Law and features of TVOC and Formaldehyde pollution in urban indoor air

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ABSTRACT

There are several categories of indoor air pollutants. Organic pollutants are the most common ones. This study chooses TVOC and Formaldehyde, two of the typical pollutants, as indicators of evaluating household indoor air pollution and improves the TVOC concentration prediction model through the samples of indoor air taken from 3122 households. This study also categorizes and explains the features of household indoor air pollution based on the TVOC and Formaldehyde models as well as a large amount of sample measurement. Moreover, this study combines the TVOC model with the Formaldehyde model to calculate and verify the critical values of each type of indoor air pollution. In this study, indoor air pollution is categorized into three types: decoration pollution, consumption pollution and transition pollution. During the first 12 months after decoration, decoration pollution is the primary pollution type, both TVOC and Formaldehyde are highly concentrated while sometimes seriously over the standard. Pollutants mainly come from volatile sources. After the first 12 month but before 24 months the indoor air pollution is transition pollution. Both decoration materials and human activities affect the indoor air quality. 24 months after decoration, it transits into consumption pollution. In this stage, the main pollutants come from combustion sources, and concentration of pollutants fluctuates with the appearance and disappearance of the sources.

Keywords: Indoor air pollution type TVOC Formaldehyde Feature Law

1. Introduction

Due to the improved living standard, people pay more attention to indoor construction and decoration now. This leads to a very serious problem of indoor air pollution, especially organic pollution. VOCs are the main cause for detrimental quality of indoor air. There are over 500 types of VOCs have been detected. Many researches only focus on measuring certain types of VOCs. However, the total quality concentrations of volatile organic compounds in indoor air (TVOC, Total Volatile Organic Compounds) (Molhave et al., 1997) can better evaluate indoor air quality, which can be an important index (Wong et al., 2006). Formaldehyde is a typical pollutant of VOCs, also the most commonly-known indoor air pollutants (Salthammer et al., 2010). Logue et al. (2011) compiled summary results from 77 published studies reporting measurements of chemical pollutants in residences in the United States and identified key

chemical contaminants of concern in residential indoor air, including Formaldehyde and other VOCs. Therefore, this study will also discuss Formaldehyde as an important factor in evaluating indoor air pollution in detail.

Many studies have been conducted in these few years to measure TVOC and Formaldehyde concentrations (Lee et al., 2002; Lai et al., 2004; Sakai et al., 2004) and tentatively explore pollutant sources of VOCs. There are many sources of VOCs pollution. Based on the production mechanism of pollutants, sources of VOCs pollution can be divided into volatile sources (Guo et al., 2011; Liu et al., 2014), combustion sources (Lee et al., 2002) and composite sources (Lee et al., 2001, Destailats et al., 2008 ; Morawska et al., 2009).

Along with more and more proceeding of regular monitor and identification of pollutant sources, many researchers turned their focus from monitoring to studying the laws of TVOC and Formaldehyde. They also concluded that the concentration of TVOC and Formaldehyde and the features and usage of pollutant sources are highly relevant (Gilbert et al., 2006; Marchand et al., 2007). Seasons, temperature, humidity and other environmental factors can affect the concentration of TVOC and Formaldehyde, too (Hoffmann et al., 2000; Schneider et al., 2001; Dingle; Franklin, 2002; Schlink et al., 2004; Jarnstrom et al., 2006; Jo et al., 2009; Huang et al., 2015). Son et al. (2003) discovered that the concentration of some VOCs in indoor air and the air exchange rate are negatively correlated. Later, other scholars confirmed this theory through experiments (Salthammer et al., 1995; Gilbert et al., 2008; Jia et al., 2008)

Liu et al (2011) further explored affecting factors based on previous researches. His research revealed that concentration of TVOC is closely related to time from the end of decoration and time the door and windows closed before sampling. The concentration also has a nonlinear correlation with humidity and temperature.

