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.
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 2019). 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; Goeury 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 carried 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 we 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 (Dreolín 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 Kow (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 refining 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, DBB: 11.2 mg/L and DEHP: 0.27 mg/L) and have a particular affinity for fats and alcohols (Grinbaum et al. 2013; 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 preconcentrate 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, preconcentration and clean-up from co-existing species (Gao et al. 2015). A preconcentration 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-Wasylka 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-Wasylka 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

| Detected analyte (s) | Extraction method | Chromatographic technique | LODs (μg/L) | LOQs (μg/L) | Reference |
|----------------------|-------------------|--------------------------|-------------|-------------|-----------|
| BPA                  | SPE               | UFLC–MS/MS               | 0.004–0.055<sup>a</sup> | 1.4 × 10<sup>−2</sup> | Zhou et al. (2019) |
| BPA                  | SBSE              | HPLC–UV/Vis              | 0.02        | 0.06        | Gorji et al. (2019) |
| BPA                  | UAE-MIP-μ-SPE     | HPLC–DAD                 | 0.07        | 0.15        | Rozaini et al. (2017) |
| BPA                  | MDMIP- SPE        | HPLC–DAD                 | 0.083       | 0.114       | Chang et al. (2017) |
| BPA                  | SPME              | HPLC–DAD                 | 0.20        | Not stated  | Mohammadnezhad et al. (2017) |
| BPA                  | USAE-MIP-μ-SPE    | HPLC–DAD                 | 0.54        | 1.8         | Karayaka et al. (2019) |
| BPA                  | SLLME             | GC–MS                    | 0.07        | 0.15        | Karayaka et al. (2019) |
| BPA; BBP; DBP; DEHP  | LLE               | LC–MS/MS GS–MS           | Not stated  | Not stated  | Wu et al. (2019) |
| BBP; DBP             | MIP-SPE           | HPLC–MS                  | 0.16; 0.84  | 0.55; 2.81  | Barciela-Alonso et al. (2017) |
| BBP; DBP             | IT-LLME           | GC–MS                    | 1.67; 0.75  | 5.50; 2.46  | Farahani et al. (2017) |
| BBP; DBP             | m-μDME            | UHPLC–MS/MS              | Not stated  | 6 × 10<sup>−3</sup>; 11 × 10<sup>−3</sup> | Santana-Mayor et al. (2018) |
| BBP; DBP; DEHP       | MSPE              | HPLC–UV/Vis              | 0.0103; 0.003<sup>b</sup>; 0.0167 | 0.0342; 0.022<sup>b</sup>; 0.0556 | Yin et al. (2019) |
| BBP; DBP; DEHP       | GC–MS/MS         | 1.0; 1.0; 0.5            | 3.0; 3.0; 0.15 | Li et al. (2019) |
| BBP; DBP; DEHP       | GC–MS/MS         | 5.0; 1.0; 5.0            | Not stated  | Not stated  | Wei et al. (2018) |
| DBP; DEHP            | TSP–LLME         | GC–MS                    | 0.007       | 0.021       | Chen et al. (2019) |
| DBP; DEHP            | SVA–LLME         | GC–MS                    | 0.15        | 0.50        | Mohebbi et al. (2017) |
| DBP; DEHP            | DSPE–DLLME       | GC–FID                   | 1.24        | 4.11        | Farajzadeh et al. (2019) |
| DBP; DEHP            | SPE              | HPLC–UV/Vis              | 2.4         | 7.9         | Salazar-Beltrán et al. (2017) |
| DBP; DEHP            | MISPE            | HPLC–UV/Vis              | 3           | 10          | Sohelifar et al. (2018) |
| DBP; DEHP            | m-μDME           | GC–MS/MS                 | Not stated  | 0.009       | González-Sálimo et al. (2017) |
| DBP; DEHP            | LLE              | HPLC–UV/Vis              | Not stated  | Not stated  | Dada et al. (2018) |
| DBP; DEHP            | HF–LPME          | GC–MS/MS                 | Not stated  | Not stated  | González-Sálimo et al. (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 et al. (2019) |
| DBP; DEHP            | MEPS             | GC–FID                   | 0.01; 0.15  | 0.10; 0.25  | Manzo et al. (2019) |
| DBP; DEHP            | LLE              | GC–FID                   | Not stated  | Not stated  | Szendi et al. (2018) |
| DBP; DEHP            | DMIS–SPE         | GC–MS                    | 0.000309    | 0.0013      | Özer et al. (2017) |
| DBP; DEHP            | SPE              | GC–FID                   | 0.02        | Not stated  | Chaikhani & Amiri (2019) |
| DBP; DEHP            | DLLME            | GC–FID                   | 2           | 4           | Notardonato et al. (2018) |

