# Aldehydes and VOCs in newly-built unoccupied houses in Tokyo

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## ABSTRACT

The Ministry of Health, Labor and Welfare in Japan have published the guideline values for 14 chemical compounds including formaldehyde, toluene, xylenes etc., in order to solve the "Sick house syndrome" problem. In this study, field measurements on indoor air quality were conducted in newly-built houses which had not been occupied.

The concentrations of aldehydes and VOCs, the ventilation rate, the emission rate of formaldehyde from each surface material, temperature and humidity were measured in three houses in summer and in two houses in winter. The emission rate of formaldehyde from each material was measured by TEA-dish method developed by us.

The results showed that the concentrations of formaldehyde measured were lower than the guidelines. However, when the concentrations of formaldehyde measured were converted into the concentration under 25$^{o}$C and 50%rh, it was found that some of the converted concentrations were higher than the guidelines. Regarding the relative emission rate of formaldehyde, the inside surfaces and the shelves of the closets measured in summer showed higher values than the other surface, e.g. walls, ceilings and floors. The concentrations of toluene, acetaldehyde and TVOC were higher than the guidelines.

## INDEX TERMS

Aldehydes, TVOC, Emission Rate, Measurement Technique, Toluene

## INTRODUCTION

In order to solve the "Sick house syndrome" problem, the guideline values for 14 chemical compounds including formaldehyde, toluene, xylene etc. have been published by the Ministry of Health, Labor and Welfare in Japan. The emission rate of those chemical compounds from building materials decreases with the time elapsed from completion. The concentration of those chemical compounds is affected not only by the kinds of building materials used, but also by the ventilation rate and other emission sources, which are determined by occupants' behavior. For this reason, field measurements on indoor air quality were conducted in newly-built houses which had not been occupied to avoid effects of occupants' behavior.

## METHODS

### Outline of houses

Table 1 shows the outline of the houses where measurements were carried out. Houses A, B and C, whose lots are subdivided from a larger lot, are built of the same materials. House D was a custom-made house and House E was ready-built housing. Each had 2 or 3 bedrooms. The measurements in House A, B and C were conducted in August, 2001 and the measurements in House C and D were in November, 2001.

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Table 1. Outline of the newly-built houses

| | City | Structure | Floors | Construction period |
|---|---|---|---|---|
| Houses A -C | Tokyo | Wooden frame | 2 stories + basement | 2001.Feb.-2001.July |
| House D | Tokyo | Wooden frame | 2 stories + basement | 2001.May.-2001.Nov. |
| House E | Tokyo | Wooden frame | 2 stories | 2001.July.-2001.Oct. |

### Ventilation

Twenty-four hours before the measurement, after fully ventilating, all the doors and windows were closed and ventilating systems were switched off. To measure air change rate, the decay of SF6 which was added as tracer gas was monitored by the photoacoustic infrared detection method at 1.2m height from the floor. Air temperature and relative humidity were recorded every 5 minutes.

### Concentration of aldehydes and VOCs

In order to measure concentration of formaldehyde, DNPH-HPLC method was used. The sampling speed was 300mL/min and the sampling period was 30 minutes. For VOCs, activated charcoal was used as sorbent. The sampling speed was 100mL/min and the sampling period was 21 hours. VOCs were analysed by gas chromatograph.

### Emission rate of formaldehyde

The emission rate of formaldehyde from each material was measured by TEA-dish method in which filter paper containing 10% 2, 2', 2''-nitrilotriethanol (triethanol amine: TEA) was used as adsorbent[Hori, Tsukahara and Iwata, 2001]. In an aluminium dish (197 mm in diameter and 10 mm in depth), TEA filter paper was fixed with magnet as shown in Figure 1. The dish was attached to the floors, the walls, the doors, the ceilings, and the inside surfaces and the shelves of the closets as shown in Figure 2. The sampling period was 20 hours. After the sampling period, the filter paper was put in a bag, which was sealed air-tightly. Radiant temperature near the sampling point was measured by spot thermometer (Minolta).