By integration the source characteristics and environmental factors, the regression was developed as follows:

$$\ln(C_{TVOC})=0.212\ln(t_1+1)-5.42\times10^{-3}\times t_2^2-1.52\times273.15/(T+273.15)-44.1/RH+0.952d+1.40 \quad (1)$$

T : indoor temperature (°C), RH : the relative humidity (%), t₁: time the door and windows closed before sampling (h), t₂: time from the end of decoration to sampling (months), The diffusion sources of pollutants are divided into three levels, non-dimensional values of -1, 0, and 1 are assigned to levels -A,-B and -C, respectively, and these are expressed as parameter d.

In addition, Guo et al (2013) advocated using humidity-and-temperature coefficient to define the combined effect of humidity and temperature on diffusion of Formaldehyde, which is calculated as below.

$$TH(T, RH) = -20.2\times273.15/(273.15 + T) -27.6\times1/RH \quad (2)$$

Guo et al (2013) defined it as the humidity-and-temperature effect of indoor Formaldehyde pollution and put forward a new household Formaldehyde concentration model:

$$\ln(C_{HCHO})=-20.2\times273.15/(273.15+T) -27.6\times1/RH +0.124\times\ln(DC+1)-2.85\times10^{-3}\times DR^2+0.8\times d+16. \quad (3)$$

T : indoor temperature (°C), RH : the relative humidity (%), DC: time the door and windows closed before sampling (h), DR: time from the end of decoration to sampling (months), The diffusion sources

of pollutants are divided into three levels, non-dimensional values of -1, 0, and 1 are assigned to levels -A, -B and -C, respectively, and these are expressed as parameter d.

These two models are great breakthroughs from the old models. They consider all different factors and collaborate with real data. But because the features of pollutant sources are the major factors affecting indoor air pollution, simplifying the features into three levels and assigning non-dimensional numbers (-1,0,1) to the levels reduces the fitness of the models.

Therefore, this study investigated TVOC and Formaldehyde concentrations in more than three thousand newly decorated rooms as well as older rooms. Through further clarifies the characteristics of pollutant sources, this study modifies the model for TVOC diffusion based on the previous researches and measurement results, which can improve accuracy of predication, categorizes and explains the features of indoor air pollution. This study will further reveal the characteristics and laws of indoor air pollution

2 Materials and Methods

2.1 Sampling sites

Hangzhou is the second largest city in terms of economy in the Yangtze River Delta, Chin. Hangzhou has four distinctive seasons with representative temperature and humidity. Because Real estate in Hangzhou is relatively developed, the decoration level in Hangzhou also has certain representativeness. Therefore, this study considers Hangzhou as an example to investigate the rule and patterns of indoor air.

Air samples were collected in 3122 residential rooms at Hangzhou, china, from January 2007 to December 2014 (Table 1).

2.2 Sampling program

All the rooms were kept under normal conditions before sampling. Windows and doors were closed during sampling. The air sampler for was set 1.5 m above the floor.

VOCs were collected into seamless stainless steel tubes packed with 0.20 mg of Tenax-TA adsorbent (60–80 mesh) using an electronically controlled air sampler (DDY-1.5, Xingyu, Jiangsu, China) at 0.4 L/min for 10 min. This sampling tube was activated using N₂ for 1 h at 350°C before sampling, then sealed with seal film and a plastic cap.

HCHO was sampled at 0.5L/min for 20 min. When sampling was complete, both ends of the sampling tube were sealed, and samples were brought back to laboratory for immediate processing.

The temperature (T), relative humidity (RH), time the windows and doors closed for before sampling (DC) (t_1), time from the end of decoration to sampling (DR) (t_2), and atmospheric pressure (P) were recorded at the sample collection time (Table 2).

2.3 Experimental methods

VOCs in the sample were separated by gas chromatography, and then analyzed by thermal desorption at 250°C for 10 min. The chromatographic conditions and analytical methods are detailed in GB 50325-2001.

HCHO was determined by the 3-methyl-2-benzothiazolinone hydrazone method through a UV-Visrecording spectrophotometer (detector UV@630 nm, UV-2401PC, Shimadzu, Japan). The reagent

preparation and analytical methods are detailed in the Chinese National Standard GB/T 18204.26-2000.

