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# Occurrence of Eight Bisphenol Analogues in Indoor Dust from the United States and Several Asian Countries: Implications for Human Exposure

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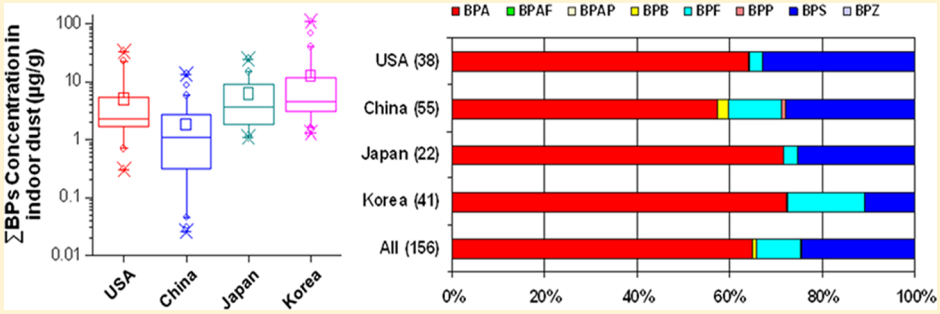
Supporting Information




**ABSTRACT:** Bisphenol A has been reported to be a ubiquitous contaminant in indoor dust, and human exposure to this compound is well documented. Information on the occurrence of and human exposure to other bisphenol analogues is limited. In this study, eight bisphenol analogues, namely 2,2-bis(4-hydroxyphenyl)propane (BPA), 4,4′-(hexafluoroisopropylidene)-diphenol (BPAF), 4,4′-(1-phenylethylidene)bisphenol (BPAP), 2,2-bis(4-hydroxyphenyl)butane (BPB), 4,4′-dihydroxydiphenylmethane (BPF), 4,4′-(1,4-phenylenediisopropylidene)bisphenol (BPP), 4,4′- sulfonyldiphenol (BPS), and 4,4′-cyclohexylidenebisphenol (BPZ), were determined in indoor dust samples ($n$ = 156) collected from the United States (U.S.), China, Japan, and Korea. Samples were extracted by solid−liquid extraction, purified by automated solid phase extraction methods, and determined by liquid chromatography-tandem mass spectrometry (LC-MS/MS). The total concentrations of bisphenols (∑BPs; sum of eight bisphenols) in dust were in the range of 0.026−111 μg/g (geometric mean: 2.29 μg/g). BPA, BPS, and BPF were the three major bisphenols, accounting for >98% of the total concentrations. Other bisphenol analogues were rare or not detected, with the exception of BPAF, which was found in 76% of the 41 samples collected in Korea (geometric mean: 0.0039 μg/g). The indoor dust samples from Korea contained the highest concentrations of both individual and total bisphenols. BPA concentrations in dust were compared among three microenvironments (house, office, and laboratory). The estimated median daily intake (EDI) of ∑BPs through dust ingestion in the U.S., China, Japan, and Korea was 12.6, 4.61, 15.8, and 18.6 ng/kg body weight (bw)/day, respectively, for toddlers and 1.72, 0.78, 2.65, and 3.13 ng/kg bw/day, respectively, for adults. This is the first report on the occurrence of bisphenols, other than BPA, in indoor dust.

## INTRODUCTION

Due to endocrine disrupting chemicals' (EDCs) potential to adversely affect human health, concern about human exposure is growing.[1,2] EDCs consist of a wide range of substances, including organochlorine pesticides, organotins, polychlori-

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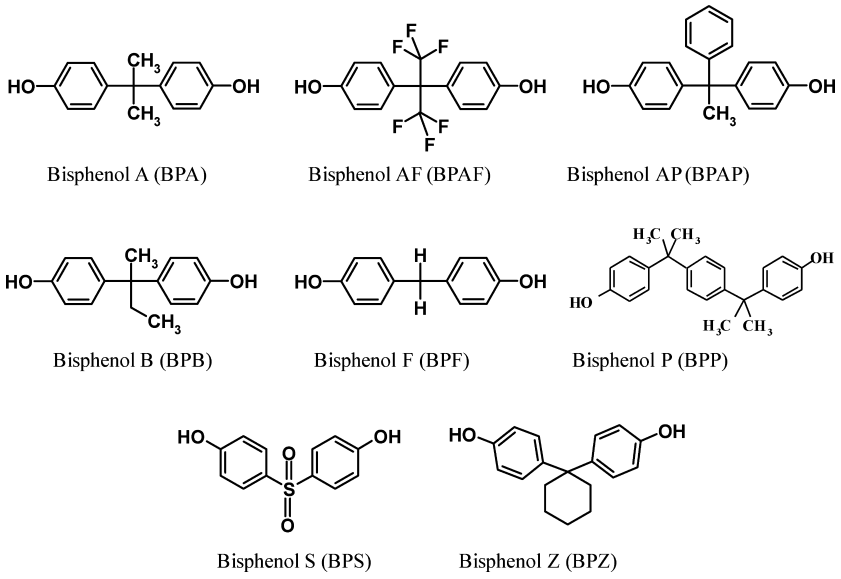


