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An investigation of formaldehyde concentration in residences and the development of a model for the prediction of its emission rates

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Highlights:

- CO₂ concentration was used as the tracer gas to calculate the indoor ventilation airflow rate;
- Monte-Carlo simulations were conducted for sensitivity analysis;
- A time-averaged effective emission rate predicts the formaldehyde emission rate in residences;
- Occupant activity was taken into account to calculate the emissions.

Abstract: Indoor air pollution caused by formaldehyde associated with building materials imposes a variety of acute and chronic adverse effects on people's health. The aim of this research is to investigate the concentrations of formaldehyde in residences and develop emission rate prediction model in residential buildings. On-site measurements including the indoor and outdoor concentrations of formaldehyde and CO₂ were carried out in 42 urban residences in Chongqing. The people occupancy schedule in different functional rooms was obtained by observing the change in CO₂ concentration. A robust model for the estimation of formaldehyde emission rates using

CO₂ as the tracer gas; associated with a Monte-Carlo simulation of occupant activities and the characteristics of residences; has been developed. It is revealed that the mean indoor formaldehyde concentration was 30.12 $\mu\text{g}/\text{m}^3$, which was slightly higher than the outdoor concentration of 27.80 $\mu\text{g}/\text{m}^3$. The emission rates of 61.82 \pm 52.39 and 49.69 \pm 42.13 $\mu\text{g}/\text{h}/\text{m}^2$ (mean \pm SD) during the daytime and nighttime, respectively with a daily average of 57.20 \pm 48.79 $\mu\text{g}/\text{h}/\text{m}^2$. The significant contribution to indoor formaldehyde concentration was from indoor sources. Indoor formaldehyde source control is suggested to be an efficient way to control the indoor concentration.

Key words: indoor air quality (IAQ); formaldehyde; air exchange rate; emission rate

1. Introduction

Volatile organic compounds (VOCs) are one of the main sources of indoor air pollution. As the most commercially used aldehyde, formaldehyde is widely used in construction, wood processing, furniture, textiles, carpeting, and the chemical industry^{[1][2]}. China is the single-largest market for formaldehyde, accounting for 47% of world demand in 2017^[3]. Due to the economic boom in recent decades, intensive interior decoration and renovation of homes became very popular in China^[4] for the purpose of creating more aesthetically pleasing home environments. More than 65% of formaldehyde production goes to produce synthetic resins used in building materials^[1]. For example, interior decoration materials such as wood, wallpaper, paint, and household consumer products like floor cleaning agents, candles, and electric air fresheners^[5] would release formaldehyde into the indoor environment. Meanwhile,

formaldehyde has been detected in exhaled air using modern analytical techniques and has even been linked to various diseases. Compounds, particularly aldehydes, have to be considered with great care since their elevated exhaled level might reveal a relationship to exposure to air pollutants^[6].

Making buildings airtight in an attempt to improve energy efficiency by decreasing the infiltration of unconditioned outdoor air^[7] could potentially contribute to lower-quality indoor air due to lack of ventilation.

It is reported that the indoor air pollution caused by formaldehyde associated with building materials imposed a variety of acute and chronic adverse effects on people's health^{[1][4][8]} such as symptoms of Sick Building Syndrome (SBS), lower respiratory and eye irritation, acute poisoning, dermal allergies, allergic asthma, neurotoxicity, pulmonary function damage, and potential carcinogenic effects. The situation has attracted considerable public attention in recent years in terms of their health and wellbeing. The mechanism for the emission and release of pollutants from materials has been extensively studied in recent decades^{[9][10]} including onsite measurements and numerical modelling aimed at developing methods that can reduce exposure to indoor VOCs and improve IAQ^{[11][12][13][14][15]}.

Environment chamber experiments and modeling are usually employed to study the formaldehyde emission characteristics such as the decay time^{[16][17]}. However, it is hard to use such experimental data to predict the real conditions in buildings because of uncertainties such as the interaction of human activities and the nature of dynamic

infiltration in real cases^[18]. On the other hand, measurement of the emission rates of formaldehyde in an actual indoor environment could be costly and impractical.

One alternative method is to measure the concentration of formaldehyde in the indoor and outdoor environment and then to back-calculate the effective emission rates^{[19][20][21]}. According to the standard mass-balance model^[21], indoor formaldehyde concentration is inversely proportional to the air exchange rate. A developed mass-balance model was used to differentiate indoor-outdoor concentration ratios and to separate indoor and outdoor sources, but it did not contain building characteristics^{[22][23][24][25][26][27]}. Riley *et al.*^[28] developed the model by specifically considering building operational characteristics (i.e. filtration, penetration, deposition, and ventilation), but some of the parameters, such as air exchange rates, were assumed and the indoor sources of emissions were ignored. This model was then used to predict the proportion of pollutants outdoors and indoors considering natural ventilation. The limitation of these studies is the ignorance of occupant activities^[29]. For example, the model developed by Rackes *et al.*^[30] based on the 24-hour time-average pollutant concentration and occupant respiratory effects was used to predict the indoor pollutant emission rate which accounts for all indoor pollutant sources. In addition, the occupancy schedule was not considered.

Turk *et al.*^[31] investigated 40 houses approximately one-year old or newer with air exchange rates of at least 0.13/h using passive techniques and found the average formaldehyde emission was $90 \pm 55 \mu\text{g/h/m}^2$. The outcomes of this study revealed that

the emission rate of formaldehyde in new buildings was relatively high. The effective emission rate of formaldehyde in typical new houses in North America was estimated to be $44 \pm 16 \mu\text{g/h/m}^2$ using the backward calculation method, but this research assumed that all the formaldehyde is released from building materials^[32]. Kim *et al.*^[33] calculated the average emission rates of formaldehyde using 5 different sources in 19 private reading rooms. This was $45 \pm 38 \mu\text{g/h/m}^2$ and concluded that it was impossible to find all the emission sources.

Because the impact of various sources would significantly influence the accuracy of the results, the emission rate obtained from traditional methods is usually not directly applicable to real buildings.

It was demonstrated in a previous study^[34] that formaldehyde posed the highest risk to people's health. Previous studies have found that the concentrations of formaldehyde in buildings in Chongqing were higher than for other types of VOCs^[35]. It remains questionable whether the formaldehyde concentration in residences considers human interactions and what the emission rate would be. In order to fill the research gap, this study aims to gain a better understanding of the current situation of indoor formaldehyde pollution in residences and to reveal its emission mechanism using onsite measurements and a numerical approach. Furthermore, this study attempted to test whether the formaldehyde emissions from humans will affect the indoor concentration. The work then concentrated on the following aspects: 1) to collect CO₂ real-time data and formaldehyde concentration data via on-site measurement in real residences as well as

their occupancy pattern; 2) to calculate ventilation rates using CO₂ emission as the tracer gas; 3) to develop a robust model for the estimation of formaldehyde emission rates.

Detailed descriptions are presented in the following sections.

2. Method

The onsite measurements were carried out in 42 residences in Chongqing during the period between November 2015 and January 2016. The information about the building characteristics of these residences and the structure of the families living there were obtained through a questionnaire survey involving a brief information sheet. The measurements of the parameters included room dimensions, indoor/outdoor air temperatures and the indoor/outdoor concentrations of CO₂ and formaldehyde.

