

Potential risk of BPA and phthalates in commercial water bottles: a minireview

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ABSTRACT

The global water bottling market grows annually. Today, to ensure consumer safety, it is important to verify the possible migration of compounds from bottles into the water contained in them. Potential health risks due to the prevalence of bisphenol A (BPA) and phthalates (PAEs) exposure through water bottle consumption have become an important issue. BPA, benzyl butyl phthalate (BBP), di-*n*-butyl phthalate (DBP) and di (2-ethylhexyl) phthalate (DEHP) can cause adverse effects on human health. Papers of literature published in English, with BPA, BBP, DBP and DEHP detections during 2017, by 2019 by liquid chromatography and gas chromatography analysis methods were searched. The highest concentrations of BPA, BBP, DBP and DEHP in all the bottled waters studied were found to be 5.7, 12.11, 82.8 and 64.0 µg/L, respectively. DBP was the most compound detected and the main contributor by bottled water consumption with 23.7% of the Tolerable Daily Intake (TDI). Based on the risk assessment, BPA, BBP, DBP and DEHP in commercial water bottles do not pose a serious concern for humans. The average estrogen equivalent level revealed that BPA, BBP, DBP and DEHP in bottled waters may induce adverse estrogenic effects on human health.

Key words | bisphenol A, estrogenic effects, phthalates, risk assessment, water bottles

HIGHLIGHTS

- DBP was the most compound detected.
- An estimated intake of BPA, BBP, DBP and DEHP was far below their TDIs.
- The risk assessment of BPA, BBP, DBP and DEHP does not raise serious concern for humans.
- The average estrogen equivalent level for BPA, BBP, DBP and DEHP may induce adverse estrogenic effects on human health.
- BPA, BBP, DBP and DEHP in bottled water need more accurate data to avoid their effects on human health.

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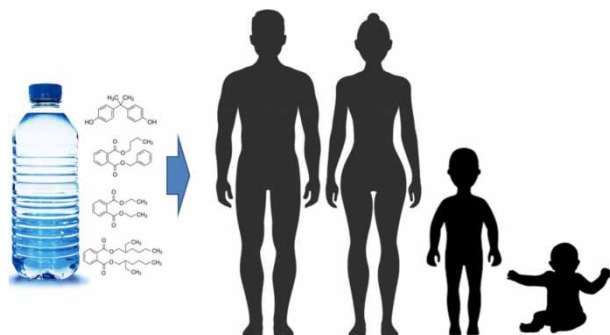
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doi: 10.2166/wh.2021.202

GRAPHICAL ABSTRACT



INTRODUCTION

Reports show that, in 2018, 64% of produced bottles were made of polyethylene terephthalate (PET), 34% of high-density polyethylene (HDPE), 1.8% of polypropylene and 1% other (polycarbonate (PC) included here) (ACC 2019). According to the American Chemistry Council (ACC), in 2018, 0.31 million pounds of postconsumer PC bottles were collected for recycling. PET and HDPE continued to dominate as selected resins to produce plastic bottles (97.1% by weight of produced bottles has made of PET or HDPE) (ACC 2019).

The bottled water industry is a phenomenon in practically every region of the world. First, bottled water became a mainstream commercial beverage category in Western Europe and later grew into a truly global beverage (IBWA 2018). The bottled industry produces mainly two types of packaged water: packaged natural mineral drinking water and packaged drinking water. The last is water derived from any source of a potable water (ground, well, bore well water, etc.), which must be subjected to different treatment processes such as filtration, aeration, decantation, and reverse osmosis (Jain *et al.* 2019). In 2018, for the first time, global bottled water consumption has surpassed that 100 billion gallons is estimated to, and the per capita consumption exceeded 42 gallons (158,987 liters). It should be stressing that per capita consumption by individual regions or countries can differ from the global average (IBWA 2019). In 2018, the rank of the 10 leading countries' consumption was China, United

States, Mexico, Indonesia, Brazil, India, Thailand, Germany, Italy and France, respectively (IBWA 2019).

In 2018, approximately 7.7% (27.64 million tons out of the total plastic production of 359 million tons) of the plastic demand was constituted by PET worldwide was used in bottles for water, soft drinks, juices, and cleaners (Plastics Europe 2020). PET is the packaging most used in water bottles (Coniglio *et al.* 2020). PET and PC as the packing materials have been widely used for Chinese bottled water (Wang *et al.* 2020).

Bisphenol A (BPA), benzyl butyl phthalate (BBP), di-*n*-butyl phthalate (DBP) and di (2-ethylhexyl) phthalate (DEHP) have recently been detected in commercial water bottles raising concerns and discussions on possible risks for human health (Dada *et al.* 2018; Pinsrithong & Bunkoed 2018; Karayaka *et al.* 2019; Wu *et al.* 2019). Many countries included BPA, BBP, DBP and DEHP in the priority list of pollutants (Pignotti *et al.* 2017; Goeuru *et al.* 2019; Li *et al.* 2019; Fard *et al.* 2020). Acceptable exposure levels for these compounds have been created to protect human health (Čelić *et al.* 2020; Fard *et al.* 2020). The maximum contaminant level (MCL) is the highest level of a contaminant that is allowed in drinking water (US EPA 2021). The MCL for drinking water for BPA, BBP, DBP and DEHP is in the section 'Extraction techniques for detection'.

According to Hassan *et al.* (2020), BPA and PAEs exhibit similar toxicogenomics and health effects. How BPA and PAEs are not bound to the matrix, they can leach out into

the surroundings by delicate changes in the environment, like temperature, pH and pressure alterations (Hassan *et al.* 2020). The Regulation (EU) No. 10/2011 (EC 2011) defines the Specific Migration Limit (SML) as the maximum permitted amount of a given substance released from a material or article into food or food simulants. The SML values by the EU for BBP, DBP and DEHP are 30, 0.3 and 1.5 mg/kg, respectively (EFSA 2019). The detection of very low BPA, BBP, DBP and DEHP in water can be carry out by high-performance liquid (HPLC) and gas (GC) chromatography (Gorji *et al.* 2019; Karayaka *et al.* 2019; Li *et al.* 2019; Yin *et al.* 2019). The detection power can be improved by preconcentrating analytes before instrumental measurement and the type of detector (Kumar *et al.* 2014; Chang *et al.* 2017; Farajzadeh *et al.* 2019; Karayaka *et al.* 2019; Li *et al.* 2019).

In this context, due to the increasing popularity of bottled water consumption, the potential health effects of possible migration of chemical compounds from the bottles into the water can pose a health risk to consumers. The purpose of this minireview is to verify if recent BPA, BBP, DBP and DEHP detections in commercial water bottles around the world using HPLC and gas GC may pose a risk to human health.

Papers of literature published in English, that detected BPA and PAEs (BBP, DBP and DEHP) in commercial bottles during 2017, by 2019 were searched. Papers with storage studies were also taken into account. For data sources for further analysis were identified a total of 41 publications from 17 countries. PC bottles were not considered. Thus, this work hopes to aid decision-making in future research focusing on BPA, BBP, DBP and DEHP in commercial water bottles using HPLC and GC. Moreover, this review hopes to avoid consumer exposure to these chemicals and to guarantee consumer safety.

BPA AND PAES IN PET BOTTLED WATER

The production process of water bottles uses PC plastics containing BPA (antioxidant or monomer) (Alfarhani *et al.* 2019; Fikarová *et al.* 2019; Liu *et al.* 2019). Although BPA is not used in the manufacture of PET, it should consider the use of recycled PET (R-PET) as a possible source of BPA

coming from cross-contamination, not only during the recycling process but also during the manufacture of virgin PET (Dreolin *et al.* 2019). BPA leachable from polymer packaging due to its moderate water solubility (120–300 mg/L: pH 7.0 at 25 °C) and low log K_{ow} (3.32) in water (Borrirukwisitsak *et al.* 2012; Fikarová *et al.* 2019). Guart *et al.* (2011) not detected BPA in PET bottles cut in pieces, but on the other hand, detected BPA in HDPE caps at concentrations of 0.145 µg/dm². Bach *et al.* (2012) also indicated that the containers' caps, in PET bottled water, could be a source of BPA.

The manufacturing of beverage bottles widely uses PAEs (Li *et al.* 2019) and like they are not chemically bound to polymers, they may also enter drinking samples. This process can occur through the production, packaging and storage (bottling lines and water refinement centers) (Manzo *et al.* 2019; Pacyga *et al.* 2019). According to Bach *et al.* (2014), background pollution, as a source of PAEs, cannot be excluded. PAEs' presence in PET bottled water can be associated with PAEs in the source of water (groundwater or tap water) used to fill in the bottles (Jeddi *et al.* 2015). The type of closure ('cap') on the bottles could be a more important source of PAEs than the bottle material (glass or PET) (EFSA 2019). The caps of plastic bottles are made of high- and low-density polyethylene (HDPE and LDPE) and polystyrene (PS) (Guart *et al.* 2011). Guart *et al.* (2011) identified BPA in HDPE, LDPE and PS plastics. The adhesive used for sticking the bottle labels could thus be considered one of the sources of PAEs in water samples (Cincotta *et al.* 2018). Aznar *et al.* (2011) identified DBP and DEHP in adhesive based on vinyl acetate-ethylene.

PAEs are hydrophobic organic compounds under normal conditions (25 °C), very insoluble in water (BBP: 2.69 mg/L, DBP: 11.2 mg/L and DEHP: 0.27 mg/L) and have a particular affinity for fats and alcohols (Grinbaum *et al.* 2019; PubChem 2020). However, exposure to these low levels in water may also cause significant risks to humans under long-term chronic exposure by resulting in a considerable total health risk (Abtahi *et al.* 2019; Chen *et al.* 2019a; Abdelghani *et al.* 2020). Exposure to that low level can cause problems such as spasms in arms and legs, bronchial obstruction in children, irritation of the eyes and endocrine disruption (Abdelghani *et al.* 2020).

