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# Distribution of 58 Semi-Volatile Organic Chemicals in the Gas Phase and Three Particle Sizes in Indoor Air and House Dust in Residential Buildings During the Hot Season in Japan

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**A variety of semi-volatile organic chemicals (SVOCs), such as plasticizers and flame retardants, are released into indoor air and dust from building materials, furniture, and housekeeping products in residential housing. In this study, we measured 58 SVOCs in indoor air and dust from 50 and 48 dwellings, respectively, from 19 prefectures across Japan during the hot season (from July to September). In order to reveal the current situation regarding these compounds in indoor air, we obtained indoor air samples using a newly designed four-stage multi-nozzle cascade impactor and measured the concentrations of the chemicals in the indoor air in three different particle size ranges (<2.5, 2.5-10, and >10μm), as well as a gas phase. From the results obtained using the multi-nozzle cascade impactor, smaller compounds were mainly detected in the gas phase and larger compounds were found in the particle phases. However, the three cyclic polysiloxanes, including decamethylcyclooctasiloxane (D5), with large molecules were detected in the gas phase in all of the houses. Among the 58 chemicals, D5 showed the highest median concentration (1.1 μg/m³) in the range from 0.2 to 36 μg/m³ in the indoor air samples. Our analysis of house dust revealed that di-2-ethylhexyl phthalate (DEHP) was present in all samples at the highest median concentration (590 μg/g) in the range from 200 to 6,200 μg/g. These results suggest that the residential indoor environment in Japan is mainly polluted with siloxanes in the gas phase of indoor air and by DEHP in house dust.**

**Key words** indoor air chemicals, plasticizer, organophosphorus flame retardant, semi-volatile organic compounds, house dust

## INTRODUCTION

The development of new building materials, furnishings, and consumer products in recent decades has resulted in a corresponding increase in new chemicals in indoor environments.[1–3] Indoor chemical concentrations in the 1990s were higher than those found 50 years earlier because of the wider variety of chemicals used and the suppression of air exchange rates in buildings to improve their energy efficiency when regulating residential thermal conditions. Health problems suspected to be induced by indoor air polluted with volatile organic chemicals (VOCs) have been observed in Japan, particularly in the 1990s.[4,5] However, these problems have shown general improvement as a result of the Japanese Ministry of Health, Labor and Welfare setting guideline values for 13 compounds between 1997 and 2002. Nevertheless, "non-regulated" chemicals have been used in place of the 13 regulated compounds and have caused sporadic health problems.[6,7]

Plasticizers and flame retardants are indispensable as they add strength, plasticity and safety to synthetic resins and fibers widely used in modern buildings, electrical devices, and household products. In addition, large amounts of these chemicals are used in a variety of building materials and furniture. Many of these chemicals are classified as semi-volatile organic compounds (SVOCs), which are a group of chemicals that have higher boiling points than VOCs. As a result of the lower volatility of SVOCs, their concentrations in indoor air are generally much lower than those of VOCs.[8–10] A number of SVOCs have also been detected in indoor dust.[11,12] Although the level of consumption and diversity of these chemicals have been increasing, information on the concentrations of these chemicals in indoor air is limited to only a small number of compounds. On the other hand, SVOCs, such as pesticides, plasticizers, and flame retardants, have been suggested to cause adverse health effects, including ocular and nasal symptoms, allergic diseases and endocrine-disrupting effects.[13–19]

SPM (suspended particulate matter) such as $PM_{10}$ and $PM_{2.5}$ (particles less than 10 and 2.5 micrometers in diameter, respectively) can enter the respiratory tract through respiration. $PM_{2.5}$, in particular, can penetrate deeply into the lungs, and may irritate and corrode the alveolar wall, consequently impairing lung function.[20] Fine particulate matter problem

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is a common topic in indoor air science today.[21] If indoor air chemicals exist as SPM, they may result in human exposure *via* the lungs. However, there is only limited information on the particle size and/or gas distributions of indoor air chemicals in actual dwellings.

In our previous study,[22] we developed a new sampling method to collect samples in three different size particulate ranges (<2.5, 2.5–10, and >10 μm) and the gas phase in indoor air. In the present study, we measured 58 SVOCs, including plasticizers (phthalates, adipates, and others) and organophosphorus flame retardants (OPFRs) in the indoor air and house dust from the living rooms of 50 and 48 dwellings, respectively, during the hot season (from July to August) in Japan. By comparing the results of a previous study in which samples were collected during the cold season (from October to January),[22] the influence of temperature on the presence of SVOCs in the various particle sizes and gas phase in indoor air and their concentrations in house dust samples could also be examined.

## MATERIALS AND METHODS

**Chemicals** The chemicals used in this study were of the highest available purity and purchased or provided as analytical standards from the companies listed in Table 1. The chemical names and their abbreviations for every category are also listed in Table 1. Anthracene-$d_{10}$, purchased from Wako Pure Chemical Industry (Osaka, Japan), was used as an internal control in the GC/MS analysis in this study.

