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Risk assessment of population inhalation exposure to volatile organic compounds and carbonyls in urban China

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ABSTRACT

Over the past three decades, China has experienced rapid urbanization. The risks to its urban population posed by inhalation exposure to hazardous air pollutants (HAPs) have not been well characterized. Here, we summarize recent measurements of 16 highly prevalent HAPs in urban China and compile their distribution inputs. Based on activity patterns of urban Chinese working adults, we derive personal exposures. Using a probabilistic risk assessment method, we determine cancer and non-cancer risks for working females and males. We also assess the uncertainty associated with risk estimates using Monte Carlo simulation, accounting for variations in HAP concentrations, cancer potency factors (CPFs) and inhalation rates. Average total lifetime cancer risks attributable to HAPs are 2.27×10^{-4} (2.27 additional cases per 10,000 people exposed) and 2.93×10^{-4} for Chinese urban working females and males, respectively. Formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene are the major risk contributors yielding the highest median cancer risk estimates, $>1 \times 10^{-5}$. About 70% of the risk is due to exposures occurring in homes. Outdoor sources contribute most to the risk of benzene, ethylbenzene and carbon tetrachloride, while indoor sources dominate for all other compounds. Chronic exposure limits are not exceeded for non-carcinogenic effects, except for formaldehyde. Risks are overestimated if variation is not accounted for. Sensitivity analyses demonstrate that the major contributors to total variance are range of inhalation rates, CPFs of formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene, and indoor home concentrations of formaldehyde and benzene. Despite uncertainty, risks exceeding the acceptable benchmark of 1×10^{-6} suggest actions to reduce exposures. Future efforts should be directed toward large-scale measurements of air pollutant concentrations, refinement of CPFs and investigation of population exposure parameters. The present study is a first effort to estimate carcinogenic and non-carcinogenic risks of inhalation exposure to HAPs for the large working populations of Chinese cites.

1. Introduction

Public concern regarding organic hazardous air pollutants (HAPs) continues to grow world-wide. HAPs have a wide range of sources, effects and exposure routes. Their sources include indoor sources (Clarisse et al., 2003; Kim et al., 2001), outdoor vehicle emissions and industrial combustion (Guo et al., 2004b; Ohura et al., 2009). Their adverse health effects range from irritation of eyes, skin, mucous membranes and respiratory tract (Jones, 1999; WHO, 2010) to serious chronic illnesses such as asthma (Weisel, 2002), chronic obstructive pulmonary disease (Viegi et al., 2006), cardiovascular disease and cancer (Lewtas, 2007; WHO, 2010). Because of their relatively low boiling points and high vapor pressures, the main exposure pathway to most HAPs is through inhalation (Ramirez et al., 2012).

Due to rapid industrial and economic development over the past three decades, China has experienced large migration from rural areas to cities; urban growth and modernization; and a concomitant increase in urban air pollution (Fang et al., 2009; Zhang et al., 2013b). From 1990 to 2010, China's urban population more than doubled; net urban residential building area grew from 4 to 21 billion m^2; and the number of motor vehicles increased from 5 to 78 million (NBS, 2011). By 2007, China's formaldehyde production had reached a staggering 12,000 kt, about 4000 times what it was five decades earlier. More than 65% of formaldehyde produced is used to synthesize resins used in construction materials which are therefore a source of indoor formaldehyde pollution (Tang et al., 2009).

However, qualitative and quantitative changes in population exposure to HAPs with urbanization in China have not been characterized sufficiently, nor have the health risks of these HAP exposures. The few studies that have assessed health risks posed by HAPs in China have been limited to either indoor or outdoor exposure, or to a small number of carcinogenic HAPs, or to localized cases within small sample sizes. Zhou et al. (2011) monitored personal exposure to volatile organic compounds (VOCs) and estimated associated cancer risks (median, 4.4×10^{-5}, which means 4.4 additional cases per 100,000 people exposed) in Tianjin. However, their results were limited because formaldehyde and acetaldehyde were not considered and data was only

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collected for 12 participants. Other studies have only considered risks caused by exposure to a small suite of VOCs in indoor environments, such as residences (Guo et al., 2004a; Liu et al., 2013; Ohura et al., 2009; Weng et al., 2010), commutes (Feng et al., 2010; Li et al., 2009; Pang and Mu, 2007), public places (Weng et al., 2009), hotels (Feng et al., 2004), and hospitals (Lu et al., 2006), and hence could not give a complete picture of risk. Some studies have considered high exposure scenarios, such as recent renovation, cooking and indoor combustion, but which only evaluate a narrow suite of pollutants (Du et al., 2014; Huang et al., 2011). So far, no broad analyses of the personal exposure and associated cancer risks posed by a wide range of organic HAPs in multiple microenvironments have been done for China.

The paucity of information on HAP risk assessment has hindered risk-based regulatory decision making and regulatory actions aimed at protecting public health in China. In 2003, China's national indoor air quality standard GB/T 18883 was put into effect, which set guideline values for formaldehyde, benzene, toluene, xylenes and total volatile organic compounds (TVOCs) respectively (GB/T, 18883, 2002). However, for probable carcinogens like trichloroethene, tetrachloroethene, and for possible carcinogens like ethylbenzene, carbon tetrachloride and chloroform (IARC, 2011), the concentrations in both indoor and outdoor air is not yet regulated in China, probably due to lack of information on exposure and risk assessment. Therefore, information is required for regulatory policy.

In this study, we first summarize recent HAPs measurements in urban China, focusing on those HAPs with frequent occurrence in different microenvironments, and then derive a personal exposure model for working adults. Then, cancer risks due to inhalation are calculated using the method proposed by the California Office of Environmental Health and Hazard Assessment (OEHHA), with risks apportioned between indoor and outdoor sources. Because a great deal of uncertainty is often incorporated with risk assessment due to the variations in HAP concentrations, inhalation rates among individuals and toxicity of the HAPs, uncertainty in the risk estimates is also assessed by using a probabilistic risk assessment approach.

2. Methods

2.1. Urban China

In the present study, we focused on HAP exposure modeling and risk assessment in three urban areas of China, the Pearl River Delta (PRD) region, the Yangtze River Delta (YRD) region and the Bohai Rim (BR) region. All three regions have high population density and a developed economy: in 2007, the three regions accounted for more than 40% of China's entire gross domestic product (GDP). While nearly 20% of China's total population lived in these areas, they account for only about 5% of China's land area (Ness et al., 2010). Due to industrial and vehicle emissions in cities, outdoor air in these regions tends to be more polluted than in rural and suburban areas of China (Chan and Yao, 2008; Zhang et al., 2013b). The megacities Beijing, Hong Kong, Shanghai, Guangzhou, Hangzhou, Nanjing, Tianjin, and Dalian in these regions were selected as target cities for the present study.

2.2. Literature review and data collection

We searched the literature for peer-reviewed studies reporting measurements of HAPs in various microenvironments using the ISI Web of Science database for articles published prior to August 2013. The HAP measurements selected, preferably from the last 10 years (2003–2013), but at least within the last 15 years so as to reflect recent HAP emission resources and characteristics in urban China, presented useful data on HAP concentrations in one or more microenvironments.

Research studies reporting HAP measurements in different microenvironments were considered regardless of measurement duration and HAP source. Thus, studies included both short- and long-term measurements. Also, the HAPs considered included those mainly emitted from either outdoor or indoor sources, with some having both outdoor and indoor sources. Microenvironments used to determine air pollutant exposures in the present study included indoor residences (home), indoor work environment (office), commute and outdoor/other, which were defined as the representative microenvironments (Dodson et al., 2007; Weisel, 2002). The miscellaneous "other" microenvironments were assigned the same concentration distributions as the outdoors. Table 1 shows the selected references with information on study location, sampling method, sampling duration, sample size, microenvironment and the types of pollutants.

Sixteen HAPs with high abundance in different microenvironments were considered, covering the most high-risk pollutants as listed by Payne-Sturges et al. (2004) and Sax et al. (2006). These were formaldehyde, acetaldehyde, 1,3-butadiene, 1,4-dichlorobenzene, benzene, carbon tetrachloride, chloroform, ethylbenzene, styrene, tetrachloroethene, trichloroethene, toluene, m,p-xylene, o-xylene and 1,2,4-trimethylbenzene.

As raw data, namely the concentrations of individual compounds in individual samples collected during each study were not available for most studies; the reported statistics were used for analyses. To properly weight statistics from individual studies of a given city, we first compared each study's parameters, including percentiles, arithmetic mean, standard deviation, median, min, and max. We weighted equally the studies for each city/geographic region as per Loh et al. (2007). If there was more than one study for a city, the studies were weighted according to the number of unique measurements. Usually, the weighting was by the number of sampling sites; less frequently, the weighting was by the number of samples.

