A survey of indoor air chemical contaminants in newly built detached houses

Takekuma M[1], Hayashi M[2], Kim H[2], Osawa H[2]

[1]Saitama Prefectural Institute of Public Health, Saitama, Japan, 355-0133, takekuma.mikiko@pref.saitama.lg.jp; [2]National Institute of Public Health, Saitama, Japan, 351-0197

Introduction

We spend most of the day indoors and usually spend at home. The average daily home time of Japanese people is 15.8 hours a day[1]. Since the average respiratory rate of Japanese men and women is 17.3 m^3/day[1], it means that we have been inhaling 11.4 m^3/day at home. Exposure to chemicals through the respiratory tract is generally said that the absorption efficiency is higher because the chemicals flow directly from the alveoli to the bloodstream and circulate throughout the body. Indoor air should be kept as clean as possible. On the other hand, in Japan, the energy conservation standards for residential buildings have been revised in 2013, and highly insulated and airtight houses are spreading. The component and concentration of indoor air chemical contaminants differ from each house due to constructional materials, lifestyle, airtightness, ventilation volume and other things. Moreover, many chemicals in the air are unintentionally present. Filed survey is required in order to evaluate the exposure to each chemicals. We examined indoor air chemical contaminants in detail in newly built detached houses based on Japanese next generation energy conservation standards.

Materials and methods

Surveyed houses: A newly wooden house was built from October 2014 to December 2016. 10 housing suppliers (A-J companies). 19 non-occupancy houses (A1, A2, A3, A4, B1, B2, D1, D2, E1, E2, E3, E4, F1, F2, C, G, H, I, J).

Sample collection: Three places on each house: the living room and the bedroom on the first and second floor, and the outdoor. Air samples for A1, B1 and C were taken twice at each places for seasonal observation. Other air samples were collected once from the completion date to 7 months after completion. Air samples were collected at 44 places indoors and 22 places outdoors. The whole indoor spaces were ventilated for 30 minutes from 8:00 am to 9:00 am, and air samples were collected at each places for 10 or 30 minutes from 2:00 pm to 3:00 pm. The air sampling pumps were used SP208-20Dual, SP208-100Dual and SP208-1000Dual (GL Sciences Inc.). Sampling volumes were 30L (at 1.0L/min) for carbonyl compounds with InertSep mini AERO DNPH cartridge (GL Sciences Inc.) and 3L (at 100mL/min) for volatile organic compounds (VOCs) with Tenax TA sorbent in inert stainless tube (Camsco Inc.). Double sampling was done for VOCs and two tubes were deployed tandem, and additionally one more tube collected air volume of 0.1L (at 10mL/min) in order to check breakthrough.

Airtightness performance: An airtightness performance test was carried out according to JIS A 2201 (Japanese Industrial Standards, 2017) after collecting air samples. The airtightness, "c-value" (cm^2/m^2) was measured after sealing up opened spaces such as the kitchen hood, sewage inlet etc.

Sample analysis: 12 carbonyl compounds were analyzed by DNPH sorbent tube / solvent extraction / HPLC (LC-2000, Jasco Inc.) using Carb Method 1004 DNPH Mix 2 (Sigma-Aldrich Inc.) as the standard. VOCs were analyzed by sorbent tube / thermal desorption (Turbo Matrix 650, Perkin Elmer Inc.) -GC-MS (7890B/5977A, Agilent Technologies Inc.) using Toluene-d_8 (Takachiho Chemical Industrial Inc.) as the internal standard gas. 49 VOCs were quantified using 50 Component Indoor Air Standard, (+)-3-Carene (Sigma-Aldrich Inc.) and γ-Terpinene (Wako Pure Chemical Industries Inc.) as the standard. Other VOCs were searched by the chromatogram deconvolution function and the library of NIST 11 with MassHunter Qualitative Analysis B.06.00 (Agilent Technologies Inc.). The top-five detected compounds for each house were quantified in terms of toluene.

