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Levels of volatile organic compounds in homes in Dalian, China

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Abstract This paper measured selected individual volatile organic compounds (VOCs), including formaldehyde, in residences in Dalian, evaluated the association between the apartment characteristics and VOC concentrations, and explored the associations between chemicals and sick building syndrome (SBS). Higher VOC concentrations were measured indoors than outdoors in summer (August to September) and winter (January to March) in Dalian, and there were no strong correlations between the indoor and outdoor concentrations of most VOCs. This indicates the dominance of indoor sources as compared to outdoor sources. Formaldehyde was the most abundant compound in this study, followed by toluene, benzene, xylene, and styrene. These pollutants increase the occurrence of SBS. Thus, the VOC levels in dwellings in Dalian should be regulated, in view of SBS risks.

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Introduction

With the advancement of modern construction technology, synthetic building materials and chemical products have been extensively used in new buildings. In addition, the air-tightness of buildings has been improved in order to save energy, and the exchange between outdoor fresh air and indoor air has been reduced. The low ventilation rate and increased sources of synthetic chemicals jointly lead to significant indoor air pollution. Moreover, people generally spend 20 out of 24 h in enclosed spaces (houses, offices, schools, public buildings, vehicles, etc.), and 70 % of indoor time is spent at home (Matz et al. 2014). Many health problems, such as allergies, asthma, sick building syndrome (SBS), and even cancer, are considered to be related to poor indoor air quality in both developing and developed countries (Tunsaringkarn et al. 2015). Therefore, indoor air pollution has become a major health problem all over the world. In particular, the volatile organic compound (VOC) concentrations in homes and their adverse effects on the health of residents have increasingly become a cause for concern.

VOCs have been recognised as one of the principal constituents of indoor air pollutants. A large variety of building materials, consumer products, and human activities can contribute to indoor levels of VOCs, including vinyl tiles, paints, cleaners, frying foods, smoking, dry-cleaned clothing, and photocopiers or printers (Shin and Jo 2013; Son et al. 2013; Steinemann 2015). Due to the numerous indoor sources, many VOCs are present indoors at concentrations substantially higher than outdoors.

Many reports have indicated that VOCs are the most important pollutants in an indoor environment and they are the main risk factors for adverse effects on a residents' health (Lee et al. 2014; Shin and Jo 2013). For example, benzene, styrene, tetrachloroethylene, etc. are mutagenic and/or carcinogenic. Thus, long-term exposure to these compounds is linked to an increased risk of cancer. Additionally, many indoor VOCs can cause sensory irritation and central nervous system symptoms. The level of total VOCs (TVOC) indoors has been used as an indicator of building healthiness, in view of the correlation of the prevalence rate of SBS symptoms or complaints with TVOC concentration (Godish 2000).

China has experienced rapid economic growth in the past three decades. Real estate in China is currently thriving. The massive construction of buildings has provided more and more common Chinese with new apartments. Decoration and refurbishment of apartments have become popular in urban China. VOCs are released from decorations and furniture and accumulate in indoor air. The indoor air of Chinese residences with abundant decorations and furniture may be heavily polluted by VOCs. However, thus far, information about VOCs in the indoor air of Chinese residences is still very limited. In particular, there have been few systematic investigations of VOC concentrations and the factors influencing them in indoor air in the northeast of China.

Dalian is a moderately large city in the northeast of China as well as an important commercial and industrial centre in China. In this paper, we measured indoor and outdoor air chemical concentrations and used questionnaires to gather information on residents' health complaints and apartment characteristics in summer and winter periods in Dalian. The main objectives of our study were to (1) quantify selected individual VOCs, including formaldehyde, in residences in Dalian; (2) evaluate the relations between the apartment characteristics and VOC concentrations; and (3) explore the relation between chemical substances and subjective symptoms.

Materials and methods

Sampling sites

Air sampling was conducted in Dalian, China (shown in Fig. 1). Dalian (latitude 38°55′ N, longitude 121°38′ E) has a temperate monsoon climate (average temperature of 11 °C) and about 6,690,000 inhabitants (2010) and is typical of a commercial city in China. The area surveyed is a typical neighbourhood consisting of approximately 2000 apartments located in an urban centre in Dalian. Sampling was conducted from August to September in 2007 for summer and from January to March in 2009 for winter. Fifty-three houses were sampled in summer and one hundred in winter.

Fig. 1 Map of China and the location of Dalian city, Liaoning Province

All participants volunteered for this study and signed the form indicating informed consent, which was approved by the Institutional Ethics Review Board of Dalian Medical University.

Sampling methods

Air samples were collected in the bedroom, kitchen, and outside for each surveyed apartment. A passive sampler was placed in the middle of the bedroom and kitchen, at a height of 1.2–1.5 m above the floors and a minimum distance of 0.5 cm to the neighbouring pieces of furniture. Formaldehyde was collected by a passive sampler packed with 2,4-dinitrophenylhydrazine (DNPH)-coated silica cartridges (Sep-Pak DNPH-Silica cartridge, Waters, Milford, MA, USA) in duplicate for 24 h. VOCs were collected by a passive sampler packed with activated charcoal (Sibata Chemicals, Co. Ltd, Tokyo, Japan) in duplicate for 24 h. The two passive samplers were simultaneously exposed in the same location. At the same time, travel blank samples were obtained to determine the sample contamination during travel to the laboratory.

The outdoor samplers were hung by strings from the window frames. They were faced away from exhaust ducts and heat sources of the apartment and protected from rain and direct solar irradiation. The temperature and relative humidity (RH) of indoor air were also measured with an Assman psychrometer. After sampling was completed, the samplers were stored at −4 °C in sealed aluminium bags.

