



## VOC characteristics and inhalation health risks in newly renovated residences in Shanghai, China



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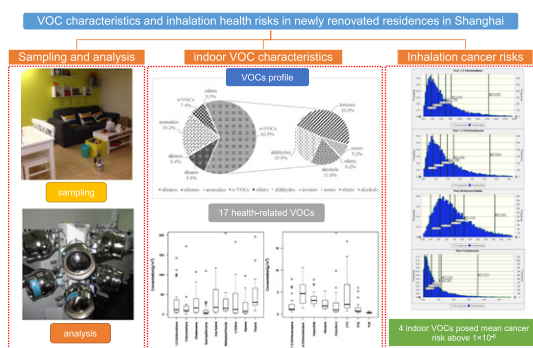
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### HIGHLIGHTS

- Measurements of up to 101 VOCs were made in 8 newly renovated homes in Shanghai, China.
- The dominant VOC groups by mass concentration were oxygenated VOCs, aromatics, alkanes and halogenated VOCs.
- The median individual concentrations of 17 health-related VOCs ranged from 0.35  $\mu\text{g}/\text{m}^3$  to 30.64  $\mu\text{g}/\text{m}^3$ .
- The concentrations of 1,2-dichloroethane, 1,4-dichlorobenzene, methylene chloride, and ethylbenzene presented a mean cancer risk above the acceptable risk level of  $1 \times 10^{-6}$ .

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 4 June 2016

Received in revised form 2 September 2016

Accepted 10 October 2016

Available online 4 November 2016

#### Keywords:

Volatile organic compounds

Renovation

BTEX

Chlorinated hydrocarbons (CL-VOCs)

Health risk assessment

### ABSTRACT

**Background:** Exposure to indoor VOCs is expected to link to a variety of negative health outcome. The popularity of decorations and refurbishment in homes in China has given rise to indoor elevated VOC levels, potentially posing health threats to residents.

**Methods:** In this study, concentrations of 101 VOC compounds and associated health risks were investigated in newly renovated homes in Shanghai. The potential excess inhalation health risks from home exposure of 17 health-related VOCs were estimated by the Inhalation Unit Risk (IUR) and Reference Concentration (RfC) proposed by US EPA. Monte Carlo simulation and sensitivity analysis were used to assess the uncertainty associated with the estimates of health risks.

**Results:** The dominant groups by mass concentration were oxygenated VOCs (o-VOCs), aromatics, alkanes and halogenated VOCs (x-VOCs). 12 VOCs with IARC's confirmed or probable carcinogens ratings were detected with a >60% detection frequency in the total samples. The mean concentrations of BTEX (benzene, toluene, *m/p*-xylene, *o*-xylene, ethylbenzene) were 2.32  $\mu\text{g}/\text{m}^3$ , 200.13  $\mu\text{g}/\text{m}^3$ , 39.56  $\mu\text{g}/\text{m}^3$ , 32.59  $\mu\text{g}/\text{m}^3$  and 26.33  $\mu\text{g}/\text{m}^3$  respectively, generally higher than those in older homes reported in previous studies except benzene. The mean concentration of methylene chloride (47.43  $\mu\text{g}/\text{m}^3$ ) and 1,2-dichloroethane (33.83  $\mu\text{g}/\text{m}^3$ ) were noticeably higher than the levels reported in previous studies in Hong Kong, Japan and Canada. Whereas the mean concentration of 1,4-dichlorobenzene (5.53  $\mu\text{g}/\text{m}^3$ ) were similar to the results of Canadian national survey but lower than those in Japan. The concentrations of 1,2-dichloroethane, 1,4-dichlorobenzene, and methylene chloride, ethylbenzene presented a mean cancer risk at  $7.39 \times 10^{-6}$ ,  $1.95 \times 10^{-6}$ ,  $1.62 \times 10^{-6}$ ,  $1.04 \times 10^{-6}$  respectively, above the US EPA proposed acceptable risk level of  $1 \times 10^{-6}$ . Sensitivity analyses indicated that the VOC exposure concentration have a greater impact than the IUR values on the risk assessment.

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**Conclusion:** This study highlights the characteristics of VOCs in recently renovated homes and has implications for the adverse health effects that result from exposure to chlorinated hydrocarbons in indoor air.

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## 1. Introduction

Indoor Air Quality (IAQ) has been a major public concern for several decades given that city dwellers spend approximately 80–90% of their time indoors. Volatile organic compounds (VOCs), as a common group of indoor air pollutants with large variety of species, continued to get worldwide attention due to their ubiquitous occurrence in indoor environments and their potentially important implications for human health. It has been widely reported that some VOCs may have both short- and long-term adverse health effects on occupants, including sensory irritation, allergies, sick building syndrome (SBS), decrements in lung function, asthma, and even leukemia, particularly in vulnerable groups such as children (Norback et al., 1995; Daisey et al., 2003; Rumchev et al., 2004; Cakmak et al., 2014; Gao et al., 2014).

Due to their potential health effects, studies on investigating the levels of indoor VOCs as well as screening the priority toxic pollutants have been carried out in many countries. In the United States, efforts on clarifying the exposure and source of VOCs were made since the late 1970s. Air toxins such as carbonyls (formaldehyde, acetaldehyde), chlorinated hydrocarbon (1,4-dichlorobenzene, chloroform, carbon tetrachloride), benzene, naphthalene have been identified as pollutants of potential concern indoors (POPCs) in view of several risk-based analyses (US EPA, 2004; Loh et al., 2007; Jia et al., 2008a). In Europe, European Commission's INDEX study classified BTXS (benzene, toluene, xylene, styrene), naphthalene, carbonyls (acetaldehyde, formaldehyde), terpene (limonene,  $\alpha$ -pinene) as priority pollutants to be regulated in dwellings and public buildings. Other EU-funded studies (Sarigiannis et al., 2011; Karakitsios et al., 2015) revealed that for carcinogenic contaminants in EU countries, attributed risk is one to two orders of magnitude higher than the acceptable one of  $10^{-6}$ . In Japan, recent nationwide survey (Azuma et al., 2016) on indoor chemicals exposure assessment showed that the highest risk pollutants included acrolein, benzene, propanal, acetaldehyde, and 1,4-dichlorobenzene.

Household microenvironments have been shown to be the driving factor determining personal exposure to VOCs (Guo et al., 2004; Stocco et al., 2008; Delgado-Saborit et al., 2011; Du et al., 2014a, 2014b; Karakitsios et al., 2015). Previous studies (Lee et al., 2002a; Zhou et al., 2011; Maisey et al., 2013; Wheeler et al., 2013; Gao et al., 2014) identified that recent renovation was an important factor influencing indoor VOC levels due to emission of the building materials, furniture, paints, glues, floor covering and other decorating materials (Hodgson et al., 2002; Ni et al., 2005; Jarnstrom et al., 2008; Guo, 2011; Liu et al., 2014). By utilizing low-emission building materials and eco-friendly consumer products, the indoor air quality appeared to have been improved over time in some developed countries (US EPA, 2011; Zhu et al., 2013). However, in most cases, homes undergone recent renovation had significantly higher VOC concentrations than those in non-renovated homes. These compounds may go through various changes in relation to time, some studies (Park and Ikeda, 2006; Liu et al., 2012; Shin and Jo, 2013; Liu et al., 2013) have shown that VOCs in new or recently renovated dwellings remain elevated for a range from 1 to 5 years.

