde/acetaldehyde (C_1/C_2) is used to identify sources of carbonyl compounds. Usually in urban areas, C_1/C_2 is between 1 and 2, and in rural or forest areas it is 10. In the present study, C_1/C_2 was 1.52–2.50, indicating that most ambient carbonyl compounds are emitted in urban areas. However, this also indicates that rural or forest sources cannot be ignored.

2.3. Ozone formation potential

Atmospheric ozone and PM have long been a serious problem in China. Ozone is not emitted directly, and is formed by photochemical interactions of VOCs and nitrogen oxides (NO_x). Ambient VOCs mainly include NMHCs and carbonyl compounds. NMHCs are precursors producing ozone through OH radical-initiated oxidation and subsequent reactions with NO_x (Warneck, 1988). Carbonyl compounds are sources of free radicals and precursors of ozone and peroxyacetyl nitrates. Ambient VOCs are very important to atmospheric chemistry, and studies on the OFP of VOCs in China are still limited (Duan et al., 2008; Pang and Mu, 2006; Barletta et al., 2005; Wang et al., 2003). To estimate the photochemical reactivity of NMHCs and carbonyls, the maximum incremental reactivity (MIR) is often used (So and Wang, 2004; Carter, 1994). In this paper, the OFP of ambient VOCs is calculated by Eq. (1):

$$OFP_i = C_{VOCs} \times MIR_i, \quad (1)$$

where, OFP_i ($\mu g/m^3$) is the ozone formation potential of the i VOCs, C_{VOCs} ($\mu g/m^3$) is the concentration of the i VOCs, and MIR_i ($g\ O_3/g\ VOCs$) is the maximum incremental reactivity of the i VOCs. The MIR is cited from Development of the SAPRC-07 Chemical Mechanism and Updated Ozone Reactivity Scales by Carter (2010).

The percent of OFPs of various ambient VOC species in China are shown in Fig. 2. OFPs of ambient VOCs in Chinese

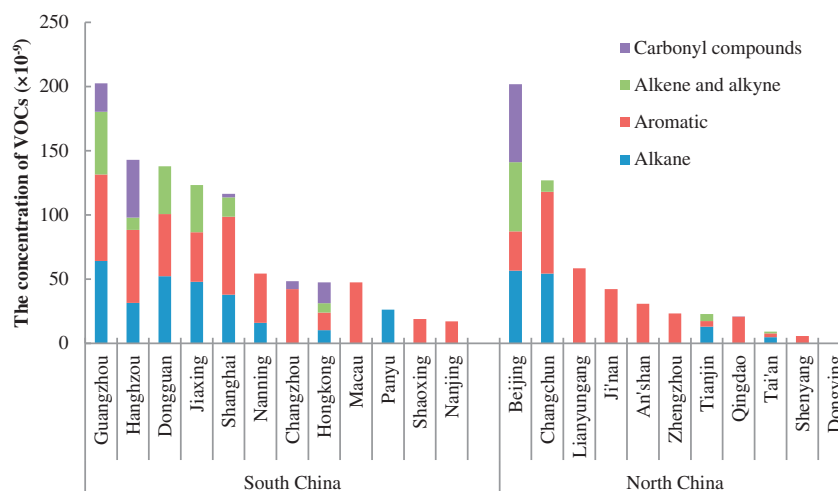


Fig. 1 – Spatial distribution of ambient volatile organic compounds in China.

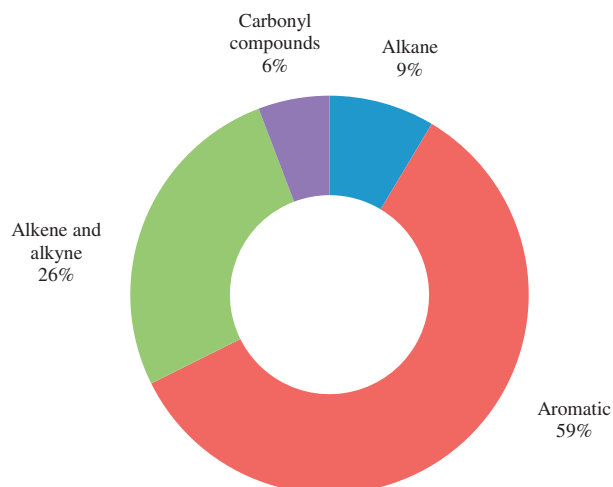


Fig. 2 – The percentage of ozone formation potential of various ambient VOCs species in China.