**Figure 1.** Chemical structures of several bisphenols analyzed in the present study.

nated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), phthalates, and bisphenols, among others. Mounting evidence suggests that EDCs interfere with the body's endocrine system and adversely affect reproduction and development, neural network, and cardiovascular, metabolic, and immune systems in humans.[1,2]

Bisphenols are a group of chemicals with two hydroxyphenyl functionalities and include several analogues such as bisphenol A [BPA; 2,2-bis(4-hydroxyphenyl)propane], bisphenol B [BPB; 2,2-bis(4-hydroxyphenyl)butane], bisphenol F (BPF; 4,4′-dihydroxydiphenylmethane), bisphenol AF [BPAF; 4,4′-(hexafluoroisopropylidene)diphenol], and bisphenol S (BPS; 4,4′-sulfonyldiphenol). Bisphenols have been widely used as the base chemical in the manufacturing of polycarbonate plastics and the resin lining of food and beverage cans.[3−6] Although studies have reported the ubiquitous occurrence of BPA in environmental and human samples,[7−10] information on the occurrence of other bisphenol analogues is scarce. One study reported the occurrence of BPF in surface water, sewage, and sediments.[11] BPF was reported to occur in soft drinks at a mean concentration of 0.180 $\mu$g/L.[12] BPS has been reported to occur in canned foodstuffs at concentrations ranging from below the limit of quantitation (LOQ) to 175 ng/g.[13] BPB has been found not only in canned foodstuffs but also in human serum from Italy at concentrations on the order of several tens of nanograms per gram.[14,15]

Diet is considered an important source of human exposure to contaminants such as PCBs, perfluorochemicals, phthalates, and BPA.[16−19] In addition to diet, indoor air and indoor dust can be a significant source of human exposure to these contaminants. Elevated concentrations of BPA has been found in dust samples from daycare centers, homes, offices, and research laboratories, at concentrations on the order of several micrograms per gram; and ingestion of indoor dust has been demonstrated to be an exposure pathway to BPA in humans, especially children.[20−23] The 2010 joint Food and Agriculture Organization (FAO) and World Health Organization (WHO) expert meeting estimated soil/dust ingestion at 0.0001−0.03 $\mu$g BPA/kg body weight (bw)/day for the general population.[24] The contribution of dust ingestion to exposure to other bisphenols is not known.

BPA is a weak estrogen receptor agonist and its toxicity to laboratory animals has been well documented.[3,4,8] Considerable controversies surround the safety of BPA, and this compound is being replaced with alternatives in certain products. Some of the alternatives include compounds that are similar in structure to BPA. Limited studies have shown that BPS, BPB, and BPF possess toxicities (e.g., acute toxicity, genotoxicity, and estrogenic activity) similar to BPA.[12,13,25−27] One study showed that BPB and BPAF are potent human pregnane X receptor agonist.[28] Another study compared biodegradation of a series of bisphenols and showed that BPF and BPS are more resistant to degradation than BPA.[29] Given the similarity in molecular structure of the bisphenol analogues, it is expected that these compounds will have similar environmental behavior, toxicity, and exposure pathways to those of BPA, the prototype compound of this class.

In view of the potential for occurrence and toxicity, determination of bisphenol analogues in indoor dust and evaluation of human exposure arising from the ingestion of dust are imperative to the assessment of risks and for the development of strategies to mitigate exposures. In this study, eight bisphenol analogues, including BPA, BPF, BPB, and BPS, were measured in 156 indoor dust samples collected from houses, offices, laboratories, and dormitories in the United States (U.S.), China, Japan, and Korea. The baseline concentrations of bisphenol analogues in indoor dust were established, and geographic distribution patterns were explored. Bisphenol exposures via dust ingestion for various age groups (infants, toddlers, children, teenagers, and adults) were estimated on the basis of measured concentrations. This is the first study to report the occurrence of bisphenol analogues other than BPA in indoor dust from the U.S. and three Asian countries.

## MATERIALS AND METHODS

**Chemicals.** Bisphenol analogues, including BPA (purity: 97%), BPAF (97%), bisphenol AP [BPAP; 4,4′-(1-phenylethylidene)bisphenol; 99%], BPF (98%), bisphenol P [BPP; 4,4′-(1,4-phenylenediisopropylidene)bisphenol; 99%], BPS (98%), and bisphenol Z [BPZ; 4,4′-cyclohexylidenebisphenol; 98%] were purchased from Sigma-Aldrich (St. Louis,

Table 1. Elemental Composition and Ions Monitored for Several Bisphenol Analogues Analyzed in This Study

| target compounds | CAS no. | elemental composition | molecular weight | retention time (min) | MS/MS ions ($m/z$) |
|---|---|---|---|---|---|
| BPA | 80−05−7 | $C_{15}H_{16}O_2$ | 228.29 | 19.1 | 227 > 212 |
| BPAF | 1478−61−1 | $C_{15}H_{10}F_6O_2$ | 336.23 | 20.5 | 335 > 265 |
| BPAP | 1571−75−1 | $C_{20}H_{18}O_2$ | 290.36 | 20.9 | 289 > 274 |
| BPB | 77−40−7 | $C_{16}H_{18}O_2$ | 242.32 | 20.2 | 241 > 212 |
| BPF | 620−92−8 | $C_{13}H_{12}O_2$ | 200.23 | 17.2 | 199 > 93 |
| BPP | 2167−51−3 | $C_{24}H_{26}O_2$ | 346.46 | 22.6 | 345 > 330 |
| BPS | 80−09−1 | $C_{12}H_{10}O_4S$ | 250.27 | 13.3 | 249 > 108 |
| BPZ | 843−55−0 | $C_{18}H_{20}O_2$ | 268.35 | 21.2 | 267 > 173 |