**Sampling and Analysis of Indoor Air Chemicals** Air samples were collected from 50 houses in 19 prefectures across Japan (Fig. 1), with the houses aged between three and 48 years old (median 13 years). They included 32 detached houses and 16 condominiums, and the 26 of the 32 houses were constructed from wood. PVC (poly vinyl chloride) wallpaper, which is a known source of phthalate ester emissions, was used in 16 houses. The interior in 9 houses had been renovated more than six years before the air sampling in this study. The indoor air and house dust sampling were performed in the same way as in a previous study.[22] Briefly, the air sampling rate was 10 L/min, and the sampling was performed for 24 h with the residents living their everyday life. The sampling was performed during the hottest season (from July and September) in 2014. A multi-nozzle cascade impactor (air sampling cartridge) equipped with a four-stage filter (Tokyo Dylec Corp., Tokyo, Japan) was used for the separation of the indoor air chemicals. The air sampling cartridge is designed to be able to capture indoor air chemicals based on their form (three sizes of particles and gas). A 47-mm quartz fiber filter (Tokyo Dylec Corp., Tokyo, Japan), which can catch particles larger than 10 nm, was used to catch particulate chemicals in three size ranges (larger than 10 μm, 2.5-10 μm, and smaller than 2.5 μm), and a solid-phase extraction disk (Empore 2215 FF $C_{18}$ Disk; 3M Company, Saint Paul, MN, U.S.A) was used as the last (fourth) stage to catch gaseous chemicals that were not captured by the three previous stages. The quartz fiber filters were baked at 400°C for 3 h for cleaning. The solid-phase extraction disk, sampling cartridges and all of the glassware tools were washed in ultra-purified acetone (acetone for pesticide residue and polychlorinated biphenyl analysis produced by Wako Pure Chemical Industries Ltd., Osaka, Japan) using an ultrasonic cleaner to remove pollutants, such as phthalates and organophosphorus flame retardants, prior to use. After indoor air sampling, each stage of the filters was rolled up and placed in a 10-mL test tube along with 10 μL of 100 μg/mL anthracene-$d_{10}$ added as an internal control. Compounds were extracted from the filters by ultra-sonication for 15 min with 10 mL of acetone and then concentrated to one mL using a rotary evaporator. The concentrated extracts were subjected to GC/MS.

**Sampling and Analysis of Indoor Dust** Indoor dust samples were collected from 48 of the 50 houses using a compact vacuum cleaner equipped with a Teflon fiber bag (adequate dust samples could not be collected in two of the 50 houses) as previously reported.[22] After removing contaminants, such as pieces of food and hair from the dust samples, 10 mg of the dust was placed in a pre-cleaned 10-mL centrifuge test tube. Two mL of acetone and 10 μL of 100 μg/mL anthracene-$d_{10}$ were added and chemicals were extracted by ultra-sonication for 15 min. The extracts were centrifuged at 1,000 rpm for five minutes and the supernatants were subjected to GC/MS.

**Instruments and Analytical Conditions** Analysis of the test compounds was performed using a Shimadzu QP-2010 GC/MS system equipped with a DB-5MS column (30 m × 0.25 mm i.d. × 0.25 μm) as previously reported.[22] The following conditions were used for quantitative detection in this study: injection volume, 2 μL (1 μL for dust samples); carrier gas, helium; and column head pressure, 72 kPa. The GC oven was initially maintained at 40 °C for 2 min, then increased by 25 °C/min to 200 °C and 40 °C/min to 280 °C, where it was maintained for 6 min, and finally increased by 10 °C/min to 320 °C, where it was maintained for 7 min.

The limit of detection (LOD) was defined as the amount of each test compound that yielded S/N = 3 in the analysis. The limits of quantification (LOQ) of the indoor chemicals were calculated from their LODs (except for DEP, DBP, DEHP, Txol, and TBP, which were calculated based on the "travel blank" values described below), the area of the peaks in the analysis of the test compounds extracted from the filters, and the air sampling volume (14.4 $m^3$). To check for contamination by chemicals during the transport of the sampling materials and air sampling, we used a "travel blank," which was an additional cartridge equipped with the same types of filters used for the air sampling that traveled together with the sampling materials. The nine compounds (DEP, DBP, DEHP, Txol, TBP TXIB, D4, D5, and D6) were detected at low concentrations even in the extracts from the cleaned filters, including those of the travel blanks. Therefore, the LOQs of those compounds were set at a value three-fold that of the highest concentration among the travel blanks in this study. The indoor air concentrations of these compounds were calculated by subtracting the travel blank values from the indoor air sample values.

Recovery tests were performed and revealed that the recovery rates were within the range of 50-150% (data not shown). Laboratory blanks and travel blanks were also checked regularly. Thirty-three percent of breakthrough was found for 2EH under the air flow condition of 10 L/min for 24 h by the breakthrough test using two stages of the solid-phase extraction disk, although breakthrough rates of the other test compounds were within 10%.

