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Indoor occurrence and health risk of formaldehyde, toluene, xylene and total volatile organic compounds derived from an extensive monitoring campaign in Harbin, a megacity of China

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HIGHLIGHTS

- SConducted an extensive monitoring campaign in Harbin, from May 2013 to March 2018.
- There were correlations between the indoor temperature, relative humidity, finish time of decoration and concentration of formaldehyde.
- The predicted carcinogenic risk of formaldehyde was greater than the threshold value at all microenvironments.

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ABSTRACT

Human exposure to formaldehyde, toluene, xylene (FTX) and other volatile organic compounds (VOCs) are associated with negative health impact. To characterize the exposure and health effects of FTX and TVOC from indoor environments, we conducted an extensive monitoring campaign involving 1278 measurements of 472 indoor locations in Harbin, a megacity in China from May 2013 to March 2018. The results showed that household had the highest mean formaldehyde concentration (0.171 ± 0.084 mg m-3) among all types of indoor environments. Meanwhile, there was no significant differences in formaldehyde concentration of the living room, master bedroom, secondary bedroom and study room ($p > 0.05$), as well as toluene and xylene. The highest mean concentration of toluene, xylene and TVOC was measured in public bath center. Great difference was found between formaldehyde concentrations in 2013 and other years, except 2015. There were great positive nonlinear correlations between the indoor temperature and concentration of formaldehyde ($p < 0.01$), good negative nonlinear correlations between the finish time of decoration and concentration of formaldehyde ($p < 0.01$), good positive linear correlations between the relative humidity and concentration of formaldehyde ($p < 0.01$). A risk assessment methodology was utilized to evaluate the potential adverse health effects of the individual FTX compounds according to their carcinogenicities. The predicted carcinogenic risk of formaldehyde was greater than the threshold value 1E-06 at all environments. The non-carcinogenic risk of TX compounds in the population is negligible. For estimating human health risk exposure, sensitivity analysis showed that more attention should be given to the influential variables such as the level of pollutants.

1. Introduction

A variety of chemicals are used indoors and emitted to indoor air. Of all the chemicals with indoor sources, formaldehyde, toluene and xylene (FTX) are the most commonly detected due to

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their wide uses in building materials, furniture, flooring, wallpaper, and adhesives (An et al., 2011; Du et al., 2014; Durmusoglu et al., 2010). With multiple indoor sources, and limited air exchange of indoor and outdoor, indoor concentrations of these contaminants are generally higher than that of outdoors (Gilbert et al., 2008). Airborne levels of selected VOCs including toluene and xylene that were measured in both indoor and outdoor air of 75 residential houses in Canada (Zhu et al., 2005). The results showed that the VOCs including toluene and xylene were present at significantly higher levels and the indoor/outdoor ratios (I/O) > 1, which indicating that the presence of indoor sources.

Indoor air pollution has become a public health concern due to the elevated indoor chemical concentrations and the majority of time spent in indoor environments by the general public. Human exposures to FTX are associated with adverse health effects such as induction and possible neurological responses including weakness, loss of appetite, fatigue, confusion, and nausea (World Health Organization, 2010). Long-time exposure to these contaminants can cause asthma, affect the reproductive and immune systems and even cause cancer (Du et al., 2014; Sax et al., 2006; Sousa et al., 2011). An assessment of health risk was taken from VOCs in three primary schools in I'zmir, Turkey (Sofuoglu et al., 2011). Suggested formaldehyde had the highest chronic toxic and carcinogenic risk than naphthalene, benzene, and toluene. The average lifetime cancer risks of formaldehyde in Bangkok, Thailand exceeded 1E-06 (Tunsaringkarn et al., 2014), which is the safety limit of exposure established by the US EPA.

To protect the general public from excessive exposure to indoor FTX and TVOC, China has issued indoor air quality standards (GB/T18883-2002) based on health risk associated with long-term exposures. The standards regulate indoor air concentrations of FTX and TVOC to 0.08, 0.2, 0.2 and 0.5 mg m^{-3} respectively. Despite of the regulations, indoor air concentrations of these toxic chemicals in certain indoor locations can still exceed the indoor air quality standards (Kim et al., 2001; Sousa et al., 2011; Tang et al., 2009). Depending to a study on indoor VOCs in Xi'an, China, 67% and 78% of measured formaldehyde and TVOC concentrations in underground malls exceeded the national indoor air quality guidelines (Chang et al., 2017). Because of the concerns on the exposure to the elevated levels of VOCs and associated health risk, there is a requirement to identify the indoor environments such as the hospital where elevated levels of VOCs tend to occur. Indoor VOCs concentrations and associated health risks are varied from indoor environments (Guo et al., 2004). Variations in environmental parameters such as temperature, humidity and air exchange rate can also cause variety in VOCs concentrations and health risk (Tao et al., 2015; Zhang et al., 2007). It has been observed that ventilation and heating systems can affect formaldehyde concentration (Hadei et al., 2018; Guo et al., 2013). Since the variety of indoor sources and environments, extensive surveys covering a wide range of indoor locations are needed to assess the health risk associated with indoor human exposures and to evaluate factors that cause elevated health risks.

In the present study, we seek to assess human exposure to FTX and associate health risk of different types of indoor environments. To gain the data onto the evaluation, we conducted a monitoring campaign from 2013 to 2018 in 13 types of public indoor locations and 396 residential flats in Harbin, a megacity in Northeast China. The exposure and risk assessment based on the extensive monitoring survey would provide valuable information on the practice of source mitigation and regulation of indoor FTX in China.

2. Materials and methods

2.1. Sampling sites

Concentrations of formaldehyde, toluene, xylene and TVOC in 396 households and 13 types of public places were monitored in Harbin, China from May 2013 to March 2018. The selected sites were decorated over the past 1–37 months. The types of public places are shown in Table 1. In residential flats, our measurements were conducted in the living room, master bedroom, secondary bedroom, study and others (containing bathroom, cloakroom, kitchen, piano room, storage room and recreation room).

2.2. Sampling and analysis

Room doors and windows were kept closed for 12 h before sampling process. The samplers were placed in the middle of the sampled rooms with a height of 1.5 m above the floor and 1 m away from walls. During sampling, general descriptions including indoor temperature, housing type, decoration time, and furniture density were recorded. After sampling is complete, seal both ends of the sampling tube and bring the sample back to the lab for immediate processing.