MO). BPB (98%) was purchased from TCI America (Portland, OR). The molecular structures of bisphenol analogues are shown in Figure 1. $^{13}C_{12}$-labeled BPA (99%) was purchased from Cambridge Isotope Laboratories (Andover, MA). Formic acid (98.2%) was obtained from Sigma-Aldrich, and methanol (HPLC grade) was from Mallinckrodt Baker (Phillipsburg, NJ). Milli-Q water was provided through an ultrapure water system (Barnstead International, Dubuque, IA). The stock solutions of bisphenol analogues and $^{13}C_{12}$−BPA were prepared at 1 mg/mL in methanol. All stock solutions were stored at −20 °C. The working solutions were prepared from the stock solutions through serial dilution with methanol before use.

**Sample Collection.** Dust samples from the U.S. ($n$ = 38) were collected from houses, offices, and laboratories in Albany, New York, in 2006 and 2010. Dust from China ($n$ = 55) was collected from houses, student dormitories, offices, and laboratories in six cities, Beijing, Jinan, Guangzhou, Shanghai, Qiqihaer, and Urumchi, in 2010. Dust from Japan ($n$ = 22) was collected from houses, student dormitories, and offices in five cities, Kumamoto, Nagasaki, Fukuoka, Saitama, and Saga, in 2012. Dust from Korea ($n$ = 41) was collected from houses, offices, and laboratories in two cities, Ansan and Anyang, in 2012. Floor dust samples were obtained from vacuum cleaner bags in each of the sampling sites, with the exception of samples from China (which were obtained by sweeping the floor). All samples were sieved through a ≤2 mm sieve, homogenized, packed in clean aluminum foil, and stored at 4 °C until analysis.

**Sample Preparation.** Dust samples were extracted and analyzed by following the method described elsewhere, with some modifications.[30] Briefly, 0.05−0.1 g of sample was weighed and transferred into a 15 mL polypropylene conical tube (PP tube). After spiking with 20 ng $^{13}C_{12}$−BPA, a dust sample was extracted with a 5 mL solvent mixture of methanol and water (5:3, v/v) by shaking for 60 min. The mixture was centrifuged at 4500$g$ for 5 min (Eppendorf Centrifuge 5804, Hamburg, Germany), and the supernatant was transferred into a glass tube. The extraction step was repeated one more time with 3 mL solvent mixture, and the extracts were combined and concentrated to ∼4 mL under a gentle nitrogen stream. The sample solution was diluted to 10 mL with 0.2% formic acid (pH 2.5), and the extracts were purified with a RapidTrace SPE workstation (Caliper Life Sciences, Inc., Hopkinton, MA). The extract was loaded onto an Oasis MCX cartridge (60 mg/3 $cm^3$; Waters, Milford, MA), preconditioned with 5 mL of methanol and 5 mL of water. The cartridge was rinsed with 15 mL of 25% methanol in water and 5 mL of water. The target compounds were eluted with 5 mL of methanol, and the eluate was concentrated to 1 mL before liquid chromatography-tandem mass spectrometry (LC-MS/MS) analysis.

**Instrumental Analysis.** The concentrations of bisphenol analogues in sample extracts were determined by an Applied Biosystems API 2000 electrospray triple quadrupole mass spectrometer (ESI-MS/MS; Applied Biosystems, Foster City, CA), coupled with an Agilent 1100 Series HPLC (Agilent Technologies Inc., Santa Clara, CA) equipped with a binary pump and an autosampler. An analytical column (Betasil C18, 100 × 2.1 mm column; Thermo Electron Corporation, Waltham, MA), connected to a Javelin guard column (Betasil C18, 20 × 2.1 mm), was used for LC separation. The injection volume was 10 μL. The mobile phase was methanol and water at a flow rate of 0.3 mL/min with a gradient as follows: 0−2 min, 15% methanol; 2−5 min, 15−50% methanol; 5−8 min, 50% methanol; 8−20 min, 50−90% methanol; 20−21 min, 90−99% methanol; 21−25 min, 99% methanol; 25−30 min, 15% methanol. The negative ion multiple reaction monitoring (MRM) mode was used; the transitions of ions monitored are listed in Table 1. The MS/MS parameters were optimized by infusion of individual compounds into the mass spectrometer through a flow injection system (Supporting Information (SI) Table S1). Nitrogen was used as both a curtain and a collision gas.