According to *Time use patterns in China* (NBS, 2010), the automobile is the most common mode of commuting for Chinese urban working adults. On average, working adults spend about 60% of their commuting time in an automobile. Therefore, when compiling HAP concentrations in commute microenvironments, we mainly focused on automobiles. Outdoor HAP concentrations near roadways and HAP concentrations in subways were treated as surrogates only when there was no available data. Additionally, due to lack of sufficient indoor acetaldehyde concentrations for offices, residential acetaldehyde concentrations were used as surrogates. Table 2 presents the summary statistics for the 16 HAPs in home, office, commute and outdoor/other. Table S1 (Supplementary material) shows the means and SDs for each outdoor measurement.

2.3. Exposure model and risk characterization

Fig. 1 presents the schematic diagram of risk characterization for hazardous air pollution. The current standard approach for health risk assessment was originally proposed by the U.S. National Academy of Sciences (National Academy of Sciences, 1983). The four steps of risk assessment are hazard identification, dose–response assessment, exposure assessment, and risk characterization. To estimate risk, we followed the guidance proposed by OEHHA (2003).

Risk information for the target HAPs, including cancer classification, cancer potency factor, cancer benchmark concentration and inhalation reference concentration, is presented in Table 3. Based on the weight of evidence presented by the International Agency for Research on Cancer (IARC) and Integrated Risk Information System (IRIS), all target HAPs are known to be human carcinogens, or probable and possible human carcinogens, except for toluene 1,2,4-TMB and the three xylenes. Cancer potency factors are defined as the upper-bound probability of developing cancer, assuming continuous lifetime exposure to a substance at a dose of one milligram per kilogram of body weight per day. Cancer benchmark concentrations ($\mu g/m^3$) are the exposure levels for cancer risk of one-in-one-million over a lifetime of exposure. A cancer risk of 1.0×10^{-6} is considered negligible risk (Caldwell et al., 1998; Woodruff et al., 1998). For non-cancer hazard assessment, inhalation chronic reference exposure limits (*RELs*) are used as an indicator of potential non-cancer health impacts. *REL* is defined as the concentration at

Table 1
Recent publications reporting HAP measurements in urban China.

Study	City/region	Sampling method	Sampling duration	Sample size	Home	Office	Commute	Outdoor/other	VOCs	Carbonyls
Pearl River Delta (PRD) region										
Chao and Chan (2001)	Hong Kong	Canister sampling	4 h	10		√			√	
Ho et al. (2002)		Canister/active sampling	24 h	62/41				√	√	√
Lee et al. (2002a)		Canister sampling	1–8 h	6–23	√	√		√	√	√
Lee et al. (2002b)		Canister sampling	8 h	6	√			√	√	
Guo et al. (2003)		Canister sampling	1–8 h	4–6	√	√		√	√	
Guo et al. (2004b)		Canister sampling	24 h	120				√	√	
		Passive sampling	24 h	100	√				√	√
Guo et al. (2009)	Mainland of China	Passive sampling	24 h	94	√				√	√
Chan et al. (2003)	Guangzhou	Active sampling	30–60 min	12–20			√		√	
Feng et al. (2004)		Active sampling	3.5 h	25				√		√
Zhao et al. (2004)		Active sampling	30 min	48			√		√	
Feng et al. (2005)		Active sampling	2–3 h	7				√		√
Tang et al. (2005)		Active sampling	30 min	57					√	
Yuan et al. (2012)		Canister/active sampling	3/1 h	10/23				√	√	√
Zhang et al. (2013a)		Canister sampling	60 min	67				√	√	
Bohai Rim (BR) region										
Pang and Mu (2006)	Beijing	Active sampling	1 h	388				√		√
Song et al. (2007)		Active sampling (direct reading instrument)	30 min	1257				√	√	
Pang and Mu (2007)		Active sampling	1 h	29			√			√
Liu et al. (2013)		Active sampling	60/30 min	210/7	√			√	√	√
Zhou et al. (2011)	Tianjin	Passive sampling	5 days	10/6/6/8	√	√	√	√	√	
Guo et al. (2013)	Dalian	Passive sampling	24 h	59	√				√	√
Yangtze River Delta (YRD) region										
Feng et al. (2010)	Shanghai	Active sampling	3 h	12				√		√
Zhang et al. (2012)		Canister sampling	60 min	45			√	√	√	
Li et al. (2009)	Hangzhou	Active sampling	20 min	48			√		√	
Ohura et al. (2009)		Active sampling	30 min	21	√			√	√	
Weng et al. (2009)		Active sampling	3–6 h	36				√		√
Weng et al. (2010)		Active sampling	10 h	–	√					√
Wang and Zhao (2008)	Nanjing	Active sampling	12 h	430				√	√	

or below which no adverse non-cancer health effects are anticipated following long-term exposure (OEHHA, 2003).

Human exposure to HAPs occurs in different microenvironments such that HAP concentrations vary temporally and spatially. In this paper, to predict personal exposure, a time-weighted exposure model was used, which integrates the time fraction spent in each microenvironment and the concentration of each microenvironment visited (Dodson et al., 2007), as shown in Eq. (1):

$$E_i = \sum_j C_{ij} t_j \tag{1}$$

where E_i is the time-weighted personal exposure to pollutant i (mg/m^3); C_{ij} the concentration of pollutant i in microenvironment j (mg/m^3); t_j the time fraction spent in microenvironment j. The modeled exposure is a representative of the true exposure provided that all microenvironments significantly contributing to the total exposure are included and the concentration assigned to the microenvironment is appropriate for the time spent there.

The chronic daily intake (*CDI*, mg/kg/day) attributable to inhalation was calculated as

$$CDI_i = \frac{E_i \times IR \times EF \times ED}{BM \times AL \times NY} \times 90\% \tag{2}$$

where CDI_i is the daily inhalation intake of pollutant i (mg/kg/day); E_i is the modeled personal exposure of pollutant i (mg/m^3); *IR* the inhalation rate (m^3/day); *EF* the exposure frequency (day/year); *ED* the exposure duration (years), given as the working lifetime for adults; *BM* body mass (kg); *AL* the average lifetime (70 years); *NY* the number of days per year (365 days/year); and 90% the absorption factor of VOCs for humans (Guo et al., 2004a; Lerner et al., 2012; Li et al., 2009).

Lifetime cancer risk (*LCR*) is the increased probability of developing cancer as a result of a specific exposure and was calculated by multiplying the intake of a toxic substance by the cancer potency factor, as follows:

$$LCR_i = CDI_i \times CPF_i \tag{3}$$

where LCR_i is the cancer risk associated with compound i; CDI_i the daily intake of compound i; and CPF_i represents the inhalation cancer potency factor of compound i. The cumulative cancer risks were determined by adding all the known, possible, and probable carcinogens (Caldwell et al., 1998; Payne-Sturges et al., 2004).

Non-cancer chronic inhalation health impacts were assessed by a direct comparison of the average personal exposure (E) with a substance-specific *REL*. The hazard quotient (HQ) of each compound was calculated:

$$HQ_i = \frac{E_i}{REL_i} \tag{4}$$

where HQ_i is the hazard quotient for compound i; E_i is the modeled personal exposure to compound i; and REL_i is the reference exposure limits for compound i. An HQ value of one or less indicates that adverse health effects are not expected to result from exposure to this HAP. As the HQ increases above one, the probability of human health effects increases by an undefined amount, such that a particular HAP could be of public health concern. Cumulative non-cancer risks from multiple substances were assessed by summing the HQs of the VOCs that affected the same target organ system, using hazard index (HI) as an indicator. An HI greater than one suggests that there is the potential for adverse chronic health impacts at this receptor location (OEHHA, 2003).

In this study, the modeled population was working adults (male and female) in urban China. Distribution of the time spent in each microenvironment for working adults were extracted from the *Time use patterns*

Table 2
Summary statistics for HAP concentrations in different microenvironments (μg/m^3).

