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Evaluation and characterization of volatile air toxics indoors in a heavy polluted city of northwestern China in wintertime

Yu Huang [a,b,*], Ting Su [a,b], Liqin Wang [a,b], Nan Wang [a,b], Yonggang Xue [a,b], Wanting Dai [a,b], Shun Cheng Lee [c], Junji Cao [a,b], Steven Sai Hang Ho [a,d,**]

[a] Key Laboratory of Aerosol Chemistry & Physics (KLACP), State Key Laboratory of Loess and Quaternary Geology (SKLLQG), Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710061, China

[b] Shaanxi Key Laboratory of Atmospheric and Haze-fog Pollution Prevention, Institute of Earth Environment, Chinese Academy of Sciences, China

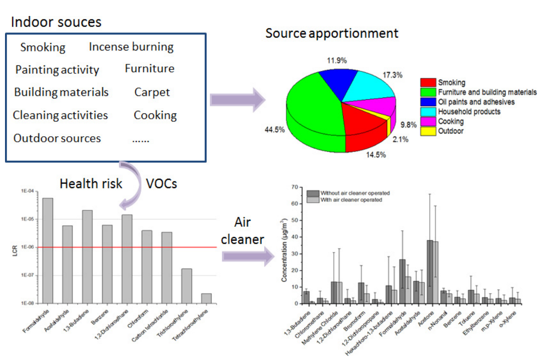
[c] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong

[d] Division of Atmospheric Sciences, Desert Research Institute, Reno, NV 89512, United States

HIGHLIGHTS

- Hazardous VOCs and carbonyls were evaluated in typical dwellings in northwestern China.
- High levels of the pollutants were associated with characteristic pollution sources.
- Paints, adhesives, decoration, and household products are major indoor pollution contributors.
- Cancer risks for formaldehyde, 1,3-butadiene and 1,2-dichloroethane exceeded acceptable level.
- Household air cleaner can be efficiently reduced pollutant levels in residential airs.

GRAPHICAL ABSTRACT



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ABSTRACT

Hazardous volatile organic compounds (VOCs) and carbonyls were evaluated in typical dwellings in Xi'an in northwestern China in wintertime. High indoor concentrations were observed for formaldehyde, acetone, naphthalene, methylene chloride and acetaldehyde, associated with characteristic pollution sources. In comparison, many of the target VOCs were higher in Chinese dwellings than those in other countries, suggesting the significances of indoor pollutions in China. Source apportionment with receptor model shows that furniture and building materials (44.5%), paints and adhesives (11.9%), household products (17.3%), smoking (14.5%), and cooking (9.8%) are the major contributors to the indoor VOCs and carbonyls. The health risk assessment shows that the cancer risks for formaldehyde (5.73×10^{-5}), 1,3-butadiene (2.07×10^{-5}) and 1,2-dichloroethane (1.44×10^{-5}) were much higher than the acceptable level of 1×10^{-6} recommended by International Register for Certified Auditors (IRCA). The hazard quotient (HQ) of target VOCs were far less than the threshold (HQ = 1). Moreover, the practical efficiency of household air purifier in removal of the VOCs and carbonyls was examined first time in dwellings in northern China. The results prove that most of the indoor organic pollutants and their cancer risk to humans can be efficiently reduced, particularly for formaldehyde and 1,3-butadiene. The findings of the

* Correspondence to: Y. Huang, Key Laboratory of Aerosol Chemistry & Physics (KLACP), State Key Laboratory of Loess and Quaternary Geology (SKLLQG), Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710061, China.

** Correspondence to: S.S.H. Ho, Division of Atmospheric Sciences, Desert Research Institute, Reno, NV 89512, United States.

E-mail addresses: huangyu@ieecas.cn (Y. Huang), stevenho@hkpsrl.org (S.S.H. Ho).

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study offer useful preliminary and updated information on current indoor air toxics levels, dominant pollution sources and their potential health risks to residents in northwest China.

1. Introduction

The importance of indoor air quality (IAQ) alerts public in recent decades world-widely. Surveys show that people spend ~80–90% of their daily time indoors on average (Leech et al., 2002). Volatile organic compounds (VOCs) are one of major class of pollutants due to their ubiquity in indoor environments, significantly impacting on human health. Exposure to hazardous volatile air toxics has potential carcinogenic and other toxicological effects, while long-term exposure can harm the respiratory, neurological and reproductive system, or even lead to death (Delfino, 2002; Windham et al., 2006; Wu et al., 2007).

Indoor VOCs and carbonyls can be originated from a variety of sources. Household products were the major contributor (44%), followed by combustion processes and environmental tobacco smoke (ETS) (10.5%), deodorizers (8.4%) and off-gassing of building materials (5.9%) in residences of Edmonton, Alberta (Bari et al., 2015). Seasonal variation on VOCs levels could be seen indoors. Higher indoor levels of alkanes, alkenes, aromatics were reported due to low ventilation rates in heating period (Duan et al., 2014). Pekey and Arslanbaş (2008) also found that most quantified VOCs had higher concentrations in winter than summer in Turkey. The winter values could be even double of those in summer in Edmonton, Canada (Bari et al., 2015). The indoor air can be greatly impacted by coal combustion and biomass burning when household warming is required (Abeleira and Farmer, 2017; Duan et al., 2014). However, in winter, indoor and outdoor air exchange efficiency is much poorer than other seasons, leading to the accumulation of pollutants indoors.

A lot of studies have been conducted to screen and measure those priority toxic VOCs as well as to assess their health-related potentials indoors in China. Duan et al. (2014) quantified nearly one hundred VOCs to obtain the seasonal variations, indoor and outdoor relationships, and potential sources at residential units in Beijing, China (Duan et al., 2014). The results showed that formaldehyde, acetone, acetaldehyde, toluene, ethane and propane were the most dominant indoor airborne organic species. Wang et al. (2007) measured carbonyls simultaneously in twelve urban dwellings in Chinese megacities including Beijing, Shanghai, Guangzhou, and Xi'an (Wang et al., 2007). Formaldehyde was the most abundant compound, accounting for ~46.0% of the quantified carbonyls and ranging from the lowest of 19.3 $\mu g/m^3$ in Xi'an to the highest of 92.8 $\mu g/m^3$ in Beijing during summer.

Higher indoor VOCs and carbonyls levels are always seen in China than other countries. Their concentrations and composition can be varied by interior decorations, activities, ventilations and locations. Guo et al. (2009) conducted a comprehensive study at 100 homes in Hong Kong, reporting that the total VOC and formaldehyde concentration was 46.1 ± 8.8 $\mu g/m^3$ and 112.3 ± 9.5 $\mu g/m^3$, respectively, much higher than other East Asian cities (Guo et al., 2009). Furthermore, higher levels of 1,2,4-trimethylbenzene, styrene, nonane and heptane were found in gas-use families rather than in electricity-use homes in their study.

Due to the importance of VOCs, Du et al. (2014) accessed sixteen highly prevalent Hazardous Air Pollutants (HAPs) in urban cities in China and reported the average total lifetime cancer risks attributable to HAPs are 2.27×10^{-4} and 2.93×10^{-4} for Chinese females and males, respectively. Over 70% of the risk was found due to exposure to indoor air at home and formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene are the major contributors to health hazard.

Xi'an (33°N and 107°E) is a key city in the northwest China and the capital of Shaanxi province. With the supports by the national policies, it has been rapidly developing since 1980s. The growth economy elevates the living standard and also alerts residents to concern their health regarding air pollutions. To our best knowledge, there is still a lack of comprehensive study to evaluate both VOCs and carbonyls in dwellings in the northwest China. The objectives of this work are to compare indoor and outdoor VOC levels, to explore the potential effects of VOC levels indoor and to quantify exposure risks. This study was designed to cover as many compounds as possible under the premise of experimental condition, because of the lack of VOCs data in Xi'an residence.

