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Research article

Evaluation of Bisphenol A content in food contact PVC cling film

Hamidreza Pourzamani[1,2], Karim Ebrahim[2], Maryam Mirlohi[3], Nasrin Mirzaei[4], Ahmad Reza Bahrami[5], Ebrahim Rahimi[6], Bayan Saeidi[3], Majid Falahati[2], Bahram Nasr Esfahani[7], HajiehGhasemian Safaei*[7]

[1]*Environment Research Center, Research Institute for Primordial Prevention of Non-communicable Disease, Isfahan University of Medical Sciences, Isfahan, Iran.*
[2]*Department of Environmental Health Engineering, School of Health, Isfahan University of Medical Sciences, Isfahan, Iran.*
[3]*Food Security Research Center, School of Nutrition and Food Science, Isfahan University of Medical Sciences, Isfahan, Iran.*
[4]*Department of Microbiology, Tonekabon Branch, Islamic Azad University, Tonekabon, Iran.*
[5]*School of Veterinary Medicine, Shahrekord Branch, Islamic Azad University, Shahrekord, Iran.*
[6]*Department of Food Hygiene and Quality Control, School of Veterinary Medicine, Shahrekord Branch, Islamic Azad University, Shahrekord, Iran.*
[7]*Department of Microbiology, School of Medicine, Isfahan University of Medical Sciences, Isfahan, Iran.*

Key words: Bisphenol A, cling film, food, packaging.

***Corresponding Author: Hajieh Ghasemian Safaei,** Department of Microbiology, School of Medicine, Isfahan University of Medical Sciences, Isfahan, Iran. Email id- ghasemian@med.mui.ac.ir

Abstract

Bisphenol A is commonly used as the plastic and metal material in the food and drink packaging industry and can be found in an abundant of material intended to come into contact with food such as polyvinyl chloride plastic film or stretch cling wrap. As Bisphenol A was classified as a substance of external origin with a detrimental effect on human health, it is aimed to evaluate the occurrence of Bisphenol A in food contact cling film. Twelve cling film samples from various countries along with six standard samples of Bisphenol A with different concentrations were evaluated. Bisphenol A was extracted from cling film and a small fraction of that was injected to GC–MS for quantitative analysis. Then, identification of Bisphenol A was carried out by GC-MS. GC Bisphenol A peaks were identified by comparison of their mass spectra with that of the Bisphenol A standard and with a spectrum. The Bisphenol A level among samples varies between 3.3 µg/L and 402.3 µg/L while, the daily intake of Bisphenol A ranges between 0.16 µg/kg and 20.1 µg/kg. Additionally, the average level of Bisphenol A concentration and daily intake were 81.46 µg/L and 4.072 µg/kg, respectively. Since the concentration of Bisphenol A varied in all the samples due to their countries produce cling film, it is required to establish a worldwide standard Bisphenol A limits specifically for food packaging cling films due to its importance and international trade as well.

Introduction

Bisphenol A (BPA), 2,2-bis(4-hydroxyphenyl) propane, has one of the highest volumes between chemical substances in the world. BPA is widely used in the manufacture of resistant plastic polycarbonate and epoxy resins [1-2]. As such, it is commonly used as the plastic and metal material in the food and drink packaging industry and can be found in an abundant of material intended to come into contact with food such as reusable plastic bottles, plates, cups, tableware, containers, trays, and in the protective linings in cans [3-4]. It is also found in polyvinyl chloride plastic film or stretch cling wrap [5], which extends the shelf life of products by keeping food fresh and creating greater food safety assurance from microorganisms along with biological and chemical changes that can reduce or change food quality [6-7].

While the plastics industry claims that BPA, using in accordance with current regulations, is nontoxic and un-harmful to human health; BPA was classified as a substance of external origin with a detrimental effect on human health by the European Commission in 1996 [8-9]. Numerous studies carried out on side effects caused by human exposure to BPA have linked it with Type 2 diabetes, obesity, abnormal liver functions, hormone disruption called endocrine disrupting chemical (EDC), prostate and breast cancer, neurodevelopmental impairment, neurobehavioral, neurological problems such as attention deficit hyperactivity disorders and autism and most importantly, various reproductive disorders affecting off springs[10-11].

Since, BPA is used as an additive in the production of PVC products, there is the possibility of its transferring from the plastic packaging into the food products [12]. Therefore, a specific migration limit (SML) of 3 mg/ kg of food was set

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by the European Commission Directive 90/128 (1990) for the use of BPA as a monomer or starting substance in the manufacture of plastic materials and or articles intended to come into contact with food, a limit treated as equivalent to 0.5 mgdm^{-2} when the plastic is a container, container lining or film [9]. The European Union re-evaluated the specific migration limit for BPA from food contact plastic materials to 600 ng/g in 2004 [1]. In a study conducted by Sajiki et al. [13], it was reported that the amount of BPA in foodstuff relies on the type of container used for storing food as it is 0-14 ng/g, 0-1 ng/g and 0-842 ng/g for plastic, paper containers and canned food products, respectively [13].