Guo et al. (2009) have confirmed that the diffusive sampling rates of formaldehyde and VOCs provided by the manufacturers of the two types of diffusive samplers are appropriate for a sampling period of 24 h. The diffusive sampling rate of formaldehyde is 0.112 μg/ppm min, whilst those of the other VOCs range from 0.05 to 0.3 μg/ppm min.

Extraction and analytical

Formaldehyde and 12 VOCs that are widely monitored in indoor environments and suitable for passive sampling were selected for chemical analysis in this study. The sum of the concentrations of the 12 target VOCs was defined as TVOC. The formaldehyde and VOC samplers were then transported to Japan by air for analysis as soon as possible. The analysis was done at the Nagoya City Public Health Research Institute. Formaldehyde was eluted with 3 mL of acetonitrile before analysis with a high-performance liquid chromatograph (HPLC, Hewlett Packard Series 1100, Hewlett Packard, Palo Alto, CA, USA) equipped with a photodiode array detector (Hewlett Packard, USA). Chromatic separation was performed using solvent gradient elution at a flow rate of 0.2 mL/min in an analytical column (Hypersil ODS; 250 mm, 2.1 mm i.d., 5 μm; Agilent Technologies, Palo Alto, CA, USA). The mobile phase was acetonitrile and distilled water. The column was kept a constant temperature of 40 °C, using a thermostat. The detection wavelength was 360 nm.

VOCs were extracted with carbon disulphide and analysed by a gas chromatograph equipped with a mass spectrometer (GC-MS). The GC-MS was equipped with a 60 × 0.25 mm i.d. capillary column coated with a 1.5-μm film of NB-1 (GL Sciences, Tokyo, Japan). First, the GC oven temperature was maintained at 45 °C for 5 min, then programmed to 300 °C at a speed of 10 °C/min, and held at 300 °C for 7 min. The analysis was performed under the selected ion monitoring mode at a helium flow rate of 0.9 mL/min to the 12 target chemicals.

Quality assurance

A calibration standard curve of formaldehyde was firstly prepared within the concentration range of 0.005 to 1.0 μg/mL, which showed a good linearity ($r > 0.99$). A calibration standard curve of 12 VOC was also prepared, respectively, which showed a good linearity ($r > 0.99$) within the concentration range of 0.125 to 8.0 μg/mL. Quality control samples were analysed with calibration standards on 6 days (intra- and inter-day precision and accuracy). Precision was evaluated by calculating coefficients of variations (% CV) from the measured concentrations. Percent relative error (% RE) was calculated as a measure of accuracy. The precision was better than 5 %, and the accuracy did not exceed 10 % at all concentrations of quality control samples.

Questionnaire study

We distributed self-administered questionnaires during home visits for environmental monitoring. The questionnaire included questions on personal characteristics (e.g. gender, age, smoking habits, time spent at home, alcohol habits, history of allergies), on apartment characteristics (e.g. building date, apartment area, materials used to construct the floor and paint the walls, and the presence or absence of plants in the apartment), and on subjective symptoms. The subjective symptoms consisted of optical symptoms (eye irritation), nasal symptoms (rhinitis, blocked nose, and sneezing), gular symptoms (hoarseness, dry throat, coughing, and wheezing), dermal symptoms (itching, dryness, and erupted skin), and general symptoms (fatigue, feeling heavy-headed, headache, nausea, dizziness, and difficulty concentrating). A representative of the household answered the questions about the characteristics of the apartment. Other occupants of each dwelling answered the questions about subjective symptoms and health problems. If the participants could not read or write, another family member answered the questionnaires for them. Participants were asked to refer to their health complaints during the last 3 months in choosing responses to each symptom: occurring ≥3 times per week (always), occurring once or twice per week (sometimes), and never occurring. Participants were also asked whether their symptoms could be attributed to the home environment. Any "always" or "sometimes" symptoms related to the home environment were defined as positive SBS symptoms. Those who complained about more than one positive SBS symptom were classified as suffering from SBS.

Statistical analysis

VOC concentrations below LOD were substituted by the LOD/2 to estimate the means and standard deviations. Because VOC concentrations were approximately log normally distributed, their average concentrations were calculated as geometric means. The Mann–Whitney *U* test was performed to test the differences between sample sites or seasons. Spearman's correlation coefficient was calculated to assess the relationship between the concentrations in different sites. To identify the apartment characteristics affecting VOC concentrations, multiple linear regression analysis was conducted. Logistic regression analysis was conducted to determine the relation between SBS and VOCs, and adjusted odds ratios of VOCs were estimated. All analyses were performed with SPSS 13.0 for Windows (SPSS Inc., Chicago, IL, USA).

