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# Volatile organic compounds from residential solid fuel burning in Guanzhong Plain, China: Source-related profiles and risks



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Jian Sun <sup>a</sup>, Jinhui Wang <sup>b, \*\*</sup>, Zhenxing Shen <sup>c, \*</sup>, Yu Huang <sup>d</sup>, Yue Zhang <sup>c</sup>, Xinyi Niu <sup>d, e</sup>, Junji Cao <sup>d</sup>, Qian Zhang <sup>f</sup>, Hongmei Xu <sup>c</sup>, Ningning Zhang <sup>d</sup>, Xuxiang Li <sup>a</sup>

<sup>a</sup> School of Human Settlements and Civil Engineering, Xi'an Jiaotong University, Xi'an, 710049, China

<sup>b</sup> NICU, Xi'an Children's Hospital, Xi'an, 710003, China

<sup>c</sup> Department of Environmental Science and Engineering, Xi'an Jiaotong University, Xi'an 710049, China

<sup>d</sup> Key Lab of Aerosol Chemistry & Physics, SKLLQG, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, 710049, China

<sup>e</sup> The Jockey Club School of Public Health and Primary Care, The Chinese University of Hong Kong, Hong Kong, China

f School of Environmental & Municipal Engineering, Xi'an University of Architecture and Technology, Xi'an 710055, China

# HIGHLIGHTS

• Profiles of toxic VOCs in 2 kitchens and 5 typical bedrooms were investigated.

• Kitchen with tradition wood stove showed the highest toxic VOCs concentrations.

• Cancer (noncancer) risk exposure to VOCs for women highly exceeded USEPA threshold.

• Indoor toxic VOCs were demonstrated mostly derived from indoor solid-fuel burning.

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# ABSTRACT

Characteristics of indoor volatile organic compounds (VOCs) and their health risks were investigated in kitchens and bedrooms during the heating season in rural Guanzhong Plain, China. Toxic-VOC concentrations in kitchens with traditional wood ( $299 \pm 38.8 \,\mu g \, m^{-3}$ ) and liquefied petroleum gas (LPG) stoves  $(187 \pm 54.6 \,\mu g \, m^{-3})$  were considerably higher than those in bedrooms. High levels of toxic VOCs in traditional kitchens were strongly correlated with wood combustion (R = 0.72). The coefficient of determination of VOC profiles between the kitchen and wood combustion was 0.27, indicating that VOCs in traditional kitchens are mainly derived from wood combustion. For women, who do most of the cooking, noncancer risk from exposure to toxic VOCs could reach 7600 and 2550 in traditional and LPG kitchens, respectively. Noncancer risks were much lower in bedrooms than in kitchens, but still two orders of magnitude higher than the United States Environmental Protection Agency (USEPA) threshold. Cancer risk from exposure to VOCs for women was  $8.98 \times 10^{-4}$  and  $1.67 \times 10^{-4}$  in both traditional and LPG kitchens, respectively, and ranged from  $2.51 \times 10^{-6}$  to  $3.85 \times 10^{-5}$  in bedrooms—all exceeding the USEPA threshold. Thus, during the heating season indicated that the rural Guanzhong residents were exposed to toxic VOCs from indoor heating and cooking at levels higher than the recommended safety levels. Moreover, traditional cooking and heating styles in rural Guanzhong need to be urgently updated to improve the indoor air quality for residents.

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# 1. Introduction

According to The Global Burden of Disease Study 2014 by World Health Organization (WHO), indoor air pollution had been the fifth largest contributor to human disease. Among the indoor air pollutants, volatile organic compounds (VOCs) are being proven hazardous to human health by the United States Environmental

<sup>\*</sup> Corresponding author.

<sup>\*\*</sup> Corresponding author.

*E-mail addresses:* jinhuiwang75@163.com (J. Wang), zxshen@mail.xjtu.edu.cn (Z. Shen).

Protection Agency (USEPA, 2009) and massive researchers. In general, benzene and many benzene-series VOCs, the main products of solid-fuel burning, are highly toxic and have been widely investigated (Colman Lerner et al., 2012; Liu et al., 2008, 2017). Carbonyl compounds have received more attention than other VOCs because of their potential adverse health effects on human and crucial roles in atmospheric chemistry (Ho et al., 2006). Some alkenes (e.g., 1,3-butadiene) and halogenated hydrocarbons (e.g., chloroform) are carcinogenic, and thus, they pose high risks to human health (USEPA, 1998).

High VOC levels in indoor environments can be derived from domestic sources, such as cleaning products, furnishing, cooking, and solid-fuel (biomass and coal)-burning heating systems (Chang et al., 2017; Lee et al., 2002b; Ohura et al., 2009; Soto-Garcia et al., 2015). During heating season, residential burning (heating and cooking) of solid fuels was the dominant indoor VOC source especially for those with stoves used in indoors. However, numerous researches have focused on the adverse effects of particulate matter (PM) and particle-bound components (e.g., polycyclic aromatic hydrocarbons) emitted from residential solid fuel burning (Andreae et al., 2001; Ho et al., 2016; Li et al., 2009). Research on gaseous emissions from cooking and space heating have been relatively lacking. Guo et al. (2004) reported that concentrations of nonmethane hydrocarbons (NMHCs) in restaurants were higher than in residential living rooms or bedrooms, indicating that cooking emissions are substantial indoor sources of NMHC pollution in enclosed spaces. Stabile et al. (2018) measured VOC emissions from biomass-burning heating systems and evaluated their effects on indoor environments, thus demonstrating that indoor burning of biomass fuels was the dominant source of VOCs, adversely affecting human health, particularly through respiratory system.

Understanding of the indoor VOC levels in rural dwellings is necessary not only as a basis for exposure assessment to the residents, but also to help in identifying the main indoor VOC source and making mitigations targetedly. However, exposure risks to indoor VOCs vary considerably because of the different toxicities, VOC abundance and most importantly the different guidelines. In the 1990s, the USEPA (1995) set several risk assessment guidelines for carcinogenicity. In the 2000s, the WHO (2000) and European Union (EU, 2008) both established annual concentration limits for many carcinogenic VOCs. In China, the guidelines (i.e., limits) for indoor VOC levels, however, are not comprehensive.

In the rural area of Guanzhong Plain, China, solid-fuel-burning heating systems are currently considered an indoor heating solution because of their low cost with compared with electricity and coal as well as unavailability of natural gas (Sun et al., 2018b). Moreover, in this area, many residents have planted apple trees; this has led to abundance of wood biomass, and therefore, higher wood consumption for heating when compared with other areas in China (Hou et al., 2017). Outdated heating and cooking stoves (namely heated kang, traditional cooking stove, and indoor heating stove)-which are generally used for combustion indoors-introduce a substantial amount of pollutants into the living spaces. The relatively low temperatures in winter lead to more time spent in indoors for the rural Guanzhong residents. Collectively, these conditions lead to the high VOC exposure and risks of indoor solid-fuel burning during the heating season for these residents.

In this study, the indoor profiles of toxic VOCs originating from main heating and cooking processes in seven typical indoor dwellings were determined. Moreover, lifetime cancer and noncancer risks related to exposure to VOCs, with their explicit carcinogenicities and toxicities, for male and female adults were evaluated.

# 2. Methodology

# 2.1. VOC sample collection

VOC samples were collected from three typical agricultural villages in the rural Guanzhong Plain (Figure S1) (Supplementary Material). A detailed introduction of fuel consumption and stove use are described in section S1 (Supplementary Material). Descriptions of the seven indoor sampling types are provided in Table S1 (Supplementary Material). Bedrooms and kitchens were selected as sampling sites because the rural residents spend majority of their time in these two places among the various indoor activities. According to the traditions there, the living room was not regularly used and the function of it was generally undertaken by bedroom. 4–6 dwellings were selected for each room type to get a more reliable and representative database (Table S1).

