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Hazardous volatile organic compounds in ambient air of China

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Abstract: Volatile organic compounds (VOCs) are ubiquitous in the atmosphere and the majority of them have been proved to be detrimental to human health. The hazardous VOCs were studied highly insufficiently in China, despite the enormous emissions of VOCs. In this study, the concentrations and sources of 17 hazardous VOCs reported in literature were reviewed, based on which the health effects were assessed. In-depth survey indicated that benzene and toluene had the highest concentrations in eastern China (confined to the study regions reviewed, same for the other geographic generalization), which however showed significant declines. The southern China featured high levels of trichloroethylene. Dichloromethane and chloroform were observed to be concentrated in northern China. The distributions of 1,2-dichloropropane and tetrachloroethylene were homogeneous across the country. Basically consistent with the spatial patterns of ozone, the summertime formaldehyde exhibited higher levels in eastern and northern China, and increased continuously. While transportation served as the largest source of benzene and toluene, industrial emissions and secondary formation were the predominant contributors of halogenated hydrocarbons and aldehydes (formaldehyde and acetaldehyde), respectively. While chronic non-cancer effects of inhalation exposure to the hazardous VOCs were insignificant, the most worrying result was that the probabilities of developing cancers by inhaling the hazardous VOCs in ambient air of China were quite high. Formaldehyde was identified as the primary carcinogenic VOC in the atmosphere of most regions. The striking results, especially the high inhalation cancer risks, alerted us that the emission controls of hazardous VOCs were urgent in China, which must be grounded upon full understanding on their occurrence, presence and health effects.

Keywords: Volatile organic compounds, hazardous air pollutants, health effects, risk assessment, China

1. Introduction

With the increasing concerns of the public and research communities on environmental issues, volatile organic compounds (VOCs) have been recognized to be atmospherically relevant, through serving as the precursors of ozone and secondary organic aerosols (Carter, 1994; Kroll and Seinfeld, 2008). At the same time, the atmospheric oxidative capacity can be enhanced by the participation of VOCs in photochemical reactions (Lelieveld et al., 2008). Furthermore, many VOCs pose direct harm to human health (Brown et al., 1981; Huang et al., 2014), which are collectively named as hazardous VOCs hereafter. Hazardous VOCs are important members of the 187 hazardous air pollutants (HAPs) controlled by the US Environmental Protection Agency (EPA) (USEPA, 2019a), as shown in Table S1.

The species and abundances of hazardous VOCs are in general of highly spatial and temporal dependence. For example, toluene has long been recognized as the most abundant and primarily emitted hazardous VOC in Chinese cities (Liu et al., 2008a; Ou et al., 2015), which however is overwhelmed by tetrachloroethylene within dry-cleaning areas (Verberk et al., 1980). The temporal patterns of hazardous VOCs are related to the seasonal emission characteristics and the reactivity of the compounds. The northern China generally suffered from elevated air pollutants, including hazardous VOCs, in winter heating period (Yang et al., 2018). The commonly reported lower abundances of 1,3-butadiene and toluene at noon and in early afternoon are partially attributable to the quicker photochemical consumptions of these "C=C" bond containing VOCs (Tang et al., 2007; Lyu et al., 2016). This is opposite to the diurnal pattern of formaldehyde, which often presents a peak at noon caused by the secondary formation (Li et al., 2010; Lu et al., 2010). In general, anthropogenic emissions are the largest sources of hazardous VOCs (Li et al., 2010; Chen et al., 2017). The widespread sources of hazardous VOCs include agriculture, industries, power plants, transportation and residential sectors (Wei, 2008). However, the source contributions vary largely for different hazardous VOCs and in different regions (Guo et al., 2011; Wang, 2017). For example, while industrial emissions have been identified as the largest contributor to most halogenated hydrocarbons (Guo et al., 2009; Yuan, et al., 2013), tetrachloroethylene is mainly emitted from dry-cleanings (Raisanen et al., 2001).

China has been experiencing rapid economic growth for ~40 years since 1980s, with more than 80 folds increase in the nominal Gross Domestic Product (GDP). A sacrifice of the fast development is the arising of various environmental issues. The total emissions of non-methane VOCs (NMVOCs) in China have been increasing since 1990 (Figure 1 (Zheng et al., 2018; USEPA 2019b; EEA, 2019)). Until now no obvious reduction in emissions of NMVOCs has been achieved, though the emissions of many other air pollutants began to decrease in the mid-2000s or early 2010s (Lu et al., 2000; Zheng et al., 2018). According to the emission inventories, the total emissions of NMVOCs in China have overwhelmed those in Europe since 1997 and exceeded US emissions since 2004 (Figure 1). The current total NMVOC emissions in China are more than two and four folds higher than those in US and Europe, respectively. On one hand, scientists have already pointed out the urgency of controlling VOC emissions in China, for the sake of improving air quality (Zheng et al., 2009; Huang et al., 2014). On the other hand, the hazardous VOCs account for 20 – 40% of the total NMVOCs in China (Wei, 2009), imposing great threats to the health of residents. However, there has been no law or regulation specifically aiming to the control of hazardous VOCs in China, until the Ministry of Ecology and Environment of the People's

Republic of China (MoEE, PRC) released a "List of HAPs" in January 2019 (PRCMoEE, 2019). Even so, only six hazardous VOCs are included in the list, *i.e.* formaldehyde, acetaldehyde, dichloromethane, chloroform, trichloroethylene and tetrachloroethylene. Lack of systematic research on hazardous VOCs in China is also prominent in scientific community, though the individual species were separately discussed in a wide range of studies (Zhao et al., 2004; Zhou et al., 2011; Zhang et al., 2013).

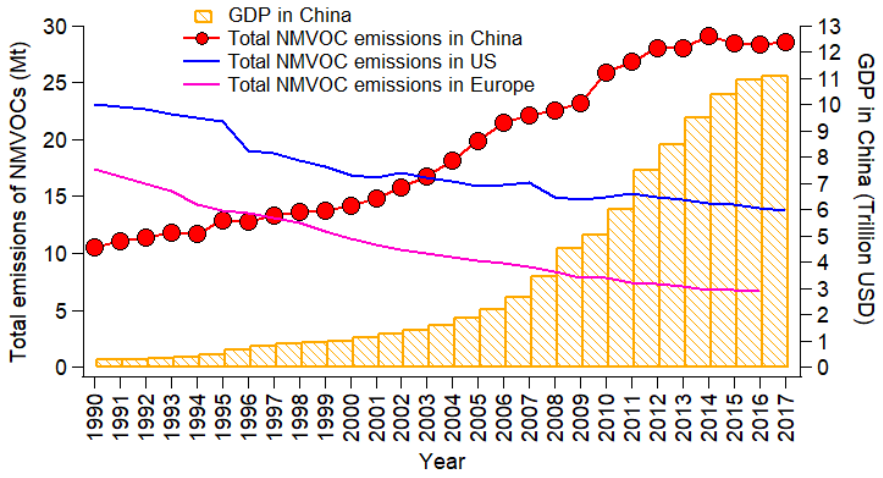

Figure 1 Total emissions of NMVOCs in China, US and Europe between 1990 and 2017.

Under the National Key R&D Program of China, the project "Towards an Air Toxic Management SYstem in China (ATMSYC, http://www.atmsyc.cn/)" inspired us to review the studies on hazardous VOCs in China. Our objectives were to advance the understanding on the spatial distributions and sources of hazardous VOCs in China, and the health risks of inhalation exposure to them. Due to the diversity and different characteristics of hazardous VOCs, we put our emphases on 17 hazardous VOCs in this review (Table 1), which belong to the 30 urban air toxics identified by US EPA due to their greatest threats to public health in most urban areas (USEPA, 2019c).

Table 1 Hazardous VOCs included in this review and their carcinogenicity, Reference Concentrations (RfCs) and Inhalation Unit Risks (IURs). Bold and italic fronts are the VOCs required to be controlled by MoEE, PRC.

