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High Levels of Bisphenol A in Paper Currencies from Several Countries, and Implications for Dermal Exposure

Chunyang Liao and Kurunthachalam Kannan*

Wadsworth Center, New York State Department of Health, and Department of Environmental Health Sciences, School of Public Health, State University of New York at Albany, Empire State Plaza, P.O. Box 509, Albany, New York 12201-0509, United States

Supporting Information

ABSTRACT: The sources of human exposure to bisphenol A (BPA) are not well characterized. Little is known about the presence of BPA in paper products, especially in paper currencies, and its implications for human exposure. In this work, paper currencies from 21 countries ($N = 156$) were analyzed for BPA, which was measured in 19 mm punches taken from three spots on the paper currencies. BPA was found in all paper currencies at concentrations ranging from 0.001 to 82.7 μg/g (mean 4.94; median 1.02) and the concentrations in samples taken from the middle portion of the currencies were higher than those taken from peripheral portions. We also examined the transfer of BPA from thermal receipt paper to currencies by placing currencies in contact with thermal receipt papers for 24 h in a wallet. Concentrations of BPA dramatically increased after 24 h of contact with thermal receipt papers, suggesting that thermal receipt paper is an important source of BPA in paper currencies. The estimated daily intake of BPA through dermal absorption from handling paper currencies was on the order of a few nanograms per day.

INTRODUCTION

Bisphenol A (2,2-bis(4-hydroxyphenyl)propane; BPA) is one of the high production volume chemicals, with over 8 billion pounds produced every year worldwide.[1,2] BPA is used mainly as a monomer in the production of polycarbonate plastics and epoxy resins.[1] Polycarbonate plastic is used in a variety of consumer products, including baby and water bottles, sports equipment, medical devices, dental fillings, sealants, and household electronics. Epoxy resin is commonly used as a protective lining for canned foods and beverages and as a coating on metal lids for glass jars and bottles.[3] Due to its widespread use in many industrial and commercial products, BPA has frequently been detected in environmental matrices, including air, water, sewage sludge, soil, dust, foodstuffs, and drinks as well as in human samples.[4−6]

A number of studies have demonstrated that BPA can mimic the body's endogenous hormones, by binding to estrogen receptors, making BPA an endocrine disruptor.[4,7,8] Prenatal BPA exposure has been linked to changes in vaginal and uterine histology and altered ovarian morphology in laboratory animals.[9−12] Prenatal exposure to BPA at doses of 0.1−1000 μg/kg body weight, during critical periods of cell differentiation, caused long-term reproductive and carcinogenic effects.[13] BPA may be a causal agent in cardiovascular diseases, diabetes, metabolic disorders, prostate cancer, neurobehavioral and neuroendocrinal effects, and immune system alterations.[5,14−16] Urinary concentrations of BPA reported by the National Health and Nutrition Examination Survey (NHANES) from 2003 to 2006 showed an association with heart diseases in the adult U.S. population, including heart attacks and angina.[17] Epidemiological studies also have found that high exposure to BPA is associated with male sexual dysfunction.[18,19]

BPA has been detected in canned foods and fresh foods, and diet has been considered the major source of human exposure to BPA.[20−24] Additionally, inhalation and dermal routes are presumed to be important sources of exposure, especially in occupational settings.[4] BPA has been reported to occur in the general populations in the U.S. and in several other countries.[25−27] It is estimated that more than 90% of the US population is chronically exposed to BPA [25] and this statistic can be applied to people living in other countries around the world.[28]

BPA has been used as a component in the production of thermal papers (as a weakly acidic color developer) for more than 50 years.[29,30] Due to its low cost, thermal papers containing BPA are still produced in massive quantities for a wide variety of commercial applications, including daily cash register receipts, luggage tags, books, faxes, and labels. BPA has been detected in thermal paper at extremely high levels of up to 3−22 g/kg.[29−31] Studies have shown that, because BPA is applied on thermal papers as a powdery film, it can be absorbed into human skin when handling receipts.[30,31] BPA can be transferred from receipts to paper currencies every time a receipt is placed near the currency in a cash register or wallet or whenever one handles a receipt before handling money. Although several studies have estimated sources of BPA exposure in humans, information on BPA contamination in paper currencies is still limited. In the present work, we investigated BPA concentrations in paper currencies collected

