ORIGINAL ARTICLE

Release of Bisphenol A From Polycarbonate and Polyethylene Terephthalate Drinking Water Bottles Under Different Storage Conditions and Its Associated Health Risk

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ABSTRACT

Introduction: Bisphenol A (BPA) is a controversial plastics ingredient used mainly in the production of polycarbonate plastics (PC) and epoxy resins that widely used nowadays in food and drink packaging. Even though BPA is not involved in polyethylene terephthalate (PET) manufacturing, recent study had reported the present of BPA in PET water bottle. This study was conducted to investigate effects storage conditions on release of BPA from PC and PET bottled water as well as to assess health risks associated with consumption. **Methods:** Solid phase extraction (SPE) was used to extract the samples, followed by analysis using ultra high performance liquid chromatography with fluorescence detector (UHPLC-FLD). The possibility of developing chronic non-carcinogenic health risk among consumers of bottled water was evaluated using hazard quotient (HQ). **Results:** Results showed that BPA migrated from PC and PET water bottles at concentrations ranging from 9.13 to 257.67 ng/L and 11.53 ng/L to 269.87 ng/L respectively. Concentrations of BPA were higher in PET bottled water compared to PC bottled water across all storage conditions. Higher storage temperature and longer storage duration increased BPA concentrations in PC and PET bottled water. Concentrations of BPA in bottled water which were kept in a car and were exposed to sunlight were higher than control samples which were stored indoor at room temperature. **Conclusion:** No significant chronic non-carcinogenic health risks were calculated for daily ingestion of BPA-contaminated bottled water; calculated HQ was less than one.

Keywords: Plastic bottles, Ultra-high-performance liquid chromatography (UHPLC), Solid phase extraction (SPE), Health risk assessment

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INTRODUCTION

Bisphenol A (2,2-bis(4-hydroxyphenyl)propane); otherwise known as BPA, is a monomer mainly applied in the production of polycarbonate plastics (PC) and epoxy resins. Polycarbonate plastics exhibit high transparency, heat resistance, light weight and good impact resistance; these advantages had led to its incorporation into production of many household items (1).

Besides PC, polyethylene terephthalate (PET) is also commonly used in the packaging industry particularly as drinking water bottles. PET is a long-chain polymer of polyesters consisting of two monomers; terephthalate acid (TPA) and ethylene glycol (EG) which are derived from oil feedstock (2). BPA is not a monomer for PET. As such, its occurrence in PET products is not anticipated (3). However, recent studies have reported BPA leaching from PET water bottles (4-6). It was suggested that PET water bottles were contaminated with BPA during recycling (3).

On a daily basis, humans' major exposure to BPA is through their diets, including the consumption of contaminated beverages and food. U.S. Centers for Disease Control and Prevention reported that urinary BPA concentrations were detected in 92.6% of the U.S. general population at an average level of 2.6 ng/L (7). Therefore, exposure to BPA due to its migration from water bottles into drinking water is alarming. Consumers were concerned as they consume daily from water bottles.

BPA is an endocrine disruptor that is gaining public attention due to its potential health effects. Several laboratory animal models were exposed to BPA at concentrations comparable to those faced by human in an attempt to gauge potential adverse effects in human (8). Pérez-Albaladejo et al. (9) has suspected BPA to be toxic and was able to disrupt the endocrine system of human. It was also able to diffuse across the placental barrier

causing harm during embryonic and fetal development. BPA mimics estrogenic activity in animals and human by modifying synthesis of endogenous hormone, altering hormone concentrations and hormone metabolism in blood (10). As a precautionary measure, legislations in United States had imposed a ban on BPA in children's products at both states and federal constitutions (11). In addition, European Commission (EC) Directive (EU Regulation No. 10/2011) had set a specific migration limit for BPA to food at 600 ng/g (12).

Sugiura-Ogasawara et al. (13) reported that exposure to BPA was associated with recurrent miscarriage. Mean BPA levels detected in blood serum of women who experienced three or more consecutive miscarriages were 2.59 ng/mL; which was three times higher compared to those without fertility problems. In addition, Lang et al. (14) reported association between high concentrations of urinary BPA and incidences of diabetes and cardiovascular disease in United States.