All VOC samples were collected through field experiments. The sampling platform was set 3-5 m from the stove at a 1.5-m height-the typical inhalation height for adults in China (Figure S2). To eliminate the emissions originating from cooking process, cooking activities were limited to only boiling water (tapwater meets the national standard of GB5749-2006) during VOC sampling. For VOC sampling, we used a 1/4" o.d. stainless-steel, multibed adsorbent tube filled with Tenax-TA, Carbograph 1-TD, and Carboxen 1003 (Markes International Ltd., Llantrisant, UK) using a low-flow module pump (ACTI-VOC, Markes International Ltd.) at a flow rate of  $50 \text{ mLmin}^{-1}$  for 60 min. A Teflon filter assembly (47 mm, Whatman, Clifton, NI, USA) and a homemade ozone scrubber, both constructed from a 1-m-long, 1/4" o.d. saturated potassium iodide (KI)-coated copper tube, were installed in the air upstream to remove any influences from PM and O<sub>3</sub>, respectively. The sorbent tubes were precleaned in a thermal conditioner (TC-20, Markes International Ltd.) at 330 °C for 20 min. All preconditioned and sampled tubes were capped and shipped at 0 °C. Nonsignificant breakthrough (<5%) was observed both in field and laboratory demonstrations, under this sampling flow and volume (Ho et al., 2017). Each test was performed three times to avoid experimental error and determine standard deviations. Breakthrough tests were conducted for the absorbent tube to ensure sufficient absorption for VOCs in this study. Source-emitted VOC samples were also collected; sample collection is described in Section S1 (Supplementary Material). Background VOC samples were also collected in outdoor air, 5–10 m away from the chimney. Background VOC samples were analyzed using the same protocol as the source sample collection. The background concentrations of VOCs were subtracted when calculating the source emitted VOC profiles; the data are listed in Table S2.

#### 2.2. VOC analysis

The absorbent tubes were analyzed using a thermal desorption (TD) unit (Series 2 UNITY-xr system, Markes International Ltd.) coupled with a gas chromatograph (GC)—mass spectrometric detector (MSD, Models 7890A/5977B, Agilent, Santa Clara, CA, USA). A tube was inserted into the TD unit at room temperature (~25 °C) and purged with ultrahigh-purity (UHP) helium (He) gas at a flow rate of 40 mL min<sup>-1</sup> 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 then maintained at 320 °C for 5 min. The analytes were passed through a heated transfer line at 160 °C and refocused onto a cold GC capillary column head (Rtx-1, 105-m × 0.25-mm × 1-µm film,

Restek Corporation, Bellefonte, PA, USA) at -45 °C by using liquid nitrogen in the GC oven. After the second-stage desorption was completed, the oven temperature program was started at an initial temperature of -45 °C for 4 min and subsequently increased to 230 °C at a rate of  $6 \circ C \cdot min^{-1}$  and then maintained at 230 °C for 5 min. The flow rate of He carrier gas was maintained constant at 1.0 mL min<sup>-1</sup> throughout the GC analysis. The MSD was operated in selective ion monitoring mode at 230 °C and 70 eV for electron ionization. The compounds were identified by comparing the mass spectra and retention times of the chromatographic peaks with certified standard sample. Certified Photochemical Assessment Monitoring Stations standard mixtures (Restek Corporation) were used in calibrations. A multipoint calibration curve was established to quantify each target compound with a linearity of >0.999. The minimum detection limits (MDLs) for 98 target analytes were in the range of 0.001–0.159 ppbv with a sampling volume of 3 L; the MDLs for all VOC species are listed in Table S3. The measurement precision level for the analysis of eight replicates of standard samples at 2 ppbv was  $\leq$ 5%. Duplicate samples were collected, and the reproducibility was >95%. Additional details on sampling and analytical methods were published by Ho et al. (2017) and (2018).

#### 2.3. Cancer and noncancer risk assessment

According Guo et al. (2004) and Huang et al. (2011), cancer risk from daily exposure of an individual to inhalation can be calculated by Eqs. (1) and (2):

$$CR = CDI * SF$$
(1)

where CR is the cancer risk, CDI is the chronic dose of intake through inhalation  $(mg \cdot kg^{-1} \cdot day^{-1})$ , and SF the slope factor of VOCs  $(kg \cdot d \cdot mg^{-1})$ . The SFs of main carcinogens, obtained from the Integrated Risk Information System (IRIS) (USEPA, 1998), are shown in Table S4. The chronic intake dose of a carcinogenic contaminant

Uncertainties =  $\sqrt{(\text{Error fraction } \times \text{Concertaintion})^2 + (0.5 \times \text{MDL})^2}$ 

was controlled by various factors, such as exposure frequency, exposure duration, and body weight of the receptor. The equation used to calculate CDI ( $mg \cdot kg^{-1} \cdot day^{-1}$ ) is

$$CDI = C_i * IR * EF * ED/(BW * AL * NY)$$
<sup>(2)</sup>

where  $C_i$  is the contaminant concentration  $(mg \cdot m^{-3})$ , IR the inhalation rate  $(m^3 \cdot h^{-1})$ , EF the exposure time  $(h \cdot day^{-1})$ , ED the total number of exposure days, BW the body weight (kg), AL the average lifetime (year), and NY the total number of days in 1 year (365 day · year^{-1}). Table S5 lists the activities data and relative parameters for risk assessment in this study based on our survey in the Guanzhong area and statistical data. Compounds with cancer risk higher than  $1 \times 10^{-4}$  are considered as "definite risk", between  $1 \times 10^{-6}$  and  $1 \times 10^{-4}$  as "probable risk" and less than  $1 \times 10^{-6}$  as "negligible risk" (An et al., 2014; He et al., 2015).

The noncancer risk of VOC exposure is quantified as hazard quotient (HQ) which are calculated by Eqs. (3) and (4) (Durmusoglu et al., 2010):

$$HQ = ADI/RfD$$
(3)

where ADI is the average daily intake  $(mg \cdot kg^{-1} \cdot day^{-1})$  and RfD the

reference dose for VOC species ( $mg \cdot kg^{-1} \cdot day^{-1}$ ). RfD values refer to the USEPA (USEPA, 1998) and are listed in Table S6. ADI differs from CDI because the acute effect does not require calculation as a lifetime average. Thus, ADI is calculated as follows:

$$ADI = (C^{1} * IR * EF) / BW$$
(4)

where  $C_i$  is the contaminant concentration  $(mg \cdot m^{-3})$ , IR the inhalation rate  $(m^3 \cdot h^{-1})$ , EF the exposure time  $(h \cdot day^{-1})$ , and BW the body weight (kg). For parameters, refer to Table S5. When the HQ is  $\geq$  1.0, an adverse health effect is posed to human beings (He et al., 2015).

Inhalation exposure is a simple multiple of the mean concentrations of certain VOCs and the corresponding exposure duration (Guo et al., 2004). For risk assessment, some parameters were established from assumptions, whereas others were based on professional judgement and questionnaire data. Also shown in Table S5, body weight and inhalation rate followed the IRIS guidelines (USEPA, 1998). An expected lifetime of 70 years was adopted for both the male and female groups. According to the questionnaire results, the heating season lasted for 100 days and home meals were prepared on average 300 days per year. In winter, residents in rural Guanzhong spent approximately 10 h per day in bedrooms and approximately 3 h per day in the kitchen to prepare three meals for the family; cooking was mainly performed by women.

# 2.4. Uncertainty

According to Norris et al. (2014), uncertainties from VOC measurement could be calculated by Eqs. (5) and (6), and the total uncertainty could be calculated by error propagation which was expressed as Eq. (7). When the concentration exceeded the MDL, the uncertainties were calculated as:

When the concentration measured was below the MDL, the uncertainties were calculated as:

Uncertainties 
$$=\frac{5}{6} \times MD$$
 (6)

For uncertainties derived from multiple sources, the calculation should be the combination of uncertainties as

$$Uncertainties = \sqrt{sum(Unc_i)^2}$$
(7)

where Unc<sub>i</sub> denotes the uncertainties in different sources i.