**Quality Assurance and Quality Control (QA/QC).** The extraction efficiency of bisphenols from dust samples was examined by performing a third extraction (after the first two extractions) with 3 mL solvent mixture of methanol and water (5:3, v/v) for 20 randomly selected samples. BPA was detected in the third extraction in 4 of 20 samples, at concentrations ranging from 0.13 to 0.57 ng/mL. In comparison with the high concentrations of BPA detected in these dust samples (ranging from 203 to 2035 ng/g), the residual BPA found in the third extraction was only 1.1 ± 0.6% (mean ± SD) of the total concentration. BPF was detected in the third extraction in only one sample (0.58 ng/mL), and the residual BPF found in the third extraction was 1.0% of the total concentration in the corresponding dust sample (1890 ng/g). No other bisphenols were detected in the third extraction. With each set of 20 samples, a procedural blank, a spiked blank (containing water in place of dust), a pair of matrix spike samples (50 ng of BPA and 20 ng of other individual bisphenols), and duplicate samples were analyzed. Trace levels of BPA, BPF, and BPS (approximately 0.01, 0.3, and 0.01 ng/mL, respectively) were found in procedural blanks in some batches, and background subtraction was performed in the quantification of concentration in samples. Recoveries of bisphenols in spiked blanks ranged from 89 ± 10% (mean ± SD) for BPAF to 109 ± 6% for BPS (SI Table S2). Recoveries of bisphenols in spiked matrices ranged from 51 ± 11% for BPZ to 137 ± 45% for BPS (SI Table S2). Duplicate analysis of randomly selected samples showed a coefficient variation of <20% for all bisphenols. The LOQs were 0.5 ng/g for BPA, BPAF, BPAP, and BPS, 1.0 ng/g for BPB and BPP, and 2.0 ng/g for BPF and BPZ (SI Table S2), which were calculated from the value of the valid lowest

Table 2. Concentrations of Several Bisphenols (μg/g) in Indoor Dust Collected from the United States and Three Asian Countries

| | | BPA | BPAF | BPAP | BPB | BPF | BPP | BPS | BPZ | ∑BPs |
|---|---|---|---|---|---|---|---|---|---|---|
| U.S. | GM[a] | 1.70 | 0.00035 | 0.00038 | 0.00071 | 0.022 | 0.00071 | 0.62 | 0.0014 | 3.03 |
| (n = 38) | median | 1.60 | 0.00035 | 0.00035 | 0.00071 | 0.049 | 0.00071 | 0.63 | 0.0014 | 2.54 |
| | range | 0.20−9.38 | nd[b] | nd−0.0071[c] | nd | nd−0.24 | nd | 0.0056−25.5 | nd | 0.30−33.4 |
| | detection rate | 100% | 0% | 2.63% | 0% | 68.4% | 0% | 100% | 0% | |
| China | GM | 0.36 | 0.00035 | 0.00035 | 0.00099 | 0.021 | 0.00080 | 0.13 | 0.0014 | 0.83 |
| (n = 55) | median | 0.63 | 0.00035 | 0.00035 | 0.00071 | 0.038 | 0.00071 | 0.17 | 0.0014 | 1.11 |
| | range | nd−8.35 | nd | nd | nd−0.030 | nd−1.89 | nd−0.63[c] | 0.00083−12.6 | nd | 0.026−13.5 |
| | detection rate | 96.4% | 0% | 0% | 10.9% | 56.4% | 1.82% | 100% | 0% | |
| Japan | GM | 2.83 | 0.00046 | 0.00035 | 0.00071 | 0.045 | 0.00071 | 0.82 | 0.0014 | 4.06 |
| (n = 22) | median | 2.70 | 0.00035 | 0.00035 | 0.00071 | 0.057 | 0.00071 | 0.81 | 0.0014 | 3.79 |
| | range | 0.50−21.8 | nd−0.011 | nd | nd | nd−2.78 | nd | 0.25−2.55 | nd | 1.08−24.7 |
| | detection rate | 100% | 9.09% | 0% | 0% | 81.8% | 0% | 100% | 0% | |
| Korea | GM | 4.07 | 0.0039 | 0.00035 | 0.00071 | 0.50 | 0.00071 | 0.43 | 0.0014 | 6.21 |
| (n = 41) | median | 3.26 | 0.0048 | 0.00035 | 0.00071 | 0.45 | 0.00071 | 0.36 | 0.0014 | 4.48 |
| | range | 0.98−39.1 | nd−0.091 | nd | nd | nd −107 | nd | 0.090−26.6 | nd | 1.29−111 |
| | detection rate | 100% | 75.6% | 0% | 0% | 97.6% | 0% | 100% | 0% | |
| all | GM | 1.33 | 0.00069 | 0.00036 | 0.00080 | 0.054 | 0.00074 | 0.34 | 0.0014 | 2.29 |
| (n = 156) | median | 1.58 | 0.00035 | 0.00035 | 0.00071 | 0.096 | 0.00071 | 0.36 | 0.0014 | 2.66 |
| | range | nd−39.1 | nd−0.091 | nd−0.0071 | nd-0.030 | nd−107 | nd−0.63 | 0.00083−26.6 | nd | 0.026−111 |
| | detection rate | 98.7% | 21.2% | 0.64% | 3.85% | 74.4% | 0.64% | 100% | 0% | |

[a]GM = geometric mean. [b]nd = not detected. [c]BPAP and BPP were detected in only one sample.

acceptable calibration standard and a nominal sample weight of 0.1 g. The quantification was based on an external calibration method and corrected for the recoveries of the internal standard ($^{13}C_{12}$−BPA). A midpoint calibration check standard was injected as a check for instrumental drift in sensitivity after every 20 samples, and a pure solvent (methanol) was injected as a check for carry-over of bisphenols from sample to sample. Instrumental calibration was verified by injection of 10 calibration standards (ranging from 0.02 to 100 ng/mL), and the linearity of the calibration curve ($r$) was >0.99.

