



Emission rates of substances from low-volatile-organic-compound paints

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Abstract

Globally, sick building syndrome remains a considerable social issue. It is primarily caused by the emission of volatile organic compounds (VOCs) from construction materials. Among these, paints and sealers are used over wide swaths of area and their use causes chemical substances to be released in a room. To prevent sick building syndrome, it is necessary to clarify the cause of air quality changes due to the different types of paints and sealers available. Recently, low-VOC paints (containing 0.3–1.0% VOCs) have been used widely; however, the effects of reducing VOC levels have not yet been fully quantified. This study aims to clarify the chemical compositions by conducting an emission rate test of these low-VOC paints and sealers. We prepared six specimens and used them for chamber testing. Three types of paint with and without a sealer were tested. The minimum and maximum total volatile organic compound (TVOC) concentration was 96 and 4750 $\mu\text{g m}^{-2} \text{h}^{-1}$, respectively. Differences in the VOC content of the paints and sealers could be confirmed. The main TVOC was TexanolTM and glycerol ethers. The results demonstrate that using paints containing less than 0.3% VOCs can decrease the VOC emission rate considerably compared to paints classified as less than 1% VOCs. Moreover, because different kinds of sealers affected the results, it is possible to decrease VOC emission rate using a variety of sealers.

Keywords Indoor air quality · Low-volatile-organic-compound paints · Sealer · Small chamber test

Introduction

The enactment of interior concentration guideline values by the Ministry of Health, Labor, and Welfare of Japan (MHLW) and the revision of the Building Standard Law have been accompanied by the declining reports of sick building syndrome (SBS); nevertheless, the SBS problem remains unresolved. The symptoms of SBS vary greatly between individuals. Additionally, the concentration of every chemical substance that causes SBS symptoms and the sensitivity to each chemical varies between individuals

(Norbäck et al. 1990a, b; Nordström et al. 1995). For these reasons, specifying the causes of SBS is extremely difficult and it is still treated as a social problem.

The major cause of SBS is suspected to be volatile organic compounds (VOCs) emitted by building materials (Mori and Todaka 2011; Heinrich 2011; Herberth et al. 2009; Azuma et al. 2016; Brinke et al. 1998; Hodgson 2002; Molhave 2003; Schneider et al. 2003; Kim et al. 2012; Alevantis 1996; Zhang and Ying 2003; Fang et al. 1999; Deng and Kim 2004). Technological advances in recent years have led to an increase in highly airtight and highly insulated homes and have significantly reduced home energy consumption for heating and cooling. Meanwhile, increasing the airtightness and building insulation levels have caused the chemical substances emitted from building materials and interior finishing to remain in homes for a longer duration, deteriorating interior air quality. It is hypothesized that the large quantities of chemical substances are emitted inside rooms from paints and sealers as they are liberally applied over large surface areas. Although paint is a familiar building material that is handled casually, correlations between air quality and paint types used should be clarified to prevent SBS. The Japanese

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Ministry of Environment requests the industry to reduce the release of VOCs into the environment and recommends the use of low-VOC paints as a VOC reduction strategy (2007). However, very little investigation has been performed in Japan concerning the emissions of VOCs from so-called low-VOC paints. Additionally, Park et al. (2017) examined the effect of low-VOC, water-based paint on allergic diseases of schoolchildren in Korea. They concluded that the paint used in their study might not be a major aggravating factor for the allergic diseases in schoolchildren. It is necessary to investigate the relationship between the contents indicated and emission rates also of low-VOC paints and their adverse effects on public health.

The Japan Paint Manufacturers Association (JPMS) has proposed that the water-based paints should be classified from W1 to W3 according to their VOC and aromatic hydrocarbon-type solvent contents (Table 1). Using W1 class paints (defined to have VOC contents less than 1%) is recommended for reducing the VOC concentrations in rooms. In recent years, low-VOC water-based paints with VOC contents lower than 0.2% or 0.3% have become available; however, even among W1 class paints, large differences in VOC content are seen. This study aims to clarify the characteristics of chemical substances emitted by W1 class low-VOC paints and sealers by conducting emission rate tests and investigate whether they contain chemical substances which may have adverse effects on human

health. Sealers are always used in combination with paints to improve paint performance (Boxall et al. 1992). Kobayashi et al. (2010) have pointed out that sealers also emit Texanol™, which could be a cause of SBS. Hence, in addition to paints, we also performed comparative testing with and without different types of sealers. The experiments were conducted in November 2015 at Chiba University. Precise analyses were conducted by the Tokyo Kenbikyoin Foundation.