Air pollutants	Arithmetic mean (standard deviation)			
	Home	Office	Commute	Outdoor/other
Carbonyls & very volatile organic compound (VVOC)				
Formaldehyde	54.0 (17.2)	16.1 (2.0)	25.2 (8.4)	12.5 (2.5)
Acetaldehyde	13.6 (10.8)	13.6 (10.8)	18.4 (6.1)	7.1 (1.8)
1,3-Butadiene	0.5 (0.3)	0.3 (0.1)	0.6 (0.3)	0.4 (0.5)
Aromatic hydrocarbons				
Benzene	5.8 (5.7)	3.7 (5.8)	11.1 (5.3)	5.3 (2.0)
Toluene	16.6 (33.0)	32.9 (39.7)	33.7 (16.6)	17.2 (10.9)
m,p-Xylene	3.1 (1.9)	7.1 (13.6)	8.6 (3.8)	6.0 (3.7)
o-Xylene	3.1 (2.5)	5.5 (6.9)	8.5 (5.0)	4.5 (1.7)
Ethylbenzene	3.1 (1.1)	4.2 (6.1)	8.6 (3.4)	4.4 (2.7)
Styrene	5.0 (1.1)	2.6 (7.1)	0.8 (0.7)	0.6 (1.1)
1,2,4-Trimethylbenzene	5.8 (0.2)	2.2 (2.4)	8.6 (7.8)	4.2 (6.4)
Chlorinated hydrocarbons				
Chloroform	1.4 (0.6)	0.5 (0.8)	0.6 (0.3)	0.3 (0.4)
Carbon tetrachloride	0.4 (0.1)	0.1 (0.1)	1.7 (5.4)	0.5 (0.2)
Trichloroethene	1.8 (0.2)	5.6 (9.6)	3.6 (7)	1.6 (0.9)
Tetrachloroethene	2.5 (0.2)	5.2 (9.2)	4.5 (6.9)	0.8 (0.3)
1,4-Dichlorobenzene	20.6 (3.7)	10.2 (17.8)	3.9 (1.6)	3.1 (1.7)

in China (NBS, 2010), which were provided as week-weighted values (Table 4). Exposure parameters derived from existing statistical reports, including activity pattern, inhalation rate, body weight and exposure duration, are also presented in Table 4.

2.4. Uncertainty and sensitivity analysis

High uncertainty accompanies risk assessment especially when only single point values are used to estimate the risk for a population (Sielken and Valdez-Flores, 1999). A stochastic risk assessment approach that can deal with the uncertainty and variability in each parameter was used in this study (Sielken and Valdez-Flores, 1999). Monte Carlo simulation and sensitivity analysis were implemented using Oracle Crystal Ball software. Monte Carlo simulation was used to characterize ranges of exposures and risks. Sensitivity analysis ranks input assumptions according to their importance to the outcome. The stochastic risk modeling provides a clearer picture of population risk estimates than risk assessment using single point data.

Probability distributions reflecting the variations in dose–response relationships, exposure concentrations and exposure factors among individuals and over time within individuals were derived using existing statistical reports (studies). According to OEHHA (2003), the cancer potency factors (CPFs) for HAPs are based on linear extrapolation from animal data or occupational studies, and therefore have considerable uncertainty. Triangle distribution was selected to characterize the CPF parameters (Zhou et al., 2011). The CPF values recommended by OEHHA (2003) representing upper-bound excess lifetime cancer risk potencies were assumed to represent the maximum and the most likely values, and 0 was assigned to the minimum. As lognormal curves best fit the distributions of naturally occurring pollutants (Ott, 1990), a lognormal distribution was used to describe the cumulative frequency distributions of HAP concentrations. To consider the potential effect of inhalation rate on the risk estimates, inhalation rate distributions were also assumed to fit a lognormal distribution (Liao and Chiang, 2006; Lonati and Zanoni, 2012). Standard deviations were taken as 20% of the means.

3. Results

3.1. Major VOCs and carbonyls in urban China

Table 2 lists the sixteen high prevalence HAPs in each microenvironment along with their central tendency data derived from available studies. Of the carbonyls listed, formaldehyde and acetaldehyde, both of which are carcinogens, are most frequently detected in each microenvironment. Their emission sources are well-documented for both indoors and outdoors. In particular, formaldehyde is produced by wood-based materials (Bohm et al., 2012), environmental tobacco smoking (Nazaroff and Singer, 2004) and ozone-initiated secondary chemistry (Nazaroff and Weschler, 2004). It is noteworthy that most of the total formaldehyde in wood-based materials becomes indoor formaldehyde pollution (Tang et al., 2009). This suggests an explanation for why residences and offices have higher formaldehyde concentration than other microenvironments. In urban China, indoor home formaldehyde concentration varies by more than a factor of 100, from less than 1 μg/m^3 to more than 100 μg/m^3, but its typical weighted value is about 50 μg/m^3. Outdoor formaldehyde concentration, on the other hand, is typically in the range of 5–20 μg/m^3. The mean concentrations of acetaldehyde are 13.6 and 7.1 μg/m^3 in residential indoor and outdoor environments. While acetone and acrolein also have adverse effects on human health, there are few measurements in urban China. A recent study has reported residential indoor and outdoor average concentrations of 10.7 and 8.6 μg/m^3 for acetone, and 2.8 and 3.8 μg/m^3 for acrolein in Beijing (Liu et al., 2013).

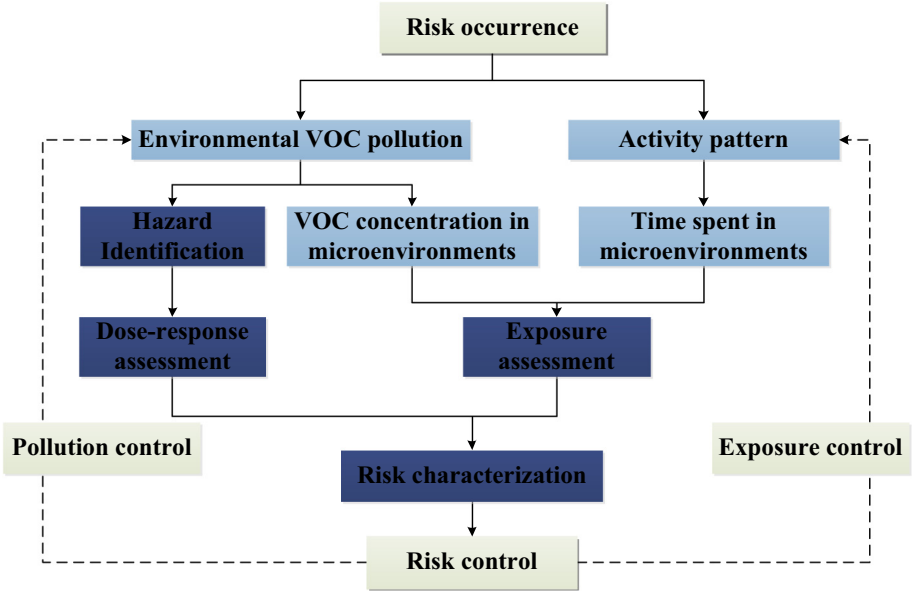

Fig. 1. Characterization of risk occurrence for hazardous air pollutants.

Table 3
Target HAPs and associated toxicity values.
Source: [a]Guo et al. (2004a), [b]U.S. EPA's Cumulative Exposure Project (Woodruff et al., 1998).

Compounds	Weight of evidence		Cancer potency factor $(mg/kg/day)^{-1}$	Cancer benchmark concentrations ($\mu g/m^3$)	Inhalation chronic *RELs* ($\mu g/m^3$)	Target organs
	IARC	IRIS				
Carbonyls & VVOC						
Formaldehyde	2A	B1	2.1×10^{-2}	0.17	9	Eye, respiratory
Acetaldehyde	2B	B2	1.0×10^{-2}	0.37	140	Respiratory
1,3-Butadiene	2A	B2	6.0×10^{-1}	0.01	20	Reproductive
Aromatic hydrocarbons						
Benzene	1	A	1.0×10^{-1}	0.03	60	Developmental, hematologic, nervous
Toluene	–	–	–	–	300	Developmental, nervous, respiratory
Xylenes	–	–	–	–	700	Nervous, respiratory
Ethyl benzene	2B	NC	8.7×10^{-3}	0.40	2000	Alimentary, developmental, endocrine, kidney
Styrene	2B	B2, C	5.7×10^{-4} [a]	2.00[b]	900	Nervous
1,2,4-TMB	–	–	–	–	–	–
Chlorinated hydrocarbons						
Chloroform	2B	B2	1.9×10^{-2}	0.19	300	Alimentary, developmental, kidney
Carbon tetrachloride	2B	B2	1.5×10^{-1}	0.02	40	Alimentary, developmental, nervous
Trichloroethene	2A	B2, C	7.0×10^{-3}	0.50	600	Eye, nervous
Tetrachloroethene	2A	NA	2.1×10^{-2}	0.17	35	Alimentary, kidney
1,4-Dichlorobenzene	2B	NA	4.0×10^{-2}	0.09	800	Alimentary, kidney, nervous, respiratory

Abbreviations: IARC, International Agency for Research on Cancer; group 1, carcinogenic to humans; group 2A, probably carcinogenic to humans; group 2B, possibly carcinogenic to humans. IRIS, Integrated Risk Information System; group A, human carcinogen; group B1, probable human carcinogen; group B2, probable human carcinogen; group C, possible human carcinogen; and group NA, not assessed under the IRIS program.