Concentrations of BPA in PC and PET in bottled drinking water had been reported previously (5-6, 8, 15-22). Most research methodology was similar with minor variations of storage durations and temperature. Unhealthy consumer behavior such as reuse of water bottle for storage of warm water and exposing them to sunlight had led to elevated health risk. Thus, this study aims to (i) investigate effects of storage conditions (temperature, duration, and storage in car exposed to sunlight) on BPA migration among different types of water bottles (PC and PET) and (ii) assess their associated health risks due to consumption.

MATERIALS AND METHODS

Reagents and chemicals

BPA standard (≥99%) was purchased from Sigma-Aldrich (St. Louis, MO, USA). A standard stock solution of BPA was prepared in HPLC grade methanol at concentration of 1000mg/L monthly. Meanwhile, 10mg/L standard working solution was prepared weekly by diluting the stock solution. Both solutions were stored in amber screw-capped vials at temperature of -20°C.

HPLC grade methanol, analytical grade dichloromethane (DCM) and 98% formic acid were obtained from R & M Marketing (Essex, UK). Analytical grade acetone was purchased from Sigma-Aldrich (Germany). Analytical grade methanol was purchased from J.T. Baker (USA). All ultrapure water (18.2 MΩ) used in this study was produced by Milli-Q® Advantage A10 Water Purification System (France).

An SPE vacuum manifold with 12 positions from Phenomenex Inc. (USA) was used for extraction. Sep Pak® Vac 3cc (200 mg) C18 cartridges were purchased from Waters (Massachusetts, USA). Two mL amber screw-capped vials with PTFE/silicone septa liner caps were purchased from Thermo-Line (Australia). Nylon syringe filters (0.22 µm, 13 mm) were purchased from Thermo Scientific (USA).

Sample collection

PC and PET drinking water bottles were selected as they are used and manufactured in Malaysia. Empty drinking water bottles were purchased intentionally to prevent any cross-contaminations from source of water. Caps for PC and PET were made of low-density polyethylene (LDPE) and high-density polyethylene (HDPE), respectively.

A total of 72 empty PC and PET bottles without labelling were purchased for this study. All PC bottles purchased from one manufacturer while all PET bottles purchased from another manufacturer in Malaysia. PET bottles usually will be disposed after single use but there are consumers who reuse them. PC bottles usually will be reused by consumers on daily basis as drinking water bottles. Three different storage conditions were studied: i) storage temperature, ii) storage duration, and iii) storage in a car. All bottles were filled with 500 mL ultrapure water and stored in standing position. The pH of the ultrapure water ranged from 6.9-7.0. However, pH was not the parameter of interest in this study.

Sample preparation

Methodology for sample preparation was adapted from Fan et al. (22) with some slight modifications. Effects of storage duration on the migration of BPA were studied at week 1, 2, 4 and 8. Meanwhile, storage temperatures studied were 4°C (chiller temperature), 25°C (room temperature), 50°C and 75°C (interior temperature of a vehicle). Temperature to emulate storage in car was determined based on study by Manning and Ewing (23); which interior temperature of a car was postulated to be 75.1°C given ambient temperature of 32.5°C.

Effects of storing water bottles in a car on leaching of BPA were studied at day 1, 2, 4, and 8. Meanwhile, control samples were established by storing additional set of water bottles indoor at room temperature. Hourly UV radiation and temperature data in the car were recorded to serve as supporting data.

Sample extraction and analysis

Water samples were extracted using SPE and analyzed using UHPLC-FLD, as outlined in Fan et al. (22) with slight modifications. A Sep-Pak Vac C18 cartridge (200 mg/3 mL) was used to extract BPA for analysis. Prior to sample loading, cartridges were conditioned with 4 mL methanol (1 mL/min) and 4 mL water (1 mL/min) twice. Then, 500 mL of sample was loaded into SPE cartridge and thereafter followed by drying for 10 mins at pressure of 10 mm Hg. Four mL of methanol was added to elute analytes of interest (24). The eluent was dried using nitrogen and then reconstituted with 1 mL of methanol:water (1:3). All extracts were filtered through a 0.22 µm nylon syringe filter and stored in a 2 mL amber

glass vial at temperature of -20°C until analysis.

