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The influence of indoor environmental factors on toluene uptake rate of a tube-type diffusive sampler

Yan Wang [a,b], Tao Yu [c,d], Jinhan Mo [a,b,e,*]

[a] Department of Building Science, Tsinghua University, Beijing, 100084, China
[b] Beijing Key Laboratory of Indoor Air Quality Evaluation and Control, Beijing, 100084, China
[c] Wuhan Second Ship Design and Research Institute, Wuhan, 430205, China
[d] School of Energy and Power Engineering, Beihang University, Beijing, 100191, China
[e] Key Laboratory of Eco Planning & Green Building, Ministry of Education (Tsinghua University), Beijing, 100084, China

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ABSTRACT

Obtaining effective and reliable uptake rates is crucial for applying diffusive samplers in monitoring indoor gaseous pollutants. Diffusive uptake rates are mainly reported for the workplace and environmental monitoring in a short-term exposure (such as 8 h) at high concentrations or long-term exposure (such as 7d, 14d) at lower concentrations. There are relatively few reports on the 24 h uptake rate and the changes in uptake rate under different environmental conditions. This study explores the influence of indoor environmental factors such as temperature, humidity and concentration on the 24 h toluene uptake rate of Tenax TA diffusive samplers. Fifteen humidity control experimental cases are conducted under three concentration ranges (0.2–0.8 mg/m^3, 1–1.5 mg/m^3, 2–3 mg/m^3) with temperature recorded simultaneously. Through correlation analysis and linear regression, it is found that temperature, humidity and concentration have statistically significant effects on the toluene uptake rate. The increase in temperature and humidity would cause a decrease up to 25.9% and 46.2% in toluene uptake rate respectively, while concentration has a positive impact. Accordingly, an empirical equation is proposed to predict the uptake rate under environmental effects. The reported effect of environmental factors will help obtain the accurate uptake rates of tube-type diffusive samplers for gaseous pollutants.

Abbreviations

UR	uptake rate
UR_{ideal}	the ideal uptake rate
UR_a	active sampling rate
UR_p	diffusive uptake rate
$UR_{p,24h}$	the 24 h diffusive uptake rate
VOC	volatile organic compound
GC-MS	chromatography-mass spectrometry
RH	relative humidity
RSD	relative standard deviation

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* Corresponding author. Department of Building Science, Tsinghua University, Beijing, 100084, China.
E-mail address: mojinhan@tsinghua.edu.cn (J. Mo).

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1. Introduction

Monitoring indoor volatile organic compounds (VOCs), such as toluene, is crucial to evaluate their exposure risk [1,2]. Volatile organic compounds found in indoor air investigations come from sources such as building materials, paints, furnishing materials, tobacco smoke, personal care, and household and cleaning materials [3]. Most VOCs are considered as potential candidates involved in the establishment of sick building syndromes and often associated with clinical manifestations like allergy, rhinitis, asthma and conjunctivitis. Continuous exposure to indoor air pollutants may even cause respiratory and cardiovascular diseases, eventually contributing to the so-called 'sick building syndrome' (SBS) and 'building-related illnesses' (BRI). Toluene is a recognized cause of neurodevelopmental disorders and subclinical brain dysfunction. Exposure to toluene during early fetal development can cause brain injury at doses much lower than those affecting adult brain function [4]. Curative treatment of these problems requires the precise identification and assessment of the VOC pollutants. Passive or diffusive sampling using sorbent tubes has become a widely accepted method [3,5–8]. It offers certain advantages over the active and pumped sampling, including no electrical power, simplicity of use, reduction of operation cost, and suitability for personal exposure measurements [9–11]. Some passive sampling devices were developed and used in environmental or workplace applications [12–14]. However, the challenge in using passive sampling is how to convert the pollutant mass determined in the collection medium to the concentration in the air [15]. Because no pump is used, the concept of sampling rate or sample volume commonly used in active sampling is not suited for passive sampling. Instead, passive sampling uses the term uptake rate to calculate the average concentration over the exposure time. The uptake rate often expressed in units of mL/min or as ng/(ppm•min), is the speed at which a VOC diffuses from surrounding air onto the collection medium [16].

Based on Fick's first law of diffusion [17], the ideal uptake rate of a passive sampler can be calculated and is constant throughout the sampling time, provided there are no variations in temperature and the pollutant concentration [16,18]. However, the actual uptake rate will be more or less different from the ideal one [15]. The actual uptake rate varies according to several aspects, including sampling time [16,19], environmental factors (temperature, relative humidity, and wind speed) [20,21], VOC concentrations [22,23]. It was not very reliable to directly use the constant uptake rates provided by manufacturers to calculate VOC concentrations, which will lead to quantification errors [24]. For example, some researchers have studied the radial sampler behavior in laboratory-controlled atmospheres and outdoors [24,25]. Plaisance et al. found that the temperature and humidity significantly influence the O_3 uptake rate on the radial diffusive sampler [26]. And the uncertainty of benzene uptake rate was found mainly caused by environmental temperatures [27]. However, there were few studies on the influence of environmental factors on the tube-type Tenax TA diffusive samplers. Existing studies about Tenax TA diffusive samplers mainly focused on the influence of different adsorbents and sampling time on the uptake rate [1,18,19]. Some studies measured the uptake rates of tube-type Tenax TA diffusive samplers in laboratory-controlled atmospheres or real environments [1,18,25,28]. However, the uptake rates in these studies were obtained under specific environmental conditions and often differed, which may impede the application of these uptake rates to other environmental conditions. Furthermore, the diffusive uptake rates in standards and literature are mainly reported in the environmental monitoring for

