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Indoor Air Pollution Levels in Decorated Residences and Public Places over Xi'an, China

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ABSTRACT

To investigate the indoor air quality (IAQ) over Xi'an, the concentrations of volatile organic compounds (VOCs, including formaldehyde, benzene, toluene, o-xylene, p-xylene, n-butyl acetate, ethylbenzene, styrene, n-undecane, and total VOCs) in 471 residential rooms and 58 public rooms during 2014–2015 were determined. All the data were measured at a variety of 6–48 months after the decorations of these rooms. The results showed that formaldehyde was the most serious pollutant in almost all the monitored rooms. The concentrations of formaldehyde in residences and public places ranged from 0.02 mg m^{-3} to 0.45 mg m^{-3} and 0.05 mg m^{-3} to 0.32 mg m^{-3}, respectively. And the concentration levels in the 83.6% selected residences and 44.8% public places exceeded the Chinese National Indoor Air Quality Standard (GB/T 18883-2002) of formaldehyde value (0.1 mg m^{-3}). However, the TVOC concentrations in most sites were lower than the Chinese National Standard (GB/T 18883–2002) value. In residences, the formaldehyde and TVOC concentrations in bedrooms were slightly higher than those in living rooms and other rooms. The relationships among formaldehyde and TVOC concentrations with indoor temperature, relative humidity (RH), and decorative materials (curtain, wall decoration, wood floor, and panel furniture) were also investigated. Formaldehyde levels showed strong positive correlation with indoor temperature and RH. However, the TVOC levels had a relatively weak correlation with indoor air temperature and RH. The wall decoration and panel furniture were the main sources of indoor formaldehyde, while wood floor and panel furniture were the main sources of TVOC. In addition, indoor air pollution of three selected newly decorated houses with 11 rooms was monitored monthly for one year to evaluate the relationship between indoor pollution levels and ventilation time. It was found that the concentrations of formaldehyde and TVOC decreased with ventilation time, and the duration was one year after decoration especially after summer ventilation.

Keywords: Xi'an; Indoor air pollution; Formaldehyde; TVOC.

INTRODUCTION

Recent studies revealed that people spend more time inside buildings rather than outdoors, and the indoor concentrations of pollutants have already exceeded those of outdoors due to the presence of strong emission sources and lack of ventilation (Lee and Chang, 2000; Klepeis *et al.*, 2001; Lee *et al.*, 2001; Li *et al.*, 2001; Lee *et al.*, 2002a, b; Yrieix *et al.*, 2010; Jovanović *et al.*, 2014). Thus, the poor indoor air quality (IAQ) poses a heavy threat to human health. In these few years, concerns about IAQ have increased in developing countries like China, which has issued the relevant standards of IAQ since the Ninth Five-Year Plan (1996–2000), such as Indoor Environment Quality Evaluation Standard, constituted by the State Environmental Protection Administration and the Ministry of Health.

VOCs including benzene series, aliphatic hydrocarbon, polycyclic aromatic hydrocarbon, organic acid and others, are a group of major indoor air pollutants and the total volatile organic compounds (TVOC) has been widely used as an important index, which can better evaluate the overall IAQ (Brown., 2002). Exposure to VOCs has been associated with acute and chronic health problems, such as asthma and sick building syndrome symptoms (Madureira *et al.*, 2015; Mendes *et al.*, 2015; Huang *et al.*, 2016; Tham, 2016). Bocskay, *et al.* (2010) reported that prenatal exposures to carcinogenic polycyclic aromatic hydrocarbons (PAHs) were positively associated with stable chromosomal aberrations. The health effects of VOCs were shown detailed in Qian and Dai (2013). Formaldehyde is the most ubiquitous and typical indoor air pollutant, and was proved to be the

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carcinogen (Tao *et al.*, 2015), which can irritate the skin and the respiratory tract, leading to the occurrence of dysosmia, allergy, and immune dysfunction.