Results

Characteristics of apartments and inhabitants

Data from 247 individuals living in 153 dwellings were included in this analysis. Table 1 shows that 19.43 % of subjects suffered from SBS. All 153 dwellings were concrete apartments that were <8 years old (mean = 3.39 years old), and

Table 1 The characteristics of inhabitants and their apartments

No.	Item	[Code] levels	*N* (%)
Inhabitants characteristics (*N* = 247)			
1	What is your sex?	[1] Male	114 (46.15)
		[2] Female	133 (53.85)
2	How old are you?	[1] <20 years old	7 (2.83)
		[2] ≥20 and <39 years old	56 (22.67)
		[3] ≥40 and <59 years old	137 (55.47)
		[4] ≥60 years old	47 (19.03)
3	Do you drink alcohol (≥1/week)?	[0] No	209 (84.62)
		[1] Yes	38 (15.38)
4	Do you smoke cigarettes now?	[0] No	173 (70.04)
		[1] Yes	74 (29.96)
5	How many hours do you spend per day in your apartment on average?	[1] <8 h	40 (16.19)
		[2] ≥8 and <16 h	169 (68.43)
		[3] ≥16 and ≤24 h	38 (15.38)
6	Do you have more than any one symptom of SBS?	[0] No	199 (80.57)
		[1] Yes	48 (19.43)
7	Do you have any allergies?	[0] No	222 (89.88)
		[1] Yes	25 (10.12)
Apartment characteristics (*N* = 153)			
8	How old is your apartment?	[1] <1 year	32 (20.92)
		[2] ≥1 and <2 years	28 (18.30)
		[3] ≥2 and <3 years	24 (15.69)
		[4] ≥3 years	69 (45.09)
9	What is the living space of your apartment?	[1] <60 m^2	42 (27.45)
		[2] ≥60 and <120 m^2	78 (50.98)
		[3] ≥120 and <180 m^2	33 (21.57)
10	How many hours does your apartment ventilate per day?	[1] 0 h	14 (9.15)
		[2] ≥0 and <12 h	123 (80.39)
		[3] ≥12 h	16 (10.46)
11	Do you have any plants in your apartment?	[0] No	51 (33.33)
		[1] Yes	102 (66.67)
12	How is your apartment decorated?	[1] Simple decoration	45 (29.41)
		[2] Complex decoration	108 (70.59)
13	What kind of coating was used to paint the interior walls?	[1] Water-based coating	91 (59.47)
		[2] Solvent-based coating	38 (24.84)
		[3] Others	24 (15.69)
14	What kinds of floor materials are used in your apartment?	[1] Wooden floor	75 (49.02)
		[2] Plywood floor	51 (33.33)
		[3] Others	27 (17.65)
15	What kind of door material is used in your apartment?	[1] Wooden door	65 (42.48)
		[2] Plywood door	63 (41.18)
		[3] Steel door	25 (16.34)

their living rooms had wood or plywood flooring. The number of inhabitants in each dwelling was 1.8 ± 1.0 and ranged from 1 to 3 persons. The mean of the living space was 68 m^2 (ranged from 48 to 194). Over one half (70.59 %) of the surveyed dwellings had been decorated complexly. The average indoor temperatures ± SD for 2007 and 2009 were 22.8 ±

2.8 and 23.4 ± 3.3 °C, respectively. The average indoor humidities ± SD for 2007 and 2009 were 58.7 ± 8.4 and 56.2 ± 7.7 %, respectively.

VOC concentrations

In summer, formaldehyde, m,p-xylene, and toluene were detected in 100 % of the investigated apartments; benzene, o-xylene,1,2-dichloroethane, n-hexane, and butyl acetate were all detected in above 90 %, whilst styrene was detected in only approximately 30 %. In winter, the detection rates were as high as in summer (Table 2).

In summer, TVOCs ranged from 61.10 to 405.60 μg/m^3, with averages of 93.10 μg/m^3 in the bedroom and 90.46 μg/m^3 in the kitchen. For outdoor sites, they were in the range of 40.22–140.74 μg/m^3, with an average of 77.66 μg/m^3. Thus, there is a significant difference between indoor and outdoor values ($P < 0.05$). Amongst the target VOCs in summer, formaldehyde was the most abundant in the bedrooms in all surveyed sites, followed by toluene, n-butanol, and m,p-xylene (Table 2). For most VOCs, higher concentrations of VOCs were detected in bedrooms than in kitchens although the difference was not statistically significant, except for styrene, 1,1,1-trichloroethane, and butyl acetate in summer. Generally, most indoor VOC levels were significantly higher than those outside ($P < 0.05$). However, only four chemicals (p-dichlorobenzene, 1,2-dichloroethane, 1,1,1-trichloroethane, and carbon tetrachloride) showed no significant differences between indoor and outdoor concentrations.

In winter, the average concentrations of TVOCs in bedrooms, kitchens, and outdoors were 120.27, 116.23, and 75.56 μg/m^3, respectively; the difference between indoor and outdoor concentrations was significant ($P < 0.05$). As shown in Table 2, formaldehyde was also the most abundant pollutant in indoor air, followed by toluene, butanol, benzene, and 1, 2-dichloroethane. In short, most VOC levels in different rooms were almost the same in winter, but significantly higher than those outside ($P < 0.05$).

We also compared the indoor VOC concentrations during summer and winter. Figure 2 reveals that the concentrations of VOCs varied significantly with seasons. Overall, the indoor concentrations of most target VOCs are significantly higher in winter than those in summer ($P < 0.05$).

Significant correlations were observed between concentrations in bedrooms and kitchens, especially for formaldehyde, toluene, and m,p-xylene, which showed stronger correlations ($r = 0.790$, 0.791, 0.790) in winter (Table 3). Amongst indoor rooms, the concentrations in bedrooms were consistent with or slightly higher than concentrations in kitchen rooms (Table 2). Nevertheless, correlations between outdoor and indoor (kitchen or bedroom) concentrations are mostly weak even though some correlations are statistically significant. Toluene showed stronger correlations between outdoors levels and kitchen/bedroom levels in summer as well as in winter.