In correspondence with the points above, a maximum acceptable dose of 50 µg/kg bw/day for BPA was established by the United States Environmental Protection Agency (1993), while Health Canada (2010) and the European Food Safety Authority (2015) set a temporary total daily intake (t-TDI) of 25 and 4 µg/kgbw/day, respectively. These discrepancies in total intakes set by the three legislative organizations reflect the fact that BPA toxicity continues to be controversial [3, 14].

Although, BPA concentrations in the nanomolar range in other published documents are classified as the risk alert limit by American legislative organizations [15], but some studies reported that BPA concentrations lower than 1 ng/L have estrogenic activity. Exposure to BPA may lead to irreversible changes, some of which might not surface immediately such as a disturbance in the distribution seen in the chromosomes in fetuses daughter cells of mice exposed to BPA concentration as low as 20 ppm in drinking water [8]. The occurrence of BPA in various environmental and human samples as well as foodstuffs has been reported extensively, but not much work has been done on the evaluation of BPA in food contact cling film. This is the first study in our country with that specific objective.

Experimental

Twelve samples from various countries including Iran, Poland, Germany, Korea, Canada and USA with different brand names were chosen randomly. Two samples were chosen from each country. Additionally, six standard samples of BPA with different concentrations were examined and studied for this purpose. The product names are not mentioned here.

Chemicals

BPA (99%) was purchased from Sigma-Aldrich. Methanol (GC grade) and methyl tert-butyl ether (MTBE, 99.9%) was purchased from Merck (Germany). Ultra-pure water (Millipore, Bedford, MA, USA) was used as a simulant.

Samples

The reels of all polyvinyl chloride cling film were obtained from the local supermarkets of different countries. The average length and width of all samples was 155.07 m and 30.53 cm, respectively. The average thickness of the samples, after assessment with thickness measurement gauge, was 10±2 µ. In order to prevent any kind of contamination, one meter of cling film was removed before analysis.

A stock solution of approximately 100 mg/l of BPA (Sigma-Alderich, USA) was prepared by weighing and dissolving 100 mg BPA into 100 ml deionized H_2O in a volumetric flask. All standard solutions were processed exactly the same as sample. Standard series (between 1µg/L to 1000µg/L) were prepared using deionized H_2O to achieve the desired concentrations. A six-point standard curve was plotted using BPA concentrations of 1, 50, 100, 250, 500 and 1000 µg/L. Two samples of each standard were each run three times.

BPA extraction from cling film

In order to evaluate BPA concentration, it is required to extract BPA from the cling film sample. First, 5 dm^2 of cling film sample submerged in 100 ml distilled deionized water in an Erlenmeyer flask, which was hermetically closed. Then, containers were kept at 100°C for one hour. Subsequently, 20 ml of water from the Erlenmeyer flask was passed through C18 cartridge (500 mg; Chromabond, Germany) with silica-based bonded phase (4-5 inches/Hg), which was conditioned using 5 ml methanol (99%) and 10 ml ultra-pure water. Methanol was used for washing cartridge under 1 inch/Hg pressure. The eluted methanol was collected and dried under nitrogen gas (with 99% purity) at ambient temperature. The dried extract was resolved in 100 µL MTBE from which 3µL of that was injected to GC–MS for quantitative analysis. All the stages of experiments were done under sterile condition and in duplicate.

Identification of BPA by GC-MS

For quantifying the BPA concentration, an Agilent Technologies system containing a 5975C inert MSD, a triple axis detector equipped with a 7890A gas chromatograph and a split/splitless injector was used. Helium (99.995% purity) as the carrier gas at a flow rate of 1 ml/min was used to accompany with a fused silica column (HP-5 ms, 5% phenyl and 95% dimethylpolysiloxane, 30 m × 0.25 mm I.D and 0.25- µm film thickness).

The column temperature was as follow: 100°C for 1 min increased to 210°C at a rate of 10°C/min, then to 250°C at a rate of 5°C/min and finally to 280°C at a rate of 30°C/min with holding for 1 min. The injection and transfer line temperatures were 260 and 280°C, respectively. The total running time was set at 25 min. GC BPA peaks were identified by comparison of their mass spectra with that of the BPA standard and with a spectrum.

