Measurements of indoor concentrations of aldehydes, VOCs and fungi in newly built apartment houses in Tokyo

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ABSTRACT
The purpose of this study is to identify the causality explaining indoor concentrations of formaldehyde, VOCs and fungi which have significant health effects. In the rainy season (July), summer (August), autumn (October) and winter (December), measurements were carried out in three newly built apartment complexes. In each complex the same building materials were used. Temperature, relative humidity, concentrations of formaldehyde, VOCs and CFU (colony forming unit), were measured and questionnaire surveys were conducted. The concentrations of formaldehyde and VOCs were measured by passive samplers. Formaldehydes, acetaldehyde, benzene, toluene, ethylbenzene, *o*-, *m*-, *p*-xylene, *p*-dichlorobenzene, styrene and limonene were analysed.

The concentration of formaldehyde did not decrease with the elapsed time within 6 months. However, for toluene and limonene, which were mainly emitted from building materials, the emission rates decreased with the elapsed time. A high CFU was detected in some apartments and no negative correlation was found between the concentration of formaldehyde and airborne fungi and between the concentrations of VOCs and airborne fungi.

INDEX TERMS
Newly built apartment house; VOC; Formaldehyde; Fungi; Questionnaire

INTRODUCTION
Indoor air quality (IAQ) is an important determinant of population health and well-being but in spite of this the control of IAQ is difficult because of a wide variety of air pollutants, e.g. VOCs, formaldehyde and fungi (Mϕlhave, 2000). It has been reported that chemical compounds were emitted not only from building materials but also from furniture, electric devices, aromatic and insecticide (Wensing *et al.*, 2002). It has been also reported that the emission rate of VOCs decreased with the age of the house while fungi concentration increased with the age.

In this study, the concentrations of formaldehyde, VOCs and airborne fungi were measured in newly built apartment houses. Since the same building materials are used in each complex, it is expected that the effect of building materials on the concentrations of formaldehyde and VOCs can be eliminated. The questionnaire survey was also conducted in order to examine the effect of lifestyle on concentrations of formaldehyde, VOCs and airborne fungi.

METHODS
Measurements were carried out in three newly built apartment complexes (Complex A, four houses; B, two houses; C, one house) in the rainy season (July), summer (August), autumn (October) and winter (December), 2002. Table 1 shows the outline of the houses. Formaldehyde, VOCs, suspended fungi, temperature and relative humidity were measured as shown in Table 2. Formaldehyde and VOCs were sampled at 1.2 m above the floor by DSD-DNPH and VOC-SD for 24 h. Both passive samplers were hung at the centre of the rooms. Formaldehyde was

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analysed by HPLC. VOCs were analysed by gas chromatography. Suspended fungi were sampled at 1.2 m above the floor by centrifugal samplers, RCS standard (40 l/min) and RCS high-flow (100 l/min) and a sieve sampler, Bio Samp (100 l/min). The samples were put in an incubator at 25°C for 96 h and then CFU was counted. Temperature and relative humidity were measured continuously for a week. Questionnaire survey asking their lifestyle was conducted.

Table 1 Outline of houses

House	Year & month of conpletion	Living Room		Bedroom		Bathroom		Floor Material			Orientation of the Room			Floor	Number of occupant	Pet	When moving into
		A	H	A	H	A	H	Living Room	Bed room	Bath room	Living Room	Bed room	Bath room				
A-1	2002.3.	28.0	2.4	2.4	2.4	3.0	2.0	w	w	ti	S	N	NW	4	2	no	2002.4.
A-2		28.0	2.4	2.4	2.4	3.0	2.0	w	w	ti	S	N	NW	1	2	no	2002.3.
A-3		23.0	2.5	2.5	2.5	2.0	1.9	w	w	b	SW	N	S	1	1	no	2002.6.
A-4		22.0	2.5	2.5	2.5	3.0	2.0	w	w	b	SE	NE	SE	1	4	no	2002.3.
B-1	2002.5.	21.0	2.2	2.2	2.2	2.0	2.1	w+t	w	ti	S	NW	N	1	2	no	2002.6.
B-2		23.0	2.2	2.1	2.1	3.0	2.2	w	t	ti	S	SE	N	2	3	Cat	2002.6.
C-1	2002.3.	8.0	2.4			1.0	2.0	w		ti	S		E	3	1	no	2002.4.