3. Results

3.1 TVOC and Formaldehyde concentrations in urban indoor air

TVOC and Formaldehyde concentrations in the examined rooms were listed in Table 3

3.2 TVOC models modification

Firstly, we require a confirmation that which pollutant source diffusion model provides the best fitness and expression. The existing researches has already compared and analyzed the fitness of different pollutant source diffusion models. Zhu et al (2001) conducted experiment on different pollutant sources in active experiment module. He compared the fitness of first-order model and power law decay model, and discovered the decay coefficient using the first-order model to estimate is below the experiment outcome for a long period. On the other hand, the power law decay model turned out to be a better fit with the experiment results. Power law decay model can be defined by two parameters:

$$E = a \times t^{(-b)} \quad (4)$$

t is defined as emission time, h^{-1} ; a is the initial value, equals to the diversion coefficient of pollutant source at one hour. $\text{mg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$; b is the dimensionless constant.

$$b = \log(ef_1) - \log(ef_2) / (\log t_2 - \log t_1) \quad (5)$$

$$a = ef_1 \times t_1^b \text{ Or } a = ef_2 \times t_2^b \quad (6)$$

As is shown in Formula (6), a equals to the value of ef when $t=1$. And the value of ef is related to the concentration of pollutants; C , rate of air exchange; N , the loading ratio; L , ratio of total surface area of the interior materials and rooms volume.

Han et al (2012) compared the fitness of typical double-exponential decay model, power law decay model and mechanistic diffusion model for indoor volatile source. His research revealed that power law decay model and mechanistic diffusion model fits better for long-term diffusion. These two models provide better description of diffusion within the materials. Mechanistic diffusion model is more complex in application, while power law decay model is more efficient in calculation.

As a result, power law decay model is better for describing and predicting long-term diffusion of VOCs from indoor materials than first-order model, double-exponential decay model and mechanistic diffusion model.

We fit data of TVOC and Formaldehyde concentrations measured in actual room (newly decorated within 8 months)(Brown, 2002) with the power law decay model, and compare the predicted value with actual value. (With the actual value as x value, predictive value as y value, the curve slope k is more close to 1, the predicted results will be better). The final result shows that the power law decay model can well describe and predict the attenuation law of TVOC and Formaldehyde concentration for a long time in actual room.(Table 4)

After selecting the decay model, this study now describes the features of source with reference to parameter a in the power law decay model. Based on statistics from experiment, A is defined as the

combined value of pollutant sources in decoration pollution (related to decoration material, load of material and air exchange rate), which is estimated by two-point method. Each factor is adjusted based on factor A and four environmental factors (t_1 , t_2 , T and RH), and fitted into the TVOC concentration by means of stepwise regression analysis.

Stepwise regression analysis starts with one variable, and introduces each variable into the regression equation by relevancy with Y . However, a variable has to be deleted if sequent introduction of new variable renders its relevancy null.

After the attempted fitting of each of the five factors (A , t_1 , t_2 , T and RH) with the concentration of TVOC, it appears that correlation coefficient R , reduces significantly after RH is introduced into the calculation when conducting stepwise regression analysis. Therefore, RH is deleted from the calculation to maximize the correlation coefficient. The equation for this model is:

$$\ln(C_{\text{TVOC}}) = C_0 + C_1 \times \ln A + C_2 \times \ln t_1 + C_3 \times t_2^2 + C_4 \times 273.15 / (T + 273.15) \quad (7)$$

A is defined as variables of pollutant sources features; t_1 : time the door and windows closed before sampling (h), t_2 : time from the end of decoration to sampling (months), T is defines as indoor temperature °C. C_1 , C_2 , C_3 , C_4 is the coefficient of each parameter. C_0 is constant. The fitness is listed below in Table 5