VOCs was collected in seamless stainless steel tubes packaged with electronically controlled air sampler (DDY-1.5, Xingyu, China), 0.20 mg Tenax-TA adsorbent (60–80 mesh) and kept for 20 min at a speed of 5 L min^{-1}. Activation of the sampling tube with ultra-pure N_2 (>99.999%) requires that the tube be kept at 350 °C for 1 h before sampling, then sealed with a sealing film and covered with a plastic cap. VOCs were analyzed by a system consisting of a Thermal Desorber (TD) (Markes Unity, USA) coupled with a gas chromatography (GC) (Thermo, Trace 1300) fitted with a flame ionization detector (FID) (Thermo). The GC/FID is equipped with a TG-624 capillary column (30 mx0.23 mmx1.40 μm, Thermo). The sampling tubes were thermally desorbed at 300 °C for 10 min with pure helium gas passing through, and carrying the desorbed gases was carried to a pre-concentration trap at −10 °C. After the tube desorption, the trap was thermally desorbed at 300 °C for 3 min and then the gas was transferred to the GC/FID for measurement. The GC oven temperature was started at 40 °C and held for 5 min, then heated to 240 °C at a rate of 5 °C min^{-1}, and finally held at 240 °C for 5 min. The flow rates for H2 and air were 40 mL min^{-1} and 400 mL min^{-1}, respectively. The quantification of target VOCs was accomplished by using multipoint external standard curves. To prepare the calibration solution, nine kinds of VOC mixed series III solution (IERM, China) in methanol was used. A seven-point calibration (0.5, 1, 5, 10, 20, 50, 100, and 200 μg mL^{-1}) was performed for quantifying the pollutants in the samples. The correlation coefficients were calculated and >0.99 was deemed to be acceptable. A calibration standard was run on a daily basis to ensure the stabilization of the instrument. Detailed information is shown in GB50325-2010.

Formaldehyde was sampled at 0.5 L min^{-1} for 20 min, and was determined by the 3-methyl-2-benzonthiazolinone hydrazine (MBTH) method through a UV–Visrecording spectrophotometer (detector UV@630 nm, UV-2401PC, Shimadzu, Japan). The reagent preparation and analytical methods are detailed in the Chinese National Standard GB/T 18204.26-2000.

2.3. QA/QC

The quality control program followed during the present study included field blanks and parallel samples. To collect the field

Table 1
Numbers and mean concentrations of FTX and TVOC (mg m^{-3}) at different microenvironments.

Site	Numbers	Formaldehyde			Toluene			Xylene			TVOC		
		Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max
Beauty parlor	1	0.090 ± 0.000	0.09	0.09	0.150 ± 0.000	0.15	0.15	0.170 ± 0.000	0.17	0.17	0.550 ± 0.000	0.55	0.55
Fitness Center	3	0.093 ± 0.006	0.09	0.10	0.123 ± 0.063	0.05	0.16	0.157 ± 0.040	0.11	0.18	0.543 ± 0.046	0.49	0.57
Public bath center	2	0.085 ± 0.007	0.08	0.09	0.155 ± 0.007	0.15	0.16	0.180 ± 0.000	0.18	0.18	0.555 ± 0.007	0.55	0.56
Kindergarten	23	0.076 ± 0.028	0.04	0.17	0.094 ± 0.042	0.03	0.16	0.106 ± 0.050	0.04	0.18	0.390 ± 0.124	0.20	0.59
Hospital	1	0.070 ± 0.000	0.07	0.07	0.070 ± 0.000	0.07	0.07	0.070 ± 0.000	0.07	0.07	0.360 ± 0.000	0.36	0.36
Stock exchange	3	0.103 ± 0.035	0.07	0.14	0.120 ± 0.000	0.12	0.12	0.143 ± 0.012	0.13	0.15	0.410 ± 0.036	0.37	0.44
Office	8	0.096 ± 0.051	0.03	0.17	0.103 ± 0.031	0.06	0.15	0.103 ± 0.029	0.08	0.17	0.460 ± 0.067	0.41	0.58
Research laboratory	8	0.139 ± 0.059	0.08	0.22	0.114 ± 0.033	0.07	0.17	0.133 ± 0.025	0.10	0.18	0.373 ± 0.023	0.36	0.41
Plastic card manufacturing plant	6	0.147 ± 0.012	0.13	0.16	0.143 ± 0.037	0.10	0.18	0.130 ± 0.030	0.10	0.16	0.552 ± 0.013	0.54	0.57
Elderly activity room	2	0.125 ± 0.007	0.12	0.13	0.095 ± 0.007	0.09	0.10	0.065 ± 0.007	0.06	0.07	0.420 ± 0.000	0.42	0.42
Health center	2	0.020 ± 0.000	0.02	0.02	0.055 ± 0.007	0.05	0.06	0.060 ± 0.000	0.06	0.06	0.370 ± 0.000	0.37	0.37
Primary school	13	0.141 ± 0.056	0.09	0.26	0.103 ± 0.015	0.08	0.12	0.105 ± 0.027	0.04	0.13	0.418 ± 0.036	0.39	0.46
Government	4	0.068 ± 0.005	0.06	0.07	0.058 ± 0.005	0.05	0.06	0.050 ± 0.012	0.04	0.06	0.435 ± 0.010	0.43	0.45
Household	396	0.175 ± 0.084	0.01	0.78	0.093 ± 0.029	BDL	0.29	0.092 ± 0.029	0.02	0.29	0.411 ± 0.029	0.28	0.48

blanks, the sampling tubes were opened on-site of the participants for a very short period, and then was transported to the laboratory along with other samples for immediate processing. The blank was tested and no significant contamination was found. Good agreement was found in parallel samples, with a relative standard deviation of less than 15%. The pump was calibrated using a flow controller (Bios Defender 510, USA) before each sample. The flow was recorded at the beginning and end of the sampling to ensure that the error in the calculated variation of the flow was less than 10%. Limit of Detection (LOD) was calculated for each analyte using five field blanks with values were 1.8E-03 μg mL^{-1}, 2.12 μg m^{-3}, and 0.42 μg m^{-3} for FTX respectively.

2.4. Health risk assessment

Formaldehyde is a hazardous air pollutant that can induce adverse effect on human health. It has been classified as a probable human carcinogen (group B1) (National Research Council, 2011). In contrast, toluene and xylene are not classified as human carcinogen (Group 3; IARC, 2014). Individual FTX compounds may lead to serious health hazards. Hence, the adverse health effects of exposure to these compounds should be characterized.

The carcinogenic risk posed by formaldehyde was estimated as a simple multiple of the CDI and a slope factor proposed by USEPA (2013):

$$CR = CDI \times SF \tag{1}$$

where CDI and SF are the chronic daily intake (mg kg^{-1} d^{-1}) and slope factor (mg kg^{-1} d^{-1}) $^{-1}$for the interest compound, respectively.