Materials and methods

Test material

We prepared six specimens and used them for chamber testing. All paints and sealers were water-based synthetic resin emulsion. Three kinds of paint, i.e., Paint A (PA), Paint B (PB), and Paint C (PC), that conform with class 1 composite emulsion paints categorized as W1 (VOC content lower than 1%), and three types of sealers, i.e., Sealer A (SA), Sealer B (SB), and Sealer C (SC), were tested in assorted combinations. Table 2 shows the combinations of sealers and paints. Under usual application conditions, paints and sealers made by the same manufacturer are typically used together if combined; therefore, in this study, we also used combinations of paints and sealers from the same manufacturer.

Table 1 Classification of paints based on the VOC content

Classification	VOC content (%)	Aromatic hydrocarbons solvent content (%)	Interior	Exterior
W1	Less than 1	Less than 0.1	◎	◎
W2	2 or more but less than 5	Less than 1	○	◎
W3	More than 5	Less than 1	△	○

Table 2 Study samples

Test body	I	II	III	IV	V	VI	Types of paint	VOC content
	PA	PA+SA	PB	PB+SB	PC	PC+SC		
Paint A	○	○					Synthetic resin Emulsion paint	Less than 0.2%
Paint B			○	○			Synthetic resin Emulsion paint	Less than 0.3%
Paint C					○	○	Synthetic resin Emulsion paint	Less than 1%
Sealer A		○					Synthetic resin Emulsion sealer	-
Sealer B				○			Synthetic resin Emulsion sealer	-
Sealer C						○	Synthetic resin Emulsion sealer	-



Emission rate test

The emission rate test was conducted in a 20-L chamber in accordance with the JIS A 1901 small chamber method (JSA. JIS A 1901, based on ISO 16000-9). The chamber and seal boxes were washed with water and baked in an oven at 260 °C before they were set up to eliminate any pollutants from the chamber itself. All test conditions were adjusted such that the test temperature was 28 °C, relative humidity was 50%, ventilation frequency was 0.5 times h⁻¹, and the test loading rate was 2.25 m² m⁻³. The emission rates for 62 VOCs and 15 aldehydes were measured. The chemical substances measured are presented in Table 3. The paints were applied to one side of 165 × 165 mm² glass plates (2

plates) such that the coverage would be 150 × 150 mm² (2 plates) with a total painted area of 0.045 m² (0.0225 m², 2 plates). The painted plates were dried for 1 day at room temperature (23–28 °C). After the paints had set, the painted glass plates fixed in a sealed box that was then placed inside the small chamber.

Measurements were taken on the 7th day after the plates were confined in the small chamber. Precise analyses were conducted by the Tokyo Kenbikyuu-in Foundation. Figure 1 show views of the small chamber test. Formaldehyde and acetaldehyde were measured using high-performance liquid chromatography (two LC-20AD liquid pumps and SIL-20AC auto-sampler from Shimadzu). The separation column used was the Ascentis RP-Amide (150 × 4.6 mm²;

