SVOC Concentrations in House dust and Residential Environment in Japanese Houses

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Abstract. This study aims to investigate the relevance of between SVOC concentrations in house dust and living environments. Using the newly proposed dust collecting and SVOC analysis method, we determined concentrations of the nine SVOC substances by dust size and made simultaneously the questionnaire about residential environments. Collected dusts were screened with sieves and then analyzed on nine components (DEP, DMP, BBP, DEHP, DINP, DBP, DNOP, DIDP, DIBP) by LC/MS/MS method. The results leaded to selecting less than 100 μ m and 100-250 μ m as the target size of dusts to be investigated intensively by considering the concentration distribution and the oral intake of humans. DEHP and DINP accounted for 96 to 97% of determined amount of SVOC and only the small amounts in the other seven components were detected. There was no significant difference in the component ratio depending on particle size. Significant difference was obtained between a few substances of SVOC and several living environment items.

1 Introduction

Several phthalate esters are added as plasticizers to increase flexibility and moldability of plastic products, and classified as SVOC (semi-volatile organic compounds) with a high boiling point. These are widely used in our life from building materials to toys, children's goods, daily necessaries and cosmetics. It is said that many SVOC substances are hardly present in the air but rather on the surface of the objects and dust etc. since it has low vapor pressure and strong adsorptivity.

There is also a possibility of endocrine disruption and relevance to asthma and allergosis of children [1]. Furthermore, an increase in the usage of products containing SVOC ingredients and long-term contamination due to their persistent property are reinforcing concerns about health effects [2] [3].

This study aims to investigate the actual condition of SVOC concentration in house dust and to ascertain the relevance of between them and living environments.

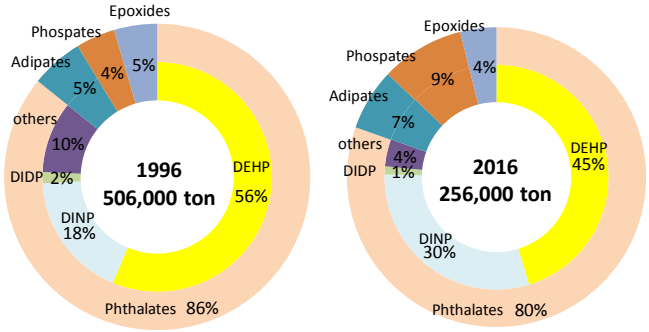

Fig 1. Production amount and composition ratio of plasticizer in Japan [4].

As a preliminary test, dusts were collected in ten houses and a detailed chemical analysis was conducted for four particle sizes. Based on the results, two particle sizes were selected to be examined intensively and then the determination of SVOC concentrations was conducted for the dusts collected in approximately 60 houses. Simultaneously we made the questionnaires about residential environments.

2 Methods

2.1 Questionnaire survey

The questionnaire concerning living environments included family characteristics and health conditions, house outline, indoor and surrounding environments, and details of daily life etc. Bivariate analysis was performed with the significance level 5% for correlation between SVOC concentrations and living environments.

2.2 Dust collection and size selection

There has been no unified method to collect house dust, and a few collecszting methods have been proposed. In this study, we conducted a background and blank test of three types of filters and selected the PET - nonwoven fabric filter as considering the applicability in the field.

In practice the dust accumulated in the vacuum cleaner as a backup specimen as well as filter collection in the field was collected since sufficient dust quantity

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should be ensured to classify dust sizes and to determine the chemical concentrations. For the preliminary study, ten houses provided their house dust and the dusts were sieved into the four particle sizes of less than 100μm, 100-250μm, 250-500μm and more than 500μm. For one dust specimen, from n=5 to 7 samples were prepared by each particle size, and the concentration distribution and deviation were investigated. The preliminary test resulted in selecting the two dust size of less than 100μm and 100-250μm and then dusts of 63 houses were analyzed for these two particle sizes. The outlines of the houses (in Fig. 2) showed the data of 71 houses that gave the cooperation for our research, and there were 59 (in less than 100μm) and 63 (in 100-250μm) dust samples respectively available for chemical analysis in the final analysis.