As in the fitness results, the determinative coefficients $R^2=0.8943$, which means the four variables can explain the over 89% of causes for variation of TVOC concentration. This is higher than what was in the previous researches. Therefore, the elaboration of pollutant sources features reduces errors caused by simplified measurements. The remaining uncertainty is caused by other factors, for example, air exchange rate and adsorption effect of indoor decoration materials. The fitness results indicate that this model can calculate variations of TVOC concentration in decoration pollution period. The finalized equation is shown as below:

$$\ln(C_{\text{TVOC}}) = -1.3208 + 0.8195 \times \ln A + 0.0329 \times \ln t_1 - 0.0076 \times t_2^2 + 1.0591 \times 273.15 / (T + 273.15) \quad (8)$$

A is defined as variables of pollutant sources features; t_1 : time the door and windows closed before sampling (h), t_2 : time from the end of decoration to sampling (months), T is defines as indoor temperature °C.

In order to verify the fitting effect of this model, we compared the predicted values of the model with its actual monitoring values (Fig.1). The actual monitoring values were practically measured within 1 to 6 months after the decoration in newly decorated houses in Hangzhou, China. Each house was measured 3 or 4 times, and then values derived from the first and second measurements were used to estimate the values of pollutant sources features(A) before utilizing the model to make predictions. Using the monitoring values as the abscissa and the predicted values as the ordinate, results shown in the Fig.1 were derived, with the slope of the curve as $k=0.887$ whereas $R^2=0.882$. Such results implied that the predictions of the model are correlated to the actual monitoring values to some extent. (This is a supplementary experiment)

We also compare the predicted value under the modified model and Liu's model with the measured value: the measured value is the value of TVOC concentration within 1~6 months after decoration. The samples are selected from 3122 measured households (number of samples is 48), with 3~4 times of measurement for each household in Hangzhou, China. The first two-measurement value will be used to estimate the pollutant source feature factor A. Apply A to the model and acquire predicted result 1. Then apply Liu's model to get predicted result 2. Compare the two predicted results based on the equation: relative error value=Abs (Predicted Value-Measured Value)/ Measured Value (See results in Table 6)

The average value of relative error for model 1 is 0.2049, while the average value of relative error for model 2 is 0.3746. This indicates that the modified model leads to more accurate prediction than Liu's model.

3.3 Types of indoor air pollution and their features

To apply the new model to predict the time needed for TVOC concentration to reach the Chinese national standard (0.5 mg/m^3), we firstly set the time the doors and windows closed before sampling as one hour, the temperature as the average temperature of Hangzhou, China in the seven decades, from 2007 to 2014 and the values of source feature factor A in table 7 were derived from above measurement results. When choosing the minimum value of A, the result shows that for TVOC concentration to go below the 0.5 mg/m^3 standard, it takes about 8 months in average. When the source feature factor A is maximum, in other words, when the decoration pollution is the most severe, for TVOC concentration to go below the standard, it takes about 16 months. The average time is 12 months.

To apply the equation (3) (Guo et al., 2013) to predict the time needed for Formaldehyde concentration to reach the Chinese national standard 0.08 mg/m^3 . We firstly set the time the doors and windows closed before sampling as one hour, and the temperature as the average temperature of Hangzhou, China in the seven decades. The result shows that for Formaldehyde concentration to go below the 0.08 mg/m^3 standard after decoration, it takes about 24 months in average. To make the conclusion more convinced, we also added The European indoor Formaldehyde standard 0.05 mg/m^3 into calculation as stricter circumstance. The result shows that it takes about 27 months Formaldehyde concentration to go below the 0.05 mg/m^3 standard after decoration. The result is close to 24 months

We chose the statistics with complete information after filtering the results, including decoration time (table 8).

We used the above-mentioned data of TVOC and Formaldehyde concentrations to verify the critical points we calculate from the models (Fig.2 and Fig.3).

Based on the features of indoor pollutant sources, features of indoor air pollution, and a large amount of measured statistics, we categorized indoor air pollution into three types: decoration pollution, consumption pollution and transition pollution. This study also concluded that the critical points for the three stages of pollution are 12.5 months and 24 months based on prediction model and measurement. Within 12.5 months after decoration, the indoor pollution is decoration pollution. After the first 12.5 month but before 24 months the indoor pollution is transition pollution. And 24 months after decoration, it transits into consumption pollution.