**Calculation of Daily Exposure Doses.** Based on the median and 95th percentile concentrations of bisphenols measured in indoor dust samples, we estimated the daily intake (EDI; ng/kg bw/day) of bisphenols through dust ingestion as shown in eq 1:[31−33]

$$\mathrm{EDI} = \frac{C \times \mathrm{DIR} \times \mathrm{IEF}}{\mathrm{BW}} \quad (1)$$

where $C$ is the concentration of bisphenols in dust samples (ng/g), DIR is dust ingestion rate (g/day), IEF is indoor exposure fraction (hours spent over a day in an indoor microenvironment), and BW is body weight (kg). We assumed an absorption efficiency of 100% for bisphenols from dust to systemic blood circulation. Details of the parameters used for EDI calculation are shown in SI Table S3.

**Statistical Analysis.** Statistical analysis was performed with Origin, Version 7.5, software. Concentrations below the LOQ were substituted with a value equal to LOQ divided by the square root of 2 for the calculation of geometric mean (GM). Differences between groups were compared by a one-way ANOVA with the Tukey test. A value of $p < 0.05$ was considered significant.

## RESULTS AND DISCUSSION

**Concentrations.** GM and median concentrations of bisphenols in indoor dust samples collected from four countries, the U.S., China, Japan, and Korea, are shown in Table 2. BPA was found in 99% of the 156 dust samples analyzed (the entire sample set) at concentrations ranging from <LOQ to 39.1 μg/g, with a GM of 1.33 μg/g. The highest concentration of BPA was found in dust from Korea (GM: 4.07, median: 3.26 μg/g); followed, in decreasing order, by Japan (2.83, 2.70 μg/g), the U.S. (1.70, 1.60 μg/g), and China (0.36, 0.63 μg/g; Table 2). BPS was detected in all dust samples at concentrations ranging from 0.00083 to 26.6 μg/g (GM: 0.34 μg/g). The GM and median concentrations of BPS in dust from Japan, the U.S., Korea, and China were 0.82 and 0.81 μg/g, 0.62 and 0.63 μg/g, 0.43 and 0.36 μg/g, and 0.13 and 0.17 μg/g, respectively (Table 2).

The finding of high concentrations of BPS in dust samples from Japan and the U.S. is consistent with those reported in our recent studies that showed high concentrations and detection frequencies of BPS in paper products and human urine collected from these two countries.[34,35] Japan ceased the application of BPA in thermal receipts in 2001 and made an effort to develop alternatives, including BPS.[36] A major manufacturer of thermal receipt papers in the U.S. announced replacement of BPA with BPS in 2006.[37,38] Similar to BPS, BPF is being proposed as a replacement for BPA in many commercial applications.[29,39] BPF was frequently found (74%) in the 156 dust samples analyzed (the entire sample set) and the concentrations were in the range of <LOQ to 107 μg/g (Table 2).

The highest GM concentration of BPF was found in dust from Korea (GM: 0.50, median: 0.45 μg/g), which was 1 order of magnitude higher than that detected in samples of the other three countries, although the differences were not statistically significant ($p > 0.05$, one-way ANOVA). The other five bisphenols were seldom detected in dust samples (detection rates for the entire sample set: all <4%, with the exception of BPAF, for which the detection rate was 21%). BPAF was detected in 76% of samples from Korea at concentrations ranging from <LOQ to 0.09 μg/g (GM: 0.005 μg/g). BPAF was found in 2 of 22 samples (9%) from Japan but not found in


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samples from the U.S. and China. A few samples from China (11%) contained BPB (range: <LOQ to 0.03, GM: 0.001 μg/g). BPAP and BPP were found in only one sample from the U.S. (0.007 μg BPAP/g) and China (0.63 μg BPP/g). BPZ was not found in any of the dust samples (Table 2).

The total concentration (sum of bisphenols: ∑BPs) of the eight target compounds was compared among the four countries studied. Dust samples from Korea contained the highest ∑BP concentrations (range: 1.29−111, GM: 6.21 μg/g), followed by those of Japan (1.08−24.7, 4.06 μg/g), the U.S. (0.30−33.4, 3.03 μg/g; $p < 0.05$), and China (0.026−13.5, 0.83 μg/g; $p < 0.001$; Table 2). The percentage composition of individual bisphenols to ∑BP concentrations is shown in Figure 2. BPA was the predominant analogue, representing 65

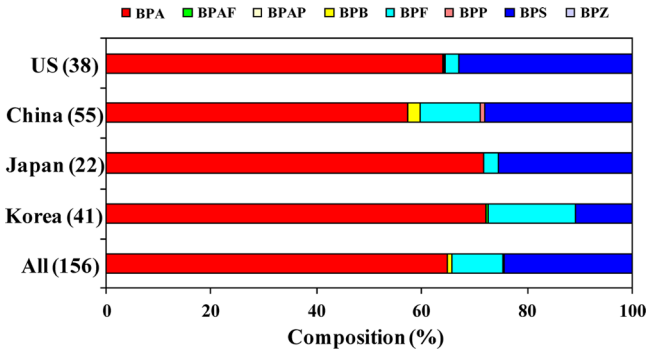


**Figure 2.** Composition profiles of several bisphenols in indoor dust. The numbers within parentheses on *x*-axis represent the number of samples analyzed.