2. Methodology

2.1. Sampling locations

Eleven dwellings in Yanta, Weiyang, Xincheng and Yanliang districts were selected in this study (Fig. 1). The locations represent typical residential areas in urban and suburban Xi'an where the residents are concentrated. The sampling campaign was conducted from mid-November 2016 to mid-February 2017 during the regular regional heating supply period. The average ambient temperature was -1 ± 5 °C. All selected dwellings have not been renovated in the past three years. No particular pollution sources (e.g., industrial sector or power plant) were near the sampling areas.

2.2. Sample collection

Indoor and outdoor samples were collected simultaneously. For indoor, samplers were placed in the center of living room with an inlet height of 1.5 m above the floor. The living room is the center of the room and the place where people undertakes most activities. All doors and windows were closed when the sampling conducted. The sampling time was between 09:00 and 11:00 when the impact from household cooking was minimized. Additional comparison tests were carried out indoors on the days when an in-house air purifier was operating in each dwelling. The air purifier had worked for 9 h before the first sample was collected. It has operated continuously for four consecutive days (96 h) while the samples were collected daily. Other sampling conditions were the same as before. Two sets of four indoor samples were collected in each dwelling when the air purifier was on or off respectively. Outdoor samples were collected spontaneously on the balcony by extending the sampling tubes outside when indoor sampling was conducted.

A total of sixty-five VOCs classified as “Air Toxics” by United States Environmental Protection Department (VOC_{Toxic}) (USEPA, 1999b) and seventeen carbonyls (including mono- and di-carbonyls) were quantified in this study. The VOC_{Toxic} was collected into a stainless-steel multi-bed adsorbent tube filled with Tenax-TA, Carbograph I TD and Carboxen 1003 (C3-DXXX-5266, ¼″ o.d., Markes International Ltd., Llantrisant, U.K.) using a low-flow module pump (ACTI-VOC, Markes International Ltd.). The sampling flow rate was 50 mL/min and each sample was collected for 120 min. Prior to the sampling, the sorbent tubes were thermally cleaned in a conditioner (TC20, Markes International Ltd.) at 330 °C for 20 min. The pre-conditioned and sampled tubes were sealed with Difok caps (Markes International Ltd.) and stored in pollutant-free desiccators at −4 °C for a maximum of 14 days. The pump was calibrated with a mass flow calibrator (Defender 510, Bios, Torrance, CA, USA) before and after each sampling event. A Teflon filter assembly (47 mm, Whatman, Clifton, NJ, USA) and coiled potassium iodide (KI)-coated copper tubing (¼″ o.d., 1 m in length) were installed in

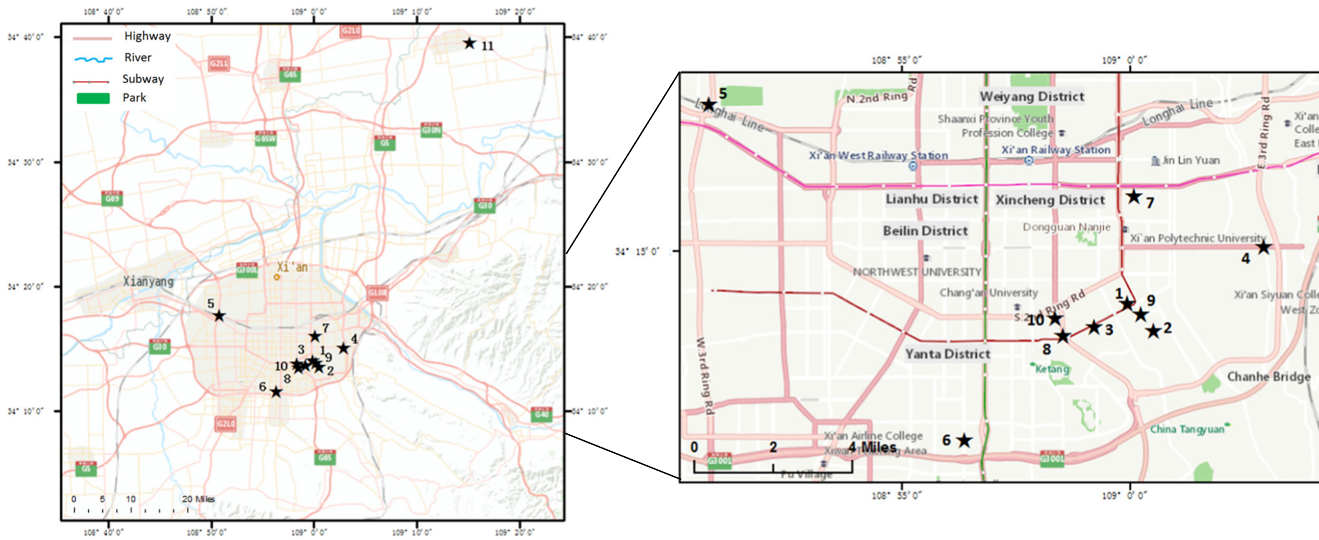

Fig. 1. Map shown dwellings in districts in Xi'an.

upstream to remove particle and ozone (O_3) influences, respectively (Ho et al., 2017, 2018).

The carbonyls were collected into silica cartridges impregnated with acidified 2,4-dinitrophenylhydrazine (DNPH) (Sep-Pak DNPH-silica, 55–105 μm particle size, 125 Å pore size; Waters Corporation, Milford, MA) at a flow rate of 0.6 L/min for 120 min (USEPA, 1999a). Detailed sampling procedures were shown in our previous publications (Spaulding et al., 1999; Ho et al., 2011).

An absorbent tube and a cartridge were reserved to serve as field blanks on each sampling trip and were handled in the same way as the samples. The amounts of target compounds were corrected for the field blank. All samples were shipped and stored in a refrigerator at <4 °C until the chemical analyses.

2.3. Analytical methods

The absorbent tubes for collection of VOC_{Toxic} were analyzed using a thermal desorption (TD) unit (Series 2 UNITY-xr system, Markes International Ltd.) coupled with a gas chromatograph/mass spectrometric detector (GC/MSD, Models 7890A/5977 B, Agilent, Santa Clara, CA, USA). A tube was connected into the TD unit at room temperature (~25 °C) and purged with ultra-high purity (UHP) helium (He) gas at a flow rate of 40 mL/min for 10 s to eliminate air and oxygen intrusion. For the primary desorption stage, the analytes were desorbed at 330 °C for 5 min and refocused onto a cryogenic-trap (U-T1703P-2S, Markes International Ltd.) to capture high volatility target compounds at 15 °C. For the secondary desorption stage, the trap was dry-purged for 10 s and rapidly heated from 15 °C to 320 °C and maintained for 5 min. The analytes were passed via a heated transfer line at 160 °C, and re-refocused onto a cold GC capillary column head (Rtx®-1, 105 m0.25 mm × 1 μm film thickness, Restek Corporation, Bellefonte, PA, USA) at −45 °C with an aid of liquid nitrogen (N_2) in GC oven. Once the second desorption is completed, the oven temperature program started at an initial temperature of −45 °C for 4 min, ramped to 230 °C at a rate of 6 °C/min, and maintained at 230 °C for 5 min. The constant flow rate of helium carrier gas was 1.0 mL/min throughout the GC analysis. The MSD was operated in selective ion monitoring (SIM) mode at 230 °C and 70 eV for electron ionization. Identification was achieved by comparing the mass spectra and retention times of the chromatographic peaks with those of authentic standards. Certified Air Toxics standard mixtures (Restek Corporation) were used in calibrations. A multi-point calibration curve was established to quantify each of the target compounds with linearity > 0.995. The minimum detection limits (MDL) were in the range of 0.1–0.158 ppbv with a sampling volume of 6 L. The measurement precision for the analysis of eight replicates of standard samples at 2 ppbv were <25%.

The carbonyls in DNPH-silica were eluted with acetone-free acetonitrile (ACN) and the extract was injected into a high-pressure liquid chromatography (HPLC) system (1200; Agilent Technology) equipped with a photodiode array detector (DAD). Details on extraction, calibration, and chromatographic conditions were shown elsewhere (Dai et al., 2012). The limit of detections (LOD) of the target carbonyls ranged from 0.002 to 0.010 μg/mL.

