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Original article

Total odor threshold ratio can be a new method to evaluate indoor air quality

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Abstract

Sick building syndrome (SBS) is a range of symptoms that include eye irritation, pain in the nose and throat, headaches when entering a newly constructed or remodeled house or building, and in many cases, the patients claim odor. Volatile organic compounds (VOCs) are suspected to be one of the major causes of SBS. The Ministry of Health, Labor and Welfare of Japan (MHLW) has set guideline values for 13 VOCs and the interim target value of total VOCs (TVOC). However, there are people who still claim SBS. VOC levels are important to know to evaluate the indoor air, but even if the TVOC is quite low, some people claim odor, which is difficult to quantify. Therefore, in this study, we tried to quantify odor by using odor threshold ratio (OTR) and investigated the relationship between total OTR (TOTR) and TVOC with the data obtained in laboratory houses in Chemiless Town of Chiba University. As the result, it became clear that TVOC correlate with TOTR when TVOC level was higher than 500 $\mu g/m^3$, but below 500 $\mu g/m^3$, the correlation became weak. TOTR varies with the building materials, and even if TVOC was very low, in some cases, TOTR was higher than others. Our study suggests that odor can be quantified with OTR objectively, and TOTR can be a useful new method to evaluate indoor air quality in addition to TVOC.

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《**Key words**》 sick building syndrome (SBS), evaluation method, indoor air quality, total odor threshold ratio (TOTR), total volatile organic compounds (TVOC)

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Ⅰ. Introduction

Sick building syndrome (SBS) is a range of symptoms that include eye irritation, pain in the nose and throat, dizziness, headaches, fever, sore joints, feelings of listlessness, or nausea when entering a newly constructed or remodeled house or building[1], and in many cases, the patients claim odor[2, 3]. Volatile organic compounds (VOCs) are suspected to be one of the major causes of SBS. In 2003, the Ministry of Land, Infrastructure, Transport and Tourism, which is responsible for construction standards, revised the Building Standards Law in July 2003 to address the SBS problem. In the revision, the standard value for formaldehyde, one substance responsible for the syndrome, was set at under 0.08 ppm (or 100 $\mu g/m^3$) for indoor air[4]. The Ministry of Health, Labor and Welfare of Japan has set non-binding guideline values for 13 VOCs and the interim target value of total VOCs (TVOC) as 400 $\mu g/m^3$ by 2001[1, 5]. The adoption of this regulation and guideline gave everyone, especially those in the construction and interior design industries, the impression that the SBS problem was over[1].

However, the problem of SBS has not been solved completely[6]. The industry has decreased the use of those chemicals which were regulated by the law; instead, they started using other chemicals which were not regulated. It resulted in new problems that SBS has been occurred by unregulated chemicals[7]. Therefore, it is important to evaluate the indoor air quality, not only with the concentration level of regulated VOCs, but with the TVOC.

According to our previous studies[8], it became clear that if the amount of TVOC was low enough, the number of people who claimed the symptoms clearly decreased. However, there were people who still claimed unpleasant odor in indoor air despite the fact that the TVOC level was very low.

Meanwhile, there are many reports that odor is often a primary determinant in the assessment of the quality and acceptability of indoor air[2, 9, 10]. Sensory irritation and odor are common symptoms and complaints that may be experienced simultaneously and may interact with each other[11]. Thus it seems that odor is also a very important aspect to evaluate the indoor air quality. Odor is perceptible with human sense, and some people are sensitive with odor, and others are not. However, if there is a method to quantify the odor objectively, it will become possible to measure it.

Odor threshold of indoor air VOCs are very different. Even if the concentration level is quite low, some chemicals smell strongly. Therefore, in this study, odor threshold ratio (OTR) was investigated if it would be useful to evaluate odor in indoor air. OTR of a chemical is obtained by dividing concentration level by its odor threshold value[12], and higher OTR means stronger smell.

The purposes of this study were to quantify the odor in indoor air and investigate the relationship with TVOC to establish a new evaluation method of indoor air quality.