2.3 Chemical analysis

It has been difficult to quantify DINP and DIDP by using usual GC-MS method since the method spreads broadly saw-like low peaks on chromatograph for the both sub stances. We examined newly the method of solvent extraction-LC-MS/MS and could quantify stably the nine SVOC substances (in Fig. 3) -DEP(Diethyl phthalate), DMP(Dimethyl phthalate), BBP(Butyl Benzyl Phthalate), DEHP(Di(2-ethylhexyl)phthalate), DINP(Diisononyl phthalate), DBP(Dibutyl phthalate), DNOP(Di-n-octyl phthalate), DIDP(Diisodecyl phthalate), DIBP(Diisobutyl Phthalate)-.

3 Result

3.1 Residential environments

Most of the houses are in urban area, in detail 45 houses were in Kanto region (near Tokyo), 15 ones in Tohoku, 6 ones in Kyushu and 5 ones in Hokkaido.

In Fig. 2, for the cooling and heating system, the room air conditioners made up the majority in living rooms, main bedrooms, and the children's rooms. As other heating systems at living room, oil stove / fan heater accounted for 25% (18 cases), floor heating 18% (13 cases), electric carpet and electric kotatsu did 11% (8 cases) respectively. In the case of main bedroom, no heating was reported as 11% (8 cases), oil stove / fan heater was 7 cases, electric stove / fan heater was used at 4 rooms. Children's rooms were not much different from main bedrooms and showed the same breakdown.

At both living and bed rooms, wood floors accounted for the largest proportion of 60 to 70% and a lot of rooms were carpeted. About 30% (21 cases) used carpets at the living room, and the percentage at the main bedrooms was 11% (8 cases). In addition, there were a small number of straw mat, linoleum and vinyl sheets.

For the wall material, there was not a little of difference among living room, main bedroom and children's room, and the ratio of plastered walls at living room was slightly high. The ratio of wallpapers was close to 90% as vinyl or paper accounting for a half of the materials. Only four houses had a wood-based wall finish.

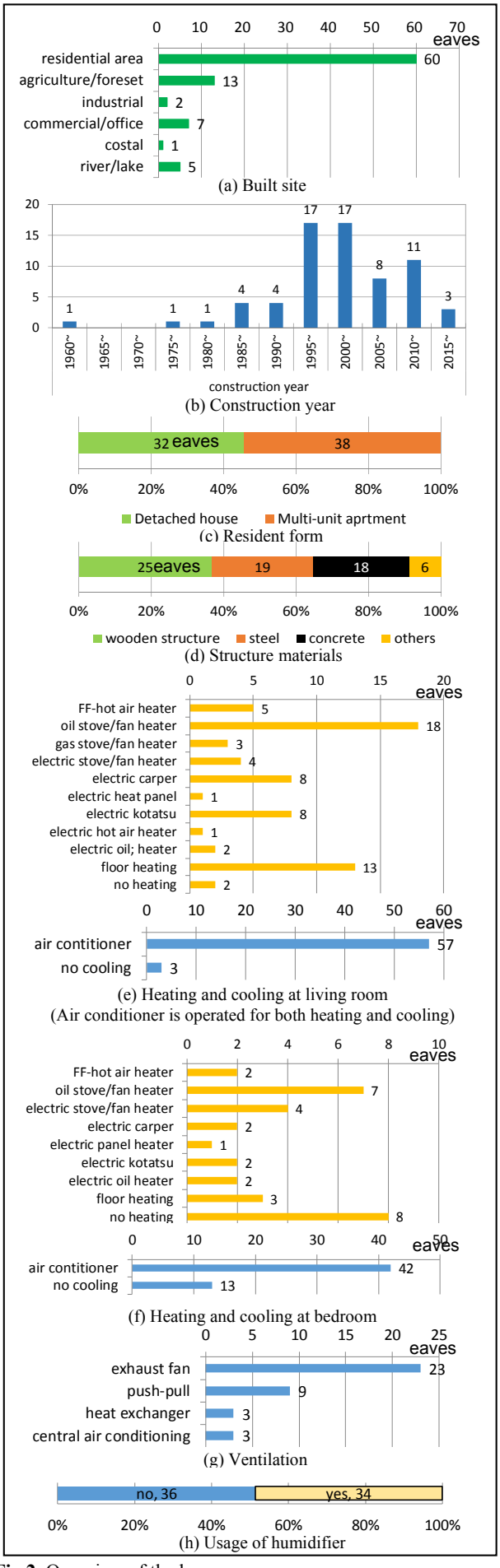

Fig 2. Overview of the houses.