Table 3 VOCs and aldehydes analyzed in this study

62 VOCs and aldehydes analyzed by GC/MS		15 aldehydes analyzed by HPLC
2-Propanol	2-Butoxyethanol	Formaldehyde
Pentane	Nonane	Acetaldehyde
Ethyl acetate	Tricyclene	Acetone
Dichloromethane	α-Pinene	2-Furanacrolein
1-Propanol	3-Ethyltoluene	Propionaldehyde
Ethyl acetate	Camphene	2-Butanone
Hexane	4-Ethyltoluene	Butanal
Chloroform	1,3,5-Trimethylbenzene	Cyclohexanone
1,2-Dichloroethane	2-Ethyltoluene	Benzaldehyde
2,4-Dimethylpentane	β-Pinene	Pentanal
1,1,1-Trichloroethane	1,2,4-Trimethylbenzene	Hexanal
Butanol	D4	Heptanal
Benzene	Decane	Octanol
Carbon tetrachloride	Isododecane	Nonanal
Cyclohexane	<i>p</i> -Dichlorobenzene	Decanal
1,2-Dichloropropane	2-Ethyl-1-hexanol	
Bromodichloromethane	3-Carene	
Trichloroethylene	1,2,3-Trimethylbenzene	
Isooctane	<i>p</i> -Cymene	
Heptane	Limonene	
4-Methyl-2-pentanone (MIBK)	4-Ethyl-1,2-dimethylbenzene	
Methylcyclohexane	Undecane	
Toluene	1,2,4,5-Tetramethylbenzene	
Dibromochloromethane	D5	
Butyl acetate	Dodecane	
Octane	Tridecane	
Tetrachloroethylene	D6	
Ethylbenzene	Texanol	
<i>m</i> , <i>p</i> -xylene	Tetradecane	
Styrene	Pentadecane	
<i>o</i> -Xylene	Hexadecane	