Over the past two decades, because of rapid urbanization in China, the popularity of decoration and renovation in homes has led to elevated levels of indoor VOCs (Liu et al., 2012). Thus, adverse health effects associated with these compounds has become a major public concern (Zhang et al., 2013). A number of studies (Zhao et al., 2008; Guo et al., 2013; Dong et al., 2013, 2014; Gao et al., 2014) carried out in cities across China reported that recent renovation or use of new furniture in homes were associated with increased likelihood of sick building

syndrome, respiratory symptoms, allergic rhinitis, asthma and leukemia in occupants, especially in children and women. Furthermore, a recent survey in city of Changsha (Deng et al., 2015) revealed that early life exposure to new furniture and home decoration contribute the rapid development of childhood asthma in China.

Thus far, the characteristics of VOCs and their inhalation-related health risks in newly renovated homes have not been well studied in China (Kulmala, 2015). Most studies of indoor air pollution in China (Wang et al., 2007; Weng et al., 2010; Huang et al., 2013; Liu et al., 2013; Zhu and Liu, 2014; Liu et al., 2014; Du et al., 2014a, 2014b) have focused on benzene, toluene, xylene and ethylbenzene (BTEX) or carbonyls (formaldehyde), and little is known about the levels of other VOCs and their potential health impacts. Thus, it remains important to better understand the profile and potential health impacts of VOCs in newly renovated residences.

The objective of this study was to understand the concentration of VOCs in newly renovated homes in Shanghai and the health risks related to inhalation exposure to VOCs. 101 VOCs were measured and their compositions were analyzed. The inhalation cancer risk and chronic toxicity for specific VOCs were calculated using the inhalation unit risk (IUR) and Reference Concentration (RfC) from US EPA as well as OEHHA of CalEPA. Uncertainty and sensitivity analyses were also conducted using a Monte Carlo simulation to determine the overall uncertainty associated with the predicted risks.

## 2. Methods

### 2.1. Sampling area and study design

Shanghai is one of the largest cities in China, with an area of over 6340 km<sup>2</sup> and with population of over 24 million people. It has a humid subtropical climate and experiences four distinct seasons. Home interior decoration and renovation are very popular in Shanghai due to the boom in the urban real estate industry in this megacity.

This study was carried out in May 2015 in Shanghai, China. Eight residences that had been renovated within the past year were studied. Information was collected regarding the construction area, type of decoration materials, and time of decoration.

### 2.2. Sampling method and analysis

The sampling process followed the national standard Technical Specifications for Monitoring of Indoor Air Quality (HJ/T167-2004). Three sampling sites were used in each participating residence: the living room, bedroom, and study. The latter two areas generally had a high amount of pressed-wood furniture. 3.2-Liter Summa canisters (Entech Instruments, Inc.) were used to sample indoor air VOCs. The sampling time of the Summa canisters was set to 45 min by controlling the QT valve. The relative sampling height was 0.5 to 1.5 m, which represents the breathing zone. Before sampling, the rooms were closed for approximately 12 h to simulate bad ventilation during daily life. The rooms were also closed during sampling. No smoking or cooking occurred during the measurements. The temperature, air pressure and other weather conditions were recorded during sampling.

VOC samples were analyzed using a gas chromatograph with a mass spectrometer and a flame ionization detector (GC-MS/FID). The VOC samples were first pumped into a cryogenic pre-concentrator (TH\_PKU-300, Tianhong, China) and then concentrated at  $-150^{\circ}\text{C}$  in two traps. The concentrated VOCs were desorbed at  $100^{\circ}\text{C}$  and injected into the gas chromatograph (7820A, Agilent, USA). The C2–C5

hydrocarbons were separated on a PLOT capillary column (0.32 mm × 15 m, Dikma, USA) and quantified by the FID. The C5–C10 hydrocarbons, halogenated VOCs (x-VOCs), and oxygenated VOCs (o-VOCs) were separated on a DB-624 (0.25 mm × 60 m, Agilent, USA) and quantified using a quadrupole mass spectrometer (MS-5977E, Agilent, USA). The source temperature in the MS was 200 °C and the scan mass ranged from 30 to 300 amu.

The VOC species were identified based on their retention time and mass spectra. A commercial standard gas (Spectra, USA) containing a Photochemical Assessment Monitoring System (PAMS), o-VOCs, and x-VOCs was used to confirm the compound retention time and help identify the compounds. In this study, 101 species were identified, including 27 alkanes, 11 alkenes, acetylene, 16 aromatics, 16 o-VOCs, 29 x-VOCs and acetonitrile.

During the field survey, 2 blank samples and 3 parallel samples were taken for quality control. The relative deviation of the difference between a single measured value and the average of the parallel samples should not exceed 20%.

The target species were quantified using the multipoint internal calibration method. Calibration curves for all species were made before and after analysis and showed good linear regressions ( $R^2 > 0.99$ ). For each batch of 6 samples, a high-purity nitrogen sample and a standard sample (1 ppb) were analyzed to check the peak time and signal intensity. The method detection limits (MDLs) of the various VOC species ranged from 2 to 70 pptv.

### 2.3. Risk assessment method

The risk characterization for indoor VOC inhalation exposure was conducted by combining published toxicity data with the exposure concentrations estimated in this study.

#### 2.3.1. Selection of health-related target VOCs

17 of the VOC species were selected as health-related target chemicals based on the following principles: 1) Agents classified by the IARC in the 1, 2A and 2B groups. 2) Chemicals with confirmed IUR or RfC inhalation toxicity in the US EPA's Integrated Risk Information System (IRIS) or OEHHA of CalEPA. 3) Hazardous chemicals reported by previous research. 4) Chemicals with a detection frequency above

60% in this study; 1,3-butadiene, vinyl chloride and 1,2-dibromoethane were not included in our health-related list due to their low detection frequency in our study. The chemical names and associated toxicity values are presented in Table 1. The inhalation cancer risk or non-cancer hazard risk were calculated based on these parameters.

#### 2.3.2. Cancer and non-cancer risk calculation model

To calculate inhalation risks, an adjusted air concentration ( $EC_a$ ) proposed by US EPA was estimated to represent continuous exposure.

$$EC_a = C_m \times ET \times 1\text{day}/24\text{hours} \times EF \times ED/AT \quad (1)$$

where  $C_m$  is the measured VOC concentration in the residences ( $\mu\text{g}/\text{m}^3$ ); ET is exposure time (hours/day); EF is the exposure frequency (days/year); ED is exposure duration (years); AT is averaging time (days). For cancer and chronic hazard assessments, lifetime (70 years) is substituted for AT.

We adjusted exposure air concentration by incorporating time-activity data of Chinese residents. Based on Exposure Factors Handbook of Chinese Population (MEP, China, 2013) and Wang et al.'s (Wang et al., 2012) study, average exposure time was estimated as 15 h/day for residents. Exposure frequency was estimated as 354 day/year, assuming 11-day annual leave for Chinese residents to go vacation outside. Park and Ikeda (2006) showed that VOCs in new homes decreased dramatically in a few months and closed to the levels of older homes after 1–3 years, varying with different species. Some domestic research (Liu et al., 2013; Gao et al., 2014; Dong et al., 2013, 2014) reported that VOC levels in homes (renovation age < 5 years or changing furniture in the last 5 years) were significantly higher than those in old homes. Based on these studies, exposure duration was estimated as 5 years to calculate inhalation cancer risk attributable to elevated VOCs from residential renovation.

The indoor inhalation cancer risk at residences was calculated with the methodology proposed by US EPA (, 2004)

$$R_j = EC_{aj} \times IUR_j \quad (2)$$

where  $R_j$  is the estimated inhalation cancer risk from chemical j;  $EC_j$  is the adjusted inhalation exposure concentration of chemical j;  $IUR_j$  is

**Table 1**

Target health-related VOCs and related toxicity values.