**Table 1.** Chemical Names and Abbreviations of the 58 Compounds Measured in This Study

| No. | Chemical name | Abbreviation | Supplier |
|---|---|---|---|
| 21 Phthalates | | | |
| 1 | Dimethyl phthalate | DMP | Wako[a] |
| 2 | Diethyl phthalate | DEP | Wako |
| 3 | Diisopropyl phthalate | DiPP | TCI[b] |
| 4 | Diallyl phthalate | DAP | Wako |
| 5 | Dipropyl phthalate | DPP | Wako |
| 6 | Diisobutyl phthalate | DiBP | Wako |
| 7 | Dibutyl phthalate | DBP | Wako |
| 8 | Dipentyl phthalate | DPeP | Kanto[c] |
| 9 | Diisohexyl phthalate | DiHP | Kanto |
| 10 | Butyl phthalyl butyl glycolate | BPBG | Wako |
| 11 | Dihexyl phthalate | DHP | Wako |
| 12 | Butyl benzyl phthalate | BBzP | Kanto |
| 13 | Diisoheptyl phthalate | DiHpP | S-A[d] |
| 14 | Di (2-ethyl-1-hexyl) phthalate | DEHP | Wako |
| 15 | Diheptyl phthalate | DHpP | Wako |
| 16 | Dicyclohexyl phthalate | DcHP | Wako |
| 17 | Diphenyl phthalate | DPhP | Wako |
| 18 | Dioctyl phthalate | DOP | Wako |
| 19 | Dibenzyl phthalate | DBzP | Wako |
| 20 | Diisononyl phthalate | DiNP | Wako |
| 21 | Diisodecyl phthalate | DiDP | Wako |
| 23 Non-phthalate plasticizers | | | |
| 1 | Octamethylcyclotetrasiloxane | D4 | Wako |
| 2 | 2-Ethyl-1-hexanol | 2EH | Wako |
| 3 | 1-Methyl-2-pyrrolidinone | 1M2Pd | Wako |
| 4 | Decamethylcyclopentasiloxane | D5 | Wako |
| 5 | Dimethyl adipate | DMA | Wako |
| 6 | Dodecamethylcyclohexasiloxane | D6 | TCI |
| 7 | Diethyl adipate | DEA | Wako |
| 8 | 2,2,4-Trimethyl-1,3-pentanediol monoisobutyrate | Txol | S-A |
| 9 | Diisopropyl adipate | DiPA | Wako |
| 10 | 2,2,4-Trimethyl-1,3-pentanediol diisobutyrate | TXIB | S-A |
| 11 | Diopropyl adipate | DPA | Junsei[e] |
| 12 | Dibutyl adipate | DBA | Wako |
| 13 | Diisobutyl adipate | DiBA | Wako |
| 14 | Isopropyl myristate | iPMs | Wako |
| 15 | Dibutyl sebacate | DBSb | Wako |
| 16 | AcetylTributyl citrate | AcTBCt | Wako |
| 17 | Di (2-ethyl-1-hexyl) adipate | DEHA | Kanto |
| 18 | Di (2-ethyl-1-hexyl) terephthalate | DEHT | Wako |
| 19 | Di (2-ethyl-1-hexyl) azelate | BEHAz | Wako |
| 20 | 1,2-Cyclohexane dicarboxylic acid diisononyl ester | DINCH | Wako |
| 21 | Bis (2-ethyl-1-hexyl) sebacate | BEHSb | Wako |
| 22 | Dioctyl azelate | DOAz | Wako |
| 23 | Tris (2-ethyl-1-hexyl) trimellitate | TOTM | Wako |
| 14 Phosphorous flame retardants | | | |
| 1 | Trimethyl phosphate | TMP | Wako |
| 2 | Triethyl phosphate | TEP | Wako |
| 3 | Tripropyl phosphate | TPrP | S-A |
| 4 | Tributyl Phosphate | TBP | Wako |
| 5 | Tris (2-chloroethyl) phosphate | TCEP | Wako |
| 6 | Tris (2-chloro-1-methylethyl) phosphate | TCIPP | Wako |
| 7 | Tris (1,3-dichloro-2-propyl) phosphate | TDCIPP | Wako |
| 8 | Tris (2-butoxyethyl) phosphate | TBEP | Wako |
| 9 | Triphenyl phosphate | TPhP | Wako |
| 10 | Tri (2-ethyl-1-hexyl) phosphate | TEHP | Wako |
| 11 | 2-Ethyl-1-hexyl diphenyl phosphate | EHDPhP | TCI |
| 12 | Cresyl diphenyl phosphate | CsDPhP | TCI |
| 13 | Tricresyl phosphate | TCsP | Wako |
| 14 | Trixylenyl phosphate | TXP | DH[g] |

[a]Wako: Wako Pure Chemical Industry (Osaka, Japan)
[b]TCI: Tokyo Chemical Industry (Tokyo, Japan)
[c]Kanto: Kanto Chemical Co. Inc. (Tokyo, Japan)
[d]S-A: Sigma-Aldrich Co. (St. Louis, MO, USA)
[e]Junsei: Junsei Chemical Co. Ltd. (Tokyo, Japan)
[f]DH: Daihachi Chemical Industry (Osaka, Japan)

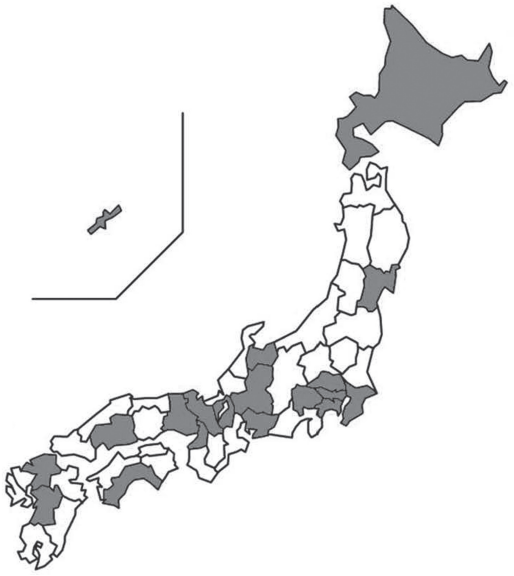

**Fig. 1.** The 19 Prefectures in Japan Where the Indoor Air and House Dust Samples Were Collected in This Study

## RESULTS

**Levels of Plasticizers and OPFRs in the Indoor Air from 50 Japanese Houses** Table 2 shows the concentration ranges for 36 of the 58 measured compounds detected in the indoor air from the 50 houses by the order of the molecular weight.