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Table 1. BPA Concentrations (μg/g) in Different Regions of the Paper Currencies from Various Countries

country	no.	LLC[a]	M[b]	URC[c]	mean	total amount in currency bill (μg)
USA	#1	0.751	1.47	0.726	0.981	0.863
	#2	10.3	2.86	3.68	5.63	5.70
	#3	2.32	2.14	1.86	2.11	2.84
	#4−1[d]	0.036	0.019	0.024	0.026	0.253
	#4−2[d]	0.013	0.008	0.008	0.010	0.094
Canada	#1−1	5.38	10.4	2.60	6.13	6.02
	#1−2	5.38	8.10	3.47	5.65	5.61
Europe	#1	1.56	3.08	0.578	1.74	1.06
Czech Republic	#1	23.0	40.5	24.3	29.2	22.2
Russia	#1	1.18	6.87	1.97	3.34	2.66
Turkey	#1	0.001	0.001	0.001	0.001	0.001
Australia	#1	6.29	12.0	9.49	9.26	5.14
Brazil	#1	67.5	81.2	82.7	77.1	58.7
	#2	20.3	38.8	23.3	27.5	18.9
	#3	5.20	2.00	3.61	3.61	2.92
Egypt	#1	<LOD	0.005	<LOD	0.002	0.002
South Africa	#1	0.841	0.624	0.535	0.667	0.509
China	#1	0.338	10.8	9.90	7.02	3.51
	#2	0.999	6.15	3.44	3.53	1.58
	#3	1.18	17.3	3.87	7.46	5.59
	#4	0.708	0.772	0.546	0.675	0.412
	#5	0.943	1.07	1.08	1.03	0.758
	#6	0.364	0.573	0.438	0.458	0.346
India	#1	0.525	0.547	0.413	0.495	0.347
	#2	0.170	0.520	0.379	0.357	0.223
	#3	0.551	1.58	5.86	2.66	1.47
	#4−1	0.365	0.573	0.387	0.442	0.355
	#4−2	0.208	0.093	0.145	0.149	0.139
	#4−3	0.209	0.244	0.641	0.365	0.337
Japan	#1	0.494	0.509	0.573	0.525	0.421
Korea	#1	0.470	1.31	0.253	0.678	0.497
	#2−1	2.42	7.04	3.57	4.34	3.80
	#2−2	3.10	8.51	2.44	4.68	3.77
	#2−3	3.13	2.55	1.22	2.30	1.98
	#3	4.22	6.53	4.35	5.03	3.64
	#4−1	1.27	3.16	1.04	1.82	1.44
	#4−2	1.19	1.76	1.07	1.34	1.26
	#4−3	0.624	1.91	1.31	1.28	1.08
Kuwait	#1	0.597	0.148	0.617	0.454	0.325
	#2	2.23	3.85	2.45	2.84	2.39
Malaysia	#1	3.14	3.02	3.05	3.07	1.95
	#2−1	0.029	0.059	<LOD	0.029	0.038
	#2−2	0.052	0.058	<LOD	0.037	0.026
	#3−1	0.028	0.135	0.050	0.071	0.077
	#3−2	0.062	0.036	0.062	0.054	0.038
	#4	1.33	1.34	0.668	1.11	0.654
	#5	3.25	4.11	1.08	2.81	1.52
	#6	1.21	2.34	0.763	1.44	0.74
	#7	23.9	41.3	14.3	26.5	15.8
Philippines	#1	0.041	0.257	0.030	0.110	0.114
	#2−1	0.012	<LOD	<LOD	0.004	0.024
	#2−2	0.060	0.004	<LOD	0.021	0.016
	#3	0.324	0.508	0.385	0.406	0.353
Singapore	#1	2.38	2.40	1.55	2.11	1.24

