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Compounds in Indoor Air of Newly Built Houses
Complying with the Amended Japanese
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Synopsis: This study was performed to investigate the concentrations of ethylbenzene, styrene, toluene, and total xylenes in indoor air and their changes in three newly built houses. Air was trapped with TENAX tubes and analyzed quantitatively by GC/FID using the absolute calibration method. In these houses, the concentrations of target compounds were below the guidelines of the Ministry of Health, Labor, and Welfare. The differences among the houses were influenced by the patterns, but not by the types of compounds. In this study, a mechanical ventilation system reduced concentrations of volatile organic compounds in indoor air by 10% to 40%.

Keywords: Survey, Volatile Organic Compounds, Indoor Air, Newly Built House, Amended Japanese Building Standard Law of 2003

Introduction

To save energy, buildings in Japan are constructed to be highly airtight and adiabatic. Therefore, it is difficult for formaldehyde or volatile organic compounds (VOCs) emitted from building materials or furniture to diffuse out of the buildings. This can result in sick building syndrome caused by chemical pollution of indoor air, which has become a serious social problem in Japan.

As a measure to prevent sick building syndrome, the Ministry of Health, Labor, and Welfare (MHLW) of Japan has prepared guidelines for the concentrations of 13 compounds in indoor air. However, these are only guidelines¹⁾ and have no legal backing. On the other hand, the Building Standard Law was revised and enforced by the Ministry of Land, Infrastructure, and Transport (MLIT) on July 1, 2003. This law forbids the utilization of building materials with added chlorpyrifos in buildings with rooms that are likely to be used continuously²⁾.

Moreover, this law places limitations on the amounts of materials belonging to each emission rate of formaldehyde that can be used based on the ventilation frequency of buildings. All buildings must

have mechanical ventilation systems that can disperse the volatile compounds emitted from furniture, *etc*²⁾.

The concentrations of these two compounds in indoor air will be reduced, so that they have legal backing to be utilized them for building.

According to the survey conducted by the MLIT since 2000, actually, the concentration of formaldehyde decreased year after year, and there were less than 1% of houses whose concentrations of these compounds were higher than the guidelines in the 2004's survey³⁾.

Many surveys of chemical compounds as indoor air pollutants have been performed with the objective of indoor air quality assessment^{4) 5) 6) 7)}, but there have been few surveys of the compounds except formaldehyde.

Few researches has been reported to investigate the actual indoor air pollutants or such changes in concentrations of formaldehyde or VOCs with the mechanical ventilation system ON, such investigation is required as the basic data of chemical pollutant in indoor air.

Therefore, this study was performed to evaluate the chemical contamination of indoor air and the change of concentration of VOCs under the mechanical ventilation system ON in newly built houses that comply with the amended Building Standard Law.

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1 Methods

1.1 Houses Measured

The measurement was performed in three houses that were able to be measured before moving in. From September 2002 to March 2003, we performed measurements in one newly built house in each of Tokyo, Miyagi, and Tochigi, as shown in Table 1. The houses were all highly airtight and highly adiabatic and were compliant with the Building Standard Law enforced in 2003 after completion of construction and before people had moved in.

1.2 Target compounds

Six types of target compound were examined: four VOCs –i.e. ethylbenzene, styrene, toluene and total xylenes, and two carbonyl compounds –i.e. formaldehyde and acetaldehyde. They were selected in the compounds which had the guidelines of concentrations by MHLW.

Total xylenes were determined from the total concentrations of *o*-xylene and *m,p*-xylene. The indoor air concentration guidelines of these compounds were defined by the Ministry of Health, Labor, and Welfare as 880 ppb, 50 ppb, 70 ppb, 200 ppb, 80 ppb, and 30 ppb, respectively¹⁾.

1.3 Air Sampling

Sampling tubes were conditioned by heat purge at 200°C for half an hour to remove contamination, and then cooled to room temperature. After purging, tubes were sealed by caps made of brass and Teflon until sampling. Toluene, ethylbenzene, styrene, and total xylenes were trapped in a wooden-floored room on the 2nd floor at a rate of 100 ml/min for 10 min as determined using an automatic sampler attached to a 12-channel pump with a flux meter, using a trap tube after fixed time intervals of 0.5 h in house A, and 1.0 h in both houses B and C. These compounds in outdoor air were trapped twice with the 1-channel pump with a flux meter in the

same way and the data were averaged. The trap tubes are glass tubes 160 mm in length and 6 mm in external diameter with an internal tube 3 mm in diameter filled with 0.15 g of TENAX TA.