Association between apartment characteristics and VOC concentrations

The associations between the apartment characteristics and the log VOC concentrations were analysed by multiple linear regression analysis (Table 4). For the five VOCs, the factors documented in this study explained the variations of 30–40 % in concentrations in indoor residential air. The indoor VOC levels became lower as the apartments became older and the ventilation time increased. The plants in the apartments had a significant effect on the levels of formaldehyde and toluene, but not on those of benzene, o-xylene, and m,p-xylene. The decoration materials also influenced the concentrations of the five VOCs. For example, solvent-based coatings had a significant effect on the levels of formaldehyde ($P = 0.008$) and o-xylene ($P = 0.000$) relative to water-based coatings, whilst the plywood floor had a significant effect on the levels of benzene ($P = 0.030$). Amongst the eight factors, the ventilation time had the greatest influence on the indoor levels of formaldehyde ($\beta' = -0.320$), benzene ($\beta' = -0.317$), and toluene ($\beta' = -0.169$). The main factor influencing the levels of o-xylene and m,p-xylene was the apartment age ($\beta' = -0.300$ and $\beta' = -0.258$, respectively).

Health risks associated with each chemical substance

Forty-eight (19.43 %) subjects reported more than one SBS symptom (Table 1). After adjusting for other possible risk factors, most of these chemicals tended to increase the odds ratio (OR) of SBS symptoms significantly (Table 5). The highest OR of SBS symptoms was observed amongst those subjects exposed to m,p-xylene (adjustment OR = 1.562, 95 % CI: 1.385–1.823), followed by o-xylene (adjustment OR = 1.535, 95 % CI: 1.121–1.777), and benzene (adjustment OR = 1.488, 95 % CI: 1.104–2.005).

Discussion

In the present study, indoor concentrations of most target VOCs were higher in winter than in summer. This seasonal change of VOCs is consistent with the results of other studies (Bari Md et al. 2015). All homes surveyed utilised central heating in winter. Thus, the average indoor temperature in winter was not lower than that in summer. Additionally, some decorative materials or furniture (e.g. floors, wardrobes, and cupboards) were close to the heating radiators and thus emitted more VOCs in the heat. Such a scenario does not occur in summer. In addition, the ventilation time is an important factor. Schlink et al. (2010) concluded that the

Table 2 Summary of concentrations (μg/m^3) of target VOCs at each monitoring site in summer and winter

Season	VOCs	Bedroom					Kitchen					Outdoor				
		G mean	GSD	Min	Max	DR (%)	G mean	GSD	Min	Max	DR (%)	G mean	GSD	Min	Max	DR (%)
Summer $n = 53$	Formaldehyde	48.46^{b}	14.81	13.00	180.23	100.0	42.86^{b}	14.12	12.08	116.38	100.0	17.94	10.58	12.01	69.56	100.0
	Benzene	3.42^{b}	1.45	1.01	15.67	98.3	3.23^{b}	1.79	1.01	9.90	98.3	2.51	0.91	1.77	5.05	94.9
	Toluene	15.0	9.66	2.35	97.39	100.0	15.05	5.26	6.20	80.04	100.0	10.88	6.77	4.79	41.79	100.0
	o-Xylene	1.0	1.87	0.57	9.60	93.2	2.28^{b}	1.67	0.64	9.88	91.5	1.85	1.02	0.96	3.65	76.9
	m,p-Xylene	6.64^{b}	3.76	0.72	23.55	100.0	6.17^{b}	2.74	0.99	14.55	100.0	5.44	3.16	1.74	13.76	97.4
	Styrene	$1.61^{b, d}$	0.44	0.79	2.33	30.2	1.35	0.47	0.97	2.84	28.3	1.34	0.82	1.22	2.45	19.9
	p-Dichlorobenzene	1.74	2.27	0.53	11.40	52.5	1.68	2.22	0.52	59.0	49.1	1.98	4.51	1.72	8.5	9.0
	1,2-Dichloroethane	1.67	1.50	0.59	65.39	93.2	1.68	1.46	0.55	6.87	96.6	1.82	2.53	0.77	2.41	84.6
	1,1,1-Trichloroethane	0.84^{c}	0.45	0.52	2.95	66.4	0.79	0.18	0.48	1.07	67.8	0.74	2.35	0.59	0.78	59.0
	Carbon tetrachloride	1.18	1.39	0.39	10.03	72.9	1.48	1.68	0.43	16.06	78.0	1.43	5.36	0.59	22.27	74.4
	n-Hexane	1.85^{b}	1.11	0.55	6.25	94.9	1.85^{b}	1.09	0.67	7.99	94.9	1.71	0.92	1.01	4.26	97.4
	Butyl acetate	$4.16^{b, d}$	2.44	1.19	27.34	96.6	3.73	1.65	1.09	8.99	96.6	3.62	2.23	1.22	9.93	87.2
	n-Butanol	9.91^{b}	4.17	4.78	65.17	89.8	9.51^{b}	4.04	4.54	20.85	94.9	7.56	1.83	5.28	11.63	59.0
	TVOC	93.10^{b}	32.62	61.10	400.50	–	90.46^{b}	30.58	62.34	405.60	–	77.66	26.47	40.22	140.7	–
Winter $n = 100$	Formaldehyde	58.37^{b}	14.81	10.78	178.13	100.0	56.25^{b}	18.78	45.06	196.23	100.0	10.68	8.30	13.92	61.98	100.0
	Benzene	11.37^{b}	3.66	2.53	29.55	98.0	8.85^{b}	3.80	5.94	10.91	98.0	3.89	2.82	2.53	11.16	98.0
	Toluene	18.73^{b}	6.97	4.83	89.89	100.0	18.18^{b}	5.06	5.73	83.55	100.0	7.08	10.56	3.49	12.78	100.0
	o-Xylene	2.89^{b}	1.72	0.24	14.63	92.0	2.45^{a}	1.89	1.19	3.15	91.0	1.39	6.69	0.58	2.47	77.0
	m,p-Xylene	8.64^{b}	2.12	1.17	18.72	100.0	7.07^{b}	3.55	1.77	29.54	100.0	2.09	5.65	0.54	4.87	87.0
	Styrene	3.48^{b}	2.93	0.35	11.44	32.0	2.73^{b}	1.65	1.52	8.28	33.0	1.45	3.56	1.23	1.68	38.0
	p-Dichlorobenzene	2.85	0.79	0.82	16.68	49.0	2.74	2.84	1.41	19.07	45.0	2.56	6.59	0.98	1.89	10.0
	1,2-Dichloroethane	$9.36^{b, d}$	1.69	0.40	41.85	97.0	2.49	1.38	1.42	5.87	98.0	1.56	1.18	1.48	1.67	83.0
	1,1,1-Trichloroethane	1.21	0.41	1.17	1.31	61.0	1.18	1.36	1.16	1.19	62.0	1.30	4.46	0.56	1.30	67.0
	Carbon tetrachloride	1.37^{d}	1.41	0.82	2.42	71.0	1.24	0.43	0.92	2.66	74.0	1.32	4.76	0.91	2.00	72.0
	n-Hexane	3.87^{b}	2.42	1.08	20.15	94.0	3.70^{b}	2.48	1.61	9.24	94.0	1.39	1.38	1.29	1.48	94.0
	Butyl acetate	4.44	3.15	0.87	62.75	96.0	4.05	1.91	1.54	6.08	96.0	4.25	6.56	3.56	5.12	90.0
	n-Butanol	$14.08^{b, d}$	5.34	7.16	30.16	91.0	12.03^{b}	3.13	4.24	18.88	91.0	9.10	2.90	6.64	12.56	41.0
	TVOC	120.2^{b}	40.56	52.63	480.56	–	116.23^{b}	39.75	69.86	420.55	–	75.56	23.64	41.22	160.7	–