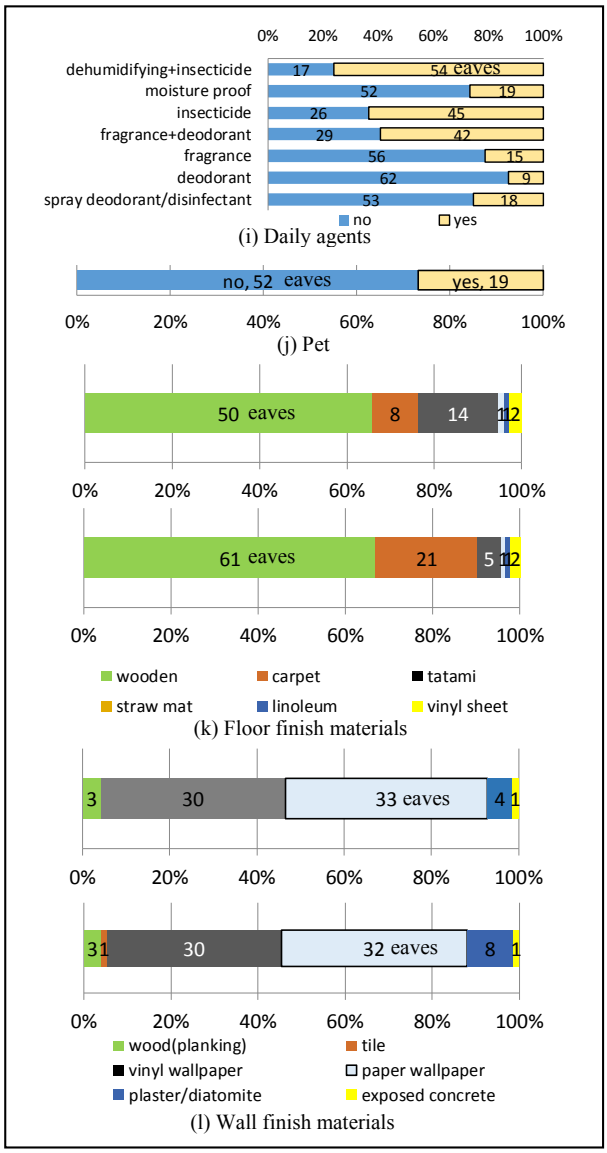

Fig 2. Overview of the houses. (sequel)

3.2 Preliminary test for 10 house dust

Table 1 shows a part of results of the preliminary study.

There is a substantial difference in determined SVOC amounts and components depending on houses. BBP, DIBP, DBP, DEHP and DINP were detected in all houses, and DIDP and DNOP were in large variation by houses. DEHP accounted for 70-80% of the total SVOC, and DINP followed as 10-20%. The two substances accounted for 90% or more as ratio. BBP, DNOP, DMP and DEP were minute amount or below the quantitative limit.

The results leaded to selecting less than 100μm and 100-250μm as the target size of dusts to be investigated intensively by considering the concentration distribution and the oral intake of humans.

3.3 SVOC concentrations in house dusts

SVOC concentrations and component ratio of SVOC concentrations in house dusts for the two particle sizes were shown in Table 2 and in Figure 4. 59 specimens in less than 100μm and 63 specimens in 100-250μm were

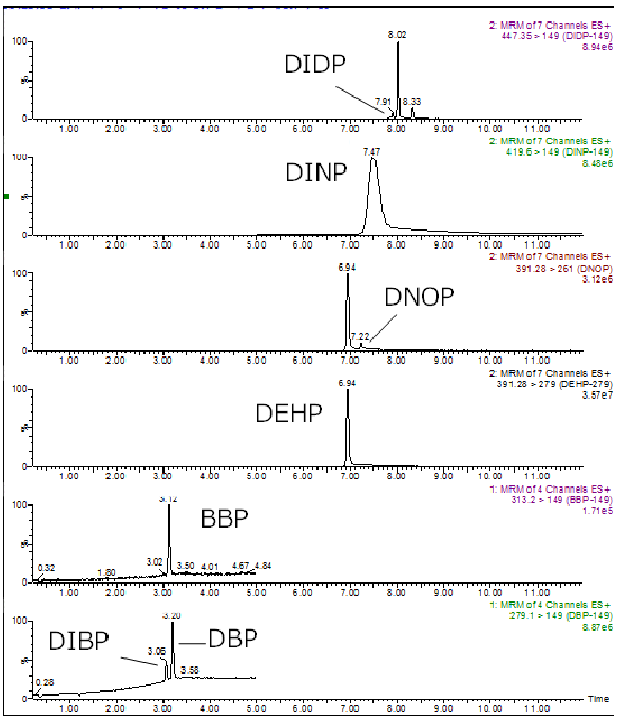

Fig 3. Chromatogram of SVOCs in dust (Solvent extraction, LC-MS/MS).