1,3-Butadiene is mainly emitted outdoors by vehicles (Guo et al., 2004b) and is transported indoors. Chronic inhalation of 1,3-butadiene may affect the reproductive system (OEHHA, 2003). Table 2 shows that the weighted mean concentration of 1,3-butadiene (0.6 $\mu g/m^3$) is the highest in the commute microenvironment.

BTEX (benzene, toluene, ethylbenzene, xylenes) are abundant and detectable in all the environments. The sources of BTEX vary widely. Household products, smoking, solvents and detergents are the main indoor contributors of BTEX while vehicle exhausts and industrial emissions are the main outdoor sources. Commuting microenvironments have the highest BTEX levels. Of BTEX, toluene is the most abundant compound in all microenvironments in urban China. Benzene is classified by the IARC and IRIS as a human carcinogen. The weighted concentrations of benzene in each microenvironment were 5.8, 3.7, 11.1 and 5.3 $\mu g/m^3$ for home, office, commute and outdoors respectively. Notably, significantly higher BTEX concentrations may be present in homes with recent renovations (Du et al., 2014; Liu et al., 2013).

Styrene emitted from household products (styrene butadiene rubber and styrene-butadiene latex), cigarette smoking and industrial sources, is ubiquitous. The IARC has designated styrene as possibly carcinogenic to humans (group 2B). Guo et al. (2009) found styrene levels of 10.9 and 3.9 $\mu g/m^3$ in homes for Hong Kong and the mainland of China, respectively; whereas outdoor styrene concentrations are typically below 1 $\mu g/m^3$. Trimethylbenzenes (TMB) are a mixture of three isomers: 1,2,3-, 1,2,4- and 1,3,5-TMB. TMBs are produced during petroleum refining, and 1,2,4-TMB makes up approximately 40% of the C9 fraction of petroleum which is used as a component of gasoline. Because of this, vehicle emissions are a major source of TMBs. Commuters are thus exposed to the highest concentrations of 1,2,4-TMB.

Chlorinated hydrocarbons are another group of organics frequently detected in each microenvironment with widely scattered point sources. Chloroform, carbon tetrachloride, trichloroethene, tetrachloroethene and 1,4-dichlorobenzene are representative organics and long-term exposure to them is a concern in terms of cancer risks. Concentrations of these compounds typically vary from less than 1 to 10 $\mu g/m^3$ in different microenvironments. In indoors, use of moth repellants and/or deodorizers is associated with higher levels of 1,4-dichlorobenzene (Lee et al., 2002b; Tang et al., 2005).

Weschler (2009) has reported increased concentrations of terpenoids (limonene and α-pinene) in indoor environments due to increased use of terpenoid solvents and scents. This deserves attention because long-term exposure to terpenoids could provoke irritation of the eyes, nose, throat and skin. Also, due to their reactive isoprenic structure, they might be oxidized by ozone or other oxidants to more harmful secondary products (Nazaroff and Weschler, 2004). Recent measurements by Guo et al. (2009) found high indoor home levels of limonene (18.1–49.3 $\mu g/m^3$) and α-pinene (11–19.2 $\mu g/m^3$) in Hong Kong and the mainland of China. However, more insights into terpenoid concentrations and associated sources in different microenvironments in urban China require investigation.

Fig. 2 shows the concentration distributions of HAPs in different microenvironments across urban China. It indicates that homes have the largest total HAP concentrations and that HAP concentrations in indoor environments (typically >100 $\mu g/m^3$) are significantly higher than in

Table 4
Population factors related to inhalation exposure for Chinese urban working adults.

Parameters		Male	Female	Source
Time distribution in different microenvironments (min/day)	Home	932	1018	*Time use patterns in China*
	Office/working place	290	217	NBS (2010)
	Commute	88	81	
	Outdoor/other	130	124	
Body weight (kg)		62.7	54.4	Wang et al. (2009)
Inhalation rate (m^3/day)		19.0	14.2	Wang et al. (2009)
Exposure duration (year)[a]		42	37	China State Council (1978)

Note: lognormal distribution (mean; SD) for inhalation rate, SD was taken as 20% of the mean.
[a] Estimated as the duration from 18-year-old to the legal retirement age (male: 60-year-old, female: 55-year-old).

outdoor environments. Overall population inhalation exposure to carbonyls and 1,3-butadiene mainly occurs in homes while commuters are exposed to higher concentration of aromatic hydrocarbons as these are associated with vehicle emission.

3.2. Modeled personal exposures

Table 5 shows summary statistics for the modeled personal exposures to each HAP. Mean personal exposure to individual compounds for urban working adults in China ranges from <0.5 μg/m^3 for 1,3-butadiene and carbon tetrachloride, to >40 μg/m^3 for formaldehyde. On average, formaldehyde, toluene, 1,4-dichlorobenzene and acetaldehyde contribute most to the total personal HAP exposures (32.5–34.0%, 15.6–16.6%, 12.7–13.1% and 10.4–10.5% respectively). Mean personal HAPs concentrations are larger than the median personal HAPs concentrations, because the former include the full distribution of values including extremes, whereas the median lack physical meaning and would underestimate the exposure (Ott, 1994). Thus, the mean has more public health relevance.

For most HAPs, the mean concentrations that people are exposed to are equal to or lower than indoor concentrations, and greater than outdoor concentrations. For example, the mean concentrations of formaldehyde that females and males are exposed to are 43.0 and 40.8 μg/m^3, compared with mean indoor and outdoor formaldehyde concentrations of 54.0 and 12.5 μg/m^3, respectively. However, toluene, trichloroethene and tetrachloroethene are exceptions; mean personal concentration exposures were greater than mean residential and outdoor concentrations, indicating the strong presence of their sources in office and/or vehicle.

Importantly, the modeled HAP personal exposures are higher than cancer benchmark concentrations by one or two orders of magnitude. This indicates cancer health impacts may be anticipated following long-term exposure to these HAPs in urban China.

3.3. Risk estimates

Fig. 3 presents the estimated lifetime excess cancer risks based on inhalation daily doses and inhalation cancer potency factors. The box plots indicate the 5th, 25th, 50th, 75th, and 95th percentiles of cancer risks for individual compounds and the total cumulative risk. As shown in Fig. 3, formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene rank in the first category, that is, with high median cancer risk estimates, greater than 1×10^{-5}. Acetaldehyde, carbon tetrachloride, tetrachloroethene, ethylbenzene, chloroform and trichloroethene rank in the second category, with medium cancer risk estimates between 1×10^{-6} and 1×10^{-5}. Styrene is in the lowest category, with median cancer risk estimates less than 1×10^{-6}. Cancer risks estimated at the 95th percentile for formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene are $1.44/1.79 \times 10^{-4}$, $1.01/1.30 \times 10^{-4}$, $1.18/1.52 \times 10^{-4}$, $5.12/6.65 \times 10^{-5}$ for working females and males, respectively. All are in the ranges for which the U.S. EPA suggests taking action to reduce exposures.

For all HAPs, the cancer risks for working males are slightly higher than those for working females, but the differences do not alter the ranking of the pollutants. Fig. 4 compares the mean risk from each HAP. Formaldehyde accounts for the greatest portion of the total risk, contributing 33%. 1,4-Dichlorobenzene, benzene and 1,3-butadiene are other significant contributors to the total risk, accounting for 55%. The mean cumulative cancer risks obtained by summing the risk of each HAP are 227 per one million for the urban working female population of China (median, 216), compared with 293 per one million for the urban working male population of China (median, 278). The cumulative cancer risks span from 1.27×10^{-4} to 3.65×10^{-4} for females and from 1.65×10^{-4} to 4.71×10^{-4} for males. However, when variations in the inhalation rate and cancer potency factor are not accounted for, the median and mean cumulative cancer risks increase to 3.29×10^{-4} and 3.39×10^{-4} for working females, and 4.25×10^{-4} and 4.38×10^{-4} for working males. This indicates that risks are overestimated when variation is not accounted for. Fig. 5 shows the cumulative frequency distributions of risks with and without considering variations. The overestimation of risk is clearly evident when variation is not included.

For non-cancer health impacts, all HAPs have a mean HQ of less than one except for formaldehyde, indicating that only the formaldehyde concentration exceeded the *REL*. Thus, an HI of one was exceeded for the eyes and the respiratory system, which suggests potential adverse chronic health impacts at these receptor locations. However, the HIs for other target organs were all <1, with average HIs ranging from 2×10^{-3} for endocrine system to about 0.2 for nervous system.