During the decoration pollution period, the Formaldehyde and TVOC in the indoor air mainly come from decoration material. And the concentrations of Formaldehyde and TVOC are generally beyond safety level at the very beginning. However, concentrations of pollutants significantly reduce when time passes. Formaldehyde concentration is sensitive to humidity and temperature. The main source of pollution is volatile sources at this stage. Therefore, the above-mentioned model for concentration of Formaldehyde and TVOC is suitable for determining concentrations at decoration pollution period.

During consumption pollution period, Formaldehyde and TVOC in indoor air come from daily consumption and human activities. In addition, the concentrations of pollutants vary a lot because of the start or cease of human activities, such as cooking and smoking. Difference in living habits can lead to changes in Formaldehyde and TVOC concentrations. But these changes are only temporary. The amount of pollutants can exceed safety standard in a very short period of time and Formaldehyde is not so sensitive to humidity and temperature. The main pollutant source is combustion source and this stage will not exist if we do not introduce new consumption pollutant sources into indoor environment, but what for sure is that decorated materials can be neglected in this period.

During the transition pollution period, concentrations of Formaldehyde and TVOC in some households are over safety standard, but not excessive in some other households. At this stage, the situation can be complicated. It is a transition that is the continuation of decoration pollution period and the beginning of consumption pollution period, if there exists this period. Anything from decoration materials to daily consumption and human activities can contribute to concentrations of Formaldehyde and TVOC in indoor air. The sources of pollution are mainly composite sources in this stage.

4. SUMMARY

The main source of indoor pollution is organic pollutants. What draws most attentions is TVOC and Formaldehyde pollution.

(1)According to the production mechanism of pollutants, pollutant sources can be categorized into volatile sources, combustion sources and composite source.

(2)Indoor air pollution can be divided into three types based on the law and features of indoor air pollution: Decoration pollution, Consumption pollution, Transition pollution.

(3)The features and critical values of Different types: Decoration pollution is within about the first year after decoration with volatile sources being the main pollutant sources. Formaldehyde concentration is sensitive to humidity and temperature. The concentrations of Formaldehyde and TVOC will decay regularly and can be predictable. Consumption pollution is the period of about two years after decoration, with combustion sources being the main pollutant sources, and the concentrations of two pollutants vary so irregularly that they generally cannot be predicted. Transition pollution is what happens during the second year after decoration with composite sources being the main source of pollution. The duration of transition pollution is related to the intensity of decoration pollution and the introduction of new combustion sources or not, both factors cannot be ignored.

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Table 1 Number of air samples collected each month from 2007 to 2014

Table 2 Sampling conditions

Table 3 TVOC and Formaldehyde concentrations and percentage of samples exceeding the China standard in 2007-2014

Table 4 Fitting result of power law decay model

Table 5 Fitting result of TVOC concentration with A, t₁, t₂ and T

Table 6 Comparison of measured value and predicted value using two models (ug/m³)

Table 7 The value of A according to monitoring results

Table 8 TVOC and Formaldehyde concentrations for verification (mg/m³)

Table 1 Number of air samples collected each month from 2007 to 2014

	2007	2008	2009	2011	2012	2013	2014	Total
January	49	48	69	1	4	1	18	190
February	42	12	30	3	3	3	20	113
March	29	26	44	3	3	3	21	129
April	55	46	77	8	6	9	55	256
May	18	95	66	10	8	11	63	271
June	24	103	73	15	13	15	70	313
July	42	109	103	23	18	23	100	418
August	114	130	67	15	12	13	66	417
September	72	88	29	14	14	14	55	286
October	36	86	91	9	8	9	40	279
November	28	115	73	2	2	2	20	242
December	43	108	62	1	4	2	18	238
Total	552	966	784	104	95	105	546	3122