cities are shown in Fig. 3. Based on the MIR scale, the leading contributors to O_3 formation in the country are aromatics (59%), alkenes and alkynes (26%), alkanes (9%) and carbonyl compounds (6%). Ambient VOCs in the cities of Beijing, Guangzhou, Changchun and Jiaxing have much higher OFP ($2517.25 \mu\text{g}/\text{m}^3$, $1751.77 \mu\text{g}/\text{m}^3$, $1885.57 \mu\text{g}/\text{m}^3$ and $1663.90 \mu\text{g}/\text{m}^3$). These results mean that more VOC measurements should be taken, especially of aromatics and alkenes, to control O_3 pollution.

2.4. Secondary organic aerosol formation potential

Recently, haze pollution has been heavy in China, and $\text{PM}_{2.5}$ is the main pollutant in haze. SOAs constitute a very large portion of the mass concentration of ambient fine particles ($\text{PM}_{2.5}$), and VOCs are one of the most important precursors of

SOA formation. Parameterization is a practical approach in estimating the SOAFP (Grosjean, 1992), and therefore in identifying the relative contributions of VOC species to $\text{PM}_{2.5}$. We calculated the SOAFP of ambient VOCs by the following equation,

$$\text{SOAFP} = \text{VOC}_{\text{so}} \times \text{FAC} \quad (2)$$

where, VOC_{so} ($\mu\text{g}/\text{m}^3$) is the initial VOC and FAC is the fractional aerosol coefficient (Yuan et al., 2013; Nakao et al., 2011; Barbara and Sonia, 2007; Grosjean, 1992). In our study, SOAFP was calculated for 17 ambient VOC species, using concentration.

The aforementioned SOAFPs are shown in Fig. 4. The SOAFP of toluene is highest, about $57.42 \mu\text{g}/\text{m}^3$; followed by those of ethylbenzene, m/p-xylene, and o-xylene. Fig. 5 depicts the SOAFPs by city. Based on the available data, the SOAFPs of the 17 ambient VOC species in Hangzhou, Guangzhou and Changchun were the highest, and that in Tai'an was the lowest. In the first three cities, there is a well-developed painting industry that emits aromatics, one reason for the higher SOAFP there. Analysis of OFP and SOAFP should consider more factors such as UV radiation and NO_x emission.

3. Conclusions

According to literature statistics of various ambient VOC species in 23 Chinese cities over the last 20 years, up to 100 VOC species including alkanes, aromatics, alkenes, alkynes and carbonyl compounds were quantified. The VOC levels averaged $66.37(\pm 61.66) \times 10^{-9}$; among them alkanes and aromatics were the most abundant species, and compared with the available standards, benzene pollution was serious; The concentrations of ambient aromatics, alkanes, alkenes and alkynes in southern cities were slightly higher than those in the north, but concentrations of carbonyl compounds in the south were slightly lower than those in the north; Aromatics

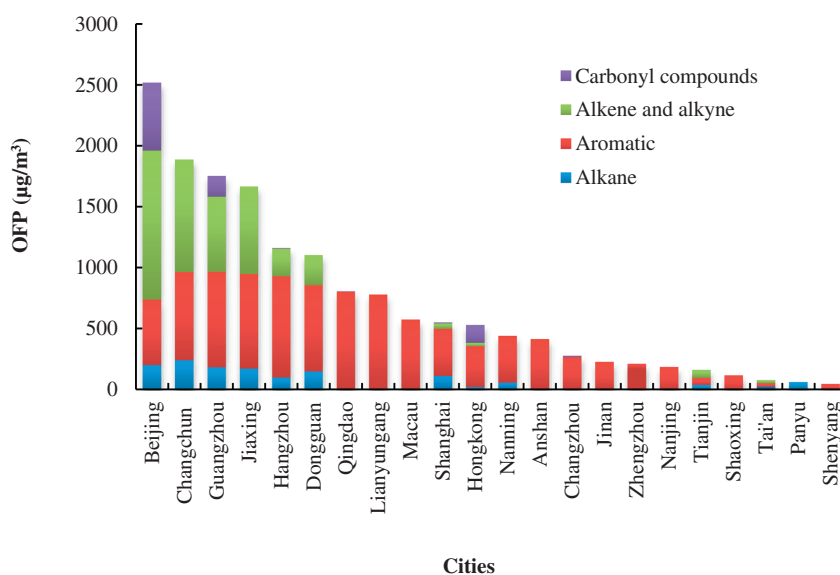


Fig. 3 – The ozone formation potential of ambient VOCs in Chinese cities.