(continued)
### Without migration study

| Detected analyte (s) | Extraction method | Chromatographic technique | LODs (µg/L) | LOQs (µg/L) | Reference |
|----------------------|-------------------|---------------------------|-------------|-------------|-----------|
| BBP; DBP; DEHP       | LLE               | GC-MS                     | 0.00031; 0.00025; 0.00042 | 0.00096; 0.0008; 0.00122 | Yang et al. (2017) |
| BBP; DBP; DEHP       | Not stated        | GC-MS                     | 0.02; 0.02; 0.03 | 0.06; 0.06; 0.94 | Suriho et al. (2017) |
| BBP; DBP; DEHP       | LLE               | GC-MS                     | 0.025; 0.017; 0.031 | Not stated | Abtahi et al. (2019) |
| BBP; DBP; DEHP       | Not stated        | GC-MS                     | 0.015 | Not stated | Hashemi-Moghaddam & Maddah (2018) |
| BBP; DBP; DEHP       | PT-µ-SPE          | HPLC-FLD                  | 0.001 | 0.0032 | Kaykhaii et al. (2020) |
| BBP; DBP; DEHP       | m-SPE             | GC-FID                    | 0.64 | Not stated | Surhio et al. (2017) |
| BBP; DBP; DEHP       | AALLME            | GC-MS/MS                  | 0.64; 0.60; 0.94 | Not stated | Yousefi et al. (2019) |
| BBP; DBP; DEHP       | UA-DLLME          | GC-MS                     | 0.3 | Not stated | Annamalai & Namasivayam (2017) |
| BBP; DBP; DEHP       | SPE               | GC-MS                     | 0.015 | Not stated | Sulentic et al. (2018) |
| BBP; DBP; DEHP       | UA-DLLME          | GC-MS                     | 0.043; 0.062 | Not stated | Zaki & Shoeib (2018) |
| BBP; DBP; DEHP       | MIP-SPE           | GC-FID                    | 0.12 | Not stated | Yang et al. (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; MDMP, magnetic dummy molecularly imprinted polymer; SPME, solid-phase microextraction; SLLME, switchable liquid-liquid microextraction; LE, liquid-liquid extraction; IT-UAA, in tube ultrasonic and air-assisted; LLME, liquid-liquid microextraction; m-µ-SPE, magnetic micro-dispersive solid-phase extraction; MSPE, magnetic solid-phase extraction; TSP, temperature-sensitive polymer; SVA, solvent vapor-assisted; 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 detector; FID, flame ionization detector; UV/Vis, ultraviolet/visible; EC, external quality control; UHPLC, ultra-high-performance liquid chromatography; UPLC, ultra-performance liquid chromatography; FLD, fluorescence detector.

### With migration study

| Detected analyte (s) | Extraction method | Chromatographic technique | LODs (µg/L) | LOQs (µg/L) | Reference |
|----------------------|-------------------|---------------------------|-------------|-------------|-----------|
| BBP; DBP; DEHP       | LLE               | GC-MS                     | 0.00031; 0.00025; 0.00042 | 0.00096; 0.0008; 0.00122 | Yang et al. (2017) |
| BBP; DBP; DEHP       | Not stated        | GC-MS                     | 0.02; 0.02; 0.03 | 0.06; 0.06; 0.94 | Suriho et al. (2017) |
| BBP; DBP; DEHP       | LLE               | GC-MS                     | 0.025; 0.017; 0.031 | Not stated | Abtahi et al. (2019) |
| BBP; DBP; DEHP       | Not stated        | GC-MS/MS                  | 0.015 | Not stated | Hashemi-Moghaddam & Maddah (2018) |
| BBP; DBP; DEHP       | PT-µ-SPE          | HPLC-FLD                  | 0.001 | 0.0032 | Kaykhaii et al. (2020) |
| BBP; DBP; DEHP       | m-SPE             | GC-FID                    | 0.64 | Not stated | Surhio et al. (2017) |
| BBP; DBP; DEHP       | AALLME            | GC-MS/MS                  | 0.64; 0.60; 0.94 | Not stated | Yousefi et al. (2019) |
| BBP; DBP; DEHP       | UA-DLLME          | GC-MS                     | 0.3 | Not stated | Annamalai & Namasivayam (2017) |
| BBP; DBP; DEHP       | SPE               | GC-MS                     | 0.015 | Not stated | Sulentic et al. (2018) |
| BBP; DBP; DEHP       | UA-DLLME          | GC-MS                     | 0.043; 0.062 | Not stated | Zaki & Shoeib (2018) |
| BBP; DBP; DEHP       | MIP-SPE           | GC-FID                    | 0.12 | Not stated | Yang et al. (2017) |