No.	Compound	Chemical formula	Group *	Carcinogenicity	RfC (mg/m^3)	IUR ($m^3/\mu g$)
1	Benzene	C_6H_6	1	Carcinogenic	3×10^{-2}	7.8×10^{-6}
2	1,2-Dichloropropane (Propylene dichloride)	$C_3H_6Cl_2$			4×10^{-3}	1.0×10^{-5} *
3	***Trichloroethylene***	C_2HCl_3			2×10^{-3}	4.1×10^{-6}
4	Vinyl chloride (Chloroethylene)	C_2H_3Cl			1×10^{-1}	8.8×10^{-6}
5	***Formaldehyde***	CH_2O			9×10^{-3} #	1.3×10^{-5}
6	1,3-Butadiene	C_4H_6			2×10^{-3}	3×10^{-5}
7	Ethylene oxide	C_2H_4O			3×10^{-2} #	3×10^{-3}
8	***Dichloromethane (Methylene chloride)***	CH_2Cl_2	2A	Probably carcinogenic	6×10^{-1}	1×10^{-8}

9	***Tetrachloroethylene***	C_2Cl_4			4×10^{-2}	2.6×10^{-7}
10	***Chloroform***	$CHCl_3$	2B	Possibly carcinogenic	3×10^{-1} #	2.3×10^{-5}
11	1,3-Dichloropropene	$C_3H_4Cl_2$			2×10^{-2}	4×10^{-6}
12	1,2-Dichloroethane (Ethylene dichloride)	$C_2H_4Cl_2$			4×10^{-1} #	2.6×10^{-5}
13	1,1,2,2-Tetrachloroethane	$C_2H_2Cl_4$			-	5.8×10^{-5}
14	***Acetaldehyde***	C_2H_4O			9×10^{-3}	2.2×10^{-6}
15	Acrylonitrile	C_3H_3N			2×10^{-3}	6.8×10^{-5}
16	Quinoline	C_9H_7N			-	-
17	Toluene	C_7H_8	3	Not classifiable	5	-

* Classification according to the carcinogenic risks to humans identified by International Agency for Research on Cancer (IARC), referring to (IARC, 2019).

Values recommended by California Office of Environmental Health Hazardous Assessment (OEHHA, 2019). The others are adopted from Integrated Risk Information System (IRIS) of US EPA (USEPA, 2019d).

2. Methodology

2.1. Scope of the review

Figure 2 shows the locations of the cities where the concentrations and/or sources of at least one hazardous VOC were reviewed. To facilitate the discussions, the cities were grouped into several city clusters, including the North China Plain (NCP), the Yangtze River Delta (YRD), and the Greater Bay Area (GBA). Cities outside the three clusters were collectively assigned to the other regions. The NCP, YRD and GBA were the three most populated city clusters in China, with fast-growing economy, dense industries and busy traffics. Comparisons were made among the three city clusters throughout the review.

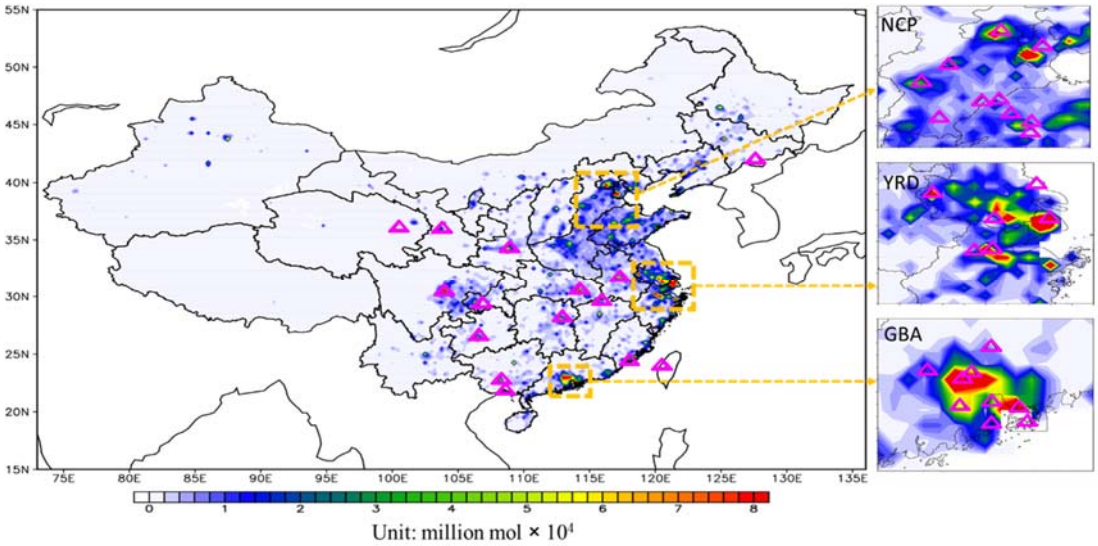

Figure 2 Geographical locations of the Chinese cities involved in this review (open triangles). Color on the map represents the average emissions of VOCs according to the Multi-resolution Emission Inventory for China (MEIC), 2016 (MEIC, 2016).

2.2. Review materials

Figure 3 summarizes the distributions of the sampling years of VOCs in more than 150 publications reviewed. Inevitably, there was a gap between the sampling years and the current period, with the median sampling year of 2013, 2008, 2007 and 2012 in the NCP, YRD, GBA and other regions in China, respectively, though the latest publications were given priority in our review. The earlier median sampling years in YRD and GBA were mainly due to the pioneering work done in Shanghai, Guangzhou and Hong Kong. In comparison, the results of sources of hazardous VOCs were more up-to-date, and the peak year around 2008 was also ascribed to studies in YRD and GBA. To enhance comparability, the sources reported in different publications were reasonably unified in this review, by combining or separating some factors, *e.g.* combination of gasoline and diesel vehicle exhausts into transportation. It should be noted that some hazardous VOCs were only reported in a few papers published more than a decade ago, such as 1,3-dichloropropene in GBA. We do not recommend over-interpretation on these compounds. Because of the lag of scientific publications, the results presented in this review cannot be directly regarded as an evaluation of the current air quality and air toxicity in China, but are still valuable given the fact that there is no decline for the national total emissions of VOCs (Figure 1). The spatial patterns were established by averaging the concentrations of hazardous VOCs and the source contributions to them by regions. To avoid misinterpretation, we reviewed the publications spanning a wide range of land uses (*i.e.* industrial, urban, suburban and rural areas) in each region. Though the median sampling year in YRD and GBA was 4-5 years earlier than that in NCP and other regions, the impacts on the conclusions of spatial patterns were multifaceted, due to the diverse trends of different hazardous VOCs in last decade. The trends of the concentrations and source contributions were discussed wherever they were available, based on which the actual spatial patterns were inferred. However, this should not waver in our conclusions on the nationwide average situations, because no state-level intervention has been imposed to decrease VOC emissions in China.

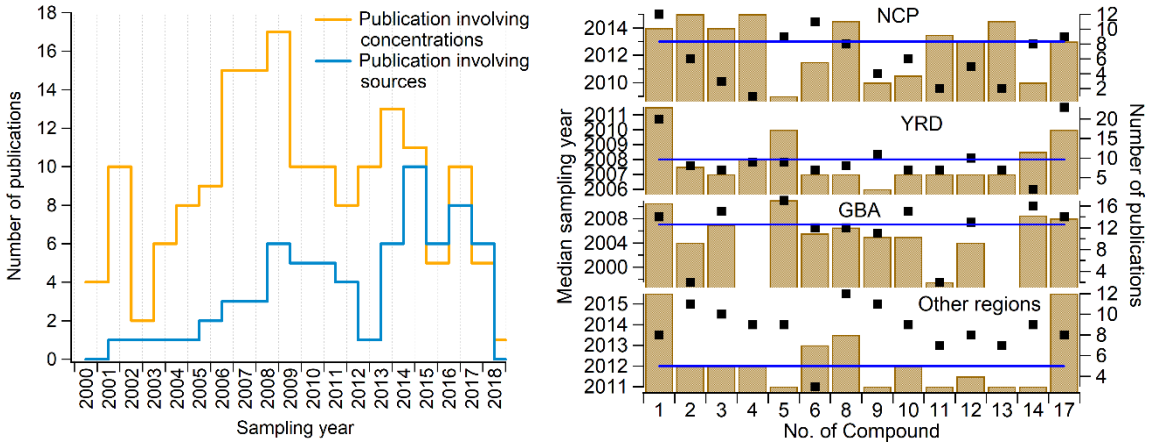

Figure 3 Left panel: Number of publications involving hazardous VOCs in sampling years in China. Right panel: Compound specified median sampling years (brown column) and number of publications (black square) in different regions. Blue line shows the median sampling year for all compounds. See Table 1 for details of No. of compound in right panel.