2.4. Questionnaire

Information of selected dwellings characteristics and potential sources for VOCs and carbonyls were obtained from site investigation and self-administered questionnaire (Table S1). It included details of room description (i.e., area, age, type of wall, refurbishment, and pet), ventilation and heating systems, frequency and fuel of cooking, smoking activities, cleaning activities (detergent and frequency). The occupants were further interviewed on their other daily activities to identify any additional potential exposure to the target compounds as show in Table 1.

2.5. Positive matrix factorization (PMF) receptor model

Positive matrix factorization (PMF) (USEPA, PMF3.0) receptor model was applied to distinguish dominant sources in the indoor environments (Anderson et al., 2002). The PMF model can be expressed as a chemical mass balance equation in terms of contributions from *p* independent sources to *n* chemical species measured in a given sample (Miller et al., 1972):

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{1}$$

where x_{ij} is the jth chemical species concentration determined in the ith sample, g_{ik} is the species contribution of the kth source to the ith sample, f_{kj} is the loading of jth species on the kth factor, e_{ij} is the residual resulting from bias in the measurement of g_{ik} and f_{kj}, and *p* represent the total number of independent sources (Paatero, 1997). Every data point can be individually weighed in PMF, so that the retainment of data below detection limit with its associated uncertainty was permissible. The stability of the solution can be evaluated by means of examining

Table 1
Statistical data of the general information and activities in the sampled dwellings obtained from the questionnaires.

Site#	Floor#	No of rooms	No of smokers[a]	Ventilation time per day	Cooking per week	Cleaning per week	Incense burning[a]	Insecticide[a]	Types of household chemicals consumed[a]	Fuel
1	26	4	1	<1 h	3–4	2–3	–	+	Laundry, dishwashing, and toilet detergent	LPG
2	6	4	–[b]	1-3 h	7	1–2	–	–	Laundry, dishwashing, and toilet detergent	Natural gas
3	18	5	–	<1 h	3–4	7	–	–	Laundry, dishwashing, and toilet detergent	LPG
4	23	6	1	<1 h	7	7	–	–	Laundry, dishwashing, and toilet detergent	Natural gas
5	4	5	–	<1 h	7	7	–	–	Laundry, dishwashing, and toilet detergent	Electricity
6	23	4	–	<1 h	7	7	–	+	Dishwashing detergent	Electricity, LPG
7	5	4	–	9–12 h	–	7	–	–	Laundry and dishwashing detergent	–
8	2	2	–	1–3 h	7	4–5	–	–	Laundry and dishwashing detergent	Electricity, natural gas
9	8	3	1	1–3 h	1–2	2–3	–	–	Dishwashing and toilet detergent	LPG
10	2	4	–	3–6 h	7	2–3	–	–	Laundry, dishwashing, and toilet detergent, bleach	Electricity, LPG
11	5	4	2	1–3 h	7	7	+	–	Laundry and dishwashing detergent	Electricity, natural gas

[a] No record on the quantity consumed daily due to limitation.
[b] No activity conducted in the dwelling.

the proportion of each source undertaken in terms of the object function Q:

$$Q = \sum_{t=1}^{n}\sum_{i=1}^{m}\left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{\mu_{ij}}\right]^2 \quad (2)$$

where μ_{ij} represents the uncertainty of jth species in ith sample. For the PMF input, the uncertainty caused by sampling and analytical errors was calculated using the following equation suggested by Polissar et al. (1998):

$$U = \sqrt{(EF \times conc)^2 + (MDL)^2}(conc > MDL) \quad (3)$$

where EF represent the error fraction, which is the result of the relative standard deviations of the instrument multiply 100, and we set it as 0.10, equal to the average percent uncertainty in our study. For values below detection limit, the uncertainties were replaced by 5/6 times of the detection limit values. Any missing data is replaced with the median concentration of that species and the uncertainty are expressed as four times the median concentration (USEPA, 2008). For the selection of chemical species, typical tracers of different sources and those in high indoor concentrations were taken into account in the receptor modeling. In addition, those species with >50% of samples below LODs were screened out.

2.6. Cancer and non-cancer risk calculation model

The risk characterization for indoor VOC inhalation exposure was conducted by combining published toxicity data with the exposure concentrations estimated in this study. To calculate inhalation risks, an adjusted air concentration (EC_i) was calculated using the following equation according to United States Environmental Protection Agency (USEPA) Superfund program (USEPA, 2009; Office of Solid Waste and Emergency Response, 1991).

$$EC_i = \frac{CA_i \times ET \times EF \times ED}{AT} \quad (4)$$

where EC_i is the exposure concentration; CA_i is the measured VOC concentration in the residences (μg/m^3); ET is exposure time (h/day); EF is the exposure frequency (days/year); ED is exposure duration (years); AT is averaging time (h). For cancer and chronic hazard assessments, lifetime (70 years) is substituted for AT (lifetime in years × 365 days/year × 24 h/day).

We adjusted exposure air concentration by incorporating time-activity data of Chinese residents. Based on Exposure Factors Handbook of Chinese Population (Duan, 2013) and Wang et al.'s (2012) study, average exposure time was estimated as 15 h/day for residents (Dai et al., 2017). Exposure frequency was estimated as 350 days/year, exposure duration was estimated as 24 years for adults to calculate inhalation cancer risk attributable to indoor VOCs. The indoor inhalation cancer risk at residences was calculated with the methodology proposed by USEPA (2004).

$$LCR_i = IUR_i \times EC_i \quad (5)$$

where LCR_i is the cancer risk associated with compound i; EC_i the daily average inhaled concentration of compound i; and IUR_i is the estimated inhalation unit risk (m^3/μg) for compound i from USEPA, IRIS (Integrated Risk Information System) or OEHHA (Office of Environment Health Hazard Assessment), which is the excess lifetime cancer risk estimated to result from continuous exposure to an individual VOC via inhalation per μg/m^3.

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

$$HQ_i = \frac{EC_i}{RfC_i} \quad (6)$$

where HQ_i is the hazard quotient for compound i; EC_i is the modeled personal exposure to compound i; and RfC_i is the reference exposure limits for compound i.

The results of LCR_i and HQ_i were judged according to USEPA's (2004) approach. Namely, cancer risks no higher than 1×10^{-6} for an "ample margin of safety" and an HQ value of one or less indicates that adverse health effects are not expected to result from exposure to this VOC.

3. Results and discussion

3.1. Characteristics of VOCs and carbonyls

3.1.1. Indoor and outdoor levels of VOC_{Toxic} and carbonyls

The average concentrations of VOC_{Toxic} and quantified carbonyls indoors and outdoors are listed in Table 2. For the indoors, acetone (35.55 ± 24.34 μg/m^3) was the most dominant species, followed by formaldehyde (21.45 ± 13.72 μg/m^3), naphthalene (16.64 ± 18.96 μg/m^3), methylene chloride (13.13 ± 18.64 μg/m^3), acetaldehyde (12.92 ± 6.36 μg/m^3), hexachloro-1,3-butadiene (9.41 ± 15.78 μg/m^3), bromoform (9.21 ± 8.16 μg/m^3), toluene (7.23 ± 7.00 μg/m^3), n-nonanal (6.86 ± 2.02 μg/m^3), methyl butyl ketone

Table 2
Concentration of different categories of carbonyls and VOCs in indoor and outdoor (μg/m^3).