With the improved living standard, decoration, refurbishment activities, remodeling and new furniture have become very popular in houses, commercial offices and similar places. Harmful compounds emitted from decoration materials and new furniture tended to accumulate and cause potential adverse health effects (Hesaraki *et al.*, 2015; McGill *et al.*, 2015; Wang *et al.*, 2015; Parajuli *et al.*, 2016; Zorpas and Skouroupatis, 2016). Brown S K. (2002) reported that the presence of attached garages, site contamination and wool carpet contributed to the deterioration of IAQ. Also, the researcher found that the mass concentration of VOCs was one to two orders of magnitude lower in established dwellings than in new and renovated buildings. As for formaldehyde, the main indoor sources were building materials, furnishings and household products such as wood panels, resins, adhesives, and carpeting (Asere *et al.*, 2016; Chi *et al.*, 2016; Shang *et al.*, 2016).

Xi'an is the biggest city in Northwest China, with high population density caused by tourist and servicing industry development. And many districts are full of high-rise buildings near industrial premises and heavy traffic roads, resulting in deterioration and poor ventilation in these buildings. Therefore, it's crucial to determine the concentration of indoor VOCs and identify the potential influencing factors contributing to the VOCs emission. The objective of this study was to examine the indoor formaldehyde and VOCs pollution levels in residences and public places, discussing the current indoor air pollution status of Xi'an. The 9 compounds including formaldehyde, benzene, toluene, o-xylene, p-xylene, n-butyl acetate, ethylbenzene, styrene, and n-undecane were targeted, and TVOC was chosen as an important index of IAQ. The effect of indoor temperature, RH, decorative materials (curtain, wall decoration, wood floor, and panel furniture) on formaldehyde and TVOC levels were investigated also. Finally, the variations of indoor concentrations of formaldehyde and TVOC in newly decorated houses as a function of time were estimated, and some pollution prevention advices according to the findings were proposed.

MATERIALS AND METHODS

Samples Collection

The concentrations of formaldehyde and TVOC in 471 residential rooms and 58 public rooms were tested respectively from March 2014 to October 2015 in Xi'an. All the selected sites were newly decorated within 6–48 months. In addition, 3 newly decorated rooms were selected to monitor from March 2015 to March 2016 continuously.

Before sampling, the flow of constant current sampling apparatus was calibrated by a mass flow meter (TSI 4140). The stainless steel tubes were activated using N_2 by the thermal desorption unit for 1 h at 350°C before sampling, then both ends of these tubes were sealed with seal films and plastic caps. Moreover, all the monitored rooms were kept under normal conditions and closed for 12 h before sampling. Doors and windows were closed during the whole sampling process. During sampling, the gas tightness was examined and general descriptions including indoor temperature, RH, pressure, home type, decoration time, and decoration materials were recorded.

Formaldehyde samples were collected using a sampler (QC-2, Beijing Municipal Institute of Labour Protection) with bubble sampling tubes and then analyzed by the spectrophotometer (UV-2600AH, Unico Instruments). As for VOCs, samples were collected by the constant current samplers and stainless steel tubes filled with 200 mg Tenax-TA absorbents (60–80 mesh). The sampling apparatus was mounted on a tripod about 1.5 m above the ground level which was close to the breathing height. The samples were set at a flow rate of 0.5 L min^{-1} for 20 min. After sampling, formaldehyde samples would be kept refrigerated and unexposed to light, and the sampling stainless steel tubes for VOCs absorption were sealed tightly. Then, the samples were immediately transported to the laboratory for analysis. During indoor sampling, a control sample outside and a blank sample were measured also.

Analysis Methods

Formaldehyde samples were determined by phenol reagent spectrophotometry. Formaldehyde reacted with phenol reagent to generate azine. And then high-valence iron ions were added into the above liquid to form blue-green compounds in acid solution. Finally, the concentrations of formaldehyde samples were determined by colorimetric method. The details of reagent preparation and analytical methods are prescribed in the Chinese National Standard (GB/T 18204.26-2000). Also of note was that the sample bottles were allowed to stand for 15 minutes after the color developing agents added and should be heated if the ambient temperature was pretty low.