A: area (m^2), H: height (m), w: wooden board, t: Japanese Tatami Mat, ti: tile, b: synthetic resin board,

Table 2 Place and methods

	Formaldehyde*1	VOCs	Suspended Fungi								Temp. & R.H.
	DSD-DNPH	VOC-SD	RCS standard			RCS high-flow			Bio Samp		
			YM*2	PDA*3	DG-18*4	YM*2	PDA*3	DG-18*4	PDA	DG-18	
Living Room	>	>	>	>	>	>	>	>	>	>	>
Bedroom	>	>	>	>	>	>	>	>	>	>	>
Bathroom	-	-	>	>	>	>	>	>	>	>	>
Outside*5	>	>	-	-	-	-	-	-	>	>	>

*1: Aug.–, *2: –Aug., *3: Oct.–, *4: Only Oct., *5: Aug.–
YM: Yeast and molt agar (use of yeast and fungi), PDA: potato dextrose agar (use of anaerobic fungi), DG-18: dichloran glycel 18% agar (use of aerobic fungi).

RESULTS AND DISCUSSION

Formaldehyde

Temperature and relative humidity influence formaldehyde emission rate. The concentration of formaldehyde was converted to that under 25°C and 50% RH by Inoue's equation (Inoue, 1997) as Eqn (1). Figure 1 shows the concentration of formaldehyde under 25°C and 50% RH. All values were lower than the guidelines in Japan (the Ministry of Health, Labour and Welfare in Japan, 100 μg/m^3). The concentration of formaldehyde did not decrease with the elapsed time within 6 months:

$$C_0 = C \frac{1.09^{(t_0 - t)}}{1 + 0.01(H - H_0)} \qquad (1)$$

where C is the measured concentration, C_0 the standard concentration (under t_0 °C, H_0 %RH), t the measured temperature, t_0 the standard temperature (=25°C), H the measured relative humidity and H_0 the standard relative humidity (=50% RH).

To identify the sources of formaldehyde, analysis of variance was used. Table 3 shows the ANOVA table. Besides seasonal difference, a significant difference was found between Complex A and Complex B, which implied the effects of building materials on formaldehyde concentration. No correlation was found between the concentrations of formaldehyde and amount of cigarette smoked, the frequency of opening windows or the frequency of using aromatic and insecticide.

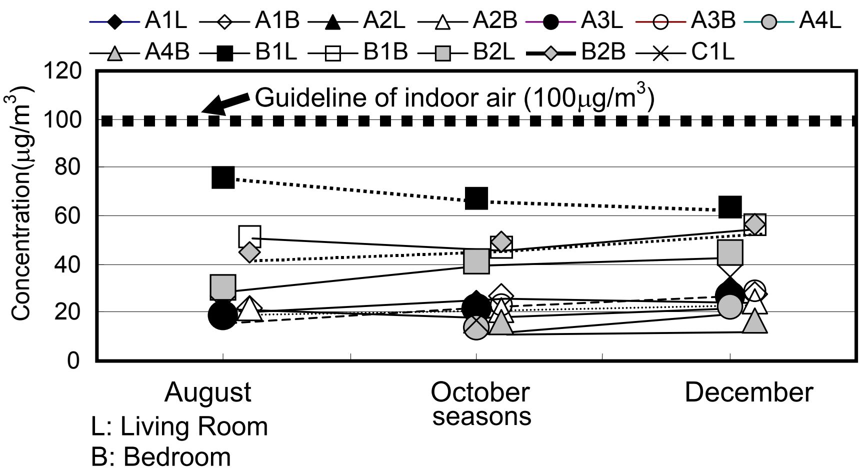

Figure 1 Modified concentration of formaldehyde.

Table 3 ANOVA table of formaldehyde

		SS	df	MS	F	p
Aug.vs Oct. vs Dec.	between	4791.0	2	2395.5	6.2	**
	within	12752.1	33	386.4		
	total	17543.1	35			
Complex A vs Complex B	between	8477.5	1	8477.5	30.2	**
	within	8695.3	31	280.5		
	total	17172.8	32			
A1 vs A2 vs A3 vs A4	between	64.1	3	21.4	0.7	
	within	485.4	17	28.6		
	total	549.5	20			
B1 vs B2	between	584.7	1	584.7	0.8	
	within	7561.1	10	756.1		
	total	8145.8	11			

**:$p < 0.01$.

VOCs

Table 4 show the concentrations of VOCs and the ratio of VOCs concentration to the guidelines (%). The concentrations of ethylbenzene, xylenes and styrene were lower than the guidelines while the concentrations of benzene and limonene were higher than the guidelines in some houses (benzene: houses A-3, B-1, B-2 and C-1; limonene: houses A-1, A-3 and B-1).