3.4. Risk apportionment

We compared mean risks as a function of total HAP exposure, regardless of whether the sources are indoor or outdoor (Fig. 6). On average, 73% and 67% of the total risks come from exposures inside the home, 12% and 18% from the office, 8% and 9% from commuting time, and 6% and 6% from outdoor, for working females and males respectively. For formaldehyde, 1,4-dichlorobenzene, chloroform and styrene, indoor home exposures are above 80% of total exposure for working adults.

Following the methodology of Loh et al. (2007), we calculated the indoor and outdoor contribution of each pollutant to indoor risk by using the indoor to outdoor concentration ratio as a source perspective. As shown in Table 6, about 60% of the risk comes from indoor sources (nearly 50% from the homes), and 34% from outdoor sources, on average. It should be noted that for benzene, ethylbenzene and carbon tetrachloride, exposures from indoor environments contribute more to risk than outdoor exposures; however, from a source perspective, outdoor sources of these organics contribute more to risk than indoor sources.

4. Discussion

Reviewing urban population inhalation exposure to HAPs and the associated risk is a demanding task in China, especially given that China has experienced huge changes in indoor and outdoor environments affecting hundreds of millions of people, and that few studies have reported population-based exposure modeling and risk assessment for Chinese cities. This study is a first effort to estimate the cancer and non-cancer risks from a wide range of air pollutants for the large population of working adults in urban China. In addition, by summarizing HAP measurements in urban China, this study reports their recent emissions and characteristics on an across-region scale.

In China, the decade since 2003 has witnessed increasing concern about the indoor air pollution. Most significantly, the national indoor air quality standard GB/T 18883 was enforced to regulate the emissions of a number of indoor air pollutants. The guideline values set for the limited regulated VOCs are 100, 110, 200, 200, and 600 μg/m^3 for formaldehyde, benzene, toluene, xylenes and TVOC respectively (GB/T, 18883, 2002). However, the limited available studies indicate that HAP concentrations indoors and outdoors in urban China remain at relatively constant and plateau levels (Guo et al., 2009; Lee et al., 2002a; Liu et al., 2013; Weng et al., 2009; Weng et al., 2010; Zhang et al., 1994), even though many interventions have been put into action during the past decade. This may be due to the fact that during the period, China is experiencing fast and rapid urbanization with the largest rural to urban migration in history and that accompanying this trend is an increasing use of furniture, building materials and man-made consumer products indoors, as well as increasing vehicle emissions and industrial combustion (Zhang et al., 2013b).

Average total lifetime cancer risks attributed to HAPs are 2.27×10^{-4} and 2.93×10^{-4} for urban working females and males in China. The risks exceed (1) the U.S. EPA guideline value of 10^{-6} (Caldwell et al., 1998; Woodruff et al., 1998); (2) the upper-bound excess lifetime cancer risk recommended by the WHO for carcinogens in drinking water of 10^{-5}

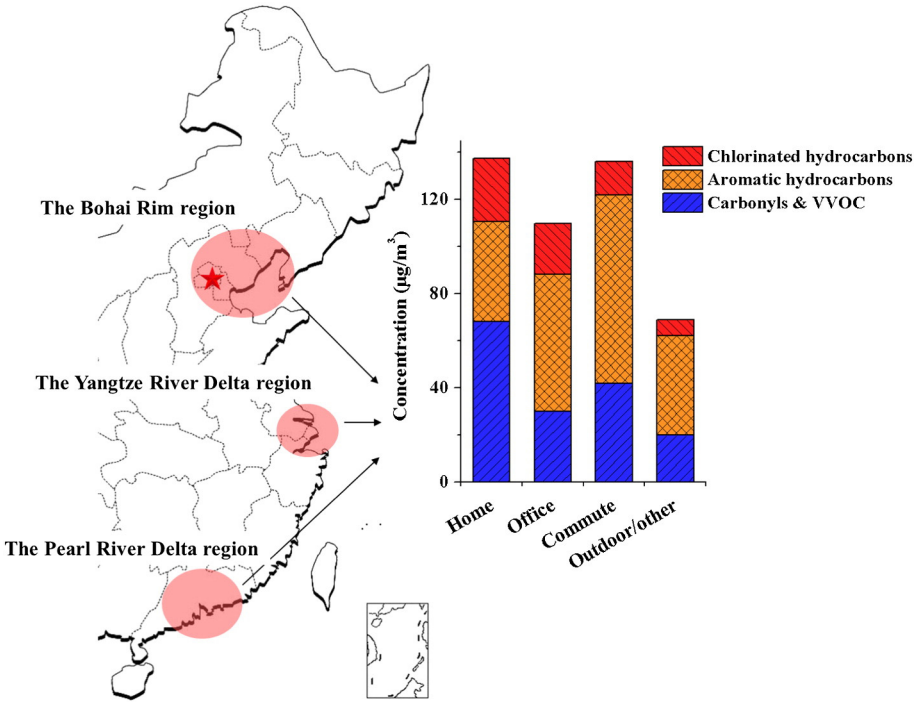

Fig. 2. Distributions of HAP concentrations across urban China (only part of China is shown and the three light red circle shadows indicate the regions considered in this study). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(WHO, 2008); and (3) the level (10^{-4}) at which the U.S. EPA would consider some action or intervention to reduce exposures (Payne-Sturges et al., 2004). According to the WHO Air Quality Guidelines for Europe, each country must determine its own acceptable risk levels (WHO, 2000). Indeed, China must also quantify its own acceptable risk levels based on scientific data, expected public health, environmental and socio-economic impacts, political factors and perceived benefits (Sarigiannis et al., 2011). The cancer risks induced by these HAP exposures for the urban working population in China are not negligible and should be taken into account for future health risk management.

Our study provides an overall picture of the cancer risks attributable to population inhalation exposure to HAPs and comparisons with other studies. However, there are still some limitations and uncertainties inherent in exposure and risk assessment in the present study.

4.1. Comparison with other studies

Exposures and risks estimated in the present study were compared with those of previous studies conducted in China and abroad. The major VOC contributors to risk are formaldehyde (54.0 ± 17.2 μg/m^3), 1,4-dichlorobenzene (20.6 ± 3.7 μg/m^3), benzene (5.8 ± 5.7 μg/m^3) and acetaldehyde (13.6 ± 10.8 μg/m^3) in urban Chinese homes. Most of the HAP concentrations in urban China as shown in Table 2 are greater by a factor of 1–9 than those calculated by Loh et al. (2007) for corresponding microenvironments in the United States. Even when compared to measured concentrations for large metropolitan cities such as New York City and Los Angeles (Sax et al., 2006), higher levels are found in homes and in the outdoors in urban China for most HAPs. Sarigiannis et al. (2011) compiled measurements of HAPs (BTEX, styrene and

Table 5
Estimated personal exposure levels for females and males (μg/m^3).

Compound	Personal exposure level for female					Personal exposure level for male				
	Mean	SD	Median	5%	95%	Mean	SD	Median	5%	95%
Carbonyls & VVOC										
Formaldehyde	43.0	12.2	41.2	26.4	65.2	40.8	11.2	39.2	25.6	61.0
Acetaldehyde	13.3	7.6	11.3	5.6	27.5	13.3	7.1	11.5	5.9	26.8
1,3-Butadiene	0.5	0.2	0.4	0.2	0.9	0.5	0.2	0.4	0.2	0.8
Aromatic hydrocarbons										
Benzene	5.8	4.1	4.6	2.1	13.3	5.7	3.9	4.7	2.1	12.6
Toluene	19.8	21.3	13.9	5.7	52.1	20.8	20.5	15.2	6.3	52.8
m,p-Xylene	4.3	2.6	3.7	2.0	8.2	4.5	3.2	3.8	2.1	8.9
o-Xylene	3.9	2.0	3.4	1.7	7.7	4.0	2.1	3.5	1.8	8.0
Xylenes	8.1	3.3	7.4	4.6	13.6	8.5	3.9	7.7	4.8	14.7
Ethylbenzene	3.7	1.2	3.4	2.2	5.7	3.8	1.4	3.5	2.2	6.0
Styrene	4.0	1.2	3.9	2.7	5.8	3.9	1.4	3.6	2.5	5.8
1,2,4-TMB	5.3	0.8	5.1	4.4	6.7	5.1	0.9	4.9	4.2	6.7
Chlorinated hydrocarbons										
Chloroform	1.2	0.5	1.1	0.6	2.1	1.1	0.5	1.0	0.6	2.0
Carbon tetrachloride	0.4	0.2	0.4	0.3	0.7	0.4	0.3	0.4	0.3	0.7
Trichloroethene	2.5	1.6	2.1	1.5	4.6	2.7	2.1	2.1	1.4	5.5
Tetrachloroethene	2.9	1.4	2.5	1.9	4.9	3.0	1.8	2.5	1.8	5.7
1,4-Dichlorobenzene	16.7	3.6	16.2	12.1	22.6	16.0	4.1	15.3	11.3	22.7