#### 3. Results and discussion

#### 3.1. Profiles of indoor toxic VOCs

Table 1 showed the 61 individuals of toxic VOCs determined for each sample collected from two kitchens and five bedrooms. In the kitchen during the cooking process, total VOC concentrations were  $299 \pm 38.8$  and  $187 \pm 54.6 \,\mu g \, m^{-3}$  when traditional cooking stoves and liquefied petroleum gas (LPG) stoves were used, respectively. The traditional cooking stoves using wood branches as fuel emitted

Table 1	
Profiles of VOCs in different indoor environments ( $\mu g$	-m <sup>-3</sup> ).

VOCs Kitchen		Bedroom					
	Wood branch stove	LPG stove	Anthracite	Residue Heated Kang	Electric heater	Clean stove with plumbing system	Air conditioner
Hexachloro-1 3-butadiene	$429 \pm 607$	802+113	<dl< td=""><td>8 68 + 0 06</td><td>451+637</td><td><di.< td=""><td><dl< td=""></dl<></td></di.<></td></dl<>	8 68 + 0 06	451+637	<di.< td=""><td><dl< td=""></dl<></td></di.<>	<dl< td=""></dl<>
1 2-Dichlorobenzene	<di< td=""><td><di< td=""><td><di< td=""><td><di< td=""><td>∠DI</td><td><di< td=""><td><di< td=""></di<></td></di<></td></di<></td></di<></td></di<></td></di<>	<di< td=""><td><di< td=""><td><di< td=""><td>∠DI</td><td><di< td=""><td><di< td=""></di<></td></di<></td></di<></td></di<></td></di<>	<di< td=""><td><di< td=""><td>∠DI</td><td><di< td=""><td><di< td=""></di<></td></di<></td></di<></td></di<>	<di< td=""><td>∠DI</td><td><di< td=""><td><di< td=""></di<></td></di<></td></di<>	∠DI	<di< td=""><td><di< td=""></di<></td></di<>	<di< td=""></di<>
1 4-Dichlorobenzene	$682 \pm 0.03$	<di< td=""><td>589 + 833</td><td><math>681 \pm 0.00</math></td><td><math>343 \pm 485</math></td><td><di< td=""><td><math>590 \pm 834</math></td></di<></td></di<>	589 + 833	$681 \pm 0.00$	$343 \pm 485$	<di< td=""><td><math>590 \pm 834</math></td></di<>	$590 \pm 834$
Actone	$486 \pm 492$	185 + 579	<di< td=""><td><math>1.48 \pm 1.09</math></td><td><di< td=""><td>401 + 568</td><td><math>2.30 \pm 0.07</math></td></di<></td></di<>	$1.48 \pm 1.09$	<di< td=""><td>401 + 568</td><td><math>2.30 \pm 0.07</math></td></di<>	401 + 568	$2.30 \pm 0.07$
Freon-12	$1.17 \pm 0.05$	$10.5 \pm 0.75$		$1.40 \pm 1.00$		$4.01 \pm 0.00$	2.25 ± 0.07
Chloromethane	$1.17 \pm 0.05$ $1.19 \pm 0.59$	0.0 < D1.03	$0.02 \pm 0.02$	$0.33 \pm 0.13$ 0.32 ± 0.11	$0.01 \pm 0.01$	$0.31 \pm 0.73$	$0.12 \pm 0.04$
Freon-114	-DI	$0.22 \pm 0.31$	<pre>0.02 ± 0.02</pre>	0.52 ± 0.11	0.01 ± 0.01	< <u>DI</u>	0.12 ± 0.04
Vinyl chloride	$\sqrt{DL}$ 0.71 $\pm$ 0.02	$1.13 \pm 0.05$	$0.49 \pm 0.69$	$\sqrt{55} \pm 0.01$	$(33 \pm 0.47)$	$0.53 \pm 0.74$	$\sqrt{DE}$
1 3-Butadiene	$11.4 \pm 9.65$	$3.28 \pm 4.64$	0.45 ± 0.05	$1.41 \pm 0.65$	0.55 ± 0.47	$0.05 \pm 0.07$	$0.30 \pm 0.00$ 0.72 ± 0.10
Bromomethane	$11.4 \pm 0.03$	$3.20 \pm 4.04$ $3.57 \pm 0.11$	$175 \pm 2.48$	$0.01 \pm 0.02$	$175 \pm 2.48$	$3.49 \pm 0.00$	$3.57 \pm 0.04$
Chloroethane	<pre>0.05 ± 0.04</pre>	∠DI	∠DI	0.01 ± 0.02	-DI	-DI	2.57 ± 0.04
Ethanol	$146 \pm 0.74$	$437 \pm 2.84$	$179 \pm 113$	$275 \pm 0.28$	$\sqrt{25} \pm 1.06$	$240 \pm 152$	$565 \pm 0.86$
Acrolein	$126 \pm 126$	$103 \pm 145$	<di< td=""><td><math>1.06 \pm 0.20</math></td><td><math>0.75 \pm 1.00</math> 0.16 ± 0.22</td><td><math>0.42 \pm 0.59</math></td><td><math>1.09 \pm 0.92</math></td></di<>	$1.06 \pm 0.20$	$0.75 \pm 1.00$ 0.16 ± 0.22	$0.42 \pm 0.59$	$1.09 \pm 0.92$
Freon-11	$327 \pm 023$	$59 \pm 102$	$522 \pm 0.03$	$2.92 \pm 0.01$	$2.60 \pm 3.68$	$5.12 \pm 0.00$ $5.36 \pm 0.21$	$5.22 \pm 0.00$
Isopropyl Alcohol	$0.21 \pm 0.3$	$0.56 \pm 0.15$	$0.19 \pm 0.05$	$0.42 \pm 0.04$	0.1 < DI 14	$0.29 \pm 0.34$	$0.48 \pm 0.08$
1 1-Dichloroethene	$0.21 \pm 0.03$	$2.30 \pm 0.15$	$2.13 \pm 0.20$	$0.42 \pm 0.04$	$1.17 \pm 1.62$	$1.12 \pm 1.58$	$2.40 \pm 0.00$
Methylene Chloride	$0.03 \pm 0.03$	$2.24 \pm 0.05$	$2.22 \pm 0.01$	$0.05 \pm 0.00$	$1.17 \pm 1.02$ 0.22 $\pm 0.03$	$1.12 \pm 1.58$ $1.57 \pm 1.95$	$2.51 \pm 0.02$
Carbon disulfide	$0.03 \pm 0.01$	0.7 < D1.30	$0.24 \pm 0.00$	0.2 < D1.01	$0.22 \pm 0.03$	$1.57 \pm 1.55$	$0.22 \pm 0.04$
Freen-113	$12.2 \pm 0.55$	$0.38 \pm 0.70$ $34.8 \pm 0.90$	$0.04 \pm 0.01$	$0.03 \pm 0.05$	$0.01 \pm 0.01$	$0.04 \pm 0.00$	$0.01 \pm 0.01$
trans_1.2_Dichloroethene	12.2 ± 13.8	$0.81 \pm 0.90$	$2.27 \pm 1.98$	$0.78 \pm 0.00$	$0.03 \pm 0.20$ 0.43 ± 0.58	$0.50 \pm 0.22$	$0.84 \pm 0.01$
1 1 Dichloroothano		0.01 ± 0.00	0.00 ± 0.01	20.05 ± 0.05	0.45 ± 0.56	0.41 ± 0.55	0.05 ± 0.00
Mothyltort butyl othor		<dl 1.40 ± 0.06</dl 	<dl 2 17 - 1 00</dl 			<dl 1.55 + 0.01</dl 	
Vipul Acotate	0.7 < DL.45	$1.49 \pm 0.00$	$3.17 \pm 1.00$	0.7 < DL.02	$0.93 \pm 0.97$	$1.55 \pm 0.01$	2.0 < DL.07
Mothyl athyl katono	$20.3 \pm 23.0$ 2 19 + 0 12	$07.4 \pm 1.00$	$9.09 \pm 13.99$	$33.0 \pm 4.03$	$10.4 \pm 20.0$ 1 20 + 1 51	$42.0 \pm 21.3$	$33.3 \pm 39.0$
sia 1.2 Disblaraathana	$3.18 \pm 0.13$	$2.16 \pm 1.49$	$0.27 \pm 0.38$	$0.96 \pm 0.22$	$1.39 \pm 1.51$	$0.74 \pm 1.05$	4.1 < DL.57
cis-1,2-Dichloroethene	$0.14 \pm 0.16$	$2.54 \pm 0.19$	$2.64 \pm 0.02$	$0.17 \pm 0.04$	$1.47 \pm 1.89$	$2.48 \pm 0.13$	$2.77 \pm 0.05$
Ethyl Acotato	$2.09 \pm 1.00$ 2.76 ± 0.10	$0.33 \pm 0.47$	$0.42 \pm 0.6$	$2.12 \pm 0.53$	$0.04 \pm 0.06$	$0.11 \pm 0.15$	$0.34 \pm 0.08$
Chloroform	$5.70 \pm 0.19$	$1.95 \pm 2.70$	$4.01 \pm 0.74$	$4.07 \pm 0.71$	$5.54 \pm 0.97$	$2.55 \pm 5.52$	$4.70 \pm 1.21$
Tatashudashuran	$0.16 \pm 0.02$	$0.18 \pm 0.04$	0.1 < DL.07	$0.13 \pm 0.00$	0.11±0.01	0.1 < DL.02	$0.17 \pm 0.01$
	<dl< td=""><td><math>0.28 \pm 0.35</math></td><td></td><td><dl 0.10 - 0.02</dl </td><td><dl< td=""><td><dl 0.14 - 0.00</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<>	$0.28 \pm 0.35$		<dl 0.10 - 0.02</dl 	<dl< td=""><td><dl 0.14 - 0.00</dl </td><td><dl< td=""></dl<></td></dl<>	<dl 0.14 - 0.00</dl 	<dl< td=""></dl<>
1,2-Dichloroethane	$0.19 \pm 0.04$	$0.13 \pm 0.18$	0.2 < DL.20	$0.10 \pm 0.03$	$0.24 \pm 0.06$	$0.14 \pm 0.09$	$0.26 \pm 0.04$
1,1,1-1richioroethane	U.I < DL.I3	$0.18 \pm 0.18$	$0.31 \pm 0.11$	$0.12 \pm 0.00$	$0.13 \pm 0.04$	0.1 < DL.14	$0.18 \pm 0.03$
Garban Tatra ablarida	$148 \pm 30.8$	<dl< td=""><td><math>9.02 \pm 0.33</math></td><td><math>15.04 \pm 5.85</math></td><td><dl< td=""><td><dl 0.74 - 0.12</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<>	$9.02 \pm 0.33$	$15.04 \pm 5.85$	<dl< td=""><td><dl 0.74 - 0.12</dl </td><td><dl< td=""></dl<></td></dl<>	<dl 0.74 - 0.12</dl 	<dl< td=""></dl<>