CHROMATOGRAPHIC AND EXTRACTION TECHNIQUES FOR DETECTION

Chromatographic techniques for detection

A wide range of methods analyzes BPA and PAEs. The liquid chromatography (LC) and gas chromatography (GC) analysis methods for detection and respective extraction techniques used for the determination in commercial water bottles are presented in Table 1. The choice of the detector and extraction influences the detection limit (LOD) and the quantification limit (LOQ) values obtained.

HPLC coupled with diode-array detection (HPLC–DAD) was the most used in BPA detections. HPLC is adequate for the analysis of BPA since it is a relatively polar compound. The DAD detector allows simultaneous collection of chromatograms over a range of wavelengths during a single run, providing more information on sample composition than is provided by the use of a single wavelength detector (Waksmundzka-Hajnos & Sherma 2010). DAD is preferable since it is sufficiently selective for compound identification (McGowin 2006).

GC coupled with mass spectrometry (GC–MS) was the most used technique in PAEs detections. GC can separate volatile and semi-volatile compounds with high resolution, and its combination with MS can identify them, providing detailed structural information on most compounds such that they can be identified correctly (Hussain & Maqbool 2014). Only Karayaka *et al.* (2019) analyzed BPA by GC–MS and derivatization is not used. BPA has volatility and thermal stability suitable for detection and quantification by GC–MS. However, derivatization can improve the sensitivity, selectivity and performance of the chromatographic properties (Nollet 2005).

BPA analysis underivatized by GC–MS can be found in the literature because sensitivity can be improved using pre-concentration and liquid–liquid extraction (Oca *et al.* 2013). Karayaka *et al.* (2019) used the switchable liquid–liquid microextraction (SLLME) to pre-concentrate BPA and improving the detection power of GC–MS. Microextraction methods are eco-friendly because they use too small quantities of chemicals, no compromising extraction efficiency and agree with green chemistry (Armenta *et al.* 2015).

Extraction techniques for detection

The sample preparation has been considered as the Achilles' heel (Fumes *et al.* 2015). Matrix-related compounds can be co-extracted and can interfere in the analysis; so, the sample preparation has a multifarious role related to target analyte extraction, pre-concentration and clean-up from co-existing species (Gao *et al.* 2015). A pre-concentration step is usually necessary before the final analysis of compounds (Gao *et al.* 2015; Feizi *et al.* 2017). However, some methods often require high amounts of organic solvents that are harmful to the environment (Gao *et al.* 2015; Feizi *et al.* 2017; Plotka-Wasyłka *et al.* 2017). A concept that has been approached is the green analytical chemistry, which decreases or eliminates organic solvents during the extraction procedure (Fumes *et al.* 2015; Plotka-Wasyłka *et al.* 2017). Karayaka *et al.* (2019) developed a method to extract BPA from drinking water bottles using a switchable polarity solvent (*N,N*-dimethylbenzylamine), which is a green solvent. Also, it is very important to use a proper sample preparation to reach the required lower LODs (Gao *et al.* 2015). Discoveries in materials science may supply new tools for the preparation of samples (Jalili *et al.* 2020). Mohammadnezhad *et al.* (2017) developed ionic liquid-bonded fused silica as a new solid-phase microextraction (SPME) fiber for the liquid chromatographic determination of BPA in mineral water bottled in PET. Wei *et al.* (2018) synthesized a novel magnetic solid-phase extraction (MSPE) for the determination of six phthalic acid esters in mineral water (including BBP, DBP and DEHP). The development of natural sorbents has also been investigated, which are cheap and readily available and sometimes their performance was comparable with synthetic sorbents (Sajid *et al.* 2016).

Some works in Table 1 developed extraction methods. González-Sálamo *et al.* (2017) used the first application of core-shell poly (dopamine) magnetic nanoparticles as a sorbent for the extraction of a group of 11 phthalic acid esters of interest. Pinsrithong & Bunkoed (2018) synthesized a hierarchically porous composite nanostructure of polypyrrole, reduced graphene oxide, magnetite nanoparticles and alginate hydrogel microspheres (PPy-rGOx-Fe₃O₄). They applied as a magnetic solid-phase extraction

Table 1 | Extraction methods for the determination of BPA, BBP, DBP and DEHP in commercial water bottles**Without migration study**

Detected analyte (s)	Extraction method	Chromatographic technique	LODs ($\mu\text{g/L}$)	LOQs ($\mu\text{g/L}$)	Reference
BPA	SPE	UFLC-MS/MS	0.004–0.055 ^a	1.4×10^{-2} – 1.2×10^{-2a}	Zhou <i>et al.</i> (2019)
BPA	SBSE	HPLC-UV/Vis	0.02	0.06	Gorji <i>et al.</i> (2019)
BPA	USAE-MIP- μ -SPE	HPLC-DAD	0.07	0.15	Rozaini <i>et al.</i> (2017)
BPA	MDMIP- SPE	HPLC-DAD	0.083	0.114	Chang <i>et al.</i> (2017)
BPA	SPME	HPLC-DAD	0.20	Not stated	Mohammadnezhad <i>et al.</i> (2017)
BPA	SLLME	GC-MS	0.54	1.8	Karayaka <i>et al.</i> (2019)
BPA BBP; DBP; DEHP	LLE	LC-MS/MS GC-MS	Not stated	Not stated	Wu <i>et al.</i> (2019)
BBP; DBP	MIP-SPE	HPLC-MS	0.16; 0.84	0.55; 2.81	Barciela-Alonso <i>et al.</i> (2017)
BBP; DBP	IT-UAA-LLME	GC-MS	1.67; 0.75	5.50; 2.46	Farahani <i>et al.</i> (2017)
BBP; DBP	m- μ dSPE	UHPLC-MS/MS	Not stated	6×10^{-3} ; 11×10^{-3}	Santana-Mayor <i>et al.</i> (2018)
BBP; DBP; DEHP	MSPE	HPLC-UV/Vis	0.0103; 0.003 ^b ; 0.0167	0.0342; 0.022 ^b ; 0.0556	Yin <i>et al.</i> (2019)
BBP; DBP; DEHP	SPE	GC-MS/MS	0.18; 0.021; 0.036	0.60; 0.070; 0.12	Li <i>et al.</i> (2019)
BBP; DBP; DEHP	LLE	GC-MS/MS	1.0; 1.0; 0.5	3.0; 3.0; 0.15	Tran-Lam <i>et al.</i> (2018)
BBP; DBP; DEHP	MSPE	GC-MS	5.0; 1.0; 5.0	Not stated	Wei <i>et al.</i> (2018)
DBP	TSP-LLME	GC-MS	0.007	0.021	Chen <i>et al.</i> (2019b)
DBP	SVA-LLME	GC-MS	0.15	0.50	Mohebbi <i>et al.</i> (2017)
DBP	DSPE-DLLME	GC-FID	1.24	4.11	Farajzadeh <i>et al.</i> (2019)
DBP	SPE	HPLC-UV/Vis	2.4	7.9	Salazar-Beltrán <i>et al.</i> (2017)
DBP	MISPME	HPLC-UV/Vis	3	10	Soheilifar <i>et al.</i> (2018)
DBP	m- μ dSPE	GC-MS/MS	Not stated	0.009	González-Sálamo <i>et al.</i> (2017)
DBP	LLE	HPLC-UV/Vis	Not stated	Not stated	Dada <i>et al.</i> (2018)
DBP	HF-LPME	GC-MS/MS	Not stated	Not stated	González-Sálamo <i>et al.</i> (2018)
DBP; DEHP	MEPS-DLLME	GC-FID	0.001; 0.005	0.003; 0.015	Amiri & Ghaemi (2017)
DBP; DEHP	MSPE	GC-MS/MS	0.005; 0.008	0.02; 0.03	Pinsrithong & Bunkoed (2018)
DBP; DEHP	RDSE	GC-MS	0.01; 0.03	0.04; 0.10	Manzo <i>et al.</i> (2019)
DBP; DEHP	LLE	GC-MS	0.01–0.05 ^c	0.03–0.15 ^c	Tri <i>et al.</i> (2018)
DBP; DEHP	MEPS	GC-FID	0.05; 0.10	0.10; 0.25	Amiri <i>et al.</i> (2017)
DBP; DEHP	LLE	GC-FID	Not stated	Not stated	Szendi <i>et al.</i> (2018)
DEHP	DMIMS-SPE	GC-MS	0.00039	0.0013	Özer <i>et al.</i> (2017)
DEHP	MSPE	GC-FID	0.02	Not stated	Chahkandi & Amiri (2019)
DEHP	DLLME	GC-FID	2	4	Notardonato <i>et al.</i> (2018)
Detected analyte (s)	Extraction method	Chromatographic technique	LODs (mg/kg)	LOQs (mg/kg)	Reference

(continued)

Table 1 | continued

Without migration study

Detected analyte (s)	Extraction method	Chromatographic technique	LODs ($\mu\text{g/L}$)	LOQs ($\mu\text{g/L}$)	Reference
BBP; DBP; DEHP	LLE	GC-MS	0.00031; 0.00025; 0.00042	0.00096; 0.0008; 0.00122	Yang <i>et al.</i> (2017)
With migration study					
Detected analyte (s)	Extraction method	Chromatographic technique	LODs ($\mu\text{g/L}$)	LOQs ($\mu\text{g/L}$)	Reference
BPA	PT- μ -SPE	HPLC-FLD	0.001	0.0032	Kaykhaii <i>et al.</i> (2020)
BBP; DBP; DEHP	μ SPE	GC-FID	0.025; 0.017; 0.031	Not stated	Abtahi <i>et al.</i> (2019)
BBP; DBP; DEHP	Not stated	LC-MS/MS	0.20; 0.20; 0.30	0.64; 0.60; 0.94	Surhio <i>et al.</i> (2017)
DBP	MIP-SPME	GC-FID	0.12	Not stated	Hashemi-Moghaddam & Maddah (2018)
DBP	HS-SPME	GC-MS	0.17	0.57	Cincotta <i>et al.</i> (2018)
DBP; DEHP	SPE	GC-MS	0.015 ^d	Not stated	Sulentici <i>et al.</i> (2018)
DBP; DEHP	LLE	GC-MS/MS	0.043; 0.062	Not stated	Zaki & Shoeib (2018)
DBP; DEHP	AALLME	GC-MS	0.3; 0.2	Not stated	Yousefi <i>et al.</i> (2019)
DEHP	UA-DLLME	GC-MS	10–100 ^e	50–500 ^e	Annamalai & Namasivayam (2017)