Comparing the VOCs concentrations with these outdoor (Mann–Whitney *U* test): $^{a}P < 0.05$, $^{b}P < 0.01$. Comparing the VOCs concentrations between bedroom and kitchen (Mann–Whitney *U* test): $^{c}P < 0.05$, $^{d}P < 0.01$

DR detection rate

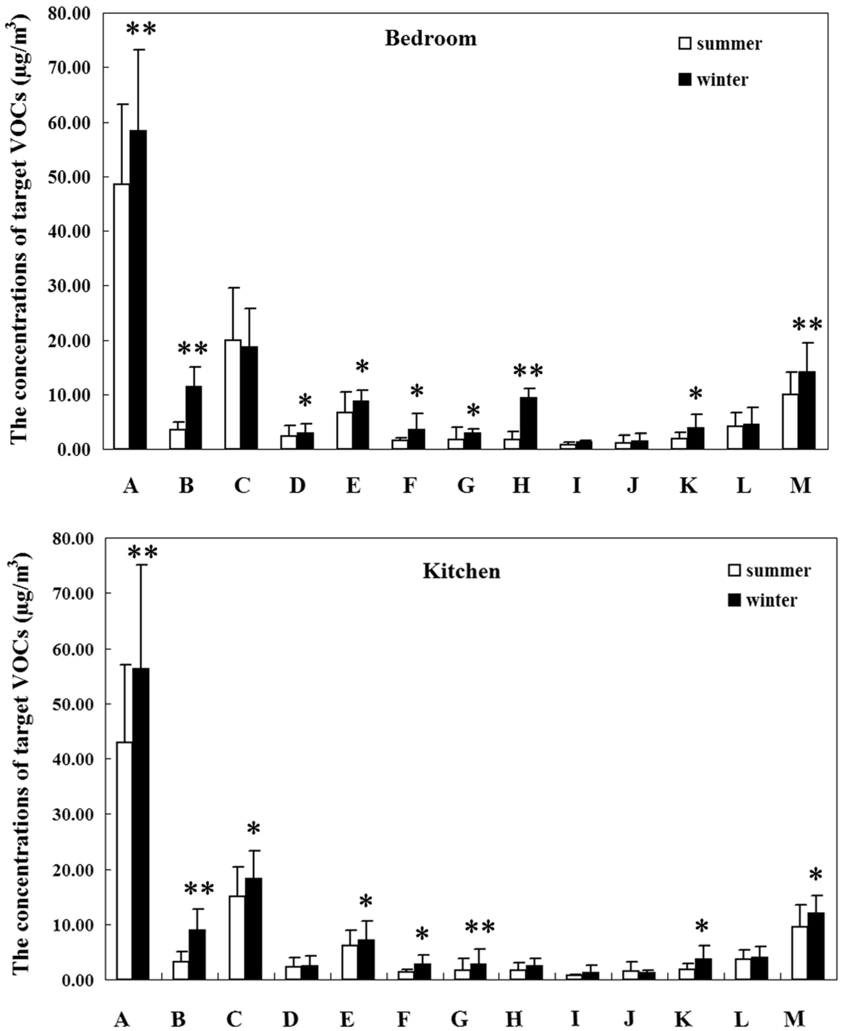

Fig. 2 Comparison of average concentrations of target VOCs in indoor air between summer and winter. *A* Formaldehyde, *B* benzene, *C* toluene, *D* o-xylene, *E* m,p-xylene, *F* styrene, *G* p-dichlorobenzene, *H* 1,2-dichloroethane, *I* 1,1,1-trichloroethane, *J* carbon tetrachloride, *K* n-hexane, *L* butyl acetate, *M* n-butanol ($P < 0.05$ (*asterisk*) and $P < 0.01$ (*two asterisks*), statistically significant for summer vs. winter)

season is the most important modifying factor influencing the concentration of each VOC component in indoor air. Seasonality is identified as a proxy for the ventilation behaviour of the inhabitants. Because of the low temperature in northeast China in winter, the windows of rooms were kept closed for longer periods to maintain thermal comfort. This may have contributed to the greater accumulation of VOCs indoors in winter.