A PerkinElmer Flexar FX-15 UHPLC system with Series 200a fluorescence detector equipped with Fortis C18 column (50 mm x 2.1 mm, 1.7 µm) was used for analysis. The mobile phase was methanol-water (70:30, v:v) with 0.1% formic acid and run under isocratic condition. The flow rate and injection volume was 0.3 mL/min and 10 µL respectively. Total runtime for analysis was 4 min. Excitation and emission for fluorescence detector was set at 278 nm and 315 nm respectively. Data acquisition and analyses were performed using Chromera software from Perkin Elmer (USA).

Migration rate of BPA from PC and PET water bottles
The BPA release kinetics curve was plotted using Eq. 1 and SigmaPlot version 12.

$$C = C_{max} \times (1 - e^{-bt}) \quad \text{Eq. 1}$$

where C is concentration of BPA leached into drinking water at time t, Cmax is the maximum concentration of released BPA (ng/L), b is the kinetics constant ($week^{-1}$ or day^{-1}), and t is the storage duration (week or day).

C_{max} and b values can be calculated by substituting the obtained results into Eq. 1 or through plotting the kinetics curve for more accurate values. Through differentiation of Eq. 1, Eq. 2 was obtained to compute the migration rate of BPA at certain times.

$$\text{Migration rate } (week^{-1} \text{ or } day^{-1}) = Cmax \times b \times e - bt \quad \text{Eq. 2}$$

where C_{max} is the concentration of BPA leached into drinking water (ng/L), b is the kinetics constant ($week^{-1}$ or day^{-1}), and t is storage duration (week or day).

Quality control (QC)
In every batch of sample analysis, a sample blank was process alongside to rule out possible background contamination during sample extraction ultrapure water was used as the SPE sample blank. Triplicate samples were analyzed for each condition.

UHPLC-FLD was calibrated with BPA standards producing a five-point external calibration curve. The linearity of calibration curve was evaluated by coefficients of determination (R2).

Instrumental detection limits (IDL) and method detection limits (MDL) were determined as concentration producing a signal-to-noise ratio (S/N) of 3. Instrumental quantification limits (IQL) and method quantification limits (MQL) were determined as concentration with S/N of 10. IDL and IQL were determined using direct injections of decreasing volume of BPA solutions at the lowest observed concentration, whereas MDL and MQL were estimated by spiking each matrix with known concentrations of BPA. Spiked matrices undergone similar extraction procedures and instrumental analysis (25).

Percentage of extraction recovery was used to determine percentage of BPA recovered during extraction. 0.2 mg/L of BPA were spiked into ultrapure water; which ultrapure water is the standard reference matrix. Percentage of extraction recovery is defined as ratio of BPA concentration recovered from sample which was spiked prior to SPE extraction to sample which was spiked after SPE extraction; minus the background concentration if any (Eq. 3) (26).

$$\text{Recovery } (\%) = \frac{C_p - C_{qc}}{C_a - C_{qc}} \times 100 \quad \text{Eq. 3}$$

where Cp is BPA concentration spiked prior SPE; Ca is BPA concentration spiked after SPE; and Cqc is BPA concentration in ultrapure water which serve as standard reference matrix.

Instrumental's precision intra-day and inter-day were determined using the same operating conditions. 0.2 mg/L of BPA was injected five times a day to determine intra-day precision; while once a day injection for 5 consecutive days was performed to determine the inter-day accuracy.

Health Risk Assessments
Hazard quotient (HQ) was calculated to determine possible chronic non-carcinogenic health effects among consumers due to consumption from PC and PET water bottles. The hazard quotient (HQ) is defined as ratio of chronic chemical daily intake (mg/kg/day) to the reference dose (RfD) (mg/kg/day) (27). The HQ was calculated according to Eq. 4.

$$HQ = \frac{CDI}{RfD} \quad \text{Eq. 4}$$

where HQ is the calculated hazard quotient, CDI is the chronic daily intake of BPA (mg/kg/day), and RfD is the reference dose for BPA in respect to non-carcinogenic health effects (0.05 mg/kg/day) (28).

Based on US EPA (27), the chronic daily intake (CDI) of BPA (mg/kg/day) through consumption of drinking water was obtained by using Eq. 5.

$$CDI = \frac{C \times IR \times EF \times ED}{BW \times AD} \quad \text{Eq. 5}$$

where CDI is the calculated chronic daily intake of BPA (mg/kg/day), C is the concentration of BPA leached into drinking water (mg/L), IR is the ingestion rate (L/day for water), EF is the exposure frequency (days/year), ED is the exposure duration (years), BW is body weight (kg), and AT is the averaging time (equal to ED x 365 days/ year for non-carcinogens).