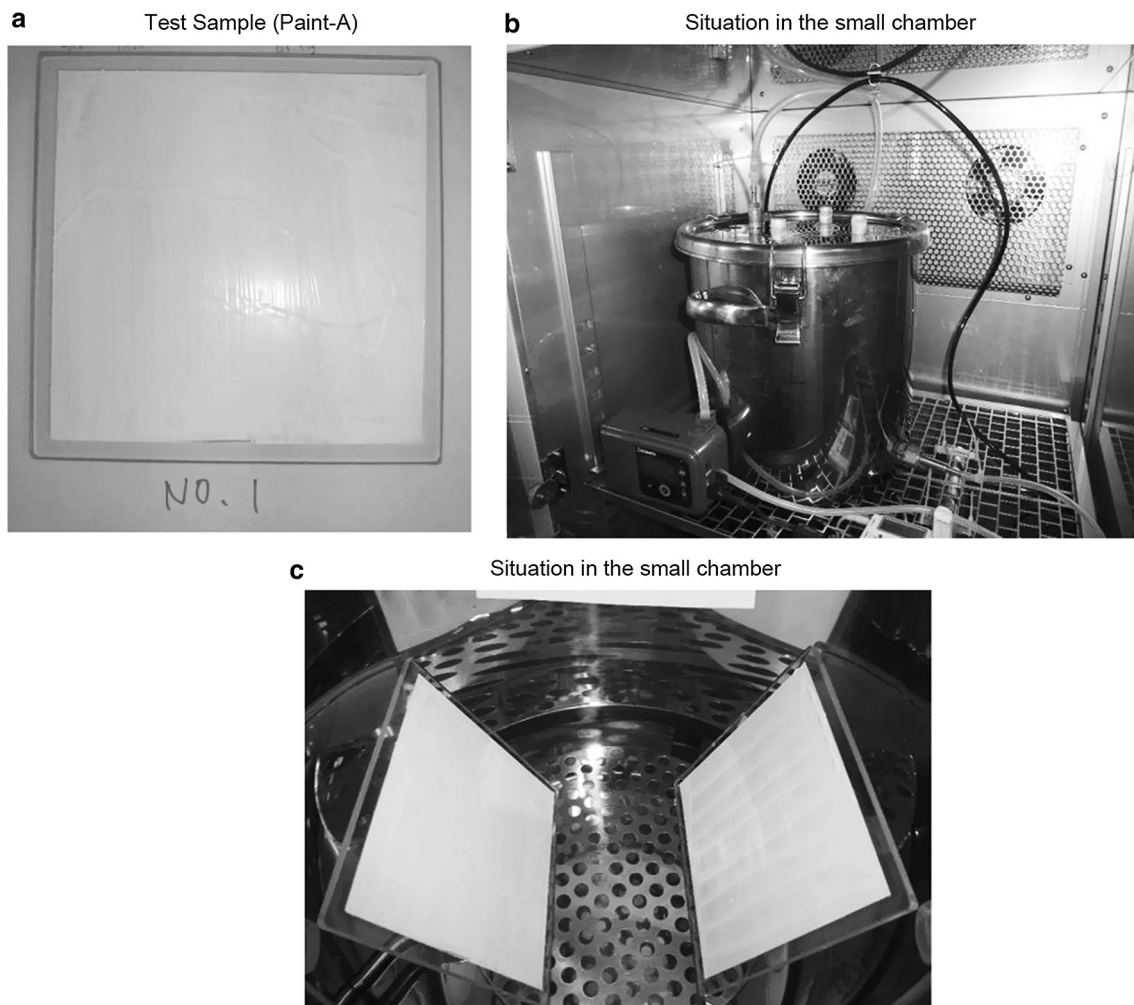


Fig. 1 Small chamber test. **a** PA test sample; **b** situation of the small chamber; **c** plate setup in the small chamber

i.d., 3 μm) made by Sigma–Aldrich. VOCs were measured using a GC6890 and MSD5973N from Agilent. The separation column was the DB-1 (0.25 mm; i.d \times 60 m, 1.0 μm) from GL Sciences. For known substances with standards, individual quantities were calculated. For substances that did not have standards, a library search was performed to infer the substance and quantities were calculated by toluene conversion. In this study, the TVOC was calculated as the toluene equivalent of all substances between C6 and C16, as defined by ISO.

Results and discussion

Results

Table 4 shows the emission rates of each specimen and the toluene-converted TVOC value.

The emission rates of the guideline value substances stipulated by the MHLW were extremely low. Formaldehyde was 1.0 $\mu\text{g m}^{-2} \text{ h}^{-1}$ in the combination sample of PB-SB, but not detected (ND) in other combinations. Compounds for which guideline values were set, such as acetaldehyde, toluene, xylene, ethylbenzene, paradichlorobenzene, and tetradecane, were all classified as ND.

Styrene was detected in all the samples except those of PB, PB + SB, and PC; 1.2 $\mu\text{g m}^{-2} \text{ h}^{-1}$ styrene in the sample of PA, 1.3 $\mu\text{g m}^{-2} \text{ h}^{-1}$ in that of PA + SA, and

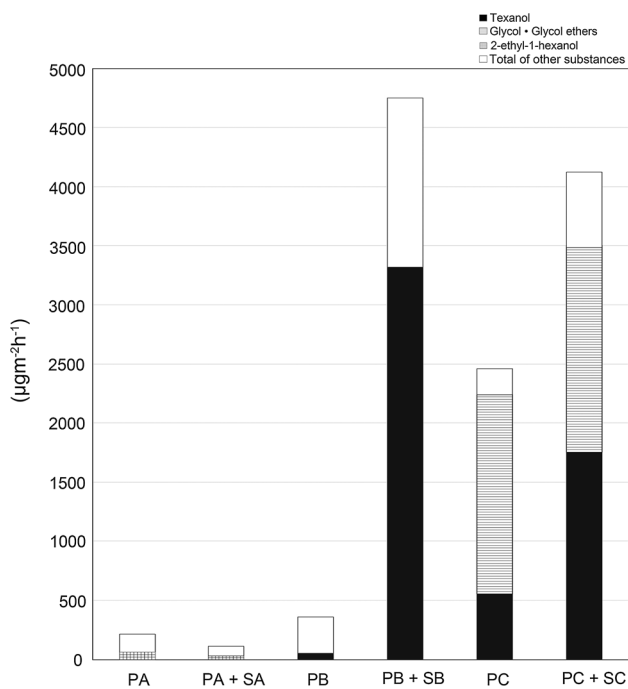


Table 4 Detection levels of selected VOCs and aldehydes analyzed in the test samples