We found that most VOC concentrations in bedrooms were similar to or higher than those in kitchens in summer as well as in winter. However, this does not mean that cooking behaviours do not influence VOC levels. Firstly, there is usually one kitchen ventilator in the kitchens in our surveyed apartments. When cooking, the equipment is operated and removes cooking oil fumes, due to which cooking does not cause a significant increase of VOC levels in the kitchen. Secondly,

Table 3 Correlations between the concentrations of five compounds measured outdoors, in the kitchen, and in the bedroom, separately for summer and winter

VOCs	r_s in summer			r_s in winter		
	B vs. K	B vs. O	K vs. O	B vs. K	B vs. O	K vs. O
Formaldehyde	0.592**	0.278*	0.294**	0.790**	0.189	0.020
Benzene	0.247*	0.251	0.532**	0.381**	0.206	0.198
Toluene	0.762**	0.744**	0.679**	0.791**	0.637**	0.514**
o-Xylene	0.555**	0.468**	0.256	0.491**	0.155	0.095
m,p-Xylene	0.742**	0.571**	0.370	0.790**	0.198	0.066

B bedroom, *K* kitchen, and *O* outdoors

*$P < 0.05$ and **$P < 0.01$

Table 4 Associations between 5 main logs of VOC concentrations and apartment characteristics: multiple linear regression

Pollutants	Variables	Standardised coefficient (β')	*P* value
Formaldehyde $R^2 = 0.402$	Apartment age	−0.352	0.000
	Apartment size	−0.042	0.610
	Ventilation time	−0.320	0.000
	Indoor plants	−0.238	0.007
	Degree of decoration	−0.108	0.190
	Coating material		
	Solvent-based coating	0.243	0.008
	Others	0.023	0.253
	Water-based coating		
	Floor material		
	Plywood floor	0.048	0.248
	Others	−0.014	0.156
	Wooden floor		
	Door material		
	Plywood door	0.055	0.505
	Steel door	0.038	0.264
	Wooden door		
Benzene $R^2=0.398$	Apartment age	−0.162	0.098
	Apartment size	−0.267	0.067
	Ventilation time	−0.317	0.001
	Indoor plants	−0.011	0.913
	Degree of decoration	−0.144	0.143
	Coating material		
	Solvent-based coating	0.031	0.767
	Others	0.013	0.800
	Water-based coating		
	Floor material		
	Plywood floor	0.209	0.030
	Others	0.089	0.252
	Wooden floor		
	Door material		
	Plywood door	0.102	0.270
	Steel door	0.097	0.325
	Wooden door		
Toluene $R^2=0.350$	Apartment age	−0.026	0.797
	Apartment size	−0.039	0.703
	Ventilation time	−0.169	0.097
	Indoor plants	0.235	0.035
	Degree of decoration	−0.031	0.758
	Coating material		
	Solvent-based coating	0.019	0.934
	Others	0.013	0.791
	Water-based coating		
	Floor material		
	Plywood floor	0.068	0.495
	Others	0.059	0.216
	Wooden floor		
	Door material		
	Plywood door	0.166	0.108
	Steel door	0.087	0.244
	Wooden door		
o-Xylene $R^2=0.337$	Apartment age	−0.300	0.002
	Apartment size	0.049	0.579
	Ventilation time	−0.167	0.064
	Indoor plants	0.012	0.891
	Degree of decoration	0.048	0.597
	Coating material		
	Solvent-based coating	0.388	0.000
	Others	0.045	0.554
	Water-based coating		
	Floor material		
	Plywood floor	0.062	0.478
	Others	0.039	0.192
	Wooden floor		
	Door material		
	Plywood door	0.068	0.466
	Steel door	0.017	0.775
	Wooden door		
m,p-Xylene $R^2=0.325$	Apartment age	−0.258	0.005
	Apartment size	−0.006	0.944
	Ventilation time	−0.030	0.745
	Indoor plants	−0.011	0.906
	Degree of decoration	0.057	0.551
	Coating material		
	Solvent-based coating	0.361	0.000
	Others	0.145	0.393
	Water-based coating		
	Floor material		
	Plywood floor	0.062	0.478
	Others	0.039	0.192
	Wooden floor		
	Door material		
	Plywood door	0.096	0.301
	Steel door	0.054	0.482
	Wooden door		

there are more emission sources of VOCs in bedrooms than in kitchens. In our surveyed apartments, the bedrooms were decorated with more types of decoration materials (floors, wardrobes, beds, and doors made of composite wood and walls painted with latex paint). In comparison, the kitchens were decorated simply. Their floors and walls were covered by tiles, and there was only a small number of cupboards. On the other

Table 5 Odds ratios of the relationships between VOC concentrations and subjective symptoms of SBS

