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DISTRIBUTION CHARACTERISTICS OF FORMALDEHYDE IN NEWLY DECORATED ROOM IN NINGBO, TAIZHOU AND WENZHOU CITY, CHINA

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ABSTRACT

The indoor air samples taken from 768 newly decorated rooms in Ningbo, Taizhou and Wenzhou between November 2014 and October 2016 were analyzed to detect the concentration of formaldehyde, benzene, toluene, ethylbenzene and xylene. Meanwhile, the temperature, relative humidity and air pressure of those rooms were also recorded. The results showed that the formaldehyde concentration ranged from 0.000476 to 0.789 mg/m³, with the mean concentration of 0.136 ± 0.116 mg/m³. 47.9% of the rooms exceeded the permitted formaldehyde concentration defined in Chinese national standard for indoor air quality (GB/T 18883-2002). Significant positive correlations were found between the formaldehyde and temperature (r=0.358, P < 0.01), humidity (r=0.128, P<0.05), and the significant negative correlation was found between the formaldehyde and air pressure (r=-0.291, P<0.01). The highest formaldehyde concentration was observed in June and July, which could be due to the high temperature and humidity in these two months. The formaldehyde concentration in the drawing rooms and bedrooms was significantly higher than that of classrooms and halls. Besides, no positive correlations were found between the formaldehyde and benzene, toluene, ethylbenzene and xylene, suggesting that they might come from different sources.

KEYWORDS:

Indoor, Decorated, Formaldehyde, Distribution, Correlation

INTRODUCTION

The indoor air quality (IAQ) has attracted more and more public concern since people spend most of their time within the enclosed living space [1-6]. The poor IAQ is linked to sick building syndrome (SBS) [7]. On the other hand, due to the improved living standard, people pay much more attention to the indoor decoration, especially in the developing countries. This could increase the exposure of humans to the air pollutants including formaldehyde, benzene, methylbenzene, ethylbenzene, dimethylbenzene, etc. Among the various pollutants, formaldehyde is of particular interest due to its abundance and adverse health effects [8]. The low level formaldehyde could provoke eye and throat irritation, chest tightness and shortness of breath. High-dose exposure to formaldehyde could lead to the acute poisoning, chronic toxicity and even cancer [9-10], hence it is classified as carcinogenic to humans (Group 1) by the World Health Organization.

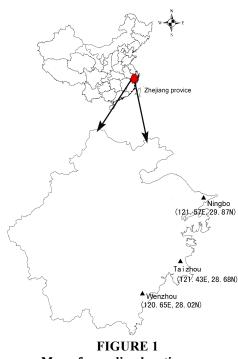
The indoor formaldehyde is connected with several indoor sources associated with the indoor decoration. For example, it can be emitted by the wood-based products used during the decoration, which is assembled with the urea-formaldehyde and phenol-formaldehyde resins. In addition, formaldehyde is also presented in the paints, varnishes and carpets [11, 12]. Chi et al. [13] reiterated that decoration pollution was the primary indoor pollution within the first 12 months after decoration. To avoid the healthy risk of the formaldehyde, the Chinese indoor air quality standard was issued in 2002, in which 0.1 mg/m³ of formaldehyde was considered preventive of negative effects on humans (GB/T 18883-2002). However, there are still a large magnitude epidemiology studies showing the adverse health is associated with indoor air pollution in terms of morbidity and mortality. It is estimated that about 110,000 people die from the indoor air pollution in China every year. Ningbo, Taizhou and Wenzhou, the three eastern coastal cities of Zhejiang province, are recognized as nouveaux riches regions in East China. The fast development of the economy in the last three decades has made people rich and eager to improve quality of their life. Lots of people there choose to promote the indoor decoration by using large quantities of building materials, such as wood, paints and wool products. However, this style of living may aggravate the indoor pollution. Up to the date, several studies have been done to investigate the level and the emission source of indoor formaldehyde around the world [14-18]. However, it is still necessary to investigate the distribution characteristics of formaldehyde in newly decorated rooms in these regions, which are identified as nouveaux

riches regions in the subtropical zone. As it is known, different lifestyles and meteorology conditions can result in different distribution characteristics of indoor formaldehyde. It is impossible to help the occupants escape from the negative health effect of indoor formaldehyde before the distribution characteristics are figured out.