Nine (DMP, DEP, DiBP, DBP, BBzP, DHP, DEHP, DOP and DiNP) of the 21 phthalates measured were detected at different detection frequencies from indoor air of the living rooms in the 50 houses. Among these nine phthalates, DMP, DiBP, DBP and DEHP were detected from all the 50 houses. We found that DBP was present in the indoor air at concentrations ranging from 0.2 to 3.6 μg/m$^3$, and it showed the highest median concentration (0.65 μg/m$^3$) among the 21 phthalates. The order of the median concentrations was as follows; DBP > DEHP > DiBP > DMP.

The compound detected at the highest concentration was D5 (36 μg/m$^3$), which was detected from all 50 houses, and D5 was the only compound with median value exceeding 1 μg/m$^3$ (1.1 μg/m$^3$). Other compounds, which were detected at concentrations higher than 5 μg/m$^3$, were 2EH, DMA, Txol, DiPA, and D6. Two adipates, DMA and DiPA, were detected at 7.2 μg/m$^3$ and 6.5 μg/m$^3$, respectively. Txol was detected from indoor air at the third highest concentration (14 μg/m$^3$), and its detection rate was 100%.

Nine of the 14 OPFRs were detected in the indoor air of the 50 houses, and TCIPP was detected at the highest concentration (0.25 μg/m$^3$) among the OPFRs. OPFRs were detected at lower concentrations (<0.25 μg/m$^3$) than those of PPs (<3.6 μg/m$^3$) and NPPs (<36 μg/m$^3$) in the indoor air.

Separate sampling in the gas phase and three sizes of particle phases was conducted in this study. The 36 compounds detected are listed by their molecular weight in Table 3 and it shows the distribution of the detection ratios of the 36 compounds. Fig. 2 summarizes the average distribution patterns of particle/gas of the 36 compounds in indoor air. These results show that most of these compounds were found to exist as the

**Table 2.** Concentrations of the 36 Compounds Detected in the Indoor Air Samples from the 50 Houses

| Chemical | Med[a] (μg/m³) | Max[b] (μg/m³) | DF[c] (%) | LOQ[d] (μg/m³) |
|---|---|---|---|---|
| *Phthalate plasticizers* | | | | |
| DMP | 0.07 | 0.44 | 100 | 0.002 |
| DEP | 0.11 | 0.39 | 98 | 0.004 |
| DiBP | 0.27 | 3.0 | 100 | 0.003 |
| DBP | 0.65 | 3.6 | 100 | 0.011 |
| BBzP | 0.0037 | 0.044 | 60 | 0.003 |
| DHP | N.D.[e] | 0.027 | 10 | 0.00007 |
| DEHP | 0.6 | 1.3 | 100 | 0.02 |
| DOP | N.D. | 0.062 | 32 | 0.00007 |
| DiNP | 0.011 | 0.14 | 54 | 0.0002 |
| *Non-phthalate plasticizers* | | | | |
| 1M2Pd | 0.062 | 0.98 | 56 | 0.05 |
| 2EH | 0.98 | 28 | 100 | 0.04 |
| DMA | 0.064 | 7.2 | 98 | 0.007 |
| Txol | 0.99 | 14 | 100 | 0.02 |
| DiPA | 0.008 | 6.5 | 94 | 0.001 |
| DBA | 0.0079 | 1.1 | 80 | 0.0009 |
| DiBA | 0.039 | 0.29 | 88 | 0.01 |
| iPMs | 0.33 | 3.8 | 100 | 0.008 |
| TXIB | 0.73 | 4.1 | 100 | 0.01 |
| D4 | 0.034 | 1.0 | 100 | 0.01 |
| DBSb | N.D. | 0.082 | 48 | 0.002 |
| DEHA | 0.018 | 0.11 | 98 | 0.003 |
| D5 | 1.1 | 36 | 100 | 0.01 |
| DEHT | N.D. | 0.021 | 34 | 0.00007 |
| AcTBCt | 0.01 | 0.11 | 98 | 0.0103 |
| BEHSb | 0.0023 | 0.061 | 64 | 0.0002 |
| D6 | 0.52 | 6.0 | 100 | 0.01 |
| TOTM | N.D. | 0.017 | 8 | 0.002 |
| *Phosphorous flame retardants* | | | | |
| TEP | 0.0046 | 0.051 | 100 | 0.00007 |
| TBP | 0.036 | 0.22 | 88 | 0.006 |
| TCEP | 0.0059 | 0.11 | 70 | 0.00007 |
| TPhP | 0.0046 | 0.088 | 80 | 0.0007 |
| TCIPP | 0.025 | 0.25 | 100 | 0.003 |
| EHDPhP | 0.0071 | 0.048 | 84 | 0.0007 |
| TCsP | N.D. | 0.017 | 40 | 0.00007 |
| TBEP | N.D. | 0.089 | 34 | 0.0002 |
| TDCIPP | N.D. | 0.028 | 28 | 0.0002 |