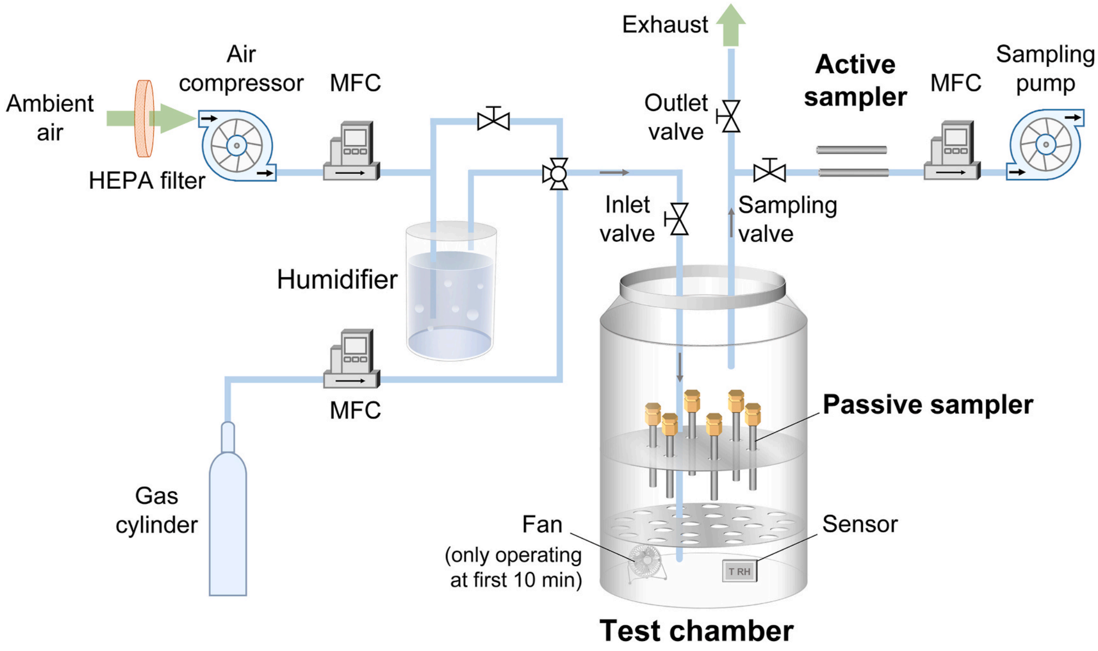

Fig. 1. Schematic of the experimental setup.

occupational safety and health in a short-term exposure (such as 2 h, 8 h) at high concentrations or in a long-term exposure (such as 7d, 14d) at lower concentrations. There are relatively few reports on the 24 h uptake rate. However, some workplaces require people to stay 24 h a day (such as in submarines, underground workstations) [29]. Long-term environmental monitoring also requires the 24 h uptake rate to obtain daily exposure.

In this study, we explore the influence of environmental factors (temperature, humidity and concentration) on the gas-phase 24 h toluene uptake rate in Tenax TA diffusive samplers. Statistical analysis is performed to quantify the effects of temperature, humidity and concentration on the toluene uptake rate. A comparison between the results in this study and previous literature is addressed. We propose an empirical equation to amend the effect of environmental factors accordingly, which can promote the accurate uptake rates of tube-type diffusive samplers.

2. Methods

2.1. Experimental design

The configuration of the experimental setup is shown in Fig. 1. Compressed ambient air is filtered by a high-efficiency particulate air (HEPA) filter and then passes through a mass flow controller (MC 20SLPM, Alicat Scientific, USA) with an airflow rate of 20 L/min. The compressed air is then divided into two streams. One stream passes through an airflow control valve and the other passes through a bottle-wash humidifier to adjust the humidity. The two streams are mixed with another specific pollutant stream from a standard gas cylinder (Beijing Zhaoge Gas Technology Co., Ltd, China) and then are supplied to a stainless-steel chamber. As one of the typical organic compounds indoors [30], toluene is used as the target pollutant in this study. The toluene concentration is adjusted by another mass flow controller (MC1SLPM, Alicat Scientific, USA).

The chamber is 400 mm in inner diameter, 510 mm in height and 64 L in total volume. A lid is used to cover the top of the chamber. A seal ring is placed between the chamber and lid. Then the chamber is air-tight sealed by a lathedog. Two stainless steel tubes with a diameter of 6 mm are welded symmetrically on the lid for the conditioned and polluted air purging. The tube lengths inside the chamber are 140 mm and 430 mm, respectively, with 70 mm exposed outside of the chamber. There is an inlet valve in the long tube, while an outlet valve and a sampling valve are connected to the short tube. The three valves are used to turn on/off the airways. As shown in Fig. 1, there is a stainless-steel mounting bracket inside the chamber. Six Camsco axial Tenax TA cartridges (3.5 inches/89 mm length, 60/80mesh, USA) are vertically placed at the bracket, 400 mm from the bottom. The adsorbent is retained in place by means of stainless steel gauzes at both ends. Analyte molecules diffuse through the sampling end, and the diffusion distance is typically 15 mm before they are trapped in the adsorbent [31]. A sensor is set inside the chamber to simultaneously record temperature and relative humidity (RH). Temperatures in the experiments fluctuated within a range of 23.8 ± 2.5 °C. RH was controlled in the range of 20–90%.