In this study, 768 indoor air samples were taken from newly decorated rooms in Ningbo, Taizhou and Wenzhou city over two years. The concentrations of formaldehyde, benzene, toluene, ethylbenzene and xylene in the samples were determined. Besides, the correlation between the formaldehyde and sampling time, type of room, temperature, humidity, air pressure, as well as benzene, toluene, ethylbenzene and xylene was also discussed to reveal the distribution characteristics of formaldehyde in the newly decorated rooms.

MATERIALS AND METHODS

Sampling period and sites. 768 indoor air samples, collected between November 2014 and October 2016, were taken from the newly decorated rooms in Ningbo, Taizhou and Wenzhou city, which were three adjoining coastal cities located in Zhejiang province in China (Fig. 1). The three cities feature a subtropical monsoon climate, with a permanent population of 19,952,297 distributed over a 31292 km² land. Samples were collected between November 2014 and October 2016. The types of the rooms include the drawing room, bedroom, Hall, conference room and classroom.



Map of sampling location

Sampling methods. The sampling was carried out according to the Standard of Indoor Environmental Pollution Control of Civil Engineering (GB 50325-2010). Windows and doors were closed during sampling. Samples were taken at a height of 1.5 m above the floor. Formaldehyde was sampled at 0.5 L/min for 20 minutes. After the sampling, both ends of the sampling tube were sealed, and samples were brought back to lab for immediate analysis. Benzene, toluene, ethylbenzene and xylene were sampled at 0.2 L/min for 60 minutes using charcoal sorbent tubes. Samples were stored at -20 °C and analyzed within 48 h. Besides, the temperature, relative humidity and air pressure were recorded during the sampling.

Analytical methods. Formaldehyde was analyzed by the MBTH method through a UV-VIS spectrophotometer (GB/T 18204.26-2000). Benzene, methylbenzene, ethylbenzene and dimethylbenzene were extracted from charcoal tubes using 2 mL carbon disulfide. The solvent was transferred into GC vials and analyzed by a gas chromatograph (GC Agilent 7890A), which was equipped with FID detector using the capillary column (30 m×0.32 mm×1 μ m, CD-WAS). Aliquots of 1.0 µL were injected into the capillary column. Injector and detector temperatures were set at 150 and 250 °C, respectively. Oven temperature was programmed at 65 °C for 10 min and then 5 °C/min to 90 °C.

Statistical methods. Statistical analysis was performed by SPSS 19.0. Independent samples t-test was conducted to elucidate the equality of means for the formaldehyde concentration in different months and room types. Correlation was analyzed between the formaldehyde and temperature, humidity, air pressure, benzene, toluene, ethylbenzene and xylene.

RESULT AND DISCUSSION

Concentration of formaldehyde. The average formaldehyde concentration of the sampled rooms was 0.137 ± 0.116 mg/m³ (Fig. 2). The maximum concentration was 0.794 mg/m³, which was approximately 7 times higher than the permitted concentration defined in the Chinese standard for indoor air quality (GB/T 18883-2002, formaldehyde ≤0.10 mg/m^3). The formaldehyde concentration obtained in this study is higher than the result of Guo et al. [15], which shows the average formaldehyde concentration of newly decorated rooms is 0.107±0.095 mg/m³ in Hangzhou city of Zhejiang province, China. Besides, it is also higher than that of Canada, France, Japan and Sweden [14, 16, 19, 20]. Among the 768 samples, 47.9% of them exceeded the permitted concentration. The above results suggest that the newly decorated rooms in Ningbo, Taizhou and Wenzhou city are so polluted that special measures



must be taken to control the pollution. However, it seems that the indoor pollution has not attracted serious attention. If the study is divided into two phases, including the phase I from the November 2014 to October 2015 and the phase II from the November 2015 to October 2016, no significant difference could be found between the two phases. The average formaldehyde concentration was 0.134 ± 0.114 mg/m³ for the phase I and 0.139 ± 0.119 mg/m³ for phase II. That is to say that no valid measures have been taken to reduce the indoor formaldehyde pollution for the newly decorated rooms.