VOCs	CAS no.	Group (IARC)	IUR ( $\mu\text{g}/\text{m}^3$ ) <sup>-1</sup>		RfC ( $\text{mg}/\text{m}^3$ )	
			Value	Source	Value	Source
Benzene	71-43-2	1	$(2.2-7.8) \times 10^{-6}$	IRIS	0.03	IRIS
Toluene	108-88-3	3	—	—	5	IRIS
m/p-Xylene	106-42-3	3	—	—	0.1	IRIS
	/108-38-3					
o-Xylene	95-47-6		—	—		
Ethylbenzene	100-41-4	2B	$2.5 \times 10^{-6}$	OEHHA	1	IRIS
Styrene	100-42-5	2B	—	—	1	IRIS
Isopropylbenzene	98-82-8	2B	—	—	0.4	IRIS
1,4-Dichlorobenzene	106-46-7	2B	$11 \times 10^{-6}$	OEHHA	0.8	IRIS
Chloromethane	74-87-3	3	—	—	0.09	IRIS
Methylene chloride	75-09-2	2A	$4.1 \times 10^{-6}$	IRIS	0.6	IRIS
1,2-Dichloroethane	107-06-2	2B	$26 \times 10^{-6}$	IRIS	0.4*	OEHHA
1,2-Dichloropropane	78-87-5	1	$10 \times 10^{-6}$	OEHHA	0.004	IRIS
Chloroform	67-66-3	2B	$23 \times 10^{-6}$	IRIS	0.3*	OEHHA
Carbon tetrachloride	56-23-5	2B	$6 \times 10^{-6}$	IRIS	0.1	IRIS
Trichloroethylene	79-01-6	1	$6.8 \times 10^{-6}$	IRIS	0.002	IRIS
Tetrachloroethylene	127-18-4	2A	$0.58 \times 10^{-6}$	IRIS	0.035*	OEHHA
Acetonitrile	75-05-8	—	—	—	0.06	IRIS

Notes: CAS no., Chemical abstracts service number.

IARC, International Agency for Research on Cancer.

Group 1, Carcinogenic to humans; Group 2A, Probably carcinogenic to humans; Group 2B, Possibly carcinogenic to humans; Group 3, Not classifiable based on carcinogenicity to humans.

IUR, Inhalation Unit Risk.

RfC, Reference concentration.

IRIS, US EPA's Integrated Risk Information System.

OEHHA, Office of Environmental Health Hazard Assessment of CalEPA, \*, Chronic REL.

the estimated inhalation unit risk ( $\text{m}^3/\mu\text{g}$ ) for chemical  $j$  (from US EPA IRIS or OEHHA), which is the excess lifetime cancer risk estimated to result from continuous exposure to an individual VOC via inhalation per  $\mu\text{g}/\text{m}^3$ .

For trichloroethylene (TCE), which is carcinogenic by a mutagenic mode of action, the inhalation cancer risk was calculated using the US EPA's procedures with an age group-adjusted specific IUR (USA EPA, 2005).

Based on current studies, additivity of effects was the major mechanisms observed in the health effect assessment for the chemical mixtures at concentrations encountered in non-occupational settings (WHO, 2013). Thus, the total cancer risk posed by target VOCs in this study is the sum of the individual VOCs risks.

The indoor inhalation non-cancer hazard risk at home was calculated using the following equation (US EPA, 2004):

$$\text{HQ}_i = \frac{\text{EC}_{a95i}}{\text{RfC}_i} \quad (3)$$

where HQ is the hazard quotient for the chemical  $i$ ;  $\text{EC}_{a95i}$  is the adjusted 95th percentile inhalation concentration of chemical  $i$ ; Here we used 95th percentile measured concentration to estimate the inhalation exposure under the most unfavorable conditions. RfC is the chronic reference concentration for chemical  $i$  (from US EPA IRIS and OEHHA), which was often derived from NOAEL (No-Observed-Adverse-Effect Levels) or LOAEL (Lowest-Observed-Adverse-Effect Levels) associated with uncertainty factors.

The results of  $R_j$  and  $\text{HQ}_i$  were judged according to US EPA's (, 2004) approach. Namely, cancer risks no higher than  $1 \times 10^{-6}$  for an "ample margin of safety" and non-cancer hazard risk  $<1$  were regarded as acceptable.

### 2.3.3. Uncertainty and sensitivity analysis

To quantify the uncertainty associated with the cancer risk assessment, we performed Monte Carlo simulation and sensitivity analysis using Crystal Ball software (v11.1.2.4). This probabilistic modeling utilizes the entire range of input data to develop a probability distribution of exposure or risk rather than a single point value, which has been commonly used in previous health risk studies (Hu et al., 2007; Zhou et al., 2011; Sofuoglu et al., 2011; Du et al., 2014a, 2014b). Probability distributions of VOC concentrations were determined by fitting continuous distributions (beta, exponential, extreme value, gamma, logistic, lognormal, normal, and Weibull) to measured data using goodness-of-fit tests (chi-squared, Kolmogorov-Smirnov, and Anderson-Darling). In this study, lognormal distribution had best fit for most measured target VOC concentrations based on Anderson-Darling test (prior distribution are listed in Table S2 in the Appendix), similar to previous studies (Loh et al., 2007; Jia et al., 2008b; US EPA, 2011), thus lognormal distribution was used to describe VOCs exposure concentrations. Triangle distribution was used to describe IUR characters. The lowest risk zero was assigned to minimum and IUR values was assigned to maximum and most likely values (Zhou et al., 2011) based on that IUR values recommended by US EPA were assumed to represent maximum and most likely values (Payne-Sturges et al., 2004; US EPA, 2004). During a single trial, values were randomly selected from the defined probabilities for each uncertain variable and then the model output was calculated. The carcinogenic risk and chronic toxicity risk were calculated. To test the convergence and the stability of the output, independent runs were performed for multiple iterations; the results showed that 10,000 iterations were sufficient to ensure simulation stability. Thus, simulated values ( $n = 10,000$ ) were used to estimate the exposure and risk distributions. Sensitivity analysis was conducted to determine the input parameters that most strongly influenced the risk level.

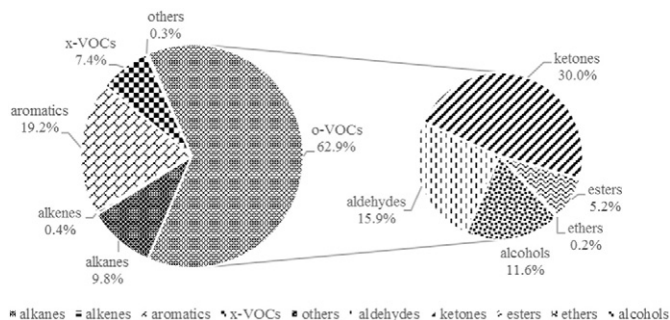


Fig. 1. VOC profile of air samples from residences renovated within the past year in Shanghai. Abbreviations: o-VOCs, oxygenated VOCs; x-VOCs, halogenated VOCs.

## 3. Results and discussion

### 3.1. VOC characteristics in newly renovated residences

Concentrations of the 101 species of indoor VOCs are listed in Table S1 in the Appendix. Of these species, 82 VOCs were found to have a detection frequency  $>60\%$  based on the proportion of samples above the method detection limit (MDL). The VOCs were grouped into 6 categories: alkanes, alkenes, aromatics, o-VOCs, x-VOCs, and others. The o-VOCs were further divided into 5 sub-categories: aldehydes, ketones, alcohols, ethers, and esters. We also calculated the average mass concentration of each category as well as its percentage of the total mass. Fig. 1 shows the mass proportions of different groups of VOCs from air samples analyzed in this study.

Fig. 1 shows that in this study, o-VOCs had the greatest mass proportion of the total VOCs (62.9%), followed by aromatics (19.2%), alkanes (9.8%) and x-VOCs (7.4%). For the o-VOCs, the dominant species by mass were ketones (30.0%), aldehydes (15.9%) and alcohols (11.6%).