effectively determined. The average of the total amount of SVOC (SUM) was 1,983 μg/g in dusts of the particle size of less than 100 μm, and 3,028 μg/g in 100-250 μm. In the component ratio, DEHP accounted for 83 to 84% of SUM and DINP was 13%. Considering other existing researches and the domestic production amount of plasticizer, it was considered to be a reasonable value. Consequently the two substances accounted for 96 to 97% of SUM and only small amounts in the other seven components were detected.

4 Discussion

Correlation analysis (in Table 3) showed there was no significant difference between the component ratio of SVOC and dust sizes. It was observed lower concentration of DEHP (100-250 μm) and SUM (100-250 μm) at housing that their location was in a residential area. Significant difference was also found for DEHP, DINP, SUM concentrations with house ages and residence years in dusts of 100-250 μm. The longer the house ages and residence years, the higher the concentrations were detected. In the case of using wooden floor, it made a decrease tendency in DEHP, DINP, SUM concentrations. In the other hand, PVC floor sheet increased significantly DEHP, SUM concentrations.

Significant difference, however, was not obtained from kinds of wall and ceiling material, frequency of running a vacuum cleaner, cleaning method and having a pet.

5 Conclusions

In order to assess the intake amount and human health risk of SVOC from indoor air, house dust, transdermal absorption, this report indicated that it is important

Table 1. Examples of SVOC determination in house dust by particle size in the preliminary study (less than 100, 100-250, 250-500, more than 500μm).

House A		Amounts (μg/g)																					
		BBP			DIBP			DBP			DEHP			DINP			DIDP			DNOP			DMP			DEP		
size (μm)	samples	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD
<500	5	12.1	±	0.9	3.5	±	0.6	14.1	±	2.3	640	±	96	469	±	48	7.9	±	12.1	0.4	±	0.9	0	±	0	0	±	0
250-500	5	23.5	±	6.2	4.9	±	1.0	18.4	±	2.6	1,076	±	517	724	±	106	4.7	±	1.4	0.6	±	1.4	0	±	0	0	±	0
100-250	5	24.9	±	1.2	3.6	±	1.1	21.9	±	2.3	1,145	±	95	848	±	32	4.7	±	0.2	0.0	±	0.0	0	±	0	0	±	0
100<	5	20.5	±	0.6	2.0	±	1.2	20.8	±	1.6	1,274	±	86	509	±	304	3.9	±	2.3	0.0	±	0.0	0	±	0	0	±	0
House C		**Amounts (μg/g)**																					
		BBP			DIBP			DBP			DEHP			DINP			DIDP			DNOP			DMP			DEP		
粒径別 (μm)	samples	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD
<500	5	1.4	±	0.2	7.6	±	2.2	27.4	±	3.4	1,759	±	219	256	±	219	260.5	±	30.8	0.2	±	0.5	0.0	±	0.0	0.1	±	0.1
250-500	5	1.6	±	0.5	9.7	±	1.6	40.6	±	9.4	2,850	±	739	304	±	274	267.0	±	14.7	0.4	±	0.8	0.1	±	0.3	0.4	±	0.6
100-250	5	1.2	±	0.3	9.5	±	1.2	75.8	±	29.1	4,694	±	911	330	±	42.6	243.8	±	38.0	0.0	±	0.0	0.0	±	0.0	0.5	±	1.0
100<	5	1.8	±	0.3	8.6	±	0.6	50.1	±	4.9	2,042	±	97.3	123	±	18.0	238.6	±	10.9	0.0	±	0.0	0.0	±	0.0	0.1	±	0.1

to investigate intensively the two dust sizes of less than 100μm and 100-250μm when considering the concentration distribution and the oral intake of human.

DEHP accounted for 83 to 84% of SUM and DINP was 13% by weight ratio to whole SVOC components, and the two substances accounted for 96 to 97% of SUM, and only the small amounts were detected in the other seven components.

Significant difference was obtained between a few substances of SVOC and several living environmental items such as residential location, type of floor material, years after construction and/or residence, fragrance, spray type deodorant and disinfectant.