In each experiment, polluted air is first supplied through the long tube to the chamber bottom, where a mini-fan will quickly and evenly disperse the polluted air inside the chamber. An orifice layer between the fan and the stainless steel mounting bracket allows the airflow to be evenly distributed upwards. After 10 min, the polluted air supply and the fan are stopped, and then the inlet, outlet and sampling valves are all closed to keep the chamber sealed for 24 h. During the 24 h, the 6 Tenax TA cartridges as passive samplers will diffusively capture toluene inside the chamber. After 24 h, the sampling valve is turned on and active sampling of two Tenax TA cartridges is individually performed with an airflow rate of 100 mL/min for 10 min in sequence. Then, the 6 Tenax TA cartridges will be taken out of the chamber, sealed with Difflok caps (Markes) and stored in a refrigerator at 4 °C for a maximum of two weeks before analysis.

2.2. Sample treatment and analysis

Prior to sampling, all Tenax TA cartridges are thermally aged by a gas chromatography-mass spectrometry (GC-MS) (QP-2010SE, SHIMADZU, Japan) at 350 °C with a nitrogen flow rate of 100 mL/min for 60 min before use.

The sampled cartridges are first thermally desorbed by a sample pretreatment platform (MPS Robotic, Gerstel, Germany) and analyzed by the GC-MS. The thermal desorption unit (TDU) of the sample pretreatment platform is programmed at 50 °C for 1 min and ramped up by 40 °C/min to 320 °C, where it is held constant for 8 min to heat the cartridges. The desorption pressure and desorption flow are set as 81.0 kPa and 50.0 mL/min, respectively. The desorbed compounds from TDU are trapped in a cooled injection system (CIS) at −20 °C by liquid nitrogen. The split ratio is set as 20:1 on CIS for the sample injection. The chromatographic column in the GC-MS is DB-5MS (0.25 mm of inner diameter, 60 m in length, 0.25 μm of film thickness, Agilent, USA). Helium is used as the carrier gas and the column flow is set at 0.74 mL/min. The oven temperature is programmed at 50 °C for 1 min and ramped up to 320 °C by 40 °C/min. Then the temperature is held constant for 8 min. The gas chromatograph is operated in electron impact (EI) mode at 70 eV electron energy. Ion source and interface temperatures are set as 250 °C and 230 °C, respectively.

The quality control/quality assurance (QA/QC) analysis of the sampling method and GC-MS measurement is shown in the supplementary materials. The stability analysis and the background measurement on GC-MS are presented in Supplementary Table S1 and Table S2, respectively. The relative standard deviation (RSDs) of toluene measurements are all less than 2%. The background amount of toluene in each cartridge is low and can be ignored. The standard curves of toluene are shown inSupplementary Figure S1. Supplementary Table S2 indicates that the background amount of toluene in each tube is low and can be ignored.

2.3. Calculation of uptake rates

The theory for diffusive sampling, primarily defined by Fick's first law, has been well established and studied [32]. For toluene in this study, the ideal uptake rate (UR_{ideal}, mL/min) is determined as Eq. (1).

$$UR_{\text{ideal}} = \frac{D \cdot A}{L} \times 60 \tag{1}$$

where D is the diffusion coefficient of toluene in the air at 25 °C, 0.0829 cm^2/s [15,33]; A is the cross-sectional area of the Tenax TA cartridge, 0.196 cm^2; L is the air gap between the sampling end of the cartridge and the surface of sorbent, 1.5 cm, and 60 is the conversion coefficient from mL/s to mL/min. Therefore, the UR_{ideal} of toluene is 0.65 mL/min. However, the actual uptake rate often differs from the UR_{ideal} due to volatility of the compound, weak sorbent, back diffusion and sorbent saturation during long-term sampling [15].

The toluene concentration in the chamber is calculated by Eq. (2).

$$C_a = \frac{m_a}{UR_a \cdot t_a} \times 10^3 \tag{2}$$

where C_a is the mass concentration of toluene, $\mu\text{g/m}^3$; UR_a is the active sampling rate, mL/min; m_a is the mass of toluene in the active cartridge, ng, detected by GC-MS; t_a is the active sampling time, min.

The uptake rate of a diffusive cartridge, UR_p (mL/min), is calculated by Eq. (3).

$$UR_p = \frac{m_p}{C_p t_p} \times 10^{-3} \tag{3}$$

where C_p is the mass concentration of toluene in the chamber, $\mu\text{g/m}^3$, equal to C_a; m_p is the mass of toluene in the diffusive cartridge, ng; t_p is the passive sampling time, min.