2.1. Sampling site

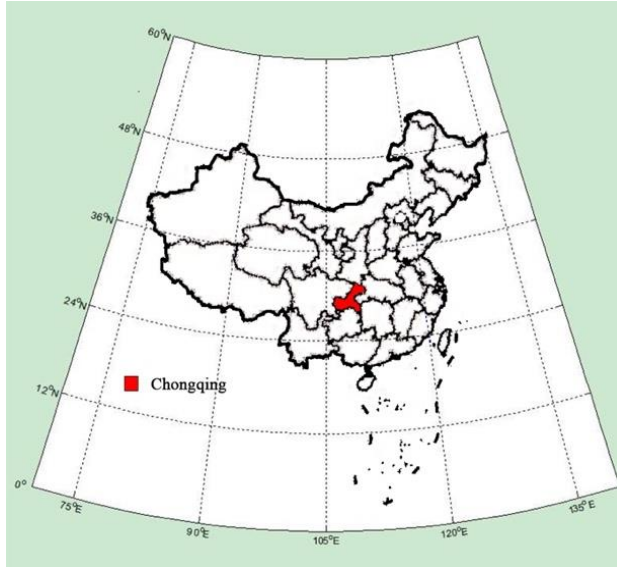
The on-site measurements were conducted in the urban districts of Chongqing, i.e. Shapingba (SPB), Jiangbei (JB), Yubei (YB), Banan (BN), Nanan (NA), Yuzhong (YZ), Dadukou (DDK), Jiulongpo (JLP), and Beibei (BB), as shown in Fig. 1. In 2005, the Chinese government published a national standard ‘Code for Indoor Environmental Pollution Control of Civil Building Engineering’ (GB50325-2005)^[36], this standard required the quality of civil buildings built after 2005 to be of better quality than those built before 2005. The age band of the buildings in this study ranged from 2005 to 2012. Nine of all the measured residences had been renovated; the most recent renovation was 4 years prior to this study. The buildings are reinforced concrete structures. The lowest residential apartment is on the first floor and the highest residential apartment is on the

27th floor. 20 residential apartments are located below the 10th floor. The characteristics of the measured residences are shown in Table 1.

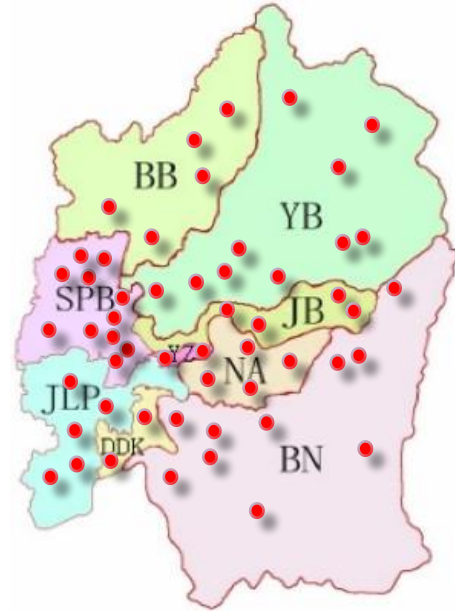
Table 1: Characteristics of residential apartments

Functional room	Living room	Bedroom	Kitchen
Floor Materials	Wood (n=14)	Wood (n=37)	Wood (n=3)
	Tile (n=28)	Tile (n=5)	Tile (n=39)
Wall Materials	Wallpaper (n=15)	Wallpaper (n=15)	Ceramic tile (n=38)
	Latex paint (n=27)	Latex paint (n=27)	Latex paint (n=4)
Ceiling Materials	Paint (n=1)	Paint (n=1)	Paint (n=1)
	Latex paint (n=41)	Latex paint (n=41)	Latex paint (n=41)
Window Materials	Single layer glass (n=39)	Single layer glass (n=37)	Single layer glass (n=37)
	double-layered glass (n=3)	double-layered glass (n=5)	double-layered glass (n=5)
	Wood (n=1)	Wood (n=1)	Wood (n=1)
Window Frame Materials	Aluminum alloy (34)	Aluminum alloy (34)	Aluminum alloy (34)
	Plastic (7)	Plastic (7)	Plastic (7)
Quantity	1 (n=42)	1 (n=5)	1 (n=42)
		2 (n=25)	
		3 (n=12)	

The selection of residences for this research was based on the following criteria: 1) the residences were occupied without any renovation within the last year to avoid excessive concentration values skewing the test results; 2) all the residences had to be at least 100m away from the main roads to avoid the influence of vehicle emissions^[37]; 3) all the residences were located in the main urban areas with relatively high population densities, which is representative of the characteristics of developing urban areas in China; 4) the house owners were informed about the details of the investigation during the whole sampling process; 5) during the field test, natural ventilation was the only way for fresh air to enter the residences.



(a) Location of Chongqing



(b) Location of sampled buildings highlighted within the urban areas of Chongqing

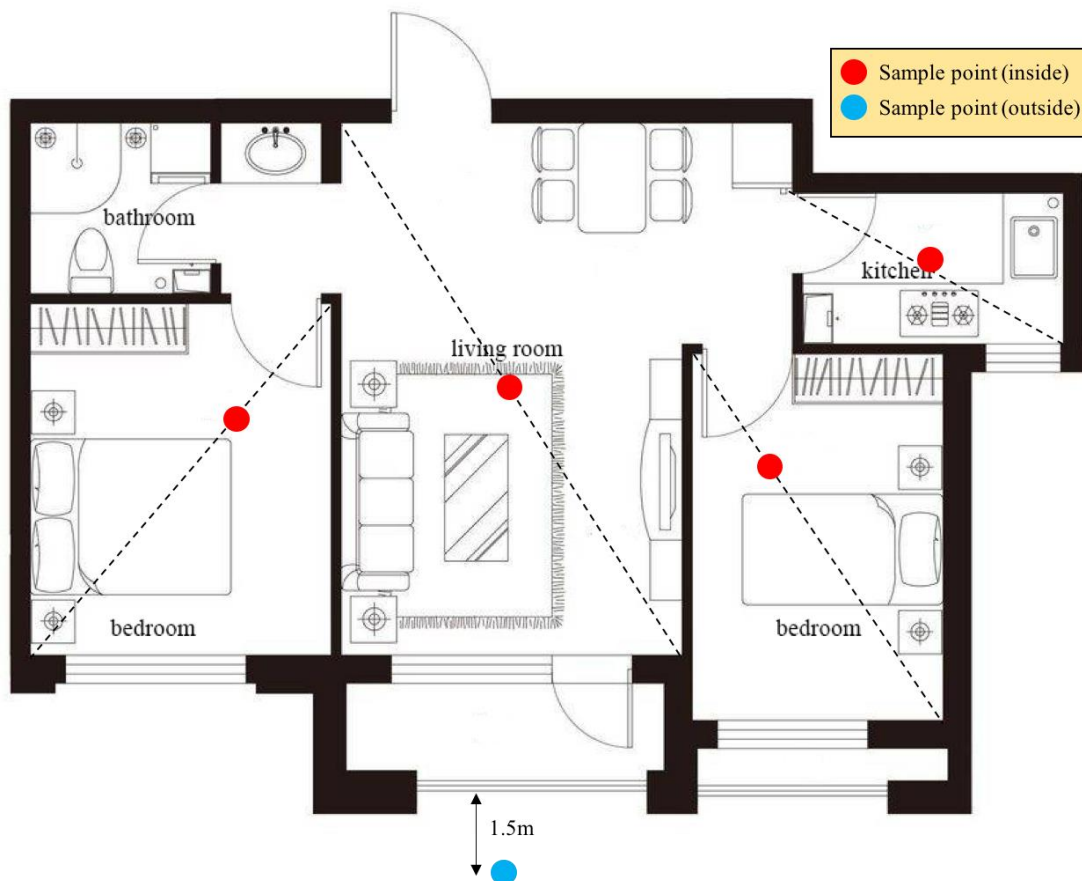
Fig. 1: Illustrates (a) the location of Chongqing and (b) the areas investigated within Chongqing.