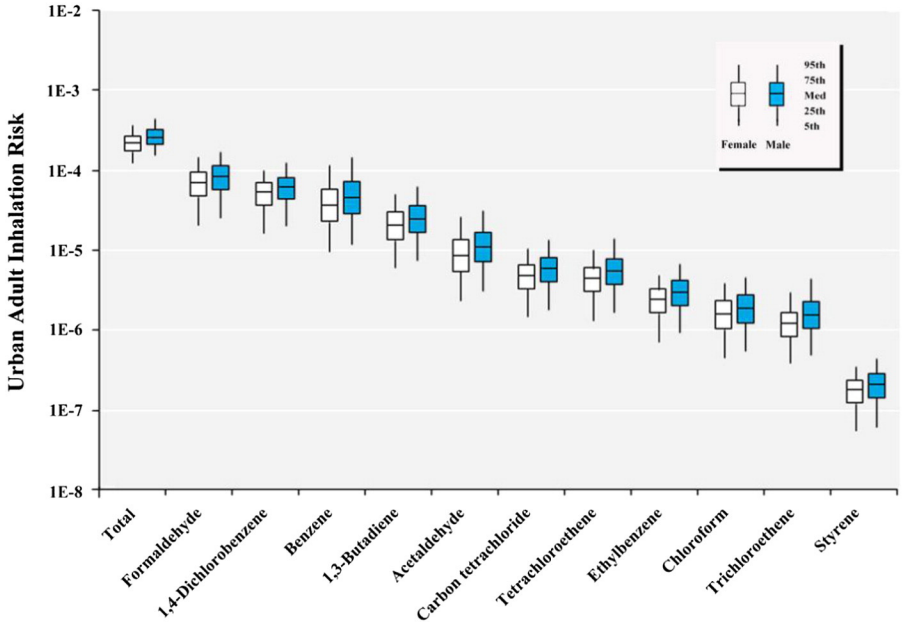

Fig. 3. Ranking of cancer risk estimates from HAPs for urban adults in China. Cancer risks are on a log scale. Bars represent 5th and 95th percentiles. Boxes represent the 25th, 50th, and 75th percentiles.

carbonyls) in European indoor environments, and reported concentrations indoors similar to those we have estimated for urban China. However, Sarigiannis et al. (2011) did not distinguish among different indoor microenvironments such as home, office, public buildings, museum, library and cars, which may explain the wider range of HAP indoor concentrations in Europe.

Few studies have estimated cancer risks in China based on personal exposures. A recent cancer risk assessment conducted in Tianjin, China, might provide a comparison to our results. Zhou et al. (2011) assessed personal inhalation exposure to 5 VOCs in Tianjin, and concluded that benzene and 1,3-butiene were the primary risk drivers among the targeted VOCs. However, their results are likely biased toward a limited suite of HAPs, not including carbonyls, and for only 12 participants. Guo et al. (2004a) considered risks associated with exposure to seven VOCs in different indoor environments in Hong Kong, calculating lower risks than our present 16 HAP evaluation.

Cancer risks from HAPs have been estimated on community-, city- and national-scale by Payne-Sturges et al. (2004), Sax et al. (2006) and Loh et al. (2007) in the United States, respectively. With high 1,4-dichlorobenzene concentration homes taken into account, Loh et al. (2007) and Sax et al. (2006) found that formaldehyde and 1,4-dichlorobenzene were the primary risk contributors, similar to our finding. In terms of the indoor/outdoor source contributions to risk, higher contributions of indoor sources were found in urban China than in the United States (Loh et al., 2007; Sax et al., 2006). Finally, it should be pointed out that these studies assessed lifetime cancer risks as a direct combination of the exposure levels and cancer unit risks with the default assumption of a standard body weight (70 kg) and an average breathing rate (20 m^3/day), which may be overestimates Chinese adults. If we follow the same methodology, *LCR*s of 7.84×10^{-4} and 7.59×10^{-4} for working females and males in urban China would be obtained respectively, which are much greater than the former results (see Section 3.3). In brief, direct comparisons with other studies on HAP risk assessment are difficult because different studies may include different compounds and risk estimates are not always calculated based on the same approach.

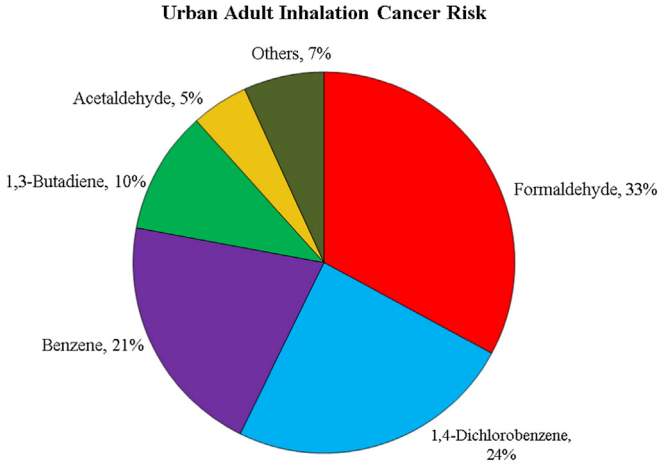

Fig. 4. Mean contribution to cumulative cancer risks from various HAPs.

4.2. Uncertainties of modeled exposures

First, the paucity of measurement data on non-home microenvironments for some HAPs, especially in workplaces, is the major difficulty in deriving exposures. Although we have eliminated the vast non-urban areas for the reason that few HAP measurements have been made in these areas, the problem of limited data still arose when we tried to compile summary statistics for urban China. For example, due to lack of sufficient indoor acetaldehyde concentrations in office, residential acetaldehyde concentrations were used as surrogates. Also, it is difficult to compare the concentrations of some HAPs in offices and commutes, because only single measurements exist. Moreover, there are few HAP measurements with big samples reflecting a wide population (Table 1). Nonetheless, because exposures in homes dominate the total risk for the baseline population, the data scarcity does not increase the uncertainty to an inestimable level. Future exposure assessment could, however, be greatly facilitated by national/regional scale measurements with sufficient size for a large population.

Close inspection of the collected data indicated wide variability for some pollutants. This might be attributable to different source characteristics. For example, 1,4-dichlorobenzene and styrene are exclusively linked to specific consumer products and behaviors (in this case, the use of mothballs, room deodorizers and styrene butadiene rubber products respectively). In contrast, for BTEX, strong outdoor traffic emission

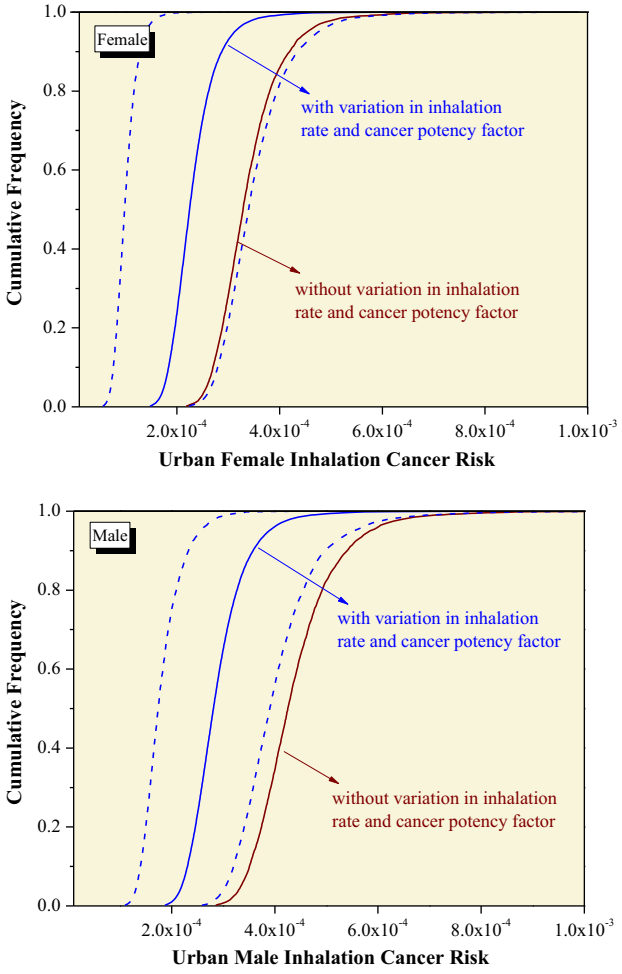

Fig. 5. Cumulative frequency distribution of total risk for working females and males in urban China. The results were derived with (blue curves) and without (brown curve) variation in inhalation rate and cancer potency factors. The medians are presented as solid lines while the 5th and 95th percentiles are presented as dashed curves. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

sources and out-to-indoor air penetration might have a more significant contribution than indoor sources. Moreover, seasonal changes, which influence climatic conditions, may result in different HAP emission strengths in indoor and outdoor environments. Carbonyls including formaldehyde and acetaldehyde presented higher indoor and outdoor levels in summer than winter in Hangzhou and Beijing, partially because of stronger temperature-related emissions and stronger photo-oxidation of VOCs in summer (Pang and Mu, 2006; Weng et al., 2010; Zhang et al., 2007). Thus, we conducted a simple comparison of year-round and short-term average concentrations of formaldehyde and acetaldehyde in Beijing. Pang and Mu measured full year levels of ambient carbonyl compounds in Beijing from November 2004 to October 2005 (Pang and Mu, 2006). The annual average concentrations of formaldehyde and acetaldehyde were 14.8 and 10.2 μg/m^3, respectively. Later, Liu monitored the ambient levels of carbonyls in Beijing during the period of November–December 2009 (Liu et al., 2013). The short-term average concentrations of formaldehyde and acetaldehyde were 16.8 and 11.2 μg/m^3, respectively. While these short-term average concentrations were close to the annual average concentrations in Beijing, we cannot check whether the selected outdoor measurements are representative of the various seasons in each city.