Eq. (2) and Eq. (3) can be combined into Eq. (4).

$$UR_p = \frac{m_p t_a}{m_a t_p} \times UR_a \tag{4}$$

The average diffusive uptake rate and the standard deviation (SD) can be calculated by Eq. (5) and Eq. (6), respectively.

$$\overline{UR_p} = \frac{\sum_{i=1}^{n} UR_{p,i}}{n} \tag{5}$$

$$SD = \sqrt{\frac{\sum_{i=1}^{n} \left(UR_{p,i} - \overline{UR_p}\right)^2}{n}} \tag{6}$$

where $UR_{p,i}$ is the diffusive uptake rate of ith cartridge, mL/min $\overline{UR_p}$ is the mean diffusive uptake rate, mL/min; n is the cartridge number. The relative standard deviations (RSD) of toluene concentration is the ratio of SD and $\overline{UR_p}$.

3. Results

3.1. Toluene uptake rates measured in different conditions

Table 1 summarizes 15 experimental cases in this study. The RSDs of toluene concentration monitored by active sampling range from 0.4 to 15%. The 15 cases are separated into three concentration groups: Group 1 (low concentrations, 0.2–0.8 mg/m^3), Group 2 (medium concentrations, 1–1.5 mg/m^3), and Group 3 (high concentrations, 2–3 mg/m^3). Temperatures in all experiments fluctuate within 21.3–26.3 °C, and the RH varies from 20% to 90%. The RSDs of temperature and RH are 0.4–10.6% and 1.4–10%, respectively.

Table 1

Toluene uptake rates (UR_p) at different environmental factors: concentration, relative humidity (RH) and temperature. All data are in the form of average ± SD.

Group	No.	Concentration (mg/m^3)	RH (%)	Temperature (°C)	UR_p (mL/min)
Group 1 low concentration	1	0.475 ± 0.0035	22.3 ± 0.7	19.3 ± 0.7	0.367 ± 0.0273
	2	0.417 ± 0.0395	37.4 ± 3.6	26.5 ± 0.5	0.358 ± 0.0227
	3	0.322 ± 0.0265	40.7 ± 2.9	26.6 ± 0.7	0.355 ± 0.0273
	4	0.393 ± 0.0207	50.4 ± 1.0	25.1 ± 0.4	0.308 ± 0.0017
	5	0.231 ± 0.0063	51.5 ± 1.0	25.1 ± 0.5	0.333 ± 0.0309
	6	0.691 ± 0.0388	66.4 ± 1.2	23.6 ± 0.5	0.330 ± 0.0260
	7	0.580 ± 0.0084	76.9 ± 1.4	23.2 ± 0.4	0.256 ± 0.0156
Group 2 medium concentration	8	1.240 ± 0.0253	21.2 ± 2.2	25.1 ± 0.7	0.457 ± 0.0219
	9	1.086 ± 0.0461	43.9 ± 1.7	25.7 ± 0.7	0.350 ± 0.0361
	10	1.366 ± 0.0453	66.5 ± 0.9	24.8 ± 0.4	0.334 ± 0.0585
	11	1.074 ± 0.0064	76.7 ± 0.6	25.0 ± 0.1	0.246 ± 0.0225
Group 3 high concentration	12	2.012 ± 0.0359	30.9 ± 2.9	18.8 ± 2.0	0.472 ± 0.0328
	13	2.350 ± 0.0097	41.9 ± 1.5	25.4 ± 0.6	0.512 ± 0.0398
	14	2.065 ± 0.0697	62.3 ± 4.1	18.5 ± 1.8	0.479 ± 0.0262
	15	2.670 ± 0.3907	85.6 ± 2.0	25.0 ± 0.2	0.384 ± 0.0599

Since the toluene concentration in indoor environments usually varies in the range of 0.1–0.6 mg/m^3 [30,34], we conducted more experiments in Group 1 to detail the influence of RH.

Correlation analysis is used to analyze the relationship between the environmental factors and the toluene uptake rate. First, the Kolmogorov-Smirnov test is applied to verify whether the RH, temperature, concentration and toluene uptake rate follow normal distributions. At the significance level of 0.05, the RH and toluene uptake rates follow normal distributions, but the temperature and toluene concentration are not normally distributed. The Pearson correlation and Spearman correlation are adopted to investigate the influence of RH, temperature, and concentration on toluene uptake rate.

Table 2 reports that the Pearson and Spearman correlation coefficients between RH and toluene uptake rate are about −0.5, while the Pearson and Spearman correlation coefficients between concentration and toluene uptake rate are about 0.5. The *p*-values of both coefficients are less than 0.05, indicating that the negative influence of RH on UR_p and the positive influence of concentration on UR_p are statistically significant. The Pearson correlation coefficient between temperature and UR_p is negative with a *p*-value less than 0.05. However, the Spearman correlation coefficient between temperature and toluene uptake rate is close to 0, and the *p*-value is larger than 0.05. A similar situation was reported in Rovetta's study [35], which indicates that although the data is not normally distributed, Pearson's coefficient is even more effective than Spearman's in some specific cases. Therefore, the increase in temperature in this study was only slightly correlated with the decrease in UR_p. The reason is that the range of temperature change is small.

3.2. Impacts of environmental factors on the uptake rate

3.2.1. Impact of humidity

Fig. 2 shows that the toluene uptake rate in three concentration groups decreases with the increase in RH. The filled circle points are the data measured in this study. The hollow circle points in Fig. 2 are the 24 h toluene uptake rates obtained by Tenax TA passive sampling in the literature [1,19,25].