Compounds (μg/m^3)	Indoor (n = 44)		Outdoor (n = 37)	
	Mean	SD	Mean	SD
Carbon disulfide	0.63	0.69	1.24	2.41
Alkane				
n-Hexane	1.76	1.65	1.83	2.12
n-Heptane	0.68	0.86	0.4	0.35
Cyclohexane	0.51	0.75	0.26	0.23
Alkene				
1,3-Butadiene	3.36	1.16	3.89	0.64
Propylene	2.91	3.38	4.54	6.28
Carbonyls				
Acetone	35.55	24.34	26.92	28.09
Formaldehyde	21.45	13.72	8.53	7.94
Acetaldehyde	12.92	6.36	7.33	4.54
n-Nonanal	6.86	2.02	4.9	2.84
Methyl butyl ketone	5.45	8.98	10.8	36.77
n-Octanal	4.68	2.1	3.6	1.87
n-Decanal	4.05	1.9	2.77	1.44
Methyl ethyl ketone	3.56	1.81	3.01	1.53
Hexanal	3.05	1.66	1.23	1.09
n-Heptanal	2.49	0.88	1.96	0.72
iso-Pentanal	1.78	1.25	1	0.54
Methylglyoxal	1.72	0.92	2.16	1.57
Glyoxal	1.56	0.81	2.49	1.44
Propanal	1.54	0.66	1.35	0.7
iso- + *n*-butanal	1.52	0.76	1.17	0.59
Acrolein	1.49	1.14	3.1	3.35
Benzaldehyde	1.24	0.68	0.9	0.38
n-Pentanal	0.92	0.5	0.53	0.23
2,5-Dimethylbenzaldehyde	0.9	0.42	0.87	0.46
Methyl isobutyl ketone	0.83	0.76	1.51	1.43
o-Tolualdehyde	0.49	0.21	0.43	0.21
p-Tolualdehyde	0.43	0.19	0.43	0.19
m-Tolualdehyde	0.31	0.05	bd	–
Others				
Ethyl acetate	4.59	3.67	3.53	2.85
Isopropyl alcohol	1.17	1.05	1.71	1.71
Vinyl acetate	0.92	1.86	0.55	1.67
Tetrahydrofuran	0.91	1.18	0.43	0.59
Methyl-*tert*-butyl ether	0.61	0.56	0.55	0.52
Methyl methacrylate	0.15	0.12	0.22	0.42
Freon				
Freon-11	1.67	2.15	0.97	1.8
Freon-12	1.18	1.19	0.65	0.63
Freon-113	0.39	0.35	0.21	0.21
Freon-114	0.01	0	0.07	0.03
Halohydrocarbon				
Methylene chloride	13.13	18.64	18.39	30.48
Hexachloro-1,3-butadiene	9.41	15.78	0.62	1.74
Bromoform	9.21	8.16	2.39	2.65
Carbon tetrachloride	2.81	5.84	2.12	8.01
1,2-Dichloroethane	2.69	4.12	1.78	3.08
Chloromethane	2.64	3.31	6.46	14.63
1,2-Dichloropropane	1.93	2.98	1.86	3.35
cis-1,2-Dichloroethene	1.32	0.8	0.53	0.32
Chloroethane	1.08	1.07	0.89	0.9
1,4-Dichlorobenzene	0.99	1.15	0.16	0.21
Benzyl chloride	0.98	1.95	0.23	0.53
Chloroform	0.84	0.51	0.55	0.31
1,1,2,2-Tetrachloroethane	0.74	1.31	0.31	0.66
1,1,1-Trichloroethane	0.71	0.56	0.32	0.32
1,3-Dichlorobenzene	0.68	0.87	0.14	0.16
1,2-Dichlorobenzene	0.56	1.04	0.06	0.1
1,1-Dichloroethane	0.47	0.44	0.34	0.32
1,2,4-Trichlorobenzene	0.47	0.38	0.03	0.04
Dibromochloromethane	0.45	0.31	0.58	0.17
Trichloroethene	0.42	0.37	0.48	0.51
Tetrachloroethene	0.42	0.32	0.23	0.21
Chlorobenzene	0.39	0.75	0.13	0.13
Bromodichloromethane	0.36	0.5	0.46	0.6
1,1,2-Trichloroethane	0.2	0.1	0.02	–
1,2-Dichlorobenzene	0.18	0.16	0.26	0.28
1,1-Dichloroethene	0.16	0.11	0.06	0.02
trans-1,3-Dichloropropene	0.08	0.06	0.04	0.02
Bromomethane	bd	–	bd	–
trans-1,2-Dichloroethene	bd	–	bd	–
1,4-Dioxane	bd	–	bd	–
cis-1,3-Dichloropropene	bd	–	0.41	0.07
1,2-Dibromoethane	bd	–	bd	–
Aromatic				
Naphthalene	16.64	18.96	5.51	6.49
Toluene	7.23	7	4.22	4.08
Benzene	3.58	3.48	3.01	2.66
Ethylbenzene	3.46	4.21	1.5	1.64
o-Xylene	3.29	5.16	1.47	1.61
m,*p*-Xylene	2.72	4.41	1.22	1.61
Styrene	1.74	1.25	0.99	0.69
1,2,4-Trimethylbenzene	0.87	0.89	0.36	0.39
4-Ethyltoluene	0.37	0.32	0.15	0.15
1,3,5-Trimethylbenzene	0.37	0.35	0.15	0.13

(5.45 ± 8.98 μg/m^3) and ethyl acetate (4.59 ± 3.67 μg/m^3). The concentrations of individual target compounds in each dwelling are shown in supporting information (Table S2). The concentrations are associated with specific situations (e.g., size, design, and ventilation rate) and indoor activities. The common indoor sources are known as paints, adhesives, synthetic fragrances and cigarette smoke (Guo et al., 2003; Polzin et al., 2007).

Owing to the uniqueness of each dwelling (e.g., size, design, and ventilation rate), it is more appropriate to present the proportion instead of absolute concentration (Fig. 2). Acetone had the highest mass proportion of 10–25%. The range is consistent among the dwellings except an extremely high value of 55% at Site 6. Acetone is used as solvent and widely present in many household products (Wang et al., 2007). High proportions of methylene chloride (7–20%) were also found in many dwellings such as Site 2, 4 and 6 where occupants frequently conducted cleaning activities with detergents. Methylene chloride is a propellant to form aerosols while spraying (Agency for Toxic Substances and Disease Registry, 2000). The indoor level of methylene chloride is thus linked with the application of spray products. These could be supported by our results that acetone and methylene chloride were in high proportions at Site 1 and 6 where insecticide had been used (Table 1). The proportion of methylene chloride in Site 1 and 3 were also up to 10% which occupants are not frequently conducted cleaning activities with detergents (1–3 times per week). Methylene chloride is a powerful solvent that often be used as active ingredient in most paint strippers and foaming agent (Riley et al., 2000), so the high proportion of methylene chloride may also from the volatilization of furniture and building materials. Furthermore, high proportion of naphthalene (0–16%) may be related to the use of mothballs but there were difficulties to statistically record their usages in each dwelling in this study (Jo et al., 2008).

Formaldehyde widely presents in paints, adhesives, synthetic fragrances and cigarette smoke (Guo et al., 2003; Polzin et al., 2007). The mass proportion of formaldehyde were not greatly varied (5–15%) but high, implying that there were consistent and rich sources in those dwellings in northern China (Salthammer et al., 2010). Wang et al. (2007) revealed that building materials and some combustion activities including tobacco smoke and incense burning are the contributors for indoor carbonyls.

Moderate compositions of BTEX (i.e., benzene, toluene, ethylbenzene, *m*/*p*-xylene and *o*-xylene) and styrene (1–20% in total) were shown in most dwellings. Their proportions were up to 20% at Sites 1 and 4, where the occupants were smokers (Table 1). It was reported that tobacco smoking could emit different degrees of benzene, toluene and *m*,*p*-xylene (Lee et al., 2002). Besides, BTEX and styrene can be produced in combustion processes, fuel evaporative losses, and uses of solvents (Buczynska et al., 2009; Ilgen et al., 2001). In general, aromatic VOCs were often found higher in China than other countries (Ohura et al., 2009).