Carbonyl compounds were trapped at a rate of 1000 ml/min for 30 minutes with a DNPH cartridge (SUPELCO) twice in the same room and outside.

1.4 Air sampling protocol

Fig. 1 shows the protocol used for air sampling. Briefly, air was sampled in accordance with “the sampling and analysis method of chemical compounds in indoor air” proposed by the MHLW of Japan⁹⁾. For ventilation, we first opened all the doors and windows that faced the outside. All the doors inside the houses, including those of storage spaces, were also opened to allow the compounds in the indoor air to reach uniform concentrations throughout the house. The measurement instruments were set in the room on the second floor and the automatic sampler was started. After more than 30 minutes of ventilation, all the doors and windows facing the outside were shut and we left the house. The automatic sampler obtained indoor air samples at regular intervals of 0.5 h in house A and 1.0 h in both houses B and C under the operation of a mechanical ventilation system. The time that the doors and windows facing the outside were shut, was defined as 0 h, and the point of starting previous ventilation was -0.5 h.

The sampling tubes and cartridges were sealed with caps, and were analyzed within 24 hours.

The temperature and the relative humidity of the room and the outside were measured with the portable thermohygrometer (thermorecorder, T&D) at every 10 minutes throughout the measurements.

Table 1 Features of the sampling sites

Sample	Sites	Date	Weather	Floor Area (m ²)	Ventilation System / Air Volume (m ³ /h)
House A	Tokyo	09/27/02	rain	Total=105.9 1F=56.3 2F=49.6	AVR-150FT, Air Water Emoto /75-100
House B	Miyagi	12/13/02	snow	Total=143.4 1F=77.2 2F=66.2	unidentified
House C	Tochigi	03/20/03	fine	Total=105.9 1F=52.9 2F=52.9	FY-Z40DB2K, Matsushita / 65

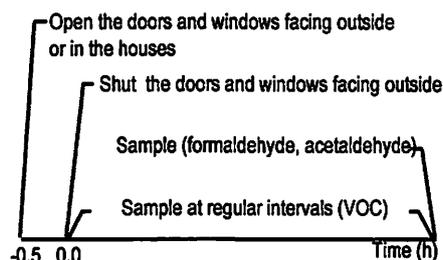


Fig. 1 Air Sampling Protocol

1.5 Analysis

Tables 2 and 3 show the operating conditions of the Gas Chromatograph with Flame Ionization Detector (GC/FID) and High Performance Liquid Chromatograph (HPLC), respectively. Analysis was begun less than 24 hours after sampling.

The samples were analyzed with standard gas (NMOG, 1 ppm, Taiyo Toyo Sanso) which was diluted to 5 ppb or standard solution (13 components, 30 $\mu\text{g/ml}$, SUPELCO) which was diluted to 0.3 mg/l.

The qualitative analysis was performed by referring to the retention time of each peak of the standard toluene, formaldehyde and acetaldehyde were analyzed quantitatively with each compound of the standard, and ethylbenzene, total-xylenes and styrene were analyzed likewise with *o*-xylene of standard gas by the absolute calibration method.

The repeatability test using standard gas containing ethylbenzene (5.0 ppb), toluene (5.8 ppb) and *o*-xylene (5.0 ppb) was carried out. The detection limits of them were 2.2 ppb, 1.7 ppb and 2.6 ppb, and the quantitation limits were 7.2 ppb, 5.5 ppb and 8.7 ppb, respectively. Moreover, the limit value of detection and quantitation of two carbonyl compounds were 0.04 mg/l and 0.13 mg/l, respectively.

The carbonyl compounds except formaldehyde and acetaldehyde, which was able to be analyzed quantitatively with standard liquid, and the sorts of them were noted in chromatograms.

2 Results and Discussion

2.1 Concentrations of target compounds

Figs. 2, 3, and 4 show GC/FID chromatograms obtained at 5.0 h in house A, at 8.5 h in house B, and at 9.5 h in house C, respectively. Figs. 5, 6, and 7 show HPLC chromatograms in house A, in house B, and in house C, respectively. The target compounds were detected in each chromatogram such as in Figs.

