


Detection of Endocrine Disruptor Bisphenol A and Bisphenol S in Bangladeshi Thermal Paper Receipts [†]

Santona Khatun ^{1,*}, Hridita Ferdous ¹, Shahriar Hossain ¹, Siddika Sultana ¹, Inja Choi ²  and Young-Sun Lee ²

¹ Environment and Social Development Organization (ESDO), House 8/1, Level 5, Block C, Lalmatia, Dhaka 1207, Bangladesh

² Wonjin Institute for Occupational and Environmental Health, 53, Sagajeong-ro 49-gil, Jungnang-gu, Seoul 02221, Korea

* Correspondence: santona@esdo.org

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Abstract: Bisphenol analogues such as bisphenol A (BPA), a high production volume chemical identified as an endocrine disruptor and toxic to reproduction, are mainly used in thermal receipt papers as a color developer. Due to its effects on health and legal restrictions, BPA is increasingly replaced by other bisphenols such as Bisphenol S (BPS). In this study, BPA and 4 alternatives including BPS, Bisphenol F (BPF), Bisphenol B (BPB), and Bisphenol AF (BPAF) were analyzed in thermal paper cash receipts using a sensitive LC-MS/MS method. The cash receipts contained almost only BPA and BPS, whereas BPA was found in 67.5% of the samples at a concentration ranging from 0.83% to 1.71%. BPS was detected at lower concentrations than BPA, found in 25% of samples at a concentration ranging from 0.61% to 0.96%. Both of the substances exceeded the EU limit in the thermal receipt paper of 0.02% per weight. No other analogues were detected from any of the samples analyzed here. As the levels identified are higher than the legal limits of the EU regulations, we should develop regulatory rules and restrictions on the use of bisphenols in cash receipts in Bangladesh.

Keywords: endocrine disruptors; bisphenol A; bisphenol S; cash receipts; thermal paper



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1. Introduction

Bisphenol analogs have already attracted worldwide concern due to the environmental and health risks, as bisphenol A (BPA) is considered a high production volume chemical [1,2]. Although BPA is mainly used as a monomer in the manufacturing of polymer products and also as an additive in plastics, it is widely found in the paper industry as the color developer in thermal paper [3,4]. As such additives are typically not part of more chemically tightly bound thermal papers, the unpolymerized monomers such as BPA can leach from the finished product [4]. This chemical could cause endocrine disruption which may arise during pregnancy under the influence of estrogens and also other adverse effects even at a low dose [5,6]. However, there are many uncertainties that remain controversial about BPA.

Due to the ubiquity of BPA, the majority of humans are exposed to this chemical mainly via diet, packaged food, and beverages, as well as migrating BPA from the packaging material [7–10]. Although diet was found to be the main source of exposure in most cases, thermal paper was the second source when considering the environmental contamination that may also occur during the production process, use, or disposal of BPA-containing substances [9,10]. Previously, several reports were published about thermal paper as a potential source of BPA exposure in many countries such as Denmark, Sweden, Spain, Italy, Norway, Brazil, Switzerland, and the USA [3,11–16]. Moreover, concerns associated with BPA exposure have led many developed countries to take practical measures to minimize the exposure of the public to BPA through diet and other products.

In developed countries, the use of BPA has been reduced due to a combination of consumer pressure and government regulations. Following the EU regulation amending

the Commission Regulation No 10/2011, the use of BPA in plastic materials and other products that come into contact with food is authorized to the specific migration limit of 0.05 mg/kg. Moreover, BPA used in infant feeding bottles, plastic drinking cups, or other bottles made of polycarbonate is also prohibited according to the regulation. Furthermore, Since January 2020, the utilization of BPA in thermal papers present in the EU market is legally restricted or limited to a maximum level of 0.02% by weight by the REACH regulation. Although the use of BPA in many products has been prohibited in some Asian countries, such as China, South Korea, Japan, and Taiwan, no precautionary principles have been taken in countries like Bangladesh.