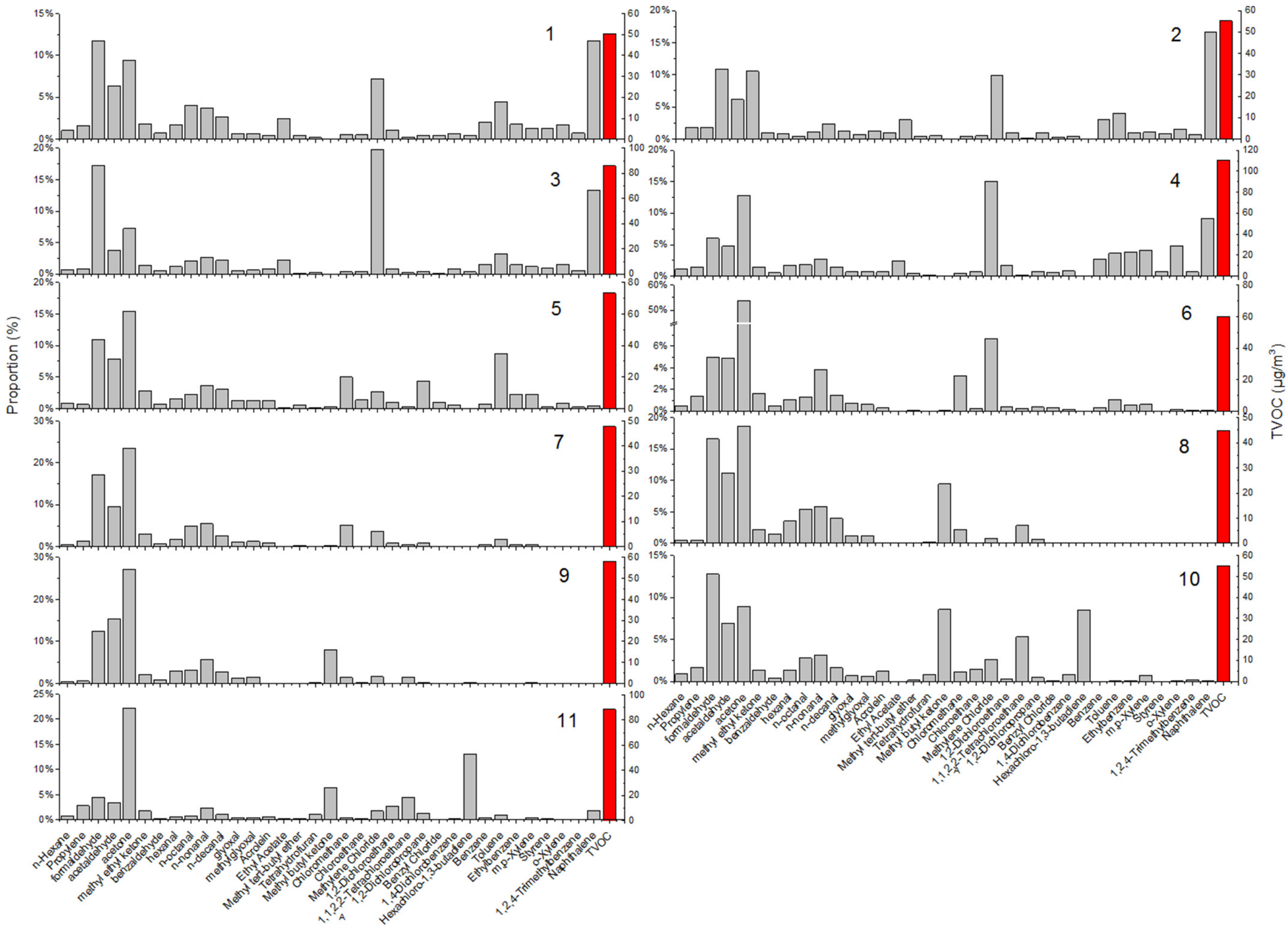

Fig. 2. Indoor mass proportions of typical compounds in each sampling site.

For the outdoors, the highest concentration was also seen for acetone (26.92 ± 28.09 μg/m^3), followed by methylene chloride (18.39 ± 30.48 μg/m^3), methyl butyl ketone (10.80 ± 36.77 μg/m^3). Formaldehyde (8.53 ± 7.94 μg/m^3) and acetaldehyde (7.33 ± 4.54 μg/m^3) were also abundant in ambient air as they can be primarily formed in any combustions (i.e., vehicle emission and industrial activities) and secondarily formed through photochemical reactions (Possanzini et al., 2002). Similar explanation was applied in explanation for high abundances of other compounds such as propylene (4.54 ± 6.28 μg/m^3), toluene (4.22 ± 4.08 μg/m^3), 1,3-butadiene (3.89 ± 0.64 μg/m^3) and benzene (3.01 ± 2.66 μg/m^3) as well (Li et al., 2017; Xue et al., 2017). Liu (2014) evaluated the carbonyls and BTEX levels in indoor air at 128 residential homes in Beijing, China, and found four major pollution contributors including outdoor incursion, building materials and paints, particle board and plywood, and household cleaning chemicals. Considering that the air can be exchanged physically, the indoor levels can be thus impacted by the outdoor sources particularly during the frequent pollution episodes in northern China.

3.1.2. Comparison of indoor levels with other studies

Table 3 compares the average indoor concentrations of selected compounds between our values and the findings in other literatures. The concentration of formaldehyde was higher in our study than the average value in Japan, but ~50% lower than that report in Beijing. Formaldehyde is mainly released to indoor air from refurbishment materials, wood-based products, flooring materials, smoking, and any indoor combustion (Salthammer et al., 2010). The variations of the indoor levels between the cities could be attributed with the differences of materials used and living styles (i.e., smoking, cleaning, cooking). In addition, formaldehyde is the only carbonyl incorporated in the Chinese national indoor air quality standard. It was found that the concentrations of formaldehyde in all dwellings were well below the standard of 100 μg/m^3 on 1-h average (GB/T, 2002). Even though the concentrations in the dwellings were below the standard, the high abundances must be an alarm to the Chinese residents. The level of acetaldehyde in our study was comparative or lower than those in Beijing and Japan, but not for acetone. High abundances of acetone in Xi'an potentially associated with more solvents used such as bleach, laundry detergent, laundry stain remover, floor glue, nail color remover, oil paint, and furniture polish (Kwon et al., 2007). For methylene chloride and chloroform, our values are much lower than the newly renovated apartment in Shanghai and Hong Kong.

For aromatics, the concentrations of benzene, toluene, ethylbenzene, *m*/*p*-xylene and *o*-xylene (BTEX) and styrene in the dwellings in Xi'an were relatively low. This could be explained by less or no smoking and cooking activities had been conducted in the residences during the sampling period. In addition, the concentrations of BTEX were much lower than the newly houses renovated in Shanghai, indicating that less evaporation or releases of solvents from the aged walls and furniture.

3.2. Indoor source identification

3.2.1. Indoor and outdoor ratios

Indoor to outdoor (I/O) ratio is a strong evidence to identify whether indoor or outdoor sources may play decisive roles in IAQ. The I/O ratios

Table 3
Comparison of selected concentrations (μg/m^3) in dwelling with other relevant studies.

Compounds (μg/m^3)	This study	Beijing, China (Duan et al., 2014)	Various cities, Japan (Azuma et al., 2016)	Shanghai, China (Dai et al., 2017)	Hong Kong, China (Lee et al., 2002)	Kocaeli, Turkey (Pekey and Arslanbaş, 2008)
Formaldehyde	21.45 ± 13.72	40.2 ± 26.2	13.00	–	–	–
Acetaldehyde	12.92 ± 636	17.0 ± 10.3	21.10	–	–	–
Acetone	35.55 ± 24.34	23.6 ± 10.7	27.10	–	–	–
Methylene chloride	13.13 ± 18.64	12.5 ± 78.5	–	47.43 ± 75.66	8.8 ± 0.8	–
Chloroform	0.84 ± 0.51	–	1.10	3.59 ± 6.66	2.6 ± 0.9	–
Benzene	3.58 ± 3.48	7.35 ± 11.6	2.40	2.32 ± 1.19	4.7 ± 0.5	13.06
Toluene	7.23 ± 7.00	23.5 ± 45.6	10.80	200.13 ± 443.89	52.1 ± 8.4	72.44
Ethylbenzene	3.46 ± 4.21	3.68 ± 2.49	5.60	26.33 ± 27.73	0.6 ± 0.8	–
m,p-Xylene	2.72 ± 4.41	6.33 ± 4.41	8.30	39.56 ± 49.81	3.9 ± 1.2	27.46
Styrene	1.74 ± 1.25	1.85 ± 2.13	–	32.59 ± 42.77	–	11.65
o-Xylene	3.29 ± 5.16	2.32 ± 1.57	3.40	–	4.5 ± 0.4	16.24
1,2,4-Trimethylbenzene	0.87 ± 0.89	1.99 ± 2.10	6.40	–	–	4.20