### Currently, MCL has not been established for BBP (US EPA 2019a), although, in 1990, US EPA proposed an MCL of 100 µg/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.

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 µg/L), but are lower than in China (10 µg/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 µg/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 lower than 3 μg/L (GB-5749-2006) and displayed DEHP levels lower 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 & Shoieb 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).

Kaykhaii 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 (Lertsirisophon 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. 2017; Guart et al. 2014). Anna- malai & Namasiyam (2017) obtained bigger values to DEHP at 4 °C and smaller values at 37 °C. To Zaki &
| Detected analyte | Sample                  | Country     | Type of bottle | Number of brands or samples | Concentration (μg/L) | Reference                      |
|------------------|-------------------------|-------------|----------------|-----------------------------|----------------------|--------------------------------|
| BPA              | Drinking water bottle   | Turkey      | Not stated     | 3                           | 5.7                  | Karayaka et al. (2019)         |
| BPA              | Mineral water bottle    | Iran        | PET            | 1                           | 5.5                  | Mohammadnejad et al. (2017)    |
| BPA              | Mineral water           | Malaysia    | Not stated     | 6                           | 1.25                 | Rozaini et al. (2017)          |
| BPA              | Plastic bottled mineral water | China | Not stated     | 1                           | 0.127                | Chang et al. (2017)            |
| BPA              | Bottled mineral water   | Iran        | Not stated     | 3                           | 0.07                 | Gorji et al. (2019)            |
| BPA              | Bottled water           | China       | Not stated     | Not stated                  | 0.05–0.08            | Zhou et al. (2019)             |
| BPA              | Bottled water           | China       | Not stated     | 17                          | 0.01                 | Wu et al. (2019)               |
| BPA              | Bottled water           | Iran        | PET            | 3                           | 2.9–5.5              | Farahani et al. (2017)         |
| BPA              | Bottled water           | China       | Not stated     | 17                          | 1.86                 | Wu et al. (2019)               |
| BPA              | Bottled water in plastic| Spain       | Not stated     | 4                           | 0.75–1.9             | Barciela-Alonso et al. (2017)  |
| BPA              | Mineral water           | China       | Not stated     | 5                           | 0.515–0.690          | Yin et al. (2019)              |
| BPA              | Mineral water           | Vietnam     | Not stated     | 14                          | 0.30–0.95            | Tran-Lam et al. (2018)         |
| BPA              | Bottled drinking water  | China       | Not stated     | 60                          | 0.019–0.032          | Li et al. (2019)               |
| BPA              | Mineral water bottled in plastic | Spain | Not stated     | 1                           | <LOQ                | Santana-Mayor et al. (2018)    |
| DBP              | Plastic bottled Water   | Nigeria     | Not stated     | 15                          | 42                   | Dada et al. (2018)             |
| DBP              | Drinking water          | Mexico      | PET            | 10                          | 20.5–82.8            | Salazar-Beltrán et al. (2017)  |
| DBP              | Mineral water           | China       | Not stated     | 5                           | 8.98–11.5            | Yin et al. (2019)              |
| DBP              | Bottled water in plastic| Spain       | Not stated     | 4                           | 4.6–8.2              | Barciela-Alonso et al. (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 et al. (2017)          |
| DBP              | Mineral water           | Cyprus      | Not stated     | Not stated                  | 4.35                 | Farajzadeh et al. (2019)       |
| DBP              | Bottled mineral water   | Iran        | Not stated     | 1                           | 4.5                  | Amiri & Ghaemi (2017)          |
| DBP              | Mineral water           | China       | Not stated     | 1                           | 2.68                 | Chen et al. (2019b)            |
| DBP              | Bottled water           | China       | Not stated     | 17                          | 1.34                 | Wu et al. (2019)               |
| DBP              | Mineral water           | Iran        | Not stated     | 3                           | 1.1–2.5              | Amiri et al. (2017)            |
| DBP              | Mineral water           | Iran        | PET            | 3                           | 1.1–1.7              | Farahani et al. (2017)         |
| DBP              | Mineral water           | Spain       | Not stated     | 1                           | <1                   | González-Sálamo et al. (2018)  |
| DBP              | Mineral bottled water   | Spain       | PET            | 1                           | 0.36                 | González-Sálamo et al. (2017)  |
| DBP              | Water packed in plastic bottle | Chile | Not stated     | 5 (2 – still, 2 – sparkling, 1 – light sparkling) | 0.353–2.756 | Manzo et al. (2019) |
| DBP              | Plastic bottled water   | Iran        | Not stated     | 10                          | 0.26–1.13            | Soheilifar et al. (2018)       |
| DBP              | Plastic bottled beverages | Vietnam | Not stated     | 8                           | 0.24–1.86            | Tri et al. (2018)              |
| 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 et al. (2018) |
| DBP              | Mineral water | Vietnam | Not stated | 14 | 0.09–0.95 | Tran-Lam et al. (2018) |
| DBP              | Bottled drinking water | China | Not stated | 60 | 0.021–0.51 | Li et al. (2019) |
| DBP              | Bottled mineral water | Hungary | PET | 4 | <0.005–0.2 | Szendi et al. (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 et al. (2018) |
| DEHP             | Plastic bottled beverages (water) | Vietnam | Not stated | 8 | 10.3–42.3 | Tri et al. (2018) |
| DEHP             | Bottled water | Turkey | Not stated | Not stated | 10.06–11.90 | Özer et al. (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 et al. (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 et al. (2019) |
| DBP              | Mineral water | Iran | Not stated | 3 | 0.5–3.5 | Amiri et al. (2017) |
| DBP              | Mineral water | Vietnam | Not stated | 14 | 0.46–1.8 | Tran-Lam et al. (2018) |
| DEHP             | Mineral water | China | Not stated | 5 | <LOQ–0.733 | Yin et al. (2019) |
| DEHP             | Bottled mineral water | Hungary | PET | 4 | <0.29–11.289 | Szendi et al. (2018) |
| DEHP             | Bottled drinking water | China | Not stated | 60 | 0.013–0.021 | Li et al. (2019) |
| BBP              | Mineral water | China | Not stated | 1 | 0.001 | Wei et al. (2018) |
| DBP              | Mineral water | China | Not stated | 0.014 | | |
| DEHP             | Mineral water | China | Not stated | 0.018 | | |
| BBP              | Mineral water | China | Not stated | 0.32 × 10⁻⁴– 1.1 × 10⁻⁴ | Yang et al. (2017) |
| DBP              | Mineral water | China | Not stated | 1.3 × 10⁻⁴– 10.2 × 10⁻⁴ | | |
| DEHP             | Mineral water | China | Not stated | 2.2 × 10⁻⁴– 43.9 × 10⁻⁴ | | |