Non-carcinogenic risk is characterized in terms of a hazard quotient (HQ). The HQs for TX was calculated proposed by USEPA (2013):

$$HQ = \frac{CDI}{RfD} \tag{2}$$

where RfD is the reference dose (mg kg^{-1} day^{-1}).

CDI in mg kg^{-1} d^{-1} was calculated with the methodology proposed by USEPA (2013):

$$CDI = \frac{CA \times IR \times ED \times EF}{BW \times AT} \tag{3}$$

where CA is the contaminant concentration (mg m^{-3}); IR is the inhalation rate (m^3 d^{-1}); ED is the length of exposure (a); EF is the exposure frequency (d a^{-1}); BW is the body weight (kg); AT is the average time of lifetime(d). According to the IRIS system, the slope factor of formaldehyde is 0.0455 mg^{-1} kg d, RfD for toluene and xylene are 1.43 and 0.029 mg kg^{-1} day^{-1} (IRIS, 2009; Durmusoglu et al., 2010),respectively. In China, the work lifetime is assumed to be 40 years. The duration of schooling are 3 and 6 years for kindergarten and primary school respectively.

2.5. Uncertainty analysis

The uncertainty and variability of the parameters is the major sources of un-predictability for exposure assessment models. The probability distribution was specified by setting up a range of values and allocating probability weight for each selected risk variable, i.e. body weight (BW), inhalation rate (IR). Lognormal destitution of FTX concentrations were observed and used in this study. Selected types of probability distribution for other input variables to predict probability density functions were listed in Table 2. In order to quantify this uncertainty and its impact on the estimation, Monte Carlo simulation and sensitivity analysis were implemented using Oracle Crystal Ball software. To obtain a true picture of the distribution of risks, 5000 iterations were performed and a histogram of risks produced.

3. Results and discussion

3.1. Concentration of FTX and TVOC in different microenvironments

FTX and TVOC were measured at different indoor locations of Harbin are summarized in Table 1. There were 13 public locations (beauty parlor, fitness center, public bath center, kindergarten, hospital, stock exchange, research laboratory, plastic card manufacturing plant, elderly activity room, health center, primary school, government, office). FTX and TVOC were found in all microenvironments, demonstrating that the indoor air in Harbin was seriously polluted. The mean concentrations of FTX and TVOC were in the range of 0.020–0.175 mg m^{-3},0.055–0.155 mg m^{-3}, 0.050–0.180 mg m^{-3} and 0.360–0.555 mg m^{-3} respectively, in all microenvironments. Household had the highest mean concentration of formaldehyde (0.171 ± 0.084 mg m^{-3}) in all indoor microenvironments. The highest concentration of formaldehyde in household was 0.78 mg m^{-3}, over 9 times higher than the Chinese indoor air quality standard of 0.08 mg m^{-3}, which was consistent with previous study performed in households in Hangzhou, China (the mean concentration of formaldehyde was 0.179 mg m^{-3}) (Chi et al., 2016). But almost an order of magnitude higher than the

Table 2
Parameters for the equations to predicted probability density functions.

Parameters	Kindergarten student	Primary school student	Adult female	Adult male
Body weight (kg)[a]	N (18.9,2.39)	N (33.27,6.74)	N (59.50, 9.06)	N (71.01, 10.91)
Inhalation rate (m^3 d^{-1})[b,c]	U=(min, 7.9; max, 9.3)	U=(min,9.7; max12.3)	T= (min, 11.21; ML, 12.59; max, 13.68)	T=(min,14.26; ML,16.20; max,17.36)
Average time of lifetime(d)[a]	27,730	27,730	28,790	26,830
Exposure frequency at school/office (d a^{-1})[b,c],	LN (50.69, 27.64)	LN (53.8, 13.6)	LN (79.71, 29.23)	LN (85.81, 32.60)
Exposure frequency at home (d a^{-1})[b, c]	–	LN (221.28, 51.20)	LN (240.27, 67.68)	LN (223.31, 65.7)

Note. N Normal Distribution and the two values in the parentheses represent mean and standard deviation.
U Uniform Distribution; T triangular distribution, ML most likely.
LN lognormal Distribution and the two values in the parentheses represent mean and standard deviation.
[a] Chinese National Physique Monitoring Communique (2010) (in Chinese). Available from: http://www.gov.cn/test/2012-04/19/content_2117320.htm.
[b] MEPPRC, 2013.
[c] USEPA, 2011.

research conducted in Perth, Western Australia (Dingle and Franklin, 2002) and Quebec City, Canada (Gilbert et al., 2006), with mean concentration of 0.028 mg m^{-3} (ranged from 0.003 to 0.0923 mg m^{-3}) and the geometric mean of 0.0295 mg m^{-3} (ranged from 0.0096 to 0.090 mg m^{-3}) respectively. Demonstrating that compared with other countries, more serious indoor formaldehyde pollution is found in China, relevant departments should take measures to solve the problem. Formaldehyde is the major indoor pollutants in Harbin. All indoor environments exceed the national standard except health center, government, hospital and kindergarten. Toluene and xylene in all environments did not exceed the national standard. For TVOC, only four indoor environments (public bath center, beauty parlor, fitness center and plastic card manufacturing plant) exceed the standard. The concentration of FTX and TVOC in public bath center, beauty parlor, fitness center and plastic card factory is greater than other places. Since various environmental factors stimulate the release of pollutants. High temperature and humidity, and limited air exchange will stimulate the emissions of FTX and TVOC in public bath center (Tao et al., 2015; Zheng and Liu, 2002). Luxurious decoration and extensive use of air fresheners, perfumes and other skin care products result in high detection concentrations in a beauty parlor. In the fitness center, large airflow and poor ventilation, coupled with a variety of fitness equipment resulting in the elevated concentration (Zhang and Yu, 2016). In the plastic card manufacturing plant, there were various glues, plastic cards and card-making instruments (Sarigiannis et al., 2011), which are the emission sources of those pollutants. The presence of elevated concentrations in such widely frequented microenvironments could therefore have appreciable implications for public health.