In addition, sponsored by the "ATMSYC" project, VOCs samples were collected in 18 cities across China (Figure S1 and Table S2) during January – February and June – August 2018. Text S1 describes the procedures of the collection and chemical analysis of the samples. The concentrations of the hazardous VOCs detected were also used to estimate the health effects of the hazardous VOCs in ambient air of China, as a validation of the review results.

2.3 Risk assessment metrics

The health risks of inhalation exposure to the hazardous VOCs were assessed by the Hazard Quotient (HQ) and Inhalation Cancer Risk (ICR), representing the chronic non-cancer effects and cancer effects, respectively. HQ and ICR are calculated following the formulas 1 – 2 (Zhou et al., 2013), where *CC* stands for the ambient concentrations of hazardous VOCs. *RfC* and *IUR* are provided in Table 1.

$$\mathrm{HQ} = \frac{CC}{RfC} \quad \text{(Formula 1)}$$

$$\mathrm{ICR} = \mathrm{CC} \times \mathrm{IUR} \quad \text{(Formula 2)}$$

The adverse chronic non-cancer effects are not likely to occur when the exposure concentrations (*CC*) are not higher than the *RfC* of an air toxic, *i.e.* HQ ≤ 1 (Zhou et al., 2013). Otherwise (HQ > 1), exposures to the air toxics are expected to cause chronic health effects other than cancer. The ICR represents the probability of developing cancers by lifetime exposure (70 years) to the air toxics under the concentration of CC (Zhou et al., 2013).

3. Spatial distributions of hazardous VOCs in China

3.1. Benzene & Toluene

As shown in Table S3, the mixing ratios of benzene were highest in YRD region, particularly in the urban and suburban areas. For example, the mixing ratio of 5.20 ppbv recorded at an urban site of Shanghai in the winter of 2007 was the highest value among all the studies we reviewed (Song et al., 2012). High levels of benzene were also documented in other studies (An et al., 2014; Han et al., 2017) conducted in Shanghai and Nanjing, two cities in YRD. Generally, comparable concentrations of benzene were observed between NCP and GBA except Hong Kong. Hong Kong, within GBA, had much lower benzene concentrations than those in the inland GBA cities, indicating the differences in local emissions of benzene. Furthermore, benzene observed at four background sites in different regions of China also followed the consistent spatial patterns. Taihu, as a background site in YRD, exhibited the highest benzene concentration (1.17±0.30 ppbv) (Zhang et al., 2015) among all the background sites, in contrast to the lowest value in background atmosphere of Hong Kong (0.51 ppbv) (Li et al., 2018). The highest concentrations of benzene in

YRD were possibly associated with the intensive petrochemical industries and transportation. For instance, the amount of crude oil refining (a large source of benzene) in YRD region was only second to that in Shandong province (CSY, 2018), and the vehicle population in YRD ranked the first in China (CSY, 2018). In contrast, there was nearly no high emission industry (such as oil refining and coking) in Hong Kong. Besides, a series of control measures have been taken in Hong Kong to reduce the emissions of VOCs (including benzene) from vehicles and solvent usages since 2000s (Lyu et al., 2017; HKEPD, 2019), which might also account for the relatively low levels of benzene in Hong Kong. However, it is striking to note that the mixing ratios of benzene in the background air of Hong Kong (0.51 ppbv) was comparable to or even higher than those in the urban areas of US (0.06 – 0.48 ppbv) (Baker et al., 2008) and Europe (0.46±0.19 ppbv) (EEA, 2018), not to mention much higher levels of benzene in other Chinese cities.

Toluene also exhibited the highest concentrations in YRD among the three city clusters (Table S4). Differently, the concentrations of toluene in GBA were at high levels, though they were slightly lower than those in YRD. A concurrent measurement of VOCs at an upwind (Conghua, 0.98±1.57 ppbv) and a downwind site (Jiangmen, 2.05±2.38) in GBA indicated that toluene concentrations were significantly built up after the air masses passed the GBA cities (Yuan et al., 2012). Thus, the local emissions to a large extent accounted for the high concentrations of toluene in GBA. Besides, the toluene concentrations in Hong Kong were comparable to those in the inland GBA cities, likely resulting from the intensive emissions from vehicles and solvent products (Guo et al., 2007; Ou et al., 2015). The ambient concentrations of toluene in NCP showed significantly seasonal and geographical variations. Beijing, as the capital city, had significantly lower levels of toluene than the other NCP cities, possibly thanks to the more stringent controls of vehicular and industrial emissions. Besides, notably higher concentrations of toluene were observed in winter. In addition to the lower boundary layer and less depletion in photochemical reactions, the stronger emissions in heating period played important role in elevating the wintertime toluene in NCP (Liu et al., 2015; Yang et al., 2018). The mixing ratios of toluene reported in the other Chinese cities were generally within the range of 1 – 2 ppbv, lower than those in the three city clusters. Overall, the ambient concentrations of toluene in China, particularly in the YRD and GBA, were higher than those in US (*e.g.* 0.12 – 1.54 ppbv in 28 US cities between 1999 and 2005 (Baker et al., 2008)) and Europe (*e.g.* 2.8 ppbv in 11 European cities during 2003-2008 (Geiss et al., 2011)).

Fortunately, benzene and toluene decreased in some Chinese megacities in the past decade. For example, a reduction rate of -5.6%/yr (-4.6%/yr) was identified for benzene (toluene) in Beijing from 2002 to 2013 (Wang et al., 2015). In Shanghai, benzene and toluene declined by -6.0%/yr and -4.9%/yr between 2007 and 2015, respectively (Gao et al., 2017). Hong Kong did not witness the significant decrease of benzene, however a downward trend (-3.4%/yr) was observed for toluene during 2005 – 2014 (Wang et al., 2017). The trends of benzene and toluene concentrations in many other Chinese cities are unknown to us. It is noteworthy that the median of statistical years in YRD was May 2011and 2010 for benzene and toluene, respectively, ~3 years earlier than those in NCP (Figure 3). Due to their high reduction rates in recent years, the concentrations of benzene and toluene in YRD may not be the highest anymore in China. In fact, the concentration of benzene in Shanghai, YRD was comparable to that in Guangzhou, GBA in the 2018 "ATMSYC" sampling

campaign, though Shanghai still had higher toluene concentration than Guangzhou and Beijing (data not shown).

3.2. Halogenated hydrocarbons

The mixing ratios of 9 halogenated hydrocarbons were reviewed and discussed in this section. As shown in Table S5, moderate and comparable levels of 1,2-dichloropropane were observed in Beijing, northern China (0.42 ppbv) (Gu et al., 2019), Shanghai, eastern China (0.39 ppbv) (Ran et al., 2009), Xinken, southern China (0.30 ppbv) (Shao et al., 2011) and Wuhan, central China (0.40 ppbv) (Hui, et al., 2018). However, the irregular and/or site-specific emissions might result in the large differences in 1,2-dichloropropane concentrations even within the same city. In NCP, the mixing ratios spanned from less than 0.1 ppbv to 1 ppbv, and the values of up to 1.01 ppbv and as low as 0.05±0.14 ppbv (Li et al., 2016) were reported in the urban atmosphere of Beijing. Furthermore, it is interesting to note that the concentrations at the background sites were not necessarily lower than those in the urban areas, *e.g.* 0.14±0.08 ppbv at a background site in Shandong province (Yucheng) (Zhu et al., 2016) and 0.05±0.14 ppbv in urban Beijing (Li et al., 2016). This might be related to the use of 1,2-dichloropropane containing pesticides in rural areas. It is noteworthy that the background levels of 1,2-dichloropropane in China were more than 100 times higher than that in North America (0.001 ppbv in Canada (Qadoumi et al., 2016)).