	$0.00 \pm 0.11$	$0.85 \pm 0.20$	$0.66 \pm 0.09$	$0.52 \pm 0.02$	$0.57 \pm 0.19$	$0.74 \pm 0.12$	$0.69 \pm 0.01$
1.2 Dichloropropaga	$0.54 \pm 0.19$	$0.41 \pm 0.33$	$0.47 \pm 0.20$	0.5 < DL.15	$0.27 \pm 0.36$	0.3 < DL.03	$0.33 \pm 0.14$
I,2-Dichloropiopane	$0.12 \pm 0.05$	$0.00 \pm 0.03$	$0.07 \pm 0.10$	$0.17 \pm 0.02$	$0.13 \pm 0.11$	$0.04 \pm 0.03$	$0.07 \pm 0.01$
	$0.24 \pm 0.07$	$0.06 \pm 0.04$	$0.15 \pm 0.00$	$0.24 \pm 0.04$	$0.15 \pm 0.00$	$0.04 \pm 0.03$	0.17 ± 0.04
Trichlers others	<dl 1.40 - 2.10</dl 	<dl< td=""><td><dl< td=""><td>5.76 ± 0.02</td><td><dl< td=""><td>0.39 ± 0.64</td><td><dl DI</dl </td></dl<></td></dl<></td></dl<>	<dl< td=""><td>5.76 ± 0.02</td><td><dl< td=""><td>0.39 ± 0.64</td><td><dl DI</dl </td></dl<></td></dl<>	5.76 ± 0.02	<dl< td=""><td>0.39 ± 0.64</td><td><dl DI</dl </td></dl<>	0.39 ± 0.64	<dl DI</dl 
Mathul Mathagradata	$1.48 \pm 2.10$	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<>	<dl .DL</dl 	<dl< td=""></dl<>
	<dl 1.02 - 0.01</dl 	<dl< td=""><td><dl< td=""><td><dl 1.10 - 0.20</dl </td><td><dl< td=""><td><dl 0.20 - 0.10</dl </td><td><ul< td=""></ul<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl 1.10 - 0.20</dl </td><td><dl< td=""><td><dl 0.20 - 0.10</dl </td><td><ul< td=""></ul<></td></dl<></td></dl<>	<dl 1.10 - 0.20</dl 	<dl< td=""><td><dl 0.20 - 0.10</dl </td><td><ul< td=""></ul<></td></dl<>	<dl 0.20 - 0.10</dl 	<ul< td=""></ul<>
Nethyl Jachytyl Ketere	$1.02 \pm 0.01$	$1.00 \pm 0.99$	$0.34 \pm 0.44$	$1.10 \pm 0.20$	$0.00 \pm 0.10$	$0.30 \pm 0.10$	$1.10 \pm 0.03$
sia 1.2 Dishlarananan	$0.01 \pm 0.02$	$0.05 \pm 0.07$	$0.08 \pm 0.06$	<dl< td=""><td><math>0.08 \pm 0.11</math></td><td><math>0.01 \pm 0.02</math></td><td><math>0.15 \pm 0.03</math></td></dl<>	$0.08 \pm 0.11$	$0.01 \pm 0.02$	$0.15 \pm 0.03$
cis-1,3-Dichloropropene	$0.01 \pm 0.02$	$0.02 \pm 0.03$	$0.09 \pm 0.01$	$0.04 \pm 0.01$	$0.05 \pm 0.05$	0.01 ± 0.01	$0.07 \pm 0.03$
1.1.2 Tricklassethans	<dl< td=""><td><dl< td=""><td></td><td><dl< td=""><td><dl< td=""><td><dl di<="" td=""><td><dl< td=""></dl<></td></dl></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td></td><td><dl< td=""><td><dl< td=""><td><dl di<="" td=""><td><dl< td=""></dl<></td></dl></td></dl<></td></dl<></td></dl<>		<dl< td=""><td><dl< td=""><td><dl di<="" td=""><td><dl< td=""></dl<></td></dl></td></dl<></td></dl<>	<dl< td=""><td><dl di<="" td=""><td><dl< td=""></dl<></td></dl></td></dl<>	<dl di<="" td=""><td><dl< td=""></dl<></td></dl>	<dl< td=""></dl<>
T, 1,2-Inchioroethane	$0.02 \pm 0.00$	$0.03 \pm 0.03$	0.1 < DL.06	$0.03 \pm 0.02$	$0.03 \pm 0.01$	<dl< td=""><td><math>0.25 \pm 0.02</math></td></dl<>	$0.25 \pm 0.02$
l'oluene	$17.5 \pm 8.43$	$0.92 \pm 1.06$	$1.41 \pm 1.82$	$4.88 \pm 0.84$	$1.68 \pm 0.73$	$0.77 \pm 0.50$	$6.88 \pm 0.23$
Dibase ashlare mathema	$0.18 \pm 0.21$	$1.04 \pm 0.12$	$0.8 \pm 0.02$	$0.05 \pm 0.06$	$0.41 \pm 0.50$	0.68 ± 0.32	$0.91 \pm 0.12$
1.2 Dibromosthere	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl .DL</dl </td><td><dl< td=""></dl<></td></dl<>	<dl .DL</dl 	<dl< td=""></dl<>
Tetre chloro ether e	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td></td><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td></td><td><dl< td=""></dl<></td></dl<>		<dl< td=""></dl<>
Chlanghannana	$0.01 \pm 0.01$	$0.01 \pm 0.01$	$0.01 \pm 0.00$	$0.02 \pm 0.01$	$0.04 \pm 0.05$	0.0 < DL.01	$0.03 \pm 0.01$
Chlorobenzene	$0.13 \pm 0.05$	$0.02 \pm 0.03$	$0.04 \pm 0.03$	$0.11 \pm 0.01$	$0.06 \pm 0.02$	$0.03 \pm 0.02$	$0.08 \pm 0.01$
Ethylbenzene	$1.84 \pm 0.23$	$1.89 \pm 0.15$	$2.01 \pm 0.26$	$0.65 \pm 0.03$	$1.27 \pm 1.01$	$1.86 \pm 0.06$	$2.93 \pm 0.06$
p-Xylene	$1.15 \pm 0.04$	2.0 < DL.17	$2.19 \pm 0.04$	$0.78 \pm 0.17$	$1.18 \pm 1.12$	$1.98 \pm 0.09$	$2.9 \pm 0.55$
m-Xylene	$1.62 \pm 0.05$	$0.78 \pm 0.17$	1.0 < DL.03	$1.21 \pm 0.2$	$0.75 \pm 0.03$	0.75 ± 0.12	1.81 ± 0.62
	<ul 17.2 0.74</ul 	<ul< td=""><td><ul< td=""><td><ul 0.45 - 0.12</ul </td><td><ul< td=""><td></td><td><dl< td=""></dl<></td></ul<></td></ul<></td></ul<>	<ul< td=""><td><ul 0.45 - 0.12</ul </td><td><ul< td=""><td></td><td><dl< td=""></dl<></td></ul<></td></ul<>	<ul 0.45 - 0.12</ul 	<ul< td=""><td></td><td><dl< td=""></dl<></td></ul<>		<dl< td=""></dl<>
Stylelle	$1/.3 \pm 0.74$	1.1±0.85	0.03 ± 10.03	0.45 ± 0.12	$0.57 \pm 0.12$	0.35 ± 0.19	1.88 ± 0.94
1,1,2,2-1etrachloroethane	<ul< td=""><td><ul< td=""><td><ul< td=""><td><ul< td=""><td><ul< td=""><td><ul< td=""><td><dl< td=""></dl<></td></ul<></td></ul<></td></ul<></td></ul<></td></ul<></td></ul<>	<ul< td=""><td><ul< td=""><td><ul< td=""><td><ul< td=""><td><ul< td=""><td><dl< td=""></dl<></td></ul<></td></ul<></td></ul<></td></ul<></td></ul<>	<ul< td=""><td><ul< td=""><td><ul< td=""><td><ul< td=""><td><dl< td=""></dl<></td></ul<></td></ul<></td></ul<></td></ul<>	<ul< td=""><td><ul< td=""><td><ul< td=""><td><dl< td=""></dl<></td></ul<></td></ul<></td></ul<>	<ul< td=""><td><ul< td=""><td><dl< td=""></dl<></td></ul<></td></ul<>	<ul< td=""><td><dl< td=""></dl<></td></ul<>	<dl< td=""></dl<>
o-Xylene	$1.36 \pm 0.07$	$0.54 \pm 0.12$	$0./1 \pm 0.01$	$0.95 \pm 0.19$	$0.56 \pm 0.08$	$0.52 \pm 0.10$	$1.33 \pm 0.51$
4-Ethyltoluene	$0.35 \pm 0.06$	1.1 < DL.02	$1.24 \pm 0.09$	$0.24 \pm 0.05$	$0.59 \pm 0.69$	$1.08 \pm 0.02$	$1.61 \pm 0.40$
1,3,5-1rimethylbenzene	U.4 < DL.01	$0.17 \pm 0.25$	$0.93 \pm 0.62$	U.4 < DL.UI	$0.12 \pm 0.18$	$0.34 \pm 0.00$	$0.76 \pm 0.30$
1,2,4-Trimethylbenzene	$2.37 \pm 0.06$	$2.17 \pm 2.78$	$1./4 \pm 0.49$	$2.29 \pm 0.13$	$0.53 \pm 0.37$	$0.21 \pm 0.01$	$2./1 \pm 1.44$
1,3-Dichlorobenzene	$5.17 \pm 0.02$	<dl< td=""><td><math>4.82 \pm 6.82</math></td><td><math>5.16 \pm 0.00</math></td><td><math>2.60 \pm 3.68</math></td><td>4.81±0.81</td><td><math>4.83 \pm 6.83</math></td></dl<>	$4.82 \pm 6.82$	$5.16 \pm 0.00$	$2.60 \pm 3.68$	4.81±0.81	$4.83 \pm 6.83$
∑ vucs	299±38.8	18/±54.6	$/1.5 \pm /./9$	129±6./5	54.5 ± 25.1	87.5±33.8	$112 \pm 1/.9$
<dl concentration<="" denotes="" td="" the=""><td>on was below detect l</td><td>imit</td><td></td><td></td><td></td><td></td><td></td></dl>	on was below detect l	imit					