Table 2. SVOC concentrations in house dusts by particle size (less than 100 and 100-250μm).

	Phthalate ester (μg/g dust)							
	<100 μm (n=59)				100-250 μm (n=63)			
	min	max	Median	Detection (%)	min	max	Median	Detection (%)
DMP	<L.D.	1.17	0.10	64.4	<L.D.	0.46	0.09	68.3
DEP	<L.D.	1.67	0.41	84.7	<L.D.	1.52	0.41	93.7
BBP	<L.D.	41.4	0.39	79.7	<L.D.	123	0.58	88.9
DBP	2.2	239	18.5	100	1.7	404	17.7	100
DIBP	<L.D.	72.1	2.82	93.2	0.63	56.3	3.40	100
DEHP	234	5900	1381	100	316	10308	1865	100
DINP	13	936	138	100	26	1488	188	100
DNOP	<L.D.	5.44	0.00	13.6	<L.D.	6.95	0.00	36.5
DIDP	1.85	125	5.98	100	<L.D.	120	4.91	74.6

L.D.: Limit of detection

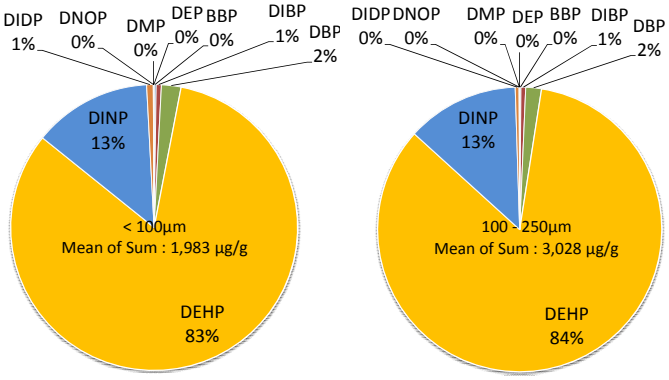

Fig 4. Ratio of SVOC components by dust particle size in 59 house dusts (less than 100, 100-250μm).

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Table 3. Correlation analysis between SVOC concentrations in house dusts and residential environments (showed only variables with a significant difference)

	objective variable	explanatory variable	estimated	t-value	p-value (Prob>|t|)
built area	DEHP 100-250μm	residential area	-1846	-2.42	0.018
	DINP 100-250μm	river/lake	362	2.10	0.040
	SUM 100-250μm	residential area	-1985	-2.45	0.017
surroundings	DINP 100-250μm	electrical train line	223	2.15	0.035
house outline	DEHP 100-250μm	house age	81	2.74	0.008
	DINP 100-250μm	residence year	100	3.13	0.003
	SUM 100-250μm	house age	88	2.79	0.007
floor finish at living room	DEHP <100μm	LF-wooden floor	-1430	-2.71	0.009
	DEHP <100μm	LF-vinyl sheet	2128	2.08	0.042
	DEHP 100-250μm	LF-wooden floor	-3389	-5.07	<.0001
	DEHP 100-250μm	LF-tatami (Japanese straw mat)	2144	2.06	0.044
	DEHP 100-250μm	LF-vinyl sheet	3727	2.34	0.023
	SUM 100-250μm	LF-wooden floor	-3590	-5.04	<.0001
	SUM 100-250μm	LF-tatami (Japanese straw mat)	2333	2.11	0.039
	SUM 100-250μm	LF-vinyl sheet	3701	2.17	0.034
floor finish at bedroom	DEHP <100μm	BF-vinyl sheet	2128	2.08	0.042
	DEHP 100-250μm	BF-vinyl sheet	3727	2.34	0.023
	SUM 100-250μm	BF-vinyl sheet	3701	2.17	0.034
wall finish at living room	DEHP 100-250μm	LW-plaster/diatomite	2000	2.24	0.028
	SUM 100-250μm	LW-plaster/diatomite	2133	2.25	0.028
cooling and heating	DINP <100μm	oil burner/fan heater	218	2.25	0.028
	SUM <100μm	FF-hot air heater	2561	2.33	0.023
	DINP 100-250μm	oil burner/fan heater	293	2.05	0.045
ventilation	DEHP 100-250μm	exhause fan	1676	2.85	0.006
	SUM 100-250μm	exhause fan	1796	2.87	0.006
groceries	SUM <100μm	fragrance	-954	-2.03	0.047
	DINP 100-250μm	spray deodorant・disinfectant	198	2.05	0.045

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