BPA, bisphenol A; BBP, benzylbutyl phthalate; DBP, di-*n*-butyl phthalate; DEHP, di(2-ethylhexyl) phthalate; SPE, solid-phase extraction; SBSE, stir bar sorptive extraction; USAE, ultrasound-assisted emulsification; MIP, molecularly imprinted polymer; μ -SPE, micro-solid-phase extraction; MDMIP, magnetic dummy molecularly imprinted polymer; SPME, solid-phase microextraction; SLLME, switchable liquid-liquid microextraction; LLE, liquid-liquid microextraction; IT-UAA, in tube ultrasonic and air-assisted; LLME, liquid-liquid microextraction; m- μ DSPE, magnetic micro-dispersive solid-phase extraction; MSPE, magnetic solid-phase extraction; TSP, temperature-sensitive polymer; SVA, solvent vapor-assisted; DSPE, dispersive solid-phase extraction; DLLME, dispersive liquid-liquid microextraction; MISPME, molecularly imprinted solid-phase microextraction; HF-LPME, hollow fiber liquid-phase microextraction; MEPS, microextraction in packed syringe; RDSE, rotating disk sorptive extraction; DMIMS, dual-template molecularly imprinted mesoporous silica; PT- μ -SPE, pipette-tip micro-solid-phase extraction; μ SPE, micro-solid-phase extraction; HS, headspace; AALLME, air-assisted liquid-liquid microextraction; UA, ultrasound-assisted; GC, gas chromatography; MS, mass spectrometry; HPLC, high-performance liquid chromatography; DAD, diode-array detection; FID, flame ionization detector; UV/Vis, dual-wavelength ultraviolet/visible; MS/MS, tandem mass spectrometry; UHPLC, ultra-high-performance liquid chromatography; UFLC, ultra-fast liquid chromatography; UPLC-MS, ultra-performance liquid chromatography-mass spectrometry; FLD, fluorescence detector.

^aCorresponds to four bisphenols – BPA; BBP: bisphenol B; BPF: bisphenol F; BPS: bisphenol S.

^bCorresponds to two phthalates – DBP: bis(2-butoxyethyl) phthalate/DBP.

^cCorresponds to nine phthalates – DEP: diethyl phthalate; DPP: dipropyl phthalate; DiBP: di-*i*-isobutyl phthalate; DBP; DCHP: dicyclohexyl phthalate; DnHP: dihexyl phthalate; BzBP: benzyl butyl phthalate; DnOP: di(*n*-octyl)phthalate; DEHP.

^dCorresponds to three phthalates – DiBP; DBP; DEHP.

^eCorresponds to 14 phthalates – BBP; DBP; DEHP; DPP; DEP; DiBP; DCHP; DNOP: di-*n*-octyl phthalate; DMP: dimethyl phthalate; DHP: dihexyl phthalate; DiNP: di-isononyl phthalate; DiDP: di-isodecyl phthalate; BBEP: bis (2-*n*-butoxyethyl) phthalate; BMEP: bis (2-methoxy ethyl) phthalate.

adsorbent for PAEs, including BBP, DBP and DEHP. Farajzadeh *et al.* (2019) developed a natural and costless adsorbent for the accomplishment of a dispersive solid-phase extraction (DSPE) procedure followed by dispersive liquid-liquid microextraction (DLLME) for the extraction and preconcentration of PAEs and alkylphenols. None of the methods of Table 1 present LOD and LOQ values lower than the MCL to BPA in drinking water by EC (0.1 $\mu\text{g/L}$), but are lower than in China (10 $\mu\text{g/L}$) (EC 2020; GB-5749-2006).

Currently, MCL has not been established for BBP (US EPA 2019a), although, in 1990, US EPA proposed an MCL of 100 $\mu\text{g/L}$ (Parks *et al.* 1993). In 2004, New Jersey State Primary and Secondary Drinking Water Standards derived the same value, multiplying the drinking water equivalent level of 7 mg/L by the relative source contribution factor of 20% and dividing the result by the additional uncertainty factor of 10 for possible human carcinogens (NJDEP 2004). All methods show LOD and LOQ below this proposed MCL value for BBP.

Almost all methods exhibit LOD and LOQ lower than DBP by China for drinking water (3 µg/L) (GB-5749-2006). All methods show LOD and LOQ lower than DEHP by US FDA for bottled water (6 µg/L) and by WHO, Codex Alimentarius, China for drinking water (8 µg/L) (Codex Alimentarius 2001; GB-5749-2006; WHO 2017; ECFR 2020). Yang *et al.* (2017) analyzed BBP, DBP and DEHP by GC-MS/LLE. The values of LOD and LOQ are given in mg/kg. The LOD and LOQ are lower than the SML values by the EU for BBP (30 mg/kg), DBP (0.3 mg/kg) and DEHP (1.5 mg/kg) (EFSA 2019).

DETECTIONS OF BPA AND PAES

The detected levels of BPA, BBP, DBP and DEHP in commercial water bottles without the storage study are present in Table 2 and Figure 1. Wei *et al.* (2018) and Yang *et al.* (2017) are not included in Figure 1 because the units are in mg/kg. The detected levels of BPA, BBP, DBP and DEHP in commercial water bottles with the storage study are present in Table 3 and Figure 2. For articles with concentration ranges, averages were used to generate Figures 1 and 2. A better understanding of the methods used in storage studies can be verified in their respective articles.

Some papers presented values above the MCL to BPA (0.1 µg/L) by EC (EC 2020). All the papers exhibited levels lower than MCL to BPA by China (GB-5749-2006). Even though there is no specific legislation for BBP so far, all the papers showed levels lower than MCL of 100 µg/L proposed by US EPA. It should be noted that this is a proposed value and has not been defined as a standard, but the proposed value serves to analyze the results for the moment. Almost all papers exhibited DBP levels low than 3 µg/L (GB-5749-2006) and displayed DEHP levels low than 6 or 8 µg/L (Codex Alimentarius 2001; GB-5749-2006; WHO 2017; ECFR 2020). The values of BBP, DBP and DEHP obtained by Wei *et al.* (2018) and Yang *et al.* (2017) are lower than the SML values by the EU for BBP (30 mg/kg), DBP (0.3 mg/kg) and DEHP (1.5 mg/kg) (EFSA 2019).

The countries with the reported highest levels of BPA, BBP, DBP and DEHP were Turkey (5.7 µg/L – Figure 1), Pakistan (12.11 µg/L – Figure 2), Mexico (82.8 µg/L –

Figure 1) and Thailand (64.0 µg/L – Figure 1), respectively. The PAE values detected were highest than those established by legislation. Thailand also was the country with the first rank with DEHP (94.1 µg/L) in bottled waters in the review by Luo *et al.* (2018). The value was obtained by Uansiri *et al.* (2016) in bottled water contained in plastic containers. DEHP is known as a dominant PAE in bottled water (Keresztes *et al.* 2013; Guart *et al.* 2014; Zaki & Shoeib 2018; Abtahi *et al.* 2019).

DBP was the most compound detected. Luo *et al.* (2018) also verified that DBP was the PAE with more detection frequency in bottled water. All the samples (10 brands) analyzed by Soheilifar *et al.* (2018) present DBP. Among 16 PAEs studied by Zhang *et al.* (2018), DBP was the most ubiquitous and dominant contaminant in the study population. Soheilifar *et al.* (2018) optimized a molecularly imprinted polymer as a highly selective sorbent toward DBP. Dada *et al.* (2018) also analyzed packaged sachet water, and DBP concentrations were almost four-time higher (160 µg/L) relative to bottled water. Sachet water is packaging in plastic bags (Semey *et al.* 2020) made of LDPE (Jnr *et al.* 2018), and it is relatively cheaper than a water bottle (Dada *et al.* 2018).