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Table 1. Continued

country	no.	LLC[a]	M[b]	URC[c]	mean	total amount in currency bill (μg)
	#2	14.1	15.2	14.4	14.6	9.40
Thailand	#1	0.067	0.227	0.040	0.111	0.098
	#2	0.007	0.109	0.005	0.040	0.035
	#3−1	0.036	0.040	<LOD	0.025	0.020
	#3−2	<LOD	0.020	<LOD	0.007	0.006
Vietnam	#1	0.296	0.679	0.465	0.480	0.305
	#2	0.062	0.064	0.029	0.052	0.039
	#3	21.9	20.8	14.3	19.0	11.9
	#4	15.7	15.4	18.9	16.7	9.59
UAE	#1	3.05	5.01	4.56	4.20	3.17
mean of all		4.18	6.26	4.37	4.94	3.54

[a] LLC = lower left corner. [b] M = middle. [c] URC = upper right corner. [d] Two sets of punches were taken from the same currency bill.

from 21 countries and estimated potential exposure doses of BPA from handling paper currencies. In addition, potential for the transfer of BPA from thermal receipt paper to currency bills was examined, to enable understanding of sources of BPA in currencies.

MATERIALS AND METHODS

Sample Collection. Currency bill samples (N = 52) were collected from 21 countries, including the United States, Canada, Czech Republic, Russia, Turkey, Australia, Brazil, Egypt, South Africa, China, India, Japan, Korea, Kuwait, Malaysia, the Philippines, Singapore, Thailand, Vietnam, and United Arab Emirates (UAE) from March 2010 to January 2011. Paper currencies were purchased mainly from currency exchange stands in international airports in the United States, Japan, and Korea. When available, we obtained "fresh" currencies. The currency samples were graded as "fresh" and "used" based on the appearance and degree of dirtiness. All of the currency samples were expected to be in circulation, although the "fresh" currencies should be handled less frequently than "used" currencies. Currency information, including name, value, serial number, dimensions, and the release date are summarized in Table S1 (Supporting Information). All samples were stored at −20 °C until further analysis.

Sample Preparation. Paper currencies were analyzed for BPA by following a method similar to that described earlier, with some modifications.[32] Three circle punches (19 mm diameter) were taken from the lower left corner (denoted as LLC), middle (M), and upper right corner (URC) of each paper currency (see Figure S1) using a punch (Uchida Corp., Torrance, CA). It is presumed that the lower left and upper right corners are frequently rubbed or pulled and that the middle region is relatively less touched. After accurately weighing each of the punches (~0.0229 g), samples were cut into small pieces and transferred into a 15 mL polypropylene conical tube (PP tube). Samples were spiked with 20 ng of $^{13}C_{12}$-bisphenol A ($^{13}C_{12}$−BPA, 99%; Cambridge Isotope Laboratories, Andover, MA) as an internal standard. Five milliliters of methanol (HPLC grade, J.T. Baker, Phillipsburg, NJ) were added to the sample, and the mixture was shaken in an orbital shaker (Eberbach, Ann Arbor, MI) at 250 oscillations/min for 30 min. After centrifugation at 4500 × g for 3 min (Eppendorf Centrifuge 5804, Hamburg, Germany), an aliquot was quantitatively transferred into a new PP tube. The residue was extracted two more times with 3.5 mL volumes of methanol by shaking. The aliquots of methanol were combined and concentrated to 10 mL under a gentle stream of nitrogen. One milliliter of the aliquot was transferred into a vial and analyzed by high performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS).