Previous studies (Park and Ikeda, 2006; Jarnstrom et al., 2007; Liu et al., 2012) have shown that indoor VOCs concentrations are generally high in newly renovated houses, especially within initial months. The emission of VOCs decrease markedly in 6–12 months if no new source is added. To verify the temporal characteristics of VOCs, the residences in this study were divided into 2 groups based on the completion time of their renovations. Group I ( $<3$  mo), residences had renovations within the past 3 months. Group II (3 mo–1 yr), residences had renovations within the past year but over 3 months prior to testing. The results for the two groups are shown in Fig. 2. The figure shows that the concentration of analyzed total VOCs in group I ( $<3$  mo) were about  $3561.88 \mu\text{g}/\text{m}^3$

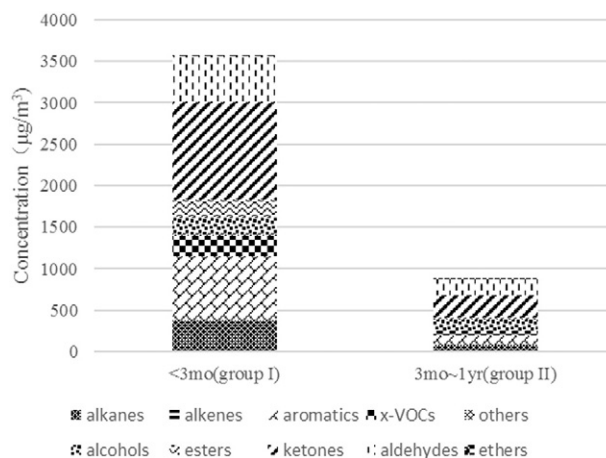
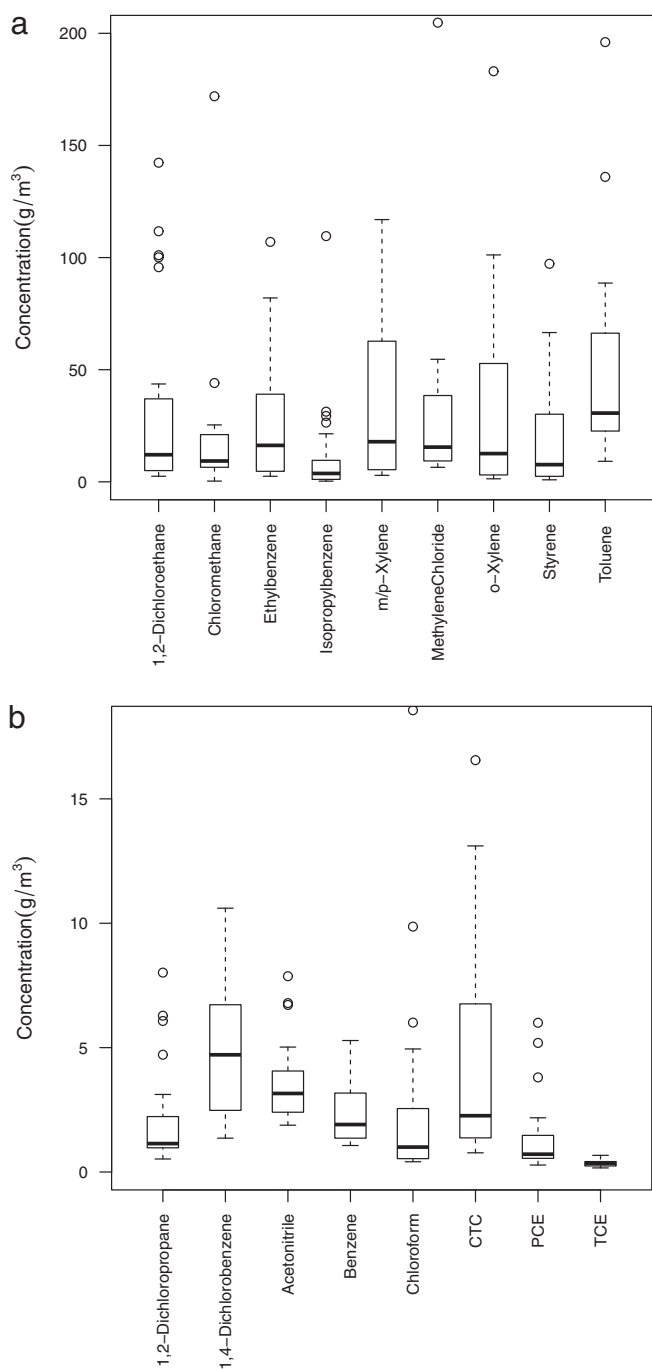


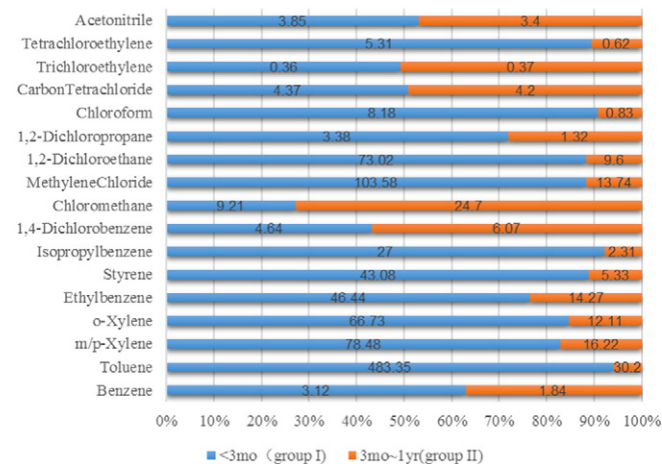
Fig. 2. VOC mass concentrations during different periods after home renovation. Group I refers to VOC levels in residences renovated within the last 3 months. Group II refers to VOC levels in residences where renovations occurred at least 3 months previously but within the last year.





**Fig. 3.** Boxplot of 17 target health-related VOCs, including (a) 9 VOCs with median concentrations ranging from 0 to 50  $\mu\text{g}/\text{m}^3$  and (b) 8 VOCs with median concentrations ranging from 0 to 10  $\mu\text{g}/\text{m}^3$ . Abbreviations: CTC, Carbon tetrachloride; TCE, trichloroethylene; PCE, tetrachloroethylene.

$\text{m}^3$ , approximately 3 times higher than group II (3 mo–1 yr). This proved that the residences renovated within the past 3 months were characterized by high indoor VOC exposure. Although the initial levels of VOCs in group I (<3 mo) decreased dramatically with time, particularly for compounds like aromatics, ketones, alkanes and esters, the VOCs levels for group II (3 mo–1 yr) still exceeded 600  $\mu\text{g}/\text{m}^3$ , which is the TVOC limits specified by national standard GB18883-2002. This suggests that indoor elevated VOCs related to renovation will last a long period.



**Fig. 4.** Percentage accumulation bar plot of indoor levels of target VOCs over two different periods. Group I refers to VOC levels in residences renovated within the last 3 months. Group II refers to VOC levels in residences where renovations occurred at least 3 months previously but within the last year. Data labels indicate mean concentrations (in  $\mu\text{g}/\text{m}^3$ ) of individual VOCs.

### 3.2. Characteristics of health-related VOCs

Fig. 3 shows a boxplot of 17 health-related VOC concentrations, among which 12 VOCs were confirmed or probable carcinogens with IARC ratings. The figure shows that indoor renovation introduced complex chemicals which could pose health threats to occupants. The median individual concentrations of these 17 VOCs ranged from 0.35  $\mu\text{g}/\text{m}^3$  for trichloroethylene to 30.64  $\mu\text{g}/\text{m}^3$  for toluene. For most chlorinated hydrocarbons, the median concentrations were below 5  $\mu\text{g}/\text{m}^3$ . For most aromatics except benzene, the median concentrations were slightly higher, ranging from 9.28  $\mu\text{g}/\text{m}^3$  to 30.64  $\mu\text{g}/\text{m}^3$ .