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

*Polyethylene terephthalate.
Shoeib (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...
| Detected analyte | Sample | Country | Type of bottle | Number of brands or samples | Storage study | Concentration (µg/L) | Reference |
|-----------------|--------|---------|----------------|----------------------------|---------------|---------------------|-----------|
| BPA             | Bottled drinking water | Iran | Not stated | 4 | Freezing temperature (24 h)<sup>a</sup> | 0.0023 | Kaykhaii et al. (2020) |
|                 |        |         |                |                            | Sunlight (for a week) | 0.007 |          |
|                 |        |         |                |                            | Boiled in a steel jar and quickly poured into the bottle (cooled to ambient temperature)<sup>a</sup> | 0.016 |          |
| BBP             | Mineral water bottle | Pakistan | Not stated | 5 | Sunlight (7 days with 10 h/day – 46–48°C) | ND<sup>b</sup> – 12.11 (median: 7.43) | Surhio et al. (2017) |
| BBP             | Bottled water | Iran | PET<sup>c</sup> | 10 | Sunlight (roof on sunny days for 1 week) | 0.03–0.13 | Abtahi et al. (2019) |
| DBP             | Mineral water bottle | Pakistan | Not stated | 5 | Sunlight (7 days with 10 h/day – 46–48°C) | ND<sup>b</sup> – 26.16 (median: 21.7) | Surhio et al. (2017) |
| DBP             | Bottled water | Iran | PET | 10 | Sunlight (roof on sunny days for 1 week) | ND<sup>b</sup> – 0.12 (median: 0.10) | Abtahi et al. (2019) |
| DBP             | Bottled water | Romania | Not stated | Not stated | Room temperature 1–4°C<sup>a</sup> | 6.11 | 5.12 | Sulentic et al. (2018) |
| DBP             | Water in plastic bottle | Iran | Not stated | 3 | Room temperature<sup>a</sup> | 5.32 | 10.12 | Hashemi-Moghaddam & Maddah (2018) |
|                 |        |         |                |                            | Freezing temperature<sup>a</sup> |          |          |
| DBP             | Bottled mineral water | Italy | PET | 15 | 6 months at 25°C | 1.23 |          | Cincotta et al. (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 et al. (2019) |
|                 |        |         |                |                            | Sunlight (23 ± 2°C at 5 days) | 5.86 | Not stated |
|                 |        |         |                |                            | 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 et al. (2017) |