Kaykhani *et al.* (2020) verified that the water sample presented more BPA migration (Figure 2) when brought to boiling in a steel jar, quickly poured into the bottle and after cooling at ambient temperature (Figure 2). Surhio *et al.* (2017) detected the highest value of BBP migration studied with the influence of sunlight in Pakistan (Figure 2). The intensity of sunlight may affect the degradation degree of PAEs (Lertsirisopon *et al.* 2009), and the occurrence of PAEs in water stored in PET bottles depended mainly on the country of origin of the bottle (Schmid *et al.* 2008; Keresztes *et al.* 2013). All the papers that specified the type of bottle demonstrated DBP levels above the MCL (3 µg/L). Yousefi *et al.* (2019) also studied PET bottled water exposed to sunlight and as well as Surhio *et al.* (2017) verified an increase in DBP concentration. DBP values at room temperature were lower than at freezing for Hashemi-Moghaddam & Maddah (2018), while the reverse occurred for Sulentic *et al.* (2018). The presence of DBP may be due to different production facilities used by the different brands tested (Al-Saleh *et al.* 2011; Guart *et al.* 2014). Annamalai & Namasivayam (2017) obtained bigger values to DEHP at 4 °C and smaller values at 37 °C. To Zaki &

Table 2 | Levels of BPA, BBP, DBP and DEHP in commercial water bottles without the storage study

Detected analyte	Sample	Country	Type of bottle	Number of brands or samples	Concentration ($\mu\text{g/L}$)	Reference
BPA	Drinking water bottle	Turkey	Not stated	3	5.7	Karayaka <i>et al.</i> (2019)
BPA	Mineral water bottle	Iran	PET ^a	1	5.5	Mohammadnezhad <i>et al.</i> (2017)
BPA	Mineral water	Malaysia	Not stated	6	1.25	Rozaini <i>et al.</i> (2017)
BPA	Plastic bottled mineral water	China	Not stated	1	0.127	Chang <i>et al.</i> (2017)
BPA	Bottled mineral water	Iran	Not stated	3	0.07	Gorji <i>et al.</i> (2019)
BPA	Bottled water	China	Not stated	Not stated	0.05–0.08	Zhou <i>et al.</i> (2019)
BPA	Bottled water	China	Not stated	17	0.01	Wu <i>et al.</i> (2019)
BBP	Mineral water	Iran	PET	3	2.9–5.5	Farahani <i>et al.</i> (2017)
BBP	Bottled water	China	Not stated	17	1.86	Wu <i>et al.</i> (2019)
BBP	Bottled water in plastic	Spain	Not stated	4	0.75–1.9	Barciela-Alonso <i>et al.</i> (2017)
BBP	Mineral water	China	Not stated	5	0.515–0.690	Yin <i>et al.</i> (2019)
BBP	Mineral water	Vietnam	Not stated	14	0.30–0.95	Tran-Lam <i>et al.</i> (2018)
BBP	Bottled drinking water	China	Not stated	60	0.019–0.032	Li <i>et al.</i> (2019)
BBP	Mineral water bottled in plastic	Spain	Not stated	1	<LOQ	Santana-Mayor <i>et al.</i> (2018)
DBP	Plastic bottled Water	Nigeria	Not stated	15	42	Dada <i>et al.</i> (2018)
DBP	Drinking water	Mexico	PET	10	20.5–82.8	Salazar-Beltrán <i>et al.</i> (2017)
DBP	Mineral water	China	Not stated	5	8.98–11.5	Yin <i>et al.</i> (2019)
DBP	Bottled water in plastic	Spain	Not stated	4	4.6–8.2	Barciela-Alonso <i>et al.</i> (2017)
DBP	Plastic bottled water	Thailand	Not stated	1	17.0	Pinsrithong & Bunkoed (2018)
DBP	Plastic bottled water	Iran	Not stated	1	5.2	Mohebbi <i>et al.</i> (2017)
DBP	Mineral water	Cyprus	Not stated	Not stated	4.35	Farajzadeh <i>et al.</i> (2019)
DBP	Bottled mineral water	Iran	Not stated	1	4.5	Amiri & Ghaemi (2017)
DBP	Mineral water	China	Not stated	1	2.68	Chen <i>et al.</i> (2019b)
DBP	Bottled water	China	Not stated	17	1.34	Wu <i>et al.</i> (2019)
DBP	Mineral water	Iran	Not stated	3	1.1–2.5	Amiri <i>et al.</i> (2017)
DBP	Mineral water	Iran	PET	3	1.1–1.7	Farahani <i>et al.</i> (2017)
DBP	Mineral water	Spain	Not stated	1	<1	González-Sálamo <i>et al.</i> (2018)
DBP	Mineral bottled water	Spain	PET	1	0.36	González-Sálamo <i>et al.</i> (2017)
DBP	Water packed in plastic bottle (still, sparkling and light sparkling)	Chile	Not stated	5 (2 – still, 2 – sparkling, 1 – light sparkling)	0.353–2.756	Manzo <i>et al.</i> (2019)
DBP	Plastic bottled water	Iran	Not stated	10	0.26–1.13	Soheilifar <i>et al.</i> (2018)
DBP	Plastic bottled beverages (water)	Vietnam	Not stated	8	0.24–1.86	Tri <i>et al.</i> (2018)

(continued)

Table 2 | continued

Detected analyte	Sample	Country	Type of bottle	Number of brands or samples	Concentration (µg/L)	Reference
DBP	Mineral water bottled in plastic	Spain	Not stated	1	0.184	Santana-Mayor <i>et al.</i> (2018)
DBP	Mineral water	Vietnam	Not stated	14	0.09–0.95	Tran-Lam <i>et al.</i> (2018)
DBP	Bottled drinking water	China	Not stated	60	0.021–0.51	Li <i>et al.</i> (2019)
DBP	Bottled mineral water	Hungary	PET	4	<0.005–0.2	Szendi <i>et al.</i> (2018)
DEHP	Plastic bottled water	Thailand	Not stated	1	64.0	Pinsrithong & Bunkoed (2018)
DEHP	Bottled water	Italy	Not stated	2	22.9–24.4	Notardonato <i>et al.</i> (2018)
DEHP	Plastic bottled beverages (water)	Vietnam	Not stated	8	10.3–42.3	Tri <i>et al.</i> (2018)
DEHP	Plastic bottled water	Turkey	Not stated	Not stated	10.06–11.90	Özer <i>et al.</i> (2017)
DEHP	Bottled mineral water	Iran	Not stated	1	3.0	Amiri & Ghaemi (2017)
DEHP	Bottled mineral water	Iran	Not stated	2	2.6	Chahkandi & Amiri (2019)
DEHP	Bottled water	China	Not stated	17	2.50	Wu <i>et al.</i> (2019)
DEHP	Water packed in plastic bottle (still, sparkling and light sparkling)	Chile	Not stated	5 (2 – still, 2 – sparkling, 1 – light sparkling)	1.258–4.321	Manzo <i>et al.</i> (2019)
DEHP	Mineral water	Iran	Not stated	3	0.5–3.5	Amiri <i>et al.</i> (2017)
DEHP	Mineral water	Vietnam	Not stated	14	0.46–1.8	Tran-Lam <i>et al.</i> (2018)
DEHP	Mineral water	China	Not stated	5	<LOQ–0.733	Yin <i>et al.</i> (2019)
DEHP	Bottled mineral water	Hungary	PET	4	<0.29–11.289	Szendi <i>et al.</i> (2018)
DEHP	Bottled drinking water	China	Not stated	60	0.013–0.021	Li <i>et al.</i> (2019)
Detected analyte	Sample	Country	Type of bottle	Number of brands or samples	Concentration (mg/kg)	Reference
BBP	Mineral water	China	Not stated	1	0.001	Wei <i>et al.</i> (2018)
DBP					0.014	
DEHP					0.018	
BBP	Mineral water Soda water	China	Not stated	Not stated	0.32 × 10 ⁻⁴ – 1.1 × 10 ⁻⁴ <LOD–1.3 × 10 ⁻⁴	Yang <i>et al.</i> (2017)
DBP	Mineral water Soda water				1.3 × 10 ⁻⁴ – 10.2 × 10 ⁻⁴ 1.6 × 10 ⁻⁴ – 63.4 × 10 ⁻⁴	
DEHP	Mineral water Soda water				2.2 × 10 ⁻⁴ – 43.9 × 10 ⁻⁴ 5.7 × 10 ⁻⁴ – 72.9 × 10 ⁻⁴	

BPA, bisphenol A; BBP, benzylbutyl phthalate; DBP, di-*n*-butyl phthalate; DEHP, di(2-ethylhexyl) phthalate.^aPolyethylene terephthalate.