Ⅱ. Materials and methods

Chemiless Town

Chemiless Town is a model town constructed under the concept of “Environmental Preventive Medicine” in a Campus of Chiba University. Environmental preventive medicine is a preventive medicine that tries to prevent sickness which is caused by pollutants in the environment by improving the environment[13, 14]. There are laboratory houses (LHs) where people can actually stay, a reinforced concrete building in which lecture rooms, office, clinic, gallery are facilitated in Chemiless Town[15]. These buildings are all laboratories which were constructed trying to reduce VOCs in indoor air as low as possible.

The study was divided into two parts. (1) 64 VOCs and 17 aldehydes in 3 LHs in Chemiless Town were analyzed and measured 2 to 4 times a year from March 2007 to July 2011. (2) OTR of each chemical was calculated by the data of its indoor concentration level and the odor threshold, and the correlation between TVOC and TOTR were investigated. The statistical analysis was performed by using Excel 2007.

1. Indoor and outdoor air sampling and data analyses

Air samples were collected in living rooms and bedrooms of 3 LHs in Chemiless Town from March 2007 when they were just constructed to July 2011. Outdoor air samples were also collected every time at a point near one of the LHs, under the eaves. Total number of air sampling was 87 times. Building materials and specifications of all the LHs are shown in Table 1.

Before the air sampling of the LHs, windows and doors were kept open for ventilation for 30 minutes, then all the windows and the doors were closed for five hours. After that, air samplings were started. During the sampling, indoor air conditions such as temperature, humidity and atmospheric pressure were recorded. Facilitated ventilation systems of LHs were operated during the sampling.

Air sampling was carried out by active sampling method for 24 hours by using Carbotrap 317 (Supelco) for capturing VOCs, and LpDNPH S10L (Supelco) for capturing aldehydes. The air passed through Carbotrap 317 sampler and DNPH sampler at flow rates of 10mℓ/h and 100mℓ/h respectively. The collected analytes of VOCs were extracted by thermal desorption and analyzed by GC/MS and those of aldehydes were by solvent extraction and by HPLC. The precise analyses were conducted by Tokyo Metropolitan Institute of

Table 1 Construction materials and interior materials of laboratory houses

Laboratory house	Construction materials	Major Interior materials
A	Light-gauge steel structure	Wall: plaster Floor: wood (chestnut) Specifications: - all stainless kitchen unit - chamber tested materials
B	Wood frame construction	Wall: photocatalytic plaster Floor: wood (heat-treated spruce) Specifications: - photocatalytic ventilation system - anti mite carpet
C	Timber framework method of construction	Wall: plaster made of the volcanic products "SHIRASU" Floor: wood (Japanese ceder)

Public Health and Tokyo Kenbikyo-in Foundation.

The analyzed chemicals were 64 VOCs and 17 aldehydes. In this study, the TVOC is defined as the total amount of these VOCs and aldehydes (Table2).

2. Odor Threshold Ratio (OTR) and the total OTR (TOTR)

OTR of each chemical can be obtained by dividing the concentration level by its odor threshold value. The odor threshold values of chemicals are obtained by the triangle odor bag method[16] and they are published by the Ministry of Environmental of Japan. The sum of all the OTRs is the total OTRs (TOTR). OTR and TOTR of each room of the LHs were calculated by using the data of concentration level of VOCs in indoor air.