Table S6 summarizes the ambient concentrations of trichloroethylene. Roughly comparable levels were observed between NCP and YRD, which were lower than those in the inland cities of GBA, such as Guangzhou. The high levels of trichloroethylene (0.4 – 0.6 ppbv) in Guangzhou were documented in many publications (Liu et al., 2008a; Ling et al., 2011; Shao et al., 2011). Even at a background site in GBA, the mixing ratios of trichloroethylene (0.17±0.13 ppbv) were higher than those in the urban areas of Beijing (NCP) and Shanghai (YRD) (Wu et al., 2016). This might be associated with the widespread manufacturing of electronics in GBA, where trichloroethylene was commonly used as the degreasers (Waters et al., 1977). In contrast, much lower levels of trichloroethylene were observed in Hong Kong (Lau et al., 2010; Guo et al., 2013; Liu et al., 2019), the most developed city in GBA, benefited from the very few industries. In the other Chinese cities, the ambient trichloroethylene was on relatively low to moderate levels. Again, the concentrations of trichloroethylene in ambient air of China were higher than those in US and European countries. The background levels of trichloroethylene measured in Canada (< 0.008 ppbv) (Yokouchi et al., 1996) were even comparable to the North-Hemisphere (NH) background levels (0.003±0.001) (Koppmann et al., 1993).

Vinyl chloride in ambient air has been seldom studied in China. The incomplete review suggested that the YRD suffered from predominantly high concentrations (Table S7). Though Hefei had the most abundant vinyl chloride among the cities reviewed, it is a city in vicinity of YRD. Therefore, some specific emissions were responsible for the high levels of vinyl chloride in YRD or eastern China. Vinyl chloride is mostly used to produce polyvinyl chloride (PVC). The statistics of nationwide PVC productions indicated that eastern China undertook the largest fraction (34.5%) of the total PVC productions in China in 2007. The percentage decreased to 10.0% in 2016;

however, it was only lower than that in the northwestern (44.0%) and northern China (31.0%) (CSY, 2018). As such, the PVC productions might partially explain the high concentrations of vinyl chloride in eastern China. The PVC productions in northern China were mainly accomplished in Shandong province and Tianjin, where the ambient concentrations of vinyl chloride were not available. By only including the data in Beijing, the ambient concentrations of vinyl chloride in the NCP were most likely underestimated. Besides, it is striking to recognize the decadal changes of PVC productions in northwestern China (mainly Xinjiang, Inner Mongolia and Shaanxi) from 10.6% in 2007 to 44.0% in 2016 (CSY, 2018), representing the significant industry migration under China's "Go West" strategy. The migration of industries in China indicated the necessity of the nationwide monitoring of HAPs (including vinyl chloride), particularly in the less developing regions.

The spatial distributions of the other 6 halogenated hydrocarbons (Tables S8-S13) with fewer weights of evidence proving the carcinogenicity are discussed in Text S2. Overall, dichloromethane and chloroform were observed to have higher concentrations in NCP, which might be attributable to their applications as solvents, such as the uses in pharmaceutical industries. 1,2-dichloroethane was higher in NCP and YRD, likely associated with the productions of PVC and/or coal combustion. The distribution of tetrachloroethylene was relatively homogeneous across the country, while the spatial patterns of 1,3-dichloropropene and 1,1,2,2-tetrachloroethane were not summarized from the limited studies. Except for tetrachloroethylene, the ambient concentrations of hazardous halogenated hydrocarbons in China were tremendously higher than those in US and Europe. What's worse, most of these halogenated hydrocarbons are not continuously or regularly monitored, so their temporal trends are unknown. Hence, the spatial patterns reviewed for individual halogenated hydrocarbons here are instructive, though they inevitably have some uncertainties.

3.3. Formaldehyde & acetaldehyde

Figure 4 presents the monthly average column concentrations of formaldehyde (HCHO) in China observed by Ozone Monitoring Instrument (OMI) on the Aura platform. The concentrations in January and July 2016 are plotted to reflect the spatial distributions of tropospheric HCHO in winter and summer, respectively. Notably, the summertime HCHO was much more abundant than that in winter, indicating the significant productions of HCHO through photochemical reactions in summer. The NCP and YRD had the highest column concentrations of summertime HCHO, consistent with the spatial distribution of ozone (not shown). In contrast, the wintertime HCHO was mainly concentrated in NCP, though the relatively high concentrations were also observed in some places of southern China and Sichuan Basin, southwestern China. Since the low temperature and weak solar radiation in winter in NCP were unfavorable for the secondary formation of HCHO, primary emissions were expected to account for the high levels of wintertime HCHO in NCP.

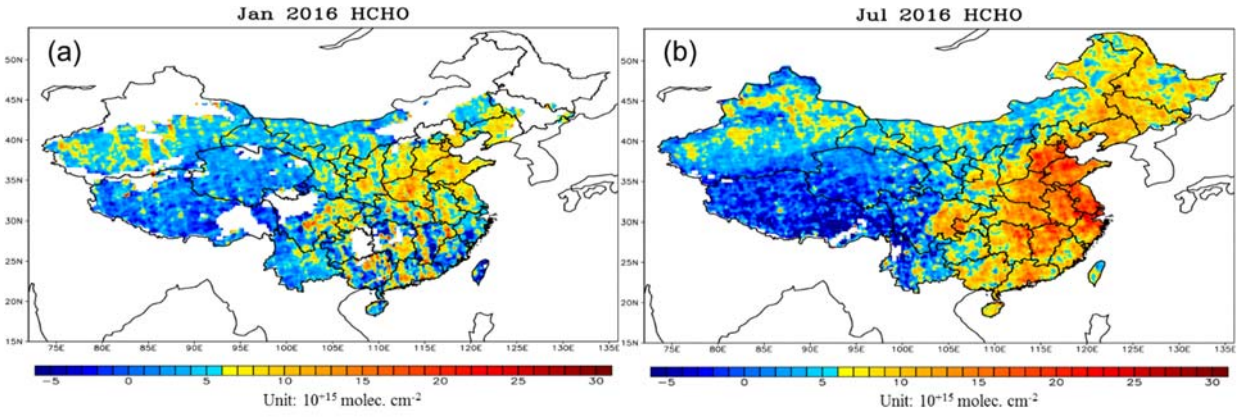

Figure 4 Distributions of the monthly average column concentrations of tropospheric HCHO across China in January 2016 (a) and July 2016 (b). Negative values are invalid readings of the very low concentrations and the blank areas in panel (a) are due to the lack of sunlight when the satellite passed these areas in winter (a drawback of the sun-synchronous orbit satellite).

Generally, the prevalence of HCHO in summer was also discernible from the ground-based observations, as shown in Table S14. The concentrations of surface HCHO followed the same spatial patterns as the tropospheric column concentrations, *i.e.* higher in NCP and YRD and lower in GBA and other regions of China. To our knowledge, the highest mixing ratios of HCHO were reported in Beijing, NCP (18.32 ppbv) (Sheng et al., 2018), followed by 16.60 ppbv in Hangzhou, YRD (Weng et al., 2009) and 15.80 ppbv in Shanghai, YRD (Huang et al., 2008). Though the concentration of up to 10.23 ppbv was ever determined for HCHO in Guangzhou, GBA (Lu et al., 2010), most studies reported the HCHO concentrations of lower than 7 ppbv in GBA. Despite the sparse industries, Hong Kong did not differentiate itself from the inland GBA cities in the ambient concentrations of HCHO, which might be due to the intensive vehicle emissions and secondary formation. The other regions in China had the HCHO concentrations roughly comparable to those in GBA.