Instrumental Analysis. Separation and detection of BPA were performed with an Agilent 1100 series HPLC coupled with an Applied Biosystems API 2000 electrospray MS/MS (Applied Biosystems, Foster City, CA). Ten μL of the extract was injected onto an analytical column (Betasil C18, 100 × 2.1 mm column; Thermo Electron Corporation, Waltham, MA), connected in a series with a Javelin guard column (Betasil C18, 20 × 2.1 mm; Thermo Electron Corporation). The mobile phase comprised methanol and water at a gradient starting from 25% methanol to 99% methanol in 4 min and was held for 10 min before it was reversed to initial conditions. The mobile phase flow rate was set at 300 μL/min, and the column temperature was set at 25 °C. The MS/MS was operated in an electrospray negative ionization mode. Instrumental parameters were optimized to transmit the $[M-H]^-$ ion before fragmentation to one or more product ions. Cone voltage was −30 V, and collision energy was 25 V. Capillary voltage was kept at −4.5 kV, and desolvation temperature was 400 °C. Data were acquired using multiple reaction monitoring (MRM) for the transitions of 227 > 212 for BPA (97%; Sigma-Aldrich, St. Louis, MO), and 239 > 224 for $^{13}C_{12}$−BPA.

Quality Assurance and Quality Control (QA/QC). The accuracy and precision of the analytical method were determined by the spiking of known concentrations of BPA into the sample matrices (paper) and passing them through the entire analytical procedure. A procedural blank, containing water in place of paper currency, was analyzed with each batch of 20 samples as a check for interferences or laboratory contamination; BPA was not found in procedural blanks. Ten samples were randomly selected, and a fourth extraction with 3 mL of methanol (after the first three extractions) was carried out to confirm that extraction of BPA from currency samples was complete. BPA was not detected in the fourth extract of the currency samples analyzed. The limit of detection (LOD) was 0.5 ng/g, which was calculated as twice the value of the lowest acceptable concentration in calibration standard. The recovery of BPA spiked at 40 ng level into selected samples was 105 ± 14.3% (mean ± SD). The recovery of $^{13}C_{12}$−BPA spiked into each currency samples was 66 ± 18%. Quantification was by the isotope-dilution method, based on the responses of $^{13}C_{12}$−BPA spiked into each currency sample. Instrumental drift in sensitivity was checked by duplicate injections of samples and the running of a continuing calibration

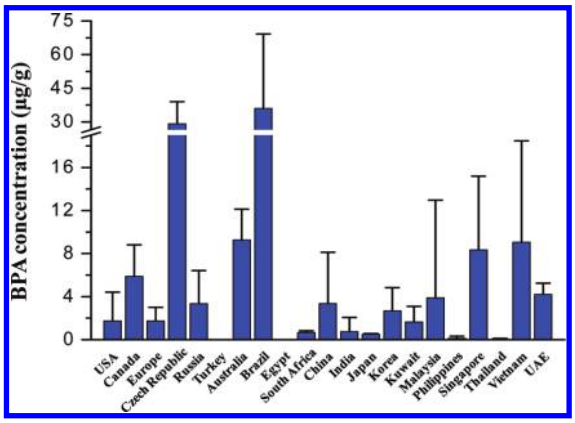

Figure 1. Comparison of BPA concentrations (mean ± SD, μg/g) in paper currencies from various countries.

check standard after every ten samples. Instrumental calibration was verified by injecting 10 μL of 0.02, 0.05, 0.1, 0.2, 0.5, 1, 5, 10, 50, and 100 ng/mL standards of BPA and $^{13}C_{12}$–BPA at the beginning and at the end of analysis of every batch of 20 samples. The regression coefficient (r) of the calibration curve was >0.99. For samples with responses lower than the linear range of calibration, extracts were concentrated 10-fold and reanalyzed. For statistical analysis, values below the LOD were set at zero. Statistical differences were tested with one-way ANOVA and the Tukey test (Origin 7.5, OriginLab Corporation, MA) at $\alpha = 0.05$, unless otherwise stated.