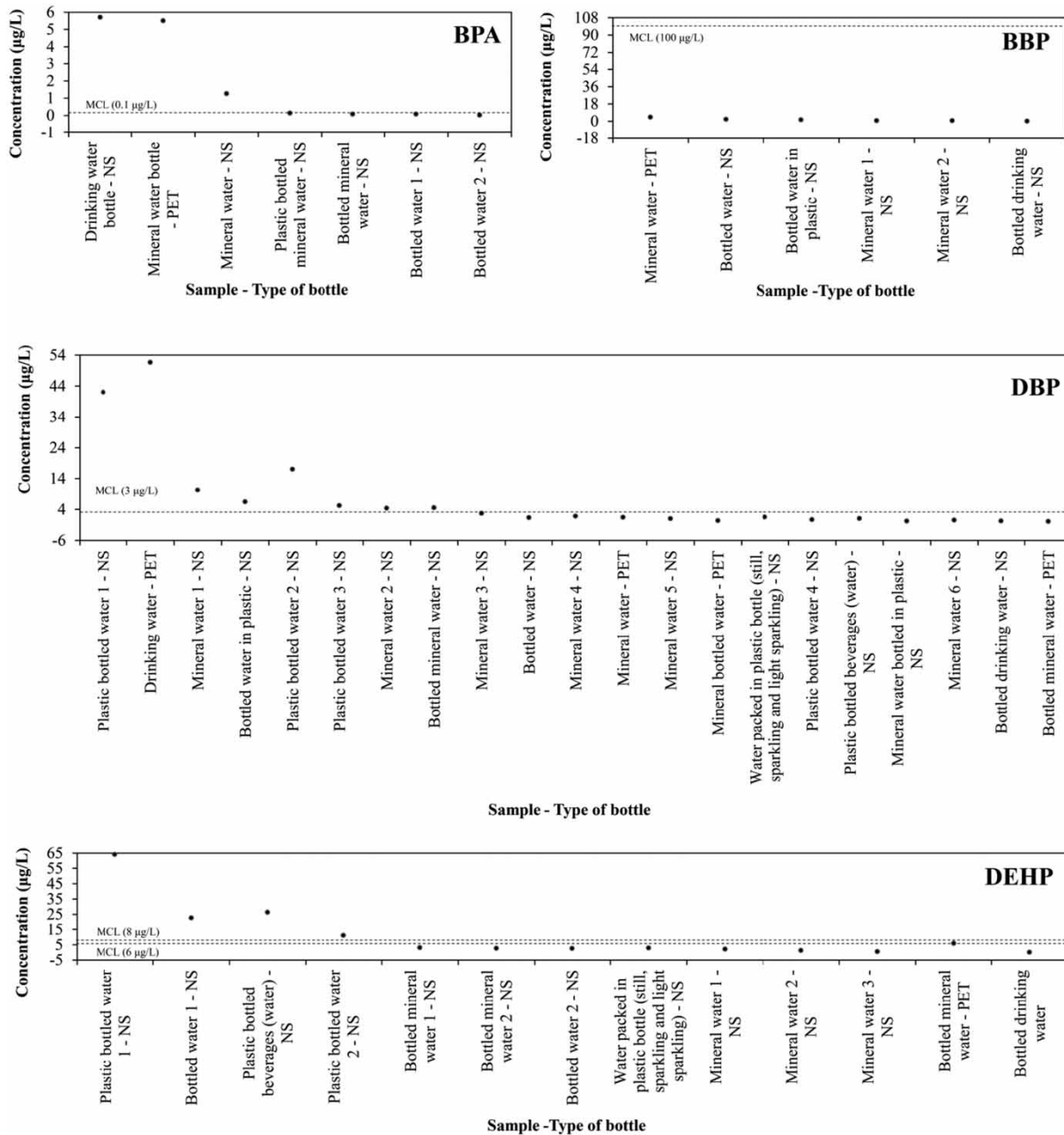


Figure 1 | BPA, BBP, DBP and DEHP concentrations detected in commercial water bottles without the storage study. The number in 'Sample-Type of bottle' represents different samples. NS is 'Not stated'.

Shoeb (2018) occurred the reverse. These authors analyzed DEHP in PET bottled water.

The migration of PAEs in bottled water results from the combined effects of multiple factors, as reported by Luo *et al.* (2018). The possible reason for the migration of PAEs is the usage of low-quality plastic as well as solubility in water

(Saeed *et al.* 2010). The plastic type is that influences the presence of specific contaminants, where the migration of plasticizers from the cap material plays an important role (Guart *et al.* 2014). Jeddi *et al.* (2016) noted that the effect of temperatures and sunlight exposure on the release of the BBP, DBP and DEHP into the water is more than the effect

Table 3 | Levels of BPA, BBP, DBP and DEHP in commercial water bottles with the storage study

Detected analyte	Sample	Country	Type of bottle	Number of brands or samples	Storage study	Concentration ($\mu\text{g/L}$)	Reference
BPA	Bottled drinking water	Iran	Not stated	4	Freezing temperature (24 h) ^a	0.0023	Kaykhai <i>et al.</i> (2020)
					Sunlight (for a week)	0.007	
					Boiled in a steel jar and quickly poured into the bottle (cooled to ambient temperature) ^a	0.016	
BBP	Mineral water bottle	Pakistan	Not stated	5	Sunlight (7 days with 10 h/day – 46–48 °C)	ND ^b – 12.11 (median: 7.43)	Surhio <i>et al.</i> (2017)
BBP	Bottled water	Iran	PET ^c	10	Sunlight (roof on sunny days for 1 week)	0.03–0.13	Abtahi <i>et al.</i> (2019)
DBP	Mineral water bottle	Pakistan	Not stated	5	Sunlight (7 days with 10 h/day – 46–48 °C)	ND ^b – 26.16 (median: 21.7)	Surhio <i>et al.</i> (2017)
DBP	Bottled water	Iran	PET	10	Sunlight (roof on sunny days for 1 week)	ND ^b – 0.12 (median: 0.10)	Abtahi <i>et al.</i> (2019)
DBP	Bottled water	Romania	Not stated	Not stated	Room temperature 1–4 °C ^a	6.11 5.12	Sulentic <i>et al.</i> (2018)
DBP	Water in plastic bottle	Iran	Not stated	3	Room temperature ^a Freezing temperature ^a	5.32 10.12	Hashemi-Moghaddam & Maddah (2018)
DBP	Bottled mineral water	Italy	PET	15	6 months at 25 °C	1.23	Cincotta <i>et al.</i> (2018)
					12 months at 60 °C	3.14	
					18 months at 60 °C	6.01	
DBP	Bottled water	Egypt	PET	5	1 months (4 ± 1 °C)	0.107	Zaki & Shoeib (2018)
					2 months (4 ± 1 °C)	0.128	
					4 months (4 ± 1 °C)	0.173	
					1 months (40 ± 5 °C)	0.124	
					2 months (40 ± 5 °C)	0.167	
					4 months (40 ± 5 °C)	0.229	
					2 months (25 ± 5 °C)	0.136	
6 months (25 ± 5 °C)	0.227						
DBP	Drinking water bottled	Iran	PET	5	First week of the production	0.80	Yousefi <i>et al.</i> (2019)
					Sunlight (23 ± 2 °C at 5 days)	5.86	
					Incubator (25 °C for 75 days)	Not stated	
					Incubator (42 °C for 15 days)	Not stated	
DEHP	Mineral water bottle	Pakistan	Not stated	5	Sunlight (7 days with 10 h/day – 46–48 °C)	20.23	Surhio <i>et al.</i> (2017)

(continued)

Table 3 | continued

Detected analyte	Sample	Country	Type of bottle	Number of brands or samples	Storage study	Concentration ($\mu\text{g/L}$)	Reference
DEHP	Bottled water	Iran	PET	10	Sunlight (roof on sunny days for 1 week)	0.7–0.12	Abtahi <i>et al.</i> (2019)
DEHP	Drinking water bottled	Iran	PET	5	First week of the production Sunlight ($23 \pm 2^\circ\text{C}$ at 5 days) Incubator (25°C for 75 days) Incubator (42°C for 15 days)	0.77 Not stated 9.62 ^d and 12.67 ^e 10.33 ^f	Yousefi <i>et al.</i> (2019)
DEHP	Bottled water	Romania	Not stated	Not stated	Room temperature 1 to 4°C ^a	0.52 2.00	Sulentic <i>et al.</i> (2018)
DEHP	Bottled water	India	PET	Not stated	2 months at 4°C 2 months at 37°C 4 months at 4°C 4 months at 37°C 6 months at 4°C 6 months at 37°C	0.303 0.081 0.40 0.2010 1.09 0.59	Annamalai & Namasivayam (2017)
DEHP	Bottled water	Egypt	PET	5	1 months ($4 \pm 1^\circ\text{C}$) 2 months ($4 \pm 1^\circ\text{C}$) 4 months ($4 \pm 1^\circ\text{C}$) 1 months ($40 \pm 5^\circ\text{C}$) 2 months ($40 \pm 5^\circ\text{C}$) 4 months ($40 \pm 5^\circ\text{C}$) 2 months ($25 \pm 5^\circ\text{C}$) 6 months ($25 \pm 5^\circ\text{C}$)	0.135 0.235 0.307 0.190 0.306 0.432 0.274 0.396	Zaki & Shoeib (2018)

BPA, bisphenol A; BBP, benzylbutyl phthalate; DBP, di-*n*-butyl phthalate; DEHP, di(2-ethylhexyl) phthalate.

^aUnspecified temperature.

^bNot detected.

^cPolyethylene terephthalate.

^dMean concentration.

^eHighest mean concentration.

^fMaximum amount.

due to storage duration. Keresztes *et al.* (2013) analyzed identical brands of water samples in PET containers having different volumes. The authors verified that how much higher is the contact surface between water and PET material, higher concentrations of BBP, DBP and DEHP were observed.

RISK ASSESSMENT

Daily intake-associated risk assessment

To compare the health risk *via* commercial water bottle consumption was used the risk assessment. The highest

levels of BPA, BBP, DBP and DEHP in commercial water bottles are presented in Table 4 and Figure 3. The BPA in PET bottled water suggests other sources of contamination beside the packaging itself. The presence of BPA in PC packaging is known. In the case of PET bottled water, BPA can result from leaching by bottle caps or contamination of the water before bottling (Guart *et al.* 2011; Bach *et al.* 2012; Rowell *et al.* 2016). The water quality intended for bottling can be affected by the leaching of pollutants from unprotected agricultural and industrial areas (Bono-Blay *et al.* 2012). Bono-Blay *et al.* (2012) studied Spanish water sources intended for bottling, where BPA was one of the most frequently