In line with HCHO, acetaldehyde (CH_3CHO) also exhibited higher concentrations in summer, resulting from the photochemical reactions. The spatial distributions of CH_3CHO were also very similar to those of HCHO (Table S15). However, the differences were not as pronounced as those for HCHO. For example, the highest value in GBA was 7.12 ppbv (Lu, et al., 2010), only slightly lower than 8.10 ppbv in YRD (Huang et al., 2012) and 7.96 ppbv in NCP (Duan et al., 2012). Relatively lower concentrations were observed for CH_3CHO in the other regions of China, compared to those in the three city clusters.

To put China in the global context, we noticed that the concentrations of HCHO and CH_3CHO in China were several to more than ten times those in US and Europe (Tables S13-S14). Furthermore, studies (Zhu et al., 2018a) indicated the increasing trends of HCHO across China, due to the rise in emissions of anthropogenic NMVOCs (MEIC, 2016) and the enhancement of the oxidative capacity of the atmosphere (Li et al., 2019). The most obvious growth was observed in YRD and NCP, with a rate of 1.5%/yr and 1.1%/yr during 2005 – 2016), respectively (Shen et al., 2019). As such, the YRD and NCP are most likely the toughest regions suffering from ambient HCHO.

3.4. Other hazardous VOCs

1,3-Butadiene is mostly used to make rubber and plastics, and can also be emitted from the combustion processes, such as fuel combustion and biomass burning. As shown in Table S16, the ambient concentrations of 1,3-butadiene were generally low (<0.4 ppbv). The distributions were relatively homogeneous across China, with the upper limit of ~0.30 ppbv in NCP and GBA, and ~0.40 ppbv in YRD. The concentrations of 1,3-butadiene were generally higher in the megacities (*e.g.* Beijing, Shanghai, Hangzhou and Guangzhou) than in the other cities, indicating the dominance of vehicle exhausts in the sources of 1,3-butadiene. The ambient concentrations of ethylene oxide, acrylonitrile and quinoline have been seldom reported in China.

4. Sources of ambient hazardous VOCs

4.1. Sources of benzene and toluene

Figure 5 shows the source contributions to ambient benzene and toluene across China (Cai et al., 2010; Gao et al., 2016; Hsu et al., 2018; Hu et al., 2018; Huang et al., 2017, 2018; Hui et al., 2018; Lau et al., 2010; Li et al., 2015, 2016, 2017; Liu et al., 2014, 2016, 2017; Lyu et al., 2016; Mo et al., 2018; Sun et al., 2016; Wang, 2017; Wang et al., 2018; Wu et al., 2016; Yan et al., 2017; Yuan et al., 2013; Zhang et al., 2014, 2017 a,b, 2018 a,b; Zhu et al., 2018b). Nationally, transportation dominated the sources of benzene, with the average contribution of 48.3%. Also as the significant contributors, solvent usage, industrial emissions and combustion accounted for 16.1%, 13.8% and 11.3% of the ambient benzene, respectively. The other sources, such as residential activities and natural sources made minor contributions to benzene. No obvious spatial pattern was distinguishable for the contributions of transportation to the ambient benzene across China. Industrial emissions served as one of the largest sources of benzene in YRD, which contributed 35.7% to the ambient benzene in Shanghai (Cai et al., 2010; Zhang et al., 2013) and 73.0% in Ningbo (Wang, 2017). Specifically, 52.4% of the ambient benzene was assigned to the steel related industrial productions in Shanghai (Cai et al., 2010) and the industrial emissions of benzene in Ningbo were mainly (54.0%) originated from the petrochemical industry (Wang, 2017). This partially explained the highest concentrations of benzene in YRD. Combustion also played a role in building up the ambient concentrations of benzene in central and northern China. For example, coal combustion was responsible for more than 50% of benzene in Wuhan (Lyu et al., 2016; Hui et al., 2018). In contrast, it only made minor or negligible contributions to the ambient benzene in eastern and southern China, except for the contribution of 30% at a background site likely due to the domestic biofuel burning in the rural areas (Wu et al., 2016).

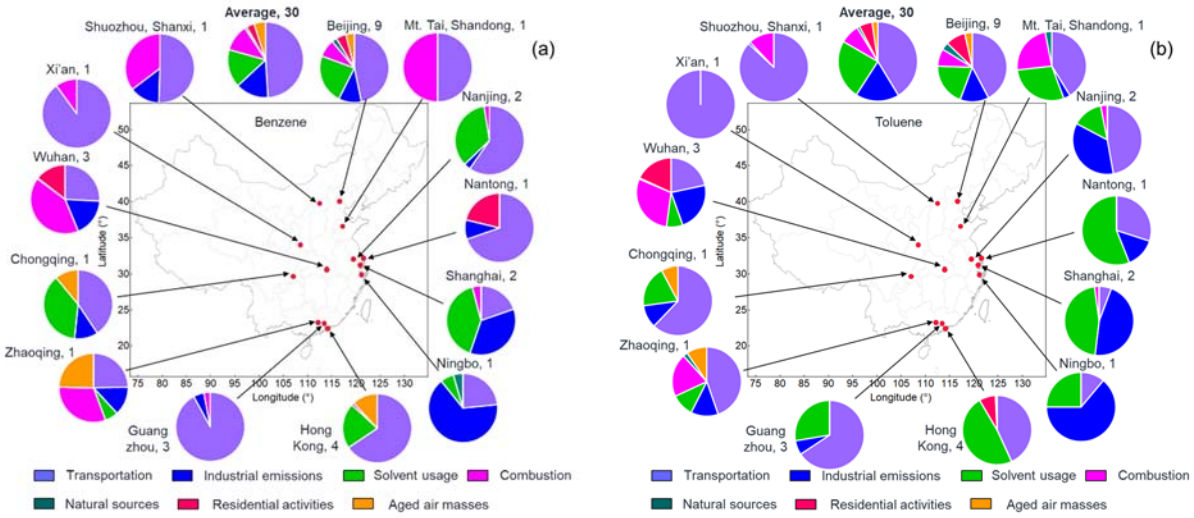

Figure 5 Source contributions to ambient benzene (a) and toluene (b) in different cities of China. Numbers following the city names indicate the numbers of publications where the source contributions were available and adopted to calculate the averages.

The ambient toluene in China was mainly originated from transportation (41.1%), solvent usage (24.1%) and industrial emissions (17.6%). The contribution of transportation to toluene (41.1%) was slightly lower than to benzene (48.3%), in contrast to the higher loadings of toluene (24.1%) than benzene (16.1%) in solvent usage. The latter coincided with the higher contents of toluene in most solvent products (Liu et al., 2008b; Lyu et al., 2017). The spatial patterns of the source contributions to toluene were similar to benzene. Transportation was the largest source of toluene in most cities, except for the higher contributions from industrial emissions in some cities of YRD. It seemed that solvent usage played more significant role in the sources of toluene in the megacities, such as the contribution of 47.7% in Hong Kong (Guo et al., 2007; Lau et al., 2010; Guo et al., 2011; Ou et al., 2015) and of 46.2% in Shanghai (Cai et al., 2010; Zhang et al., 2018). This might be due to the larger amounts of usages of solvent products (*e.g.* architectural paints and automobile coatings) in the more developed cities.

To conclude, transportation was the largest source of ambient benzene and toluene in Chinese cities with the overall contribution of 40 – 50%. Industrial emissions also made considerable contributions in YRD, eastern China. Solvent usage was more responsible for the ambient toluene, and played significant roles in building up toluene concentrations in the megacities.