RESULTS AND DISCUSSION

BPA Concentrations. All 52 paper currency bill samples, collected from 21 countries, contained BPA at concentrations ranging from 0.001 to 82.7 μg/g (Table 1). The highest BPA concentration of 82.7 μg/g was found in a currency bill collected from Brazil (Real). As noted above, we analyzed 19 mm punches taken from three spots on the paper currencies: LLC, M, and URC. Four of the 52 currency bills did not contain BPA at the URC. These four currencies, which contained relatively low concentrations (<0.1 μg/g) of BPA in the middle portion, were from Egypt, Thailand, Malaysia, and the Philippines. In general, the URC of each paper currency is frequently handled by humans, which may explain the lower levels of BPA in that corner (i.e., frequent transfer of BPA from currency to finger). BPA concentrations in the middle portion of the currencies (mean ± SD; 6.26 ± 13.2 μg/g) were greater than those found in the LLC and URC (4.18 ± 9.93 μg/g and 4.37 ± 11.4 μg/g, respectively), although the differences were not statistically significant (one-way ANOVA, $p > 0.05$). The middle potion of currency is likely to be in contact with small thermal receipts, which explains higher concentrations of BPA in samples taken from the middle portion of the currencies than those taken from peripheral portions.[30,31] The concentrations of BPA have also been reported on the basis of area and the results are shown in the Supporting Information (Table S2).

BPA concentrations in all three punches of currency bills from each country, including the replicate samples, were averaged (Figure 1). The mean and median concentrations of BPA in paper currencies from Brazil (36.1 and 23.3 μg/g) were higher than those (3.40 and 0.763 μg/g) found in currencies from other countries; concentrations of BPA in currency bills from Brazil

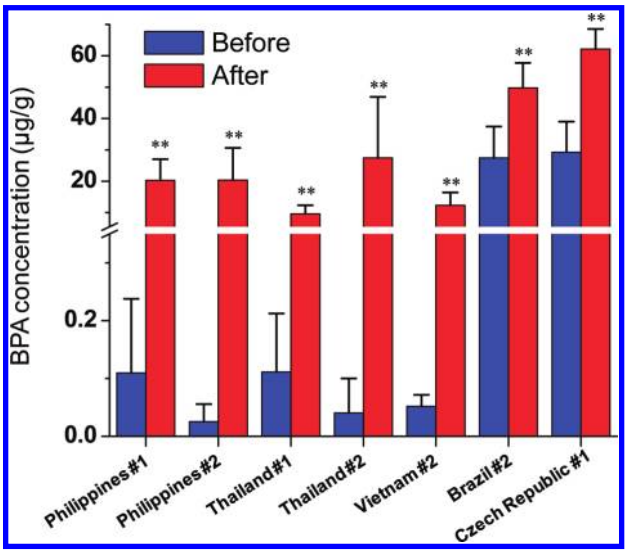

Figure 2. Comparison of BPA concentrations in selected paper currencies, before and after 24 h of contact with thermal receipt paper.

were followed by those of the Czech Republic (29.2 and 24.3 μg/g), Australia (9.26 and 9.49 μg/g), Vietnam (9.05 and 7.49 μg/g), and Singapore (8.34 and 8.25 μg/g). The only available report on BPA concentrations in paper currencies was by Washington Toxics Coalition (Seattle, WA), which reported BPA levels in U.S. dollar bills.[30] In that report, BPA was measured in 21 dollar bills, and concentrations were reported to range from 0.12 μg/g to 11 μg/g. BPA concentrations found in the U.S. dollar bills in our study were comparable to or slightly lower than those reported by Washington Toxic Coalition.[30]