Compounds	PA ($\mu\text{gm}^{-2} \text{h}^{-1}$)	PA + SA ($\mu\text{gm}^{-2} \text{h}^{-1}$)	PB ($\mu\text{gm}^{-2} \text{h}^{-1}$)	PB + SB ($\mu\text{gm}^{-2} \text{h}^{-1}$)	PC ($\mu\text{gm}^{-2} \text{h}^{-1}$)	PC + SC ($\mu\text{gm}^{-2} \text{h}^{-1}$)
Formaldehyde	ND	ND	ND	1.0	ND	ND
Acetaldehyde	ND	ND	ND	ND	ND	ND
Toluene	ND	ND	ND	ND	ND	ND
Xylene	ND	ND	ND	ND	ND	ND
Styrene	1.2	1.3	ND	ND	ND	1.0
Ethylbenzene	ND	ND	ND	ND	ND	ND
<i>p</i> -Dichlorobenzene	ND	ND	ND	ND	ND	ND
Tetradecane	ND	ND	ND	ND	ND	ND
Glycol ethers	ND	ND	ND	ND	1680	1750
Texanol	ND	ND	27	3320	555	1750
Hexane	3.2	3.2	ND	2.0	1.1	1.3
D4	1.7	1.0	ND	ND	ND	ND
2-Ethyl-1-hexanol	32	7.0	ND	ND	ND	ND
D5	1.3	ND	ND	ND	ND	ND
Acetone	ND	ND	ND	ND	ND	1.5
1-Propanol	ND	ND	ND	ND	ND	1.0
Total of other substances ^a	156	84	333	1427	221	622
TVOC	195	96	360	4750	2460	4130

ND not detected

^aValue of excluding substances that could be identified from TVOC

**Fig. 2** Emission rates of the highest emitting compounds

$1.0 \mu\text{gm}^{-2} \text{h}^{-1}$ in that of PC+SC were found. Figure 2 and Table 4 show the substances with the highest emission rates, including TexanolTM, glycol ether, and 2-ethyl-1-hexanol. The substance detected most abundant was TexanolTM, which was detected in all combinations except for PA and PA + SA. TexanolTM was detected at $27 \mu\text{gm}^{-2} \text{h}^{-1}$ in PB; $3320 \mu\text{gm}^{-2} \text{h}^{-1}$ in PB + SB; $555 \mu\text{gm}^{-2} \text{h}^{-1}$ in PC; and $1750 \mu\text{gm}^{-2} \text{h}^{-1}$ in PC + SC. The next most abundant substance detected was glycol ether at $1680 \mu\text{gm}^{-2} \text{h}^{-1}$ in PC and $1750 \mu\text{gm}^{-2} \text{h}^{-1}$ in PC + SC. In PA, PA + SA, PB, and PB + SB, glycol ether was ND. 2-Ethyl-1-hexanol was detected at $32 \mu\text{gm}^{-2} \text{h}^{-1}$ in PA and $7.0 \mu\text{gm}^{-2} \text{h}^{-1}$ in PA + SA. The toluene-converted TVOC was $195 \mu\text{gm}^{-2} \text{h}^{-1}$ in PA; $96 \mu\text{gm}^{-2} \text{h}^{-1}$ in PA + SA; $360 \mu\text{gm}^{-2} \text{h}^{-1}$ in PB; $4750 \mu\text{gm}^{-2} \text{h}^{-1}$ in PB + SB; $2460 \mu\text{gm}^{-2} \text{h}^{-1}$ in PC; and $4130 \mu\text{gm}^{-2} \text{h}^{-1}$ in PC + SC, revealing remarkable differences in the emission rates among W1 class paints.

Discussion

To the best of our knowledge, this is the first study to compare Japanese low-VOC and W1 classified paints (VOC content lower than 1%) and sealers. In this study, VOC contents