Results and Discussion

All tested cling film samples from different countries, including Iran, Poland, Germany, Korea, Canada and USA were evaluated for its BPA content. It should be noted that all samples had household consumption and were food grade cling film except Korean one, which had industrial use. Based on our results, the average of BPA content for each two samples from each country was 3.93, 3.82, 3.30, 17.67 and 57.75 µg/L for Iranian, Polish, German, Canadian and American food grade samples, respectively. Also, the daily intakes were as follow: 0.196 for Iranian, 0.191 for Polish, 0.165 for German, 0.883 for Canadian and 2.887 µg/kg for American samples. Besides, the average of BPA level and its daily intake for two industrial Korean samples was 402.30 µg/L and 20.11 µg/kg, respectively. According to the results mentioned above, the BPA content among samples varied between 3.3 and 402.30 µg/L and the daily intake of BPA ranged between 0.165 and 20.11 µg/kg.

Conspicuously, the highest level of BPA and its daily intake among cling films belong to Korean sample which had industrial consumption, while the lowest level and daily intake went for food grade German sample. It is apparent that only Korean cling film sample was not suitable for food contact wrap in comparison to all three mentioned organizations. Among food grade cling film samples, the most suitable and the less adequate samples belong to German and American cling film, respectively. Additionally, the average level of all BPA concentration and daily intake for all samples were 81.46 µg/L and 4.072 µg/kg, respectively.

Bisphenol A is a multipurpose compound, which has been extensively employed in daily life such as in polycarbonate baby bottles, reusable water bottles, polyvinylchloride stretch films and metallic food can inner coatings [16-17]. Thus, exposure of wildlife and humans to BPA is inevitable, expected to continue, and even to increase [18] that making it one of the most common concern of the modern industrial world. Typically, diffusion of BPA from polymers is reliant on a number of parameters such as concentration of BPA in packaging film and food, nature of the foods, temperature, and the time period over which duration of contact occurs [19].

As a matter of fact, the occurrence of BPA as a prevalent chemical substance in numerous environmental matrices in the ranges of 5-320 ng/L in river waters, 20-700 ng/L in sewage effluents, 0.2-199 ng/g in the dust, 0.1-384 ng/g in foodstuffs and etc have been reported, whereas the BPA concentration in food contact cling film ranges 3.3 µg/L and 402.3 µg/L in the present study. Besides, many reports have indicated that BPA has been detected from blood (0.3-4.4µg/L), urine (0.47-9.5µg/L) and other samples such as amniotic fluid and placental tissues with a detection rate above 90% in most of the studies as a result of the high daily intake of BPA [1, 6], while this value for present study varied between 0.16 µg/kg and 20.1 µg/kg which are more applicable for using in the USA and Canada according to their legislative organizations for BPA concentrations.

As it mentioned before, specific migration limits for all acceptable substances in plastic materials, including cling film is 600 ng/g for BPA since 2004 and has not been changed by the European Union, except for baby bottles since 2011 [20]. In this regard, Canada was the first country in the world to forbidden BPA use in baby bottle products as a result of being highly toxic substance in 2010 [14]. In one Iranian study, BPA concentration was between 0.49-8.58 µg/l and 0.63-2.47 µg/l for new and used baby bottles, respectively. Cao and Corriveau found this figure for used bottle was as the same as Iranian study, but BPA level for the new bottle (228-521 µg/l) was far higher than Iranian one [11]which is against of those study found that repeated use of a container as well as temperature can increase the exposure levels to human. Correspondingly, Le et al. found that leaching of BPA into the contained fluid occurred at ambient temperature and can increase to 55-fold provided that boiling water added. Although, the main mechanism lead to BPA exposure is through leaching from plastic products, sources of BPA ingestion may vary among population dependent on plastic product they use [16] such as baby milk bottle for infant or beverage bottle for adult, but food contact cling film is a kind of plastic product which have been used for all ages for packaging food. Regarding its position and importance; in a study which various film samples were tested for BPA concentration, it merely was detected in PVC-based films and its contents were between undetectable levels to almost 500 g/kg [9]. It should be noted that all cling film samples used in this study were PVC cling film and BPA level varied from 3.3 µg/L and 402.3 µg/L. Undetectable level of BPA was not seen among samples.

Another study evaluated the content of BPA and its migration in several PVC stretch films used for food packaging and resulted to have BPA level ranged from 43 µg/g and 483 µg/g [2] which is against of present study that found lower minimum and maximum level BPA in food contact cling film. Needless to say that daily intake of American (2.887 µg/kg), Canadian (0.883 µg/kg) and European (0.165 µg/kg for German and 0.191 µg/kg for Polish) cling film used in this study was far lower than a maximum acceptable dose of BPA established by United States Environmental Protection Agency (50 µg/kg), Health Canada (25 µg/kg) and European Food Safety Authority (4 µg/kg). Besides, daily intake of Iranian cling film (0.196 µg/kg) was as the same as other samples and its figure was low, whereas daily intake of Korean cling film (20.11 µg/kg) in comparison to others and three legislative organizations mentioned above, was lower than the maximum acceptable dose of BPA set by United States Environmental Protection of Agency in spite of having industrial consumption, but far higher than the daily intake of European Food Safety Authority.