The concentration of FTX and TVOC in health center, government, hospital and kindergarten is relatively low, with mean concentrations were 0.020–0.076 mg m^{-3}, 0.055–0.094 mg m^{-3}, 0.050–0.106 mg m^{-3} and 0.360–0.435 mg m^{-3} respectively. Some studies have been reported on the indoor concentrations levels of FTX and TVOC in hospital and kindergarten (Takigawa et al., 2004; Yang et al., 2009; Yang, 2015). The concentrations of formaldehyde, toluene and TVOC are 20 μg m^{-3} (range: 6–32 μg m^{-3}), 463 μg m^{-3} (range: 197–1494 μg m^{-3}) and 860 μg m^{-3} (range: 197–2679 μg m^{-3}) respectively in a newly constructed hospital in japan (Takigawa et al., 2004). The mean concentrations of TVOC and formaldehyde measured in Korean kindergartens were 0.64 mg m^{-3} (range: 0.26–1.024 mg m^{-3}) and 0.17 mg m^{-3} (range: 0.04–0.62 mg m^{-3}) respectively (Yang et al., 2009). The concentrations of toluene and xylene in the indoor air of newly built kindergarten in a district of Tianjin were 0.19 mg m^{-3} (range: 0.06–0.34 mg m^{-3}) and 0.18 mg m^{-3} (range: 0.04–0.38 mg m^{-3}) (Yang, 2015) respectively. It can be concluded that in the present study, except for the concentration of formaldehyde in hospital which is a slightly higher than that in Japan, the concentrations of other pollutants are at a low level. It indicates that indoor pollution of hospitals and kindergartens in Harbin is at a low level. Hospital and health center are inhabited by people whose health is affected, the air quality can directly affect the health and rehabilitation of patients, as well as the occurrence of infections, thus endangering the patients and employees of these institutions (Lu et al., 2006; Sousa et al., 2011). Since most of the occupants in kindergarten are children aged 3–6 years old, they have poor immunity and are vulnerable to pollutants. Therefore, indoor decoration materials are generally environmentally friendly, and most of the playing equipment is at outdoors, which reducing the pollutants discharged into the room, as well as the ventilation is ample, resulting in the better indoor air quality. The government buildings have been built for decades, and thus have less ongoing emission.

3.2. Variations in different types of indoor environments

Summary statistics of FTX and TVOC and frequencies of concentrations exceeding the indoor air quality standards at different types of household are shown in Table 3. The over-standard rate ranges of FTX are from 88.29 to 96.43%, 0.34–1.22% and 0.34–2.44%, respectively. The Independent T-test was applied to compare the difference between the concentrations of pollutants in different room types. There were no significant differences in formaldehyde concentrations between the living room, master bedroom, secondary bedroom, study and other types of room ($p > 0.05$). This finding is in line with results of Dingle and Franklin (2002) and Khoder et al. (2000), who found no significant differences between the concentrations of formaldehyde among different types of rooms. Higher formaldehyde concentration was detected in the master bedroom and living room (0.179 and 0.175 mg m^{-3} respectively), which are consistent with the results of Tang et al. (2014), who found that the concentration of formaldehyde in bedrooms was slightly higher than those in living room and other rooms. The study room had the highest exceeding rate (96.43%) followed by the master bedroom, living room, secondary bedroom and others (92.95%, 92.3%, 91.05% and 88.29% respectively). Meanwhile the mean concentrations of toluene and xylene in the study room were the highest (0.10 mg m^{-3} for both). Due to a large number of books and documents made by various organic

Table 3
Descriptive statistics of formaldehyde, toluene, xylene and TVOC (mg m^{-3}) at different room types of household.

Pollutants	Types	Number	Mean ± S.D	Min	Max	PES (%)
Formaldehyde	Living room	286	0.175 ± 0.084	0.03	0.57	92.30
	Master bedroom	299	0.179 ± 0.087	0.01	0.62	92.95
	Secondary bedroom	268	0.172 ± 0.085	0.02	0.78	91.05
	Study	84	0.171 ± 0.067	0.06	0.35	96.43
	Others	111	0.169 ± 0.099	0.06	0.66	88.29
	All	1048	0.175 ± 0.085	0.01	0.78	92.20
Toluene	Living room	281	0.091 ± 0.030	0.03	0.25	0.71
	Master bedroom	294	0.094 ± 0.028	BDL	0.29	0.34
	Secondary bedroom	263	0.094 ± 0.028	0.02	0.25	0.38
	Study	82	0.101 ± 0.027	0.05	0.21	1.22
	Others	111	0.089 ± 0.034	0.02	0.26	0.91
	All	1031	0.093 ± 0.029	BDL	0.29	0.71
Xylene	Living room	281	0.091 ± 0.027	0.03	0.25	1.07
	Master bedroom	294	0.092 ± 0.027	0.04	0.21	0.34
	Secondary bedroom	263	0.091 ± 0.029	0.02	0.29	0.38
	Study	82	0.101 ± 0.033	0.05	0.24	2.44
	Others	111	0.088 ± 0.031	0.04	0.21	0.90
	All	1031	0.092 ± 0.029	0.02	0.29	1.03
TVOC	All	308	0.411 ± 0.029	0.28	0.48	0

PES: Percent of Exceeding Standards; BDL: Below the Detection Limit.

materials, including cellulose (i.e. paper, some textile wood), synthetic materials (i.e. books, cover materials) and ink, pigments, adhesives, glue, ect (Cincinelli et al., 2016; Gibson et al., 2012), lead to the generation of air pollutants. Master bedroom and secondary bedroom had the same mean concentration of toluene (0.094 ± 0.028 mg m^{-3}) and have no statistical difference from that of the living room (0.091 ± 0.030 mg m^{-3}). Same mean concentration of xylene was found in the living room and secondary bedroom (0.091 mg m^{-3}). Other types of indoor locations (i.e., bathrooms, cloakrooms, kitchens, piano rooms, storage rooms, and recreation rooms) with fewer furniture and more concise decoration materials were found with lowest FTX concentrations are 0.169, 0.089, and 0.088 mg m^{-3}, respectively.

Formaldehyde is the main pollutant in household, and its concentration in different rooms all exceeded the national standard of 0.08 mg m^{-3}. The over-standard rate of formaldehyde in residential flats in Harbin is much higher than other cities in China, such as Haikou (52.2%) (Xiao et al., 2016), Changchun (20%) (Du, 2017), Hangzhou (56.1%) (Chi et al., 2016) and Xi'an (83.6%) (Chang et al., 2017). On the other hand, for toluene and xylene, the over-standard rate are 0.71% and 0.90%, respectively. TVOC concentration in all room did not exceed the national standard. The result is consistent with the previously reported indoor air pollution levels in decorated residences in Xi'an, China (Chang et al., 2017), indicating that the TVOC and toluene concentration in most locations was lower than the standard value. Obviously, compared with toluene, xylene and TVOC, Harbin, as a megacity in China, faced a serious situation of indoor formaldehyde pollution. High formaldehyde concentration in the household is primarily due to multiple emission sources (Du et al., 2014). Correlation analysis is a useful tool for source identification, good correlations give indication of common sources for contaminants. In the present study, the correlation coefficients between FTX concentration levels (see Table 4) are ranging from 0.304 to 0.596 and with highly statistically significant ($p < 0.01$), suggesting that the FTX could have similar and/or common sources.