Extensive research about the presence of thermal paper as well as BPA in cash receipts is highly necessary to create concerns about BPA safety in Bangladesh. In order to obtain an overview of the current situation in Bangladesh, we performed a market analysis of thermal paper receipts in the city of Dhaka, the capital of Bangladesh. Approximately 67 thermal paper receipts from 40 different sources were randomly sampled and analyzed between October 2021 and March 2022. We focused the studies to identify an alternative substance that was found during this market analysis. Another potential alternative, bisphenol S (BPS), has been included in these analyses. Our current study also included bisphenol F (BPF), bisphenol B (BPB), and bisphenol AF (BPAF).

Indeed, data about the alternatives to BPA are scarce particularly for the non-bisphenols. BPS is one of the most well-known replacements for BPA that is widely used in so-called “BPA-free” products [12]. Although BPS is considered an alternative to BPA, it has similar adverse health effects to BPA, and can bind to the estrogen receptor as well as induce cancer cell proliferation [17–20]. Another potential alternative is BPF, which is used in the production of polycarbonate products and epoxy resins as well as in thermal paper, and is also activated as an estrogen receptor, indicating that these chemicals are not safe to use either [17,21,22]. Similarly to BPS, other bisphenols, such as BPB, might be used as an alternative to BPA, but are listed as the persistent substance considered toxic to the aquatic environment and are suspected to be harmful to reproduction [23]. Bisphenol AF consists of fluorinated bisphenols which have high thermal and chemical stability and are suspected to be toxic to reproduction [24]. In light of the replacement of BPA, other bisphenols are often used in products in order to label them as BPA-free. Although BPA exposure has been studied extensively due to its high capability of endocrine disruption, scarce information is available on other bisphenol analogs. Considering such effects of the endocrine-disrupting chemicals that are being used in our daily life, we were motivated to obtain an overview of the bisphenol levels in the thermal paper receipts collected from Bangladesh.

Hence, the aim of our study was to evaluate the presence and the assessment of BPA in the cash receipts collected from different sources around Dhaka and identify the potential alternative to BPA in the Bangladeshi market. For the purpose of analyzing bisphenol analogs, liquid chromatography coupled with tandem mass spectrometry (LC-MS/MS) was used. Based on the results, the presence of bisphenols in cash receipts collected from different sources in Dhaka city was identified, and we were able to understand the exposure related to current regulatory guidelines.

2. Methodology

An extensive research study was conducted by Environment and Social Development Organization-ESDO, Bangladesh, in collaboration with the Woin Institute for Occupational and Environmental Health-WIOEH, South Korea. The data to write the report was collected from both primary and secondary sources. At first, a desk study was performed to provide a preliminary set of findings on the presence of phthalates in children’s products. The primary data collection has been divided into two parts, (1) sample collection and (2) analysis of the samples which have been elaborated on in this section. After the completion of the data analysis, data visualizations were created in order to make relevant assessments.

2.1. Sample Collection

The sample collection was performed in October 2021 amid the COVID-19 pandemic. ESDO team members collected 67 samples of cash receipts from 40 different sources in Dhaka, Bangladesh, where more than one cash receipt was collected from one source. They were categorized as public offices, local franchise stores, global franchise stores, large major supermarkets, small supermarkets or convenience stores, bank number tickets, delivery receipts of general retail shops, and others. After the collection of the cash receipts, they were kept in dark wrapping with aluminum foil, stored in a zippered storage bag, and sent to the Woin Institute for Occupational and Environmental Health-WIOEH, South Korea, for analysis.

2.2. Sample Analysis

Sample analysis was performed in two phases, (1) identification of thermal paper, and (2) chemical analysis.

2.2.1. Identification of Thermal Paper

A subsample of each receipt was tested on a hotplate (MS-100, Misung Scientific Co., Deokgye-ri, Hoecheon-eup, Yangju-gun, Gyeonggi-do, Korea) at 200 °C to confirm that it was thermal paper. Then, thermal paper samples ($n = 40$) were wrapped in aluminum foil and kept in the dark until further processing. Next, the samples were analyzed for their bisphenol content.