Temporal changes in the concentrations of 17 target health-related VOCs within the first year were shown in Fig. 4. The levels of most target VOCs in group I (<3 mo) was 0.7–15 times higher than that of group II (3 mo–1 yr), which suggested a decreasing trend for these compounds in new homes. However, there were several exceptions; for trichloroethylene (TCE), carbon tetrachloride (CTC) and acetonitrile, the concentrations in the two groups were close, whereas for chloromethane and 1,4-dichlorobenzene, their concentrations in group II were slightly higher than that in group I. These five chemicals exhibited different variation trend, indicating they may also originate from other indoor sources or infiltration from outdoor air. A previous study has shown that use of moth repellants or deodorizers is associated with higher levels of 1,4-dichlorobenzene in homes (Lee et al., 2002b). Another study carried out in Korean (Shin and Jo, 2012) suggested that six halogenated VOCs exhibited similar concentrations for indoor and outdoor air samples.

Spatial variation of the concentrations of 17 target health-related VOCs in different rooms were shown in Fig. 5. The mean concentrations of most VOCs in bedrooms were higher than those in the living room or the study, moreover, the levels of toluene exceeded the limits of national standard (GB/T 18883). The higher levels of VOCs in bedrooms was most likely due to the use of much plywood furniture like wardrobe, bed and laminated flooring in a relatively small space. This high exposure might cause potential adverse health effects in occupants considering people spend about 8 h in bedroom every day, just as Gao's study (Gao et al., 2014) had found that the use of furniture in bedroom was related to the risk of childhood acute leukemia (AL).

Tables 2 and 3 summarize the concentrations of BTEX and 7 chlorinated hydrocarbons measured in this study and compare them with the results of other studies.

BTEX are the most frequently detected compounds in indoor environments. They mainly come from decorating materials, solvents,



Fig. 5. Percentage accumulation bar plot of indoor levels of target VOCs in different rooms. Data labels indicate mean concentrations (in  $\mu\text{g}/\text{m}^3$ ) of individual VOCs.

detergents, household products and smoking (Du et al., 2014b, Liu et al., 2014; Lu et al., 2008). Current studies (see Table 2) showed that Chinese residential indoor benzene concentrations were far below the level

specified by GB50325 ( $90 \mu\text{g}/\text{m}^3$ ), although the limit value is not protective enough for chronic exposure to benzene from the point of view of public health (Sarigiannis et al., 2011). For renovated homes, the

**Table 2**  
Indoor BTEX concentrations in residences in various cities reported by this study and other studies.

Location/reference	Remarks	Sampling method	Sampling size	Statistical parameter	Concentrations ( $\mu\text{g}/\text{m}^3$ )				
					Benzene	Toluene	m/p-Xylene	o-Xylene	Ethylbenzene
Shanghai, China this study	Renovated within 1 year	Canister sampling for 45 min	8 homes	Mean (sd)	2.32 (1.19)	200.13 (443.89)	39.56 (49.81)	32.59 (42.77)	26.33 (27.73)
Guangzhou, China (Du et al., 2014a, 2014b)	Renovated within 2 years	Passive sampling for 24 h	43 homes	Median Mean (sd)	1.91 18.5 (11.6)	30.64 173.2 (129.5)	17.88 58.1 (62.8)	12.59 40.8 (39.7)	16.24 —
Beijing, China (Liu et al., 2013)	Renovated within 5 years Renovated over 5 years	Active sampling for 30 min	152 homes 255 homes	Mean (sd)	16.3 (14.7) 9.7 (5.7)	45.2 (24.7) 26.4 (13.2)	12.3 (5.3) 11.5 (3.4)	— —	— —
Daegu, Ulsan, Youngcheon, Korea (Shin and Jo, 2012)	Newly built apartments	Active sampling for 30 min	107 homes	Mean (sd)	3.9 (1.6)	184 (112)	14 (8)	2.8 (0.9)	8.2 (3.8)
Shanghai, China (Gao et al., 2014)	Case <sup>a</sup> Control <sup>a</sup>	Passive sampling for 24 h	64 homes 64 homes	Median	2.90 1.80	12.0 9.7	6.90 6.90	— —	— —
Hong Kong, China (Guo et al., 2003)	—	Canister sampling for 8 h	6 homes	Mean (sd) Median	4.99 (2.58) 4.31	59.13 (19.78) 68.19	5.27 (2.56) 4.69	3.89 (2.59) 3.41	2.72 (2.16) 2.42
Tianjin, China, (Zhou et al., 2011)	—	Passive sampling for 5 days	12 homes	Mean (sd)	6.13 (7.58)	7.47 (4.18)	1.55 (1.62)	0.47 (0.47)	1.32 (1.84)
Multiple cities, China (Du et al., 2014b)	10 studies	Canister/active/passive sampling for 1 h–5 days	—	Mean (sd)	5.8 (5.7)	16.6 (33.0)	3.1 (1.9)	3.1 (2.5)	3.1 (1.1)
National survey, Japan (Azuma et al., 2016)	Winter Summer	Passive sampling for 24 h	602 homes	Mean	2.4 1.3	10.8 12.1	8.3 5.8	3.4 2.6	5.6 4.4
Seoul, Korea (Son et al., 2003)	—	Passive sampling for 24 h	30 homes	Median	36.90	54.44	8.69 <sup>b</sup>	9.29	1.22
Three cities, UK (Delgado-Saborit et al., 2011)	—	Active sampling for 24 h	155 samples	Mean (sd)	1.97 (2.41)	17.53 (24.80)	5.84 (8.76)	2.02 (2.95)	1.74 (2.42)
Multiple cities, EU (Sarigiannis et al., 2011)	≥16 studies	Passive sampling (majority of the review)	—	Median	3.1–10.4	20.6–53.1	5.0–18.2	—	—
Multiple cities, US (US EPA, 2011)	15 studies	—	—	Median range	<RL–4.70	4.8–24.0	1.5–14	1.1–3.6	1.0–3.7
Ottawa, Canada (Zhu et al., 2005)	—	Active sampling for 100 min	75 homes	Mean	2.9	11.5	7.5	5.1	—
National survey, Canada (Zhu et al., 2013)	—	Passive sampling	3857 homes	Mean <sup>c</sup>	1.93 (1.56–2.30)	17.80 (14.10–21.51)	14.44 (10.22–18.65)	4.33 (2.97–5.68)	4.22 (2.87–5.57)
Perth, Australia (Maisey et al., 2013)	—	Active/passive sampling for 100 min–7 days	387 homes	Median range	BDL–1.31	2.62–10.10	0.42–2.08	0.32–2.05	0.20–1.36

Abbreviations: BLD, below detection limit; RL, reporting limit; sd, standard deviation.

<sup>a</sup> Case, homes with children that suffered from acute leukemia; Control, homes with normal children.

<sup>b</sup> Only p-xylene was analyzed.

<sup>c</sup> Ranges in parentheses are the 95% confidence intervals.

**Table 3**Concentrations ( $\mu\text{g}/\text{m}^3$ ) of indoor chlorinated hydrocarbons in residences reported by this study and other studies.