Table 2 VOCs and aldehydes analyzed in this study

64 VOCs analyzed by GC/MS		17 Aldehydes (Ketone) analyzed by HPLC
Ethanol	Styrene	Formaldehyde
Pentane	*o*-Xylene	Acetaldehyde
Acetone	Nonane	Acrolein
2-Propanol	Tricyclene	Propanal
Dichloromethane	α-Pinene	Butanal
1-Propanol	3-Ethyltoluene	Benzaldehyde
Acetic acid	Camphene	Cyclohexanone
2-Butanone	4-Ethyltoluene	Isopentanal
Ethyl acetate	1,3,5-Trimethylbenzene	Pentanal
Hexane	2-Ethyltoluene	*o*-Tolualdehyde
Chloroform	β-Pinene	*p*-Tolualdehyde
1,2-Dichloroethane	D4: octamethylcyclotetrasiloxane	Hexanal
2,4-dimethylpentane	1,2,4-Trimethylbenzene	Dimethylbenzaldehyde
1,1,1-Ttrichloroethane	Decane	Heptanal
1-Butanol	Isododecane	Octanal
Benzene	*p*-Dichlorobenzene	Nonanal
Carbon Tetrachloride	2-Ethyl-1-hexanol	Decanal
Cyclohexane	3-Carene	
1,2-Dichloropropane	1,2,3-Trimethylbenzene	
Bromodichloromethane	*p*-Cymene	
Trichloroethylene	Limonene	
2,2,4-Trimethylpentane	4-Ethyl-1,2-dimethylbenzene	
Heptane	Undecane	
4-Methyl-2-pentanone	1,2,4,5-Tetramethylbenzene	
Methylcyclohexane	D5: decamethylcyclopentasiloxane	
Toluene	Dodecane	
Dibromochloromethane	Tridecane	
Butyl acetate	D6: dodecamethylcyclohexasiloxane	
Octane	Texanol	
Tetrachloroethylene	Tetradecane	
Ethylbenzene	Pentadecane	
m, *p*-Xylene	Hexadecane	

Ⅲ. Results

TVOC concentration value in indoor air of the rooms of the LHs varied from 28 to 17,559 μg/m³ and the range of TOTR was from 1 to 293 (Fig. 1). There was a tendency that both of the TVOC and TOTR were higher in summer, lower in winter.

In the rooms of LH A, TVOC concentration levels ranged from 28 to 521 μg/m³ (Fig.1 A). It exceeded 500 μg/m³ in the living room when the house was just built in April 2007, but after that, the TVOC has kept decreased. TOTR values ranged from 1 to 28 (Fig.1 A). The major VOCs detected in LH A were decane, 2-ethyl-1-hexanol and toluene, however the concentration levels of all VOCs were low, and the VOCs with higher OTR value were aldehydes such as hexaldehyde, nonanal and acetaldehyde (Table 3).

In the rooms of LH B, TVOC concentration levels ranged from 80 to 2,370 μg/m³ and TOTR values ranged from 1 to 55 (Fig.1 B). Although a variety of VOCs were detected, the major VOCs were terpenes such as para-cymene, limonene, alpha-pinene and 3-carene. TOTR consisted mostly of aldehydes such as