Table 4
Pearson correlation coefficients for FTX in household samples.

Pollutants	Formaldehyde	Toluene	Xylene
Formaldehyde	1		
Toluene	0.343**	1	
Xylene	0.304**	0.596**	1

** Significant level at 0.01.

Indoor/Outdoor (I/O) ratio means the source of pollutants. When the ratio is less than 1, it is an outdoor source, and greater than 1 are an indoor source. Numerous studies have learned the I/O ratios of FTX. Khoder et al. (2000) found that formaldehyde I/O values ranged from 1.08 to 4.43 in 7 different age flats; meanwhile the 1.08 was a flat age of 43 years. Hadei et al. (2018) found that in Tehran, Iran, the I/O ratios of formaldehyde and xylene greater than one, while toluene is reversed. Zhu et al. (2005) found in Ottawa, Canada the I/O ratio of toluene and xylenes all weighty than one. Similar result was found in Guangzhou, China (Lu et al., 2006). Consequently, elevated levels of FTX in the residences were principally contributed by indoor sources. With the increasing of living standard in China, the public pays great attention to interior decoration. However, decoration materials such as wallpaper, painting and floor tend to emit various pollutants, including formaldehyde (Gunschera et al., 2013; Xiong and Zhang, 2010; Zhang et al., 2007). Meanwhile it was reported that indoor ozone chemistry could play a role in generating indoor aldehydes, which were generated by the reaction of ozone with VOCs (Dutta et al., 2009). In Harbin, the cold winter result in the lower air exchange rate causes an elevated indoor formaldehyde concentration.

3.3. Temporal changes in air quality of household

Post-hoc Tukey's test for the year's change of FTX and TVOC is listed in Table 5. Changes in FTX and TVOC concentrations from 2013 to 2018 in household are presented in Fig. 1. The concentration of formaldehyde in 2013 was significantly different ($p < 0.05$) from that in other years, except in 2015 (Table 5), the formaldehyde concentrations in 2013 and 2015 were greater than that of other years (Fig. 1). The concentrations of FTX in 2015 were significantly different from that in 2016, 2017 and 2018 ($p < 0.05$), no significant difference in the concentrations of toluene and xylene between 2016, 2017 and 2018 ($p > 0.05$), so as to formaldehyde between 2017 and 2018. However, concentrations of FTX gradually decreased from 2015 to 2018 (Fig. 1). It means that indoor air quality in Harbin had only slightly improved. There was no significant differences in TVOC concentrations in other years ($p > 0.05$) except 2016. The concentration of toluene, xylene and TVOC did not exceed the indoor air quality standards in these years. However, the concentration of formaldehyde in all years exceeded the standard,

Table 5
Post-hoc Tukey's test for the year change of FTX and TVOC.

	2013	2014	2015	2016	2017	2018		2013	2014	2015	2016	2017	2018
Formaldehyde							**Toluene**						
2013	–						**2013**	–					
2014	$p < 0.05$	–					**2014**	0.237	–				
2015	1.000	$p < 0.05$	–				**2015**	0.581	$p < 0.05$	–			
2016	$p < 0.05$	1.000	$p < 0.05$	–			**2016**	$p < 0.05$	0.084	$p < 0.05$	–		
2017	$p < 0.05$	0.192	$p < 0.05$	$p < 0.05$	–		**2017**	$p < 0.05$	$p < 0.05$	$p < 0.05$	0.269	–	
2018	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	0.349	–	**2018**	$p < 0.05$	0.254	$p < 0.05$	0.998	0.952	–
Xylene							**TVOC**						
2013	–						**2013**	–					
2014	0.739	–					**2014**	0.981	–				
2015	$p < 0.05$	$p < 0.05$	–				**2015**	0.999	0.831	–			
2016	0.920	0.058	$p < 0.05$	–			**2016**	$p < 0.05$	$p < 0.05$	$p < 0.05$	–		
2017	$p < 0.05$	$p < 0.05$	$p < 0.05$	0.134	–		**2017**	1.000	0.986	0.991	$p < 0.05$	–	
2018	0.306	$p < 0.05$	$p < 0.05$	0.752	1.000	–	**2018**	0.757	0.359	0.828	$p < 0.05$	0.602	–

$p < 0.05$ indicates significant difference.

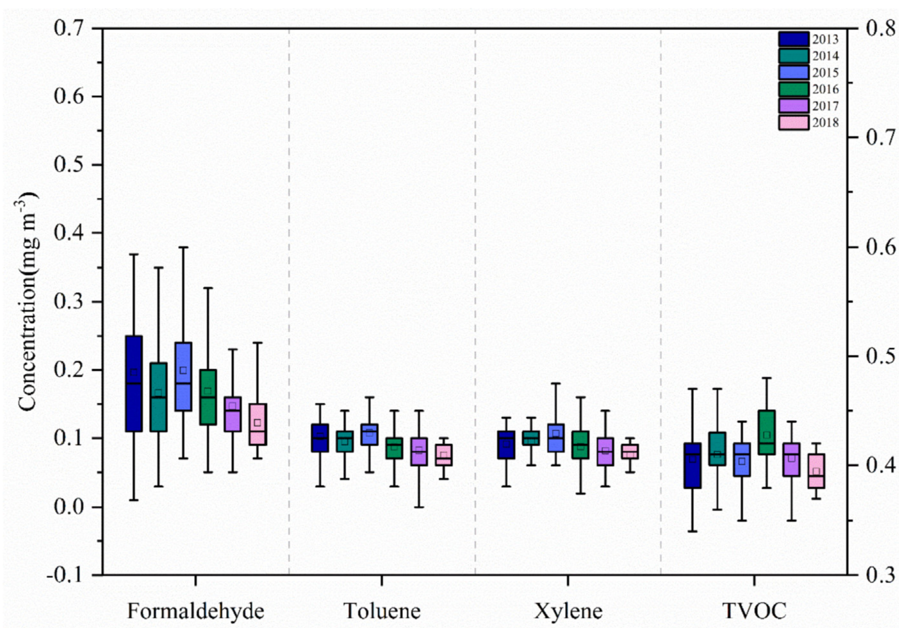

Fig. 1. Concentrations of formaldehyde, toluene, xylene and total volatile organic compounds (TVOC) in household from 2013 to 2018. Boxplots are defined as follows: center line, median; boxplot edges, 25th and 75th percentile; whiskers, 5th and 95th percentile of distribution.

again emphasizing that formaldehyde is the major indoor air pollutant in Harbin. This is likely due to materials containing formaldehyde was used for interior decoration. Often times, so-called environmentally friendly decoration materials are not free of formaldehyde. Relevant government departments should introduce stricter regulations to improve indoor air quality.