± 26% (mean ± SD; for the entire sample set) of the total bisphenol concentrations in dust, followed, in decreasing order, by BPS (24 ± 23%), BPF (9.6 ± 17%), and BPB (0.8 ± 7.9%) (Figure 2). A similar pattern was observed for samples from each country studied, with the exception of samples from Korea, for which the percentage of BPS (11 ± 8.9%) was lower than that of BPF (17 ± 21%).

Bisphenol analogues are used in a wide variety of industrial and consumer products; for example, epoxy-based floorings, adhesives, paints, toys, compact discs, electronic equipments, and printed circuit boards. Volatilization and/or release of bisphenol analogues from these products are considered a source of contamination of indoor dust. The results from this study, as well as the results of human urine analyzed in our previous studies,[35,40] suggest that BPA is still the major bisphenol used in consumer products. Our results also suggest that, in consumer products, BPA is gradually being replaced by alternatives such as BPS and BPF.[29,39]

**Comparison of Bisphenol A Concentrations in Dust with Concentrations Reported Previously.** Several factors can affect the concentrations of BPA and its analogues in indoor dust, including floor type, room furnishings, ventilation, and time of sampling.[22] The concentrations of BPA measured in dust from various microenvironments (house, office, and laboratory) in this study were compared with those reported in earlier studies (Table 3). Due to the lack of information on the microenvironment for a few U.S. and Chinese dust samples, these samples were excluded from this comparison. Dust samples collected in student dormitories from China and Japan were categorized under the "house" group. BPA concentrations (mean: 3.72, median: 2.27 μg/g) in house dust from Japan were comparable with those (2.21, 2.04 μg/g) from Korea and approximately 1 order of magnitude higher than those (0.347, 0.075 μg/g) from China ($p < 0.01$). BPA concentrations in house dust from Japan and Korea were comparable or slightly higher than those found in samples from Massachusetts, U.S. (mean: 1.65, median: 0.821 μg/g),[20] and Antwerp, Belgium (2.00, 1.46 μg/g),[23] but 3−5 times higher than those found in

**Table 3. Comparison of BPA Concentrations (μg/g) in Indoor Dust Collected from Various Microenvironments in This Study with Concentrations Reported from Other Locations around the World**

| microenvironment | location | $n^a$ | sampling year | LOQ[b] | mean | median | range | detection rate (%) | reference |
|---|---|---|---|---|---|---|---|---|---|
| house | Massachusetts, U.S. | 118 | 1999−2001 | 0.2 | 1.65 | 0.821 | nd[c]-17.6 | 86 | 20 |
| daycare center | North Carolina, U.S. | 19 | 2000−2001 | 0.02 | N/A[d] | 0.031 | nd-0.156 | 53 | 21 |
| daycare center | Ohio, U.S. | 23 | 2000−2001 | 0.02 | N/A | 0.028 | nd-0.123 | 70 | 21 |
| house | Albany, New York, U.S. | 28 | 2006 | 0.0005[e] | 0.620 | 0.701 | nd-0.169 | 93 | 22 |
| house | Albany, New York, U.S. | 9 | 2010 | 0.0005[e] | 0.836 | 0.881 | 0.135−0.232 | 100 | 22 |
| house | Murray, Kentucky, U.S. | 7 | 2010 | 0.0005[e] | 0.52 | 0.242 | 0.172−0.213 | 100 | 22 |
| house | Antwerp, Belgium | 18 | 2008 | 0.003 | 2.00 | 1.46 | 0.535−9.73 | 100 | 23 |
| house | Bavaria, Germany | 12 | 2005 | N/A | 0.661 | 0.553 | 0.117−1.49 | 100 | 41 |
| house | four cities, China[f] | 19 | 2010 | 0.0005 | 0.347 | 0.075 | nd-2.55 | 89 | this study |
| house | five cities, Japan[g] | 20 | 2012 | 0.0005 | 3.72 | 2.27 | 0.496−12.3 | 100 | this study |
| house | two cities, Korea[h] | 16 | 2012 | 0.0005 | 2.21 | 2.04 | 0.980−4.19 | 100 | this study |
| office | Antwerp, Belgium | 2 | 2008 | 0.003 | 6.53 | 6.53 | 4.69−8.38 | 100 | 23 |
| office | three cities, China[i] | 7 | 2010 | 0.0005 | 1.22 | 0.787 | 0.117−3.49 | 100 | this study |
| office | two cities, Japan[j] | 2 | 2012 | 0.0005 | 16.6 | 16.6 | 11.4−21.8 | 100 | this study |
| office | two cities, Korea[h] | 14 | 2012 | 0.0005 | 13.2 | 8.54 | 2.31−39.1 | 100 | this study |
| laboratory | Murray, Kentucky, U.S. | 5 | 2010 | 0.0005[e] | 1.28 | 1.14 | 0.445−2.95 | 100 | 22 |
| laboratory | three cities, China[i] | 6 | 2010 | 0.0005 | 1.68 | 0.732 | 0.057−5.54 | 100 | this study |
| laboratory | two cities, Korea[h] | 11 | 2012 | 0.0005 | 6.40 | 4.43 | 1.07−25.9 | 100 | this study |