The target compounds were detected in each chromatogram in Figs. Three carbonyl compounds other than formaldehyde and acetaldehyde were detected: *i.e.*, acetone in all houses, 2-butanone in houses A and C, and valeraldehyde and hexanale in house C.

Table 4 shows the guidelines and measurement results of target compounds in outdoor and indoor air.

The measurement data of VOCs were divided into the rising part and the steady-state part, and the average concentrations in the steady-state were calculated. The concentrations in indoor air are the average data in Table 4.

In outdoor air of house A, toluene, formaldehyde and acetaldehyde concentrations were higher than about 5 ppb. With the exception of these data, the outdoor concentrations of these compounds were very low or not detectable in the test houses.

The concentrations of all target compounds in all test houses were higher than those outdoors. Therefore, it was assumed that these compounds were emitted inside the houses.

Table 2 Operating conditions in GC/FID analysis

Instrument	GC-7A (Shimadzu)
Desorption Temperature	200°C
Detector	Flame Ionization Detector
Column	HP-1, Internal Diameter: 0.32 mm, Length: 50 mm, Thickness 1.0 μm
Column flow rate	Sample Injection: 20 ml/min, Analysis: 3 ml/min
Injector	200°C
Carrier Gas	Helium (20 ml/min)
Oven Temperature	-40°C, 3 min \rightarrow 30 °C/min \rightarrow 20°C \rightarrow 40 °C/min \rightarrow 170°C, 5min

Table 3 Operating conditions in HPLC analysis

Instrument	JASCO 860-CO
Detector	Ultraviolet 360nm
Column	Crest Pak C18S Internal Diameter: 4.6 mm, Length: 150 mm
Carrier	55% Acetonitrile/ Water Solution (8 ml/sec)
Injection Volume	20 μl

The concentrations of target compounds in indoor air were not greater than the guidelines recommended by the MHLW in any of the houses in the present study. In houses A and C, toluene, acetaldehyde and formaldehyde showed higher concentrations among the 6 target compounds examined. The concentrations of toluene in house A and acetaldehyde in house C were the highest, both of which were detected at concentrations of about nearly 30 ppb. The guideline of acetaldehyde is 30 ppb, therefore the concentration in house C was high level.

According to the survey conducted by the MLIT since 2000, the concentration of formaldehyde and toluene decreased actually, but the concentration of acetaldehyde were constant every year³⁾. Although there were less than 1% of houses whose concentrations of formaldehyde and toluene were higher than the guidelines, there were 10% of houses whose concentrations of acetaldehyde were higher than the guidelines in the 2004's survey³⁾.

It is necessary to consider the regulation utilizing the building materials added acetaldehyde as well as formaldehyde.

In house B, most of target compounds showed the lower concentrations than those in the other houses, especially formaldehyde and acetaldehydes showed the low concentration about 3 ppb. However ethylbenzene and total-xylenes showed higher concentrations than the others, especially the former showed approximately 10 ppb which is more than two times as the others.

In previous surveys of indoor air quality, formaldehyde and toluene were detected at high concentrations in many cases^{4) 5) 6) 7)}. In the present study, houses A and C showed similar tendencies in terms of indoor air quality. On the other hand, the concentrations of these compounds in house B were low, which was specific to this house.

There are many reports that toluene in indoor air was detected, and the source was assumed that the compound is emitted from the building materials such as diluents of paint or fixing materials⁹⁾.

Although the interior finishing materials in house B were unpainted wooden building materials, the walls and ceiling in houses A and C were covered with vinyl wallpaper. Therefore, it was assumed that the concentration of toluene, which is emitted from fixing materials, such as adhesives, was very low in house B.