In this study, values suggested by the US EPA were incorporated to calculate CDI (Table I); meanwhile, concentrations of BPA leached into drinking water (mg/L) were acquired from analysis. Calculated HQ of less than 1 indicates no adverse health effects. Vice versa, HQ value greater than 1 indicates possibility of developing adverse health effects.

Table I: Values used in risk asssessment

Name	Acronyms	Unit	Value	Reference
Ingestion Rate	IR	L/day	2.4	(29)
Exposure Frequency	EF	days/year	350	(30)
Exposure Duration	ED	years	26 (90^{th} percentile)	(31)
Average Body Weight of Malaysian	BW	kg	62.65	(32)

RESULTS

QC

An average extraction recovery for BPA was 84%. Five-point external calibration curve demonstrated good linearity over concentration range of 0.001 to 1 mg/L; producing correlation coefficient (R2) of 0.9997. IDL, IQL, MDL and MQL were 0.6 µg/L, 1 µg/L, 2 ng/L and 6 ng/L, respectively.

Concentrations of BPA in PC and PET bottled water stored at different temperature and duration

Concentrations of BPA in PC and PET bottled water kept in a car which was exposed to sunlight for different storage durations are summarized in Table II. Measured concentrations of BPA leached from PC and PET water bottles ranged from <MQL to 22.13 ng/L and <MQL to 45.07 ng/L respectively. Concentrations of BPA in PC and PET bottled water kept in a car exposed to sunlight showed steady increased with prolong storage duration (Fig. 1). Maximum concentration of BPA reported in PET bottled water was 45.1 ng/L after eight days of storage. Generally, the concentration of BPA in PET bottled water was higher than PC bottled water. Concentrations of BPA in exposed samples (stored in a car which was exposed to sunlight) were higher than control samples (stored indoor at room temperature).

The average daily car temperature and solar radiation ranged from 29.3 to 33.5°C and 81.7.6 to 124.1 W/m, respectively. The average daily control temperature and solar radiation ranged from 24.3 to 25.8°C and 0.9 to 1.5 W/m, respectively.

BPA migration rate

Increased concentration of BPA found in bottled water does not imply increasing rate of BPA migration. Thus, migration rates for both PC and PET water bottles across different storage conditions were calculated. Fig. 2 showed BPA releasing kinetics curves for both PC and PET bottled water under different storage conditions

Table II: BPA concentrations in PC and PET bottled water stored at conditions.

Storage Conditions			BPA Concentration (ng/L)	
Storage Duration	Storage Temperature (°C)	Storage in the car	PC	PET
1 Week	4°C	-	< MQL	<MQL
	25°C	-	9.13 ± 0.12	11.53 ± 0.23
	50°C	-	13.60 ± 0	16.60 ± 0.40
	75°C	-	23.47 ± 0.42	54.67 ± 0.46
2 Weeks	4°C	-	< MQL	<MQL
	25°C	-	15.07 ± 0.12	18.67 ± 0.50
	50°C	-	47.13 ± 0.12	75.13 ± 0.50
	75°C	-	109.33 ± 0.31	151.07 ± 1.00
4 Weeks	4°C	-	10.00 ± 0	18.87 ± 0.31
	25°C	-	29.27 ± 0.46	44.40 ± 0
	50°C	-	105.80 ± 0.20	131.53 ± 0.90
	75°C	-	168.67 ± 0.23	181.73 ± 0.76
8 Weeks	4°C	-	17.00 ± 0.35	23.33 ± 0.31
	25°C	-	45.07 ± 0.12	66.13 ± 0.23
	50°C	-	168.80 ± 0.60	185.07 ± 0.76
	75°C	-	257.67 ± 0.70	269.87 ± 0.92
1 Day	-	Yes	8.67 ± 0.31	13.53 ± 0.12
2 Days	-		13.07 ± 0.23	20.27 ± 0.12
4 Days	-		19.27 ± 0.31	35.27 ± 0.12
8 Days	-		22.13 ± 0.23	45.07 ± 0.70
1 Day	-	No	< MQL	< MQL
2 Days	-		< MQL	< MQL
4 Days	-		6.67 ± 0.23	7.87 ± 0.12
8 Days	-		10.47 ± 0.31	15.53 ± 0.23