The baseline exposure did not look into the common sources of indoor smoking and cooking, which might lead to underestimation of exposure to some HAPs for specific groups of the population such as a family with a smoking member, housewives and domestic helpers. However, we note that Ohura et al. (2009) investigated possible sources of indoor VOCs in homes of Hangzhou, China, using principal component analysis/absolute principal component scores. Indoor solvents, human activity (cooking, house decoration) and vehicle emissions were identified as the three main possible sources, although many homes had smoking members during the monitoring period.

Accompanying China's rapid urbanization over the past three decades has been an increase in decoration and refurbishment, which affect indoor air quality considerably (Zhang et al., 2013b). Indoor cooking may have non-negligible health risks for housewives and domestic helpers. However, the total quantities of aromatic hydrocarbons and carbonyls emitted from cooking processes may not significantly affect concentrations in whole dwellings (Huang et al., 2011).

Sampling and analytical methods play an important role in the quantification of entities in samples, and therefore affect quantification of personal exposure. As shown in Table 1, passive, active and canister sampling are all used for HAP measurement. Passive sampling is more suited to assessing long-term time-weighted averages for 24 h or even several weeks while active and canister sampling is more suitable for short-term exposure. Also, canister methodology is applicable to 150 polar and nonpolar VOCs found in ambient air from parts-per-million (ppmv) to parts-per-trillion (pptv) by volume (Wang and Austin, 2006). In particular, different sampling methods integrating different sampling durations may be subject to different biases. However, the biases are not expected to add a disproportionate amount of uncertainty in exposure modeling. Many studies have validated good agreement among sampling methods: between passive sampling and active sampling (Ballach et al., 1999; Cocheo et al., 2009), passive sampling and canister sampling (McClenny et al., 2006; Pratt et al., 2005), active sampling and automatic instrument (Wideqvist et al., 2003) for HAP measurement. Additionally, different HAP measurements have been combined to obtain a whole picture of population exposure to HAPs (Loh et al., 2007; Sarigiannis et al., 2011). For example, Loh et al. (2007) merged the short-term (2.5–3.5 h) and long-term (48 h) aldehyde concentrations provided by Zhang et al. (1994) and Sax et al. (2004) to derive the input distribution of aldehyde concentrations in indoor and outdoor residential microenvironments. Sarigiannis et al. (2011) combined the 72 h average aldehyde concentrations in Paris dwellings (Clarisse et al., 2003) and the 30–95 min average aldehyde concentrations in Strasbourg homes (Marchand et al., 2008) to represent indoor home aldehyde pollution levels of France. The U.S. EPA has provided a screening model, AERSCREEN (http://www.epa.gov/scram001/dispersion_screening.htm), which gives estimates of "worst-case" 3 h, 8 h, 24 h and annual concentrations from the maximum one-hour concentration. However, this model is only for screening purposes and requires meteorological input data and emission source input data. Therefore, this model has seldom been used for exposure modeling. Thus, there is a need for a sampling harmonization protocol considering the differences between the sampling techniques and providing guidance on data interpretation.

Another uncertainty arises from basing the release of HAPs from solid sorbents on different desorption techniques. In general, because certain solvent peaks may mask analyte peaks, solvent desorption has higher detection limits than thermal desorption (Ras et al., 2009). Larger numbers of compounds can be detected using thermal desorption than liquid desorption (Ramirez et al., 2012). In addition, Jia et al. (2012) reported four sources of variability for indoor and outdoor VOC measurements: the particular city, the particular residence, season, and measurement uncertainty. Jia et al. (2012) suggested that increased sample size covering multiple seasons and residences helps for better indoor air investigation; and more central monitoring stations could improve outdoor air investigation.

Finally, uncertainties could also arise from the input distributions of exposure factors. The working lifetime for Chinese urban females and

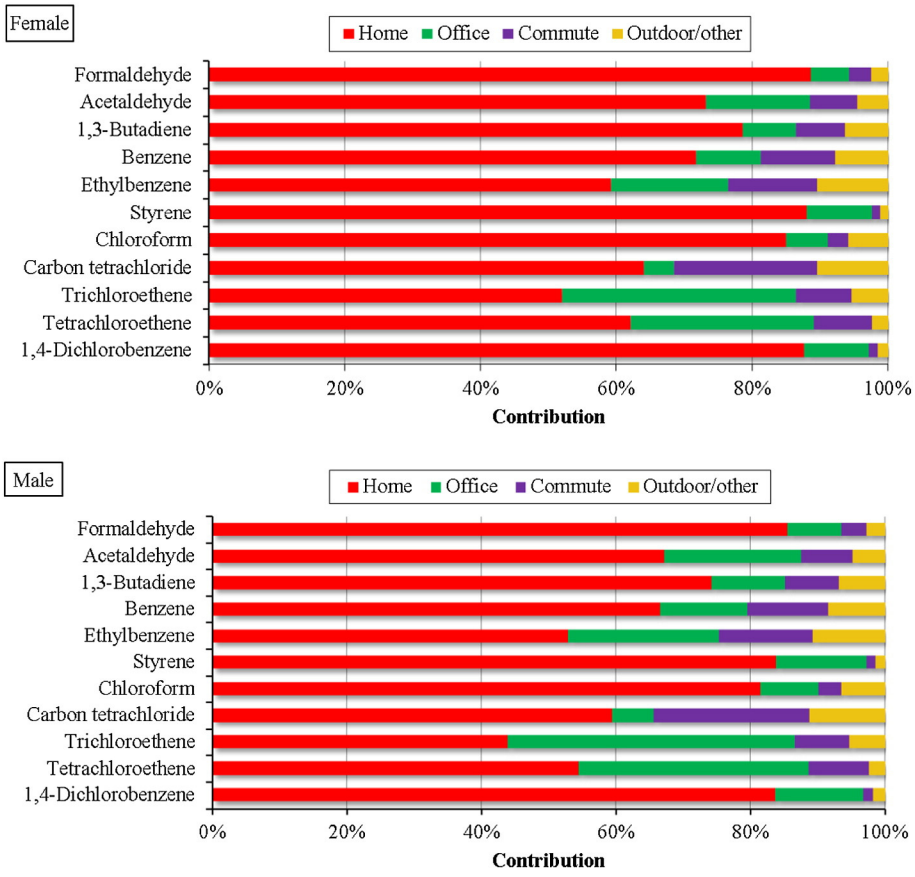

Fig. 6. Mean contributions to cumulative cancer risks from each microenvironment.

males was input as from 18 to 55 years and to 60 years respectively, but there could be considerable variability among individuals. We do not expect great differences between adult exposures and those for childhood and old age, but this assumption may create some bias in the results. Additionally, though mean values of Chinese week-long time use were used to predict the central tendency of risk, cross-individual variations in activity patterns may cause variability in risk estimates (Liu et al., 2007). Input distributions on population body weight and inhalation rate are based on limited investigations and might not yield a true picture of recent exposure parameters of urban Chinese. Future studies could benefit greatly from updated large-scale investigations of population exposure parameters in China.

4.3. Uncertainties of risk estimates

High uncertainty exists in risk assessment especially when only single point values of relevant inputs are used to estimate the risk for a population. Therefore, given the probability distributions of HAP concentrations, inhalation rates and cancer potency factors in Eqs. (2) and (3), the distributions of risks for the selected population groups were calculated using Monte Carlo simulations. As presented in Fig. 3, ranges of cancer risks for females and males are predicted to be from 1.27×10^{-4} (5th percentile) to 3.65×10^{-4} (95th percentile), and from 1.65×10^{-4} (5th percentile) to 4.71×10^{-4} (95th percentile), respectively. Lognormal distribution gives the best fitting model for the frequency distributions

Table 6
Mean contributions (%) to cumulative cancer risks from indoor and outdoor sources.