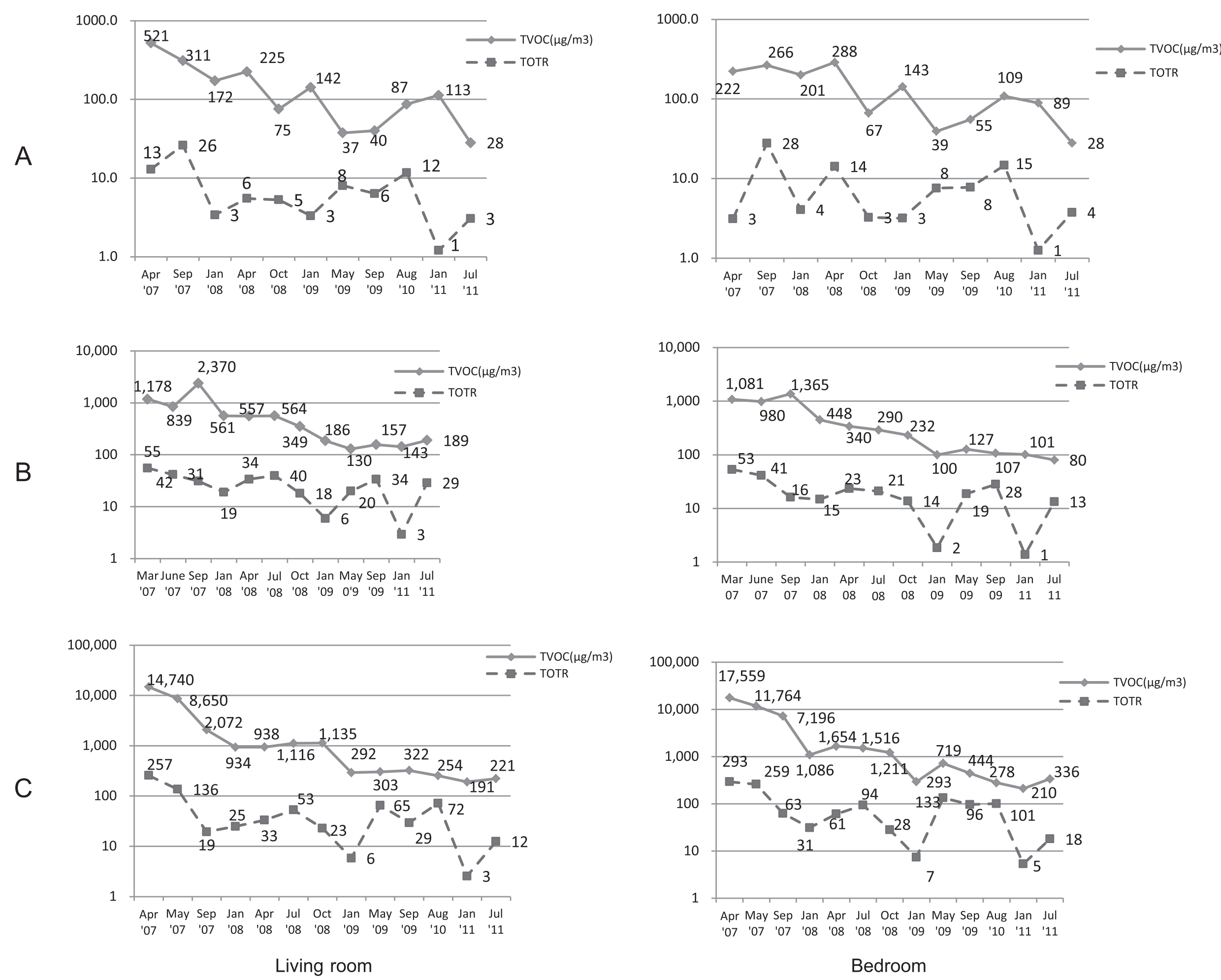

Fig.1 Trend of TVOC and TOTR in the living rooms and bedrooms of laboratory houses A, B and C. The values of Y axis shows TVOC (μg/m³) and TOTR. The values are shown in logarithm.

Table 3 VOCs top 10 concentration level and OTR in the laboratory houses A, B and C in 5 years (2007−2011)

LH	Rank	Compounds	Concentration (μg/m³)	Rank	Compounds	OTR
A	1	Decane	49	1	Hexanal	13
	2	2-Ethyl-1-hexanol	42	2	Nonanal	9
	3	Toluene	37	3	Acetaldehyde	5
	4	Dichloromethane	33	4	Propanal	3
	5	Pentane	33	5	Pentanal	2
	6	Undecane	32	6	Decanal	1
	7	1-Propanol	26	7	2-Ethyl-1-hexanol	1
	8	Ethylbenzene	24	8	Formaldehyde	0.5
	9	Butyl acetate	24	9	α-Pinene	0.2
	10	1,2,4-Ttrimethylbenzene	22	10	*p*-Cymene	0.1
B	1	*p*-Cymene	754	1	Octanal	61
	2	Limonene	404	2	Hexanal	25
	3	α-Pinene	214	3	Acetaldehyde	15
	4	3-Carene	190	4	Pentanal	13
	5	Camphene	188	5	*p*-Cymene	8
	6	2-Butanone	115	6	Nonanal	6
	7	Acetone	112	7	Butanal	4
	8	Toluene	90	8	Propanal	2
	9	Butyl acetate	74	9	α-Pinene	2
	10	Tricyclene	55	10	Limonene	2
C	1	Limonene	4460	1	Octanal	85
	2	α-Pinene	4030	2	Hexanal	69
	3	*p*-Cymene	2560	3	Decanal	66
	4	Camphene	2560	4	α-Pinene	40
	5	3-Carene	1190	5	Nonanal	27
	6	Tricyclene	804	6	*p*-Cymene	26
	7	β-Pinen	315	7	Acetaldehyde	26
	8	Acetone	244	8	Propanal	23
	9	Ethylbenzene	159	9	Limonene	21
	10	Styrene	128	10	Pentanal	21

octanal, hexanal and acetaldehyde (Table 3).