Regarding to PVC based products; BPA concentration in commercially available PVC products was reported by Sun et al. that this value was 68±3.5 µg/g for PVC wrap film,

60.5±2.8 µg/g for PVC gloves and 290.1 µg/g for PVC hose [21]. Generally, the average BPA contents for foodstuffs in glass jar whose lid covered with plastic film were determined to be between undetectable and 399.21 ± 3.26 µg/kg [22], whereas the average level of BPA in the present study was 81.46 µg/L, which is lower than the BPA concentration in foodstuffs.

The extent of BPA contamination of paper and paper products is a newer aspect of BPA consideration. Generally, BPA exposure from multiple paper products such as thermal receipts, tickets, magazines, newspapers, and business cards were used to calculate a daily dose of 0.219 ng/kg bw/day for an 80 kg adult. In comparison to recycled materials, Ozaki discovered that 0.034-0.36 µg/g can be found in food packaging paperboard from virgin materials, while food packaging paperboard from recycled materials can have ten times more BPA, at 0.19-26 µg/g [5].

Lopez-Espinosa et al. found the BPA levels in paper and cardboard containers used for take-away food varied between 0.05 and 1817 ng/g at a detection frequency of 45%. On the other hand, Kitchen roll from virgin paper contained no or insignificant BPA concentrations, whereas BPA level in kitchen roll from recycled paper was 0.6–24 µg/g [2]. In the present study, BPA concentrations were seen in all cling film samples. Besides, an abnormally high BPA value (26000 ng/g) was also reported in food packaging made from recycled paper in Japan [23].

Interestingly, BPA hazards can be absorbed into the human body by contact, which is called allergic contact dermatitis (ACD), and this is usually occupational. Most ACD occurs in workforces deal with products based on BPA such as plastic-products workers, workers exposed to epoxy adhesive tapes and foams and dental assistants [24]. As an example, it has been estimated that occupational exposure after ten hours of work as a cashier is 71 µg per day, whereas in the general population it ranges from 7.1 µg to 42.6 µg per day [25].Recently, BPA has also been detected in paper currencies, as well as in other papers and paper products. Also, an average BPA concentration in Switzerland was 13.3 g/kg for thermal printing papers [26].

According to food and drug administration (FDA) declaration in 2008, daily exposure of BPA for adults is 0.16 µg/kg per day, while the daily human intake of BPA stated to be <1µg/kg body weight/day on the basis of several studies[18, 24]. In comparison to the present study, this value for German sample is in concordance with FDA declaration, whereas all samples except Korean and American one are in consistent with other studies to have a daily intake of <1µg/kg body weight/day.

Although, BPA exposure estimated to be 0.48-1.6 g /kgbw/day from food sources by the European Scientific Committee on Food, it is believed that all canned food samples indicated detectable levels of BPA in a wide-ranging of concentrations between 3.7 ng/g and 8.3 ng/g [6, 27]. Based on BPA content in canned foods New Zealand; Thomson and Grounds reported that the mean and maximum exposure to BPA was 0.008 and 0.29 µg/kg bw/day in their country, respectively [24] that is not comparable with the data presented in this study. Other studies on canned drink analyzed from different markets showed the levels of BPA ranged from 44 ng/ L and 607 ng/L for Barcelona and 0.01 µg/L and 4.7 µg/L for Portugal, respectively, whilst, migration of BPA from canned foods in Japan was determined to be 213 ng/ml for drinks and 602 ng/g for corned beef. Although the BPA content were somewhat higher for foods stored in cans than plastic containers, this difference was not statistically meaningful [23, 28-29].

Conclusion

Taken all together, the concentration of BPA varied in all the samples due to their countries produce cling film. Ergo, the occurrence of BPA was found by the presence of BPA corresponding peak in Gas Chromatography analysis indicating the levels of BPA identified in cling film analyzed here are improbable to be of concern to public health except the Korean sample, which had industrial consumption and was unqualified for food packaging. Due to medical importance of low exposure level of BPA to human, it is better to use those food grade cling films which have a lower BPA level and daily intake. However, in the absence of any BPA legislation in some countries, it needs further investigation. This study would develop awareness among legislative organizations to establish a worldwide standard BPA limits specifically for food packaging cling films due to its importance and international trade as well.

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