2.2. Data collection

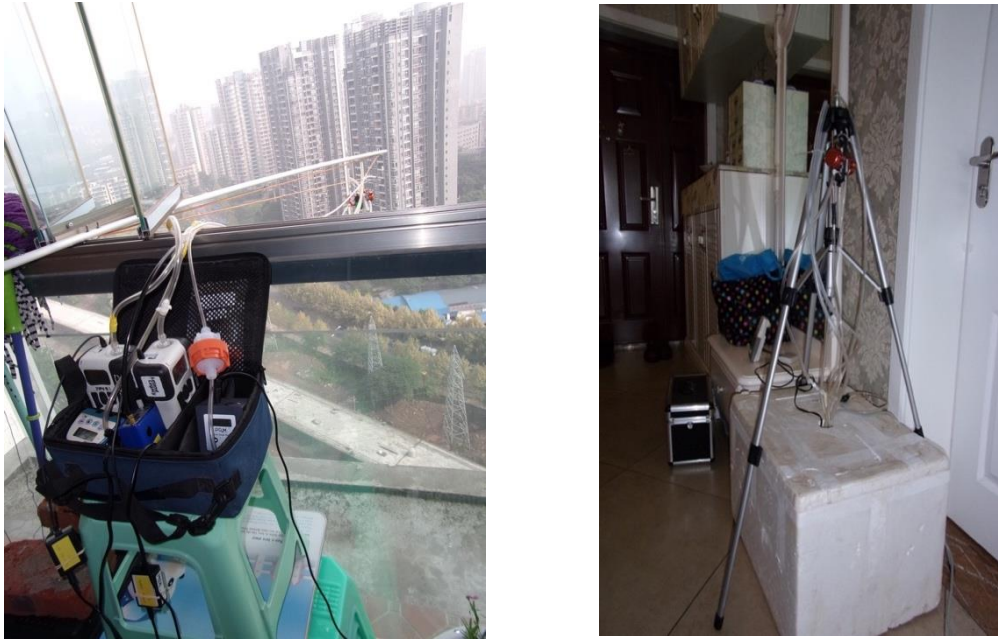
I) Onsite measurement

For formaldehyde, there was one pair of sampling points for each residence, one placed in the living room and the other outdoors. For CO₂, there were four sampling points placed in the living room, bedroom, kitchen and outdoor space respectively. Because formaldehyde concentration levels in different rooms were very similar^[34], this research only measured the formaldehyde concentration in the living room. The indoor sampling points were evenly distributed on the diagonal of each room as shown in Fig. 2 (a) i.e. as a quincunx. The number of sampling points was chosen based on the room size. If the room area was 1) less than 50m², set 1-3 sampling points; 2) 50-100m², set 3-5 sampling points; 3) more than 100m², set at least five sampling points. As all the rooms were smaller than 50m², we only used one measuring point in each room. Samplers were placed at approximately 1.5m above the floor and more than 0.5m from

the walls, windows and doors, or located as centrally as possible within these given constraints^[38]. Figure 2(b) shows a typical arrangement for the locations of sensors in a room and outdoor. The geometric parameters of the residences were measured by a laser distance meter (424D, Fluke Corp.). The outdoor sampling points were set at 1.5m away from the windows. For most Chinese residential apartments, there are only 4 different function rooms: the living room, bedroom, kitchen and bathroom. The bathroom decoration materials are different from those in the other rooms. Moreover, the bathroom always has a mechanical ventilation system and has its door closed most of the time. Hence, it was assumed that the 3 function rooms (living room, bedroom and kitchen) can represent the whole apartment.



(a) The illustration of sampling points in a typical residence



(b) The sampling points in and outside a residence

Fig. 2: The sampling points.

II) Data collection

For each residence, the temperature, relative humidity, and CO₂ concentration indoors and outdoors were recorded continuously for 24 hours, for time intervals of 1 minute. A carbon dioxide detector (Telaire 7001, General Electric Co.) was used to monitor the CO₂ concentration. An automatic temperature and humidity logger (HOBO U-12, Onset Computer Corp.) was used to monitor the temperature and humidity.

III) Air samples

Air samples were collected by a Passive Sampler (Chinese Center for Disease Control and Prevention (CDC)) for 24 hours. According to the ‘Chinese Standard Method for Hygienic Examination of Formaldehyde in the Air of Residential Areas’ - the Spectrophotometric method (GB/T 16129-1995)^[39] - formaldehyde was absorbed by a 4-amino-3-hydrazino-5-mercapto-1,2,4-triazole (AHMT) solution and analyzed using a UV-VIS spectrometer (7205, Xinmao Corp.) at 550nm. Samples must be placed in a pouch, and the pouch must be sealed, refrigerated and stored at 4°C before and after use. The sample must be analyzed within three weeks. The detection limit (MDL) of this

method was $0.01\text{mg}/\text{m}^3$ and the relative standard deviation (RSD) was less than 10%. The recovery rates for formaldehyde samplers ranged from 90%-100% and the limit of detection (LOD) value of this method is $0.22\mu\text{g}/10\text{ml}$. Table 2 shows the parameters of the measuring instruments.

Table 2: Parameters of the measuring instruments.

Model	Company	Measuring variable	Range	Accuracy	Resolution
Telaire 7001	General Electric Co.	CO ₂	0-2500 ppm	± 50 ppm or 5% of reading	± 1 ppm
HOBO U-12	General Electric Co.	Temperature	-20-70°C	$\pm 0.35^\circ\text{C}$ from 0-50°C	0.03°C
		Humidity	5%-95%RH	$\pm 2.5\%$ from 10%-90%RH	0.05%RH
Passive Sampler	CDC	Formaldehyde	10-130 $\mu\text{g}/\text{m}^3$	Lower than 10%	-
7205	Xinmao Corp.	Wavelength	325-1000nm	$\pm 2\text{nm}$	$\pm 1\%$ T
424D	Fluke Corp.	Distance	0-100m	$\pm 1\text{mm}$	$\pm 1\text{mm}$

2.3. Statistical analysis

To investigate the distribution of these data and their principal characteristics, statistical analyses were performed using SPSS (a Statistical Package for Social Science, Version 25, IBM Corp.) to ensure a significance level of $p=0.05$ as the benchmark. In addition, Pearson Correlation is used to investigate if there is a statistically significant difference between indoor and outdoor formaldehyde levels, and the Shapiro-Wilks test was applied to examine whether the pollution concentration dataset fit revealed a normal or lognormal distribution. In this research, the distribution of formaldehyde concentration matches the lognormal distribution, according to the P-P plot. Since

formaldehyde concentrations were not normally distributed, the lognormal transformed formaldehyde concentrations were used in the Monte Carlo simulations as explained in Section 2.4.