4.2. Sources of hazardous halogenated hydrocarbons

The sources of hazardous halogenated hydrocarbons are ubiquitous and complicated. As shown in Table S17, most of them can be emitted from industrial sectors. The residential activities are also potential sources, such as the emissions of tetrachloroethylene, dichloromethane, dichloroethane and trichloroethylene from dry cleaning (Wang, 2010), chloroform and dichloromethane from the obsolete air conditioners (Wang, 2010) and dichloromethane from catering (Zhang and Ma, 2011). Combustion mainly accounts for the emissions of light halogenated hydrocarbons containing 1-2 halogen atoms. For example, biomass burning is widely recognized as a source of methyl chloride

and dichloromethane (Rudolph et al., 1995), while 1,2-dichloroethane can be emitted from coal combustion (Lyu et al., 2016). Besides, the hazardous halogenated hydrocarbons emitted from transportation are generally dichloromethane, dichloroethane and chloroform (Li et al., 2015, 2016; Wang et al., 2014, 2018), which are also typical species in oceanic and plant emissions – the natural sources (Wang, 2010).

To avoid the over-complexity, the source contributions to the 9 hazardous halogenated hydrocarbons were not discussed individually. Instead, Figure 6 shows the overall contributions of different sources to the total hazardous halogenated hydrocarbons across China (Cai et al., 2010; Guo et al., 2009; Lau et al., 2010; Li et al., 2015, 2016; Liu et al., 2017; Lyu et al., 2016; Wang, 2017; Wang et al., 2018; Wu et al., 2016; Yuan et al., 2013; Zhang et al., 2014, 2018 a,b; Zhu et al., 2018b). It should be noted that some compounds, which are not belonging to the 17 hazardous VOCs (Table 1) such as methyl chloride, carbon tetrachloride and chlorobenzene, were also included in the statistics. For the reasons, it was difficult to subtract their concentrations from the total concentrations reported in many publications. In addition, they are also detrimental to human health, which are just not given priority to be controlled by US EPA.

According to Figure 6, the industrial emissions constituted the largest source of the ambient hazardous halogenated hydrocarbons in China, with the nationwide average contribution of 37.0%, followed by solvent usage (22.9%), transportation (17.1%) and combustion (15.7%). It is noteworthy that the solvent usage here denoted the solvents used in both industries and residential activities. Therefore, the contributions of industrial emissions were even underestimated. Furthermore, the industrial emissions were more significant in southern, eastern and central China, particularly in GBA where 61.5% of the total hazardous halogenated hydrocarbons were originated from industrial emissions (Yuan et al., 2013). However, solvent usage ranked the first in Hong Kong, mainly attributable to the uses of household products and dry-cleaning activities (Guo et al., 2009; Lau et al., 2010). Solvent usage also played important roles in central and eastern China, where it mainly referred to the uses of industrial solvents (Wang, 2017; Zhang et al., 2018). No obvious spatial pattern was identified for the contributions of transportation and combustion.

To sum up, the hazardous halogenated hydrocarbons in the ambient air of China were predominantly emitted from industrial sources, with considerable contributions from solvent usage, transportation and combustion. However, studies on the sources of hazardous halogenated hydrocarbons in China were quite insufficient, leading to uncertainties in the source contributions summarized above. To clearly understand the sources of hazardous halogenated hydrocarbons, more comprehensive studies are required.

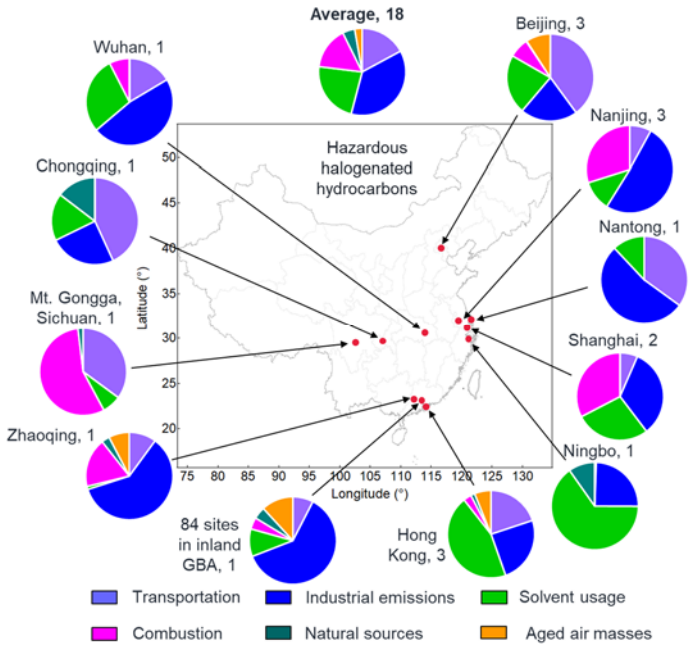

Figure 6 Source contributions to the total hazardous halogenated hydrocarbons in different Chinese cities. Numbers following the city names indicate the numbers of publications where the source contributions were available and adopted to calculate the averages.

4.3. Sources of HCHO and CH_3CHO

The ambient HCHO and CH_3CHO are derived from not only primary emissions but also secondary formation. The ever identified primary sources include vehicle exhausts, biomass burning, biofuel burning, coal combustion, industrial emissions and solvent usage, as well as the plant emissions. The HCHO directly emitted from plants and that formed with the plant emissions as precursors were separately discussed. For the integrated HCHO values reported as a whole, the primary and secondary contributions were decomposed, according to the emission ratio of HCHO/isoprene (0.98 ± 0.07 ppbv/ppbv) (Yuan et al., 2012) and the yield of HCHO from isoprene oxidation (0.57 – 0.63) (Atkinson and Arey, 2003). The HCHO and CH_3CHO assigned to the background and aged air masses might include both the primarily emitted and secondarily formed parts.

Figure 7 shows the source contributions to ambient HCHO and CH_3CHO across China (Chen et al., 2014; Han et al., 2019; Huang et al., 2019; Li et al., 2010, 2014; Liu et al., 2008, 2017; Ling et al., 2016, 2017; Ma et al., 2019; Su et al., 2019; Wang et al., 2015, 2017; Yang et al., 2017; Yuan et al., 2012; Zhou et al., 2019; Zhu et al., 2019). Secondary formation dominated the sources of HCHO, with the nationwide average contribution of 43.0%. In fact, the contribution was most likely underestimated, because the secondarily formed HCHO could also exist in the background and aged air masses, which accounted for 12.6% and 2.2% of the ambient HCHO, respectively. 33.3% of the HCHO was contributed by primary emissions, among which the contributions of vehicle exhausts (9.9%), combustion (6.1%), biogenic emissions (4.0%) and solvent usage (2.0%) were separately quantified, while the rest (11.4%) were not resolved into specific sources.

Secondary formation played more important roles in eastern and southern China. The contribution of secondary formation to HCHO reached as high as 68% in Hong Kong (Ling et al., 2016). According to the studies where the biogenic emissions were identified, the southern China also had higher biogenic emissions of HCHO (*e.g.* 21.3% in Shenzhen *vs.* 5.9% in Beijing), which might be due to the more vegetation and higher temperature in southern China. While combustion made minor contributions in most cities, it was responsible for up to 53.2% of HCHO at a suburban site in Ziyang, Sichuan, resulting from the intensive biomass burning (Li et al., 2014). There was no discernible spatial pattern for the contributions to HCHO of the other sources.

For the sources of CH_3CHO, primary emissions (51.5%) overrode secondary formation (30.7%), without the consideration of the primarily emitted and/or secondarily formed CH_3CHO in the sources of background and aged air masses. Biogenic emissions (16.1%) ranked the first among the primary sources, followed by vehicle exhausts (11.6%), combustion (3.9%) and solvent usage (3.7%). It is worth noting that the biogenic emissions might be overestimated, because the secondarily formed CH_3CHO with biogenic VOCs as precursors were not separated from the biogenic emissions in some publications. Besides, 16.3% of the primary emissions were not resolved to the specific sources. The spatial patterns of the source contributions to CH_3CHO were not distinguishable, which might be due to the limited number of studies and the inconsistencies in site categories, study periods and source definitions.