One potential source of BPA in paper currencies is the thermal paper used in inkless cash register receipts. BPA, in the form of a powdery film on thermal receipt papers, can be transferred from receipts to paper currencies each time the receipt is placed near the currency or when a receipt is touched before handling currency. To examine the transfer of BPA from thermal receipt papers to currency bills, we selected seven paper currency bill samples (two from the Philippines, two from Thailand, one from Vietnam, one from Brazil, and one from Czech Republic) and placed them aside (in contact with) seven thermal receipt papers with known concentrations of BPA in a wallet for 24 h. The thermal papers were in contact with the currency bills on both of the surfaces for 24 h. The concentrations of BPA in thermal receipt papers ranged from 6500 to 10 190 μg/g. After 24 h, three spots from each of the currency bills were punched and analyzed. As shown in Figure 2, for the currency bills with low initial concentrations of BPA (from the Philippines, Thailand, and Vietnam), BPA concentrations increased by 100- to 1000-fold after contact with thermal receipt papers for 24 h in a wallet (mean ± SD, 18.0 ± 11.1 μg/g after contact versus 0.068 ± 0.077 μg/g before contact; paired t-test, $p < 0.01$). For the currency bills with high initial concentrations of BPA (from Brazil and Czech Republic), BPA concentrations increased by 2-fold (56.0 ± 9.36 μg/g after contact versus 28.4 ± 8.85 μg/g before contact; paired t-test, $p < 0.01$). These results clearly suggest the transfer of BPA from receipt paper to currency bills and that thermal receipt paper is a major source of BPA found in paper currency bills. Furthermore, the rate of BPA transfer from thermal receipt paper to currency bills can vary depending on the

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Table 2. Estimated Daily Intake Values (ng/day) of BPA, by Way of Handling Paper Currencies, by the General Population and Occupationally Exposed Individuals, in 21 Countries

	general population exposure			occupational exposure		
country	mean	median	95th percentile	mean	median	95th percentile
USA	0.102	0.044	0.330	1.02	0.436	3.30
Canada	0.342	0.313	0.572	3.42	3.13	5.72
Europe	0.101	0.091	0.170	1.01	0.909	1.70
Czech Republic	1.70	1.41	2.26	17.0	14.1	22.6
Russia	0.194	0.115	0.371	1.94	1.15	3.71
Turkey	0.0001	0.0001	0.0001	0.0007	0.0007	0.0008
Australia	0.538	0.551	0.683	5.38	5.51	6.83
Brazil	2.10	1.36	4.77	21.0	13.6	47.7
Egypt	0.0001	0	0.0002	0.0009	0	0.0025
South Africa	0.039	0.036	0.048	0.387	0.363	0.476
China	0.195	0.060	0.686	1.95	0.601	6.86
India	0.043	0.023	0.129	0.433	0.233	1.29
Japan	0.031	0.030	0.033	0.305	0.296	0.329
Korea	0.156	0.126	0.405	1.56	1.26	4.05
Kuwait	0.096	0.083	0.203	0.958	0.827	2.03
Malaysia	0.227	0.044	1.22	2.27	0.443	12.2
Philippines	0.008	0.002	0.026	0.079	0.021	0.256
Singapore	0.485	0.479	0.873	4.85	4.79	8.73
Thailand	0.003	0.002	0.009	0.027	0.016	0.094
Vietnam	0.526	0.435	1.24	5.26	4.35	12.4
UAE	0.244	0.265	0.288	2.44	2.65	2.88

original concentration (or level of saturation) in currency bills and receipts. Although thermal receipt paper is a source of BPA contamination of paper currencies, use of BPA as an ink developer in paper currencies cannot be ruled out. High levels of BPA (up to 2.2% by weight) have been reported in thermal receipt papers.[29−31] Because the BPA used in thermal paper is not chemically bound, it can be easily transferred from thermal receipt papers to other objects, including paper currencies.[30] It has been estimated that approximately 30% of thermal papers enter recycling streams of municipal wastepaper.[33] Recycling of thermal paper can introduce BPA into the cycle of paper production.[34] Therefore, it is possible that paper currency bills may initially contain small amounts of BPA. Additionally, BPA found in indoor dust [32,35,36] can be a source of contamination in paper currencies.[35] No clear difference was found, however, in the concentrations of BPA between "fresh" and "used" paper currencies. This may be explained by the fact that a currency bill that looks "fresh" may have been used a few times and a certain amount of BPA has been transferred to the bill.