(continued)
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

| Detected analyte | Country | Concentration (μg/L)* | EDI (μg/kg-bw/day)* | Contribution via bottled water (%) | ELCR# | Reference |
|------------------|---------|-----------------------|---------------------|-----------------------------------|-------|-----------|
| BPA              | Turkey  | 5.7                   | ≈0.163              | ≈4.1                              | –     | Karayaka et al. (2019) |
| BPA              | Iran    | 5.5                   | ≈0.157              | ≈3.9                              | –     | Mohammadnezhad et al. (2017) |
| BPA              | Malaysia| 1.25                  | ≈3.6 × 10⁻²         | ≈0.9                              | –     | Rozaini et al. (2017) |
| BPA              | China   | 0.127                 | ≈3.6 × 10⁻³         | ≈0.09                             | –     | Chang et al. (2017) |
| BPA              | China   | 0.08                  | ≈2.3 × 10⁻³         | ≈0.058                            | –     | Zhou et al. (2019) |
| BPA              | Iran    | 0.07                  | 2.0 × 10⁻³          | ≈0.05                             | –     | Gorji et al. (2019) |
| BPA              | China   | 0.01                  | ≈2.9 × 10⁻⁴         | ≈7.3 × 10⁻³                       | –     | Wu et al. (2019) |
| BBP              | Iran    | 5.5                   | ≈0.157              | ≈0.03                             | –     | Farahani et al. (2017) |
| BBP              | Spain   | 1.9                   | ≈5.4 × 10⁻²         | ≈1.1 × 10⁻²                       | –     | Barciela-Alonso et al. (2017) |
| BBP              | China   | 1.86                  | 5.3 × 10⁻²          | ≈1.1 × 10⁻²                       | –     | Wu et al. (2019) |
| BBP              | Vietnam | 0.95                  | 2.7 × 10⁻²          | ≈5.4 × 10⁻³                       | –     | Tran-Lam et al. (2018) |
| BBP              | China   | 0.690                 | 2.0 × 10⁻²          | ≈4.0 × 10⁻³                       | –     | Yin et al. (2019) |
| BBP              | China   | 0.032                 | ≈9.1 × 10⁻⁴         | ≈1.8 × 10⁻⁴                       | –     | Li et al. (2019) |
| BBP              | Spain   | <LOQ (0.006)          | <1.7 × 10⁻⁴         | <3.4 × 10⁻⁵                       | –     | Santana-Mayor et al. (2018) |
| DBP              | Mexico  | 82.8                  | 2.37                | 25.7                              | –     | Salazar-Beltrán et al. (2017) |
Table 4 | continued