To identify the sources of VOCs, analysis of variance was used. Table 5 shows the results. For toluene and limonene, significant differences were found between seasons and between Complex A and Complex B. These results imply that toluene and limonene may be mainly emitted from building materials and their emission rates decreased with the elapsed time. The difference in the concentration of *p*-dichlorobenzene in the houses in the same complexion implied that the concentration of *p*-dichlorobenzene was affected not by building materials but by lifestyle.

Suspended Fungi

Figure 2 shows the relationship between temperature, relative humidity and concentration of airborne fungi. A high CFU of airborne fungi was shown under 26–30°C of air temperature and more than 60% of relative humidity.

Table 6 shows the correlation coefficient of fungi between the living rooms, the bathrooms, the bedrooms and the outside. A strong correlation was found between the living room and the bedroom (PDA: $R = 0.82$, DG-18: $R = 0.92$) and between the bedroom and the bathroom

(PDA: R = 0.73, DG-18: R = 0.78), although no strong correlation was found between the rooms and outside (PDA: R = 0.13–0.40, DG-18: R = 0.27–0.51). Figure 3 shows CFU of airborne fungi (Bio Samp, DG-18, in living room) and the guidelines. By using the regression equations by Lee *et al.* (2002) and by Iwata *et al.* (2003), the EC Concerted Action 613 guideline (Maroni *et al.*, 1995) was converted into the value shown in Figure 4. The concentrations of fungi were ‘Low’ or ‘Very Low’ except that in House B-1 in July and August.

Table 4 The ratio of VOCs concentration to the guidelines

	Guideline (μg/m³)*	Mon.	Max. conc.#1	Max. ratio#2	Min. conc.#1	Min. ratio#2	Ave. conc.#1	Ave. ratio#2
Benzene**	3	July	9	300	N.D.	N.D.	4	121
		Aug.	13	430	N.D.	N.D.	5	178
		Oct.	24	800	N.D.	N.D.	7	237
		Dec.	12	400	N.D.	N.D.	7	237
Tolene	260	July	257	99	26	10	142	55
		Aug.	260	100	17	7	81	31
		Oct.	422	162	29	11	111	43
		Dec.	168	65	25	10	85	33
Ethyl benzene	3800	July	53	1	N.D.	N.D.	26	1
		Aug.	38	1	N.D.	N.D.	11	N.D.
		Oct.	35	1	10	N.D.	13	N.D.
		Dec.	30	1	N.D.	N.D.	13	N.D.
Xylenes	870	July	83	10	N.D.	N.D.	40	5
		Aug.	76	9	N.D.	N.D.	16	2
		Oct.	63	7	N.D.	N.D.	14	2
		Dec.	43	5	N.D.	N.D.	8	1

	Guideline (μg/m³)*	Mon.	Max. conc.#1	Max. ratio#2	Min. conc.#1	Min. ratio#2	Ave. conc.#1	Ave. ratio#2
Stylene	220	July	45	20	N.D.	N.D.	15	7
		Aug.	38	17	N.D.	N.D.	4	2
		Oct.	45	20	N.D.	N.D.	6	3
		Dec.	34	15	N.D.	N.D.	5	2
p-Dichloro benzene	240	July	2279	950	N.D.	N.D.	471	196
		Aug.	397	165	N.D.	N.D.	108	45
		Oct.	1762	734	35	15	238	99
		Dec.	640	267	N.D.	N.D.	148	62
Limonene***	30	July	312	1004	N.D.	N.D.	94	313
		Aug.	135	450	N.D.	N.D.	28	93
		Oct.	428	1433	N.D.	N.D.	93	310
		Dec.	161	536	N.D.	N.D.	48	157

#1: μg/m³
#2: the ratio of VOCs concentration to the guideline(%)

*: The guidelines of indoor air (Ministry of Health, Labor and Welfare in Japan).
**: Environmental quality guidelines for air pollution value (Ministry of Environment in Japan).
***: The guidelines of indoor air in terpene (WHO).