of VOC$_{Toxic}$ and carbonyls with air purifier operated and after the use of air purifier in this study were presented in Fig. 3. Higher I/O values (>1) represent dominant indoor sources. Many VOC$_{Toxic}$ and carbonyls were significantly lower outdoors than those indoors, consistent with the findings in other literatures (Bari et al., 2015). Chlorinated compounds such as hexachloro-1,3-butadiene, 1,2,4-trichlorobenzene,1,1,2-trichloroethane and 1,2-dichlorobenzene had the highest I/O ratios, which were 13–17, 12–17, 6–15 respectively. Bleach is reported to be a contributor for indoor chloroform, and 1,2-dichlorobenzene is used to make mothballs and toilet deodorizer blocks (Shepherd et al., 1996). The I/O ratios of other chlorinated compounds were mostly >3, implying that they were originated form indoor sources such as liquid household products (Kwon et al., 2008). Huang et al. (2014) reported that chlorinated compounds often use as industrial solvent such as pharmaceutical solvents, dyes, pesticides, detergents, rubber, water disinfection, and chemical plants (Huang et al., 2014). For carbonyls, formaldehyde and acetaldehyde had the I/O ratios of ~2. Most aromatic compounds (e.g., BTEX) displayed moderate I/O ratios, revealing the contributions from both indoor and outdoor sources (Edwards et al., 2001; Jia et al., 2008; Wang et al., 2007). It should be noted that the ratios of carbon disulfide, *n*-hexane, propylene and 1,3-butadiene were below unity, indicating that these VOCs are primarily form outdoor sources. It was reported that propylene and 1,3-butadiene were the major VOCs of vehicle emission (Li et al., 2017; Xue et al., 2017), explained that the low I/O values were found in our study.

3.2.2. Source apportionment of indoor VOC

Source apportionment was conducted with USEPA PMF receptor model. The concentrations and uncertainties for the VOCs and carbonyls from those valid samples collected in the eleven dwellings were used. Calibration was run for 3–7 factors and with random seeds. We finally compared those examination results and considered that five-factor PMF solution is the best fit for further analysis, when the relevant Q value equals to 5144 in the robust mode. The residuals of the analytical results are mostly between −3.0 and 3.0 (88%). With the adjustment of the number of factors, the calculated results tend to be stable and the final determination of six factors. Some parameters of PMF model when six major factors are selected were shown in Table S3. The r^2 of majority species were >0.6 and the model fitted well. The selected compounds' average concentrations and mass contributions of each source factor are shown in Fig. 4. In Factor 1, toluene, ethylbenzene, benzene, 1,4-dichlorobenzene and styrene had the highest contribution. Lee et al. (2002) reported that tobacco smoking could explain indoor levels of benzene, toluene and *m,p*-xylene. BTEX and styrene are also found in tobacco smoke (Wallace et al., 1987). Source apportionment shows that the dwellings with two smokers (who consumed 6–9 cigarettes per

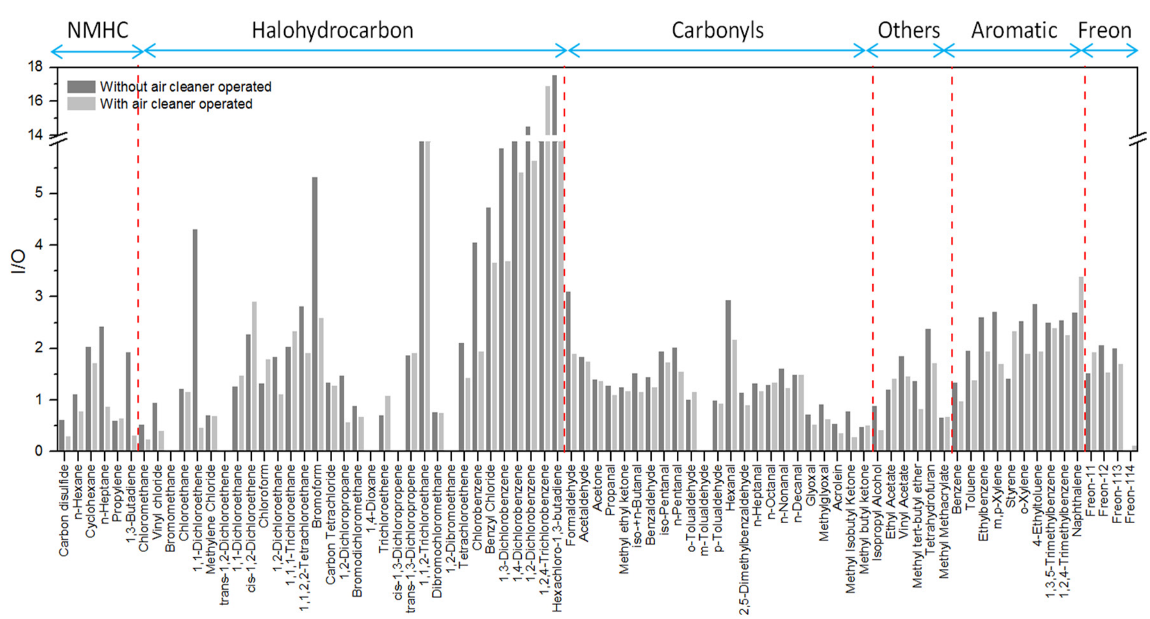

Fig. 3. I/O ratios of VOCs and carbonyls.