*MQL: Method quantification limit

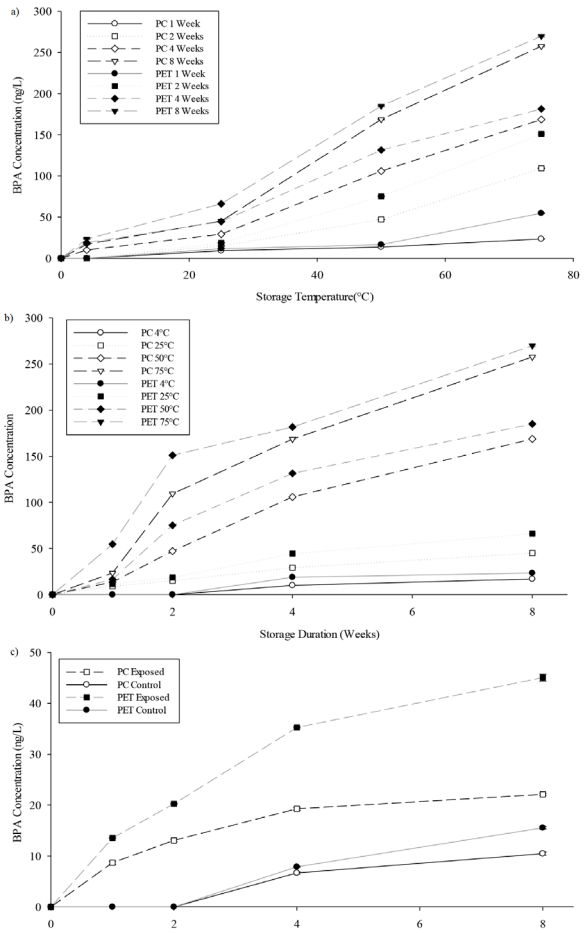

Figure 1: a) Storage temperature versus BPA concentration at different storage durations; b) storage duration versus BPA concentration at different storage temperatures; c) storage duration versus BPA concentration for control and exposed bottled water samples that stored in a car under sun-light