Table 4 Concentrations in outdoor air and in indoor air, and their guidelines

Compounds	Guideline	(ppb)					
		Outdoor Air			Indoor Air		
		A	B	C	A	B	C
ethylbenzene	880	1.2	nd	nd	4.2	9.9	3.5
styrene	50	nd	nd	nd	8.0	2.9	3.8
toluene	70	9.5	1.3	nd	25.8	5.8	16.4
total xylenes	200	1.5	1.0	nd	7.7	10.6	8.1
formaldehyde	80	4.8	nd	2.1	16.3	2.7	15.0
acetaldehyde	30	4.0	nd	3.6	12.3	2.4	27.9

*Shaded Region: the data of ethylbenzene, styrene, toluene and total-xylenes were averaged concentrations in the Steady-State

*The values which were lower than the limits of detection and quantitation were mentioned as references. *nd: lower than 1.0 ppb

Table 5 Ambient temperature and relative humidity in houses A, B, and C

Sampling	Temperature (°C)		RH ^{*)} (%)	
	Average	SD	Average	SD
A Indoor	22.3	0.26	60.9	0.93
Outdoor	18.6	0.15	72.4	3.52
B Indoor	3.3	0.56	68.8	3.09
Outdoor	0.0	1.07	92.4	9.60
C Indoor	10.1	1.59	55.3	6.84
Outdoor	5.5	3.13	69.3	14.39

^{*)} RH: Relative Humidity

^{**)} SD: standard deviation

The ambient temperatures and relative humidity in houses A, B, and C are shown in Table 5.

On sampling A, the standard deviations of indoor and outdoor temperatures were very low (0.26 and 0.15, respectively). The deviation of relative humidity showed a similar pattern (0.93 and 3.52, respectively). The measurements in house A were finished within half a day, and therefore the environmental conditions were steady.

However, sampling C showed larger temperature deviations as sampling was performed from night to morning in spring at a time when temperature varied greatly.

In house B, measurements were performed through the night in winter, and thus the ambient temperatures were extremely low, with average values of 3.3°C indoors and 0.0°C outdoors. It was snowing during measurement B, and thus the relative outdoor humidity was very high.

It has been reported that increases in temperature result in elevated concentrations of volatile compounds⁴⁾. As all measurements in the present study were carried out in cool regions of Japan from autumn to winter, the ambient temperatures were relatively low.

Therefore, the concentrations of target compounds would be higher than those reported this time if the ambient temperature is comparatively high like in summer when the buildings are finished.

2.2 Changes in target compound concentrations

Fig. 8 shows the changes in the concentrations of 4 types of target VOCs in house A. All target VOCs showed the lowest concentrations during ventilation at -0.5 h. After the doors and the windows were shut, their concentrations increased gradually and reached a steady state from 1.0 h to 1.5 h. All of the last data at 5.0 h were slightly higher than the earlier values.

The concentration of toluene changed in higher level than the other VOCs in house A, and it rose to 1.5 h and reached a steady state at about 25 ppb. Styrene and total xylenes levels showed similar changes. They reached a steady state as about 9 ppb at 1.0 h. The concentration of ethylbenzene increased to about 5 ppb at 1.0 h.

Fig. 9 shows the changes in the concentrations of the target VOCs in house B. All target VOCs showed the lowest concentrations at 0.5 h, and rose by 4.5 h or 5.5 h. Target compounds showed higher concentrations at -0.5 h, thus it is assumed that the levels of concentrations were high before measurement with some causes.

The concentrations of total xylenes and ethylbenzene reached a steady state about 10 ppb at 4.5 h. Styrene showed the increasing concentrations till 5.5 h, and showed the concentration about 3 ppb of a steady state. Toluene showed specific changes after 5.5 h, falling to about 5 ppb from 8 ppb, and then rising to about 6 ppb at