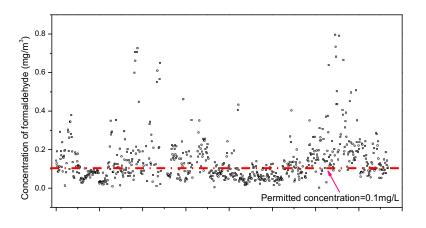
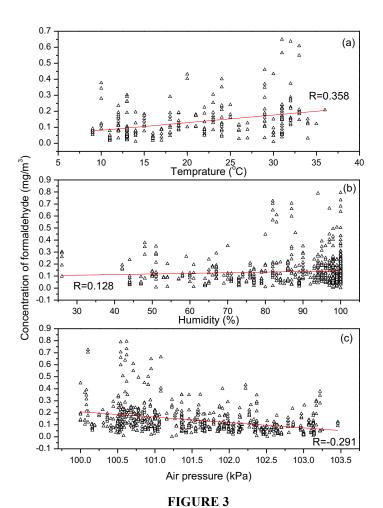


FIGURE 2 Formaldehyde concentration of 768 sampled rooms



Correlation between formaldehyde and temperature (a), relatively humidity (b) and air pressure (c)

Correlation between formaldehyde and the meteorological condition. As shown in Fig. 3(a), the formaldehyde concentration was significantly positively correlated with the room temperature (r=0.358, P<0.01). This result was consistent with the previous studies [13, 19, 21], which manifested that temperature played a key role in influencing the indoor formaldehyde. Formaldehyde is a volatile organic compound. A high temperature can facilitate the release of formaldehyde from the building materials. Zhang et al. [21] investigated the effect of temperature on the emission parameter of indoor formaldehyde, and concluded that the increase of the temperature could decrease the partition coefficient and increase the diffusion coefficient.

The positive correlation was also found between the formaldehyde and relatively humidity (Fig. 3(b), r=0.128, P<0.05). As it is known, formaldehyde has a high solubility in water. The high moisture in the air could induce the rapid absorption of formaldehyde, which will destroy the equilibrium between the building materials and adjacent air. It in turn promotes the emission of formaldehyde from the building materials. In other words, the increase of the humidity could decrease the partition coefficient and increase the diffusion coefficient of formaldehyde in building materials.

Different from the temperature and humidity, the formaldehyde was negatively correlated with the air pressure (Fig. 3(c), r=-0.291, P<0.01). High air pressure is believed to be advantageous to the convection of the indoor air. The increase of indoor air convection can help the indoor formaldehyde diffuse into a larger space. Therefore, the indoor formaldehyde concentration decreases.

It should be noted that the absolute value of correlation coefficient between the formaldehyde and temperature, air pressure was far greater than that of humidity. This result suggests that the temperature and air pressure play a more important role in influencing the indoor formaldehyde distribution, when compared with the relative humidity.

Monthly variation of formaldehyde distribution. As is shown in Fig. 4, the monthly formaldehyde concentration showed a unimodal distribution. The average formaldehyde concentration in June and July was 0.232±0.209 and 0.220±0.138 mg/m^3 , respectively, which was significantly higher than other months (P < 0.05). 75.0% of the samples collected in June and 84.8% of the samples collected in July exceeded the permitted concentration. The lowest average formaldehyde concentration was observed in January, which was 0.068±0.038 mg/m³. 20.5% of the samples collected in January exceeded the standard. This result is generally consistent with Guo et al. [15] and Weng et al. [4], which demonstrates that the formaldehyde concentration in summer is significantly higher than that in winter. In Ningbo, Taizhou and Wenzhou, compared with the other months, June and July usually have a relatively high temperature, relative humidity and low air pressure, while the meteorology characteristics are almost the opposite in January. The variation of the meteorological characteristic of each month results in the different distribution of formaldehyde. As illustrated in this study, the high temperature, relative humidity, and low air pressure are prone to seduce the release of formaldehyde from the building materials. Therefore, high formaldehyde concentration was recorded in June and July, while the low formaldehyde concentration was recorded in January. That is, the intensified release of formaldehyde probably happens in the summer season for the newly decorated rooms, which reminds us to pay more attention on the control of indoor formaldehyde pollution.

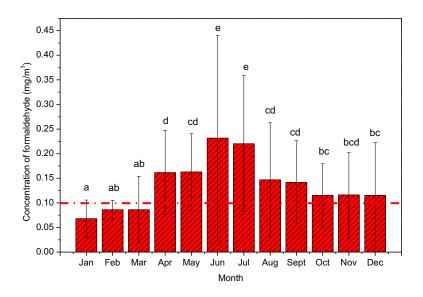


FIGURE 4 Monthly variation of formaldehyde concentration



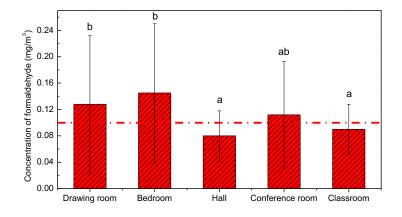


FIGURE 5 Formaldehyde concentration in different type of rooms

 TABLE 1

 Correlation between the formaldehyde and benzene, toluene, ethylbenzene, xylene

	Formaldehyde	Benzene	Toluene	Ethylbenzene	xylene
Formaldehyde	1	-0.288*	-0.020	-0.030	0.083
Benzene	-0.288**	1	0.081	0.258^{**}	0.159^{**}
Toluene	-0.020	0.081	1	0.156**	0.127^{*}
Ethylbenzene	-0.030	0.258^{**}	0.156^{**}	1	0.783^{**}
M-xylene	0.083	0.159**	0.127^{*}	0.783^{**}	1