Up to 3 significant digits were used in this table.

more concentrations of toxic VOCs into the indoor atmosphere than LPG stoves with a factor of 1.6 (p < 0.05). Wang et al. (2018) reported that VOCs emitted from LPG cooking were dominated by alkanes and alkenes because of the incomplete combustion. Moreover, Evtyugina et al. (2014) and Wang et al. (2013) indicated

that the emission factors of these VOCs were considerably lower than wood burning. Another difference between the profiles of the two types of kitchens was the species preponderance; 10 VOC species with the highest abundance in each indoor environment are listed in Table S7. For the kitchen using tree branches, benzeneseries VOCs (i.e., benzene, toluene, and styrene) represented more than 50% of the total toxic VOCs recorded. Benzene-series VOCs were the main products of residential wood burning in both our previous studies (Sun et al., 2018a) and the study by Evtyugina et al. (2014). By contrast, concentrations of benzene and benzene-series VOCs were relatively low in the LPG kitchen. Carbonyls (i.e., acetone, acrolein, and ethanol) was the most abundance species among the measured VOCs in the LPG kitchen; this is consistent with the LPG combustion emissions reported by Ho et al. (2006) and Huang et al. (2011).

For bedrooms, the toxic-VOC concentrations ranged from  $54.5 \pm 25.1$  to  $129 \pm 6.75 \,\mu\text{g m}^{-3}$ —significantly lower than those in kitchens (p < 0.05). The toxic-VOC concentrations in Bedroom 2 were lower than those in the two kitchens although with combustion sources (p < 0.05). Two reasons, namely low burning intensity and enclosed construction of the heating stove, possibly explain the low VOC levels in the bedroom using an anthracite stove for heating. Although the highest VOC level in Bedroom 2 was over twice the lowest one in Bedroom 3, the difference in toxic-VOC concentrations between any two adjacent gradient bedrooms was insignificant (p > 0.05). The 10 most abundant VOC species in the 5 bedrooms listed in Table S7, and vinyl acetate was the highest abundance among them. However, vinyl acetate is reported to be derived from multiple sources, such as combustion, furnishing and building materials (Uhde and Salthammer, 2007; Yu and Crump, 1998). Thus, residential heating activities may not be the sole contributors to toxic VOCs in the dwellings in Guanzhong. Benzene was the second most abundant in Bedrooms 1 and 2 but was not even listed as a top-10 VOCs in the other bedrooms. This is likely because Bedrooms 1 and 2 were both potentially affected by combustion smoke, in which benzene was at high concentrations (Akagi et al., 2011; Iinuma et al., 2010; Lemieux et al., 2004). This suggests that the toxic-VOC concentrations in Bedrooms 3-5, which use electricity or water for heating, should be much lower because these methods do not emit VOCs during operation. However, the result was that the average VOC concentrations in the "clean heating" bedrooms (Bedrooms 3-5,  $84.6 \pm 28.8 \,\mu g \,m^{-3}$ ) were comparable with those in combustion-heated bedrooms (Bedrooms 1 and 2,  $100 \pm 40.6 \,\mu g \, m^{-3}$ ; p > 0.05). This confirmed that indoor solid-fuel burning was not the sole contributor of indoor toxic VOCs in rural Guanzhong.