and emission rates of three types of low-VOC paints with and without sealers were investigated. Conventionally, sealers are used in combination with paints, and it is recommended to use products produced by the same manufacturer. Therefore, in addition to the performance of the paint, it is also necessary to consider the performance of their combination with the sealers. The results of our experiments revealed that there were big differences in VOCs emitted among low-VOC and W1 classification paints and various combinations of paints and sealers. They also indicate that emission rates of eight substances which are set guideline values by MHLW were low in every sample, but in some samples, other substances such as TexanolTM, glycol ether, 2-ethyl-1-hexanol, and TVOC were detected high. The result for samples of PA, PA + SA, and PB shows extremely few substances detected and low emission rates. For samples of PA and PA + SA, TexanolTM was ND and the VOC emission rates were much lower than those of the other combinations. Conversely, in samples of PB + SB, PC, and PC + SC, TexanolTM was detected at high concentrations, showing that VOC emission rates were higher than those of the other non-TexanolTM-containing combinations. Comparing TexanolTM in paint only and paint + sealer, the detected emission was $27 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PB; $3320 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PB + SB; $555 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PC; and $1750 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PC + SC. Based on these findings, the major source of TexanolTM was assumed to be sealers. Kobayashi et al. (2010) conducted the research on the SBS incidence that occurred in a newly constructed elementary school in Hokkaido, Japan in 2007, and they reported that the causative substances were suspected to be 1-methyl-2-pyrrolidone and TexanolTM emitted from water-based paints. The concentration of TexanolTM in indoor air should therefore be reduced, and attention must be paid to the use of sealers. In the samples of PC and PC + SC case, along with TexanolTM, glycol ether was also detected, and TVOC emission rates were high. The source of glycol ether was suspected to be paint C. Focusing only on paints, the concentration of TVOC was lower in the sample of PB than in that of PC. However, the results of combining paints with sealers indicated the TVOC concentration in the sample of PB + SB was higher than in that of PC + SC. The result depends on the concentration of TexanolTM in the sample of SB; i.e., TVOC concentration varies depending on the combination of paints and sealers. According to previous studies (WHO 1989; EC 1997; German Committee 2018), it was reported that the occurrence of SBS symptoms is more likely with increasing TVOC values in indoor air; therefore, it is recommended not to exceed $300\text{--}400 \mu\text{g m}^{-3}$ (Nakaoka et al.

2014). Emission rates of substances shown in Table 4 were the results of 7 days after application. It means that TVOC levels in indoor air should be reduced by intense ventilation and so on during or 7 days more after the use of some types of combination of paints and sealers.

The emission rates of styrene and 2-ethyl-1-hexanol from samples were low, and they are not regulated by the Japanese government; however, they could act as substitutes for regulated substances and have some influence on people. In recent years, reports establishing the relation between adverse health effects and styrene and 2-ethyl-1-hexanol have appeared (Loomis et al. 2018; Kamijima et al. 2002; Wieslander et al. 2010; Sakai et al. 2006). The total concentration of other substances (value of excluding substances that could be identified from TVOC) was $156 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PA, $84 \mu\text{g m}^{-2} \text{ h}^{-1}$ in the samples of PA + SA, $333 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PB, $1427 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PB + SB, $221 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PC, and $622 \mu\text{g m}^{-2} \text{ h}^{-1}$ in PC + SC. These results show that although their individual peaks are low, if they are totaled, there are cases wherein the “the totals of other substances” can account for much of the emitted VOC content. We therefore also should pay attention to the chemicals that are excluded from TVOC. Schieweck and Bock (2015) concluded from their study that a quality label of low-VOC paints just meant that a specific product fulfilled specific requirements, and it does not state that a specific product is free of chemicals or emissions. To prevent adverse health effects from chemicals in indoor air, it will be necessary to conduct studies on substances even if they are excluded from regulation targets, and the products were labeled low or free VOCs.

Conclusion

VOC contents and emission rates of W1 class, low-VOC, water-based, and emulsion paints with and without sealers were examined in this study. Great differences in VOC emission rates among them were recognized. Paints with VOC contents lower than 0.2% and 0.3% (PA and PB) showed VOC emission rates that were far lower than those of paints with 1% or less VOC content (PC). The main constituents of these TVOC were TexanolTM and glycol ether. As seen in samples of PB, PB + SB, PC, and PC + SC, the selection of paints and sealers greatly affects the VOC emission rates. Recently, many paints and sealers have been made

commercially available; however, there is not enough information for consumers regarding product emission testing. It is required finer VOC classification system in paints and sealers to assist consumers in choosing healthier environments. For the purpose of reducing SBS and protect public health, it is important to devote more effort for investigating paints and sealers.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflicts of interest.

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