Concentrations of BPA measured in paper currency samples from the United States, China, and the Philippines, released at various time periods, were examined to enable assessment of temporal variations in residue levels. The mean BPA concentrations in the U.S. dollar bill released in 2003, 2006, and 2009 were 5.63, 1.54, and 0.018 μg/g, respectively (Table 1). A similar decreasing trend was observed in BPA concentrations with the latest release date of currency samples from China and the Philippines. This may suggest prolonged exposure of older currency bills in circulation to BPA through contact with thermal receipt papers.[30] Most of the paper currency bills analyzed collected in this study were "used" and only seven were "fresh" samples. Although one might assume that there exists differences in BPA concentrations between "fresh" and "used" currency bills (because of contact with thermal receipt papers or handling by humans), samples from Singapore and Thailand showed no marked difference in BPA concentrations between "fresh" and "used" paper currencies.

The relationship between the value of the paper currency and the concentration of BPA was examined, and no notable relationship was found. BPA concentrations in currencies from the United States, Brazil, India, Korea, and Thailand generally decreased with the increasing value of the paper currency, but an opposite trend was observed for the currencies from Kuwait, Malaysia, and the Philippines (Table 1). Currency samples from several countries, including the United States, Canada, India, Korea, Malaysia, the Philippines, and Thailand, were analyzed in replicates (several punches from the same currency bill), and concentrations of BPA were found to be similar among replicate samples. For example, mean BPA concentrations in two sets of punches of Canadian dollar bills were comparable (6.13 and 5.65 μg/g, with a relative standard deviation (RSD) < 10%). A similar pattern was found for currencies from India and Korea. In contrast, replicate analysis of paper currencies of same bills from the United States, the Philippines, and Thailand yielded variable BPA concentrations. The amount of printing ink used to draw the images on one currency can vary greatly depending on the design, and this could contribute to the variation in the amount of BPA found in currencies.[37] This situation is more pronounced in currencies containing relatively low levels of BPA, such as currencies from the United States, the Philippines, and Thailand. In contrast, this could indicate that BPA contamination in paper currency originates mainly from sources other than the printing ink.[30] Overall, our results suggest that BPA contamination is widespread in paper currencies and that the concentrations vary

widely. Several factors appear to influence the concentrations of BPA in paper currencies.

Human Exposure to BPA via Handling of Paper Currencies. Because BPA in paper currencies exists as a free monomer that is mobile and transferable to objects that come in contact with each other, there is a potential for skin uptake/dermal absorption from handling of paper currencies. BPA absorbed by skin from the touching of products such as thermal cash register receipts and paper currencies can be a source of human exposure.[30,31] Little is known about human exposure to BPA through the handling of paper products.[31,38−40] Several parameters, such as method and frequency of handling of currency bills (e.g., keeping currency bills at standard pressure, pulling currency bills through fingers, rumpling currency with the whole hand), skin properties, and personal hygiene, must be taken into consideration for the estimation of intake of BPA through handling of paper currencies.[30,31] Nevertheless, considering the high concentrations of BPA found in paper currencies and the high dermal absorption coefficient of BPA,[38,40] the handling of paper currencies can be a source of human exposure, especially for people working at cash registers. Transfer of BPA from thermal receipt paper to human skin has been estimated with standard assumptions.[31] It was reported that the holding of a thermal receipt paper with 15.2 g/kg BPA (mean value) for 5 s resulted in the transfer of 1636 ng BPA on the surface of the hand.[31] Accordingly, we can calculate the paper-to-skin transfer rate as k = 1636 ng/ (15.2 g/kg·5s) = 21 522.4 ng/s. It has been estimated that 27% of BPA found on the surface of the skin penetrates and reaches the bloodstream within 2 h.[31] Another in vitro study showed that more than 27% of the BPA was metabolized/biotransformed in human skin.[40]