The toluene uptake rates in each concentration group all show a downward trend as RH increases. When RH is adjusted from the lowest to the highest, the corresponding uptake rates drop by 30.3% (Group 1, RH: 22.27%–76.86%), 46.2% (Group 2, RH: 21.24%–76.65%), and 18.6% (Group 3, RH: 30.87%–85.6%), respectively. When RH is changed from 30% to 70%, the UR_p of the three groups all change slowly. The effect of RH indicates that competitive adsorption between toluene and moisture molecules is essential to the uptake rate. Mo et al. applied a unimolecular Langmuir model to analyze the competitive adsorption effect of humidity on toluene adsorption and photocatalytic oxidation [36]. When the adsorption cartridge has the same saturation capacities for all sample components [37], a competitive Langmuir isotherm is often used, as shown in Eq. (7).

$$q=\frac{k_1 C}{1+k_1 C+k_2 C_w} \tag{7}$$

where q is the toluene concentration in the stationary phase; C is the toluene concentration in the gas phase; C_W is the moisture concentration in the gas phase; k_1 and k_2 are the Langmuir coefficients of toluene and moisture, respectively. Thus, the adsorbed moisture either removes surface sites from contention for adsorbed toluene or creates sites for competitive physical adsorption between water and toluene. Some researchers reported the effect of RH on the collection of C2–C5 aliphatic hydrocarbons [20]. Unacceptable breakthrough values for volatile C2–C5 were observed under high humidity (>60%). The breakthrough volumes for C2 aliphatic compounds were reduced 13–22 fold under 90% RH. Liu et al. studied the competing role of moisture in the adsorption of gaseous benzene on microporous carbon [38]. They found that RH sensitively decreased the maximum benzene adsorption capacities (27%–65%) compared with dry conditions. These conclusions are consistent with the experimental results reported in this study.

3.2.2. Impact of toluene concentration

When RH is about 42%, the uptake rate changes from 0.355 (Group 1, 0.322 mg/m^3) to 0.350 (Group 2, 1.086 mg/m^3) to 0.512 (Group 3, 2.350 mg/m^3). When RH is about 65%, the uptake rate changes from 0.330 (Group 1) to 0.334 (Group 2) to 0.479 (Group 3). For high concentration (Group 3), the UR_p at high humidity (RH = 86%) is obviously lower than those at other humidity conditions. However, the decrease in toluene uptake rate in Group 3 is relatively small compared to those in Group 1 and Group 2, indicating that the influence of toluene concentration on the uptake rate may be more significant than RH. It agrees with the correlation analysis in section 3.1, which shows that toluene concentration has the highest correlation with the uptake rate.

The difference in concentration will lead to the difference in the initial concentration gradient in the Tenax TA cartridges. The greater the concentration of the sampling environment, the greater the initial concentration gradient will be, which promotes the diffusion of toluene in Tenax TA cartridges. Therefore, the increase in concentration positively impacts the UR_p. The results are consistent with those found by Walgraeve [19] that the concentration level determines the mass uptake for a certain compound and sampling device.

Table 2
Correlation analysis between RH, temperature and concentration on toluene uptake rates, respectively.

Correlation coefficient	RH	Temperature	Concentration
Toluene uptake rate	Pearson		
	−**0.513** ($p = 0.000$)	−**0.268** ($p = 0.024$)	**0.546** ($p = 0.000$)
	Spearman		
	−**0.541** ($p = 0.000$)	−0.077 ($p = 0.522$)	**0.535** ($p = 0.000$)

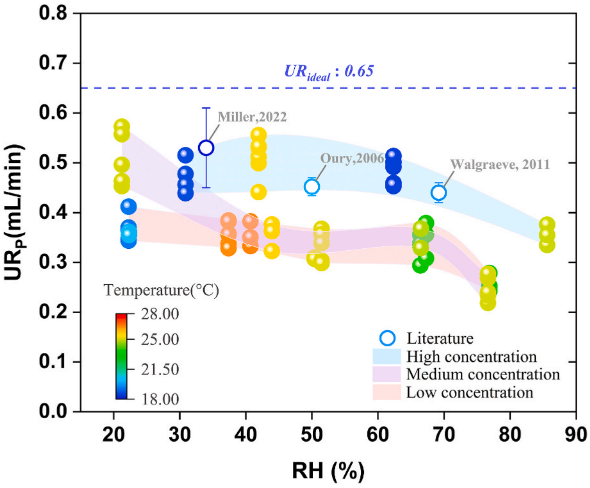

Fig. 2. The UR_p (mL/min) of toluene in 24 h. The filled points are measured in this study, and the hollow points are from other studies [1,19,25]. The color of the data point represents the experimental temperature. The shadows in pink, purple and blue present the error bands in Group 1 (low concentration, 0.2–0.8 mg/m^3), Group 2 (medium concentration, 1–1.5 mg/m^3), and Group 3 (high concentration, 2–3 mg/m^3), respectively. The dashed line of UR_{ideal} is calculated based on Eq. (1). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

3.2.3. *Impact of temperature*

There is a 25.9% decrease in toluene uptake rate from 18.5 °C to 26.6 °C. Hence the effect of temperature on UR_p should not be neglected. The diffusion coefficients (D_T) at any absolute temperature (T) are calculated as a function of D_{298} at 25 °C (298 K) [39]: $D_T = D_{298}\bullet(T/298)^{1.75}$. Therefore, the temperature increase will facilitate the diffusion process of toluene in Tenax TA cartridges. It must also be taken into account that adsorption is an exothermic process. Decreasing temperature allows for increasing the mass collected of toluene. Thus, the temperature has two opposing effects on the diffusive uptake rate. It seems that the prominent effect at a high temperature is an adsorption reduction. According to the correlation analysis, the influence of temperature on the toluene uptake rate

Table 3
Toluene uptake rates by Tenax TA cartridges reported in the literature. Some data are in the format of average ± SD.