3.4. Factors that influence indoor formaldehyde concentration

It is well known that the concentration of formaldehyde is related to the types of pollution sources (decoration materials and furniture quantity, etc.) and environmental factors (temperature, humidity, etc.) (Gunschera et al., 2013; Salthammer et al., 1995; Zhang et al., 2007), these factors affected indoor concentration of formaldehyde altogether. The present study would analyze the impact on indoor formaldehyde concentration from four perspectives: indoor temperature, relative humidity (RH), finishing time of decoration (month) (FTD) and furniture density (FD). Harbin is a typical northern city. Its heating period will be half a year. In the non-heating period (a period of time spring and late autumn), indoor temperature is very low, and the lowest temperature can reach 15 °C. Due to the short summer time in Harbin, most households do not have air conditioning, resulting in indoor temperature as high as 37 °C.

Indoor temperature, RH and FTD were measured during the survey to explore their influence on formaldehyde concentration. The indoor temperature ranged from 14.9 to 35.5 °C, the indoor RH range from 26.1 to 85.4% and the FTD range from 1 to 37 months. The regression fitting curves between indoor temperature, RH, FTD and concentration of formaldehyde were displayed in Fig. 2. Significant positive nonlinear correlations were found in indoor temperature and formaldehyde concentration ($R^2 = 0.54$, $p < 0.01$), reflecting the ability of indoor elevated temperature to increase indoor exposure to formaldehyde. The similar results were achieved in the literature (Salthammer et al., 1995; Tao et al., 2015). Good negative nonlinear correlations between the FTD and formaldehyde concentration ($R^2 = -0.28$, $p < 0.01$), demonstrating that the formaldehyde concentration will gradually dissipate from the pollution sources such as furniture and decoration materials with the increase of finishing time of decoration. Which are consist with

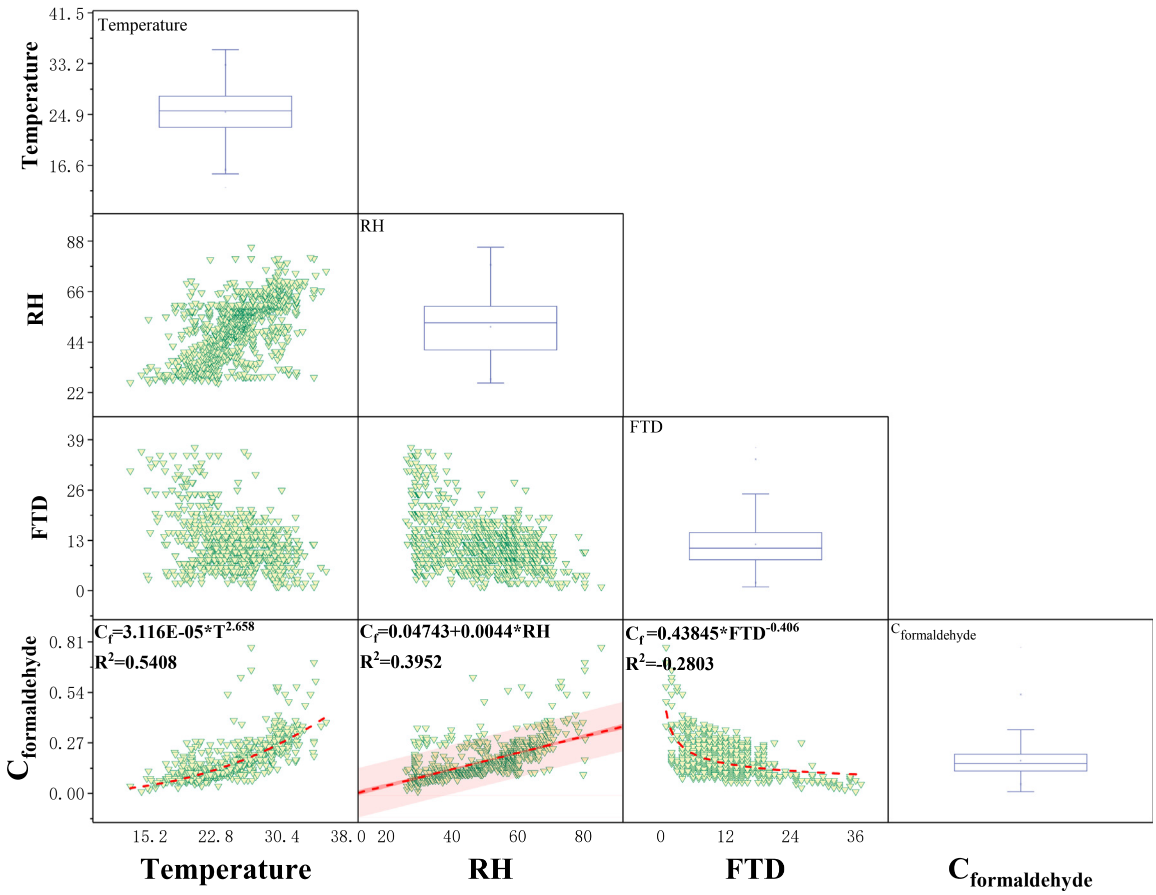

Fig. 2. The regression fitting curves between indoor temperature, RH, FTD and formaldehyde concentration.

Zhai et al. (2010), who found that the formaldehyde concentration slightly decreased with the extension of renovation completion time, the concentration of formaldehyde and over-standard rate in 1–3 years were significantly lower than 3–6 months ($p < 0.05$). Good positive linear correlations between the RH and concentration of formaldehyde ($R^2 = 0.40$, $p < 0.01$), indicating that indoor relative humidity is an important factors in stimulating the emission rate of formaldehyde. Formaldehyde is a compound that dissolves easily in water, higher relative humidity will increase the concentration of formaldehyde. It can be found that the furniture density can affect the concentration of formaldehyde too ($p < 0.01$). Urea-formaldehyde (UF) resin is the adhesive used in wood-based panels for furniture. Hun et al. (2010) found that pressed wood materials with UF resins are probably to be the dominant source of indoor pollutants. The paint on the furniture surface contains either formaldehyde or other volatile organic compounds. Temperature is the main factor affecting the concentration of formaldehyde in the room, but the influence of humidity, decoration time and furniture density on formaldehyde concentration cannot be ignored. High temperature in the indoor environment will increase the release rate of formaldehyde from decoration materials, furniture and other things, so as to RH. Therefore, controlling the indoor temperature and relative humidity well be an effective way to reduce the concentration of formaldehyde. Simultaneously, many studies have shown that air exchange was particularly notable for indoor air quality (Gilbert et al., 2008; Salthammer et al., 1995). They found that ventilation effectively decreases formaldehyde concentration. Consequently, whenever possible, choose as few pieces of furniture as possible or choose furniture made from environmentally friendly materials. It is likewise a good choice to move in the house after a long period of renovation.