Overall, secondary formation was the largest contributor to ambient HCHO in Chinese cities, while primary emissions dominated the sources of CH_3CHO. Vehicle exhausts and biogenic emissions also made certain contributions to both HCHO and CH_3CHO. To advance our understandings on the sources of HCHO and CH_3CHO, integrated studies at national scale are necessary. The sources of unresolved primary emissions, background and aged air masses should be further resolved.

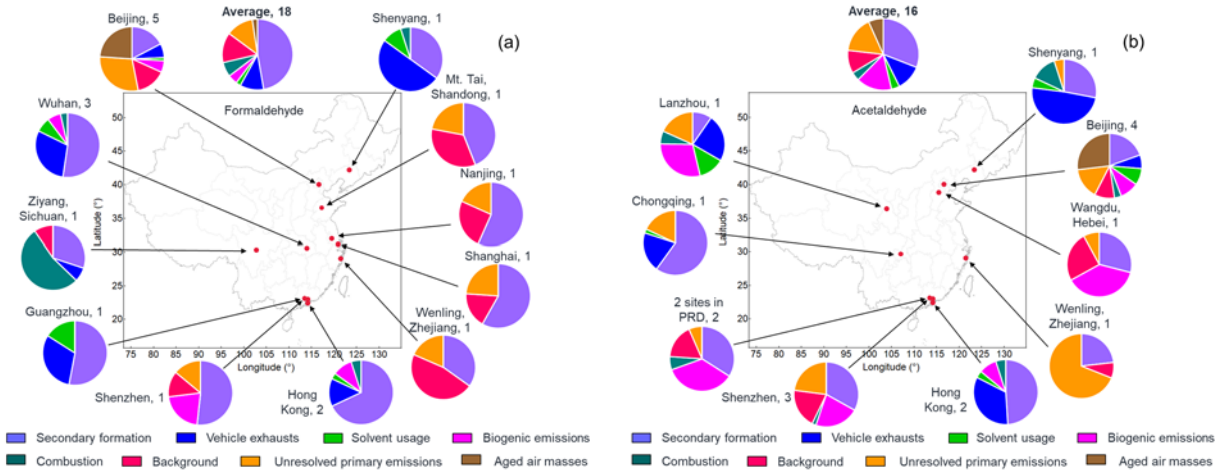

Figure 7 Source contributions to ambient HCHO (a) and CH_3CHO (b) in different Chinese cities. Numbers following the city names indicate the numbers of publications where the source contributions were available and adopted to calculate the averages.

5. Risk assessment of inhalation exposure to hazardous VOCs in China

As stated in section 2.3, the health risks of inhalation exposure to the hazardous VOCs were assessed by HQs and ICRs, reflecting the chronic non-cancer and cancer risks, respectively. Figure 8 (a) shows the average HQs of the hazardous VOCs, whose concentrations and RfCs are available. Overall, the inhalation exposures to the hazardous VOCs in ambient air of China were not likely to be associated with the non-cancer toxicity, with the HQs less than 1. However, it did not mean that the chronic health effects could be completely eliminated across the country. Specifically, the detrimental effects were expectable for the exposure to HCHO (HQ = 1.22) in the NCP and exposure to HCHO (HQ = 1.33) and CH_3CHO (HQ = 1.26) in the YRD. While the average HQs of HCHO were below 1 in the GBA, the occasionally reported high concentrations of HCHO in some cities indicated the non-cancer risks, *e.g.* HQ = 1.52 and 1.03 in Guangzhou in the summer of 2005 and 2006, respectively. The situations were the same for inhalation exposure to CH_3CHO. Attentions may also be paid to trichloroethylene in Guangzhou (HQ = 1.23 – 1.55 in the 2000s), and 1,2-dichloropropane in Beijing (HQ = 1.27 in the autumn of 2014). In fact, studies (McCarthy et al., 2009) reminded that the potential risks existed for the exposure to HAPs, provided that HQs were higher than 0.1. Accordingly, the potential concerns on the exposures to HCHO, CH_3CHO, trichloroethylene, 1,2-dichloropropane, benzene and 1,3-butadiene were not unnecessary, due to the nationally average HQs of higher than 0.1 for these compounds (Figure 8 (a)).

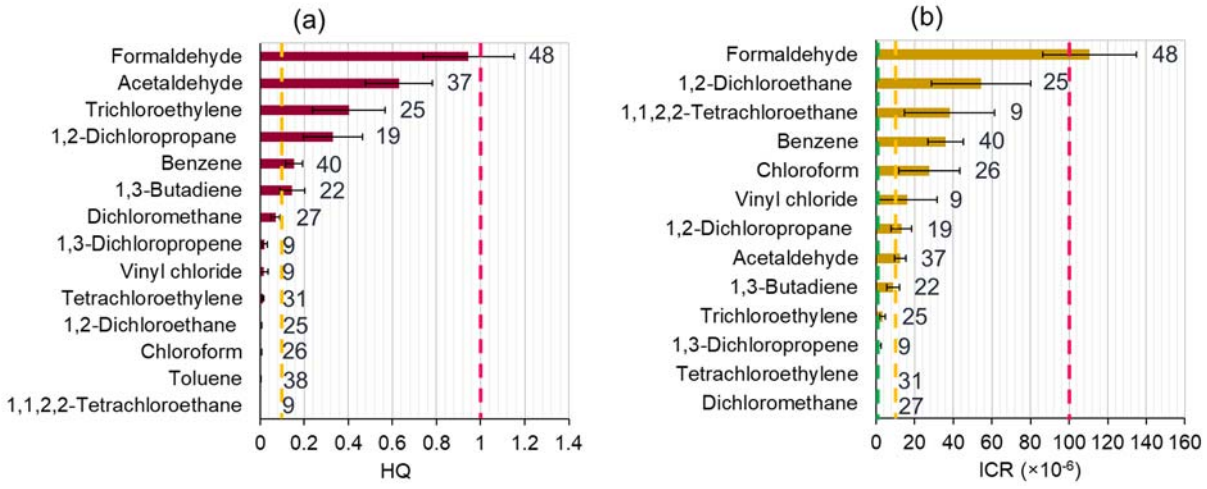

Figure 8 Nationwide average HQs (a) and ICRs (b) of hazardous VOCs in China. Red and orange lines in panel (a) stand for HQ of 1 and 0.1, respectively. Red, orange and green lines in panel (b) indicate the ICR of 1×10^{-4}, 1×10^{-5} and 1×10^{-6}, respectively. Numbers at the end of the columns are the number of publications where the concentrations of the corresponding VOCs are adopted in calculations.

The nationwide average cancer risks of exposure to the ambient hazardous VOCs are presented in Figure 8 (b). According to previous studies (Sexton, et al., 2007), the cancer risks were classified as "definite risk" (ICR > 1×10^{-4}), "probable risk" (1×10^{-5} < ICR < 1×10^{-4}) and "possible risk" (1×10^{-6} < ICR < 1×10^{-5}). ICR of 1×10^{-6} was the benchmark of acceptable exposure level recommended by US EPA (USEPA, 2019d). It is striking to note that the total probability of developing cancers through exposures to the hazardous VOCs in ambient air of China was 3.22×10^{-4}. Namely, the hazardous VOCs in the ambient air of China definitely enhanced the cancer risks

for the residents who breathe the air. HCHO was identified as the primary carcinogen in the air, with the ICR of 1.11×10^{-4} ("definite risk"), followed by 1,2-dichloroethane (0.54×10^{-4}), 1,1,2,2-tetrachloroethane (0.38×10^{-4}), benzene (0.36×10^{-4}) and chloroform (0.28×10^{-4}). The acceptable cancer risks were only identified for the exposures to tetrachloroethylene and dichlomethane. Furthermore, the ICRs of hazardous VOCs were compared among different regions of China, including the NCP, YRD, GBA and the other regions (mainly the central and southwestern China), as shown in Figure 9. The exposures to hazardous VOCs in the YRD had the highest cancer risk (3.96×10^{-4}), in contrast to the lowest risk in the GBA (1.51×10^{-4}). However, even the lowest cancer risk was at the level of "definite risk" (ICR > 1×10^{-4}). HCHO was the largest contributor to the total ICRs in most regions of China, *i.e.* 1.43×10^{-4} (36.1%) in the NCP, 0.73×10^{-4} (48.1%) in the GBA and 0.89×10^{-4} (33.6%) in the regions other than the three city clusters. In the YRD, the highest cancer risk was due to exposure to 1,2-dichloroethane (1.72×10^{-4}), slightly higher than 1.56×10^{-4} for exposure to HCHO. In fact, the ICR of 1,2-dichloroethane was only second to that of HCHO in the NCP and the regions other than the three city clusters. Benzene also significantly enhanced the cancer risks of inhalation exposure in the GBA. Although a significant reduction in benzene was observed in NCP and YRD (section 3.1), the reduction in total ICRs was estimated to be less than 2%. Therefore, the conclusion that hazardous VOCs exhibit high ICRs in YRD and NCP will be held, unless there is new evidence that other species are greatly reduced.