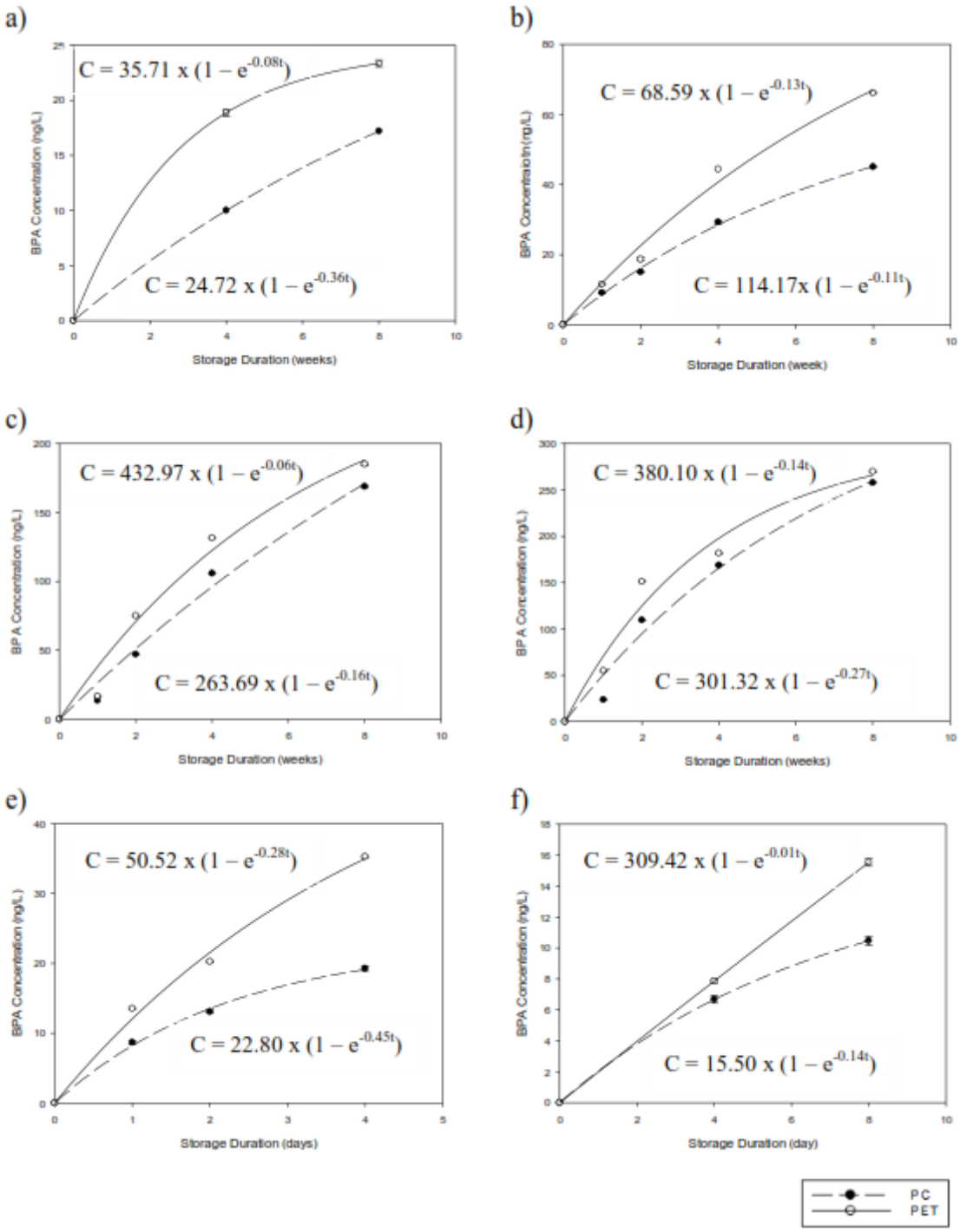

Figure 2: BPA releasing kinetic curves of PC and PET bottled waters that stored for 8 weeks under a) 4 oC, b) 25 oC, c) 50 oC, d) 75 oC; BPA releasing kinetic curves of PC and PET bottled waters that stored for 8 days e) in the car (exposed to sunlight during day time), f) indoor (unexposed to sunlight all the day) at room temperature.

(temperature, duration, and storage in a car). Values of Cmax (ng/L) and b (week-1 or day-1) are tabulated in Table III. In this study, increased storage duration adversely affect migration rates of both PC and PET bottles across varying storage conditions (Table IV).

Health risk assessment

Chronic non-carcinogenic health risks due to BPA exposure were calculated using Hazard Quotient (HQ). Magnitude of Chronic Daily Intake (CDI) was compared to reference dose (RfD) of BPA at 0.05 mg/kg/day. In this study, the highest CDI for PC and PET water bottles was 9.47×10^{-6} mg/kg/day and 9.91×10^{-6} mg/kg/day, respectively, as tabulated in Table V. HQ values for PC bottled water ranged from 4.9×10^{-6} to 1.89×10^{-4}, whereas HQ values for PET bottled water ranged from 5.78×10^{-6} to 1.98×10^{-4} across different storage conditions (Table V).

Table III: Maximum concentrations (C_{max}) and kinetic constant (b_{BPA}) of BPA under different storage conditions

Storage conditions Temperature (°C)	PC		PET	
	C_{max} BPA (ng/L)	bBPA (week^{-1})	C_{max} BPA (ng/L)	bBPA (week^{-1})
4	35.71	0.08	24.72	0.36
25	68.59	0.13	114.17	0.11
50	432.97	0.06	263.69	0.16
75	380.10	0.14	301.32	0.27
Stored in a Car	PC		PET	
	C_{max} BPA (ng/L)	bBPA (week^{-1})	C_{max} BPA (ng/L)	bBPA (week^{-1})
Yes	22.80	0.45	50.52	0.28
No	15.50	0.14	309.42	0.01