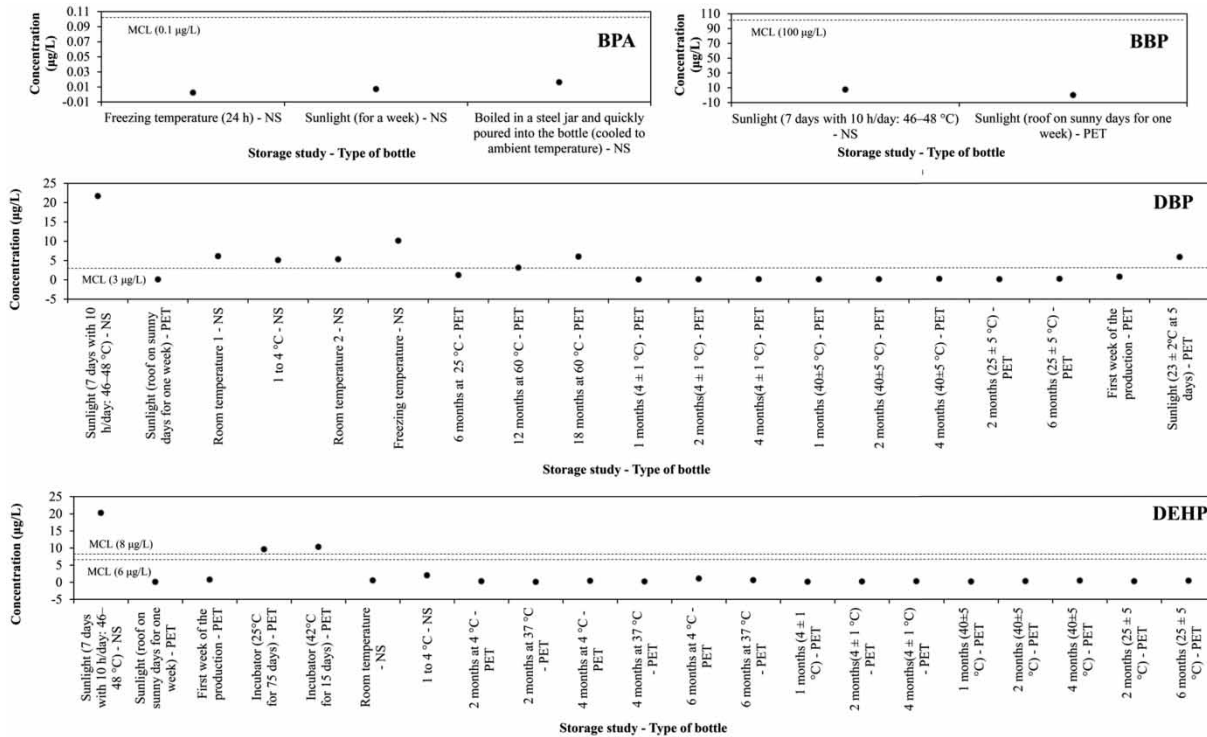


Figure 2 | Levels of BPA, BBP, DBP and DEHP variation in commercial water bottles with the storage study. The number in 'Sample-Type of bottle' represents different samples. NS is 'Not stated'.

Table 4 | Estimation of exposure to BPA, BBP, DBP and DEHP in commercial water bottles

Without the storage study

Detected analyte	Country	Concentration (µg/L) ^a	EDI (µg/kg-bw/day) ^b	Contribution via bottled water (%) ^c	ELCR ^d	Reference
BPA	Turkey	5.7	≈0.163	≈4.1	-	Karayaka <i>et al.</i> (2019)
BPA	Iran	5.5	≈0.157	≈3.9	-	Mohammadnezhad <i>et al.</i> (2017)
BPA	Malaysia	1.25	≈3.6 × 10 ⁻²	≈0.9	-	Rozaini <i>et al.</i> (2017)
BPA	China	0.127	≈3.6 × 10 ⁻³	≈0.09	-	Chang <i>et al.</i> (2017)
BPA	China	0.08	≈2.3 × 10 ⁻³	≈0.058	-	Zhou <i>et al.</i> (2019)
BPA	Iran	0.07	2.0 × 10 ⁻³	≈0.05	-	Gorji <i>et al.</i> (2019)
BPA	China	0.01	≈2.9 × 10 ⁻⁴	≈7.3 × 10 ⁻³	-	Wu <i>et al.</i> (2019)
BBP	Iran	5.5	≈0.157	≈0.03	-	Farahani <i>et al.</i> (2017)
BBP	Spain	1.9	≈5.4 × 10 ⁻²	≈1.1 × 10 ⁻²	-	Barciela-Alonso <i>et al.</i> (2017)
BBP	China	1.86	5.3 × 10 ⁻²	≈1.1 × 10 ⁻²	-	Wu <i>et al.</i> (2019)
BBP	Vietnam	0.95	≈2.7 × 10 ⁻²	≈5.4 × 10 ⁻³	-	Tran-Lam <i>et al.</i> (2018)
BBP	China	0.690	≈2.0 × 10 ⁻²	≈4.0 × 10 ⁻³	-	Yin <i>et al.</i> (2019)
BBP	China	0.032	≈9.1 × 10 ⁻⁴	≈1.8 × 10 ⁻⁴	-	Li <i>et al.</i> (2019)
BBP	Spain	<LOQ (0.006)	<1.7 × 10 ⁻⁴	<3.4 × 10 ⁻⁵	-	Santana-Mayor <i>et al.</i> (2018)
DBP	Mexico	82.8	2.37	23.7	-	Salazar-Beltrán <i>et al.</i> (2017)

(continued)

Table 4 | continued

Without the storage study

Detected analyte	Country	Concentration ($\mu\text{g/L}$) ^a	EDI ($\mu\text{g/kg-bw/day}$) ^b	Contribution via bottled water (%) ^c	ELCR ^d	Reference
DBP	Nigeria	42	1.2	12	–	Dada et al. (2018)
DBP	Thailand	17.0	≈ 0.486	≈ 4.86	–	Pinsrithong & Bunkoed (2018)
DBP	China	11.5	≈ 0.329	≈ 3.29	–	Yin et al. (2019)
DBP	Spain	8.2	≈ 0.234	≈ 2.34	–	Barciela-Alonso et al. (2017)
DBP	Iran	5.2	≈ 0.149	≈ 1.49	–	Mohebbi et al. (2017)
DBP	Iran	4.5	0.129	1.29	–	Amiri & Ghaemi (2017)
DBP	Iran	4.35	≈ 0.124	1.24	–	Farajzadeh et al. (2019)
DBP	China	2.68	$\approx 7.7 \times 10^{-2}$	≈ 0.77	–	Chen et al. (2019b)
DBP	Iran	2.5	$\approx 7.1 \times 10^{-2}$	≈ 0.71	–	Amiri et al. (2017)
DBP	Vietnam	1.86	5.3×10^{-2}	0.53	–	Tri et al. (2018)
DBP	Iran	1.7	$\approx 4.9 \times 10^{-2}$	≈ 0.49	–	Farahani et al. (2017)
DBP	China	1.34	$\approx 3.8 \times 10^{-2}$	≈ 0.38	–	Wu et al. (2019)
DBP	Iran	1.13	$\approx 3.2 \times 10^{-2}$	≈ 0.32	–	Soheilifar et al. (2018)
DBP	Spain	<1	$< 2.9 \times 10^{-2}$	<0.29	–	González-Sálamo et al. (2018)
DBP	Vietnam	0.95	$\approx 2.7 \times 10^{-2}$	≈ 0.27	–	Tran-Lam et al. (2018)
DBP	China	0.51	$\approx 1.5 \times 10^{-2}$	0.15	–	Li et al. (2019)
DBP	Spain	0.36	1.0×10^{-2}	0.1	–	González-Sálamo et al. (2017)
DBP	Hungary	<0.2	$< 5.7 \times 10^{-3}$	<0.057	–	Szendi et al. (2018)
DBP	Spain	0.184	$\approx 5.3 \times 10^{-3}$	≈ 0.053	–	Santana-Mayor et al. (2018)
DEHP	Thailand	64.0	≈ 1.83	≈ 3.66	$\approx 3.0 \times 10^{-5}$	Pinsrithong & Bunkoed (2018)
DEHP	Vietnam	42.3	≈ 1.21	2.42	$\approx 2.0 \times 10^{-5}$	Tri et al. (2018)
DEHP	China	2.50	$\approx 7.1 \times 10^{-2}$	≈ 1.42	$\approx 1.2 \times 10^{-6}$	Wu et al. (2019)
DEHP	Italy	24.4	≈ 0.697	≈ 1.39	$\approx 1.1 \times 10^{-5}$	Notardonato et al. (2018)
DEHP	Turkey	11.90	0.34	0.68	$\approx 5.6 \times 10^{-6}$	Özer et al. (2017)
DEHP	Hungary	<11.289	<0.323	<0.646	$\approx < 5.3 \times 10^{-6}$	Szendi et al. (2018)
DEHP	Chile	4.321	≈ 0.123	≈ 0.246	$\approx 2.0 \times 10^{-6}$	Manzo et al. (2019)
DEHP	Iran	3.5	0.1	≈ 0.2	$\approx 1.6 \times 10^{-6}$	Amiri et al. (2017)
DEHP	Iran	3.0	0.086	0.172	$\approx 1.4 \times 10^{-6}$	Amiri & Ghaemi (2017)
DEHP	Vietnam	1.8	5.1×10^{-2}	≈ 0.1	$\approx 8.53 \times 10^{-7}$	Tran-Lam et al. (2018)
DEHP	China	0.733	$\approx 2.1 \times 10^{-2}$	≈ 0.042	$\approx 3.4 \times 10^{-7}$	Yin et al. (2019)
DEHP	China	0.021	6.0×10^{-4}	1.2×10^{-3}	$\approx 9.9 \times 10^{-9}$	Li et al. (2019)
Detected analyte	Country	Concentration ($\mu\text{g/kg}$) ^a	EDI ($\mu\text{g/kg-bw/day}$) ^b	Contribution via bottled water (%) ^c	CR ^d	Reference
BBP	China	Mineral water (MW): 0.11 Soda water (SW): 0.13	$\approx 3.1 \times 10^{-3}$ $\approx 3.7 \times 10^{-3}$	$\approx 6.2 \times 10^{-4}$ $\approx 7.4 \times 10^{-4}$	–	Yang et al. (2017)

(continued)

Table 4 | continued

Without the storage study

Detected analyte	Country	Concentration ($\mu\text{g/L}$) ^a	EDI ($\mu\text{g/kg-bw/day}$) ^b	Contribution via bottled water (%) ^c	CR ^d	Reference
DBP		Mineral water: 1.02	$\approx 2.9 \times 10^{-2}$	≈ 0.29	–	
		Soda water: 6.34	≈ 0.181	≈ 1.8		
DEHP		Mineral water: 4.3	≈ 0.123	≈ 0.246	$\approx 5.2 \times 10^{-8}$	
		Soda water: 7.29	≈ 0.208	≈ 0.416		