Statistical analysis: A Multiple regression analysis was performed on the four factors, 1) temperature, 2) relative humidity, 3) airtightness (c-value), 4) the number of days after completion that seem to influence chemical concentrations using SPSS Statistics 19.0 (IBM Inc.). The statistical test for a significant difference was set at 5 %.

Results

Seven aldehydes and acetone in carbonyl compounds were detected from indoor air. Acetaldehyde was detected at 10 of the 44 indoor samples exceeding the indoor air guideline value in Japan (excess rate 23 %). 54 chemical compounds in VOCs were identified and quantitated, and 50 chemical compounds were detected from indoor air. No chemical compounds exceeding the current indoor air guideline values were founds, but ethylbenzene was detected exceeding the guideline values will be revised downwards (excess rate 6.8 %). Moreover dichloromethane was detected exceeding the environmental quality standards in Japan (excess rate 32 %). Because breakthrough was observed with dichloromethane and *α*-pinene detected at high concentrations in some houses, their quantifications were performed with a sampling volume of 0.1 L. Concentrations of indoor and outdoor air chemical compounds, composition rates and sampling condition in newly built detached houses are shown in Table 1. In the A1 and B1 housing completed in autumn, the concentrations of indoor air chemical compounds in the summer that will be welcomed for the first time than in winter increased with all carbonyl compounds, terpenes such as α-pinene and nonanal, and the concentrations increased by two to six times compared with the total VOCs. On the other hand, in the C housing completed in summer, the concentrations in autumn trended to be lower than immediately after completion. It decreased to about one third of total VOCs. The concentrations of total VOCs, the airtightness and the ventilation system in each house are shown in Fig. 1. The results of a main multivariate linear regression analysis (coefficient of non-standard, standard error and significant probability) are shown in Table 2. The concentrations of acetaldehyde significantly increased with temperature, relative humidity and airtightness. The temperature showed the strongest influence of the concentrations when compared to the standardization partial regression coefficients of each factors. The concentrations of terpens significantly increased when the airtightness was high that was the c-value was small. The concentrations of VOCs that were the top five compounds in terms toluene significantly increased when the temperature and the airtightness were high, and significantly decreased with the passing of the days after completion.

Discussion

Acetaldehyde, dichloromethane and *α*-pinene that is easily oxidized and changed strongly irritating aldehydes were detected at relatively high concentrations. It seems to be necessary that observe their movements in the future. However, it seems to be difficult to control acetaldehyde because the source varies from antiseptics, adhesives, paints, wood, smoking, exhalation etc. On the other hand, dichloromethane that observed at high concentrations in some house has not indoor air guideline values in Japan. It is required attention to the sampling and the measurement. Because it is not included in the definition of TVOC (total volatile organic compounds, the provisional target value of Japan; 400 $\mu g/m^3$) also, it was considered preferable to be treated as a newly observed compound indoor. Moreover the composition rates of terpenes derived from natural wood tended to be high in the indoor air of most houses, but the composition ratio of dichloromethane was higher than that of terpenes in some houses where dichloromethane was used. It was considered necessary that take some action. From the investigation of seasonal variation, it was considered necessary that call the attention to indoor air pollution for one year after completion particularly. The concentration of chemical compounds in indoor air is very high compared with outdoor air, and it tends to increase as the airtightness becomes higher. Housing suppliers should pay close attention to the use of chemical compounds in the selection of building materials and the construction process, and it is desirable to provide adequate ventilation before handover. Residents must understand the ventilation system and continue ventilation during their living.

Table 1. Concentrations of indoor and outdoor air chemical compounds, composition rates and sampling condition in newly built detached houses (μg/m^3) [a].