VOCs samples were determined by a gas chromatography with a flame ionization detector (FID) (FULI-9790), which include capillary column (SE-30, 50 m × 0.32 mm quartz capillary column) and thermal desorption unit (JX-3). The chromatographic conditions and analytical methods were detailed in the Chinese National Standard (GB 50325-2001). It was essential to test the saturated adsorption capacities of the absorbent under sampling condition, by putting two tubes together in a series and detecting their VOCs concentrations individually. Furthermore, the accuracy was detected by adding 10 µg multi-element standard solution of TVOC into the absorbent tube, and results showed that relative standard deviation of Tenax-TA was 1.2% which was in the range of the standard value.

RESULTS AND DISCUSSION

The Overall Pollution Levels of Formaldehyde and TVOC

The distribution of room quantities with different formaldehyde concentrations and TVOC concentrations were presented in Fig. 1. It was noted that formaldehyde levels in 424 rooms, which occupied 90.0% of the total detected residences and public rooms, had exceeded the standard values of the Chinese National Indoor Air Standard

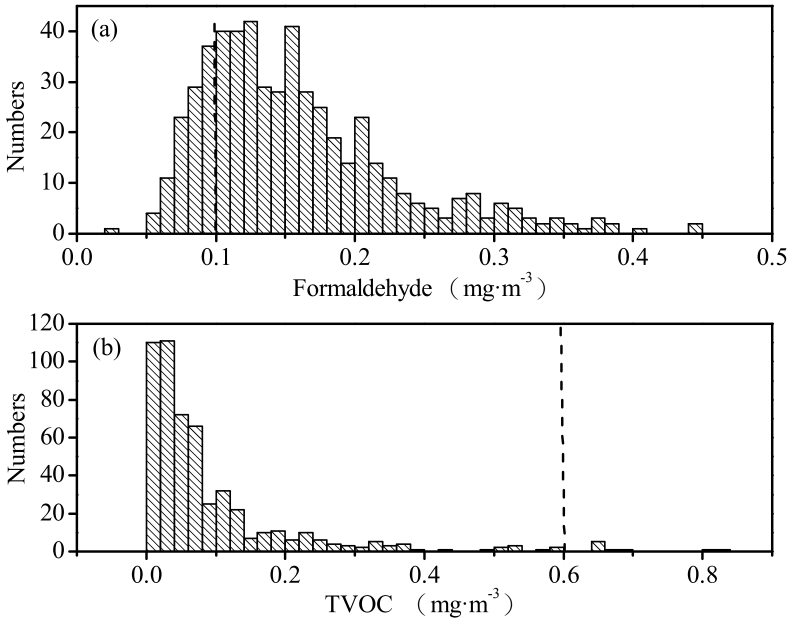

Fig. 1. The distribution of room quantities with different formaldehyde and TVOC concentrations. Note: The dashed lines in the graph represent the Chinese standard values for formaldehyde and TVOC concentrations individually.

(GB/T 18883-2002), World Health Organization (WHO) guidelines for indoor air quality (WHO, 2010), and Ventilation for Acceptable Indoor Air Quality of the United States of America (American Society of Heating Refrigerating and Air conditioning Engineer, ASHRAE, 2007). And their formaldehyde standard values are all 0.1 mg m^{-3}. In contrast, only 9 rooms exceeded the TVOC standard values of 0.6 mg m^{-3}. It was thus clear that indoor formaldehyde pollution in Xi'an was very serious in comparison with TVOC.

For the residences observation, the formaldehyde levels in 394 rooms were exceeded the Chinese standards, and the exceeding standard rate was 83.6%. While for TVOC, only 8 out of 471 rooms (1.7%) were detected to out of limits. The mean concentration of residences formaldehyde was a little higher than the Chinese National standard value, but the mean TVOC level was much lower than the standard value (Table 1). The formaldehyde concentration ranged from 0.02 mg m^{-3} to 0.45 mg m^{-3}, and the maximum concentration was 3.5 times higher than the Standard value. As for TVOC, the maximum concentration was 0.84 mg m^{-3}, which was 1.40 times higher than the Chinese National Standard (0.6 mg m^{-3}). It was noteworthy that n-butyl acetate, ethylbenzene, styrene, and n-undecane were the main pollutants in TVOC, with the exceeding standard rates of 11.6%.