[a]Med: Median
[b]Max: Maximum
[c]DF: Detection frequency
[d]LOQ: Limit of quantitation
[e]N.D.: Non detected

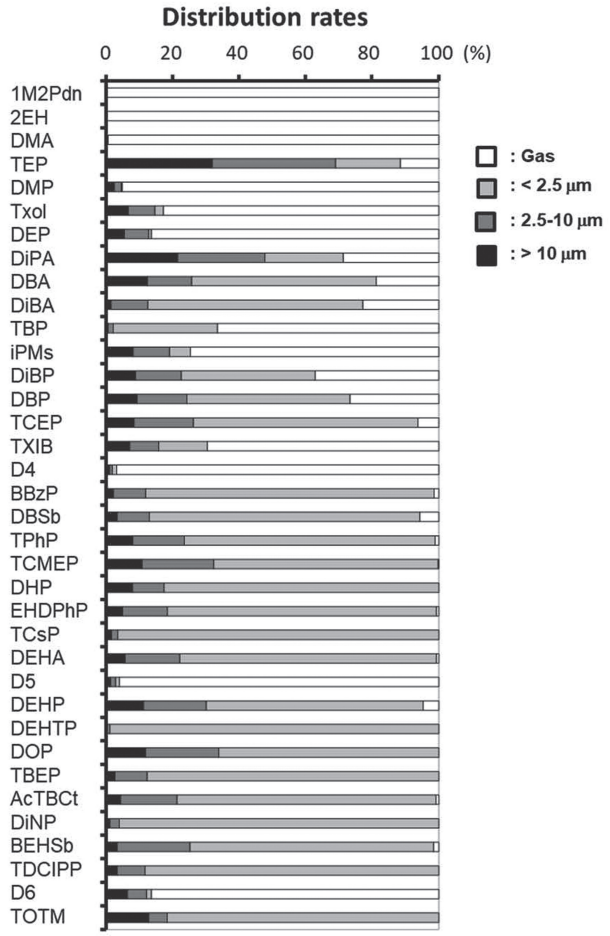


**Fig. 2.** Comparison of Distribution Patterns of Particle/Gas of the 36 Compounds Detected in Indoor Air

smallest particles (<2.5 μm), with compounds having a smaller molecular weight preferentially existing in gas phase. Interestingly, we found that three cyclic siloxanes (D4, D5, and D6) were almost always present in gas phase although they have large molecular weights compared to other gaseous compounds belonging in different chemical groups. D5, in particular, showed the highest median concentration (1.1 μg/m³; with a range from 0.2 to 36 μg/m³) among the 58 SVOCs measured in the indoor air samples.

**Levels of SVOCs in House Dust Samples from 48 Japanese Houses** The 58 compounds were also measured in house dust samples taken from 48 of the 50 houses. The 43 compounds detected are listed by their molecular weight in Table 4. The compound detected at the highest concentration from dust samples was DEHP (6,200 μg/g). DiNP was detected from dust samples at a median concentration of 100 μg/g and a maximum of 1,700 μg/g, which was the second highest concentration found in this study. TBEP was detected at the second highest concentration (100 μg/g median, 5500 μg/g maximum) in this study and its detection frequency was 100%. The detection frequencies of seven compounds (2EH, DiBP, DBP, DEHP, TBEP, DiNP, and TOTM) were also found to be 100% (Table 4).

## DISCUSSION

**Plasticizers and Organophosphorus Flame Retardants in the Indoor Air** DBP was the phthalate detected at the highest concentration in indoor air in this study (Table 2). Fromme *et al*. reported the indoor air concentrations of DBP in 59 apartments and 74 kindergartens in Berlin and the maximum concentrations were reported at 5.5 μg/m³ and 13 μg/m³, respectively.[23] The maximum concentration (3.6 μg/m³) of DBP found in the present study was lower than those in Berlin and the new Japanese guideline value (17 μg/m³) for indoor air concentrations of DBP, which was reduced from 220 to 17 μg/m³ on January 17 in 2019.

2EH was detected at 28 μg/m³ in indoor air, although this compound was also detected at the highest concentration of 5.1 μg/m³ in the previous study performed during cold season.[22] Norbäck *et al*. reported that 2EH is a hydrolysis product