Sampling time	RH (%)	Temperature (°C)	Concentration (mg/m^3)	UR_p (mL/min)	Ref.
8 h	–	25	–	0.449 ± 0.006	[15]
	50	25	–	0.444	[28]
24 h	34	14	–	0.53 ± 0.08	[1]
	50	20	0.715	0.452 ± 0.018	[25]
	69.2 ± 4.4	19.7 ± 0.3	0.049 ± 0.005	0.44 ± 0.02	[19]
	37.4 ± 3.6	26.5 ± 0.5	0.417 ± 0.0395	0.358 ± 0.0227	This study - Group 1
	51.5 ± 1.0	25.1 ± 0.5	0.231 ± 0.0063	0.333 ± 0.0309	
	76.9 ± 1.4	23.2 ± 0.4	0.580 ± 0.0084	0.256 ± 0.0156	
	21.2 ± 2.2	25.1 ± 0.7	1.240 ± 0.0253	0.457 ± 0.0219	This study - Group 2
	66.5 ± 0.9	24.8 ± 0.4	1.366 ± 0.0453	0.334 ± 0.0585	
	76.7 ± 0.6	25.0 ± 0.1	1.074 ± 0.0064	0.246 ± 0.0225	
	30.9 ± 2.9	18.8 ± 2.0	2.012 ± 0.0359	0.472 ± 0.0328	This study - Group 3
	62.3 ± 4.1	18.5 ± 1.8	2.065 ± 0.0697	0.479 ± 0.0262	
	85.6 ± 2.0	25.0 ± 0.2	2.670 ± 0.3907	0.384 ± 0.0599	
3 days	51.4 ± 5.3	17.6 ± 1.9	0.053 ± 0.006	0.51 ± 0.04	[19]
	50	20	0.715	0.397 ± 0.015	[25]
6 days	50	20	0.715	0.366 ± 0.008	[25]
7 days	62.0 ± 4.2	18.2 ± 1.1	0.049 ± 0.002	0.40 ± 0.02	[19]
	45	23	2.700	0.46	[43]
	–	25	–	0.311	[15]
	–	25	–	0.32	[18]
	67.7	6	–	0.47 ± 0.027	[1]
14 days	50	20	0.715	0.299 ± 0.007	[25]
	45	23	2.501	0.37	[43]
	0	20	–	0.36	[44]
	80	20	–	0.34	[44]
	–	20–60	–	0.324	[45]
	66	6	–	0.39 ± 0.06	[1]
28 days	69.3 ± 4.2	2.7 ± 0.6	–	0.42 ± 0.09	[1]
	–	–	2.3	0.35	[43]
	–	–	–	0.23	[42]
	–	–	1.993	0.32	[42]

is generally less than those of concentration and humidity in this study. The negative effect of temperature on diffusive uptake rates has also been reported for solid-phase microextraction (SPME) diffusive samplers [40,41].

4. Discussion

4.1. Comparison of toluene uptake rates with other studies

The toluene uptake rates were also measured by some researchers under different environmental conditions, such as temperature, relative humidity, and concentration. For example, uptake rates for 1–8 h in workplace applications had been published for a wide range of VOCs [42]. Miller et al. reported the toluene uptake rates of low VOC concentrations for 1–4 weeks [1]. Table 3 summarizes the diffusive toluene uptake rates by Tenax TA cartridges in the literature at different sampling times and experimental conditions. The average UR_p at different RH in three concentration groups in this study are selected to compare with the literature data.

Most of the existing studies reported the toluene uptake rates at high concentrations in workplaces longer than 7d [43]. These data are quite different from each other. For example, the maximum deviation between the 7d toluene uptake rate data is 51%. Moreover, many of these studies are more or less lacking in the completed information on the temperature, humidity, and concentration. In contrast, the toluene concentrations of the studies less than 7d are mainly below 1 mg/m^3. The experimental concentration in this study, ranging from 0.23 to 2.67 mg/m^3, covers the concentration range of the literature. The RH in this study, ranging from 20 to 86%, also covers most of the literature.

Generally, Table 3 also shows that the longer the sampling time, the lower the UR_p, and the UR_p at higher humidity is smaller than those at low humidity. Therefore, a comprehensive model is needed to quantify the impact of temperature, humidity, and concentration on the UR_p.