2.4. Uncertainty and sensitivity analysis

A prediction using single point values can result in great uncertainty^[40], whereas stochastic modeling can provide a more accurate emission rate compared with one that only uses field test data. The building characteristics, outdoor and indoor concentrations, and the number of people staying in a residence are useful when the Monte Carlo method is used to estimate the emission rate. Monte Carlo simulations are used to model the probability of different outcomes in a process that cannot easily be predicted due to the intervention of random variables. It is a technique used to understand the impact of risk and uncertainty in prediction and forecasting models and relies on repeated random sampling to obtain numerical results. A sensitivity analysis determines how different values of an independent variable impact on a particular dependent variable under a given set of assumptions. This technique is used within specific boundaries that depend on one or more input variables, such as the effect that changes in independent variables have on the dependent variable. In this study, the Monte-Carlo simulation and sensitivity analysis were conducted using Oracle Crystal Ball software (Fusion Edition, V. 11.1.2.4). 1,000,000 trials of Monte-Carlo simulations were performed to calculate the emission rate^[41].

2.5. Ventilation airflow rate model

The tracer gas method is one of the approaches that can be used to calculate the air exchange rate in naturally ventilated residences, however, its use is limited because 1) tracer gas equipment takes up a lot of space; 2) tracer gas and indoor air need to be evenly mixed by a hybrid fan first^{[42][43]}.

Although using CO₂ as the tracer gas may result in uncertainties, it is the best choice for field tests in residences. In this study, CO₂ was used as the tracer gas to calculate the indoor ventilation airflow rate. The CO₂ concentration rises when people stay in a room and decreases when they leave. A calculation method - the PIT (Parametric Iteration Technique) - is used to calculate the air exchange rate^[44]. The increase of CO₂ concentration (Δc) for each time interval ($\Delta \tau$) is related to the instantaneous airflow rate as in the equation below which assumes a uniform distribution of indoor CO₂:

$$\Delta c = \frac{\Delta \tau}{V_a} [F_{co_2} - N_a V_a (c_1 - c_{out})] \quad (1)$$

$$\Delta c = c_2 - c_1 \quad (2)$$

$$F_{co_2} = RQ \frac{0.00056028H^{0.725}W^{0.425}M}{(0.23RQ+0.77)} \quad (3)$$

Where N_a (h⁻¹) is the ventilation airflow rate; c_1 (ppm) is the concentration of CO₂ in the indoor environment at the beginning; c_2 (ppm) is the subsequent indoor CO₂ concentration; c_{out} (ppm) is the concentration of CO₂ in the outdoor environment; $\Delta \tau$ (s) is the time step; Δc (ppm) is the CO₂ concentration change during $\Delta \tau$; V_a (m³) is the room volume; F_{co_2} (m³/s) is the emission rate in the indoor environment; RQ is the respiratory quotient, treated as a constant = 0.83; H (cm) is the height of a person in the

indoor environment; W (kg) is the weight of a person in the indoor environment; and M (met) is the metabolic rate.

The relationship between indoor CO₂ concentrations at times τ_k and τ_{k-1} can be obtained:

$$C_1(k) = \frac{\Delta\tau}{V_a} \{FR_{all} - N_a V_a [C_1(k-1) - C_{out}]\} + C_1(k-1) \quad (4)$$

where FR_{all} (m³/s) is the CO₂ emission rate from all the occupants.

A theoretical exponential curve was obtained by iteratively calculating the concentration for each time step. Then the least squares method was used to fit the theoretical exponential curve and the measured data. At a particular moment, the actual indoor concentration is $C_1(k)$ and the predicted indoor concentration is $C_1'(k)$. From the result of the optimal fitting, the ventilation airflow rate is calculated to achieve the the smallest error.

$$\text{Error} = (C_1(k) - C_1'(k))^2 \quad (5)$$

The outdoor concentration of CO₂ barely changes during the measurements. Therefore, this study used the outdoor concentration of CO₂ recorded 30 minutes before the field test as the background concentration and recorded the occupant's time distribution in the indoor environment, the height and weight of the occupant, the volume of rooms, and the arrangement of doors and windows during the test.

The ventilation airflow rate was calculated by using MATLAB (R2015b, MathWorks, Inc.).

2.6. IAQ model

A steady-state model was used in this research since the data for the concentration of formaldehyde was not dynamic but averaged over 24 hours. Moreover, the fluctuations in the CO₂ concentration were also unsuitable for dynamic modeling. For any indoor pollutant, the change of concentration is related to its source emission and exhaust rates^{[21][30]}, see equation (6) below:

$$C_e = N(C_{in} - C_{out}) \quad (6)$$

Where C_e is the emission rate ($\mu\text{g}/\text{m}^3/\text{h}$); N (h^{-1}) is the air exchange rate, C_{in} ($\mu\text{g}/\text{m}^3$) is the indoor concentration; and C_{out} ($\mu\text{g}/\text{m}^3$) is the outdoor concentration.

The air exchange rate (N) is the total of the ventilation exchange rate (N_{vent}) and the infiltration rate (N_{inf}).

$$N = N_{vent} + N_{inf} \quad (7)$$

Also,

$$N = \frac{Q_{vent} + Q_{inf}}{V_a} \quad (8)$$

Where Q_{vent} (m^3/h) is the ventilation airflow rate and Q_{inf} (m^3/h) is the infiltration airflow rate, V_a (m^3) is the volume of the room.

$$C_e V_a = (C_{in} - C_{out})(Q_{vent} + Q_{inf}) \quad (9)$$

$$C_e V_a = E_p P_a + E_a A_a \quad (10)$$

E_p ($\mu\text{g}/\text{h}/\text{occ}$) is the emission rate per-occupant and E_a ($\mu\text{g}/\text{h}/\text{m}^2$) is the emission rate per-floor-area. P_a (occ) is the number of occupants and A_a (m^2) is the floor area.

$$C_{in} = C_{out} + \frac{E_p P_a + E_a A_a}{Q_{vent} + Q_{inf}} \quad (11)$$

In this research, the concentration levels of formaldehyde were divided into night-time and day-time, because occupants' activities are very different over a 24-hour period, meaning that time distribution must be taken into account. In this research, we separate the whole day into two parts, one is day-time, and the other night-time. We defined the night as from 18:30 pm. to 07:30 am. The rest of the time is considered as day-time.

2.7. Formaldehyde emission rate

According to the IAQ model^[30], the value of E_a can be calculated as follows:

$$E_a = \frac{(C_{in} - C_{out})(Q_{vent} + Q_{inf}) - E_p P_a}{A_a} \quad (12)$$

$$E_a = \frac{(C_{in} - C_{out})(NV_a) - E_p P_a}{A_a} \quad (13)$$

where C means the concentration of formaldehyde and E_a means the per-floor-area emission rate of formaldehyde.

This model contains the air exchange factor and in particular the human breathing factor. It was shown to provide more accurate result than previous studies.

When the indoor concentrations were smaller than the outdoor concentration, the above equation cannot give a reasonable result. Also, when the occupants' exhalation emissions were high enough to make the denominator negative, no reasonable result could be obtained. In this research, the data were filtered to ensure reasonable emission values.