2.2.2. Chemical Analysis

After the hot plate analysis, cash receipts collected from various places in Dhaka were stored separately until sample preparation for chemical analysis, as described in Björnsdotter et al., 2017 [13]. The analysis was conducted by a liquid chromatograph coupled with a tandem mass spectrometer (LCMS-8050, Shimadzu, Kyoto, Japan) equipped with an electrospray negative ionization (ESI) source. Target compounds were bisphenol A, bisphenol S, bisphenol F, bisphenol B, and bisphenol AF (Figure 1). At first, thermal papers were cut into small pieces and samples of approximately 50 mg were weighed into a glass tube and 10 mL methanol was added. The samples for chemical analysis were extracted sonication (60 min) and were then centrifuged at 2500 rpm for 5 min to allow the solids to precipitate. The supernatant was diluted with methanol (500 times) followed by adding 100 µL of isotopically labeled internal standards (ISTD) to improve the accuracy to a higher extent. Aliquots of 5 µL were injected into the liquid chromatograph coupled with a tandem mass spectrometer equipped with the electrospray negative ionization (ESI) source.

2.3. Statistical Analysis

In order to evaluate the statistical significance of the results, a commercially available statistical package for the personal computer (Microsoft Excel 2019) was used. We also conducted a Kruskal-Wallis Test in Excel in order to determine whether or not there was a statistically significant difference between the medians of the two groups. It is also considered to be the non-parametric equivalent of the One-Way ANOVA.

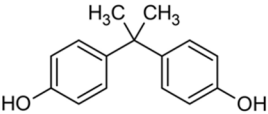
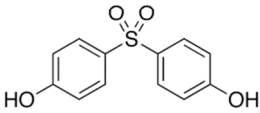
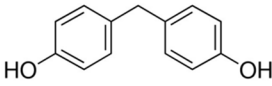
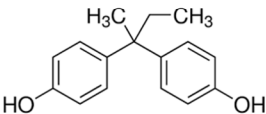
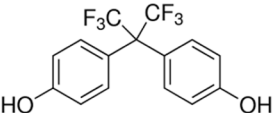
| Substances | Chemical name/ Molecular formula | Structure |
|---------------------|--|--|
| Bisphenol A (BPA) | 4,4'-isopropylidenediphenol/ $C_{15}H_{16}O_2$ |  |
| Bisphenol S (BPS) | 4,4'-sulphonyldiphenol/ $C_{12}H_{10}O_4S$ |  |
| Bisphenol F (BPF) | 4,4'-Methylenediphenol/ $C_{13}H_{12}O_2$ |  |
| Bisphenol B (BPB) | 4,4'-(Butane-2,2-diyl)diphenol/ $C_{16}H_{18}O_2$ |  |
| Bisphenol AF (BPAF) | 4,4'-(Hexafluoroisopropylidene)diphenol/ $C_{15}H_{10}F_6O_2$ |  |

Figure 1. List of substances tested.

3. Result

3.1. Thermal Paper Detection

Among the 67 collected cash receipts from 40 sources, only 2 collected cash receipts from one local franchise store were detected as non-thermal paper. Here, the two cash receipts were considered one sample, as they were collected from one source, which indicated that, among the collected receipts from 40 sources, receipts from one source were detected as non-thermal paper (Figure 2).