Concentration of formaldehyde in different types of rooms. The sequence of average formaldehyde concentration in all types of rooms was as followed: bedroom > drawing room > conference room > classroom > hall (Fig. 5). Formaldehyde concentrations of bedroom and drawing room were significantly higher than that of hall and classroom (P<0.05). 51.6% of the bedroom and 50.0% of the drawing room exceeded the permitted formaldehyde concentration defined in Chinese national standard for indoor air quality, while only 36.8% of classroom and 20.7% of the hall exceeded the standard. This result is not surprising as the decorations of bedrooms and drawing rooms are far more excessive than that of halls and classrooms according to our site investigation. People use more wood-based materials and paints for ceiling, flooring and cabinet in the bedroom and the drawing rooms. They also arrange more kartel, wooden sofa as well as the wallpaper to 'improve' the comfort of the residential living. Compared with the bedrooms and drawing rooms, the halls and classrooms have a larger space and use less formaldehyde concentrated materials. It indicates that residential pollution is a more serious issue than the public place pollution. It is advised that people in these regions need to adjust their lifestyle to mitigate the residential pollution, such as avoiding the excessive home decoration and using formaldehyde-free material.

Correlation between the formaldehyde and BTEX. BTEX, including benzene, toluene, ethylbenzene and xylene are another category of crucial indoor pollutants. The correlation analysis results showed that no positive correlation was found between the formaldehyde and benzene, toluene, ethylbenzene and xylene (Table 1). This result indicates that the formaldehyde and BTEX might come from different sources. Some studies have showed that the BTEX mainly originate from outdoors as vehicle emission and industrial combustion [22, 23]. Some other researchers pointed out that the indoor combustion contributed to the indoor BTEX pollution. For example, Hazrati et al. [1] reported that the heating system, gas stove, samovar and tobacco smoking were the main source of BTEX. Huang et al. [24] indicated that the cooking activities contributed to the indoor BTEX. These results suggest that the BTEX mainly came from the combustion, which was different from that of formaldehyde. Compared with the formaldehyde, only 11.7% sampled rooms exceed the permitted concentration of benzene, toluene, ethylbenzene and xylene defined in Chinese standard for indoor air quality (GB/T 18883-2002, benzene $\leq 0.11 \text{ mg/m}^3$, toluene $\leq 0.20 \text{ mg/m}^3$, xylene $\leq 0.20 \text{ mg/m}^3$). It further indicates that the main sources of formaldehyde and BTEX are different from each other.

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Implication. The result of this study could provide reference to prevent the human exposure to the indoor formaldehyde pollution. As approximately half of the newly decorated rooms exceed the warning threshold of the formaldehyde, it is unwise to use new rooms immediately. The pollution is especially serious in residence, which requires people to decrease the strength of the residential decoration and use more formaldehyde-free materials. The result also reveals that the high temperature and relative humidity could facilitate the release of formaldehyde. Because the intensified release of formaldehyde usually occurs in the summer, to prevent the healthy risk, newly decorated room should not be used before it goes through a summer season.

CONCLUSIONS

The newly decorated rooms are seriously polluted by the formaldehyde in Ningbo, Taizhou and Wenzhou city. 47.9% the newly decorated rooms exceeded the Chinese standard for indoor air quality with the respect of formaldehyde pollution. The bedrooms and drawing rooms posed a higher formaldehyde concentration than the classrooms and halls, indicating that residential pollution is a more serious issue than the public place pollution. Moreover, the formaldehyde concentration of samples collected in June and July were significantly higher than other months, which might be due to the high temperature, high humidity and low air pressure in the two months. No positive correlation was found between the formaldehyde and benzene, toluene, ethylbenzene and xylene, suggesting that they were from different sources. Although serious pollution is found in the newly decorated rooms, it seems that inadequate attention has been paid to mitigate the pollution. It is recommended that the newly decorated room should not be used before it goes through a summer season.

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