Location/reference	Remarks	Sampling description	Statistical parameter	Concentrations ( $\mu\text{g}/\text{m}^3$ )						
				DCA	Chloroform	CTC	TCE	PCE	Methylene chloride	1,4-Dichlorobenzene
Shanghai, China this study	Renovated within 1 year	Canister sampling for 45 min, 8 homes	Median	12.08	1.00	2.26	0.35	0.72	15.48	4.17
			Mean	33.38	3.59	4.26	0.37	2.38	47.43	5.53
			(sd)	(42.24)	(6.66)	(4.28)	(0.15)	(5.31)	(75.66)	(4.18)
Daegu, Ulsan, Youngcheon, Korea (Shin and Jo, 2012)	Newly built	Active sampling for 30 min, 107 homes	Mean	2.2	1.3	<1.1	–	0.8	–	6.7
			(sd)	(2.9)	(2.4)	(<1.1)		(2.3)		(4.1)
Shanghai, China (Gao et al., 2014)	Case <sup>a</sup>	Passive sampling for 24 h, 128 homes	Median	2.3	2.1	1.2	0.80	0.55	–	3.9
	Control <sup>a</sup>			1.5	1.6	2.2	0.80	0.55	1.3	3.5
Hong Kong, China (Guo et al., 2004)	Living room	Canister sampling for 8 h, 6 homes	Mean	–	0.30	–	0.23	0.30	1.30	–
	Kitchen		(8 h average)	–	0.38	–	0.26	0.26	0.98	–
Multiple cities, China (Du et al., 2014a, 2014b)	10 studies	Canister/active/passive sampling for 1 h–5 days	Mean	–	1.4	0.4	1.8	2.5	–	20.6
			(sd)		(0.6)	(0.1)	(0.2)	(0.2)		(3.7)
National survey, Japan (Azuma et al., 2016)	Winter	Passive sampling, 24 h, 602 homes	Mean	0.0	1.1	0.0	0.1	0.4	–	31.0
	Summer			0.2	0.7	0.1	0.0	0.2	–	117.9
National survey, Canada (Zhu et al., 2013)		Passive sampling, 3857 homes	Mean <sup>b</sup>		0.62	0.32	0.21	1.94	–	5.52
					(0.47–0.78)	(0.29–0.35)	(0.06–0.36)	(1.08–2.79)		(4.06–6.97)
Edmonton, Canada (Bari et al., 2015)		Canister sampling, 24 h, 50 homes	Mean	0.70	1.1	–	0.23	1.4	–	0.09
Windsor, Canada (Khanchi et al., 2015)	–	Canister sampling, 24 h, 50 homes	Geometric mean	–	1.667	0.591	0.615	–	1.169	0.275
Multiple cities, US (US EPA, 2011)	15 studies	–	Median range	<RL	<RL–2.4	<RL–0.68	<RL–1.2	<RL–2.2	0.68–61.00	–

Abbreviations: DCA, 1,2-Dichloroethane; CTC, Carbon tetrachloride; TCE, trichloroethylene; PCE, tetrachloroethylene; RL, reporting limit; sd, standard deviation.

<sup>a</sup> Case, homes with children that suffered from acute leukemia; Control, homes with normal children.<sup>b</sup> Ranges in parentheses are the 95% confidence intervals.

mean concentration of benzene ranged from 2.32  $\mu\text{g}/\text{m}^3$  (this study) to 18.5  $\mu\text{g}/\text{m}^3$  (Du et al., 2014b). For non-renovated homes, the benzene level ranged from 1.8  $\mu\text{g}/\text{m}^3$  (Gao et al., 2014) to 9.7  $\mu\text{g}/\text{m}^3$  (Liu et al., 2013), comparable to levels reported in European studies (Sarigiannis et al., 2011). Our results didn't show significant difference of benzene levels between renovated home and non-renovated homes, which is possibly due to the strict regulation of benzene use in solvents or dilutes in civil building engineering in China. However, as alternatives for benzene, toluene and xylene are widely used in interior decoration and home renovation in China, leading to elevated levels of these compounds in indoor air. Our results showed a high mean concentration of toluene (200.13  $\mu\text{g}/\text{m}^3$ ), similar to those reported in studies of Guangzhou (173.2  $\mu\text{g}/\text{m}^3$ ) and Korea (184  $\mu\text{g}/\text{m}^3$ ), which were approximately 10 times higher than the levels of non-renovated homes reported in Du et al.'s (Du et al., 2014a) study. Higher xylene concentrations (72.15  $\mu\text{g}/\text{m}^3$ ) were also found in this study, similar to results reported in Guangzhou (98.90  $\mu\text{g}/\text{m}^3$ ), about 20 times higher than the average level in non-renovated homes (Du et al., 2014a). Few studies have examined ethylbenzene in renovated homes in China. Our results show that ethylbenzene levels in renovated homes were approximately 6–19 times higher than results from studies for non-renovated homes in Hong Kong and Tianjin (Guo et al., 2003; Zhou et al., 2011). In general, home renovation may induce an obvious increase of indoor concentrations of toluene, xylene, ethylbenzene in China. Additionally, toluene was the most abundant BTEX compound in both renovated and non-renovated residences.

Table 3 presents data from 10 studies of indoor chlorinated hydrocarbons both in China and abroad. 2 studies have examined the levels of chlorinated hydrocarbons in newly renovated homes. Compared with homes without recent renovation, our study showed higher

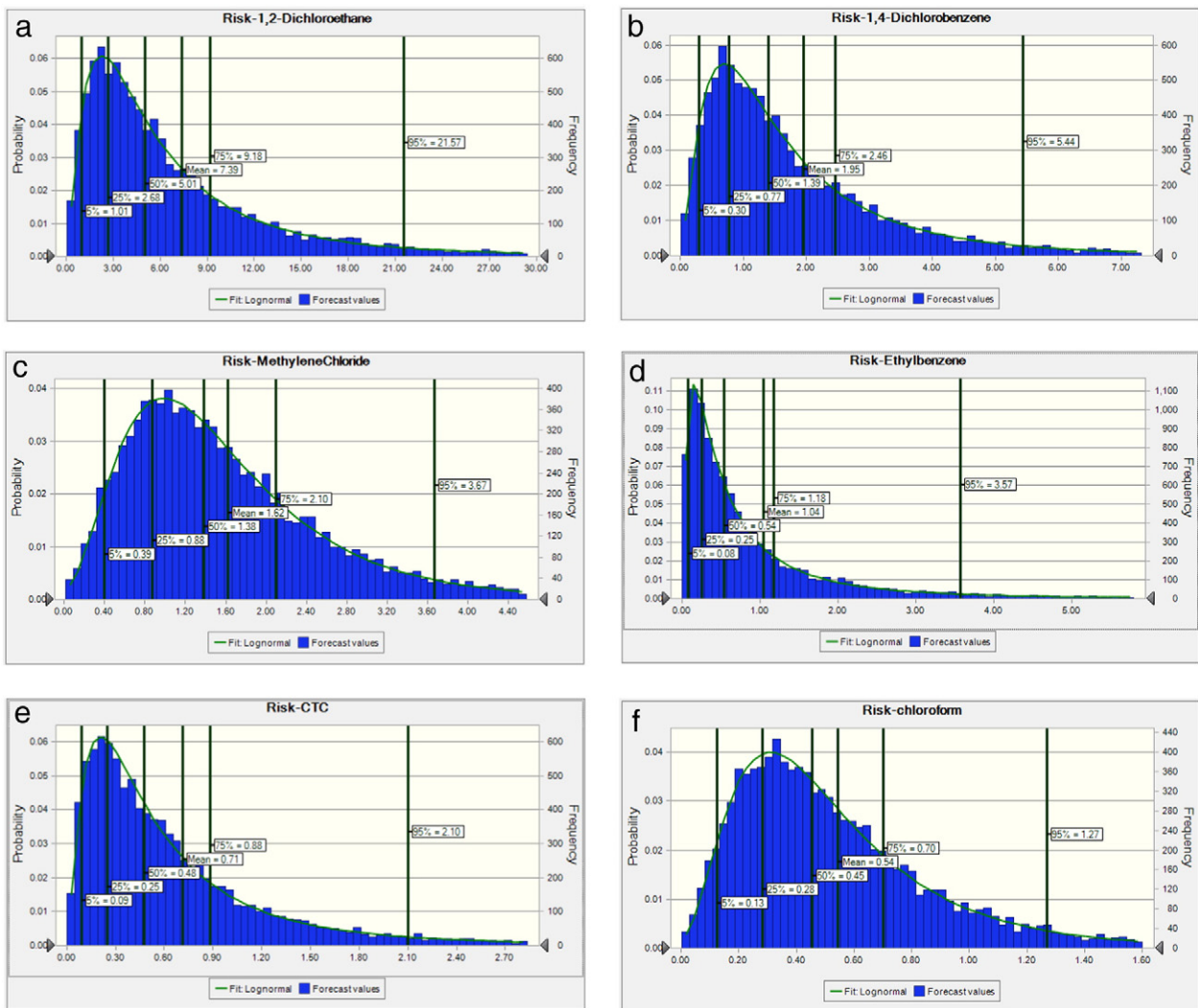
concentrations of 1,2-dichloroethane, methylene chloride, and carbon tetrachloride. The 1,4-dichlorobenzene concentrations in this study are similar to those reported by another Shanghai study (Gao et al., 2014) as well as studies in Korea (Shin and Jo, 2012) and in Canada (Zhu et al., 2013); moreover, our results were lower than those reported by study in Japan (Azuma et al., 2016) but higher than studies in Edmonton (Bari et al., 2015), and Windsor (Khanchi et al., 2015) in Canada. In general, according to current available studies, the median or mean concentration of DCA, chloroform, TCE in Chinese homes were slightly higher than those of studies in Japan, Canada and United States.