Independent variables	SBS		
	Adjustment odds ratio	95 % CI	
Formaldehyde	1.127	1.039	1.134
Benzene	1.488	1.104	2.005
Toluene	1.139	1.022	1.272
o-Xylene	1.535	1.121	1.777
m,p-Xylene	1.562	1.385	1.823
Styrene	1.203	1.078	1.505
p-Dichlorobenzene	1.156	1.085	1.356
1,2-Dichloroethane	0.991	0.893	1.100
1,1,1-Trichloroethane	0.944	0.840	1.062
Carbon tetrachloride	1.151	0.937	1.298
n-Hexane	1.231	0.781	1.941
Butyl acetate	1.256	0.855	1.495
n-Butanol	0.981	0.840	1.295

Odds ratios are for 1-U increase in the level of exposure and were computed by binary logistic regression analyses. Odds ratios are adjusted for gender, age, alcoholic drinks (≥1/week), current smoking, time spent at home (h/day), and allergies

CI confidence interval, *SBS* sick building syndrome

hand, VOCs with high liquidity, such as formaldehyde, could easily spread from room to room.

Indoors, especially in bedrooms, the concentrations of formaldehyde, benzene, toluene, o-xylene, m,p-xylene, and styrene were markedly higher than those observed outdoors in summer or winter. The indoor and outdoor concentrations of VOCs showed weaker associations, especially in winter, except for toluene. This may indicate additional indoor sources and/or accumulation of such indoor pollutants. Consequently, emission sources could be present in each indoor room rather than outdoors for most VOCs. Outdoor VOCs did not represent a dominant contribution to indoor levels, especially in cold weather, because of the lack of ventilation. The strength of sources of indoor emissions is a stronger influence than the infiltration of outdoor air for many pollutants (Yoon et al. 2011). There are numerous VOC sources indoors, including building materials and furnishings (e.g. particle board furniture, flooring, and carpets), cleaning products, and solvents (Steinemann 2015). Additionally, VOC sources are associated with many routine indoor activities, such as cooking, cleaning, painting, renovating, and smoking (Brown et al. 2015). Indoor air pollution gets worse as a result of the decreasing air change rate due to energy-saving measures in winter, which cause the accumulation of pollutants originating indoors (Langer and Bekö 2013). As a consequence, indoor concentrations of VOCs generally exceed the outdoor burden.

Amongst the indoor chemicals monitored, the concentration of formaldehyde was the highest. Its average levels in bedrooms and kitchens were 48.46 and 42.86 μg/m^3, respectively, in summer. On the other hand, in winter, the average concentrations were 58.37 and 56.25 μg/m^3, respectively, which were much higher than the average of 18.7 μg/m^3 in Japan (Ohura et al. 2006) or between 13 and 37 μg/m^3 in Finland (Jarnstrom et al. 2006) but lower than the average of 85.7 μg/m^3 in Hong Kong (Guo et al. 2009) and similar to the value of 50 μg/m^3 in Harbin, China (Zhu and Liu 2014). The differences amongst these concentrations may be due to differences in building ages, decoration complexity, the quality of furniture, and ventilation time. It was also found that the average indoor formaldehyde level in Dalian was lower than the exposure limit (100 μg/m^3) of WHO, China and Japan (Azuma et al., 2005). Nevertheless, formaldehyde is a known animal carcinogen and is classified as probably carcinogenic to humans (group 2A) by the International Agency for Research on Cancer. The indoor air quality guideline value for formaldehyde, published by Flanders, is only 10 μg/m^3 (Stranger et al. 2007). Thus, the indoor average formaldehyde level in Dalian was relatively high from the point of view of public health.

Consistent with the results of previous studies, formaldehyde was the most abundant compound amongst the VOCs measured in indoor air (Sofuoglua et al. 2011; Takigawa et al. 2012). This implies that it has strong indoor sources, such as particle board or plywood furniture, containing formaldehyde-based resins. The results of the multiple linear regression analysis showed that apartment age, ventilation time, indoor plants, and solvent-based coatings are the major factors determining the level of formaldehyde indoors. The older the flat is, the lower the indoor level of formaldehyde is. Longer ventilation times and the presence of plants inside contribute to reduced levels of formaldehyde indoors. However, the use of solvent-based coatings could result in a higher level of formaldehyde indoors. According to Guo et al. (2009), the level of formaldehyde was higher in newly built homes and decreased with the age of buildings. The levels of formaldehyde were expected to be high initially, with gradual decrease because of reduced material emissions (Shin and Jo 2014).

In addition to formaldehyde, the other abundant and frequently found VOCs were toluene, benzene, m,p-xylene, o-xylene, and xtyrene. Benzene, toluene, and xylenes (BTX) are of particular interest due to their known carcinogenic effects. The average level of toluene in bedrooms was 19.94 μg/μm^3 in summer and 18.73 μg/μm^3 in winter, and its detection rate was 100 %. As per recent reports, the mean toluene concentration indoors is 7.7 μg/m^3 in Japan, 14.5 μg/m^3 in Taiwan, 15.3 μg/m^3 in Hong Kong, 26.47 μg/m^3 in Canada, and 34.7 μg/m^{3}in Korea (Guo et al. 2009; Héroux et al. 2008). Although these toluene levels do not exceed the exposure limit (260 μg/m^3) set by WHO and Japan or the limit (200 μg/m^3) set by China, toluene pollution cannot be ignored because of its carcinogenic risks.