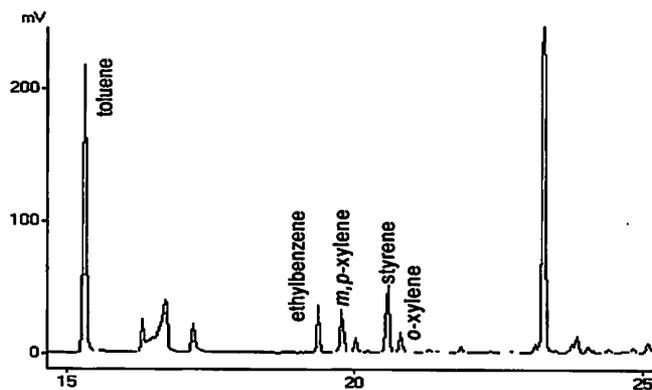


Fig. 2 GC/FID chromatogram of indoor air in house A at 5.0 h

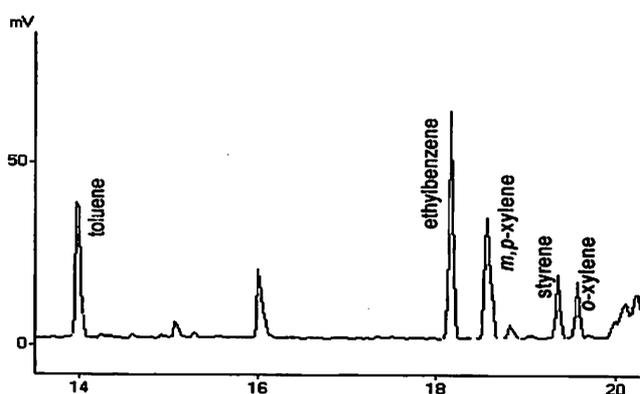


Fig. 3 GC/FID chromatogram of indoor air in house B at 8.5 h

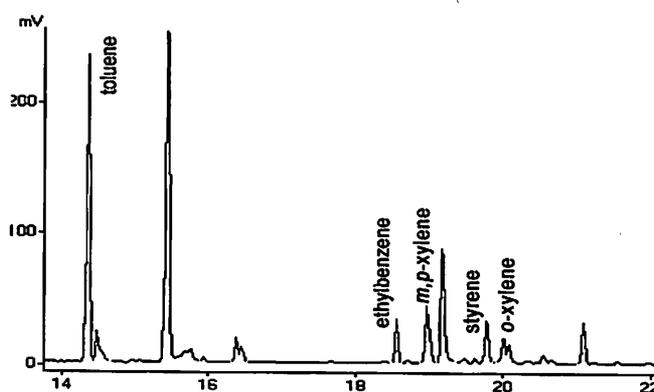


Fig. 4 GC/FID chromatogram of indoor air in house C at 9.5 h

9.5 h. However, the concentrations of the other compounds changed little from 4.5 to 9.5 h.

Fig. 10 shows the changes in the concentrations of the target VOCs in house C. All target VOCs showed the lowest concentrations during ventilation at -0.5 h. After the doors and the windows were shut, the concentrations of target compounds rose gradually until 3.5 h.

Toluene level continued to increase steadily and reached around 19 ppb at 8.5 h. While the concentrations of ethylbenzene and styrene were stable from 3.5 h to 8.5 h, total xylenes level fell to about 5 ppb from 10 ppb, from 3.5 h to 5.5 h and remained at this concentration.

The doors and windows faced outside were not opened during measurement, thus it is assumed that the causes of the decreasing of toluene in house B and total-xylenes in house C after increasing concentrations were method of measurement such as the conditions of sampling tubes. It's necessary to make the causes clear at the next stage.

The concentrations of all target VOCs rose and reached a steady state in all houses from 1.0 h to 5.5 h. Thus, the patterns of changes were similar in all houses and were dissimilar between types of compound.

Conclusion

This study was performed to investigate the concentrations of target compounds, i.e. ethylbenzene, styrene, toluene, total xylenes, formaldehyde, acetaldehyde and four Volatile Organic Compounds (VOCs), under the mechanical ventilation system ON in three newly built houses. The knowledge from this study are as follows.

The concentrations of target compounds in indoor air did not exceed the guidelines recommended by the Ministry of Health, Labor, and Welfare in any of the houses, therefore the regulations can be said to be effective.

The concentration of acetaldehyde almost reached the guidelines, so that it is necessary to consider the regulation utilizing the building materials added it.

And the patterns of changes were similar in all houses and were dissimilar between types of compound.

It is important to care about future trend of the regulation.