Given that some of the indoor VOCs decline dramatically over time, especially within the first 3 months, and because there was a significant variation in VOCs between the two groups in our study, we used the data from group II (3 mo–1 yr) as human inhalation exposure levels. We used these levels in the model to calculate health risks, assuming that these levels would be stable for five years.

### 3.3. Risk assessment results

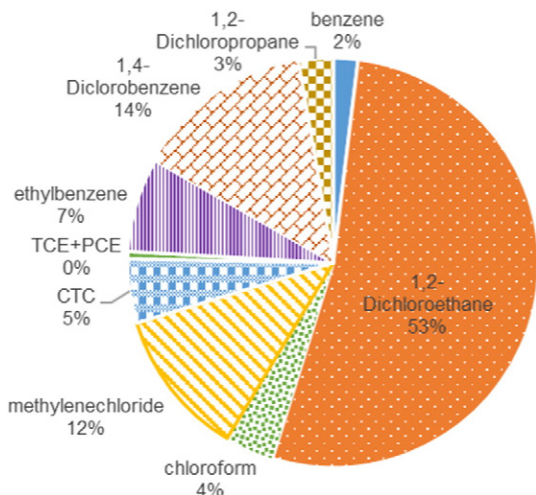
#### 3.3.1. Cancer risk assessment

The cancer risks of 10 VOCs with published IUR parameters were assessed using Eq. (2). Given the probability distributions of the  $\text{EC}_j$  and  $\text{IUR}_j$  parameters, we used Monte Carlo simulation to determine the distribution of risk for the residents. A lognormal distribution had the best fit to the frequency distribution of 9 VOC except that PCE presented a Weibull distribution. The probability distribution (mean, medians as well as several other percentiles (5th, 25th, 75th, 95th)) of the estimated inhalation cancer risk for these VOCs were shown in Fig. 6. Since the estimated cancer risk levels for 4 VOCs (TCE, PCE, benzene,



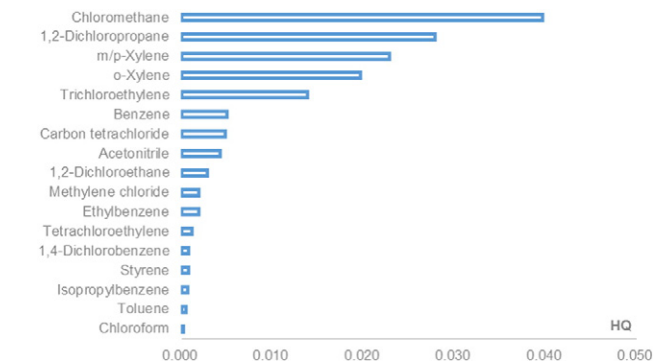
**Fig. 6.** Inhalation cancer risk for 6 VOCs from home exposure (based on an acceptable risk level of one in one million). (a) 1,2-Dichloroethane, (b) 1,4-Dichlorobenzene, (c) MethyleneChloride, (d) Ethylbenzene, (e) CTC, (f) Chloroform. Abbreviations: CTC, Carbon tetrachloride.

1,2-dichloropropane) were below the acceptable risk level of one in a million (US EPA, 2004) even at the 95th percentile, their results are not shown in the figure.



**Fig. 7.** Mean contribution to cumulative cancer risk from 10 target VOCs.

4 chemicals presented a median or mean cancer risk above the acceptable risk level of  $1 \times 10^{-6}$ . 1,2-Dichloroethane presented the highest median and mean cancer risks at  $5.01 \times 10^{-6}$  and  $7.39 \times 10^{-6}$  respectively, followed by 1,4-dichlorobenzene ( $1.39 \times 10^{-6}$  and  $1.95 \times 10^{-6}$ , respectively), methylene chloride ( $1.38 \times 10^{-6}$  and  $1.62 \times 10^{-6}$ , respectively) and ethylbenzene ( $0.54 \times 10^{-6}$  and  $1.04 \times 10^{-6}$ , respectively). For cancer risk estimate



**Fig. 8.** Estimated risk of VOCs using Inhalation Reference Concentration. The risk is estimated as the ratio of the adjusted 95th percentile concentration over Inhalation Reference Concentration (RFC) which is available from US EPA IRIS and OEHA.



at upper-bound 95th percentile, 6 VOCs had incremental risks that exceeded the US EPA benchmark of 1 per one million. 1,2-Dichloroethane still presented the highest cancer risk at  $21.57 \times 10^{-6}$  at the 95th percentile, followed by 1,4-dichlorobenzene ( $5.44 \times 10^{-6}$ ) and methylene chloride ( $3.67 \times 10^{-6}$ ).

The mean cumulative cancer risks obtained by summing the risk of 10 VOCs are 13.98 per one million (median 9.86), spanning from  $2.18 \times 10^{-6}$  to  $3.93 \times 10^{-5}$ , all above the US EPA proposed acceptable risk level of  $1 \times 10^{-6}$ . Fig. 7 compares the mean risk from each of the target VOC. 1,2-Dichloroethane accounts for the greatest portion of the total risk, contributing 53%. 1,4-Dichlorobenzene, ethylbenzene are other significant contributors to the total risk, accounting for 33%.

### 3.3.2. Non-cancer hazard risk

For VOCs with published RfCs in the IRIS or OEHHA database, the risk analysis was performed using Eq. (3). Both individual and the total HQs for all the target VOCs were far less than the threshold ( $HQ = 1$ ) (see Fig. 8). However, HQ are useful for estimates on health endpoints, but they do not provide an estimated probability of effects. Besides, our evaluations were based on the exposure levels of group II (3 mon–1 yr). Therefore, we suggest that caution should be exercised in homes renovated within the past 3 months, where high VOC exposures may exist and could induce acute health effects.