3.5. Human health risk assessment

To assess the health risks caused by FTX, a comprehensive exposure risk assessment for different indoor microenvironments and populations were conducted. The results of the calculations of the cancer risk and the hazard quotients were presented in Figs. 3 and 4.

3.5.1. Carcinogenic risk assessment of formaldehyde

Depending on the USEPA, the carcinogenic risk predicted to be less than or equal to 1.0E-6 is usually considered negligible. On the other hand, the risk expected to be greater than or equal to1.0E-3 was defined as a significant risk (Rodrics et al., 1987). Formaldehyde is a suspected carcinogen and its carcinogenic risk values in home, office and school are illustrated in Fig. 3. The median carcinogenic risk values of formaldehyde for populations from home were ranging from 9.75E-4 to 1.41E-3 (Fig. 3a), which were one and/or two orders of magnitude higher than those in office and school (from 8.27E-5 to 1.13E-4 and from 7.57E-6 to 2.08E-5 for office and school respectively) (Fig. 3b and c). It can be concluded that, within a certain group of the population, the carcinogenic risk in the home was significantly higher than in other indoor environments, since people spend most of their time in home. It is worth noting that the carcinogenic risk of formaldehyde in household of Harbin is considerably higher than countries. In Tehran, Iran, the minimum-maximum cancer risk of formaldehyde

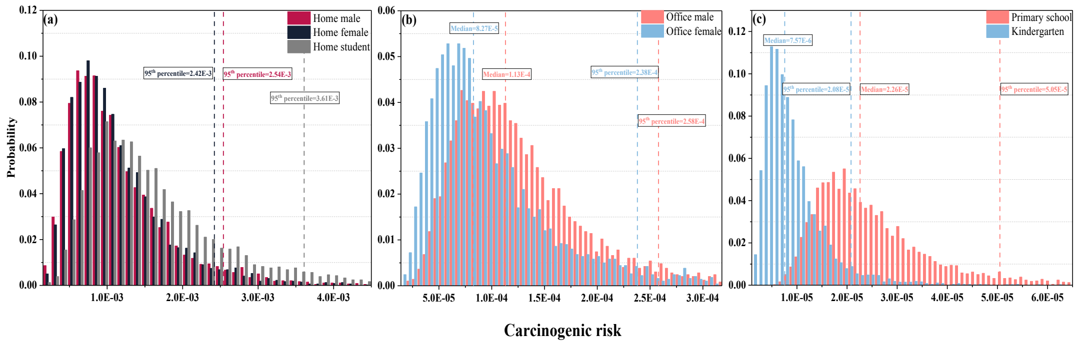

Fig. 3. Predicted probability density functions of carcinogenic risk of formaldehyde at (a) home, (b) office and (c) school. (The lines represent the median and 95th percentile values).

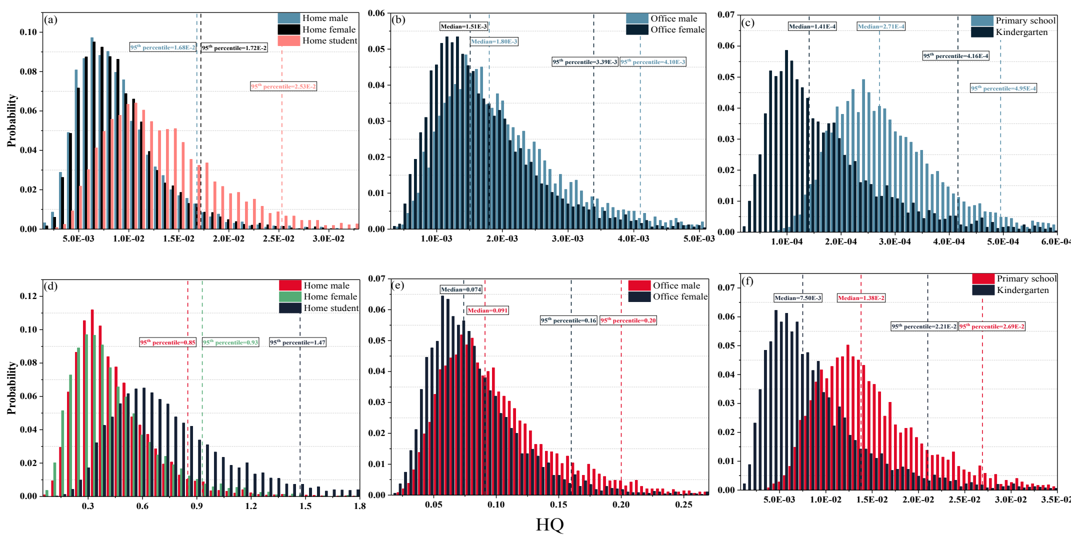

Fig. 4. Predicted probability density functions of HQ of toluene at (a) home, (b) office and (c) school and xylene at (d) home, (e) office and (f) school. (The lines represent the median and 95th percentile values).

was forecast to be 2.05E-5 to 1.24E-3 (Hadei et al., 2018). The carcinogenic risk value in the United States nearly 1E-4 (Loh et al., 2007), which were almost one order of magnitude lower than that in Harbin. On the other hand, compared to other cities, the values of carcinogenic risk in office and school are lower or the same level. In Taiwan, the carcinogenic effect of formaldehyde in the office is ranging from 2.06E-4 to 1.75E-3 (Wu et al., 2003). The health risk assessment of formaldehyde in three primary schools was conducted in Izmir, Turkey, the result showed that approximately half of the population has a carcinogenic risk great than 1.0E-5 during the kindergarten plus primary school years (Sofuoglu et al., 2011). Use of 95th percentile upper confidence limit of carcinogenic risk is emphasized by the current risk assessment guidance (USEPA, 2013). The 95th percentile of risk provides a worst-case estimate of risk and can provide a useful upper-bound estimate of potential risk. In the present study, the 95th percentile of carcinogenic risk was estimated to be ranging from 2.26E-5 to 3.31E-3 in different environments (Fig. 3a, b, c), the risk is substantially greater than the 1E-6 benchmark, indicating that the selected population is suffering noteworthy formaldehyde cancer risk.