Table IV: BPA migration rates under different storage conditions

Storage Temperature (°C)	BPA Migration Rate (ng/week)							
	PC				PET			
	1 Week	2 Week	4 Week	8 Week	1 Week	2 Week	4 Week	8 Week
4	2.70	2.49	2.29	2.11	6.21	4.33	2.1	0.50
25	8.09	7.05	6.16	5.39	11.29	10.11	8.10	5.21
50	25.54	23.98	22.52	21.15	35.25	30.15	22.06	11.80
75	47.22	40.92	35.45	30.71	61.67	47.20	27.65	9.49
Storage in a Car	BPA Migration Rate (ng/day)							
	PC				PET			
	1 Week	2 Week	4 Week	8 Week	1 Week	2 Week	4 Week	8 Week
Yes	6.85	4.17	1.69	0.28	10.80	8.13	4.61	1.48
No	1.89	1.64	1.24	0.71	1.97	1.96	1.93	1.88

Table V: Health risk assessment for ingestion of BPA in PC and PET bottled water stored at different storage conditions

Storage Conditions			CDI		HQ	
Storage Duration	Storage Temperature (°C)	Stored in the Car	PC	PET	PC	PET
1 Week	4	-	-	-	-	-
	25	-	3.36×10^{-7}	4.24×10^{-7}	6.71×10^{-6}	8.47×10^{-6}
	50	-	5.00×10^{-7}	6.10×10^{-7}	9.99×10^{-6}	1.22×10^{-5}
	75	-	8.62×10^{-7}	2.01×10^{-6}	1.72×10^{-5}	4.02×10^{-5}
2 Weeks	4	-	-	-	-	-
	25	-	5.53×10^{-7}	6.86×10^{-7}	1.11×10^{-5}	1.37×10^{-5}
	50	-	1.73×10^{-6}	2.76×10^{-6}	3.46×10^{-5}	5.52×10^{-5}
	75	-	4.02×10^{-6}	5.55×10^{-6}	8.03×10^{-5}	1.11×10^{-4}
4 Weeks	4	-	3.67×10^{-7}	6.93×10^{-7}	7.35×10^{-6}	1.39×10^{-5}
	25	-	1.08×10^{-6}	1.63×10^{-6}	2.15×10^{-5}	3.26×10^{-5}
	50	-	3.89×10^{-6}	4.83×10^{-6}	7.77×10^{-5}	9.66×10^{-5}
	75	-	6.20×10^{-6}	6.68×10^{-6}	1.24×10^{-4}	1.34×10^{-4}
8 Weeks	4	-	6.24×10^{-7}	8.57×10^{-7}	1.25×10^{-5}	1.71×10^{-5}
	25	-	1.66×10^{-6}	2.43×10^{-6}	3.31×10^{-5}	4.86×10^{-5}
	50	-	6.20×10^{-6}	6.80×10^{-6}	1.24×10^{-4}	1.36×10^{-4}
	75	-	9.47×10^{-6}	9.91×10^{-6}	1.89×10^{-4}	1.98×10^{-4}
1 Day	-	Yes	3.18×10^{-7}	4.90×10^{-7}	6.37×10^{-6}	9.94×10^{-6}
2 Days	-		4.80×10^{-7}	7.44×10^{-7}	9.60×10^{-6}	1.49×10^{-5}
4 Days	-		7.08×10^{-7}	1.30×10^{-6}	1.42×10^{-5}	2.59×10^{-5}
8 Days	-		8.13×10^{-7}	1.66×10^{-6}	1.63×10^{-5}	3.31×10^{-5}
1 Day	-	No	-	-	-	-
2 Days	-		-	-	-	-
4 Days	-		2.45×10^{-7}	2.89×10^{-7}	4.90×10^{-6}	5.78×10^{-6}
8 Days	-		3.84×10^{-7}	5.71×10^{-7}	7.69×10^{-6}	1.14×10^{-5}

DISCUSSION

Results obtained in this study were in accordance with those reported by Kubwabo et al. (17), which increased storage duration had increased concentration of BPA leached from PC bottles into water. On the other hand, Fan et al. (22) from Nanjing, China reported increased concentration of BPA leached from PET water bottles with prolong durations; which concentration increased from 23.4 ng/L (1 week of storage) to 59.4 ng/L (4 weeks of storage). BPA concentration measured in this study was higher than those reported by Fan et al. (22) after 4 weeks of storage.