[a] $n$ = the number of samples. [b] LOQ = limit of quantification. [c] nd = not detected. [d] N/A = not available. [e] LOD (limit of detection) value presented here as LOQ is not available. [f] Four cities in China represent Beijing, Jinan, Guangzhou, and Urumchi. [g] Five cities in Japan represent Kumamoto, Nagasaki, Fukuoka, Saitama, and Saga. [h] Two cities in Korea represent Ansan and Anyang. [i] Three cities in China represent Beijing, Jinan, and Guangzhou. [j] Two cities in Japan represent Kumamoto and Saitama.


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**Table 4. Estimated Daily Intakes (EDI, ng/kg bw/day) of Bisphenols by Ingestion of Indoor Dust for Various Age Groups in the United States and Three Asian Countries**

| | U.S. | | | | China | | | | Japan | | | | Korea | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | BPA | BPF | BPS | ∑BPs | BPA | BPF | BPS | ∑BPs | BPA | BPF | BPS | ∑BPs | BPA | BPF | BPS | ∑BPs |
| | | | | | | | | Median | | | | | | | | |
| infants | 5.64 | 0.17 | 2.20 | 8.95 | 2.21 | 0.14 | 0.59 | 3.91 | 9.49 | 0.20 | 2.85 | 13.4 | 11.5 | 1.57 | 1.27 | 15.8 |
| toddlers | 7.91 | 0.24 | 3.09 | 12.6 | 2.61 | 0.16 | 0.69 | 4.61 | 11.2 | 0.24 | 3.37 | 15.8 | 13.5 | 1.86 | 1.50 | 18.6 |
| children | 2.18 | 0.07 | 0.85 | 3.46 | 0.86 | 0.05 | 0.23 | 1.51 | 3.67 | 0.08 | 1.10 | 5.17 | 4.44 | 0.61 | 0.49 | 6.10 |
| teenagers | 1.36 | 0.04 | 0.53 | 2.15 | 0.52 | 0.03 | 0.14 | 0.92 | 2.24 | 0.05 | 0.67 | 3.15 | 2.70 | 0.37 | 0.30 | 3.72 |
| adults | 1.08 | 0.03 | 0.42 | 1.72 | 0.44 | 0.03 | 0.12 | 0.78 | 1.88 | 0.04 | 0.57 | 2.65 | 2.28 | 0.31 | 0.25 | 3.13 |
| | | | | | | | | 95th Percentile | | | | | | | | |
| infants | 24.2 | 0.72 | 35.6 | 42.2 | 12.5 | 1.97 | 6.14 | 16.6 | 43.1 | 2.95 | 7.16 | 52.5 | 91.2 | 49.5 | 4.74 | 142 |
| toddlers | 34.0 | 1.01 | 49.9 | 59.2 | 14.8 | 2.32 | 7.25 | 19.7 | 51.0 | 3.49 | 8.46 | 62.1 | 108 | 58.4 | 5.60 | 167 |
| children | 9.37 | 0.28 | 13.8 | 16.3 | 4.85 | 0.76 | 2.38 | 6.44 | 16.7 | 1.14 | 2.77 | 20.3 | 35.3 | 19.1 | 1.84 | 54.9 |
| teenagers | 5.82 | 0.17 | 8.55 | 10.1 | 2.96 | 0.46 | 1.45 | 3.93 | 10.2 | 0.70 | 1.69 | 12.4 | 21.5 | 11.7 | 1.12 | 33.4 |
| adults | 4.66 | 0.14 | 6.84 | 8.11 | 2.49 | 0.39 | 1.22 | 3.30 | 8.56 | 0.59 | 1.42 | 10.4 | 18.1 | 9.81 | 0.94 | 28.1 |

samples from Albany, New York, U.S. (0.836, 0.881 μg/g for 2010 samples),[22] and Bavaria, Germany (0.661, 0.553 μg/g),[41] and 2 orders of magnitude higher than those found in North Carolina and Ohio, U.S. (median: 0.031 and 0.028 μg/g).[21] A similar trend was found for BPA concentrations in office dust samples from several countries (Table 3).