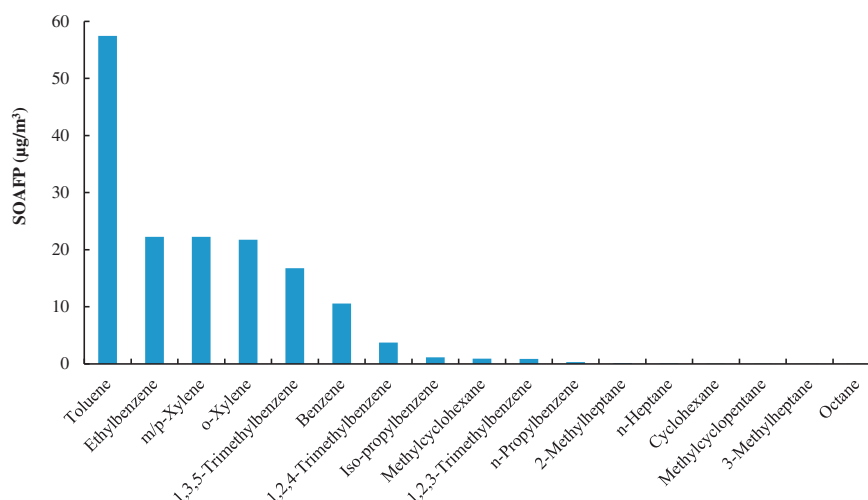


Fig. 4 – The secondary organic aerosol formation potential of 17 ambient VOCs species in China.

and alkenes had much higher OFPs, while aromatics had higher SOAFP; the OFPs of ambient VOCs in Beijing, Guangzhou and Changchun were very high, and SOAFP of ambient VOCs in Hangzhou, Guangzhou and Changchun were high. Based on the available studies, the main sources of VOCs in the ambient air in urban areas are mainly vehicle and industry emissions. Most of the studies on VOC pollution in the ambient air in China were carried out in Southeast China, and studies in Northwest China should be carried out as soon as possible. Currently, VOCs are not included in the environmental statistics system in China, so the lack of data makes it hard to fulfill environmental management requirements. We recommend future works such as investigative VOC monitoring both in the industrial emission sectors and in the ambient

air, the establishment of BAT reference documents based on up-to-date technologies, the preparation and updates of atmospheric emission standards, and amendment of NAAQS for ambient VOCs.

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.jes.2016.05.036>.

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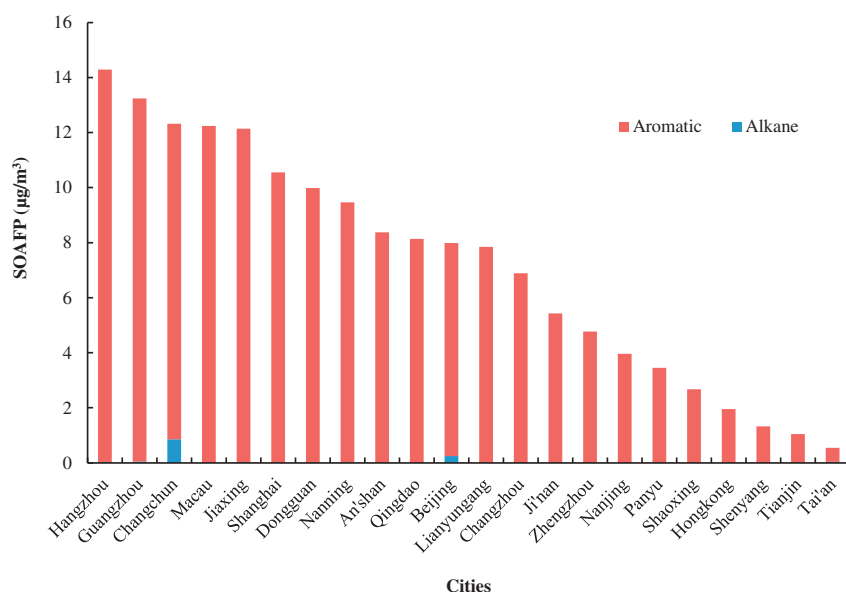


Fig. 5 – The secondary organic aerosol formation potential of 17 VOCs species in the ambient air of Chinese cities.

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