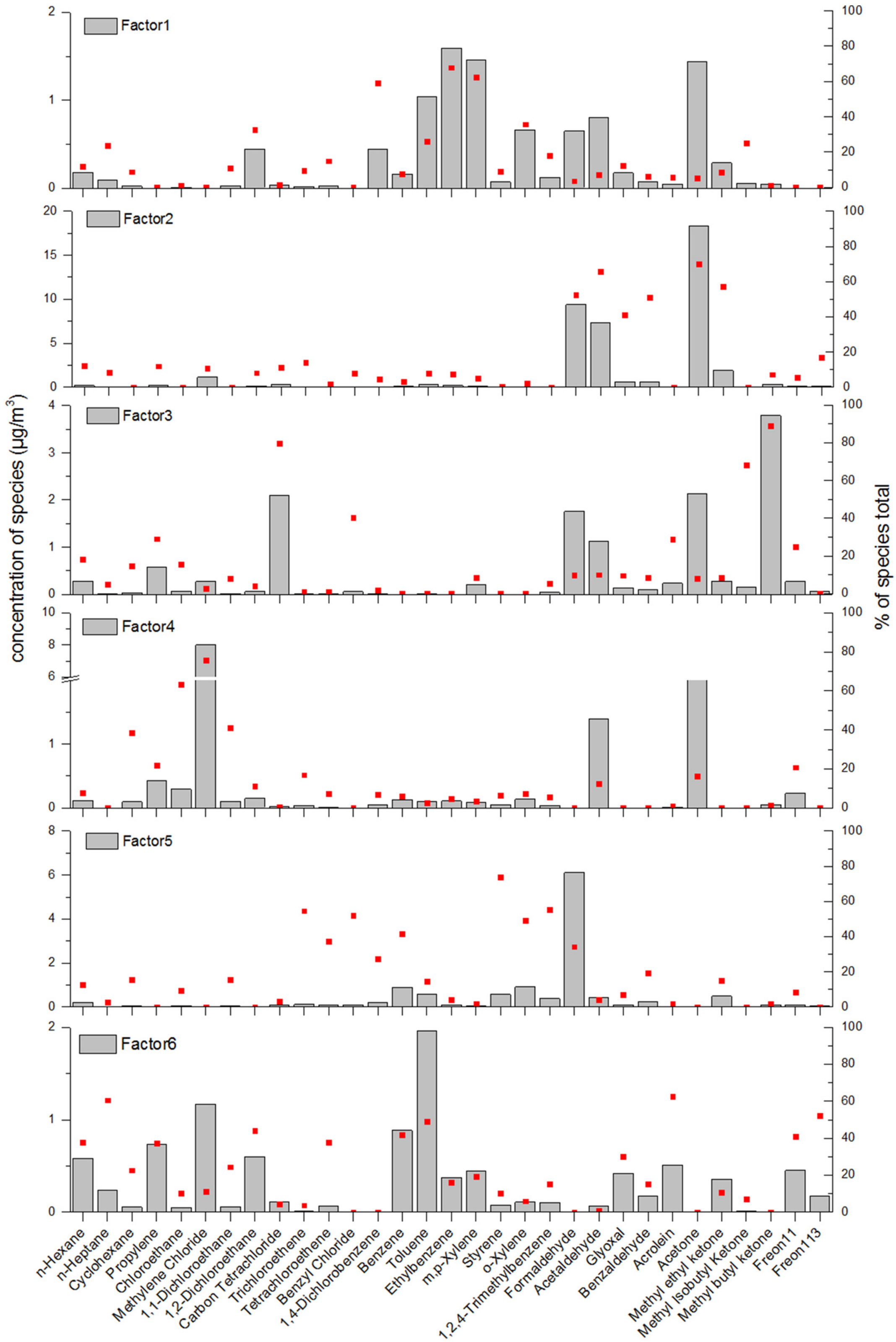

Fig. 4. PMF-resolved indoor VOC source profiles (concentration of species and % of species apportioned to the factor from base run).

day) had the contribution from this factor being up to 90%. This factor was thus identified as smoking, accounting for 14.5% of the total loading (Fig. 5).

Factor 2 should be associated with the off-gases from furniture, floor, building materials and wall coverings. The key characteristic components in this factor were acetone, formaldehyde, and acetaldehyde, also with high contribution of glyoxal, benzaldehyde and methyl isobutyl ketone. Acetone is widely utilized in lacquers for either wooden- or galvanized steel–furniture finishes (WHO, 1998). Hodgson et al. (2002) reported that several cabinetry materials, passage doors, and the plywood subfloor were the predominant sources of formaldehyde and other aldehydes. Wood-based materials used in

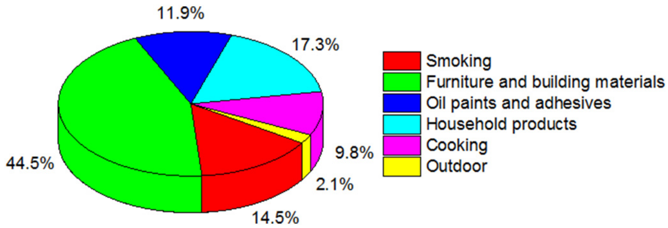

Fig. 5. Source apportionment of indoor VOCs in Xi'an in winter.

construction or in furniture production have long been the typical indoor source (Tunga, 2013). These species can be emitted from indoor decorations such as furniture, floor and wall covering materials including carpet, wallpaper, ceiling tiles, sheetrock, concrete and insulation foam (Wallace et al., 1987; Wilke et al., 2010; Yu and Crump, 1998). This common indoor source had an average loading of 44.5%, apportioned to be the most dominant indoor source.

Factor 3 was filled with methyl butyl ketone (MBK), acetone and 1,2-dichloroethane, together with high contributions of methyl isobutyl ketone (MIBK) and benzyl chloride. These species are often used as solvent in paints and adhesives (Chin et al., 2014; Yuan et al., 2010). Therefore, this factor has been assigned to oil paints and adhesives. Its contribution was 11.9% of total measured VOCs and carbonyls.

Factor 4 was characterized by methylene chloride, acetone, acetaldehyde, with high contribution of chloroethane, 1,1-dichloroethane, cyclohexane. Small amounts of BTEX also contributed to this factor. Previous studies have shown that toluene, xylenes, methylene chloride, acetone, hexane, tetrachloroethylene, 1,1,1-trichloroethane, and trichloroethylene typically exist at high abundances in household products (Sack and Steele, 1992). Formaldehyde, acetaldehyde, acetone can be released from cleaning reagents and floor cleaners (Huang et al., 2011a). Wallace et al. (1987) identified 1,1-dichloroethane and methylene dichloride in cleaning agents, pesticides, wallpaper and carpet glues (Wallace et al., 1987). Kwon et al. (2007) also investigated the emission for household products in Korea and found that acetone, *m*,*p*-xylenes, toluene, ethylbenzene, and hexane were abundant. Acetone was determined in cleaning products, glues, nail color removers, paints, and polishes. In another study, Kwon et al. (2008) also reported that many liquid household products (e.g., deodorizers, cleaners, color removers, pesticides, and polishes) can release several toxic aromatic and chlorinated organics (Kwon et al., 2008). Hence, factor 4 was interpreted as household products and had a contribution of 17.3%.

The major loadings in factor 5 was formaldehyde, other species with high contribution were *o*-xylene, styrene, benzene, 1,2,4-trimethylbenzene, benzyl chloride, trichloroethene, tetrachloroethene. Formaldehyde had the highest concentration in Hong Kong restaurants (Ho et al., 2006). The highest formaldehyde concentration in smoke from frying was also detected (Xin et al., 2016). Formaldehyde is also produced by combustion processes and heating of foods (Tunga, 2013). The concentration of aromatic hydrocarbons such as benzene, toluene and chlorinated hydrocarbons also increased during the cooking periods (Lin et al., 2014; Wang, 2011). Huang et al. (2011a, 2011b) also reported that the significant increase of aromatic was related to evaporative loss of impurities in cooking fuels (Huang et al., 2011b). As a result, this factor is marked as cooking, accounting for only 9.8% of the total VOCs since the impact from household cooking was minimized.

Factor 6 was characterized by toluene, benzene, methylene chloride, propylene, *n*-hexane, *n*-heptane, glyoxal, acrolein and freon-11. This series of compounds are highly correlated with vehicle emission, biomass burning, industrial emission and solvent usage (Li et al., 2017; Xue et al., 2017; Zhang et al., 2012). The I/O value of these species revealed they may also from outdoor. This factor was interpreted as outdoor, accounting for 2.1%.

Table 4
Health-related VOCs and related toxicity values.

Compounds	Cas no.	IARC	IUR $(\mu g/m^3)^{-1}$	RfC (mg/m^3)
Formaldehyde	50-00-0	1	1.3×10^{-5}	–
Acetaldehyde	75-07-0	2B	2.2×10^{-6}	0.009
1,3-Butadiene	106-99-0	1	3×10^{-5}	0.002
Benzene	71-43-2	1	2.2×10^{-6}	0.03
Toluene	108-88-3	3	–	5
m/*p*-Xylene	106-42-3	3	–	0.1
o-Xylene	95-47-6		–	–
Ethylbenzene	100-41-4	2B	–	1
Styrene	100-42-5	2B	–	1
1,4-Dichlorobenzene	106-46-7	2B	–	0.8
Chloromethane	74-87-3	3	–	0.09
Methylene chloride	75-09-2	2A	1×10^{-8}	0.6
1,2-Dichloroethane	107-06-2	2B	2.6×10^{-5}	–
Chloroform	67-66-3	2B	2.3×10^{-5}	–
Carbon tetrachloride	56-23-5	2B	6×10^{-6}	0.1
Trichloroethylene	79-01-6	1	4.1×10^{-6}	0.002
Tetrachloroethylene	127-18-4	2A	2.6×10^{-7}	0.04

3.3. Health risk assessments

Table 4 lists the 17 health-related chemicals catalogued at different groups by IARC (International Agency for Research on Cancer) and with confirmed IUR or RfC inhalation toxicity according to Integrated Risk Information from USEPA's. The inhalation cancer risk or non-cancer hazard risk were calculated based on these parameters.