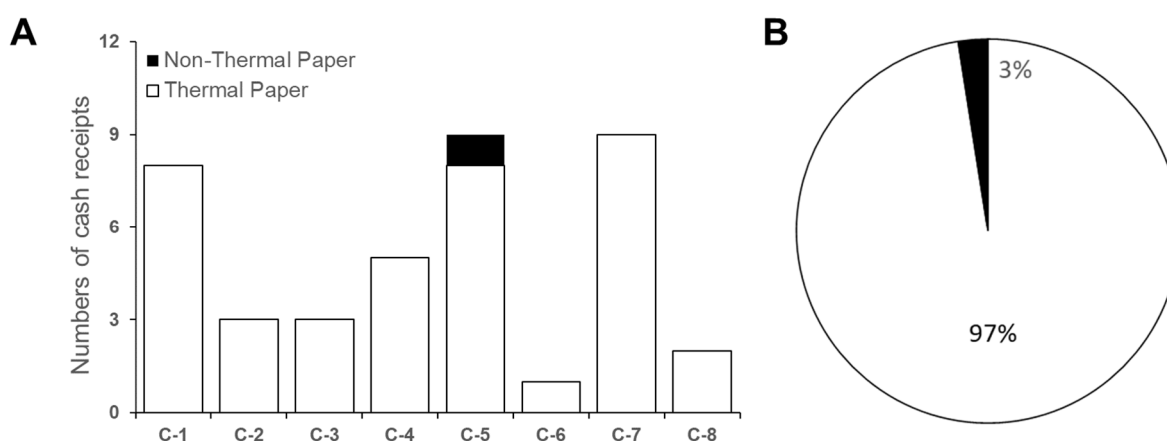


Figure 2. Result of thermal paper detection collected from Dhaka, Bangladesh. Here, panel (A) indicates the category-wise detection of thermal paper. Panel (B) indicates the place-wise detection of thermal paper that was collected from 40 sources.

We detected the types of paper category-wise, where the eight categories of the sources from where cash receipts were collected were, respectively, bank number tickets, C-1; delivery receipts or general retail shops, C-2; global franchise stores, C-3; large major

supermarkets, C-4; local franchise stores, C-5; public office, C-6; small supermarkets or convenience stores, C-7; and others, C-8. Others included online shopping and retail as well as restaurants. Among these categories, only one cash receipt from Category5 (local franchise stores) was detected as non-thermal paper (Figure 2A). Moreover, approximately 97% of cash receipts were detected as thermal and 3% were detected as non-thermal paper collected from 40 sources (Figure 2B).

3.2. Detection of Bisphenol Analogs

In this study, we analyzed 39 thermal paper receipts selected in an attempt to mirror the diversity of thermal paper available in various categories of places in Dhaka as completely as possible. The concentrations of bisphenol analogs found in these samples are summarized in Table 1. BPA was detected in 27 samples, which had levels ranging from 0.83% to 1.71%. BPS was detected in 10 samples with levels ranging from 0.61% to 0.96% (Table 1). Among the 39 thermal papers, neither BPA nor BPS were detected in two thermal paper receipts collected from Category2, delivery receipts, or general retail shops. Additionally, other bisphenol analogs such as BPF, BPB, and BPAF were not detected in any of the cash receipts.

Table 1. Detection of bisphenol analogs in the thermal paper receipts collected from Dhaka city, Bangladesh.

| Analytes | Detection Number | Detection Frequency (%) | LOD ($\mu\text{g/g}$) | Average (%) | Maximum (%) | Minimum (%) |
|----------|------------------|-------------------------|-------------------------|-------------|-------------|-------------|
| BPA | 27 | 67.5 | 20 | 1.18 | 1.71 | 0.83 |
| BPS | 10 | 25 | 23 | 0.72 | 0.96 | 0.61 |
| BPF | 0 | 0 | 34 | - | - | - |
| BPB | 0 | 0 | 34 | - | - | - |
| BPAF | 0 | 0 | 14 | - | - | - |
| ** ND | 3 | 7.5 | | | | |

** Bisphenols were not detected in two thermal papers and in one non-thermal paper.

3.3. Detection of BPA and BPS on Thermal Paper Receipts

From these results, we found that bisphenols such as BPA and BPS were commonly found in the thermal paper receipts. In all of the eight categories of places using thermal paper receipts (97.5%), bisphenols (92.5%) including BPA (67.5%) or BPS (25%) were detected in cash receipts. (Table 2). However, BPA and BPS were not present in two samples from delivery receipts or general retail shops of Category 2. Although BPA was found in cash receipts from eight categories (Table 2), BPS was found only in five categories, such as bank number tickets (2.5%), large major supermarkets (5%), local franchise stores (5%), small supermarkets or convenience stores (10%), and others (2.5%).