Figure 1. TEA-dish method


Figure 2. Sampling by TEA-dish method

AHMT (4-Amino-3- hydrazine - 5- merecapto -1,2,4-triazole) method was used for analysis. Absorption of light (550 nm of wave length) was measured by ultraviolet and visible spectrophotometer (SHIMADZU) and the concentration of formaldehyde was calculated according to Bouguer-Beer law. In this study Emission Rate (ER) (μg/m$^2$h) was defined as apparent emission rate which is computed by the following equation.

$$ER = ( \alpha - \alpha_b)\ kd / f / s / t \qquad (1)$$

$ER$: emission rate (μg/m$^2$h) $\alpha$: absorbance of sample $\alpha_b$: absorbance of blank
$kd$: dilution coefficient
$f$: conversion factor (from absorbance to weight ) decided from calibration line
$s$: area of material covered with the dish (m$^2$) $t$: sampling period (h)

## RESULTS

### Temperature, humidity and ventilation rate

Table 2 shows temperature, relative humidity and ventilation rate. Air change rate ranged from 0.1 to 0.2 (h$^{-1}$).

Table 2. Temperature, humidity and ventilation rate

| | | Temperature (degC) | | | RH (%) | | | Ventilation Rate (m$^3$/h) |
|---|---|---|---|---|---|---|---|---|
| | | Max. | Ave. (SD) | Min | Max. | Ave. (SD) | Min | |
| House A | living room | 34.4 | 30.1(1.3) | 28.2 | 72 | 65.8(2.5) | 57 | 8.0 |
| | bedroom | 29.5 | 27.3(0.3) | 26.8 | 89 | 81.2(2.7) | 71 | 2.2 |
| House B | living room | 32.5 | 30.2(1.1) | 28.4 | 71 | 64.1(1.9) | 60 | 10.6 |
| | bedroom | 28.8 | 26.8(0.2) | 26.5 | 87 | 81.8(2.2) | 73 | 2.9 |
| House C | living room | 31.4 | 29.3(0.9) | 27.7 | 77 | 66.6(1.7) | 63 | 14.3 |
| | bedroom | 30.2 | 26.7(0.4) | 26.2 | 88 | 84.6(1.9) | 72 | 4.5 |
| Outdoor (A,B,C) | | 32.9 | 27.6(2.3) | 24.4 | 87 | 74.4(8.5) | 54 | - |
| House D | living room | 19.4 | 16.7(1.8) | 13.6 | 67 | 63.2(4.7) | 48 | 5.7 |
| | bedroom | 18.3 | 16.6(0.7) | 14.5 | 73 | 67.6(2.0) | 62 | 3.1 |
| House E | living room | 18.1 | 14.5(2.2) | 10.8 | 69 | 65.4(4.4) | 52 | 6.2 |
| | bedroom | 19.9 | 14.8(1.9) | 12.3 | 71 | 67.7(2.5) | 53 | 5.0 |
| Outdoor(D,E) | | 19.9 | 10.8(4.1) | 5.3 | 99 | 79.2(18.4) | 36 | - |

### Concentration of formaldehyde

Figure 3 shows concentration of formaldehyde. Since concentrations were measured under different temperatures and humidity, they have been converted to the concentration under 25$^o$C and 50%rh according to the following equation (Inoue 1997).

$$C_o = C * 1.09^{(t_0 - t)} / ( 1 + 0.01(H - H_0)) \qquad (2)$$

$C_o$: modified concentration $C$: measured concentration
$t_o$: standard temperature(=25$^o$C) $t$: measured temperature
$H_o$: standard humidity(=50%rh) $H$: measured humidity

Outdoor concentration was 24μg/m$^3$ under 27.6$^o$C and 74.4%rh.in summer and 12 and 19 μg/m$^3$ under 10.8 $^o$C and 79.2 %rh in winter. These values are higher than the result in previous studies showing about 3 to 4 μg/m$^3$ in winter. One of the possible explanations of these high values is that the houses measured in this study were just recently completed, so construction work could have influenced the outdoor air around the houses.