With storage study

Detected analyte	Country	Concentration ($\mu\text{g/L}$) ^a	EDI ($\mu\text{g/kg-bw/day}$) ^b	Contribution via bottled water (%) ^c	CR ^d	Reference
BPA	Iran	0.016	$\approx 4.6 \times 10^{-4}$	$\approx 1.2 \times 10^{-2}$	–	Kaykhaii <i>et al.</i> (2020)
BBP	Pakistan	12.11	0.346	≈ 0.069	–	Surhio <i>et al.</i> (2017)
BBP	Iran	0.13	$\approx 3.7 \times 10^{-3}$	$\approx 7.4 \times 10^{-4}$	–	Abtahi <i>et al.</i> (2019)
DBP	Pakistan	26.16	≈ 0.747	7.47	–	Surhio <i>et al.</i> (2017)
DBP	Iran	10.12	≈ 0.289	≈ 2.89	–	Hashemi-Moghaddam & Maddah (2018)
DBP	Iran	8.45	≈ 0.241	≈ 2.41	–	Yousefi <i>et al.</i> (2019)
DBP	Romania	6.11	≈ 0.175	≈ 1.75	–	Sulentiu <i>et al.</i> (2018)
DBP	Italy	6.01	≈ 0.172	≈ 1.72	–	Cincotta <i>et al.</i> (2018)
DBP	Egypt	0.229	$\approx 6.5 \times 10^{-3}$	≈ 0.065	–	Zaki & Shoeib (2018)
DBP	Iran	0.12	$\approx 3.4 \times 10^{-3}$	≈ 0.034	–	Abtahi <i>et al.</i> (2019)
DEHP	Pakistan	20.23	0.578	≈ 1.16	$\approx 9.5 \times 10^{-6}$	Surhio <i>et al.</i> (2017)
DEHP	Iran	12.67	0.362	≈ 0.724	$\approx 6.0 \times 10^{-6}$	Yousefi <i>et al.</i> (2019)
DEHP	Romania	2.00	$\approx 5.7 \times 10^{-2}$	≈ 0.114	9.4×10^{-7}	Sulentiu <i>et al.</i> (2018)
DEHP	Egypt	0.432	$\approx 1.2 \times 10^{-2}$	≈ 0.024	$\approx 2.0 \times 10^{-7}$	Zaki & Shoeib (2018)
DEHP	Iran	0.12	$\approx 3.4 \times 10^{-3}$	$\approx 6.8 \times 10^{-3}$	$\approx 5.6 \times 10^{-8}$	Abtahi <i>et al.</i> (2019)
DEHP	India	1.09	$\approx 3.1 \times 10^{-2}$	≈ 0.062	$\approx 5.1 \times 10^{-7}$	Annamalai & Namasivayam (2017)

BPA, bisphenol A; BBP, benzylbutyl phthalate; DBP, di-*n*-butyl phthalate; DEHP, di(2-ethylhexyl) phthalate.

^aThe worst-case scenario (the maximum level of each compound) was employed.

^bEDI = (C × IR)/BW, where C is the concentration of target compounds ($\mu\text{g/L}$ or mg/kg), ingestion rate (IR) is the daily consumption rate of bottled water (L/day or g/day), and BW is body weight (Luo *et al.* 2018), and the IR was assumed to be 2.0 L/day or 2.0 kg/day for a 70 kg for adult (BW) (WHO 2005). The value of 2.0 L/day refers to all water sources that includes water from all supply sources such as community water supply (i.e., tap water), bottled water, etc.

$\mu\text{g/kg-bw/day}$: microgram per kilogram of the body weight of the person taking per day.

^cContribution via drinking water = (EDI/TDI) × 100 (Zaki & Shoeib 2018), where the TDI for BPA, BBP, DBP, and DEHP are available for reference as established by EFSA (4, 500, 10 and 50 $\mu\text{g/kg-bw/day}$).

^dELCR is the Excess Lifetime Cancer Risks due to exposure to chemicals through the use of bottled water. ELCR = DWUR × MC, where Drinking Water Unit Risk is equal to $4.7 \times 10^{-7} \mu\text{g/L}$ of DEHP in water, and MC is the maximum concentration ($\mu\text{g/L}$ or $\mu\text{g/kg}$) of DEHP in bottled water (Jeddi *et al.* 2015). Here was considered the value of $4.7 \times 10^{-7} \mu\text{g/kg}$ for papers with concentrations give in mg/kg .

detected compounds at concentrations between 0.031 and 0.203 $\mu\text{g/L}$.

Although the estimated daily intake (EDI) of BPA, BBP, DBP and DEHP detected in PET bottled waters analyzed was below the legislative values (Table 4), considering all

types of food, it may contribute to the total daily intake of these compounds. The highest contributions *via* commercial water bottles of BPA, BBP, DBP and DEHP in all the bottled waters studied were 4.1, 0.069, 23.7 and 3.66% of TDI, respectively (Figure 3). The results demonstrate that

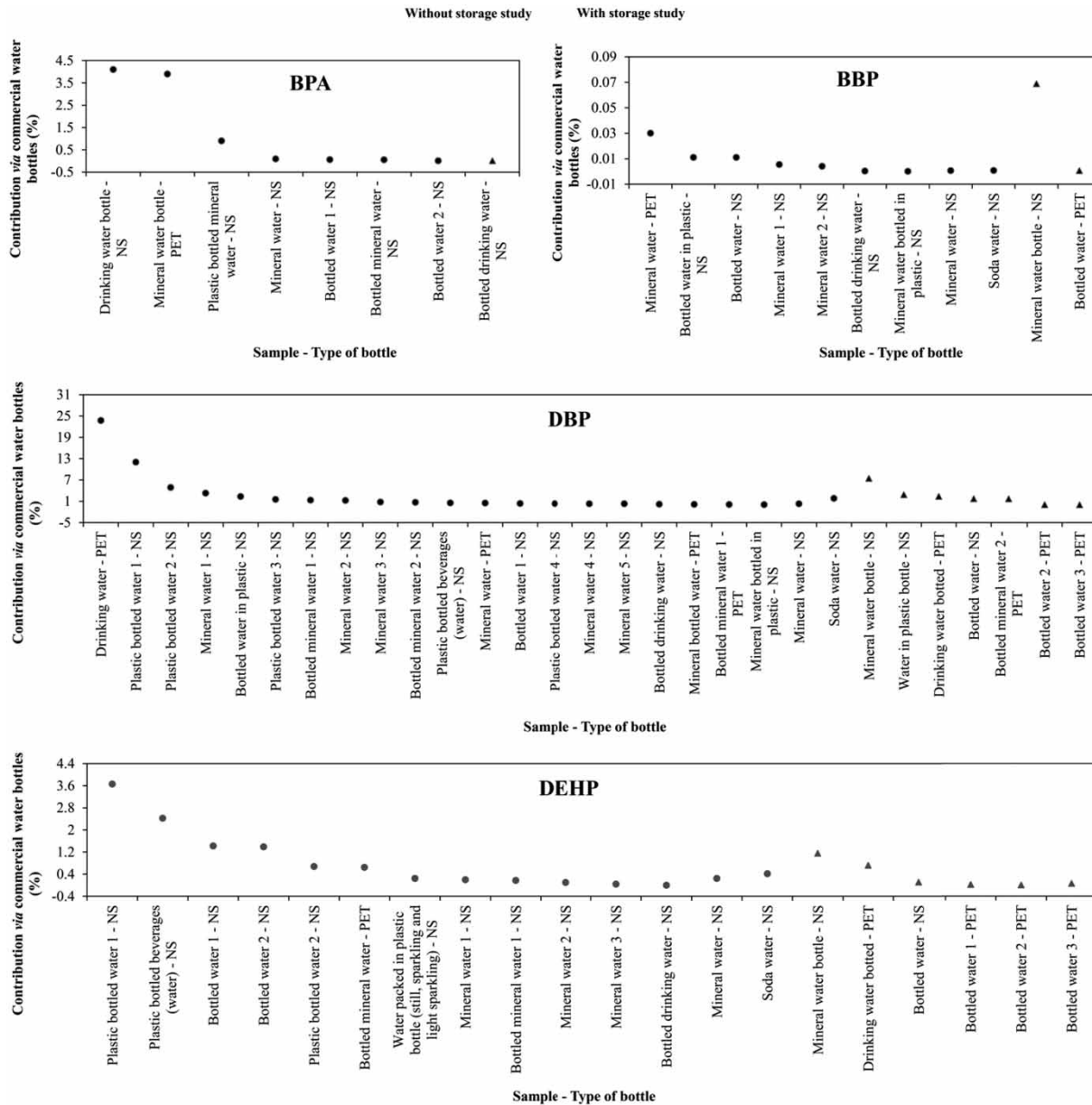


Figure 3 | BPA, BBP, DBP and DEHP contribution *via* commercial water bottles. The number in parentheses represents articles from the same country with different concentrations. MW is mineral water and SW is soda water.

contribution *via* commercial water bottles (Table 4) could represent a substantial source of exposure to these compounds (considering the highest contributions), when the daily consumption rate, of 2.0 L/day of bottled water and body weight of 70 kg, is used according to the standard WHO (2005). If Reference Dose (RfD) were considered (Table 5), which is more restrictive for DEHP, the contribution would be much higher (9.15%).