In the rooms of LH C, TVOC concentration levels were over 17,000 μg/m³ at the first measurement in April 2007, and it decreased dramatically with time (Fig.1 C). The major compounds detected were terpenes such as limonene, alpha-pinene, para-cymene and camphene. The VOCs with higher OTR were mainly aldehydes such as octanal, hexanal and decanal (Table 3).

TVOC and TOTR of each room showed strong correlation (r=0.95) when the TVOCs were higher than 500 μg/m³ (Fig.2 A) and showed weak correlation (r=0.49) when the TVOCs were below 500 μg/m³ (Fig.2 B).

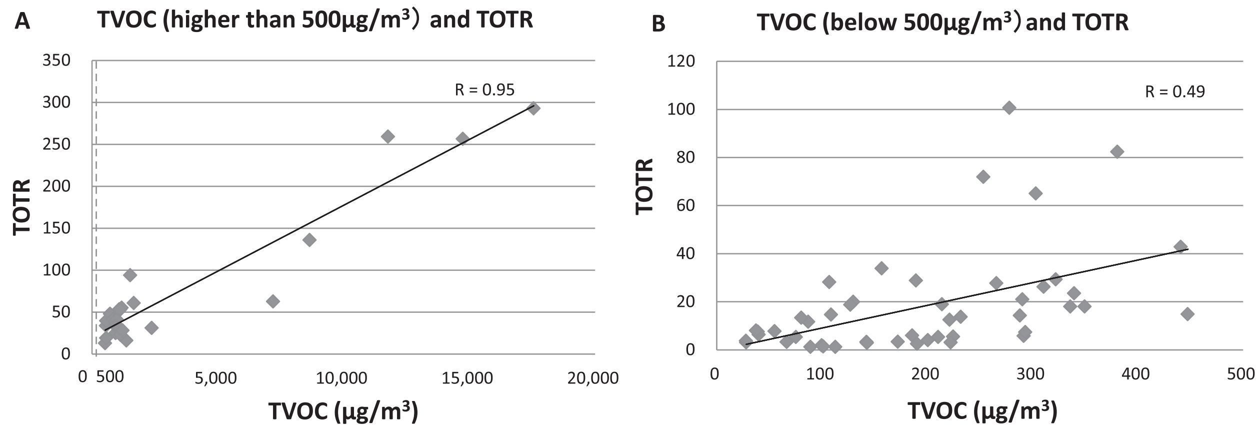

Fig.2 Correlation between TVOC and TOTR

(A) When TVOC concentration value was higher than 500 $\mu g/m^3$, There was a strong correlation.
(B) The correlation became weak when TVOC value decreased below 500 $\mu g/m^3$.

Ⅳ. Discussion

There are countless VOC in indoor air, both man-made and natural, and it is very difficult to determine cause-and-effect relationships between these substances and their impact on human health[1]. To evaluate the indoor air quality, TVOC is an important method since it is necessary to know the actual concentration level of VOCs which exist in the targeted room, not only the regulated VOCs. However, there are people who still claim unpleasant odor despite the fact that the TVOC value is very low. Odor is an important factor to affect human sense, and if odor can be quantified, it is possible to evaluate the air quality more objectively.

In this study, we calculated OTR of each VOC in the LHs with the data of the concentration levels of VOC, and the TOTR was also obtained of all LHs to investigate the relationship between TOTR and TVOC. TVOC and TOTR of each room showed correlation when TVOC was higher, but the correlation became weaker with the decreasing of TVOC. Also, even if the TVOCs were low, the TOTRs were very different depending on the LHs. If TOTR is higher, it means that it smells stronger.

Ⅴ. Conclusion

Our results in this study suggested that TOTR could be another important factor to evaluate the indoor air quality objectively. As a conclusion, TOTR could be a new method to evaluate indoor air quality in addition to TVOC.

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