4.2. Multivariate analyses of environmental factors

In order to predict the uptake rate at various environmental factors, we propose a multiple linear regression model of 24 h toluene uptake rate based on the experimental data in this study, as shown in Eq. (8). The coefficient of determination (R^2) value is 0.737. To judge whether the equation model is valid, the analysis of variance (ANOVA) and the colinear diagnosis are conducted and shown in Supplementary Tables S3 and S4. The residual plots of the three independent variables are shown in Supplementary Figure S2.

$$UR_{p,24h} = 0.479 - 0.003 \times RH - 0.0015 \times T + 0.0767 \times C \tag{8}$$

where $UR_{p,24h}$ is the 24 h uptake rate in mL/min; RH is the relative humidity in %; T is the temperature in °C; C is the toluene concentration in mg/m^3. Since Eq. (8) is obtained by fitting the experimental data, it is suitable for the range of temperature 16–28 °C, humidity 20%–90%, and concentration 0.2–4 mg/m^3.

We further use Eq. (8) to evaluate the relative change ratios of $UR_{p,24h}$ driven by humidity (at 80% RH and 20% RH) at 25 °C. Two representative scenarios were selected for comparison: indoor environments with low concentrations and long exposure time, and occupational work environments with high concentrations and short exposure time. The 1-h limit of indoor toluene concentrations in China is 0.2 mg/m^3 [46]. The indoor toluene concentration reported in different countries ranges from 0.01 to 0.6 mg/m^3 [30]. As Fig. 3 shows, the humidity increase significantly decreases the 24 h toluene uptake rate at the standard toluene concentration (0.2 mg/m^3) by 45.4% from 20% RH to 80% RH. Accordingly, under the toluene concentrations of 0.01 mg/m^3 and 0.6 mg/m^3, the predicted $UR_{p,24h}$ decreases by 47.1% and 42.1%, respectively. The toluene concentrations in the occupational environment like house painting and petrol stations have an average value ranging from 2 to 4 mg/m^3 [47]. As Fig. 3 shows, under the high concentrations range from 2 mg/m^3 and 4 mg/m^3, the predicted $UR_{p,24h}$ decreases by 33.7% and 26.2%, respectively. The curve slope in Fig. 3

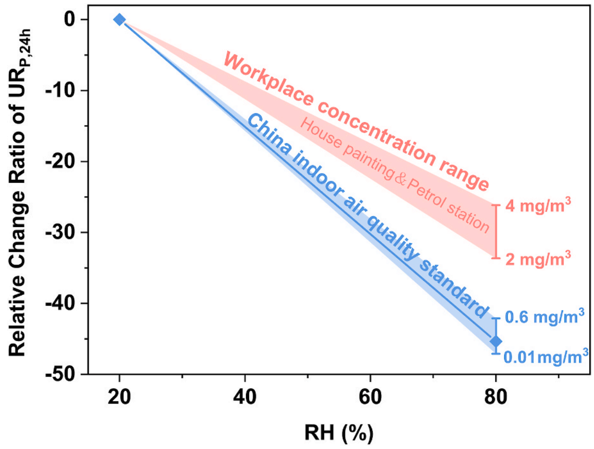

Fig. 3. Relative change ratios of toluene uptake rates from 20% RH to 80% RH. The temperature is fixed at room temperature 25 °C. The blue area represents the typical indoor concentration range of toluene (0.01–0.6 mg/m^3). The blue line is the 1-h limit of indoor toluene in China: 0.2 mg/m^3 [46]. The red area represents the concentration range of toluene in workplaces (2 ~ 4 mg/m^3).

represents how much the $UR_{p,24h}$ declines by changes in humidity. The smaller the curve slope, the less influence of $UR_{p,24h}$ by increasing humidity. The increase in concentration will reduce the impact of humidity. It is foreseeable that when the concentration is larger than a specific value, the $UR_{p,24h}$ will not be affected by humidity, that is, increasing the humidity will not cause a decline in the $UR_{p,24h}$. However, the maximum applicable concentration of Eq. (8) is 4 mg/m^3, which may not be directly extrapolated to high concentration. In the concentration range of toluene in real indoor environments and common occupational environment, it can be concluded that the impact of RH on toluene uptake rates has to be considered. And the prediction model of $UR_{p,24h}$ in Eq. (8) can help to calculate the 24 h uptake rate under different environmental factors.

4.3. Correlation between adsorption mass and uptake rate on Tenax TA cartridges

As shown in Table 3, the uptake rate declines over time. Walgraeve et al. expressed the accumulated mass (m_p) of toluene on the passive sampler as a function of the exposure dose (the product of concentration in ppm$_v$ and the sampling time in min) [19], as shown in Eq. (9).

$$m_p = k \times Dose^b \tag{9}$$

where k and b are coefficient and exponent, equal to 3.89 and 0.81, respectively [19]. The exposure dose interval used in Walgraeve's equation was between 25 and 150 ppm$_v$•min.

Similar to Eq. (3), m_p can be calculated by Eq. (10).

$$m_p = C \times UR_p \times t_p \tag{10}$$

where C is the toluene concentration, mg/m^3. The exposure dose in this study can be calculated by converting the concentration in Table 1 into ppm$_v$ and then multiplying with the sampling time, 24 h.