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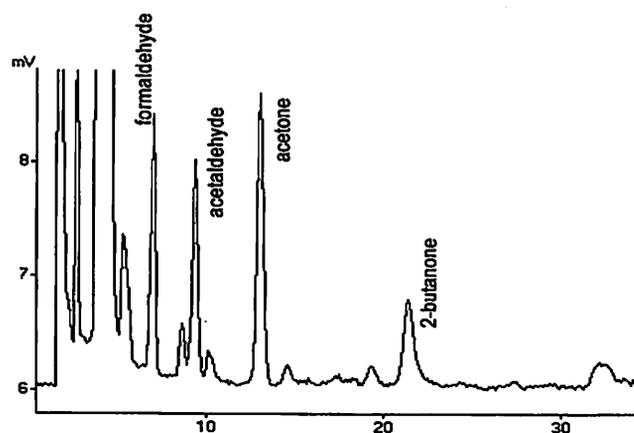


Fig. 5 HPLC chromatogram of indoor air in house A

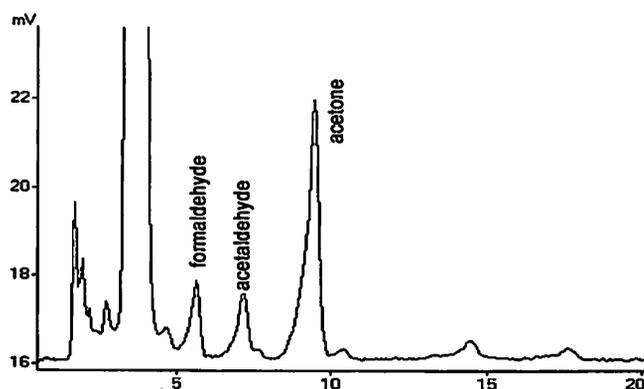


Fig. 6 HPLC chromatogram of indoor air in house B

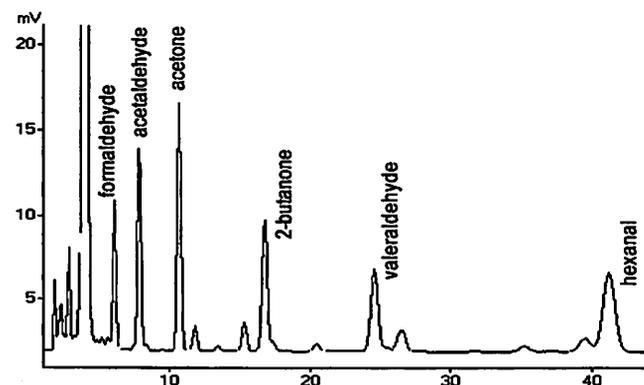


Fig. 7 HPLC chromatogram of indoor air in house C

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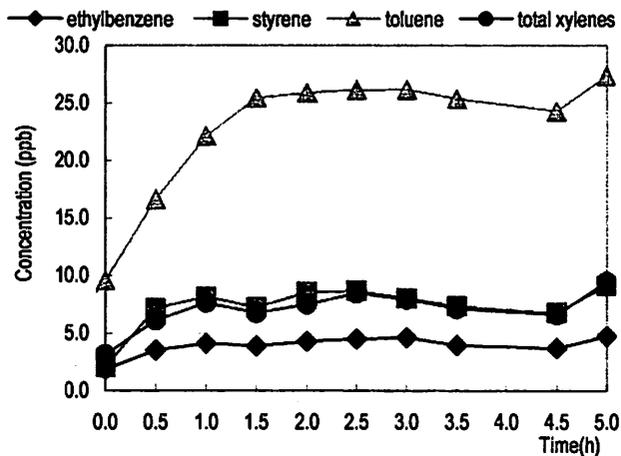