3. Results and discussion

3.1. Formaldehyde concentration

The concentration of formaldehyde was conducted by field measurements. The indoor formaldehyde sources were indoor building materials and occupant exhalation. The data set consists of 42 indoor and 42 outdoor formaldehyde concentration measurements.

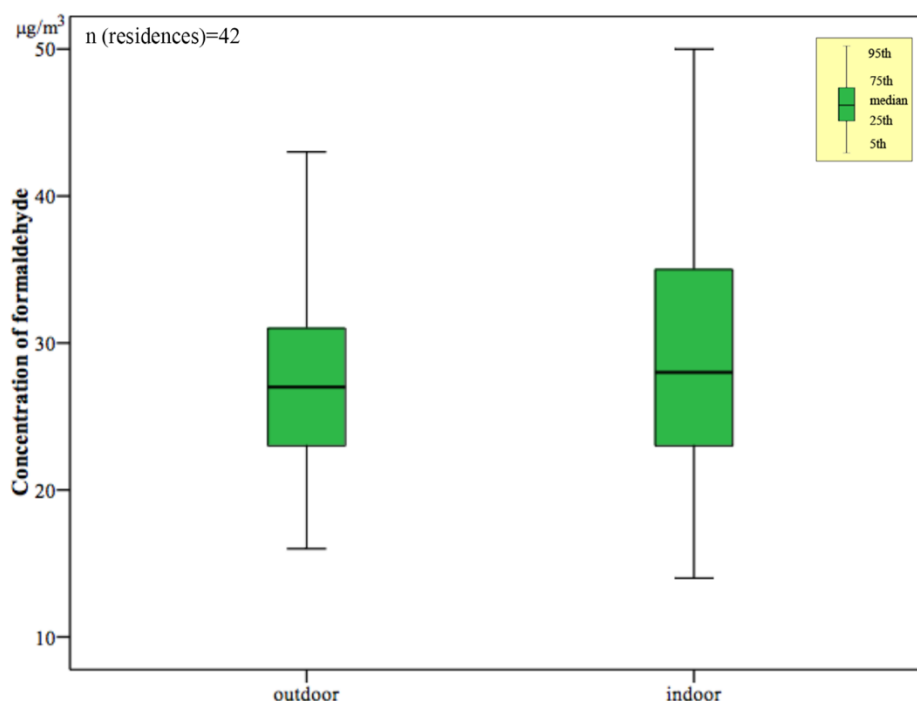


Fig. 3: The concentration of formaldehyde.

As shown in Fig. 3, the indoor concentration of formaldehyde was slightly higher than that of the outdoor environment and the mean concentrations of formaldehyde indoors and outdoors were $30.12\mu\text{g}/\text{m}^3$ and $27.80\mu\text{g}/\text{m}^3$ respectively. However, the 5th percentile value to the 95th percentile value of the indoor and outdoor formaldehyde concentrations were $18.10\mu\text{g}/\text{m}^3$ to $49.00\mu\text{g}/\text{m}^3$ and $19.10\mu\text{g}/\text{m}^3$ to $42.90\mu\text{g}/\text{m}^3$, respectively. It shows that it is possible for some residences to have a lower indoor

formaldehyde concentration than outdoor. The I/O ratios were in the range of 0.51 to 1.88, around 60% of which were greater than 1. Fig. 4 indicates the significant contribution from indoor sources. According to the statistical analyses results, the p value of Pearson correlation is 0.487, and Pearson correlation coefficient is 0.112 which means indoor formaldehyde concentration levels are not significantly associated with outdoor formaldehyde concentration levels.

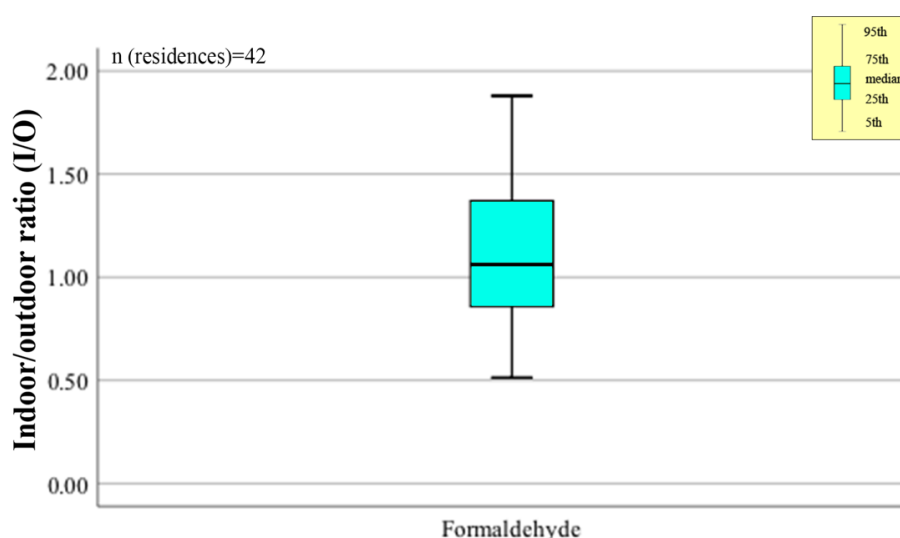


Fig. 4: The I/O ratio of formaldehyde.

Table 3: Formaldehyde concentration limits

Pollutant	Class 1 civil engineering	Class 2 civil engineering
Formaldehyde ($\mu\text{g}/\text{m}^3$)	≤ 80	≤ 100

According to the ‘Chinese Standard Code for Indoor Environmental Pollution Control of Civil Building Engineering’ (GB50325-2010)^[45], the concentration limit of formaldehyde in civil buildings is $80\mu\text{g}/\text{m}^3$ for Class 1 civil buildings, as shown in Table 3 and the concentrations of formaldehyde did not exceed this limit. China only provides guideline values for short-term (1h) exposure to the indoor environment, which are set out in the ‘Chinese Standard’ (GB/T 18883-2002)^[38], and the concentration limit for formaldehyde is $100\mu\text{g}/\text{m}^3$. In this study, the mean concentration of formaldehyde

measured in Chongqing residences was within the short-term limit. Here it should be mentioned that the short-term exposure to formaldehyde in residences might have no perceptible effect on human health.

Formaldehyde was detected with a frequency of 100%, which means that formaldehyde was one of the common pollutants in Chongqing residences. Compared with other research studies in China (Table 4), the concentration of formaldehyde in the indoor environment was similar to the concentration in Dalian ($32.9\mu\text{g}/\text{m}^3$)^[46], lower than the concentration in Tianjin ($54\mu\text{g}/\text{m}^3$)^[47], Harbin ($72.5\mu\text{g}/\text{m}^3$)^[48] and Beijing ($131\mu\text{g}/\text{m}^3$)^[4] and slightly higher than the concentration in Chongqing ($20\mu\text{g}/\text{m}^3$) obtained by another study^[34]. The lower concentration of formaldehyde in Chongqing published in a previous study may come from its use of active sampling and different chemical analysis methods^[34]. Compared to results from other countries (Table 4), except for Helsinki ($39.96\mu\text{g}/\text{m}^3$)^[49] and Strasbourg ($32.3\mu\text{g}/\text{m}^3$)^[50] which were slightly higher than the current research, other places like Quebec^[51], New York^[52], Los Angeles^[52], Houston and Elizabeth^[53] gave lower results than in the current research, especially the concentration of formaldehyde of Melbourne^[54] which was only $14.64\mu\text{g}/\text{m}^3$. Comparing the formaldehyde concentration of Chinese cities to those from other developed countries, China's indoor formaldehyde concentration level is significantly higher.