Table 2. Detection of BPA and BPS on the thermal paper receipts collected from all the eight categories of sources.

| Category | Description | Thermal Paper Detection | | Bisphenol Detection | | Detection of BPA | | | Detection of BPS | | |
|----------|---|-------------------------|------|---------------------|------|------------------|------|------------------------------|------------------|-----|------------------------------|
| | | N | % | N | % | N | % | Quantity ($\mu\text{g/g}$) | N | % | Quantity ($\mu\text{g/g}$) |
| C-1 | Bank number tickets | 8 | 20 | 8 | 20 | 7 | 17.5 | 8.33 | 1 | 2.5 | 0.82 |
| C-2 | Delivery receipts or general retail shops | 3 | 7.5 | 1 | 2.5 | 1 | 2.5 | 1.02 | - | - | 0 |
| C-3 | Global franchise stores | 3 | 7.5 | 3 | 7.5 | 3 | 7.5 | 2.99 | - | - | 0 |
| C-4 | Large major supermarkets | 5 | 12.5 | 5 | 12.5 | 3 | 7.5 | 3.16 | 2 | 5 | 1.38 |
| C-5 | Local franchise stores | 8 | 20 | 8 | 20 | 6 | 15 | 6.98 | 2 | 5 | 1.37 |
| C-6 | Public office | 1 | 2.5 | 1 | 2.5 | 1 | 2.5 | 1.71 | - | - | 0 |
| C-7 | Small supermarkets or convenience stores | 9 | 22.5 | 9 | 22.5 | 5 | 12.5 | 6.53 | 4 | 10 | 2.64 |
| C-8 | Others | 2 | 5 | 2 | 5 | 1 | 2.5 | 1.09 | 1 | 2.5 | 0.96 |
| | Total | 39 | 97.5 | 37 | 92.5 | 27 | 67.5 | | 10 | 25 | |

A Kruskal-Wallis Test was performed to compare the quantitative variables and ensure that they were the same for all the cash receipts. A total of 16 samples were used in the analysis, where BPA or BPS was found in each of the eight sources. The test revealed that the quantity of BPA or BPS was not the same ($H = 6.893$, $p = 0.005$), indicating that there was a statistically significant difference in the quantity of BPA or BPS among the cash receipts collected from different sources.

4. Discussion

In our present study, we observed that BPA is commonly used in cash receipts as a color developer. There are many studies which have investigated the presence of BPA in thermal paper receipts in different countries all over the world [11–16]. In such previous studies, BPA concentration was found up to 28 mg/g with a detection frequency of more than 44%. Similar to the detection frequencies found worldwide, we also observed a detection frequency of 67.5% in the collected thermal paper around the city of Dhaka.

As BPA is not chemically bound in thermal receipt paper, it can easily be transferred from the thermal receipts paper to other objects such as different types of papers [25–27]. Usually, people place tickets and cash receipts in their wallets, and cash receipts can be contaminated by BPA via contact with these receipts. Moreover, the recycling of thermal paper receipts and the use of BPA as binding agents in printing ink on these papers may cause BPA contamination followed by exposure to human beings via dermal contact and the environment [25,28,29].

Over the last few years, alternative substances to BPA have been developed for thermal receipt paper, as BPA has increasingly fallen into disrepute due to its negative health impact. In this present study, BPA was not found in 25% of the thermal papers collected in the eight categories of places in Dhaka, Bangladesh. Indeed, we found one of the local franchise stores was found to use non-thermal papers, and two Bangladeshi retailers used thermal paper receipts that do not contain any of the bisphenols. Therefore, it was not astonishing to find alternatives to BPA-containing thermal papers, such as non-thermal paper receipts or thermal papers with no bisphenols.