The general indoor air pollution characteristics of 130 living rooms, 293 bedrooms and 48 other rooms (containing studies, bathrooms, cloakrooms and kitchens) were listed in Tables S1–S3. The most abundant and frequently found pollutants were formaldehyde, n-butyl acetate, ethylbenzene, styrene, and n-undecane in all the monitored rooms. The exceeding times of formaldehyde in almost rooms were within 3 times, which showed that the standard-exceeding phenomena of formaldehyde were very common. The benzene series, which derived from paints, coating on the surface of walls and furniture, were also very serious. The concentrations of benzene and toluene barely exceed the Chinese National Standard in all detected rooms, and the exceeding standard rates of benzene and toluene were the lowest among benzene series. In fact, the indoor pollution of dimethylbenzene was relatively serious. According to the Chinese National Standard, the content of benzene in oil paints cannot exceed 0.5%, while the content of toluene and dimethylbenzene always ranged from 10% to 40% actually (Ma *et al.*, 2007). Therefore, many paints were under the cover of "Non-benzene", but not really environment-friendly. The total concentrations of n-butyl acetate, ethylbenzene, styrene and n-undecane were relatively high, of which the exceeding standard rates were above 10%. These four pollutants have the similar sources which mainly exist in oil paints, and there are no specific regulations issued by the government to control their indoor concentrations. Particularly, styrene detected in this study was in the same level to that of the above three pollutants and was also the main composition of TVOC, which was different to the result of Ma *et al.* (2007). As a result, among the monitored residence rooms, the mean concentrations of formaldehyde and TVOC were highest in the bedrooms, where people sleep and spend most of their time. And this was followed by the living rooms and other rooms, but the difference was not obvious among the 3 types of rooms.

This study monitored 58 rooms in public places, including schools, offices, banks and other public rooms. The mean concentrations of formaldehyde and TVOC in these public sites were similar with the residences levels. The exceeding Standard rates for formaldehyde and TVOC were 44.8% and 1.7% respectively. As shown in Table 2, the maximum concentrations of formaldehyde and TVOC in public sites were 2.2 and 1.1 times higher than the Chinese National Standard values. In public places, the profile of the monitored

Table 1. Formaldehyde and VOCs concentrations and characters of samples exceeding the China standard in residences.

Pollutants	Concentrations /mg m^{-3}				Percentage of exceeding times /%			Total exceeding standard rate/%
	Min	Max	Mean	S.D	< 1	1–3	> 3	
Formaldehyde	0.02	0.45	0.16	0.07	60.51	22.51	0.64	83.66
Benzene	0	0.02	$8.48E^{-4}$	$1.55E^{-3}$	0	0	0	0
Toluene	0	0.22	0.02	0.02	0.21	0	0	0.21
Dimethylbenzene	0	0.33	0.03	0.04	1.49	0	0	1.49
N-butyl acetate Ethylbenzene Styrene N-undecane	$2.19E^{-4}$[a]	0.62	0.05	0.08	5.94	3.61	2.12	11.67
TVOC	$9.74E^{-4}$	0.84	0.10	0.13	1.70	0	0	1.70

[a] The summary of N-butyl acetate, Ethylbenzene, Styrene and N-undecane.

Table 2. Formaldehyde and VOCs concentrations and characters of samples exceeding the China standard in public places.

Pollutants	Concentrations /mg m^{-3}				Percentage of exceeding times /%			Total exceeding standard rate/%
	Min	Max	Mean	S.D	< 1	1–3	> 3	
Formaldehyde	0.05	0.32	0.13	0.07	30.91	14.55	0	45.46
Benzene	0	0.01	$1.14E^{-3}$	$1.50E^{-3}$	0	0	0	0
Toluene	$7.42E^{-4}$	0.15	0.02	0.02	0	0	0	0
Dimethylbenzene	0	0.15	0.02	0.04	0	0	0	0
N-butyl acetate Ethylbenzene Styrene N-undecane	$7.43E^{-4}$[a]	0.49	0.04	0.08	10.91	1.82	1.82	14.55
TVOC	$2.26E^{-3}$	0.66	0.08	0.12	1.82	0	0	1.82

[a] The summary of N-butyl acetate, Ethylbenzene, Styrene and N-undecane.