The average concentrations of benzene in dwellings in Dalian are approximately 3 μg/m^3 in summer and 11 μg/m^3 in winter, which are somewhat higher than the guideline values for benzene (2 μg/m^3) in indoor air set by the Flanders region in Belgium but far lower than the guideline in China (90 μg/m^3). The Chinese limit seems to be met commonly in our survey; however, the existence of concentrations at these levels would suggest carcinogenic risks 2 to 3 orders of magnitude higher than the commonly acceptable risk of 10^{-6} (Sarigiannis et al. 2011). Thus, the Chinese limit is not protective enough for chronic exposure to benzene from the point of view of public health (Sarigiannis et al. 2011). Therefore, the concentration of benzene in our dwellings should be further reduced in order to avoid its carcinogenic risks.

Similar to formaldehyde, the indoor concentrations of all of these compounds, either in summer or winter, were generally higher than those measured outdoors. The indoor and outdoor concentrations showed weak associations, except for toluene. These results indicate that these compounds have strong indoor sources. Toluene, benzene, and xylenes have all been used as solvents in a variety of household products, such as paints, thinners, cleaning agents, coatings, nail polish and other cosmetics, adhesives, resins, and printing products (Wang et al. 2014). Styrene is a very widely used VOC. In household products, styrene-butadiene rubber (SBR) and styrene-butadiene latex are the main materials that emit styrene. SBR is used in almost all carpets that have a synthetic backing, rendering them a significant source of styrene indoors. However, the association between indoor and outdoor toluene concentrations was stronger in summer. Therefore, indoor toluene may come from additional outdoor pollutant sources as well as indoor sources. It is well known that toluene in outdoor air is associated with vehicle emissions and industrial activities (Bauri et al. 2016).

These results show that the ventilation time and plywood floor are major factors affecting the level of benzene indoors. Longer ventilation times contributed to reduced levels indoors. However, the use of plywood doors could result in a higher level of benzene indoors. Indoor plants can help to eliminate toluene in air. The apartment age and solvent-based coatings can have an impact on the level of xylenes indoors, similar to their impact on the formaldehyde level. Considering the influence of the above factors on the concentrations of these pollutants, firstly, in view of an emission control strategy, the selection of low-emission materials is more beneficial to indoor air quality. Secondly, inhabitants should allow a period of time for ventilation after dwellings furnished with materials of decoration, before moving into the apartment. Mechanical ventilation systems or higher ventilation rates are without a doubt beneficial to indoor air quality, but Park and Ikeda (2006) found that indoor-produced compounds in new homes will be more influenced by decreases of emission source strengths with time than ventilation systems.

In our study, 48 (19.43 %) subjects reported at least one SBS symptom. Although the definition of SBS differed amongst studies, other researchers have reported the prevalence of SBS to be between 3.0 and 23.3 % of the studied population (Kubo et al. 2006; Takigawa et al. 2012). The prevalence of SBS amongst participants in our study was in accordance with this range. The subjective symptoms observed in the present study could be attributed to indoor chemicals. After adjusting for other possible risk factors, most of the chemicals tended to increase the OR of SBS symptoms significantly. Formaldehyde, benzene, toluene, xylene, and styrene were all associated with the occurrence of SBS. Because many of these compounds coexist in normal building air, it is speculated that the effect of a mixture of pollutants on SBS should be more powerful than a single chemical although it is impossible and impracticable to isolate the clinical effects of a single substance. These VOCs influence sensory perception through irritation of the eyes and upper respiratory tract. For example, formaldehyde may cause irritation to the eyes, skin, nose, and throat, and higher formaldehyde levels are associated with general discomfort, lachrymation, sneezing, coughing, nausea, and dyspnea.

These results highlight the characteristics of VOCs in dwellings in Dalian, China, and significantly enhance our understanding of the factors which affect indoor concentrations of VOCs and the associations between VOCs and SBS. The findings can supply information for housing designers, builders, residents, and the government to improve indoor air quality by means of safe and environmental building materials and increased ventilation rate.

However, there are some limitations to this study. First, the size of the sample was relatively small, and the sample was only drawn from a typical neighbourhood located in an urban centre in Dalian. Thus, our findings cannot simply be generalised to the population of Dalian. Second, we only focused on the influence of characteristics of houses on VOC levels. In future research, we will consider the influence of occupants' behaviours (cooking, smoking, and the use of household products) on indoor VOC levels. Third, we did not measure the air exchange rate (AER) for an accurate evaluation of the role of ventilation in determining indoor air quality. Fourth, we analysed data using single-level logistic regression analysis, not multi-level analysis. Therefore, the results may increase the error of false positives, caused by neglecting intra-class correlations in our study.

Conclusions

Higher VOC concentrations were measured indoors than outdoors in summer or winter in Dalian, and there were no strong correlations between the concentrations of most VOC indoors and outdoors. Whilst this indicated the dominance of indoor sources, the effect of outdoor sources cannot be disregarded. Formaldehyde was the most abundant compound in this study, followed by toluene, benzene, xylene, and styrene.

These pollutants increase the occurrence of SBS. Although the VOC levels in dwellings in Dalian were below the exposure limits set by China or WHO, they should be regulated in view of SBS risks. Source removal is the most effective way to decrease chronic exposure to VOCs in existing homes. A higher ventilation rate is, without a doubt, beneficial to indoor air quality.

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Authors' contributions Guirong Song analysed the data and drafted the manuscript. Fengyuan Piao revised the manuscript. Aisong Yu and Peng Guo collected the samples. Kiyoshi Sakai, Md Khalequzzaman, Michihiro Kamijima, Tamie Nakajima, and Fumihiko Kitamura measured the samples. Kazuhito Yokoyama and Fengyuan Piao designed the study.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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