	Female			Male		
	Indoor	Outdoor	Indoor residential	Indoor	Outdoor	Indoor residential
Carbonyls & VVOC						
Formaldehyde	75	22	72	74	22	69
Acetaldehyde	58	35	48	57	35	44
1,3-Butadiene	51	42	47	49	43	45
Aromatic hydrocarbons						
Benzene	42	48	38	40	48	35
Ethylbenzene	33	54	24	33	53	22
Styrene	86	12	79	86	13	75
Chlorinated hydrocarbons						
Chloroform	56	41	54	55	42	52
Carbon tetrachloride	28	51	27	26	50	25
Trichloroethene	55	37	28	57	34	46
Tetrachloroethene	70	21	47	71	20	41
1,4-Dichlorobenzene	83	15	76	83	16	73
Total	58	34	49	57	34	46

of total cancer risk estimates. It should be noted that without the variations in inhalation rates and cancer potency factors taken into account, the risk ranges increase to 2.58×10^{-4} (5th percentile)–4.54×10^{-4} (95th percentile) and 3.34×10^{-4} (5th percentile)–5.84×10^{-4} (95th percentile) for females and males, respectively. This suggests that the risks are overestimated when variation is not included.

In addition, males have a higher risk level than females. This may be due to working males spending more time in the office and commuting, where elevated HAP concentrations easily occur; working males having greater inhalation rates than females; and males working a longer time in their life time than females.

Fig. 7 presents the contributions of 7 major parameters to the total variance of risk estimates for working females and males. Results of sensitivity analyses show that variations in the inhalation rates, cancer potency factors of formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene, and the indoor home concentrations of formaldehyde and benzene are the major contributors to total variance. The 7 major contributors account for over 90% of the total variance; inhalation rate contributes the most. These estimates of which parameters are influencing the risk estimates the most suggest the best way to refine estimates. However, it should be noted that due to lack of sufficient data, the variations in the other parameters such as body weight and time-use distribution were not considered, which may have led to underestimation of the variability in cancer risk estimates, and which could alter the rankings of the parameters.

In terms of non-carcinogenic effects, risks posed by hazardous air chemicals affecting the eye and respiratory system are of concern because the HI is higher than one. Health impacts at other receptor locations seem to be negligible, since the HIs for multiple chemicals affecting organ systems are less than the value of unity. However, there are still concerns about the non-carcinogenic effects. For example, the exposure level of formaldehyde is significantly higher than the chronic *REL* (9 μg/m^3). An epidemiological study a with a minimum of chance effects (Wolkoff and Nielsen, 2010) suggested that either the non-cancer chronic health assessment of formaldehyde needs to be refined, or measures for risk reduction need to be put into place. Also, potential interactions of multiple chemicals should be considered. Ragas et al. (2011) found that estimates which considered potential interactions always yielded higher risk estimates than when these interactions were ignored. Finally, we note that the uncertainties in the *RELs* and exposure levels of HAPs might also lead to wider ranges of HIs.

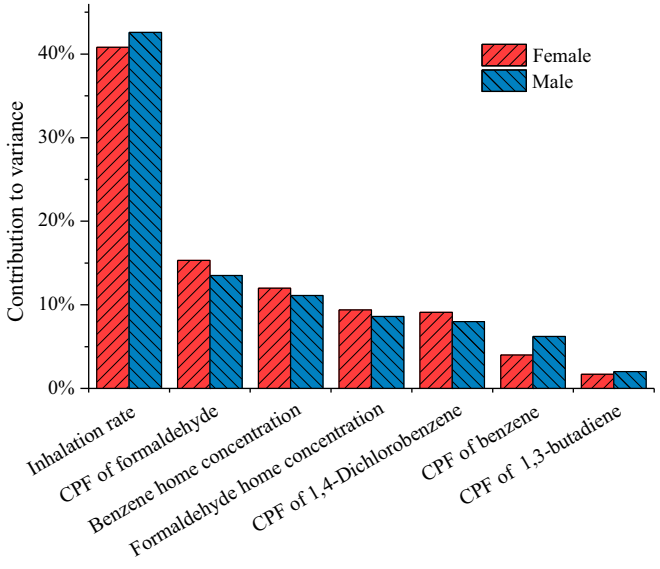

Fig. 7. Contributions of 7 major parameters to the total variance in risk estimates for females and males.

The limited and incomplete available toxicological information contributes to the uncertainties in risk assessment. For cancer risk estimates, cancer potency factors are derived by linear extrapolation from a high-dose animal or human studies to the low-dose environmental exposure using the upper 95% confidence limit on the dose response function (OEHHA, 2003). There are also uncertainties in low-dose exposure scenarios and, moreover, a clear understanding of mechanisms has not yet been achieved. Formaldehyde has a highly non-linear exposure–response relationship for nasal cancer in rats; however, the linear unit risk approach that is based on conservative ("worst-case") considerations is used for risk characterization of formaldehyde exposures (Nielsen and Wolkoff, 2010). In addition, inhalation cancer potency factors recommended for determining cancer risk instead of unit risk factors allows for integration of inhalation rate, body weight and air concentrations to estimate dose in mg/kg/day, however, default assumptions for adults with body weight of 70 kg, inhalation rate of 20 m^3/day, lifetime of 70 years should be carefully considered. Default assumptions might bias our estimates but would not alter the ranking of compounds. Hence, a calculation using realistic ranges of body weights and inhalation rates for different population groups or different races would produce a more refined analysis. For non-cancer risk estimates, HQ calculations are useful for health endpoints, but they do not provide an estimated probability of effects, and chronic reference benchmarks are often derived from No-Observed-Adverse-Effect Levels or Lowest-Observed-Adverse-Effect Levels associated with uncertainty factors (Ramirez et al., 2012).

In addition, interactive effects of exposure to more than one carcinogen or toxicant are not quantified in our study. Herein, cancer risks from all emitted carcinogens are typically added, and hazard quotients for substances impacting the same target organ/system are added to determine the hazard index. However, some authors have suggested that possible mixture-related effects, such as, antagonistic and synergistic effects may occur in complex mixtures (Monosson, 2005; Ragas et al., 2011). For substances that act synergistically, the present study could underestimate the risks. For substances that may have antagonistic effects (that is, lessen the toxic effects produced by another substance), our study could overestimate the risks.

5. Implications for policy and research

Our study provides insight not only into the prevalence of high-risk compounds in urban China but also into the exposures and associated risks for these compounds. Though risk estimates generated should not be interpreted as expected rates of disease due to the uncertainty associated with the process of risk assessment, they represent estimates of potential risks and can be used as a metric to compare one source with another and to prioritize concerns.

Our results will have implications for improving the health status of urban Chinese adults and reducing the uncertainty of risk estimates. First, about 70% of the risks come from exposures occurring indoor homes while nearly 60% of the risks comes from indoor sources. This is mainly because of the larger fraction of time spent indoors (Table 4) and the higher indoor concentrations (Table 2). Thus, environmental policy should focus on approaches which improve indoor air quality. Second, formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene are the major risk contributors. Therefore the greatest health gains can be realized by reducing indoor and outdoor emissions of these HAPs. Third, modeled HAP concentrations are appropriately used as surrogates of monitored personal exposures for risk assessment. However, more measured indoor and outdoor pollutant concentrations are needed to minimize the gaps between modeled and measured data. Finally, the cancer potency factors of HAPs and the inhalation rates of subgroup populations contribute a lot to the uncertainty of risk estimates. Therefore, refining these factors is expected to reduce the uncertainty associated with risk estimates greatly.

In terms of research needs, first, the quality and quantity of available monitored data on indoor and outdoor air pollutants are of particular importance for a probabilistic approach to exposure modeling. Importantly, the paucity of HAP measurements in vast rural and suburban areas in China has hindered national exposure and risk assessment. Therefore, large-scale air measurements in China, covering multiple seasons, microenvironments and cities, are urgently needed. Second, evidence-based air sampling harmonization protocols would increase data reliability, and should include comparisons between different sampling methods and different desorption techniques, sampling duration and target locations for measuring representative exposures. Protocols should also emphasize measurement uncertainties. Third, census reports of exposure factors for Chinese people would improve exposure modeling. Finally, a better mechanistic understanding of the interactive effects of multi-stressors is important. Deeper understanding about hazard identification of HAPs will allow us to refine the predictive assessment tools and to reduce the uncertainty associated with cancer potency factors.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2014.06.014.

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