3.5.2. No-carcinogenic risk assessment of TX

To evaluate the non-carcinogenic effects of toluene and xylene, it is generally considered that the risk is negligible if the HQ is less than or equal to one. The non-carcinogenic risks that predicted for TX at the home, office and school are negligible because all of the median HQ values are less than one. The 95th percentile of non-carcinogenic risk for toluene was estimated to be ranging from 4.16E-4 to 2.53E-2 at sampling sites (Fig. 4a, b, c), which are 2–4 orders of magnitude lower than the baseline value (HQ = 1). For xylene, the 95th percentile of non-carcinogenic risks is great than toluene, however, all the values are lower than the baseline except student in the home (the 95th percentile = 1.47), suggesting that young children were more sensitive to xylene exposure than adults. The maximum HQs of TX in the home of in Izmir, Turkey was 1.50 E−2, and 3.17E-1, respectively (Sofuoglu et al., 2011). The HQs of TX in the indoor air of homes at different microenvironments of a terai zone in north India were ranged from 7.5E-2 to 3.3E-1 and 1E-4 to 5.6E-4 (Masih et al., 2017), respectively. Consequently, TX compounds have negligible non-carcinogenic risks in the population.

3.5.3. Sensitivity analysis

A sensitivity analysis was conducted to investigate the contribution of the most important individual factors to the output of the inhalation exposure risk assessment (Fig. 5). This enables us to assess the degree of uncertainty for each input of the entire target output. The factors that could potentially contribute to the variation in output, including concentrations of FTX, inhalation rate (IR), exposure frequency (EF), and body weight (BW). Overall, the concentration of FTX was the strongest factor that influences on all

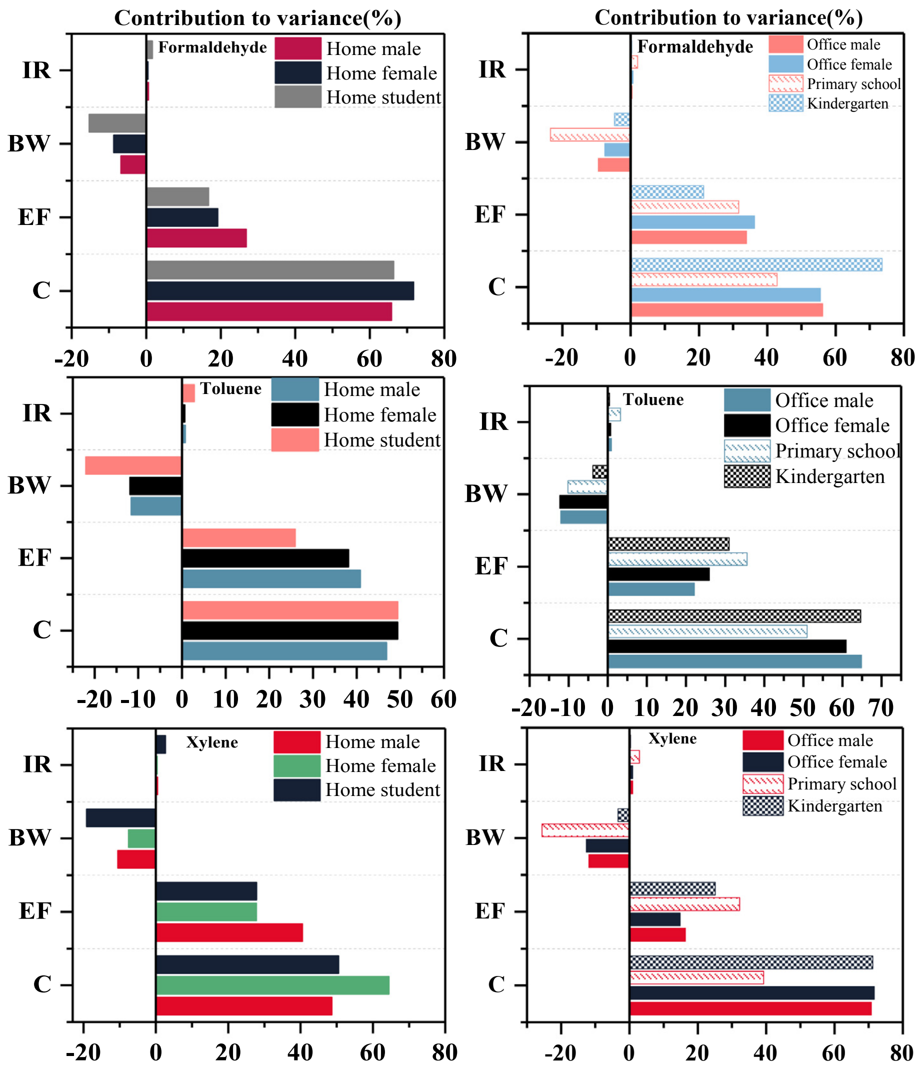

Fig. 5. Sensitivity analysis for the risk simulation.

exposure risk models, the contribution of the variance with a range of 39.26–73.62% for the population in different environment, followed by BW (range from 14.83 to 40.82%). On the other hand, IR contributed to a minor fraction with a range of 0.27–3.24%, while BW provided a negative effect on the risk simulation (range from −23.47 to −3.37).

4. Conclusion

Concentrations of FTX and TVOC in different microenvironments were monitored in Harbin, China. Household had the highest formaldehyde concentration (0.171 ± 0.084 mg m^{-3}) among all the types of tested indoor locations. Concentrations of toluene, xylene and TVOC (0.155 ± 0.007, 0.180 ± 0 and 0.555 ± 0.007 mg m^{-3} respectively) were highest at public bath center. The concentration of formaldehyde had no significant difference between the living room, master bedroom, secondary bedroom and study; meanwhile there were no statistical differences in concentrations of toluene and xylene in those rooms. TVOC concentration did not exceed the national standard in any of the tested locations. Some positive signs that concentrations of formaldehyde, toluene and xylene were gradually decreasing from 2015 to 2018 and concentration of TVOC were decreasing from 2016 to 2018. Despite of the issue with formaldehyde, this study also revealed that temperature, relative humidity, finish time of decoration and furniture density could affect indoor concentration of formaldehyde. Whenever possible, choose as few pieces of furniture as possible or choose furniture made from environmentally friendly materials. It is also a good choice to move in the house after a long period of renovation. Indoor formaldehyde pollution was posing a health risk to the general population in Harbin. On the other hand, toluene and xylene are within the safety threshold. Concentration of FTX and BW contributed most to the output of exposure risk assessment through sensitivity analysis.

Declaration of competing interest

The authors declare no competing financial interest.

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