Without the storage study

| Detected analyte | Country    | Concentration (μg/L) | EDI (μg/kg-bw/day) | Contribution via bottled water (%) | ELCR | Reference          |
|------------------|------------|----------------------|--------------------|-----------------------------------|------|--------------------|
| DBP              | Nigeria    | 42                   | 1.2                | 12                                | -    | Dada et al. (2018) |
| DBP              | Thailand   | 17.0                 | ≈0.486             | ≈4.86                             | -    | Pinsritpong & 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                 | ≈7.7 × 10⁻²        | ≈0.77                             | -    | Chen et al. (2019b)   |
| DBP              | Iran       | 2.5                  | ≈7.1 × 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                  | ≈4.9 × 10⁻²        | ≈0.49                             | -    | Farahani et al. (2017) |
| DBP              | China      | 1.34                 | ≈3.8 × 10⁻²        | ≈0.38                             | -    | Wu et al. (2019)      |
| DBP              | Iran       | 1.13                 | ≈3.2 × 10⁻²        | ≈0.32                             | -    | Soheilifar et al. (2018) |
| DBP              | Spain      | <1                   | <2.9 × 10⁻²        | <0.29                             | -    | González-Suárez et al. (2018) |
| DBP              | Vietnam    | 0.95                 | ≈2.7 × 10⁻²        | ≈0.27                             | -    | Tran-Lam et al. (2018) |
| DBP              | China      | 0.51                 | ≈1.5 × 10⁻²        | 0.15                              | -    | Li et al. (2019)      |
| DBP              | Spain      | 0.36                 | 1.0 × 10⁻²         | 0.1                               | -    | González-Suárez et al. (2017) |
| DBP              | Hungary    | <0.2                 | <5.7 × 10⁻³        | <0.057                            | -    | Szendi et al. (2018)  |
| DBP              | Spain      | 0.184                | ≈5.3 × 10⁻³        | ≈0.053                            | -    | Santana-Mayor et al. (2018) |
| DEHP             | Thailand   | 64.0                 | ≈1.83              | ≈3.66                             | ≈3.0 × 10⁻⁵ | Pinsritpong & Bunkoed (2018) |
| DEHP             | Vietnam    | 42.3                 | ≈1.21              | 2.42                              | ≈2.0 × 10⁻⁵ | Tri et al. (2018)    |
| DEHP             | China      | 2.50                 | ≈7.1 × 10⁻²        | ≈1.42                             | ≈1.2 × 10⁻⁶ | Wu et al. (2019)     |
| DEHP             | Italy      | 24.4                 | ≈0.697             | ≈1.39                             | ≈1.1 × 10⁻⁵ | Notard donato et al. (2018) |
| DEHP             | Turkey     | 11.90                | 0.34               | 0.68                              | ≈5.6 × 10⁻⁶ | Özer et al. (2017)   |
| DEHP             | Hungary    | <11.289              | <0.323             | <0.646                            | <5.3 × 10⁻⁶ | Szendi et al. (2018) |
| DEHP             | Chile      | 4.321                | ≈0.123             | ≈0.246                            | ≈2.0 × 10⁻⁶ | Manzo et al. (2019)  |
| DEHP             | Iran       | 3.5                  | 0.1                | 0.2                               | ≈1.6 × 10⁻⁶ | Amiri et al. (2017)  |
| DEHP             | Iran       | 3.0                  | 0.086              | 0.172                             | ≈1.4 × 10⁻⁶ | Amiri & Ghaemi (2017) |
| DEHP             | Vietnam    | 1.8                  | 5.1 × 10⁻²         | 0.1                               | ≈8.53 × 10⁻⁷ | Tran-Lam et al. (2018) |
| DEHP             | China      | 0.733                | ≈2.1 × 10⁻²        | ≈0.042                            | ≈3.4 × 10⁻⁷ | Yin et al. (2019)    |
| DEHP             | China      | 0.021                | 6.0 × 10⁻⁴         | 1.2 × 10⁻³                         | ≈9.9 × 10⁻⁹ | Li et al. (2019)     |

| Detected analyte | Country    | Concentration (μg/kg) | EDI (μg/kg-bw/day) | Contribution via bottled water (%) | CRI | Reference          |
|------------------|------------|----------------------|--------------------|-----------------------------------|-----|--------------------|
| BBP              | China      | Mineral water (MW): 0.11 | ≈3.1 × 10⁻⁴       | ≈6.2 × 10⁻⁴                       | -   | Yang et al. (2017) |
|                  |            | Soda water (SW): 0.13  | ≈3.7 × 10⁻⁴       | ≈7.4 × 10⁻⁴                       |     |                    |
Table 4 | continued

Without the storage study

| Detected analyte | Country             | Concentration (μg/L)a | EDI (μg/kg-bw/day)b | Contribution via bottled water (%)c | CRd | Reference                  |
|------------------|---------------------|-----------------------|---------------------|-------------------------------------|-----|---------------------------|
| DBP              | Mineral water: 1.02 | 2.9 × 10^{-2}         | 0.29                |                                     |     |                           |
|                  | Soda water: 6.34    | 0.181                 |                     |                                     |     |                           |
| DEHP             | Mineral water: 4.3  | 0.123                 | 0.246               | 5.2 × 10^{-8}                      | 3.4 |                           |
|                  | Soda water: 7.29    | 0.208                 | 0.416               |                                     |     |                           |