In indoor air of kitchen, the concentrations of toxic VOCs were much higher in bedrooms (p < 0.05). It indicated that the open-fire stoves (cook stoves) resulted in much higher influence to indoor environments than enclosing ones (heating stove). More severe case was reported by Lui et al. (2017) who in kitchens using an open stove without chimney identified carbonyls concentrations in ranges over 1000  $\mu$ g m<sup>-3</sup>. While in the present study, the VOC level in LPG kitchen was still much higher than those in bedrooms although LPG was generally deemed as clean energy (Goldemberg et al., 2004; Gould and Urpelainen, 2018). It verified that an enclosing structure of stove could efficiently prevent the VOCs in smoke leak into indoors.

#### 3.2. Correlations with combustion sources

To further investigate the influence of indoor combustion sources on concentrations of indoor VOCs, the correlations between VOC source profiles and the relevant indoor environment were calculated (Fig. 1). For the kitchens and bedrooms, with combustion sources (wood- and anthracite-burning stoves, respectively), the correlations were significant with R values of 0.72 and 0.75, respectively (p < 0.05). The slope factor of the fitted line for the wood stove kitchen was 0.56, indicating the severe influence of wood combustion on VOCs in kitchens. For the bedrooms with an anthracite-heating stove, the slope factor was as low as 0.05, indicating a low VOC emission from the stove to the bedroom. For the two bedrooms without combustion sources, the correlations were insignificant between indoor and source-emitted VOCs (R = 0.08 and -0.04, respectively, p > 0.05), indicating that removing the stove and using electric or water heating could effectively eliminate the VOCs emitted during combustion processes, providing a feasible solution to rural Guanzhong residents, thus enhancing their indoor air quality during the heating season.

The coefficient of divergence (CD) was employed here to evaluate the similarity of VOC profiles between source-emitted and relevant indoor levels. Calculations of the CD are shown in Section S2 (Supplementary Material) and the results were listed in Table 2. For VOC profiles in Kitchen 1 and Bedroom 1, the CDs were 0.27 and 0.29, respectively, with their corresponding source VOC profiles, confirming the results from correlation analysis that the indoor use of stoves (including cooking and heating stoves) had crucial effects on indoor VOC concentrations. CDs were slightly higher than 0.2 (cut point for similar) (Menchaca-Torre et al., 2015; Wongphatarakul et al., 1998); therefore, other crucial VOCs, such as those from furnishings (Uhde and Salthammer, 2007; Yu and Crump, 1998) and outdoor air (Adgate et al., 2004; Jia et al., 2008), must have influenced Kitchen 1 and Bedroom 1. The CD between VOCs in Bedroom 2 and from the residue burning in the heated kang was 0.53, slightly lower than that calculated for Bedroom 3 (0.59). The smoke tunnel under the heated kang may also lead to smoke leakage indoors, whereas this leakage could completely be avoided by plumbing-based heating. The relatively high level of VOCs in Bedroom 2 also indicated that heated kangs may release some burning-related VOCs into bedrooms; however, this is not as serious as that of indoor heating stoves.

## 3.3. Noncancer risk assessment

The individual HQs of average daily exposure to indoor VOC species and the total HQ were estimated and listed in Table 3. Differences were apparent in HQs between sexes. In kitchens, women were at major noncancer risks because they undertook the main household duties in the family. For men, HQs were lower than those for women also because of their higher average body weights of men. Therefore, the actual noncancer risk of female exposure to indoor VOCs, including from both kitchens and bedrooms, was much higher than that of men, who only exhibited notable risk in the bedrooms. Nevertheless, the noncancer risk for men was higher than 100, two orders of magnitude higher than the safe threshold of 1 (USEPA, 2009). For women, the total noncancer risks were as high as 8368 (the sum of 7600 and 768), more than 10 times higher than the safe threshold of heating season in rural Guanzhong requires urgent improvement.

Noncancer risk from VOC exposure was significant higher in kitchens than in bedrooms (p < 0.05). Without the HQ from benzene, noncancer exposure risk in the LPG kitchen was one-third that of the traditional kitchen for women (2550 vs. 7600); it remained 3–20 times higher than that in bedrooms. Huang et al. (2011) found VOC concentrations in kitchens could increase 10fold during oil-based cooking. In that case, the noncancer risk should be even higher and more detrimental to human health. The noncancer risk of exposure was the highest in the bedrooms with heated kangs ( $\Sigma$ HQ = 767.6); it was mainly contributed by benzene and acrolein. Similarly, in the bedrooms with anthracite stoves, the high  $\sum$  HQ was also mainly derived from benzene and acrolein. This similarity was due to the indoor solid-fuel combustion sources. The noncancer risks from VOC exposure in Bedrooms 3 and 4 were the lowest because the electric and plumbing-based heating system did not emit VOCs. Although clean heating measures were taken, it



Fig. 1. Correlations between VOCs profiles from sources and in relevant indoor environments.

# Table 2 CD between VOCs profiles from sources and in relevant indoor environment.

VOCs source	Branch burning in tradition cooking stove	Anthracite burning in traditional heating stove	Corn residue burning in Heated Kang	Wood burning in clean stove
Indoor environment	Kitchen 1	Bedroom 1	Bedroom 2	Bedroom 3
CD	0.27	0.29	0.53	0.59

remained in excess of the safe threshold, indicating the presence of other VOCs from sources, such as furnishing and tobacco smoking (Andreae et al., 2001; McAuley et al., 2012). The air conditioner was an exception among clean heating devices; in that, the  $\sum$ HQ in Bedroom 5 was high (420), indicating that it emitted VOCs during operation. Some studies have found that air conditioning systems could enhance indoor VOC levels if not maintained regularly (Lee et al., 2002a; Yu et al., 2009).

More than half of the 14 VOCs with confirmed noncancer risk demonstrated HQ values of higher than 1; in other words, they posed obvious noncancer risk (USEPA, 2009). In general, benzene, acrolein, and carbon tetrachloride have high HQs, which can be two or three orders of magnitude higher than those of other species. These three species are mainly derived from combustion sources (Chagger et al., 1999; Evtyugina et al., 2014) and were commonly found in the VOC profiles in our previous studies (Table S2). Clean heating methods, such as electric heaters, plumbing-based systems, and air conditioners, can help eliminate some toxic VOC species (e.g., benzene) but the remaining toxic VOCs (e.g., acrolein and carbon tetrachloride) can still lead to very high noncancer risks. Therefore, only by combination of both, stove upgrade and VOC elimination measures in rural Guanzhong Plain, the high exposure risk could be reduced completely (Chen et al., 2016; Li et al., 2013).

# 3.4. Cancer risk assessment

The individual lifetime cancer risk from average daily exposure to indoor VOC species and the total cancer risks were listed in Table 4. Similar to noncancer risk, the cancer risk of exposure in kitchens was mainly noted for women, and the risks in the two kitchens both exceeded the acceptable threshold  $(10^{-4})$  established by the USEPA (USEPA, 1998), indicating that more than 100 cases of cancer per million people would be caused by VOC exposure. Women are exposed toxic VOCs in both the kitchen and bedroom and therefore have higher cancer risk from exposure to indoor VOCs than men do. However, although men did not cook much in the kitchens, they demonstrated certain cancer risks from exposure to VOCs ( $2.97 \times 10^{-5}$  to  $1.94 \times 10^{-6}$ ), higher than the risk tolerance value of  $10^{-6}$ , defined by the USEPA (2009)—strongly suggesting that precautions are need to avoid indoor VOCs during the heating season in rural Guanzhong dwellings.