3.3.1. Cancer risk assessment

The estimated inhalation cancer risks for nine VOCs are shown in Fig. 6. Formaldehyde had the highest cancer risk of 5.73×10^{-5}, followed by 1,3-butadiene (2.07×10^{-5}) and 1,2-dichloroethane (1.44×10^{-5}). They are all higher than the acceptable risk level of 1×10^{-6} but lower than the tolerable risk level of 1×10^{-4}. Formaldehyde and 1,3-butadiene are all classified in group I as a human carcinogen by IARC groups. The major exposure route of formaldehyde is inhalation from indoor air, impacting on nasal and upper airways. Long-term exposure to formaldehyde increases the risk of developing multiple myeloma, myelogenous leukemia and other special cancers. 1,3-butadiene is a characteristic of vehicle exhaust, while 1,2-dichloroethane is often used as a solvent, such as resin, rubber, dry cleaning agent and detergent. Therefore, the best health gains can

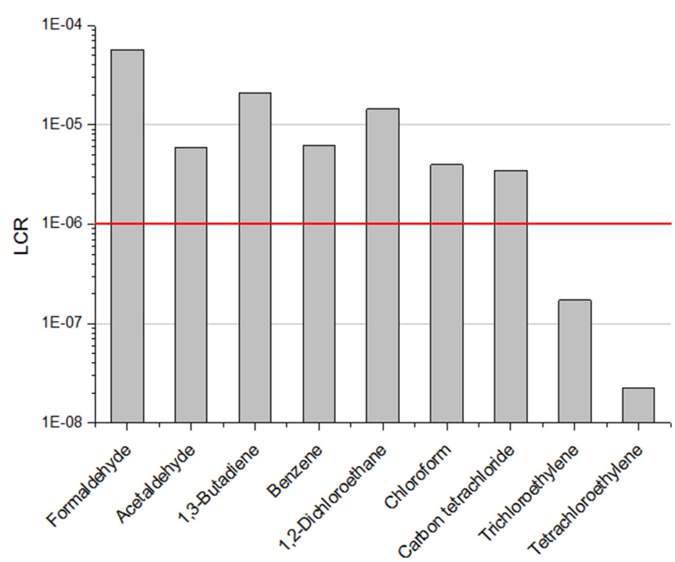

Fig. 6. Inhalation cancer risk evaluation for nine toxic compounds.

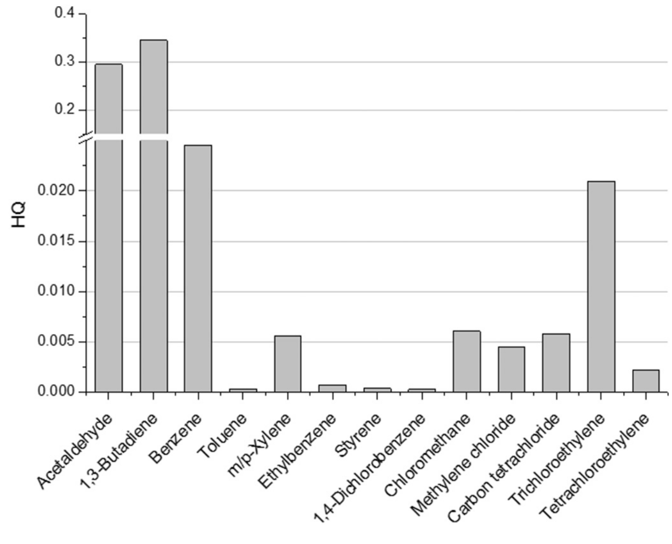

Fig. 7. Non-carcinogenic estimated risk of VOCs using Inhalation Reference Concentration.

be realized by reducing both indoor and outdoor emissions of these VOCs.

Four chemicals of acetaldehyde (5.84×10^{-6}), chloroform (3.96×10^{-6}), carbon tetrachloride (3.47×10^{-6}) and benzene (1.62×10^{-6}) presented median cancer risks but all were also higher than the acceptable risk of 1×10^{-6}. Trichloroethylene (1.72×10^{-7}) and tetrachloroethylene (2.26×10^{-8}) were well below the acceptable risk level.

3.3.2. Non-cancer hazard risk assessment

1,3-Butadiene presented the highest HQ value at 0.34, followed by acetaldehyde (0.29), but they were below the threshold value (HQ = 1). The other target VOCs with HQs values were far less than the 1 (Fig. 7). Adverse health effects are not expected to result from exposure to these VOCs according to the estimation.

3.3.3. Improvement with air purifier

The air purifier combines high efficiency particulate air filter (HEPA) with ambient temperature catalysis technology. The air flow driven by the top fan passes through the HEPA network, catalyst filling layer and inner filter layer and successively be purified hierarchically. At room temperature (15–35 °C), formaldehyde and other VOCs react with

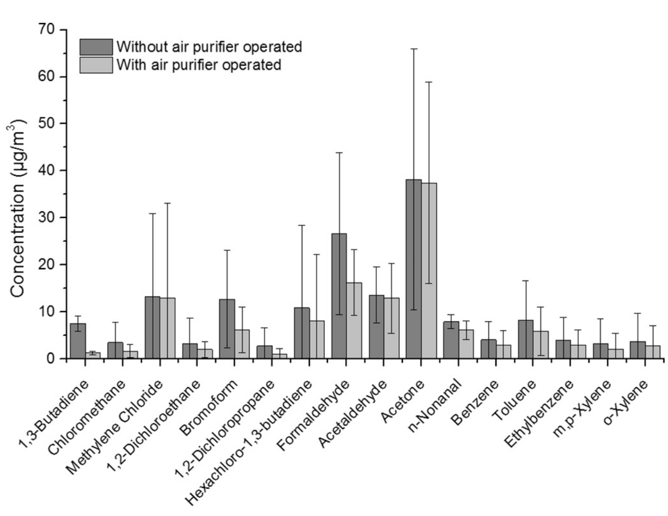

Fig. 8. The comparison of selected VOC concentrations with operation of indoor air purifier.

catalyst and rapidly decomposes into CO_2 and H_2O, which can effectively remove VOCs (Li et al., 2018). The I/O values of VOCs and carbonyls before air purifier operated and after the use of air purifier were shown in Fig. 3. It was obvious that most compounds' I/O values before air purifier operated were higher than the after the use of air purifier especially 1,1-dichloroethene, bromoform, 1,3-dichlorobenzene, chlorobenzene, formaldehyde and hexanal. The air purifier effectively reduces the indoor concentration of air pollutants. Sixteen selected VOCs and carbonyls at high indoor abundances were selected to compare the impact of operation of air purifiers to purify the indoor air (Fig. 8). Obvious declines in concentration were shown for both target compounds. The greatest improvement was seen for 1,3-butadiene and formaldehyde, which were from of 7.54 ± 1.57 and 26.66 ± 17.22 μg/m^3 to 1.26 ± 0.38 and 16.29 ± 13.41 μg/m^3, respectively. Correspondingly, their average estimated cancer risks have been also reduced from 4.65×10^{-5} and 7.12×10^{-5} to 7.8×10^{-6} and 4.35×10^{-5}. In addition, good purification efficiencies were also seen for the removal of chloromethane, 1,2-dichloroethane, bromoform, benzene, toluene and *m*,*p*-xylene. Even though the health risks for few of them are still higher than the acceptable value, the substantial reduction could benefit the human health.

4. Conclusions

Substantially high indoor VOCs and carbonyls concentrations were observed in dwellings in Xi'an during wintertime. Most of the targeted species were more abundant in China than other countries. The results from source apportionment conclude that both smoking, decoration, furniture and household products are dominated sources at the dwellings. The health risk of formaldehyde, 1,3-butadiene and 1,2-dichloroethane were much higher than the acceptable risk level, even though the hazard quotient of few target VOCs were far less than the threshold at non-cancer risk assessment. Preliminary data shows that the use of air purifier can effectively reduce most of the indoor organic pollutants, leading to decline in cancer risk to humans. The findings of this study provide solid data to policy makers for understanding of characteristic pollution sources, importance of IAQ management, and establishment of effective ambient pollution control strategies.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.01.250.

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