### 3.3.3. Uncertainty and sensitivity analysis

Fig. 9 shows the sensitivity analysis results for inhalation cancer risk for 10 indoor VOCs. The exposure concentration for most VOCs accounted for >50% (ranged from 58.9% to 85.6%) contribution to the estimated risk at home, indicating that the exposure concentration has a greater impact than the IUR values on the risk assessment for these compounds. While for TCE, PCE, benzene, the concentrations in home contributed <50% (ranged from 45% to 49.1%) to the risk estimates, suggesting that IUR values are slightly more influential than VOCs concentrations to the risk estimates. This is possibly because the low concentration level of these 3 compounds measured in this study, moreover, the limited sample size may underestimate the potential variations of these VOCs concentration in home.

In this study, the inhalation cancer risk was expressed simply as the product of the adjusted VOCs concentration with the IUR (US EPA or OEHHA). The uncertainties could arise from the estimates of inhalation exposure. VOCs emission caused by home decoration is a long and complex decaying process, we only capture their concentration levels within a year based on a small sample, which would possibly bias the results. A longer monitoring period study should be designed in the future to obtain more accurate data of the variation of the contaminants for exposure assessment. In addition, in spite of the inhalation concentration was adjusted by time–activity pattern based on Chinese studies, cross-individual variations in activity patterns may cause variability in risk estimates. Furthermore, for the exposure duration, the assumption of

5 years would arouse uncertainties considering the actual variations of different VOCs in newly renovated homes. For BTEX, 5 years may overestimate the exposure risks for their dramatically decrease character, whereas for chlorinated hydrocarbon, 5 years may not enough to estimate the real inhalation risk the residents faced due to their strong persistence in indoor environments. For example, carbon tetrachloride is very stable in the air with a lifetime of 30–100 years.

Uncertainty could also arise from the variable IUR. First, there may be uncertainties in the IUR values because the risk parameters were derived from observable dose responses to estimated low exposure risks through linear extrapolation. Second, the unit risk or RfC provided by US EPA is the risk per  $1 \mu\text{g}/\text{m}^3$  of life-time continuous exposure to the pollutants assuming that a 70 kg adult breathes  $20 \text{ m}^3/\text{d}$ . According to exposure factors handbook of Chinese population, the recommended value of long term inhalation rate in Chinese population is  $15.7 \text{ m}^3/\text{d}$  (male  $18.0 \text{ m}^3/\text{d}$ , female  $14.5 \text{ m}^3/\text{d}$ , respectively). The difference between standard inhalation rate defined by US EPA and the actual status of Chinese population would likely produce uncertainties. Du et al. (2014a, 2014b) reported that the risks were overestimated when variation in inhalation rates and cancer potency factors was not included. However, the default assumptions might bias our estimates but would not alter the ranking of compounds.

### 3.3.4. Implication for practice

The risk assessment results in this study showed that the highest risk chemicals were three chlorinated hydrocarbons (Cl-VOCs). This is consistent with the results of previous studies (Gao et al., 2014; Guo et al., 2013), which demonstrated that higher indoor Cl-VOCs concentrations were associated with an increased risk of childhood AL or adverse symptoms in adults. Cl-VOCs are a group of organic compounds that are frequently detected in indoor air. As mentioned before, interior decorating, household chemicals as well as outdoor infiltration may all introduce chlorinated hydrocarbons. Duan et al. (2016) also reported that chlorinated compounds had higher concentrations during the season without indoor heating in Beijing residences. Due to high volatility and strong persistence in the environment (Huang et al., 2014) as well as carcinogenic effects, some Cl-VOCs (such as 1,4-dichlorobenzene, chloroform, methylene chloride) were identified as indoor pollutants of potential concern in US and Japan. Health-based guidelines or source control measures have been taken in these countries to reduce the exposure to these air toxics.

Even at relatively low levels in indoor environments (median concentrations below  $15 \mu\text{g}/\text{m}^3$ ), Cl-VOCs may present a higher inhalation cancer risk above the acceptable level than other VOCs; therefore, they should arise greater concern. At present, there is no indoor air guideline for Cl-VOCs in China. The standards concerning limit of harmful substances in indoor decorating and refurbishing materials are still at an early stage. Further efforts should be made to limit the harmful substances such as Cl-VOCs in decorating and construction materials as well as household products. On the other hand, in the home, the emission of Cl-VOCs was associated with the use of paints, varnishes, finish removers, adhesives, cleaning solutions and other domestic products. Increase ventilation when using products containing these hazardous chemicals is the effective way to avoid high exposure to Cl-VOCs.

### 3.4. Strengths and limitations

One-hundred-one VOCs were examined in residential indoor air in this study. These data may help us to understand the profile of indoor VOCs in homes because of the number of species analyzed. The temporal characteristics of VOCs were observed over the course of a year, which could help residents decide when to move into newly renovated homes to prevent high VOC exposure. To understand the health risks associated with VOC exposure in recently renovated homes, the inhalation risks of 17 health-related compounds including Cl-VOCs were evaluated. These results supplement the currently limited data

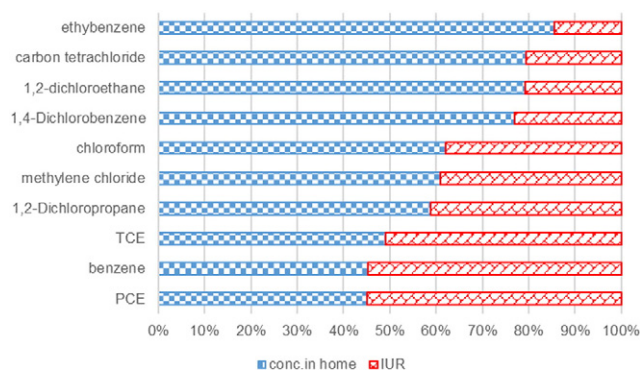


Fig. 9. Sensitivity analysis of inhalation cancer risks for 10 indoor VOCs. Abbreviations: IUR, Inhalation Unit Risk.

regarding VOC characteristics and health effects in newly renovated homes in China.

Carbonyls (formaldehyde and acetaldehyde), as principal indoor air pollutants in US and EU countries, were also priority indoor pollutants in China, they were not analyzed in this study due to limitations of the analytical method. Future research will pay more attention to this kind of compounds.

The total inhalation cancer risk in this study was calculated by summing all the target individual VOCs. Yet, there might be synergies in effect for carcinogens mixtures (benzene, formaldehyde, 1,3-butadiene) or antagonism effects for BTEX mixture. The combined effects of these chemicals are still not sufficiently elucidated so far (Sarigiannis and Gotti, 2008; Sarigiannis and Hansen, 2012; WHO, 2013; Rosch et al., 2014). More comprehensive studies of combined exposure to VOCs mixture in residential environments are needed in the future.

#### 4. Conclusions

This paper reported the results of a preliminary investigation of renovation-related indoor VOC profiles in Shanghai residences. Our results may provide information that is useful for understanding the characteristics of indoor VOCs, their exposure in homes and their potential health risks. Our results revealed that indoor renovation introduced complex chemicals that could pose health threats over certain periods of time. Specifically, we suggest that in addition to BTEX, chlorinated hydrocarbons are a group of indoor pollutants that should be a cause for concern. The sources, variability and effects of this group of VOCs should be clarified to reduce their health threats indoors. Additional studies should be performed with larger sample sizes and longer monitoring periods to characterize indoor VOCs in Chinese residences. These studies would also improve the accuracy of health risk assessments and support more effective indoor air pollution countermeasures and risk management.

#### Acknowledgements

This study was supported by grants from Shanghai Science and Technology Committee (14DZ1207400). The authors thank Wang Qian, Zhou Min, Chen Hao, Wu Mian, Gong Yuhao, and Cao Xiaoxia for assistance with sampling and data analysis. We also thank Professor Qianhua for providing valuable comments.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2016.10.071>.

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