Cancer risk from exposure to VOCs was more than 10 times higher in the kitchens than in the bedrooms (p < 0.05). The LPG kitchen also posed a much lower cancer risk to women ( $1.67 \times 10^{-4}$ ) than posed by the traditional kitchen ( $8.98 \times 10^{-4}$ ) because of the absence of benzene. Among bedrooms, the highest cancer risk was observed in the one using heated kang ( $2.97 \times 10^{-5}$ )

#### Table 3

Non-cancer risk of VOCs exposure brought by indoor solid fuel burning.

Gender VOCs		Kitchen		Bedroom				
		Wood branch stove	LPG stove	Anthracite	Residue Heated Kang	Electric heater	Clean stove with plumbing system	Air conditioner
Male	Chloroform	_	-	0.9	1.1	1.0	0.9	1.5
	Benzene	_	_	193	322	N/A	N/A	N/A
	Carbon Tetrachloride	-	-	14.1	11.1	12.3	15.8	14.8
	1,4-Dioxane	-	-	N/A	10.8	N/A	1.7	N/A
	Trichloroethene	-	-	N/A	N/A	N/A	N/A	N/A
	cis-1,3-Dichloropropene	-	-	0.3	0.1	0.2	N/A	0.2
	Ethylbenzene	-	-	1.7	0.6	1.1	1.6	2.5
	Styrene	-	-	0.3	0.2	0.2	0.2	0.8
	Acrolein	-	-	109	347	110	215	352
	Toluene	-	_	1.7	5.4	2.0	1.0	7.6
	p-Xylene	_	_	1.9	1.3	1.4	1.8	2.2
	m-Xylene	_	_	0.4	0.5	0.3	0.3	0.8
	o-Xylene	-	-	0.3	0.4	0.2	0.2	0.6
	Acetone	-	-	N/A	1.3	N/A	3.4	1.9
	∑HQ	-	-	323	702	128	242	384
Female	Chloroform	1.8	2.1	1.0	1.2	1.1	0.9	1.6
	Benzene	4150	N/A	211	353	N/A	N/A	N/A
	Carbon Tetrachloride	18.7	24.0	15.5	12.2	13.4	17.3	16.2
	1,4-Dioxane	N/A	N/A	N/A	11.8	N/A	1.9	N/A
	Trichloroethene	334	N/A	N/A	N/A	N/A	N/A	N/A
	cis-1,3-Dichloropropene	N/A	0.1	0.3	0.1	0.2	N/A	0.2
	Ethylbenzene	2.1	2.1	1.9	0.6	1.2	1.7	2.7
	Styrene	9.8	0.6	0.3	0.2	0.3	0.2	0.9
	Acrolein	3050	2510	119	380	120	236	385
	Toluene	24.8	1.5	1.9	5.9	2.2	1.1	8.3
	p-Xylene	1.9	2.3	2.0	1.4	1.6	1.9	2.4
	m-Xylene	0.9	0.4	0.5	0.6	0.4	0.4	0.8
	o-Xylene	0.8	0.3	0.3	0.4	0.3	0.2	0.6
	Acetone	5.5	12.7	N/A	1.4	N/A	3.8	2.1
	∑HQ	7600	2550	354	768	140	265	420

Up to 3 significant digits were used in this table.

N/A denotes no noncancer risks were assessed due to extremely low VOC concentrations.

## Table 4

Cancer risk of VOCs exposure brought by indoor solid fuel burning.

Gender VOCs		Kitchen			Bedroom			
		Wood branch stove	LPG stove	Anthracite	Residue Heated Kang	Electric heater	Clean stove with plumbing system	Air conditioner
Male	1,3-Butadiene	_	-	$1.84\times10^{-5}$	$1.42\times10^{-5}$	N/A	$4.87\times10^{-7}$	$\textbf{7.25}\times 10^{-6}$
	Chloroform	-	_	$\textbf{3.26}\times \textbf{10}^{-8}$	$4.09\times10^{-8}$	$\textbf{3.66}\times \textbf{10}^{-\textbf{8}}$	$3.16  imes 10^{-8}$	$5.51  imes 10^{-8}$
	Benzene	-	_	$\textbf{4.38}\times \textbf{10}^{-6}$	$7.32\times10^{-6}$	N/A	N/A	N/A
	Carbon Tetrachloride	-	_	$1.66  imes 10^{-6}$	$1.31\times10^{-6}$	$1.44\times10^{-6}$	$1.86  imes 10^{-6}$	$1.74\times10^{-6}$
	1,4-Dioxane	-	_	N/A	$6.34\times10^{-6}$	N/A	$9.94  imes 10^{-7}$	N/A
	Trichloroethene	-	_	N/A	$\text{N/A} \times 10 + 00$	N/A	N/A	N/A
	cis-1,3-Dichloropropene	-	_	$7.96  imes 10^{-8}$	$3.06  imes 10^{-8}$	$4.42\times10^{-8}$	$4.95  imes 10^{-9}$	$6.12  imes 10^{-8}$
	Ethylbenzene	-	_	$\textbf{2.93}\times \textbf{10}^{-7}$	$9.54\times10^{-8}$	$1.85  imes 10^{-7}$	$2.71 \times 10^{-7}$	$4.27\times10^{-7}$
	Styrene	-	_	$5.80  imes 10^{-9}$	$4.26\times10^{-9}$	$5.44 imes10^{-9}$	$5.08  imes 10^{-9}$	$1.80  imes 10^{-8}$
	1,3-Dichlorobenzene	-	_	$\textbf{3.23}\times 10^{-6}$	$3.46\times10^{-7}$	$1.74\times10^{-7}$	$3.23 \times 10^{-7}$	$3.24  imes 10^{-7}$
	Methylene Chloride	-	_	$5.75\times10^{-8}$	$4.76\times10^{-8}$	$\textbf{5.08}\times \textbf{10}^{-8}$	$3.69 \times 10^{-7}$	$5.25\times10^{-8}$
	∑Risk	-	_	$\textbf{2.81}\times 10^{-5}$	$2.97  imes 10^{-5}$	$1.94  imes 10^{-6}$	$4.35  imes 10^{-6}$	$9.93  imes 10^{-6}$
Female	1,3-Butadiene	$5.35\times10^{-4}$	$1.54\times10^{-4}$	$\textbf{2.38}\times10^{-5}$	$1.84\times10^{-5}$	N/A	$6.31 \times 10^{-7}$	$9.40\times10^{-6}$
	Chloroform	$2.36 imes10^{-7}$	$2.73\times10^{-7}$	$4.22\times10^{-8}$	$5.30 imes10^{-8}$	$4.74 imes10^{-8}$	$4.10  imes 10^{-8}$	$7.14  imes 10^{-8}$
	Benzene	$3.35\times10^{-4}$	N/A	$5.68  imes 10^{-6}$	$9.48\times10^{-6}$	N/A	N/A	N/A
	Carbon Tetrachloride	$7.79\times10^{-6}$	$1.00  imes 10^{-5}$	$2.15\times10^{-6}$	$1.69\times10^{-6}$	$1.87\times10^{-6}$	$2.41 \times 10^{-6}$	$\textbf{2.25}\times 10^{-6}$
	1,4-Dioxane	N/A	N/A	N/A	$8.22\times10^{-6}$	N/A	$1.29\times10^{-6}$	N/A
	Trichloroethene	$8.13  imes 10^{-7}$	N/A	N/A	N/A	N/A	N/A	N/A
	cis-1,3-Dichloropropene	$4.38\times10^{-8}$	$9.67\times10^{-8}$	$1.03  imes 10^{-7}$	$3.97  imes 10^{-8}$	$5.73 imes10^{-8}$	$6.41 \times 10^{-9}$	$7.93  imes 10^{-8}$
	Ethylbenzene	$1.25\times10^{-6}$	$1.29\times10^{-6}$	$3.80  imes 10^{-7}$	$1.24\times10^{-7}$	$\textbf{2.40}\times 10^{-7}$	$3.51 \times 10^{-7}$	$5.53 imes10^{-7}$
	Styrene	$7.73  imes 10^{-7}$	$4.90\times10^{-8}$	$7.52  imes 10^{-9}$	$5.52\times10^{-9}$	$7.06  imes 10^{-9}$	$6.58  imes 10^{-9}$	$\textbf{2.33}\times 10^{-8}$
	1,3-Dichlorobenzene	$1.62\times10^{-5}$	N/A	$4.19\times10^{-6}$	$\textbf{4.48}\times\textbf{10}^{-7}$	$\textbf{2.26}\times \textbf{10}^{-7}$	$4.19\times10^{-7}$	$\textbf{4.20}\times \textbf{10}^{-7}$
	Methylene Chloride	$6.92\times10^{-7}$	$\textbf{7.69}\times 10^{-7}$	$7.45\times10^{-8}$	$6.17\times10^{-8}$	$\textbf{6.59}\times 10^{-8}$	$4.78\times10^{-7}$	$\textbf{6.80}\times \textbf{10}^{-8}$
	∑Risk	$\textbf{8.98}\times \textbf{10}^{-4}$	$1.67\times10^{-4}$	$\textbf{3.65}\times 10^{-5}$	$\textbf{3.85}\times \textbf{10}^{-5}$	$2.51\times10^{-6}$	$5.63\times 10^{-6}$	$1.29\times10^{-5}$