Table 5 ANOVA table of VOCs

		Benzene SS	df	MS	F	p	Toluene SS	df	MS	F	p
Aug.vs Oct. vs Dec.	between	67.6	2	33.8	0.8		48855.7	2	24427.9	3.8	*
	within	1360.0	33	41.2			212981.0	33	6454.0		
	total	1427.6	35				261836.7	35			
Complex A vs Complex B	between	103.5	1	103.5	2.7		39083.7	1	39083.7	6.1	*
	within	1318.3	35	37.7			225193.0	35	6434.1		
	total	1421.8	36				264276.7	36			
A1 vs A2 vs A3 vs A4	between	170.5	3	56.8	1.3		40589.4	3	13529.8	1.4	
	within	811.8	19	42.7			168555.6	17	9915.0		
	total	982.3	22				209145.0	20			
B1 vs B2	between	0.8	1	0.8	0.0		2909.8	1	2909.8	7.6	*
	within	335.2	12	27.9			3839.5	10	384.0		
	total	336.0	13				6749.3	11			

		p-dichlorobenzene SS	df	MS	F	p	Limonene SS	df	MS	F	p
Aug.vs Oct. vs Dec.	between	215984.5	2	107992.3	0.7		55782.0	2	27891.0	3.5	*
	within	4364126.5	30	145470.9			235900.7	30	7863.4		
	total	4580111.0	32				291682.7	32			
Complex A vs Complex B	between	342243.4	1	342243.4	1.5		55430.4	1	55430.4	7.3	*
	within	8242758.6	35	235507.4			236252.3	31	7621.0		
	total	8585002.0	36				291682.7	32			
A1 vs A2 vs A3 vs A4	between	2250401.6	3	750133.9	6.1	**	43829.9	3	14610.0	1.3	
	within	2074812.9	17	122047.8			189055.6	17	11120.9		
	total	4325214.5	20				232885.6	20			
B1 vs B2	between	22124.7	1	22124.7	1.9		1683.0	1	1683.0	10.0	*
	within	117208.8	10	11720.9			1683.7	10	168.4		
	total	139333.5	11				3366.7	11			

**: $p < 0.01$; *: $0.01 < p < 0.05$.

No strong correlation was found between CFU of airborne fungi and the frequency of opening the windows and between CFU and the frequency of cleaning. In house B-1, where washing was often hung out to dry, higher humidity was shown and concentration of fungi was high.

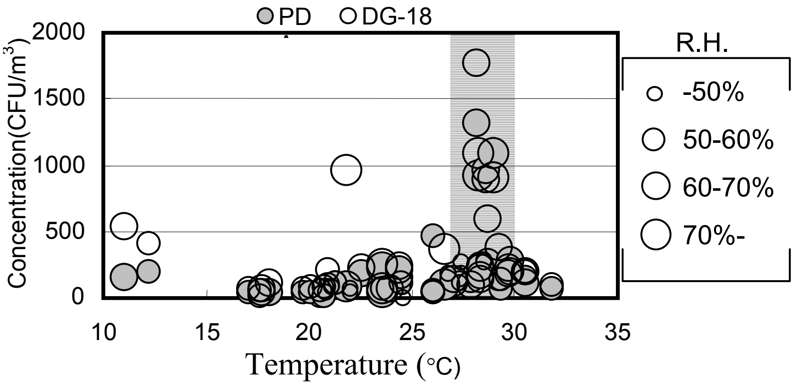

Figure 2 Effects of temperature and relative humidity on CFU.

Table 6 Correlation coefficient of fungi between the places

	L.R.	Be.	Ba.	Out.
L.R.		**0.82**	**0.67**	**0.40**
Be.	**0.92**		**0.73**	**0.13**
Ba.	**0.73**	**0.78**		**0.27**
Out.	**0.51**	**0.27**	**0.35**	

L. R.: Living Room
Be.: Bedroom
Ba.: Bathroom
Out.: Outside
: Results of PDA
: Results of DG-18

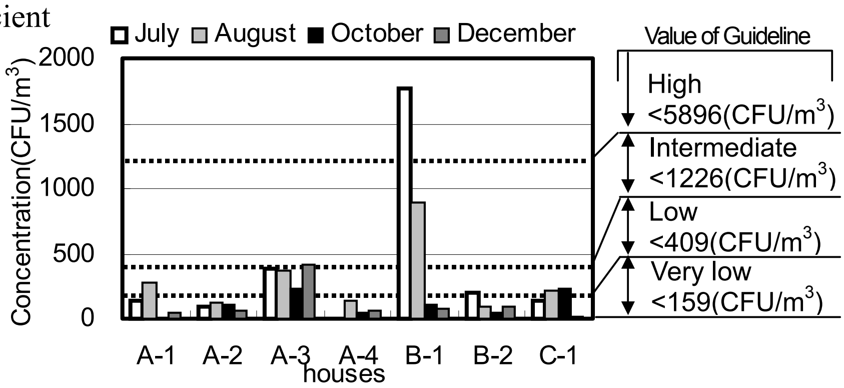

Figure 3 Concentration of viable fungi (living room, DG-18).