VOCs showed that formaldehyde was dominant, followed by n-butyl acetate, ethylbenzene, styrene, and n-undecane, which was similar with residential rooms. By comparing the pollution levels between residence and public sites, it was found that indoor air pollution in public places was slightly less serious than that in residences. It could be speculated as follows: firstly, the government paid more attention to public places with crowded population, which urged the managers of these places to put much more effort to improve the IAQ. Secondly, most public places we detected were schools, of which the decorated style was just cleanliness, simplicity and with little decorative materials. Finally, most public places may have larger space, and it's beneficial for dilution of air pollutant due to better ventilation. To better understand the pollution levels of all kinds of public places, further study is necessary.

The Effect of Indoor Temperature and RH on Formaldehyde and TVOC Levels

The influence of indoor temperature on the concentrations of formaldehyde and TVOC were shown in Fig. 2. It illustrated that formaldehyde concentrations were significantly and positively correlated with the indoor temperature, which indicated that high temperature was favored the formaldehyde vaporing from the paint or furniture. And the similar results were obtained by other literature (Tao *et al.*, 2015). While for TVOC concentrations, the influence of indoor temperature was not as significant as that of formaldehyde, although they shared the same trend.

Fig. 3 revealed the effects of indoor RH on formaldehyde and TVOC. The detected RH ranged from 26.3% to 90% with an average value of 57.4%. Formaldehyde was water-soluble substances, with the rise of RH, weakly acidic water vapor in the ambient air could react with the dissociative dimethylurea oligomer contained in urea-formaldehyde resin adhesive, or accelerate the reaction process of methylurea with wood cellulose to generate formaldehyde, and promote the hydrolysis of urea-formaldehyde resin glue to liberate formaldehyde. Therefore, indoor RH was a pivotal factor determining formaldehyde concentrations. However, the TVOC concentrations had a weak correlation with RH. This was mainly because that only a small part of TVOCs were water soluble substances, which could dissolve into water, and promote the further release of themselves in return.

The Contribution of Decorative Materials on the Concentrations of Formaldehyde and TVOC

It is well recognized that the indoor concentrations of formaldehyde and TVOC are mainly associated with decorating and furniture's materials. Figs. 4 and 5 plotted the formaldehyde and TVOC levels in different decorate materials. It found that wall decorations and panel furniture played an important role on the concentrations of formaldehyde. The average concentration of formaldehyde in rooms without wall decoration and panel furniture were reduced by 21.3% and 10.2% in compared with rooms with

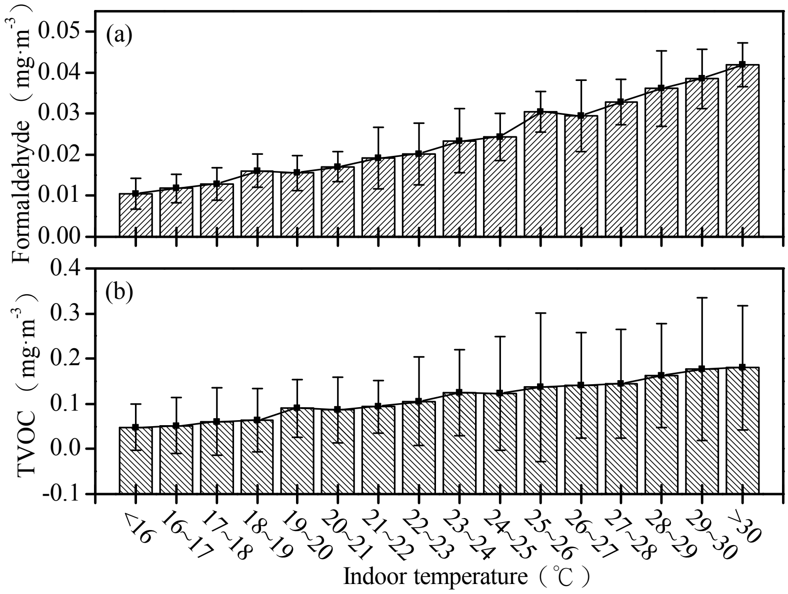

Fig. 2. Indoor temperature dependence of formaldehyde concentrations (a) and TVOC concentrations (b).