For the estimation of human exposure, we assumed that the general population handles paper currencies twice a day. This assumption is crude, as the frequency of handling of paper currencies can vary considerably among individuals. Individuals working at cash registers or banks (occupational exposure) handle paper currencies frequently on a daily basis. We assumed that the occupationally exposed individuals handle paper currencies 20 times a day. On the basis of the mean, median, and 95th percentile concentrations of BPA measured in our paper currency samples, we estimated the daily intake (EDI; ng/day) of BPA as shown in eq 1:

$$\mathrm{EDI} = k \times C \times \mathrm{HF} \times \mathrm{HT} \times \mathrm{AF}/10^6 \quad (1)$$

where k. is the paper-to-skin transfer coefficient of BPA (calculated as 21522.4 ng/s); C is the concentration of BPA in paper currency (μg/g); HF is the handling frequency (times/day) (2 and 20 times/day for the general population and occupationally exposed individuals, respectively); HT is the handling time of paper currency and is assumed as 5s for each handling; and AF is the absorption fraction of BPA by skin, which is 27%.

The mean, median, and 95th percentile values for daily intakes of BPA, through the handling of paper currencies, are summarized in Table 2. The estimated intake of BPA through dermal absorption via handling of paper currencies by the general population (calculated from the mean concentrations in paper currencies) ranged from 0.0001 ng/day (Turkey) to 2.10 ng/day (Brazil); the corresponding values for the occupationally exposed individuals were in the range of 0.0007 ng/day (Turkey) and 21.0 ng/day (Brazil). The estimated daily intake (EDI) values for BPA from paper currencies were 10-fold lower than those reported from exposures due to dust ingestion in the United States.[32] The National Toxicology Program of the United States reported BPA exposure ranges of 0.008−1.5 μg/kg bw/day for adults in the United States.[3] Similarly, several studies have generated estimates for current BPA exposures based on leaching levels from consumer products and from the consumption of canned foods. Studies have estimated that human exposures, in general, range from <1 to approximately 5 μg/kg bw/day.[5,41] The EDI values calculated for BPA from paper currencies were several orders of magnitude lower than the oral reference dose of 50 μg/kg bw/day established by the United States Environmental Protection Agency and the European Food Safety Authority.[42,43]

It should be noted that there are several uncertainties in our estimate of BPA exposures from paper currencies. A dose−response relationship for transfer coefficient of BPA from paper products to skin has not been established. The EDI can be higher when paper currencies are handled with wet or greasy fingers or by hand-to-mouth contact.[31] For occupationally exposed individuals, the duration of exposures can be much higher than what was used in our calculation. Inhalation during handling of currency papers can be an important source of BPA exposures, and this was not estimated.[30,31] Further studies are needed to assess the release of BPA from paper currencies into air during handling. The EDI of BPA from paper currencies in occupationally exposed individuals with the highest exposure scenario (e.g., Brazil currency) can be as high as 21.0 ng/day (0.35 ng/kg bw/day, based on 60 kg body weight for adults). Studies have reported adverse endocrine disruptive effects of BPA at doses as low as a few tens to a few hundreds of ng/kg bw/day.[44]

In summary, a few ng/g to several tens of μg/g concentrations of BPA were measured in paper currency samples from several countries. BPA contamination in paper currency was not dependent on the value, circulation time, or amount of ink. Contact of paper currencies with thermal receipt papers is considered a major source of BPA contamination. Although high levels of BPA were measured in paper currencies, human exposure through dermal absorption appears to be minor. Further studies are needed to assess exposure of cashiers and others who come into frequent contact with paper currencies. Further, release of BPA from paper currencies into the air and subsequent inhalation exposures should be examined.

■ ASSOCIATED CONTENT

Supporting Information. A table showing details of paper currencies used in the analysis of bisphenol A (BPA) residues and a table showing BPA concentrations in paper currencies by area (ng/cm^2) and a figure showing pictures of paper currencies with punches taken for BPA analysis. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Tel: 1-518-474-0015; Fax: 1-518-473-2895; E-mail: kkannan@wadsworth.org.

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