Our study is one of the first to find the presence of BPA and BPS in thermal paper collected from Bangladesh. The present investigation showed that either BPA or BPS were detected in the cash receipts, and were considered the most common substances out of the group of bisphenols. This is because BPA and BPS were the only bisphenols detected in most of the samples analyzed, except for the samples from one local franchise store and two retail shops. Moreover, the BPA detection was higher compared to the respective BPS among all of the samples, which is similar to previous findings of a detection frequency of 78% for BPA and 49% for BPS in thermal paper cash receipts from different sources in the Netherlands, Sweden, Norway, Spain, and Austria [13,30]. Additionally, a study of thermal paper receipts collected in Germany also revealed the abundance of BPA, where BPS was found less frequently in the receipts [31]. Regarding the detection of other bisphenols such as BPB, BPF, and BPAF, these substances were not detected in our present study. This result is similar to the results of receipts sampled in Austria, where only BPA and BPS, but not BPF, BPAF, BPE, or BPB were detected [32–34]. Actually, BPS was often used as an alternative to BPA as a color developer before legal restrictions on the use of bisphenols in thermal receipt papers in the EU were implemented.

Although there is no regulation in Bangladesh regarding the use of BPA or other bisphenols in cash receipts, the result of BPA detection was much higher than the EU standard of 0.02% per weight. Detection of BPA and BPS exceeded the EU standard limit for all of the cash receipts. The presence of BPA or BPS indicates a high health risk to the user. Indeed, studies have found that BPA, even at a low dose, can cause adverse endocrine disruptive effects and carcinogenic effects on breast cancer, suggesting the significance of identifying all potential sources of human exposure [5–8,35]. As there have been no previous studies about the presence of these endocrine-disrupting chemicals in thermal paper collected from Bangladesh, our current study fills an important knowledge gap by

identifying the thermal paper receipts as a source of bisphenols as well as the BPA exposure to our general population. Even though there are no regulatory rules and restrictions on the use of bisphenols in cash receipts, the present findings indicate the urgency of strict regulation against using such endocrine-disrupting chemicals in our daily life.

5. Conclusions

BPA and BPS were detected in thermal receipt paper at high concentrations. BPA and BPS were not found in non-thermal receipt papers from a local franchise store, and in two samples of thermal receipt paper from retail shops. The results show that no BPB, BPF, or BPAF was detected in any of the samples. As there is no information about the annual consumption of thermal paper in Bangladesh and the presence of bisphenols in cash receipts, we here estimated that approximately 67.5% of cash receipts contained BPA at a concentration range of 0.83–1.71%. All of the samples showed extremely high BPA levels, compared to the EU legal limit value of 0.02% per weight since January 2020. Additionally, BPS was detected in 25% of the thermal paper receipt samples where BPA was not detected. Therefore, use of BPA and BPS in cash receipts seem to be common in our country.

We found that these high-volume chemicals are also known as endocrine disruptors and are toxic to human reproductive systems. As they are being used in the thermal receipts papers as a color developer, the harmful effect of such chemicals may distribute to human systems and cause disabilities for future generations. Due to such detrimental effects on health and the level of BPA or BPS present in cash receipts being higher than the legal limits, we need to develop regulatory rules and restrictions on using these bisphenols in Bangladesh.

Author Contributions: H.F. performed the preliminary analysis and coordinated sample collection, preparation, and shipping for analysis. Y.-S.L. and I.C. conceived the experiment and designed the methodology, as well as conducted the laboratory analysis in Korea. S.K. performed data visualization and wrote the manuscript. S.H. and S.S., together with the other author, reviewed and edited the manuscript. All authors have read and agreed to the published version of the manuscript.

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Institutional Review Board Statement: The study was conducted in accordance with the Declaration of Helsinki, and approved by the Institutional Review Board (or Ethics Committee) of ESDO.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to further research activities.

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Conflicts of Interest: The author declares no conflict of interest.

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