Table 2 Sampling conditions

Impact	Min	Max	Mean	S.D	N
T(°C)	1.5	40.0	10	8.4	3122
RH (%)	32.0	93.0	67.5	9.7	3122
P (KPa)	98.3	104.1	101.2	0.9	3099
DC (h)	0	240	7.1	16.6	3122
DR (month)	0.25	12.0	4.4	3.6	298

Table 3 TVOC and Formaldehyde concentrations and percentage of samples exceeding the China standard in 2007-2014

TVOC(mg/m ³)							
Year	n	Min	Max	Mean	S.D	Number of samples exceeding standard	Percentage of samples exceeding standard (%)
2007	552	0.04	5.8	0.92	0.74	364	65.90
2008	966	0.05	6.8	0.51	0.56	254	26.30
2009	784	0.08	7.2	0.64	0.77	241	30.70
2011	104	0.289	1.028	0.609	0.176	70	67.3
2012	95	0.234	0.957	0.498	0.138	40	42.1
2013	105	0.304	0.933	0.539	0.120	49	46.7
2014	546	0.028	1.415	0.565	0.122	399	73.1
Total	3122	0.028	7.2	0.611	0.375	1417	45.38
Formaldehyde(mg/m ³)							
Year	n	Min	Max	Mean	S.D	Number of samples exceeding standard	Percentage of samples exceeding standard (%)
2007	557	0.006	0.664	0.149	0.111	368	66.1
2008	980	0.005	0.739	0.109	0.096	370	37.8
2009	787	0.01	0.776	0.074	0.064	167	21.2
2011	104	0.06	0.88	0.282	0.139	104	100
2012	95	0.05	0.583	0.206	0.114	94	98.9
2013	105	0.069	0.395	0.207	0.078	105	100
2014	546	0.015	0.694	0.229	0.099	544	99.6
Total	3122	0.005	0.88	0.179	0.100	1752	56.1

Table 4 Fitting result of power law decay model

	a	b	k	R ²
TVOC	1170.98	0.47	1.042	0.993
Formaldehyde	34.51	0.18	0.975	0.979

Table 5 Fitting result of TVOC concentration with A, t₁, t₂ and T

Variables	Coefficients	Regression Coefficients	Standard Regression	Partial correlation coefficient	t-statistic	P
<i>A</i>	<i>C</i> ₁	0.8195	0.8706	0.9187	39.1212	0.0000
<i>t</i> ₁	<i>C</i> ₂	0.0329	0.0562	0.1510	2.5699	0.0107
<i>t</i> ₂	<i>C</i> ₃	-0.0076	-0.4580	-0.7809	21.0313	0.0000
<i>T</i>	<i>C</i> ₄	1.0591	0.0377	0.1014	1.7154	0.0574
constant	<i>C</i> ₀	-1.3208	-	-	-	-
R ² =0.8943			Adjusted R ² =0.8927			
F-statistic : 492.29			p : 0.0000			
Durbin-Watson statistic:1.9459						

Table 6 Comparison of measured value and predicted value using two models (ug/m³)

Models	Number	Mean	Max	Min	S.D
Relative error value 1	48	0.2049	0.760	0.016	0.1463
Relative error value 2	48	0.3746	0.808	0.022	0.1909

Table 7 The value of A according to monitoring results

N	Max	Min
100	31981.423	5439.009054

Table 8 TVOC and Formaldehyde concentrations for verification (mg/m³)

	Min	Max	Mean	S.D	n
Formaldehyde	0.0126	0.585	0.149	0.0997	139
TVOC	0.12	5.5	1.044	0.8462	182

Fig.1 The relationship between fitting results and monitoring results

Fig. 2 The distribution characteristics and decay law of TVOC concentration over time

Fig. 3 The distribution characteristics and decay law of Formaldehyde concentration over time

Fig.1 The relationship between fitting results and monitoring results

Fig. 2 The distribution characteristics and decay law of TVOC concentration over time

Fig. 3 The distribution characteristics and decay law of Formaldehyde concentration over time

Graphical Abstract (for review)