In order to identify the cause of airborne fungi, Quantification theory type 1 was used. CFU is used as the criterion variable. Table 7 shows the items and categories used in the analysis and the effect of each item on CFU of airborne fungi. Figure 4 shows the results of the analysis. The correlation coefficient of 0.70 was found. The categories, which significantly increased CFU of airborne fungi, were b-2, c-1 and e-3 while the most effective category decreasing CFU was c-4. The most effective item was the frequency of cleaning.

Table 7 Used items and categories

Items		Categories		Effects: Temp	R.H.	Nourishment	Outdoor air
a	Proportion of the 25C> 30C temp	1	more than 70%	>			
		2	less than 70%				
b	Proportion of the 65%~ R.H.	1	more than 70%		>		
		2	less than 70%				
c	Direction of rooms, floor of rooms	1	North, 1st floor	>	>		>
		2	South, 1st floor				
		3	North, More than 2nd floor				
		4	South, More than 2nd floor				
d	Floor materials	1	Japanese tatami mat			>	
		2	Wooden board				
e	Frequency of cleaning	1	more than 3 times/week			>	
		2	1~2 times/week				
		3	less than 1 time/week				
f	Total occupied time/day	1	less than 10 hours		>	>	
		2	10~20 hours				
		3	more than 20 hours				
g	Ventilation times/day	1	more than 5 hours		>		>
		2	less than 5 hours				

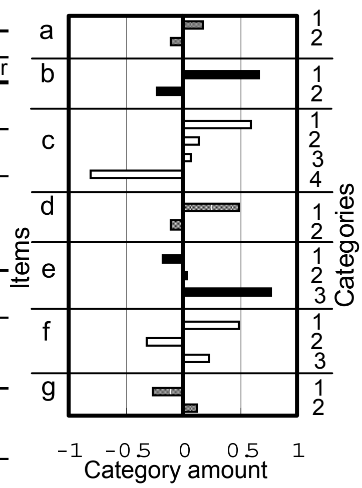

Figure 4. Results of analysis

Correlation between Formaldehyde and Airborne Fungi and between VOCs and Airborne Fungi

Table 8 shows the correlation between the concentrations of formaldehyde, VOCs and airborne fungi. The correlation coefficient of 0.50–0.66 was found between the concentrations of formaldehyde, ethylbenzene and xylene and the concentration of airborne fungi. No negative correlation was found. Although it is said that formaldehyde has a suppressive effect on growth of fungi, neither formaldehyde nor VOCs in those houses could reduce airborne fungi because of their lower concentrations.

Table 8 Correlation of formaldehyde, VOCs and fungi

Season	Agar	Formaldehyde	Benzene	Tolene	Ethylbenzene	Xylenes	Stylene	*p*-Dichlorobenzene	Limonene
July	PDA		0.14	0.44	0.33	0.56	0.43	0.13	0.20
	DG-18		0.10	0.25	0.50	0.62	0.23	0.07	0.02
Aug.	PDA	0.65	0.23	0.04	0.46	0.54	0.21	0.13	0.31
	DG-18	0.64	0.22	0.01	0.55	0.59	0.13	0.20	0.37
Oct.	PDA	0.24	0.22	0.04	0.02	0.23	0.01	0.03	0.10
	DG-18	0.17	0.03	0.07	0.03	0.16	0.12	0.07	0.28
Dec.	PDA	0.66	0.31	0.40	0.48	0.36	0.42	0.45	0.49
	DG-18	0.20	0.17	0.03	0.14	0.09	0.06	0.08	0.21

CONCLUSIONS

Measurements in newly built apartment houses were conducted and the following conclusions were obtained:

1. The converted concentration of formaldehyde showing the concentration under 25°C and 50% RH were lower than the guidelines. However, the concentration of formaldehyde did not decrease with the elapsed time within 6 months.
2. No correlation was found between the concentrations of formaldehyde and amount of cigarette smoked, the frequency of opening windows or the frequency of using aromatic and insecticide.
3. Toluene and limonene may be mainly emitted from building materials and their emission rates decreased with the elapsed time.
4. Relative humidity and the frequency of cleaning affected CFU of airborne fungi.
5. No negative correlation was found between the concentrations of formaldehyde and airborne fungi and between VOCs and airborne fungi.

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