**Table 3.** Particle Detection Ratios of the 36 Compounds Detected in the Indoor Air Samples from the 50 Houses

| No. | Chemical | MW[a] | >10 μm | | | 2.5-10 μm | | | <2.5 μm | | | Gas | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Med[b] (%) | Max[c] (%) | n[d] | Med (%) | Max (%) | n | Med (%) | Max (%) | n | Med (%) | Max (%) | n |
| 1 | 1M2Pd | 99 | | | 0 | | | 0 | | | 0 | 100 | 100 | 19 |
| 2 | 2EH | 130 | | | 0 | | | 0 | | | 0 | 100 | 100 | 38 |
| 3 | DMA | 174 | | 2 | 5 | | 0.3 | 3 | | | 1 | 100 | 100 | 37 |
| 4 | TEP | 182 | 33 | 50 | 49 | 37 | 51 | 46 | 26 | 35 | 33 | | 77 | 6 |
| 5 | DMP | 194 | | 12 | 17 | | 7 | 14 | | 1 | 5 | 100 | 100 | 37 |
| 6 | Txol | 216 | 6 | 12 | 49 | 7 | 15 | 46 | 2 | 9 | 29 | 78 | 94 | 38 |
| 7 | DEP | 222 | 5 | 11 | 40 | 7 | 13 | 38 | | 4 | 6 | 85 | 100 | 37 |
| 8 | DiPA | 230 | 24 | 47 | 45 | 29 | 50 | 43 | 20 | 42 | 34 | | 87 | 16 |
| 9 | DBA | 258 | | 73 | 14 | | 44 | 14 | 59 | 100 | 31 | | 89 | 9 |
| 10 | DiBA | 258 | | 11 | 6 | | 28 | 18 | 67 | 100 | 35 | | 67 | 15 |
| 11 | TBP | 266 | | 6 | 3 | | 12 | 4 | 22 | 100 | 29 | 77 | 100 | 26 |
| 12 | iPMs | 270 | 8 | 13 | 47 | 11 | 17 | 44 | 6 | 16 | 37 | 71 | 91 | 38 |
| 13 | DiBP | 278 | 8 | 12 | 50 | 14 | 18 | 46 | 39 | 60 | 42 | 35 | 70 | 38 |
| 14 | DBP | 278 | 8 | 13 | 50 | 15 | 20 | 46 | 51 | 68 | 42 | 26 | 58 | 38 |
| 15 | TCEP | 285 | | 26 | 15 | 21 | 43 | 19 | 65 | 100 | 24 | | 55 | 4 |
| 16 | TXIB | 286 | 6 | 21 | 48 | 8 | 19 | 44 | 11 | 33 | 42 | 70 | 91 | 36 |
| 17 | D4 | 297 | | 0.4 | 4 | | | 2 | | | 2 | 100 | 100 | 38 |
| 18 | BBzP | 312 | | 14 | 5 | | 26 | 8 | 100 | 100 | 27 | | | 1 |
| 19 | DBSb | 314 | | 15 | 6 | | 22 | 6 | 100 | 100 | 18 | | 42 | 3 |
| 20 | TPhP | 326 | | 30 | 16 | 16 | 43 | 20 | 71 | 100 | 32 | | 5 | 2 |
| 21 | TCIPP | 327 | 13 | 19 | 39 | 21 | 32 | 44 | 66 | 78 | 42 | | | 0 |
| 22 | DHP | 334 | | 24 | 2 | | 27 | 2 | 84 | 100 | 4 | | | 0 |
| 23 | EHDPhP | 362 | | 32 | 8 | | 48 | 12 | 100 | 100 | 34 | | | 0 |
| 24 | TCsP | 368 | | 2 | 1 | | 1 | 1 | 100 | 100 | 14 | | | 0 |
| 25 | DEHA | 370 | | 14 | 22 | 19 | 26 | 29 | 70 | 100 | 41 | | 8 | 3 |
| 26 | D5 | 371 | 0.1 | 3 | 42 | 0.1 | 5 | 39 | | 9 | 16 | 100 | 100 | 37 |
| 27 | DEHP | 390 | 10 | 18 | 44 | 19 | 24 | 45 | 61 | 85 | 42 | 7 | 12 | 26 |
| 28 | DOP | 390 | | 49 | 4 | | 69 | 5 | 100 | 100 | 8 | | | 0 |
| 29 | DEHT | 390 | | | 0 | | 4 | 1 | 100 | 100 | 17 | | | 0 |
| 30 | TBEP | 397 | | 23 | 2 | | 24 | 4 | 100 | 100 | 16 | | | 0 |
| 31 | AcTBCt | 402 | | 14 | 16 | 20 | 30 | 30 | 73 | 100 | 41 | | 8 | 6 |
| 32 | DiNP | 419 | | | 1 | | 24 | 3 | 100 | 100 | 25 | | | 0 |
| 33 | BEHSb | 426 | | 27 | 4 | 25 | 48 | 15 | 100 | 100 | 25 | | | 1 |
| 34 | TDCIPP | 430 | | 23 | 2 | | 36 | 4 | 100 | 100 | 11 | | | 0 |
| 35 | D6 | 445 | 3 | 20 | 44 | 4 | 20 | 44 | | 2 | 5 | 90 | 99 | 37 |
| 36 | TOTM | 546 | | 44 | 1 | | 19 | 1 | 100 | 100 | 4 | | | 0 |

[a]MW: Molecular weight
[b]Med:Median
[c]Max:Maximum
[d]n:Numberofhousesinwhichthechemicalwasdetected

of DEHP, and the presence of 2EH in indoor air could be connected to asthma symptoms in the residents.[24] It was reported that 2EH was detected from some household water-based hand-pump sprays.[25] In addition, Kamijima *et al*. reported that 2EH in indoor air is a possible cause of sick building syndrome on the basis of a comparison of chemical concentrations in indoor air and symptoms in the users of the room in some buildings.[6]

The median value for TCIPP (0.025 μg/m$^3$) in this study was found to be similar to that in US homes (0.026 μg/m$^3$).[26] The concentration range of these compounds (n.d.-0.025 μg/m$^3$) was within the ranges (0.001-1 μg/m$^3$) reported in a review paper of worldwide studies.[10] Many of these compounds detected at relatively higher concentrations in indoor air were also detected at high frequencies. Therefore, we should continue to pay attention to these compounds in order to prevent health problems.