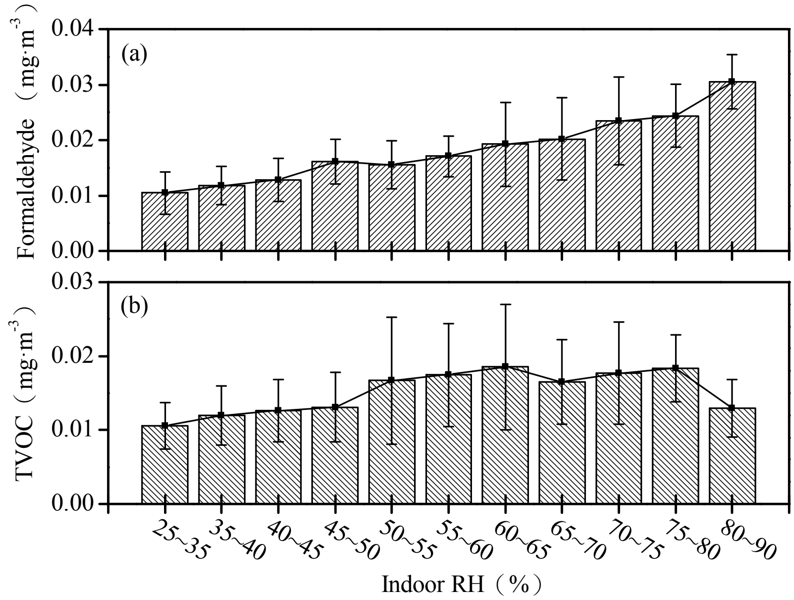

Fig. 3. Indoor relative humidity dependence of formaldehyde concentrations (a) and TVOC concentrations (b).

those materials. In fact, many adhesives containing Phenol-formaldehyde resin and urea-formaldehyde resin play an important role in the whole manufacturing process of the wall decorations and panel furniture, which could make contributions to formaldehyde emission. Chang *et al.* (2002) have reported the high emissions of formaldehyde from latex paints. Polyvinyl formal is one of the main adhesives in the white emulsion, which appear to cause large release of formaldehyde too. Wallpaper is known to be the main source for formaldehyde emission from wall covering (Suryawanshi *et al.*, 2016). The mucilage glue used to paste wallpaper, not the wallpaper itself, is the potential source of formaldehyde. In addition, wood floor and curtain had relatively less impact on the indoor concentrations of formaldehyde.

As for TVOC, wood floor and panel furniture were the dominant sources. The average concentration of TVOC in rooms no wood floor and panel furniture were lower by 15.7 and 28.2% than rooms with those materials. As we know, in the fabrication process of panel furniture and non-solid wood floor, plenty of organic reagents such as diluents, adhesives, waterproof agents and organic solvents are used. Moreover, oil paint on the surface of formed fitments and non-solid wood floor, which contains many types of TVOC such as benzene, toluene, and dimethylbenzene, lead to the significant increase of indoor TVOCs concentrations after decoration. Curtain and wall decoration had little effect on TVOC concentrations.