As Fig. 4 shows, the red dots with the exposure dose range from 0 to 1200 ppm$_v$•min are the experimental data in this study, far exceeding the range in Walgraeve's study. Thus, the gray line fitted by Walgraeve is only accurate in the range of 0–150 ppm$_v$•min, and it deviates greatly when the range is greater than 600 ppm$_v$•min. Based on the same formula in Eq. (9), we use the experimental data in this study to obtain the revised coefficients k and b as 0.68 and 1.13, respectively. The new fitting curve (red) is shown in Fig. 4. The coefficient of determination is 0.909. We also compare some literature points with a similar dose range, and find that the literature data points are closer to the data fitted in this experiment. Therefore, when the exposure dose is larger, the revised fitting curve in this study is more appropriate.

NIOSH updated the immediately dangerous to life or health concentrations (IDLH) of toluene is 1880 mg/m^3 [48]. As a safety margin, IDLH values are based on effects that might occur as a consequence of a 30-min exposure. Therefore, the corresponding exposure dose is 14,358.6 ppmv•min, which far exceeds the range of this study. For different organizations and the applications where these limits may be exceeded, the fitting curve in this study may still be inappropriate and a newly revised fitting is necessary.

Combining Eq. (8) into the new fitting formula of m_p in Fig. 4, the correlation between adsorption mass and uptake rate can be described as Eq. (11).

$$m_p = 0.68 \times \left(\frac{UR_{p,24h} + 0.003RH + 0.0015T - 0.0479}{0.0767} \times \frac{V}{M} \times t_p \right)^{1.13} \tag{11}$$

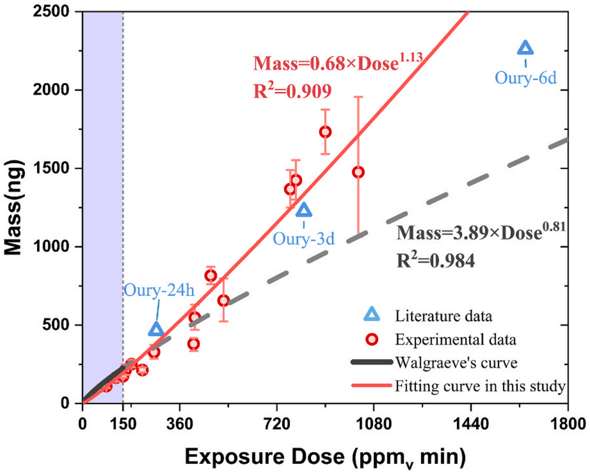

Fig. 4. The mass (ng) collected on the passive sampler as a function of exposure dose. The curve fitted by the experimental data in this study is in red, and the curve provided by Walgrave's equation is in black. Walgraeve's equation fits the exposure dose range from 0 to 150 ppm$_v$•min (the blue zone). The red dot is the adsorption mass obtained in this experiment. The blue triangle points are the data obtained from the literature [25]. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

where V is the molar volume of toluene at 25 °C and 1.01×10^5 Pa, equal to 24.5 L/mol; M is the molar mass of toluene, 92.14 g/mol.

If performing diffusive sampling in actual environments, the $UR_{p,24h}$ can be predicted by Eq. (11) when m_p, temperature and RH are all measured. Since the concentration does not appear in Eq. (11), it means that methods like active sampling are no longer needed to calibrate the $UR_{p,24h}$ when diffusive sampling is used. This will be very helpful for the application of diffusive samplers in indoor and outdoor ambient air monitoring. Similarly, the form of Eq. (11) may be extended to other VOCs, as well as the uptake rate for the corresponding sampling time.

5. Conclusions

In this study, diffusive uptake rates of toluene on Tenax TA tube-type samplers were systematically explored under different indoor environmental factors. The experiment studied the influence of different humidity, temperature and concentration on the toluene diffusive uptake rate. Correlation analysis was conducted to analyze the relation between environmental factors and uptake rate. The results show that temperature and humidity negatively correlate with the toluene uptake rate, while the concentration positively affects the rate. An empirical correlation equation between environmental factors (T, RH and concentration) and 24 h uptake rate was introduced by a multiple regression method. The empirical equation will help guide the correction of uptake rate for different environments. An improved power-law formula between adsorption mass and UR_p was developed by referring to Walgraeve's procedure. The UR_p can be predicted when the adsorption mass, temperature and relative humidity are measured.

Overall, the measured 24 h uptake rate of toluene and the improved formula can enrich the passive sampling studies by Tenax TA cartridges. The reported effect of environmental factors will help obtain the accurate uptake rates of tube-type diffusive samplers for gaseous pollutants. This study will help promote the application of passive sampling in environmental pollutant monitoring and occupational exposure monitoring.

However, due to some limitations, only toluene, a representative indoor pollutant, was selected as the target gas in this work. The influence of environmental factors on the uptake rates of other VOC gases should be studied in the future. Moreover, using the similar methods introduced in this study, we can also establish the multiple factors experiments to study the influence of other VOCs and wind speed. Furthermore, if enabling to quantify the influence of environmental factors on other VOCs, it is probable to extend similar formulas to predict the diffusive uptake rate of other VOCs. Other tube-type diffusive samplers using highly adsorptive adsorbents such as Carboxen may be used with this proposed method to calibrate the influence of environmental effects.

CRediT authorship contribution statement

Yan Wang: Writing – original draft, preparation, Methodology, Visualization, Writing – review & editing. **Tao Yu:** Investigation, Writing – review & editing, Project administration. **Jinhan Mo:** Conceptualization, Supervision, Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jobe.2022.104587.

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