Main compounds	Indoor air guideline values	Environmental quality standards	Indoors (the inside air)					Outdoors (the outside air)				
			Mean	Median	Min	Max	SD	Mean	Median	Min	Max	SD
Formaldehyde	100		20	15	4.4	75	16	4.5	4.0	1.6	10	2.3
Acetaldehyde	48		34	24	5.7	147	28	3.2	2.4	<0.4	14	3.7
Acetone			97	87	17	287	61	5.9	5.8	1.3	17	3.3
Dichloromethane		150	485	1.8	<0.3	5,878	1,249	6.9	0.20	<0.3	58	16
2-Butanone			30	13	3.9	216	43	3.6	2.9	<0.3	16	3.4
Ethylacetate			26	16	4.0	207	41	8.1	5.8	0.81	34	7.3
Toluene	260		31	27	1.4	109	23	9.6	7.2	0.98	29	7.9
Ethylbenzene	3,800 → 58		15	5.2	0.39	72	20	2.8	1.7	<0.3	21	4.5
m,p-Xylene	870 → 200		8.2	2.9	<0.3	42	10	1.7	0.80	<0.3	13	2.8
o-Xylene			20	7.7	0.72	176	31	0.48	<0.3	<0.3	4.6	1.1
Styrene	220		5.8	2.6	<0.3	59	10	0.42	<0.3	<0.3	3.2	0.76
α-Pinene			1,262	751	66	5,934	1,412	3.8	1.3	<0.3	20	5.2
3-Carene			152	106	16	402	114	0.20	<0.3	<0.3	2.6	0.57
Limonene			133	106	7.5	409	106	0.27	<0.3	<0.3	1.6	0.46
Dodecane			34	2.0	0.45	396	98	0.41	<0.3	<0.3	2.2	0.63
Main compounds in terms of toluene			Mean	Median	Min	Max	Number					
α-Pyronene			52	40	2.9	200	32	<0.3	<0.3	<0.3	<0.3	<0.3
α-Copaene			50	42	4.3	113	14	<0.3	<0.3	<0.3	<0.3	<0.3
α-Muurolene			55	29	1.5	155	30	<0.3	<0.3	<0.3	<0.3	<0.3
γ-Muurolene			92	46	9.7	318	30	<0.3	<0.3	<0.3	<0.3	<0.3
trans-Calamenene			56	37	2.1	157	14	<0.3	<0.3	<0.3	<0.3	<0.3
			Mean	Median	Min	Max	SD	Mean	Median	Min	Max	SD
Aldehydes (7 compounds)			98	73	19	256	69	12	7.5	1.6	68	15
VOCs (46 compounds)			2,426	1,678	146	8,509	2,112	57	50	8.2	191	43
VOCs (the top-five compounds in terms of toluene)			234	143	13	755	224	-	-	-	-	-
Total VOCs (58 compounds)			2,757	1,850	217	8,801	2,184	69	69	11	208	47
Terpens (5-10 compounds)			1,794	1,270	135	6,829	1,636	4.3	1.4	<0.3	22	5.9
The rate of Aldehydes per total VOCs (%)			4.8	4.1	0.62	14	3.4	19	13	3.3	60	15
The rate of Terpens per total VOCs (%)			64	72	12	95	23	7.4	2.4	0	26	9.4
The rate of Dichloromethane per total VOCs (%)			14	0.055	0	70	21	6.9	0.43	0	70	16
Temperature (℃)			21	22	10	35	6.0	22	20	9.4	39	8.6
Relative humidity (%)			49	51	19	75	12	41	37	13	98	23
Airtightness; c-value (cm^2/m^2)			1.1	0.8	0.1	3.0	0.8	-	-	-	-	-
The number of days after completion (days)			147	60	0	1,510	317	147	60	0	1,510	317

[a] Indoors: 44 places in the living room and the bedroom on the first floor and second floor. Outdoors: 22 places. The total number of houses: 22 non-occupancy houses.