Table 4: Comparison of formaldehyde level (mean) tests of air samples from residences ($\mu\text{g}/\text{m}^3$)

Reference	Country	Sampling information	type	formaldehyde
Current study	Chongqing, China	n=42		30.12
Huang <i>et al.</i> (2013)	Beijing, China	n=410		131

			Bedroom	50
			Living room	100
Zhu <i>et al.</i> (2013)	Harbin, China	n=240	Kitchen	30
			Study room	110
			Mean	72.5
Zhou <i>et al.</i> (2011)	Tianjing, China	n=10		54
			Bedroom	33.5
Guo <i>et al.</i> (2013)	Dalian, China	n=59	Kitchen	32.3
			Mean	32.9
			Bedroom	23.2
Cheng <i>et al.</i> (2018)	Chongqing, China	n=50	Living room	21.3
			Kitchen	15.6
Jouni <i>et al.</i> (2011)	Helsinki, Finland	n=15		39.96
Marchand <i>et al.</i> (2008)	Strasbourg, France	n=143		32.3
Nicolas <i>et al.</i> (2006)	Quebec, Canada	n=96		29.5
	Los Angeles, USA	n=73		
Hun <i>et al.</i> (2010)	Elizabeth, USA	n=58	Mean	20.64
	Houston, USA	n=48		
Sax <i>et al.</i> (2006)	Los Angeles, USA	n=41		19.3
Sax <i>et al.</i> (2006)	New York, USA	n=46		17.7
Molloy <i>et al.</i> (2012)	Melbourne, Australia	n=40		14.64

The concentration levels of formaldehyde in Chongqing's residences were relatively low compared to those in other Chinese cities because the measured residences were compliant with the mandatory standard and without any renovation within the last year.

3.2. Occupancy characteristics

In this study, we measured the CO₂ concentration in 3 functional rooms, namely the living room, bedroom, and kitchen. By analyzing the real-time CO₂ concentration data together with the information sheet completed by the occupants, the time distribution in residences was obtained. The ascending and descending segments of the CO₂ concentration curve can directly reflect whether the room was occupied. Almost

every measured family has one young kid, and one retired people who took care of the young kid. It is common in China that three generation live in one flat and grandparents look after their grandchild. In addition, Chinese people have a habit of eating hot dishes for lunch and sleeping after lunch at home. According to this situation, the measured residences have human activities most of the time.

Compared with the CO₂ concentration data for the 42 residences, it was easy to define the time period occupants spent in different functional rooms. Table 5 present the overall situation of occupants time distribution in different functional rooms. Occupants always stay at home during these periods.

Table 5: People occupancy schedule in Chongqing residences by real-time CO₂ concentration data

Different functional rooms	Time distribution
Living room	7:30am-8am; 12midday-1pm; 7:30pm-10:30pm
Bedroom	10:30pm-7am; 1pm-2pm
Kitchen	7am-7:30am; 11am-12midday; 6:30pm-7:30pm

The person's lifetime distribution statistical data can also be calculated from "Time Use Patterns in China"^[55] (Table 6) which provides people's time use patterns in different environments in different Chinese cities.

Table 6: People occupancy schedule in Chongqing residences by "Time Use Patterns in China"

Parameters	Male	Female
Occupancy schedule in residence (min/day)	932	1018
Living room	27.3%	30%
Bedroom	57.4%	53.2%
Kitchen	15.3%	16.8%

The time fraction for each room was calculated using real-time CO₂ concentration data and proved to be very similar to Table 6 which is based on the occupancy schedule in Chongqing residences calculated using "Time Use Patterns in China". This means the time distribution of occupants who lived in the measured residential apartments were

consistent with the time distribution data for the Chongqing urban area.

The whole day was separated into two parts in this study and the occupants' time distribution categorized accordingly into day-time and night-time. For the living room, 12:00 midday to 1p.m. and 7:30p.m. to 10:30p.m. were selected as the daytime and nighttime patterns, respectively; for the bedroom, there were only two time periods, 10:30p.m.-7a.m.; 1p.m.-2p.m.; for the kitchen, 11:00 a.m. to 12:00 midday and 6:30p.m. to 7:30p.m. were chosen as the daytime part and nighttime part, respectively. This occupancy pattern ensures that during these time period, there are always occupants staying at home.

People's metabolism makes the human body a source of pollution, and people can emit VOCs such as formaldehyde (the primary emission from the skin^[56]) within their exhaled breath^[6].

There is sufficient information in the literature to develop reasonable statistical models for human breath emissions. In this study, an individual's formaldehyde breath emission rate was obtained from the literature. The emission rate per-occupant of 4.06 $\mu\text{g/h/occ}$, which is based on data from Rackes *et al.*^[30], was used. The number of occupants in rooms was also recorded (3 ± 1 (mean \pm SD)). The number of occupants in the residences for each sampling duration were collected by questionnaire.

3.3. Air exchange rate

To compare the differences in indoor and outdoor concentrations and the differences between night and daytime, the continuous 24-hour data were averaged as shown in Fig. 5. Using the average CO₂ concentration to describe the residence CO₂ concentration level is more accurate than the time dependent values. During some periods for example, the CO₂ concentration is very low in some rooms, e.g. the kitchen when people are not cooking. The average CO₂ concentration can express the overall level of indoor CO₂ concentration. The use of CO₂ averaged over the whole household could better match the formaldehyde measurement for the whole apartment as well.

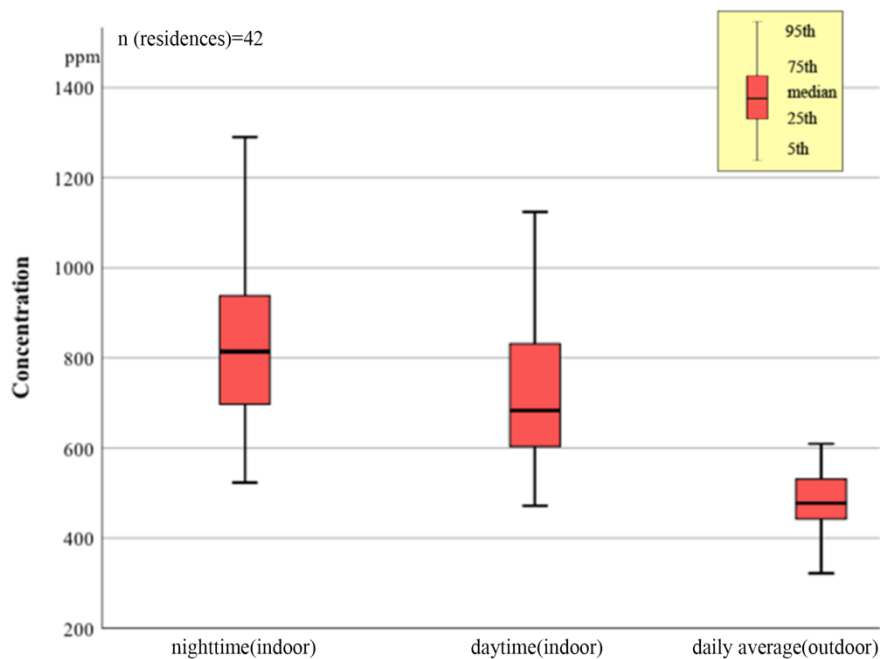


Fig. 5: The concentrations of CO₂.