The concentration of formaldehyde in the all rooms measured is lower than the guideline value in Japan, 100 μg/m$^3$. However, after modification, concentrations in three rooms measured in winter were higher than the guidelines.

Figure 4 shows the concentration of acetaldehyde. All of the rooms showed higher concentration than the guideline value in Japan, 48μg/m$^3$. It is said that the primary source of acetaldehyde is ETS. However, the reason for those high concentrations in this study could not be identified and for acetaldehyde, further study including sampling and analysing methods is required.

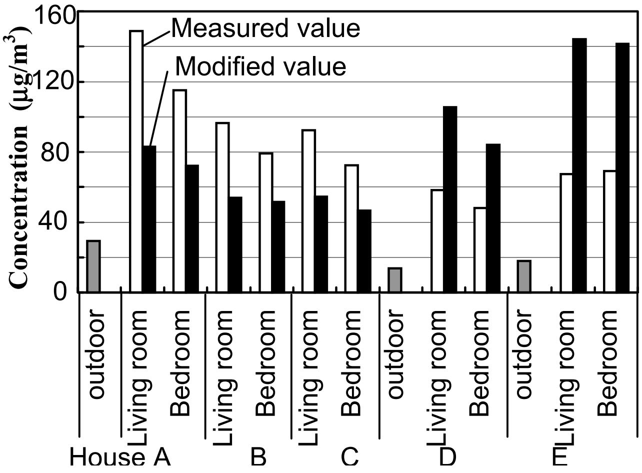


Figure 3. Concentration of Formaldehyde

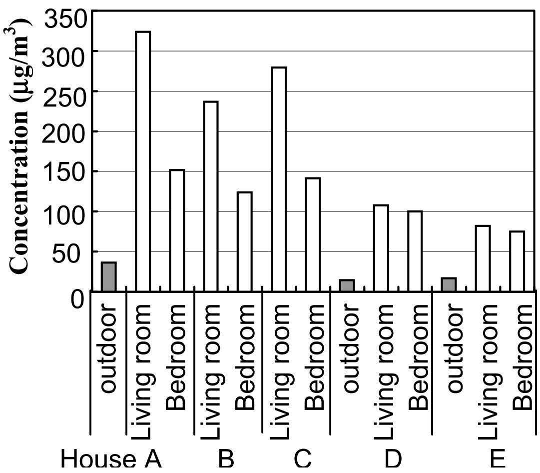


Figure 4. Concentration of Acetaldehyde

**Relative emission rate of formaldehyde**

Figure 5 shows the emission rate of formaldehyde for each material in Houses A, B, and C. Radiant temperature was also shown. In these houses, the inside surfaces and the shelves of the closets and the cupboards measured in summer showed higher values than the other surface. The emission rate of floors, walls, and ceilings ranged 1 to 5μg/m$^2$h. Houses D and E showed lower emission rates which were less than 6μg/m$^2$h except the wall and floor of a loft in House E. This can be explained by the fact that the radiant temperature in winter ranged from 5 to 15 $^o$C while that in summer ranged from 25 to 32$^o$C.

Contribution of each part to total emission was calculated according to the following equation.

Contribution of each part to total emission (%) =($ERi$ $Si$) / Σ ($ERi$ $Si$) (3)

$ERi$: Emission Rate of each material(μg/m$^2$h) $Si$: Area of each material(m$^2$)

The results showed that the storage parts e.g. the closets or the cupboards accounted for a large percentage of the total emission, which ranged from 25% (House C) to 49% (House A), although their areas were rather small.

Figure 6 shows the relationship between concentration measured and concentration calculated from emission rate for each part, area of each part and ventilation rate. The measured value was higher than the calculated value in summer while the measured value was lower in winter.