**Cyclic Siloxanes in the Indoor Air** D5 was detected at the highest concentration (36 μg/m$^3$) and detected from all 50 houses in this study. There are a very limited number of reports on the indoor air concentration of cyclic siloxanes. Tran *et al*. reported the levels of indoor air cyclic siloxanes in several categories of indoor environment such as homes, cars, hair salons and so on in USA[27] and Vietnam.[28] They demonstrated that the mean value of D5 in the USA houses was 446 μg/m$^3$ (40.4-1840 μg/m$^3$, n=20) and the median values of D5 in the houses in Vietnam were 150 (Hanoi, n=19), 21 (Bacninh, n=8), 6.8 (Thaibinh, n=6), and 6.6 (Tuyenquang, n=8) μg/m$^3$. These values are much higher than those of the present study and it may be due to large differences in the usage of cosmetic products which was reported to be a main source of D5 in indoor environment.[29] Dekant and Klaunig summarized the toxic effects of D5, such as morphological alterations in the nasal cavity at exposure to >10 ppm in Fischer F344

**Table 4.** Concentrations of the 43 Compounds Detected in the House Dust Samples from 48 Houses

| Chemical | Med[a] (μg/m$^3$) | Max[b] (μg/m$^3$) | DF[c] (%) |
|---|---|---|---|
| *Phthalate plasticizers* | | | |
| DMP | < 0.1 | 3.7 | 27 |
| DEP | 0.19 | 16 | 81 |
| DBP | 20 | 670 | 100 |
| DiBP | 3.4 | 56 | 100 |
| BBzP | 3.0 | 110 | 85 |
| DHP | < 0.1 | 2.2 | 17 |
| DBzP | < 0.1 | 4.4 | 2 |
| DHpP | < 0.1 | 12 | 2 |
| DiHpP | < 0.1 | 30 | 2 |
| DEHP | 590 | 6,200 | 100 |
| DOP | < 0.1 | 40 | 38 |
| DiNP | 100 | 1,700 | 100 |
| DiDP | < 0.1 | 390 | 10 |
| *Non-phthalate plasticizers* | | | |
| 1M2Pdn | < 0.1 | 0.29 | 8 |
| 2EH | 4.7 | 28 | 100 |
| DMA | < 0.1 | 1.3 | 2 |
| Txol | 0.87 | 5.2 | 85 |
| DiPA | < 0.1 | 0.67 | 2 |
| DBA | < 0.1 | 5.9 | 2 |
| DiBA | < 0.1 | 1.5 | 4 |
| iPMs | < 0.1 | 6.0 | 25 |
| TXIB | 0.2 | 24 | 63 |
| D4 | < 0.1 | 0.98 | 2 |
| DBSb | 41 | 200 | 98 |
| DEHA | 5.0 | 63 | 81 |
| D5 | < 0.1 | 2.7 | 2 |
| DEHT | 30 | 320 | 98 |
| AcTBCt | 5.4 | 110 | 98 |
| DOAz | < 0.1 | 23 | 8 |
| BEHSb | < 0.1 | 30 | 10 |
| D6 | < 0.1 | 32 | 35 |
| TOTM | 43 | 370 | 100 |
| *Phosphorous flame retardants* | | | |
| TEP | < 0.1 | 1.1 | 6 |
| TBP | < 0.1 | 25 | 10 |
| TCEP | < 0.1 | 63 | 15 |
| TPhP | 2.3 | 25 | 73 |
| TCIPP | 4.4 | 41 | 81 |
| EHDPhP | < 0.1 | 4.7 | 17 |
| TCsP | < 0.1 | 5.1 | 38 |
| TBEP | 100 | 550 | 100 |
| TXP | < 0.1 | 48 | 4 |
| TDCIPP | < 0.1 | 110 | 40 |
| TEHP | < 0.1 | 3.3 | 2 |

[a]Med: Median
[b]Max: Maximum
[c]DF: Detection frequency

female rats, increased liver weight (NOAEL 100 mg/kg bw/day) in male Sprague-Dawley rats, and increased incidence of uterine adenocarcinoma at exposure to 10 ppm for one year in Fischer F344 female rats, which is considered to represent a dopamine agonist effect of D5.[30]

**Phthalate in House Dust** The compound detected at the highest concentration was DEHP (6,200 μg/g) in this study. Carlstedt *et al*. reported that polyvinyl chloride floorings are related to human exposure levels of DEHP and DBP.[31] In addition, Bertelsen *et al*. found a positive correlation between DEHP exposure and the incidence of asthma in Norwegian children based on a study of urinary biomarkers of DEHP.[32] Moreover, prenatal DEHP exposure is reported to induce some adverse effects such as the disruption of adrenal androgens, glucocorticoids levels, IgE levels, and adipokine levels in cord blood, low birth weight and allergies and infectious diseases up to seven years of age.[33–35] Ait Bamai *et al*. reported the median concentration of DiNP in floor dust taken from 128 houses in Japan was 140 μg/g, with a maximum concentration of 2,100 μg/g.[36] In addition, we reported the median concentration of DiNP in house dust taken from 21 houses in Japan was 140 μg/g, with a maximum of 2,300 μg/g.[22] Therefore, the concentrations in this study (median: 100 μg/g, maximum: 1,700 μg/g) are comparable with those of the two previous studies. On the other hand, the transdermal administration of 15 mg/kg/day of DiNP was reported to induce atopic dermatitis-like skin lesions in atopic-prone NC/Nga mice by transdermal administration.[37] As DEHP and DiNP have been reported to induce some biological effects as described above, we should continue to pay attention to DEHP and DiNP in house dust in order to aid in preventing health problems.