From Fig. 5 we can see that the mean concentrations of indoor CO₂ during night-time, day-time, and outdoors were 753ppm, 705ppm, and 475ppm respectively. The outdoors CO₂ concentration had the lowest value. Sometimes, the indoor CO₂

concentration was found to exceed the threshold of 1000ppm in the ‘Chinese National Standard (GB/T 18883-2002)’^[38].

Chongqing is located in the southwest of China on the upper-middle reaches of the Yangtze River. It has a monsoon-influenced, humid, subtropical climate similar to Shanghai, experiencing very wet conditions for most of the year. Winters in Chongqing are short and somewhat mild, but damp and overcast. All the measurements in this study were conducted during winter at a time when there were no central heating systems used in Chongqing. People in Chongqing mostly opened the windows (natural ventilation) to refresh the indoor air during the daytime and closed them to keep warm when they stayed at home^[57], which may cause the CO₂ concentrations to be higher during the night-time than that in the daytime.

In this study, the *per-capita* living space in urban residences in Chongqing is $38.0 \pm 11.0 \text{ m}^2$. According to the Chinese standard ‘Design Code for Heating Ventilation and Air Conditioning Of Civil Buildings’ (GB 50736-2012)^[58], when the *per-capita* living space is between 20 m^2 and 50 m^2 , the ventilation air flow rate should be 0.5 h^{-1} ; when the *per-capita* living space is more than 50 m^2 , the ventilation air flow rate should be 0.45 h^{-1} .

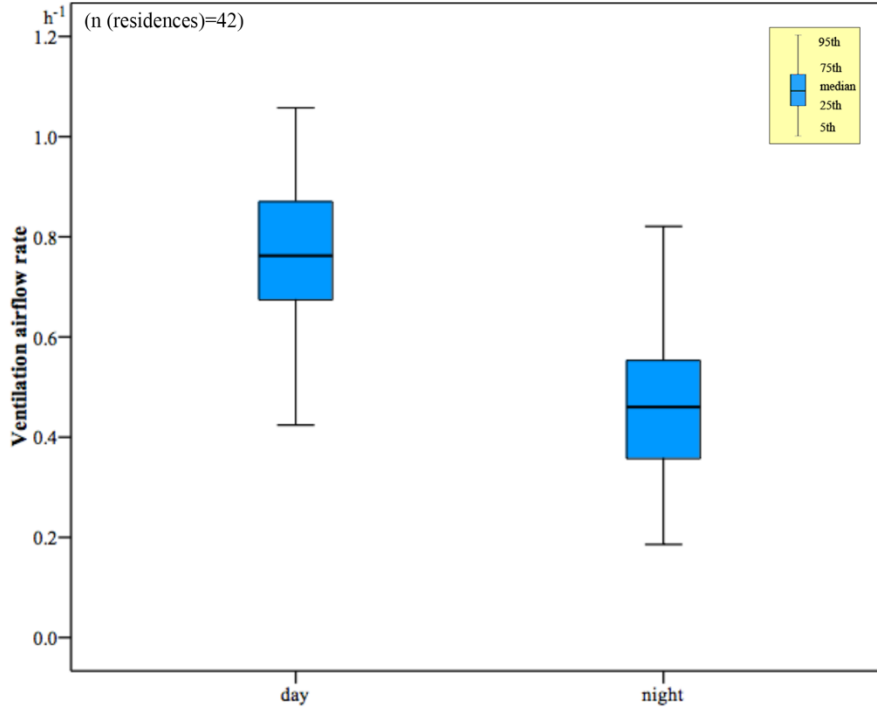


Fig. 6: Ventilation airflow rate.

Because we measured the CO₂ concentration in at least 3 rooms (the living room, bedroom, and kitchen) in each residence, we calculated ventilation airflow rate for each room and also used the average CO₂ concentration to calculate the real ventilation airflow rate for the dwelling. The relative error is around 10%. With the low ventilation airflow rate, if the concentration levels of CO₂ in different functional rooms are different, the mixing of the indoor air might not have been very good, hence the difference.

According to Fig. 6, the maximum value of the ventilation airflow rate during daytime was 1.39h⁻¹ and the minimum value was 0.43h⁻¹, giving a mean value of 0.79±0.20(SD)h⁻¹. The maximum value of the ventilation airflow rate at night was 0.83h⁻¹, and the minimum value was 0.19h⁻¹, giving a mean value of 0.46±0.13(SD)h⁻¹. At night, for most of the residences, the air exchange rate was higher than 0.4h⁻¹, but there were still 32.7% residences with an air exchange rate lower than 0.4h⁻¹. Compared to

the critical value of 0.5h^{-1} , the 25th percentile value of the daytime ventilation airflow rate was over 0.6h^{-1} , and the 5th percentile value was lower than 0.5h^{-1} ; the 5th percentile value, 25th percentile value and median value of the night ventilation airflow rate were lower than 0.5h^{-1} whilst the 75th percentile value was over 0.5h^{-1} . At night, the lower ventilation airflow rate did not meet the GB 50736-2012 standard hence the pollutants would have been retained in the indoor environment and could not effectively be exhausted to the outside.

Because people stay and sleep at home at night, the air quality at night would have more influence on human health. During the night, the windows and doors were closed and the ventilation rate low, which could lead to an increased pollutant concentration^[59].

3.4. Emission rates

Emission sources in the indoor environment are very unpredictable and consist of various categories. This makes it difficult to calculate the emission rates for every source. A time-averaged effective emission rate calculated per m^2 of floor area was used to predict the formaldehyde emission rate in residences. Table 7 lists the parameters that are used for the Monte Carlo assessments.

Table 7: Summary of the building characteristics and environmental parameters (n=25)

Parameters		Median	Mean	SD
$N_{vent} (\text{h}^{-1})$	Daytime	0.83	0.86	0.17
	Night	0.52	0.50	0.13
	Mean	0.68	0.68	0.24
The number of occupants		3	3	1

C_{in} ($\mu\text{g}/\text{m}^3$)	32.01	34.87	8.71
C_{out} ($\mu\text{g}/\text{m}^3$)	25.00	25.04	4.75
Area of residence (A_a , m^2)	100.00	98.72	31.13

To assess the emission rate, the daytime emission rate and nighttime emission rate were considered separately, and the average emission rate obtained.