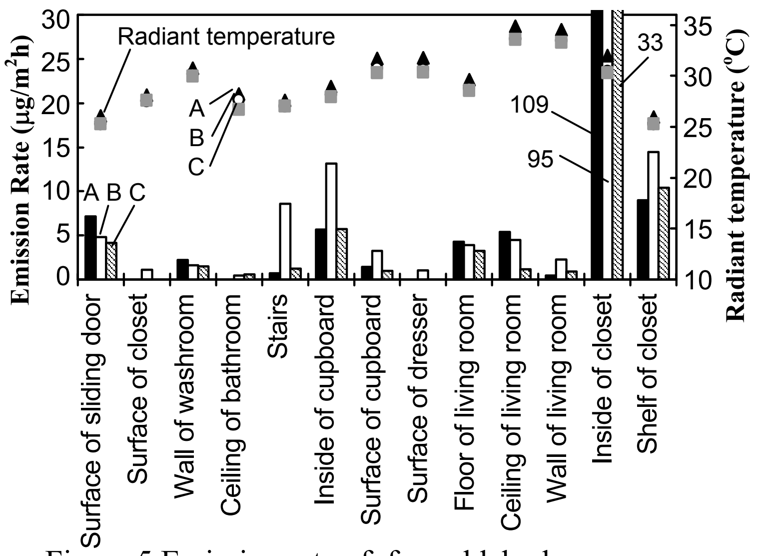


Figure 5 Emission rate of formaldehyde

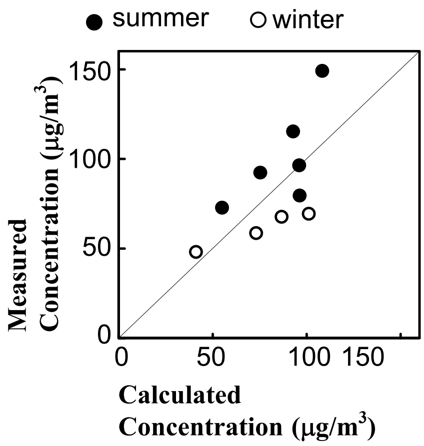


Figure 6 Calculated concentration of formaldehyde vs. measured concentration of formaldehyde

**Concentration of VOCs**

The guidelines published by the Ministry of Health, Labor and Welfare in Japan are 100μg/m³ for toluene, 870μg/m³ for xyrenes, 240μg/m³ for *p*-dichlorobenzene, 3800μg/m³ for ethyl benzene, 240μg/m³ for styrene, 330μg/m³ for tetradecane and 400μg/m³ (tentative value) for TVOC. Styrene could not be detected in any rooms. *p*-Dichlorobenzene which is used in moth crystals and room deodorizers could not be detected in 5 of the 7 rooms and detected values were low (0.9μg/m³) because sampled houses did not have occupants.

Figure 7 (1)-(5) shows the concentrations of the other compounds. All of them show higher concentration in summer. Toluene and TVOC show higher values than guidelines. Although alkanes, aromatic hydorocarbons, terpenes, halocarbons, esters, aldehydes and ketones (excl. formaldehyde) and others (toluene equivalent ) are illustrated separately in Figure 7 (5), the concentrations of halocarbons and esters are too low to indicate. Terpenes showed higher value than guidelines, because of high concentrations of α−pinene and limonene. α−Pinene, which is usually used in wax and wood products, showed high concentrations in Houses A,B,C and E. Comparing with VOCBASE (Jensen, B. and Wolkoff, P, 1996), α−pinene and nonanal showed higher concentration than the Odor detection threshold. For nonanal the Ministry of Health, Labor and Welfare suggests 41μg/m³ as a tentative guideline. In House D, nonanal showed a higher concentration than the guideline value in both the living room and the bedroom, which was greater than 2000μg/m³.