Casajuana and Lacorte (5) reported concentration of BPA in three different brands of PET bottled water sold in Spain. BPA concentration leached were minimal as only 3, 4 and 8 ng/L were detected after 10 weeks of storage at 30oC (5). Guart et al. (18) reported most PC and PET bottles experienced elevated concentrations after one year of storage. It was suggested that migration of plasticizers from bottle caps could be a potential source of BPA in PET bottles. However, concentrations of BPA originating from bottles and cap were not individually analyzed. Source of BPA remained uncertain.

Many studies had reported increasing concentrations of BPA leached with increasing storage temperature (6, 22, 33). Santhi et al. (6) reported that concentration of BPA in PET mineral water after 3 days of storage at room temperature was 3.3 ng/L and it increased to 11.3 ng/L when storage temperature increased to 50oC. In addition, Nam et al. (33) reported that concentration of BPA increased significantly when temperature was increased. PC baby bottle stored at 95°C for 30 min reported an increased from 30 ng/L to 130 ng/L.

There is no specific limit set for the migration of BPA into drinking water from water bottles. However, there is a migration limit for BPA into food which was set by the EC Directive to be 600 ng/g (34). Since BPA is not used in PET manufacturing but only serve as plasticizer and stabilizer, its concentration is anticipated to be lower than those present in PC (3, 35). However, this was contrasted in this study. Due to increasing concerns of health effects due to BPA contamination, the material is now replaced with other alternatives, such as bisphenol S (BPS; 4,4′-sulfonyldiphenol), bisphenol B [BPB; 2,2′-bis(4-hydroxyphenyl)butane], bisphenol F (BPF; 4,4′-dihydroxydiphenylmethane), and bisphenol AF [BPAF; 4,4′-(hexafluoroisopropylidene)diphenol] in the production of PC (36-37). Hence, lower concentrations of BPA found in PC could be due to the reason above.

To the authors' best knowledge, concentration of BPA leached from PC and PET water bottles kept in car exposed to sunlight has not been compared previously. Despite that, Westerhoff et al. (38) reported increasing concentration of antimony in PET plastic bottled water after 7 days of exposure to natural sunlight. In this study, both PC and PET bottled water showed significant differences between exposed samples and the control samples. Rowell et al. (20) detected a maximum BPA concentration of 1.71 µg/L in PC bottled water after 12 days of UV exposure under sunlight (540 W/m2). The UV radiation reading and the concentration of BPA in the PC bottled water in the study by Rowell et al. (20) were higher than the findings reported in this study (UV: 124 W/m2, BPA: 45.07 ng/L).

Fan et al. (22) reported decreased migration rates in 14 out of 16 PET water bottles after 4 weeks of storage, which was similar to current study. Results were also consistent with results found by De Coensel et al. (39), which BPA concentrations leached became constant after few heating cycles of PC baby bottles. Thus, migration rates of BPA stabilized over prolong storage durations.

The highest CDI for PC and PET water bottles was below tolerable daily intake (TDI) and the RfD set by the EU Commission (40) and the U.S. Environmental Protection Agency (28), which is 0.05 mg/kg/day. CDI value for adults was previously reported by Fan et al. (22) for PET bottled water, and the value ($8.94x10^{-6}$ mg/kg/day) was comparable to CDI obtained in this study. Generally, HQ values were less than 1 for all storage conditions and types of water bottles. There was no significant chronic non-carcinogenic health risk for consuming PC and PET bottled water stored in conditions tested in this study. However, the health risk assessment for BPA in bottled drinking water may not fully reflect the effect of exposure to BPA. Humans may be exposed to BPA through many other sources; such as ingestion of BPA-contaminated canned food. Previous studies have reported concentration of BPA in canned food (41-43).

CONCLUSION

Concentrations of BPA detected were below the limit set by EC Directive (600 ng/g). Concentrations of BPA in PET bottled water were higher compared to PC bottled water across all storage conditions. Its concentration increased with prolongs storage duration and higher storage temperature. BPA in samples kept in car were higher than controls which was kept in room temperature. CDI was below the RfD set by the IRIS US EPA at 0.05 mg/kg/day across all conditions. No significant chronic non-carcinogenic health risk arise due to drinking of BPA-contaminated bottled water was found in this study.

ACKNOWLEDGEMENTS

This work was financially supported by funds from Ministry of Higher Education under Fundamental Research Grant Scheme (project number 5524781). The authors would like to thank Department of Environmental and Occupational Health, Universiti Putra, Malaysia, for use of UHPLC-FLD during analysis.

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