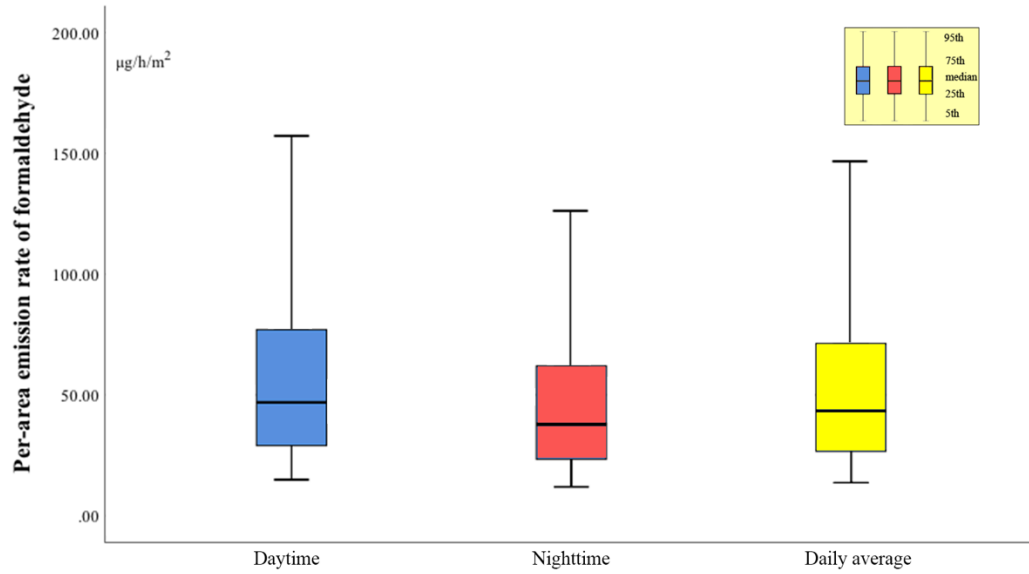


Fig. 7: Emission rates.

The value of the emission rate calculated from Equation 12 was very small and even negative in some cases; i.e. outdoor formaldehyde concentrations may be higher than indoor concentrations. To avoid this situation, the concentrations were compared before calculation, and when the indoor concentration was smaller than that outdoors, this data was removed. This situation appeared in 17 measured residences, hence we used the rest of the data on indoor and outdoor formaldehyde concentrations and ventilation rates for the rest of residences (25 in total) to calculate the emission rate.

According to Fig. 7, the daytime emission rate is $61.82 \pm 52.39 \mu\text{g}/\text{h}/\text{m}^2$ (mean \pm SD), the night emission is $49.69 \pm 42.13 \mu\text{g}/\text{h}/\text{m}^2$ (mean \pm SD) and the average emission is $57.20 \pm 48.79 \mu\text{g}/\text{h}/\text{m}^2$ (mean \pm SD). The range of emission rates was wide, one of the reasons being that the quality of building materials, such as wallpaper, wardrobes, and

cabinets, might be different in different residences. The sensitivity analysis indicates that the indoor and outdoor concentrations are two decisive factors for the emission rate. When the ventilation rate is low, the indoor emission sources are the most important for the determining the indoor formaldehyde concentration; however, if the ventilation rate is high, the outdoor concentration becomes the most important source.

In this study, we separated the indoor sources into two groups; one is the main source - like consumer products, building and decoration materials - and the other one is the human emission source. By calculating the contribution of human formaldehyde emission and comparing this with the other main sources, the human emission can in some cases be ignored. In residences, the number of occupants is mostly lower than 10, making the contribution of human emission lower than 1%.

3.5. Sensitivity analysis

Fig. 8 shows the influence of different factors on the formaldehyde emission rate. According to this figure, the concentration difference between indoors and outdoors is the most significant factor (around 64%) that influences the emission rate, whereas the contribution from the residents is around 35%, with the air exchange rate having the lowest influence (lower than 2%). That is, formaldehyde released from humans has no significant impact on the emission rate level. Hence, to control the indoor air pollutant concentration, controlling the source of the pollutant has a better effect on reducing the indoor concentration than increasing the air exchange rate, which means that using building materials that emit less formaldehyde can significantly lower the indoor pollutant concentration. Natural ventilation may not be the best way to reduce the indoor formaldehyde concentration because the indoor and outdoor concentrations are just slightly different; using an air cleaner and filter might have a better, more efficient,

effect for improving the indoor environment.

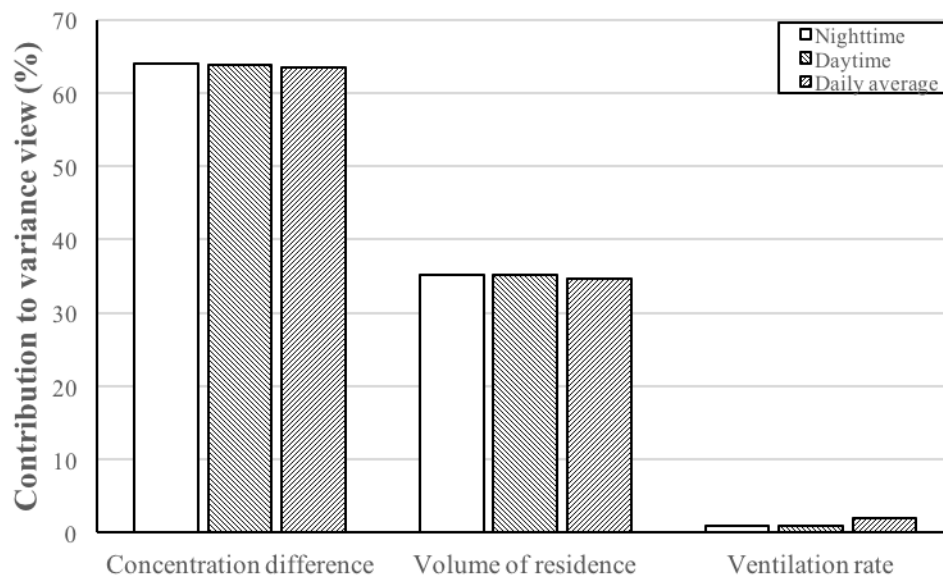


Fig. 8: Contribution to the emission rate.

4. Conclusions

This study measured the formaldehyde and CO₂ concentrations in residential buildings in Chongqing, China to provide a better understanding of indoor formaldehyde pollution and understand its emission sources using on-site measurement and a numerical approach. A robust prediction model for formaldehyde using CO₂ concentration as tracer gas has been developed. The effect of formaldehyde emissions from humans and its affect on the indoor concentration was also investigated. Results indicate that:

- The concentration levels of formaldehyde in Chongqing's residences built between 2005 and 2012 without any interior decoration were relatively low compared to other Chinese cities. The indoor concentration of formaldehyde was slightly higher than that in the outdoor environment. The significant

contribution to indoor concentration levels was from indoor sources.

- The average emission rate in urban Chongqing residences is about $57.20 \pm 48.79 \mu\text{g}/\text{h}/\text{m}^2$ (mean \pm SD), a value that is rather high compared to previous studies^{[32][33]}. During day-time hours, the emission rate is slightly higher than that at night-time. This might be due to differences in ventilation rates and personal activity during these two different periods.
- The occupants' formaldehyde emissions can be ignored in residences, due to their negligible contribution to the total emissions. However, in some spaces with a high population density, like the cabin of a plane, a train, bus or club, human emission rates of formaldehyde should be taken into consideration.

Limitations

In this study, all the measurements were conducted in winter. The emission of pollutants from building materials was highly correlated to the local temperature. Simultaneous data could be used to calculate the emission rate in future studies to investigate the real-time emission rates when the window and door status as well as occupants' activities, like cooking and smoking, are available. To study the influence of outdoor formaldehyde concentration levels on the indoor environment, the relationship between the indoor and outdoor air qualities should be further studied.

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