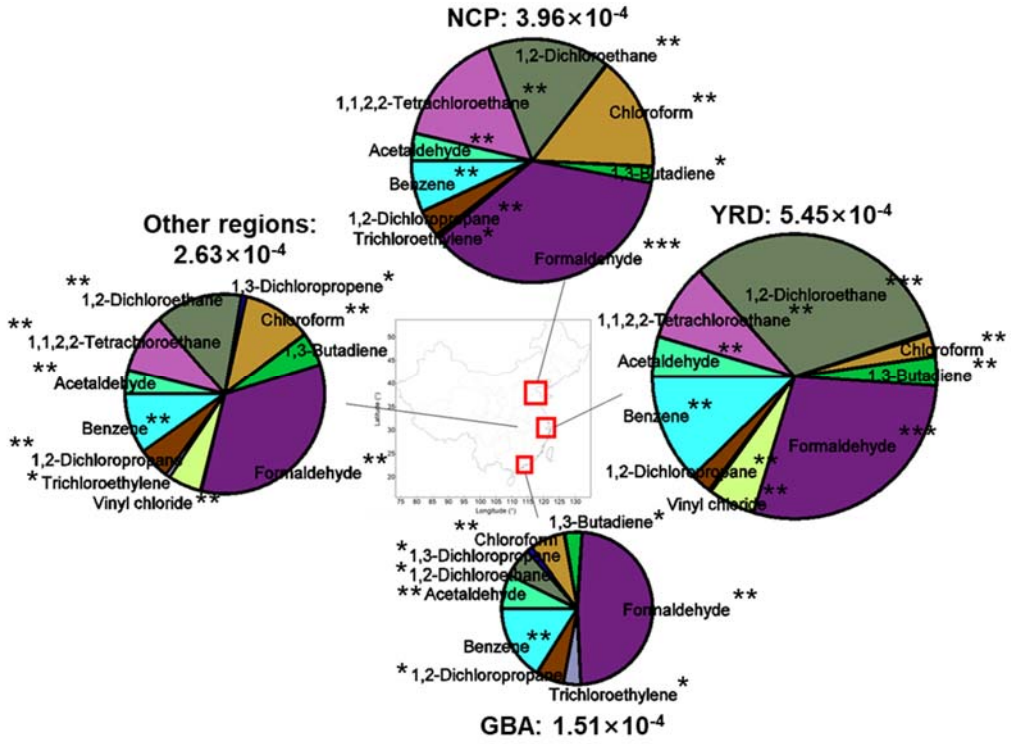

Figure 9 ICRs of exposure to hazardous VOCs among different regions in China. The areas of the pie charts represent the relative levels of ICRs. ***, ** and * denote the "definite risk", "probable risk" and "possible risk", respectively. Numbers of values for averages calculation are given in Table S18.

As verification, the health risks of inhaling the hazardous VOCs in ambient air of China were assessed based on the concentrations of VOCs collected in January – February 2018 across China under the "ATMSYC" project (Figure S2). The compounds of concern in terms of the chronic non-cancer risks mainly included HCHO, CH_3CHO, 1,3-butadiene, benzene and trichloroethylene, in the descending order of HQ. Except for 1,2-dichloropropane that was not analyzed, the compounds were exactly the same as the review results. Consistently, the ICRs of most hazardous VOCs exceeded the acceptable level (1×10^{-6}) in China, and HCHO was the main source of the total ICR. Benzene and chloroform, to which the inhalation exposure caused "probable" cancer risks according to the published data, also presented significant carcinogenic effects in the 2018 sampling campaign. The other hazardous VOCs with ICRs in the range of 1×10^{-5} – 1×10^{-4} were not analyzed, except for acetaldehyde whose ICR decreased to lower than 1×10^{-5}. It is interesting to note that the HQs and ICRs of most VOCs in the 2018 sampling campaign were lower than those calculated from the published data. For HCHO and CH_3CHO, this might be due to the lower concentrations in winter. For the other compounds, the air pollution control measures taken in recent years might work in decreasing their ambient concentrations. However, the health risks of inhalation exposure to 1,3-butadiene were even enhanced in 2018, while the causes are unknown to us. The spatial distributions of the total ICRs are not compared, because the 2018 sampling campaign did not focus on the three city clusters, but the large cities across the country. For example, Lanzhou in northwestern China was identified as the city having the highest ICRs, while the ICRs were relatively low in Beijing and Shanghai, likely due to the stringent air pollution controls in the megacities.

To sum up, HCHO imposed the greatest threat to the health of Chinese citizens among the ambient hazardous VOCs. The inhalation exposures to CH_3CHO, benzene, 1,3-butadiene, trichloroethylene and chloroform were also concerns. The other halogenated hydrocarbons with small sample sizes reviewed and not analyzed in the 2018 sampling campaign, such as 1,2-dichloroethane, 1,1,2,2-tetrachloroethane and 1,2-dichloropropane might also be detrimental to human health but with relatively high uncertainties.

6. Discussion and Conclusions

Hazardous VOCs in the ambient air are serious threats to human health, which however have not been systematically studied in China. This review discussed the spatial distributions, sources and health effects of 17 hazardous VOCs across the country. Overall, YRD in eastern China and NCP in northern China suffered from higher concentrations of most hazardous VOCs, such as benzene, toluene, vinyl chloride, dichloromethane, chloroform and 1,2-dichloroethane, mainly resulting from the industrial emissions and transportation. The summertime formaldehyde also exhibited higher levels in YRD and NCP. This was mostly explained by the secondary formation with stronger oxidative capacity of the atmosphere in both regions, as indicated by the spatial patterns of ozone. Though GBA in southern China had relatively lower concentrations of most hazardous VOCs, trichloroethylene exhibited high concentrations, likely due to the applications of

trichloroethylene as industrial solvent. Overall, the nationwide average concentrations of all the hazardous VOCs were lower than the levels causing chronic non-cancer effects. However, the potential risks could not be eliminated, particularly for the exposures to formaldehyde. What's worse, the cancer risks of inhalation exposures to ambient VOCs in China were substantially higher than the recommended benchmark. It seemed that the residents in YRD and NCP bore high risks of developing cancers by inhaling the hazardous VOCs in the air. Among all the ambient VOCs, formaldehyde presented the biggest threat to the health of Chinese citizens, regardless of non-cancer or cancer effects, and the situation is getting worse with the increase of formaldehyde concentrations. In addition to toluene whose inhalation unit risk was not available, only tetrachloroethylene and dichlomethane were identified to raise acceptable inhalation cancer risks. While 6 VOCs are specified in the list of HAPs proposed by MoEE, PRC, more VOCs, such as benzene, 1,3-butadiene and some halogenated hydrocarbons, should be included.

Despite uncertainties reminded where necessary, this is the first review of hazardous VOCs in China. However, comprehensive studies on the hazardous VOCs at national scale are required. Observations concurrently performed in different cities across the country will help to unambiguously understand the distributions of hazardous VOCs, as well as the sources. A health effect – based list of hazardous VOCs should be proposed. Regular monitoring and emission control of the compounds with significant health effects are necessary.

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