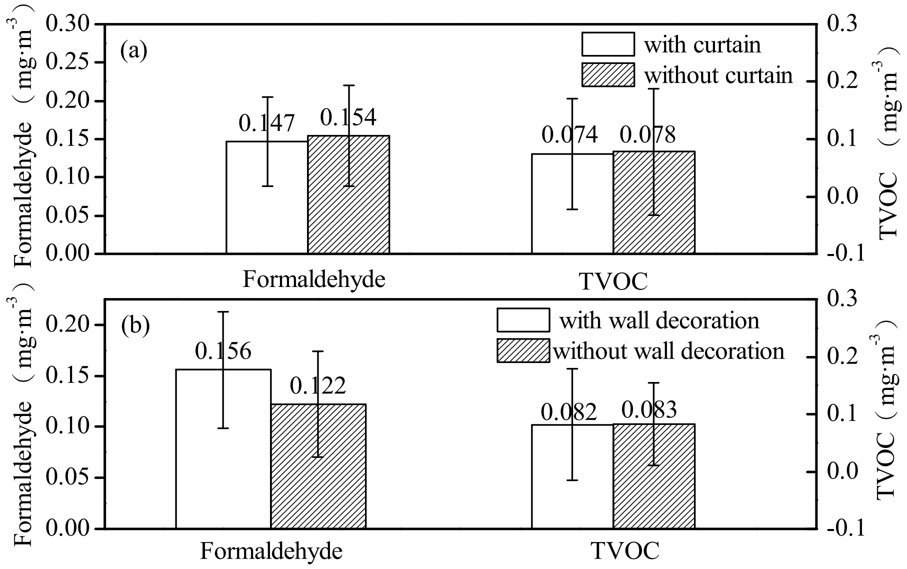

Fig. 4. The relationship between pollutants and curtain (a) and wall decoration (b).

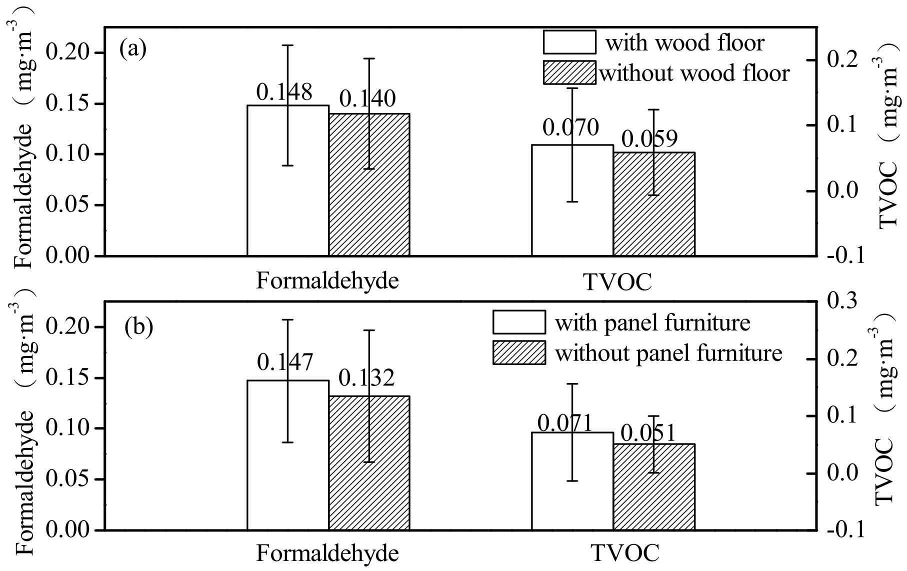

Fig. 5. The relationship between pollutants with wood floor (a) and panel furniture (b).

The Variance of the Concentrations of Formaldehyde and TVOC with Ventilation Period after Decoration

The variance of the concentrations of formaldehyde and TVOC as a function of time in 3 newly decorated houses with 11 rooms (3 living rooms and 8 bedrooms) were evaluated. These selected rooms with typically decoration styles on behalf of Xi'an, were decorated with wood floor, latex paint, wall decorations, curtains, panel furniture, etc. Except natural ventilation, no other method (such as mechanical ventilation) was utilized to enhance ventilation after decoration in these rooms. The measured data were shown in Fig. 6. Generally, the concentrations of formaldehyde and TVOC decreased with increasing of ventilation time, although the decreasing rate varied among the different kinds of rooms at a stable ventilation period. It was noted that rooms just decorated have a higher concentration of formaldehyde or/and TVOC than rooms after 1-month ventilation time, maybe resulting from that new furniture emission at the beginning. However, some rooms have higher formaldehyde or/and TVOC concentrations than one-month decoration and just decorated rooms, resulting from that some new small wooden furniture was moved into these rooms. Also of note was that the ventilation period required for the indoor concentration of formaldehyde and TVOC to decrease to meet the Chinese National Standard was varied for different kind of rooms. This can be attributed to differences of indoor air ventilation condition and decoration materials. In this study, 3–12 months of ventilation period were suggested for newly decorated rooms to meet the Chinese National Standard. Reasonably, rooms at serious pollution level need more ventilation time to satisfy the indoor air quality standard.