N/A denotes no cancer risks were assessed due to extremely low VOC concentrations.

and  $3.85\times10^{-5}$  for men and women, respectively) and the lowest one was in the bedroom using electric heater ( $1.94\times10^{-6}$  and  $2.51\times10^{-6}$  for men and women, respectively). However, all were above the risk tolerance value. The comparison of the cancer risks

in Bedrooms 1 and 2 with those in Bedrooms 3–5 indicated that indoor solid-fuel burning clearly influenced cancer risk from exposure to VOCs. Although using an air conditioner for heating (Bedroom 5) caused relatively high cancer risk ( $9.93 \times 10^{-6}$  and

 $1.29 \times 10^{-5}$  for men and women, respectively) comparable to that of the solid-fuel-burning bedrooms, this phenomenon occurs because of improper use and maintenance of the air conditioner, which introduces VOCs to the indoor environment (Lee et al., 2002a; Yu et al., 2009).

The carcinogen 1.3-butadiene was of great cancer risk in both kitchens, with cancer risks for women of  $5.35 \times 10^{-4}$  and  $1.54 \times 10^{-4}$  in traditional and LPG kitchens, respectively. It was reported that indoor 1,3-butadiene is mainly derived from combustion processes (Gustafson et al., 2007). Benzene was another crucial cancer risk source in the traditional kitchen  $(3.35 \times 10^{-4})$ . Carbon tetrachloride and ethylbenzene are two other species with cancer risks higher than  $10^{-6}$  and were mainly emitted from combustion sources (Gustafson et al., 2007). Similarly, these three VOC species were the main carcinogens with cancer risk over 10<sup>-6</sup> in bedrooms using solid-fuel burning for heating (Bedrooms 1 and 2). In Bedrooms 3 and 4, only carbon tetrachloride posed cancer risk over  $10^{-6}$ , indicating that the use of electric heaters and plumbing systems could effectively reduce the indoor VOC exposure and cancer risk. However, the total cancer risks remained above the tolerance value, suggesting that updating the heating system was insufficient to reach the indoor standard for cancer risk from exposure to VOCs.

# 4. Uncertainty analysis

Many uncertainties exist in the risk assessment of exposure. including uncertainties in measurement (Guo et al., 2004), values assigned to population exposure variables (Durmusoglu et al., 2010), and those from day-to-day or place-to-place variations in concentrations (Kim et al., 2002). Generally, because of the lack of data, risk analysis involves a series of estimations and assumptions. However, the estimations and assumptions could do not represent all individuals adequately and may lead to huge deviations (Kim et al., 2002). The uncertainties in values assigned to population exposure variables affect risk assessment, such as uncertainties in potency calculations. Cancer and noncancer risk from exposure to VOCs were determined from testing on animals, and therefore may not reflect the risk to humans (USEPA, 2009). The linear model used in risk calculation did not consider the low dose or high dose, beyond the threshold of pharmacokinetics testing. Therefore, the true risk may have been overestimated or underestimated (Guo et al., 2004).

In this study, the uncertainty in VOC measurement was calculated according to Eq. (5), and the range was from 5.1% to 12.9% with an average of 7.6%. Using 30-min average concentration to represent 24-h average concentrations ignores potential daily variations that could exert a marked influence on exposure over prolonged periods: the uncertainty could have been as high as 10% (Kim et al., 2002). Although the sampling sites were selected carefully, the limited indoor sampling environments may not accurately represent actual exposure levels to VOCs for the entire population in rural Guanzhong Plain; thus, the uncertainty of representativeness could reach 20%. The sampling campaign must be enlarged in the future to increase the representative accuracy of VOC levels by increasing sampling sites and extending sampling time. Parameters set in the noncancer and cancer risk models referred to statistical data and reasonable assumptions; uncertainty levels were both set at 20% (Zhang et al., 2007; Zhao et al., 2011). Therefore, the calculated uncertainty levels for noncancer and cancer risk estimation were 41.9% and 54.4%, respectively [according to Eq. (7)]. The cancer risk model introduced more parameters, thus presenting a higher level of uncertainty than the noncancer model. Statistical data and assumptions contributed most uncertainties, suggesting that the statistical and human activity data

should be improved to make the risk assessment more accurate and reliable.

# 5. Conclusion

Indoor VOC samples were collected in two typical kitchens and five representative bedrooms in rural Guanzhong to detect the characteristics of indoor toxic VOCs and the health risks from exposure to VOCs. Total toxic-VOC concentrations in the traditional kitchen using a wood stove were the highest  $(299 \pm 38.8 \, \mu g \, m^{-3})$ , and these were reduced by approximately 40% in the LPG kitchen  $(187 \pm 54.6 \,\mu g \,m^{-3})$ . The reduction was mainly from the low emission of benzene from LPG combustion. Toxic-VOC concentrations in bedrooms using electric or plumbing systems for heating were lower than those using solid fuels; however, the difference was insignificant (p > 0.05). Correlation analysis indicated that indoor combustion crucially influenced indoor VOCs (R > 0.7). The correlations were not found when combustion sources were removed from indoor environment. The CD also indicated that the VOC profiles were more similar between indoor VOCs with similar combustion sources (CD = 0.27 and 0.29) compared with those between Bedrooms 2 and 4 (CD = 0.53 and 0.59, respectively). Exposure risk assessment showed that exposure to kitchen VOCs posed high noncancer risks (7600 and 2550 for the traditional and LPG kitchens, respectively), mostly for women. Noncancer risks in bedrooms ranged from 140 to 768, much higher than the safe threshold, indicating that indoor toxic-VOC concentrations in the heating season are severe and must be urgently improved in rural Guanzhong Plain. Cancer risk assessment results were similar to those of noncancer risk assessment. Cancer risks exposure to VOCs in kitchens for women were  $8.98 \times 10^{-4}$  and  $1.67 \times 10^{-4}$  from tree branch burning and LPG use, respectively-much higher than the USEPA threshold, and 1,3-butadiene was the VOC with the highest cancer risk in all indoor environments using combustion for cooking and heating. The cancer risks in bedrooms using the electric heater or plumbing system were lower than those burning solid fuel: however, neither of them fulfill the USEPA safety threshold. The uncertainty levels for noncancer and cancer risk assessment were 41.9% and 54.4%, respectively, which were unavoidable due to measurement and model building. However, the considerably high noncancer and cancer risks indicate that the indoor toxic VOCs during the heating season in rural Guanzhong might harm the health of the residents and urgently require strategies for limiting them.

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# Appendix A. Supplementary data

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