Fig. 8 Changes in concentrations in house A

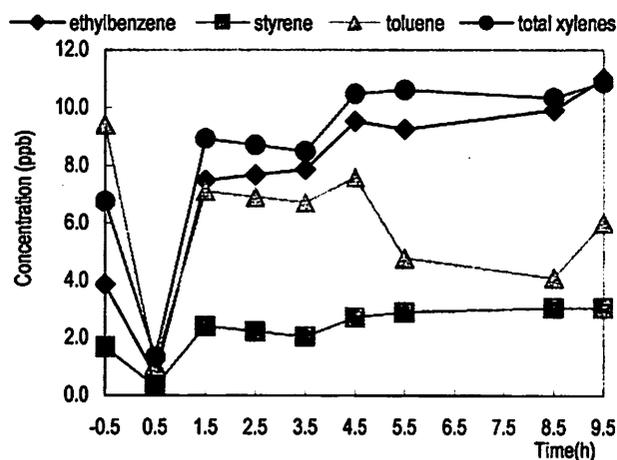


Fig. 9 Changes in concentrations in house B

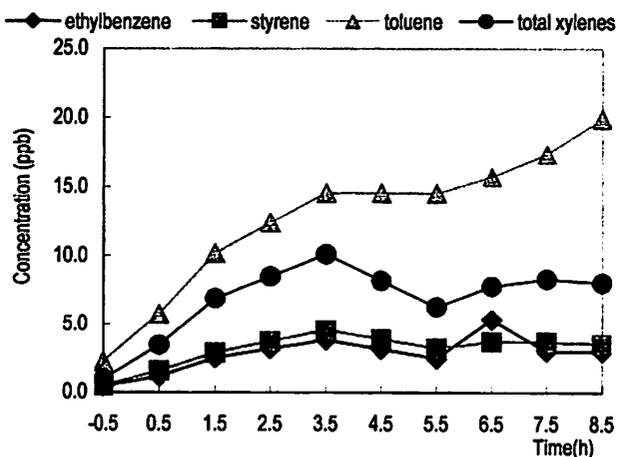


Fig. 10 Changes in concentrations in house C

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2003年改正建築基準法を満たした新築住宅室内空気における揮発性有機化合物濃度変化に関する調査

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keyword: 調査、揮発性有機化合物、室内空気、新築住宅、改正建築基準法(2003)

日本でシックハウス症候群が社会問題化して久しい。この問題を防ぐために、日本の厚生労働省は室内空気における13物質の濃度指針を示している。しかし、これらはガイドラインにすぎず法的拘束力を持たない。一方、2003年7月1日に国土交通省によりシックハウス対策を盛り込んだ建築基準法が改正・施行された。この法律は居室を持つ建築物において、chlorpyrifosを添加した建材の使用を禁止し、建築物の換気回数に基づき、formaldehyde放散量ごとに建材の使用可能面積に制限を設けている。また家具などから放散される揮発性化合物を考慮し、機械換気設備の設置を義務付けている。

室内空気質評価を目的とした室内空気の化学汚染調査、とりわけformaldehydeとする調査は数多く存在するが、それに比べてformaldehyde以外の化合物を扱う調査は多くない。また、換気設備を作動させた場合の濃度実態や濃度上昇パターンを調査した報告は少ない。

本研究では改正建築基準法を満たした新築住宅における実測をふまえて、室内空気の化学汚染実態を明らかにすること、及び揮発性有機化合物(VOCs)の経時変化を調査することを目的とした。

引渡し前の新築住宅3戸でVOCs4種(ethylbenzene、styrene、toluene、total xylene)及びカルボニル化合物2種(formaldehyde、acetaldehyde)の室内濃度と上記VOCs変化パターンを調査した。VOCsはTENAX TAを充填したガラス製捕

集管により流量計付きポンプで100 ml/minで10分間空気捕集し、GC/FIDにより分析した。カルボニル化合物はDNPH cartridgeにより流量計付きポンプで1000 ml/minで30分間空気捕集し、高速液体クロマトグラフで分析を実施した。空気採取は2回フローリング洋室及び外気で実施した。測定中、24時間機械換気設備を作動させた。

本研究の測定住宅における対象化合物濃度はすべて厚生労働省により定められた指針をすべて下回った。したがって、改正建築基準法がVOCs濃度、とりわけ使用面積制限を行っているformaldehyde濃度低減効果を示す一例といえるであろう。

しかしながら、測定対象中1戸の住宅におけるacetaldehyde濃度が指針値に非常に近い濃度を示した。したがって、この化合物に対して、建材使用制限等何らかの対策が必要であると考えられる。

また、24時間換気設備を作動させた状態でのVOCs濃度は徐々に上昇し1.0 hから5.0 hで定常に達した。経時変化のパターンは、物質の種類でなく住宅ごとに同様の傾向を示した。

本研究では、2003年建築基準法改正後の新築住宅における測定により、VOCs、カルボニル化合物の濃度実態、経時変化についての基礎的データを得た。今後、さらにデータを増やしていくことが必要であるとする。

また、規制対象2物質以外の物質に対する対策の必要性について示唆した。今後、関係省庁の規制の動向を見守りたい。

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