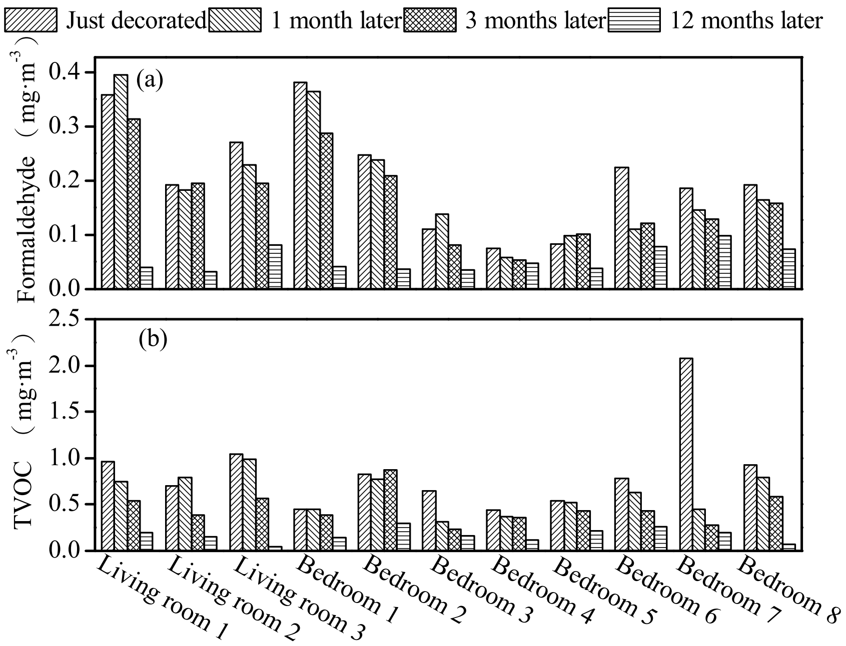

Fig. 6. The variances of formaldehyde and TVOC concentrations in newly-decorated rooms as a function of time.

As we mentioned above, the concentrations of formaldehyde and TVOC showed positive correlations with the indoor temperature, indicating that the higher temperature leads to more indoor exposure to formaldehyde and TVOC. Indoor high-temperature environment helped to increase the emission rates of formaldehyde and TVOC stemming from interior decoration materials. As a result, summer was the best ventilation time to remove formaldehyde and TVOC. And at the same time, maintaining good room ventilation was also an effective way to accelerate the release of VOCs.

CONCLUSIONS

The indoor concentration of formaldehyde in Xi'an heavily exceeded the Chinese National Standard (GB/T 18883-2002). 83.6% residences and 44.8% public places exceeded the Chinese National Standard, and the concentrations of formaldehyde in residences and public places ranged from 0.02 mg m^{-3} to 0.45 mg m^{-3} and 0.05 mg m^{-3} to 0.32 mg m^{-3}, respectively. As for TVOC concentrations, very few percentages (both 1.7%) in residences and public places exceeded the Chinese National Standard (GB/T 18883-2002), and the concentrations ranged from 0 mg m^{-3} to 0.84 mg m^{-3} and 0 mg m^{-3} to 0.66 mg m^{-3}, respectively. Among living rooms, bedrooms and other rooms, the exceeding rates of formaldehyde were all beyond 80%, which revealed that the formaldehyde pollution was a general phenomenon in Xi'an. On the Contrast, there were seldom rooms that have TVOC concentration out of limits. The public places had a lighter pollution situation compared with the residences.

The main sources of formaldehyde were wall decoration and panel furniture, while panel furniture and wood floor were the main sources for TVOC. Formaldehyde and TVOC had a positive relationship with indoor temperature, while RH favors the formaldehyde release but has a relatively weak correlation with TVOC. The concentrations of formaldehyde and TVOC decreased in newly decorated rooms with time and fell below the limits within 1 year. As mentioned above, it can thus be concluded that people should concern more to the decoration technique, choose appropriate low-emission decoration materials and furniture during decoration. In addition, keep good ventilation for a reasonable period to reduce formaldehyde and TVOC concentrations after decoration is needed and effectively, especially room ventilation more effective in summer.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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