**Non-phthalate Plasticizers in House Dust** TOTM is one of an emerging chemical which is used as an alternative of PPs and information on polluting level of TOTM in indoor environment is very limited. In this study, we detected TOTM from house dust samples at 100% of detection frequency. Christia *et al*. reported that the median TOTM concentrations in house dust from houses in Belgium (n=18), Ireland (n=6), and Netherlands (n=9) were 5.2, 2.6, and 9.8 μg/g, with maximum concentrations of 130, 3.2, and 46 μg/g, respectively.[38] The median concentration of TOTM in house dust was 43 μg/g with a maximum concentration of 370 μg/g in this study, while in the previous study in cold season the values were 25 μg/g and 240 μg/g, respectively.[22] Therefore, the TOTM concentrations in house dust in Japan were found to be higher than those in Europe.

As indoor air pollution by cyclic siloxanes has been recently reported worldwide, levels of environmental exposure to these compounds need to be continuously measured toward the prevention of health risks. Tran *et al*. summarized cyclic siloxane levels in indoor dust in 12 countries and reported the concentrations of D4, D5, and D6 in Japan to be 1.6, 13, and 15 μg/g, respectively.[39] The respective values in the present study were lower (<0.1 μg/g all for D4~D6) than those in report by Tran and colleagues. The cause of the discrepancy is not clear but it may be due to differences in sampling conditions, housekeeping and cosmetic products used, frequency of house cleaning and so on.

**Organophosphorus Flame Retardants in House Dust** Tajima *et al*. reported the median concentration of TBEP in house dust samples from 128 dwellings was 31 μg/g with a maximum concentration of 940 μg/g.[40] The value of TBEP concentration in this study was 100 μg/g in median and 550 μg/g in maximum (Table 4). They also reported that wooden flooring was a possible source of TBEP ($p<0.001$) and introduced TBEP concentrations reported in other studies in Japan (31, 1,570, and 510 μg/g) and other countries [9.4 (Spain), 2.0 (Belgium), 4.0 (New Zealand), 1.6 (Romania), and 0.73 μg/g (Germany)]. Most people in Japan take their shoes off in houses and this custom affects the floor design and materials used in Japan, and it may contribute to the higher concentrations of

TBEP.

Araki *et al*. investigated the relationship between organophosphorus flame retardants in indoor dust and health problems among residents.[41] They found that significant associations between the prevalence of atopic dermatitis and the presence of TCIPP in floor dust (odds ratio: 2.43) in a study of 182 single-family dwellings in Japan for which they reported a median concentration of TCIPP of 8.7 μg/g. The value in this study was 4.4 μg/g (Table 4) and it is slightly higher than those of homes in other countries such as the Philippines (not detected), US (2.8 μg/g), Canada (1.5 μg/g), and the Czech Republic (1.5 μg/g).[26] Therefore, careful attention should be paid to TBEP and TCIPP in house dust, especially in Japan.

**Distribution of Indoor Air SVOCs in Gas Phase, Three Sizes of Particles and House Dust** Gas/particle partitioning of organic compounds is reported to be affected by changes in atmospheric temperature[42,43] and some other studies reported gas/particle correlations between indoor air and house dust for some SVOCs.[44,45] In the previous study, sampling was performed from October to January (in the cold season),[22] while in this study it was performed from July to September (in the hot season). There was a difference of more than 10°C in the median room temperature between the two studies. From the comparison of the two study, median concentrations of eight chemicals (DMP, DEP, TBP, iPMs, DiBP, DBP, DEHA, and DEHP) in this study were approximately 3-fold higher than those of the previous study. In terms of house dust samples, 12 of the 17 compounds detected in this study were smaller than those in the previous study. These results suggest that SVOCs may increase in indoor air and decrease in house dust in hot season and higher room temperature may contribute vaporization of the chemicals in house dust and indoor materials and furniture.

## Conclusions

We investigated SVOCs in indoor air separately depend on the existence forms of three particle sizes (>10, 2.5-10, <2.5 μm in diameter) and gas. Using the method 58 SVOCs in indoor air of 50 dwellings during the hot season (from July to September) in Japan were analyzed. Of the 58 compounds, 36 were detected from the indoor air samples, with the concentration of D5 (36 μg/m$^3$) being the highest. In addition, we found that compounds with a molecular weight smaller than DBP (*m/z* 279) and cyclic polysiloxanes such as (D4~D6) were more commonly detected as a gas. On the other hand, larger SVOCs were commonly detected as the smallest particles (<2.5 μm). These results suggest that indoor air SVOCs can enter into the deepest area of the respiratory tract of residents. From a comparison with the data from previous study carried out in cold season, it suggests that higher room temperature might affect concentrations of chemicals in indoor air to increase but those in house dust to decrease.

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**Conflict of interest** The authors declare no conflict of interest.

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