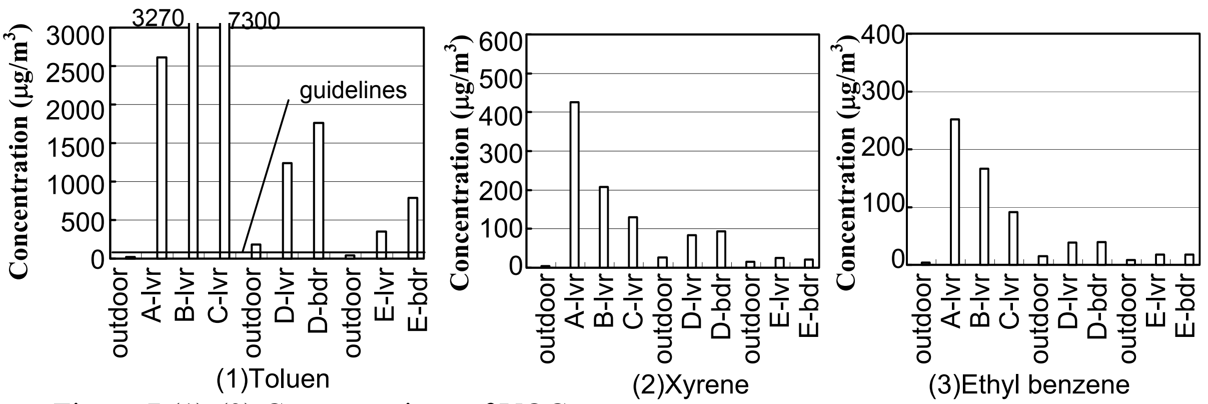


Figure 7 (1)-(3) Concentrations of VOCs (A-D:House A-D lvr: living room bdr: bedroom )

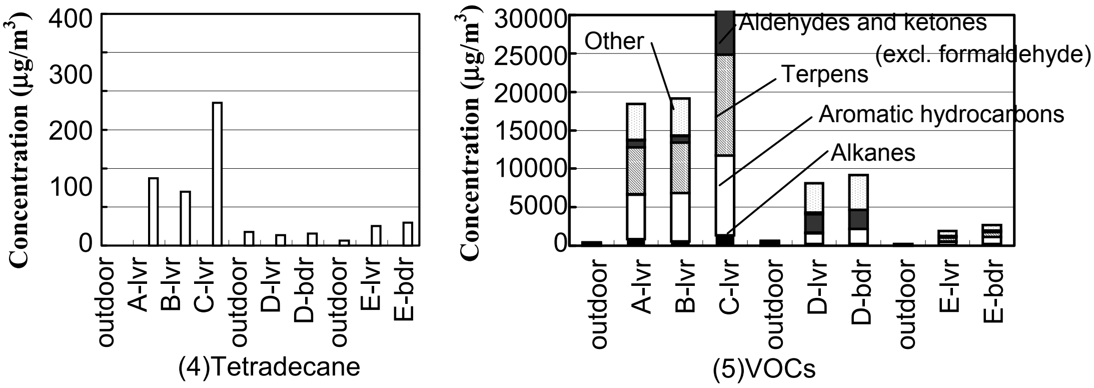


Figure 7 (4)-(5) Concentrations of VOCs (A-D:House A-D lvr: living room bdr: bedroom )

## DISCUSSION

Although the concentrations of formaldehyde measured in winter were rather low, they are higher than the guideline when they had been converted to the concentration under 25°C and 50%rh. The equation used for modification in this paper was different from the figures suggested by Mehlhorn (Seifert 1990) which illustrate the effects of temperature and humidity on formaldehyde concentration. The effects of temperature and humidity on formaldehyde concentration must be clearly identified.

The TEA-dish method proposed in this paper is helpful to find the main emission source of formaldehyde, although the relative emission rate calculated from this method may be different from the emission rate in actual conditions.

## CONCLUSIONS

Field investigation in newly-built houses were conducted and the followings were obtained.

1. The concentration of formaldehyde in all rooms was lower than the guideline value. After modification for temperature and humidity, concentrations in the three rooms measured in winter were higher than the guidelines
2. The inside surfaces and the shelves of the closets and the cupboards measured in summer showed higher emission rate of formaldehyde than the other surface.
3. Toluene and TVOC showed higher values than guidelines.


## ACKNOWLEDGEMENTS

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