With storage study

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

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 μg/ 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 × 10^{-7} μg/L of DEHP in water, and MC is the maximum concentration (μg/L or μg/kg) of DEHP in bottled water (Jeddi et al. 2015). Here was considered the value of 4.7 × 10^{-7} μg/L for papers with concentrations given in mg/kg.

dContribution via bottle 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 μg/ kg/bw/day).

detected compounds at concentrations between 0.031 and 0.203 μg/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, 25.7 and 3.66% of TDI, respectively (Figure 3). The results demonstrate that...
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

| Reference                        | BPA  | BBP  | DBP  | DEHP |
|----------------------------------|------|------|------|------|
| 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 <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)f                 | ≈0.046 | ≈0.097 | ≈0.662 | 0.512 |
| Elderly (≥65 years)g             | ≈0.057 | ≈0.120 | ≈0.821 | ≈0.635 |
| Tolerable daily intake (µg/kg-bw/day) | 4    | 500  | 10   | 50   |
| Contribution via 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)f                | ≈1.15 | ≈1.94 × 10⁻² | ≈6.62 | 1.02 |
| Elderly (≥65 years)g             | ≈1.43 | ≈1.2 × 10⁻² | ≈8.21 | ≈1.27 |
| RfD (µg/kg-bw/day)                | 50   | 500  | 100  | 20   |
| HQ (based on maximum concentration) |      |      |      |      |
| 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)f                | ≈9.20 × 10⁻⁴ | ≈1.94 × 10⁻⁴ | ≈6.62 × 10⁻³ | 2.56 × 10⁻² |
| Elderly (≥65 years)g             | ≈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 <21 years)e      | 820   | 3,880 | 113  | 29.3  |
| Adult (≥21 years)f               | 833   | 3,910 | 114  | 29.5  |

(continued)
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, older adults (≥65 years of age) and pregnant are other susceptible groups due to their physiological properties change. As reported by Gerba et al. (1999), 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 (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 79.4 kg, f0.84 L/day for 75.4 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).

Hazard Quotient (HQ) – \( \frac{EDI}{RfD} \), where HQ is associated with the exposure via the specified exposure route (unitless) (Jeddi et al. 2015).

Safety factor (SF) – \( \frac{RfD}{EDI} \) (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. 2015). 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

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−05 | 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 79.4 kg, f0.84 L/day for 75.4 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).

|     | Contribution via drinking water – \( \frac{EDI}{RfD} \times 100 \) (Zaki & Shoeib 2018), where the TD1 for BPA, BBP, DBP, and DEHP are available for reference as established by EFSA. US EPA (1987a, 1987b, 1988, 2019a).
|     |
|     | Hazard Quotient (HQ) – \( \frac{EDI}{RfD} \), where HQ is associated with the exposure via the specified exposure route (unitless) (Jeddi et al. 2015).
|     | Safety factor (SF) – \( \frac{RfD}{EDI} \) (Luo et al. 2018).
|     | EEQ – \( \frac{EP \times c}{c} \), where EP and c denote the estrogenic potency of an individual estrogenic compound (in vitro bioassays) and its corresponding concentration (Liu et al. 2009).
|     | Safety factor (SF) – \( \frac{RfD}{EDI} \) (Luo et al. 2018).
|     | EEQ – \( \frac{EP \times c}{c} \), where EP and c denote the estrogenic potency of an individual estrogenic compound (in vitro bioassays) and its corresponding concentration (Liu et al. 2009).
|     | Safety factor (SF) – \( \frac{RfD}{EDI} \) (Luo et al. 2018).
|     | EEQ – \( \frac{EP \times c}{c} \), where EP and c denote the estrogenic potency of an individual estrogenic compound (in vitro bioassays) and its corresponding concentration (Liu et al. 2009).
|     | Safety factor (SF) – \( \frac{RfD}{EDI} \) (Luo et al. 2018).
|     | EEQ – \( \frac{EP \times c}{c} \), where EP and c denote the estrogenic potency of an individual estrogenic compound (in vitro bioassays) and its corresponding concentration (Liu et al. 2009).
|     | Safety factor (SF) – \( \frac{RfD}{EDI} \) (Luo et al. 2018).

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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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