The carcinogenic risk (Excess Lifetime Cancer Risks – ELCR) posed by the highest concentration of DEHP in bottled water was negligible for all papers, with extremely below or between the accepted risk level of 10^{-6} – 10^{-4} cancer risk (WHO 2017). As mentioned by WHO (2017), daily water intake can vary significantly in different parts of the world and location-specific data on drinking water consumption are preferred. As reported by Leung *et al.*

Table 5 | Estimated human exposure and estrogenic effects of BPA, BBP, DBP and DEHP via commercial bottled water ingested for other population groups

	BPA	BBP	DBP	DEHP
Reference	Karayaka <i>et al.</i> (2019)	Surhio <i>et al.</i> (2017)	Salazar-Beltrán <i>et al.</i> (2017)	Pinsrithong & Bunkoed (2018)
Maximum concentration (µg/L)	5.7	12.11	82.8	64.0
EDI (µg/kg-bw/day)				
Infants (birth to <12 months) ^{*a}	≈0.042	≈0.089	≈0.61	≈0.471
Children (1 to <3 years) ^{*b}	≈0.106	≈0.225	≈1.539	≈1.19
Children (3 to <11 years) ^{*c}	≈0.072	≈0.154	≈1.051	≈0.813
Teenage (11 to <16 years) ^{*d}	≈0.053	≈0.112	≈0.767	≈0.593
Young adult (16 to <21 years) ^{*e}	≈0.061	≈0.129	≈0.882	≈0.682
Adult (≥21 years) ^{*f}	≈0.060	≈0.128	≈0.876	≈0.677
Pregnant (15–44) ^{*g}	≈0.046	≈0.097	≈0.662	0.512
Elderly (≥65 years) ^{*h}	≈0.057	≈0.120	≈0.821	≈0.635
Tolerable daily intake (µg/kg-bw/day)	4	500	10	50
Contribution <i>via</i> bottled water (%) [#]				
Infants (birth to <12 months) ^{*a}	≈1.05	≈1.78 × 10 ⁻²	≈6.10	≈0.942
Children (1 to <3 years) ^{*b}	≈2.65	≈4.50 × 10 ⁻²	≈15.40	≈2.38
Children (3 to <11 years) ^{*c}	≈1.80	≈3.08 × 10 ⁻²	≈1.05	≈1.63
Teenage (11 to <16 years) ^{*d}	≈1.33	≈2.24 × 10 ⁻²	≈7.67	≈1.19
Young adult (16 to <21 years) ^{*e}	≈1.53	≈2.58 × 10 ⁻²	≈8.82	≈1.36
Adult (≥21 years) ^{*f}	≈1.50	≈2.56 × 10 ⁻²	≈8.76	≈1.35
Pregnant (15–44) ^{*g}	≈1.15	≈1.94 × 10 ⁻²	≈6.62	1.02
Elderly (≥65 years) ^{*h}	≈1.43	≈1.2 × 10 ⁻²	≈8.21	≈1.27
RfD (µg/kg-bw/day) ⁱ	50	500	100	20
HQ ^j				
Infants (birth to <12 months) ^{*a}	≈8.40 × 10 ⁻⁴	≈1.78 × 10 ⁻⁴	≈6.10 × 10 ⁻³	≈2.36 × 10 ⁻²
Children (1 to <3 years) ^{*b}	≈2.12 × 10 ⁻³	≈4.50 × 10 ⁻⁴	≈1.54 × 10 ⁻²	≈5.95 × 10 ⁻²
Children (3 to <11 years) ^{*c}	≈1.44 × 10 ⁻³	≈3.08 × 10 ⁻⁴	≈1.05 × 10 ⁻²	≈4.07 × 10 ⁻²
Teenage (11 to <16 years) ^{*d}	≈1.06 × 10 ⁻³	≈2.24 × 10 ⁻⁴	≈7.67 × 10 ⁻³	≈2.97 × 10 ⁻²
Young adult (16 to <21 years) ^{*e}	≈1.22 × 10 ⁻³	≈2.58 × 10 ⁻⁴	≈8.82 × 10 ⁻³	≈3.41 × 10 ⁻²
Adult (≥21 years) ^{*f}	≈1.20 × 10 ⁻³	≈2.56 × 10 ⁻⁴	≈8.76 × 10 ⁻³	≈3.39 × 10 ⁻²
Pregnant (15–44) ^{*g}	≈9.20 × 10 ⁻⁴	≈1.94 × 10 ⁻⁴	≈6.62 × 10 ⁻³	2.56 × 10 ⁻²
Elderly (≥65 years) ^{*h}	≈1.14 × 10 ⁻³	≈2.4 × 10 ⁻⁴	≈8.21 × 10 ⁻³	≈3.18 × 10 ⁻³
SF (based on maximum concentration)				
Infants (birth to <12 months) ^{*a}	1,190	5,620	164	42.5
Children (1 to <3 years) ^{*b}	472	2,220	65	16.8
Children (3 to <11 years) ^{*c}	694	3,250	95.1	24.6
Teenage (11 to <16 years) ^{*d}	943	4,460	130	33.7
Young adult (16 to <21 years) ^{*e}	820	3,880	113	29.3
Adult (≥21 years) ^{*f}	833	3,910	114	29.5

(continued)

Table 5 | continued

	BPA	BBP	DBP	DEHP
Pregnant (15–44)* ^g	1,090	5,150	151	39.1
Elderly (≥ 65 years)* ^h	877	4,170	122	31.5
Estrogenic potency (EP)	$5.9E^{-05l}$	$2E^{-4m}$	$4.1E^{-5m}$	$3E^{-7n}$
EEQ (ng E2/L) ^k	0.336	2.42	3.39	0.0192
Total compounds				6.1652

*The values of weights and bottled water ingested are based on US EPA (2011) and US EPA (2019b): ^a0.0685 L/day for 9.3 kg, ^b0.2305 L/day for 12.45 kg, ^c0.3365 L/day for 26.5 kg, ^d0.517 L/day for 55.8 kg, ^e0.753 L/day for 70.7 kg, ^f0.84 L/day for 79.4 kg, ^g0.6 L/day for 75 kg, ^h0.749 L/day for 75.5 kg (Table 8–24, 8–25, 8–29, 3–34, 3–71, and Table A-2).

ⁱContribution via drinking water = (EDI/TDI) \times 100 (Zaki & Shoeib 2018), where the TDI for BPA, BBP, DBP, and DEHP are available for reference as established by EFSA.

^jUS EPA (1987a, 1987b, 1988, 2019a).

^kHazard Quotient (HQ) = EDI/RfD, where HQ is associated with the exposure via the specified exposure route (unitless) (Jeddi *et al.* 2015).

^lSafety factor (SF) = RfD/EDI (Luo *et al.* 2018).

^mEEQ = EP_{*i*} \times c_{*i*}, where EP and c denote the estrogenic potency of an individual estrogenic compound (*in vitro* bioassays) and its corresponding concentration (Liu *et al.* 2009).

ⁿLegler *et al.* (1999).

^oKim & Ryu (2006).

^pCavanagh *et al.* (2018).

(2013), infants and children have been subject to increased risks that are approximately six times greater than those in adolescents and adults due to their high drinking water consumption based on body weight. As specified by US EPA (2019), older adults (≥ 65 years of age) and pregnant are other susceptible groups due to their physiological properties change. As reported by Gerba *et al.* (1996), the elderly may be less able to create an effective defense against contaminants because of a pre-existing disease or weakened immune system. The risk is inherent to the pregnant and also to the fetus (Wee & Aris 2019).

Table 5 shows more detailed the risk assessment to other population groups based on only the ingestion of bottled water. The values of bottled water ingestion are based on US EPA (2019) and US EPA (2019b). US EPA (2019b) includes only bottled water consumed directly as a beverage, not including bottled water used in the preparation of foods. As stated by Hossain *et al.* (2013), the regional variability in water intake can be due to differences in weather conditions and food intake habits of the population. As can be seen in Table 5, the results demonstrate that bottled water can represent a substantial source of exposure to these compounds. Children (1 to <3 years) had a higher EDI, and as a result, a high contribution via bottled water consumption (15.4%) and high Hazard Quotient (HQ). As previously reported, if RfD for DEHP were considered, the contribution would be much higher

(5.95%). The highest HQ for the compounds via bottled water consumption was much lower than 1 for all compounds, indicating an absence of risk (US EPA 2019). The safety factor (SF) calculated for the selected compounds with the maximum concentrations was all far above 1 for all groups, denoting that the BPA, BBP, DBP and DEHP concentrations disclosed in bottled water should not represent grave safety concerns, corroborating with the review by Luo *et al.* (2018).

Potential estrogenic effect of BPA and PAEs

Despite the safety factor indicates that the levels of the compounds in bottled waters are acceptable in terms of water safety, the potential estrogenic effects of the compounds by an average Estrogen Equivalent (EEQ) level in bottled waters are based on the highest concentrations that were evaluated (Table 5). The EEQ provides valuable information on human exposure to estrogen-like compounds, aiding in the estimation of the total dietary intake of estrogenicity (Schilirò *et al.* 2013). The potential estrogenic effects of BBP and DBP in bottled water should not be ignored due to their relatively high concentrations. As can be seen in Table 5, the average EEQ level in the bottled waters is significantly at 6.1652 ng E2/L, which was 22.8 times higher than those that cause adverse estrogenic effects on zebrafish (0.27 ng E2/L) as reported by Soares *et al.* (2009). Thus, the

average EEQ level indicated that BPA, BBP, DBP and DEHP in bottled waters may induce adverse estrogenic effects on human health.

CONCLUSIONS

Although the governments have published the guideline tolerance values of bisphenol A and PAEs in drinking water, they are still detected in water bottles. HPLC–DAD was the most used in BPA detections, while GC–MS was the most used in PAE detections. New methods to improve the extraction of BPA, BBP, DBP and DEHP from commercial water bottles have been developed. DBP and DEHP have still been detected in concentrations greater than those established by legislation. Contradictory observations, with decreasing and increasing concentrations on PAE concentration in bottled water, are reported. No consistent or clear trends regarding the effects of storage conditions, on PAE concentration in bottled water, are demonstrated. Based on the risk assessment, BPA, BBP, DBP and DEHP in commercial water bottles do not raise serious concern for humans. The average EEQ level revealed that BPA, BBP, DBP and DEHP in bottled waters may induce adverse estrogenic effects on human health. Besides that, the use of bottled water kept in unsuitable conditions is not appropriate and especially for sensitive groups. Thus, the occurrence of individual BPA, BBP, DBP and DEHP and their association in bottled water need to be verified to avoid their synergistic effects on human health.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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First received 2 September 2020; accepted in revised form 29 March 2021. Available online 8 April 2021