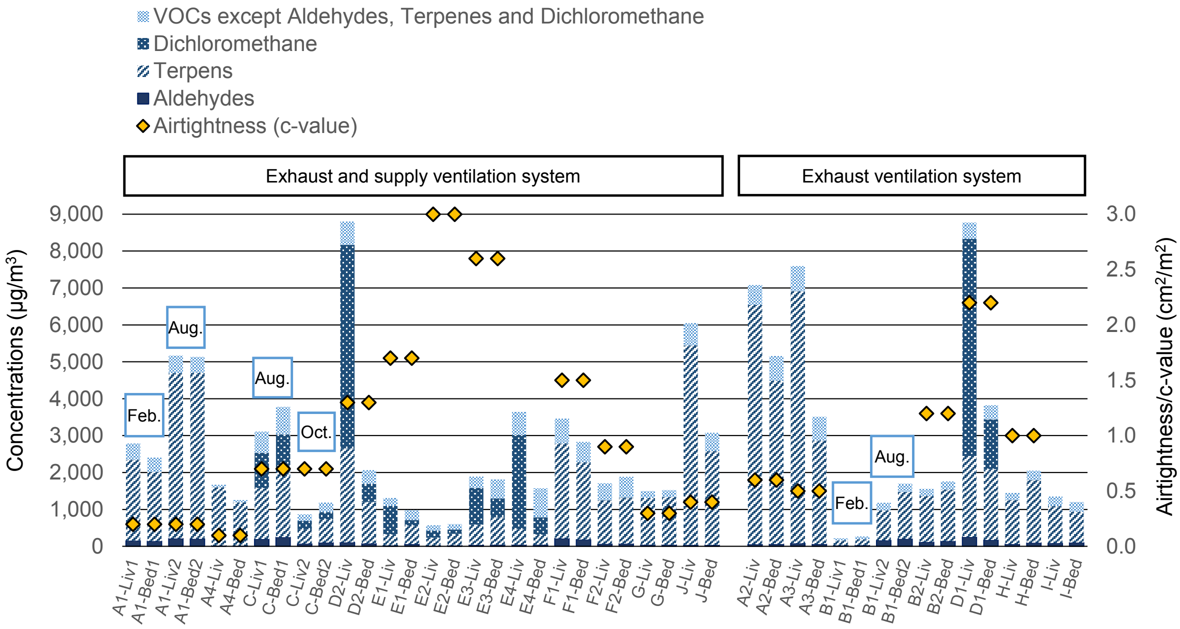

Figure 1. Concentrations of indoor air chemical compounds and airtightness (c-value) in newly built detached houses by ventilation system.

Table 2. Considering influence of various factors on concentrations of indoor air chemical compounds, and the result of multivariate linear regression analysis (n=44) [a].

Main compounds	Temperature			Relative humidity			Airtightness (c-value)		
	RC	SE	*p*	RC	SE	*p*	RC	SE	*p*
Formaldehyde	1.754	0.259	0.000	0.116	0.145	0.431	0.808	1.780	0.653
Acetaldehyde	3.593	0.563	0.000	1.278	0.315	0.000	9.402	3.865	0.021
α-Pinene	85.770	42.684	0.053	37.042	23.924	0.132	949.354	293.189	0.003
3-Carene	12.386	2.876	0.000	1.618	1.612	0.323	1.830	19.756	0.927
Limonene	3.830	2.927	0.200	1.846	1.641	0.269	81.742	20.106	0.000
Acetone	4.921	1.717	0.007	0.182	0.963	0.851	20.770	11.796	0.088
Nonanal	0.559	0.141	0.000	0.163	0.079	0.047	0.485	0.966	0.620
Aldehydes (7 compounds)	8.913	1.324	0.000	2.473	0.742	0.002	8.431	9.091	0.361
Terpens (5-10 compounds)	69.638	48.064	0.157	Δ35.762	26.939	0.194	Δ1,188.013	330.141	0.001
VOCs (the top five compounds in terms toluene)	11.702	5.717	0.049	4.393	3.204	0.180	145.056	39.266	0.001

[a] RC: Regression coefficient, SE: Standard error, Δ: Negative.

Acknowledgements
This work was supported by MEXT KAKENHI Grant Number [16H02380] and Saitama prefectural industry-academia-government cooperation project.

References

1. National Institute of Advanced Industrial Science and Technology of Japan, Chemical Risk Management Research Center. (2007); *Exposure Coefficient Handbook.*