The concentrations of BPA in dust collected from offices were similar to the concentrations found in laboratories but considerably higher than the concentrations measured in homes. For example, BPA concentrations in dust collected from Korean offices were similar to those found in laboratories ($p > 0.05$) but significantly higher than those measured in homes ($p < 0.01$; Table 3). A similar pattern was found for dust from China. ∑BP concentrations in dust samples from offices, laboratories, and homes in the U.S., Japan, Korea, and China are shown in SI Figure S1. Previous studies have reported that dust collected from offices contains higher concentrations of contaminants than does dust from homes and cars.[17,42,43]

**Exposure to Bisphenols through Dust Ingestion.** The routes and sources of human exposure to bisphenols, especially for the BPA substitutes, are not well characterized. The significance of indoor dust ingestion as a pathway for human exposure to PBDEs, perfluorochemicals, phthalates, and BPA has been highlighted recently.[17,20−23] Several parameters such as age, time spent in certain microenvironments (home, office/laboratory, car, outdoor), and amount of dust ingestion can influence exposure doses to bisphenols.[23] For the estimation of daily intakes (EDIs) to bisphenols through dust ingestion, we categorized five age groups as infants (<1 year), toddlers (1−5 years), children (6−11 years), teenagers (12−19 years), and adults (≥20 years), according to the U.S. Environmental Protection Agency (EPA).[31,32] The body weights for various age groups in China were adopted from a previous study,[33] and the values were also applied in the calculation of EDIs for the populations in Japan and Korea. Except for BPA, BPF, and BPS, other five bisphenol analogues were seldom or not detected in dust and, therefore, we only assessed exposure doses of BPA, BPF, BPS, and ∑BPs. The median and 95th percentile values of bisphenol concentrations measured in this study were used in the calculation of EDIs for median and high exposure groups, respectively.

The EDIs of BPA, BPF, BPS, and ∑BPs via dust ingestion have been summarized in Table 4. Due to relatively small body weight and high dust ingestion rates (SI Table S3), toddlers are exposed to high doses of contaminants through dust ingestion. In the U.S., the median and 95th percentile values for daily intakes of ∑BPs in toddlers were 12.6 and 59.2 ng/kg bw/day, respectively; this is followed, in decreasing order, by infants (8.95 and 42.2 ng/kg bw/day), children (3.46 and 16.3 ng/kg bw/day), teenagers (2.15 and 10.1 ng/kg bw/day), and adults (0.42 and 8.11 ng/kg bw/day). A similar intake pattern was found for various age groups in the three Asian countries studied (Table 4). Among the four countries, the highest median daily intakes of ∑BPs (calculated for toddlers from median concentrations in dusts) were estimated for Korea (18.6 ng/kg bw/day), which were similar to those for Japan (15.8 ng/kg bw/day) and the U.S. (12.6 ng/kg bw/day), and approximately 4 times higher than those for China (4.61 ng/kg bw/day).

The daily intakes of BPA (median doses) were 2−4 times higher than those estimated for BPS, and 1−2 orders of magnitude higher than those estimated for BPF in all countries, except for Korea. In Korea, the median daily intakes of BPA were approximately 1 order of magnitude higher than those estimated for both BPS and BPF (Table 4).

The U.S. National Toxicology Program reported an oral BPA exposure from foods of 0.043−14.7 μg/kg bw/day for children and 0.008−1.5 μg/kg bw/day for adults.[44] Based on the leaching levels from consumer products and consumption of canned foods, BPA exposure has been estimated to be in the range from <1 to ∼5 μg/kg bw/day.[8,10] Assuming a total BPA intake of 1 μg/kg bw/day, dust ingestion was estimated to contribute 0.79%, 0.26%, 1.12%, 1.35% (based on median values) of the total intake for toddlers in the U.S., China, Japan, and Korea, respectively. Despite the high concentrations of BPA measured in dust, dust ingestion (on the order of ng/kg bw/day) appears to be a minor contributor to the total BPA intake (on the order of μg/kg bw/day). The EDI values calculated for BPA through dust ingestion were several orders of magnitude lower than the oral reference dose of 50 μg/kg bw/day established by the EPA and the European Food Safety Authority.[45,46]

It should be noted that several uncertainties are associated with our exposure assessment of bisphenol analogues via dust ingestion. Exposure doses of bisphenols from indoor dust can be affected by factors such as personal habits, occupation, dietary preferences, application of electronic equipments, and use of plastic products. The daily intake doses estimated in our calculation could be an underestimate of the actual exposure dose because we did not evaluate the dermal absorption

through skin or inhalation exposure from ambient air, which are considered important sources of exposure. Further, our discussion on bisphenol concentrations in various microenvironments is tempered by the small sample size from individual countries; thus, further studies with large sample sizes are needed.

In summary, high concentrations of several bisphenol analogues, on the order of several micrograms per gram, were detected in indoor dust collected from the U.S., Japan, and Korea. Total concentrations of bisphenols ($\sum$BPs) in dust samples from Korea (GM: 6.21 $\mu$g/g) were higher than those from Japan (4.06 $\mu$g/g), the U.S. (3.03 $\mu$g/g), and China (0.83 $\mu$g/g). BPA was the predominant bisphenol analogue, accounting for approximately 65% of the total bisphenol concentrations in dust. BPS and BPF were prevalent in dust samples, whereas other bisphenol analogues were detected rarely. Although high concentrations of bisphenols were measured in dust, the contribution of dust ingestion to total intake appears to be small. To our knowledge, this is the first study that describes the occurrence of several bisphenols, especially BPS, BPF, and BPB, in indoor dust.

## ■ ASSOCIATED CONTENT

### Supporting Information

Additional information as noted in the text (three tables and one figure and some text material). This material is available free of charge via the Internet at http://pubs.acs.org.

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### Notes

The authors declare no competing financial interest.

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