REVIEW ARTICLE



Bisphenol A and its analogues: from their occurrence in foodstuffs marketed in Europe to improved monitoring strategies—a review of published literature from 2018 to 2023

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Abstract

In this review article, the research works covering the analytical determination of bisphenol A (BPA) and its structural analogues published from 2018 to present (February 2024) were examined. The review offers an overview of the concentration levels of these xenoestrogens in food and beverages, and discusses concerns that these may possibly pose to the human health and scrutinises, from an analytical perspective, the main biomonitoring approaches that are applied. This comes as a natural evolution of a previous review that covered the same topic but in earlier years (up to 2017). As compared to the past, while the volume of published literature on this topic has not necessarily decreased, the research studies are now much more homogeneous in terms of their geographical origin, i.e., Southern Europe (mainly Italy and Spain). For this reason, an estimated daily intake of the European population could not be calculated at this time. In terms of the analytical approaches that were applied, 67% of the research groups exploited liquid chromatography (LC), with a detection that was prevalently (71%) afforded by mass spectrometry, with over one-fourth of the groups used gas chromatography (GC)–mass spectrometry achieving comparatively superior efficiency as compared to LC. Derivatisation was performed in 59% of the GC studies to afford more symmetrical signals and enhanced sensitivity. Although the contamination levels are well below the threshold set by governments, routinely biomonitoring is encouraged because of the possible accumulation of these contaminants in the human body and of their interplay with other xenoestrogens.

Keywords Bisphenol A \cdot Endocrine disrupting chemicals \cdot Biomonitoring \cdot Food safety \cdot Food contamination \cdot Analytical chromatography

| Ab | breviations | | BPs | Bisphenols |
|-----------|-------------------|---|-----------|-------------------------------------|
| CE | ECs | Contaminants of Emerging Concern | SPE | Solid-Phase Extraction |
| PC | 2Ps | Personal Care Products | MIP | Molecular Imprinted Polymer |
| ED | DCs | Endocrine Disrupting Chemicals | MAE-MISPE | Microwave-Assisted Extraction- |
| BP | PA | Bisphenol A | | Molecularly Imprinted Polymer-Solid |
| ΒĽ | DGE | Bisphenol diglycidyl ether | | Phase Extraction |
| FC | CM | Food Contact Material | FLD | Fluorescence Detection |
| | | | MS | Mass Spectrometry |
| \square | Giacomo Russo | | MSPD | Matrix Solid-Phase Dispersion |
| | g.russo@napier.a | ac.uk | VA-DLLME | Vortex-Assisted-Dispersive Liquid |
| 1 | | | | Micro-Liquid Extraction |
| 1 | Department of P | harmacy, School of Medicine and Surgery, | UAE-SPE | Ultrasound-Assisted Extraction |
| | 80131 Naples. It | alv | LLE | Liquid–Liquid Extraction |
| 2 | Contro of Biomo | dising and Clabel Health, School of Applied | UVA-DLLME | Ultrasound-Vortex-Assisted Disper- |
| | Sciences Sighth | ill Campus Edinburgh Napier University 9 | | sive Liquid–Liquid Micro-Extraction |
| | Sighthill Ct, Edi | nburgh EH11 4BN, UK | PC | Polycarbonate |
| 3 | Consorzio Interu | niversitario INBB Viale Medaglie d'Oro | PET | Polyethylene Terephthalate |
| | 305, 00136 Rom | e, Italy | TDI | Tolerated daily intake. |

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| HDPE and LDPE | High- and Low-Density Polyethylene |
|---------------|------------------------------------|
| MC | Metallic Cans |
| PS | Polystyrene |
| PP | Polypropylene |
| PVC | Polyvinyl Chloride |
| SVHC | List of Substances of Very High |
| | Concern |
| | |

Introduction

Bisphenol A is a synthetic compound realized in the early twentieth century, emerged in the last few decades as a key monomer used as "building block" in the production of polycarbonate plastics and epoxy resins (Michałowicz 2014). The remarkable versatility, durability, and transparency of these BPA-based polymers made them the materials of choice for an array of applications, notably in the manufacturing of food and beverage packaging. As a result, the daily interaction between BPA-laden containers and the consumables they enclose has become unavoidable for billions across the globe (Usman and Ahmad 2016). BPA is found in many everyday consumer goods products made of very stable plastics, such as DVDs, plastic bowls, and thermal paper cash register receipts (Vandenberg et al. 2007), but the first evidence of its potential release from food contact material (FCM) and, of consequence, of the people exposure to this chemical due to the use of canned foods, was in 1996 by Food and Drug Administration (FDA-U.S) (Noonan et al. 2011). Produced through condensation of two parts of phenol and one part of acetone, BPA is chemically converted into a plastic polymer or epoxy resin and in the meantime firmly bonded in the polymer matrix. Unfortunately, under certain conditions, unbonded BPA residues, even if in ppm range, can be released into foodstuffs (Fattore et al. 2015; Guart et al. 2011).

The migration of BPA from packaging to food and beverages is a consequence of the dynamic interplay between the chemical composition of the packaging material and the contents it encases (Kawamura et al. 2001). This migration can be exacerbated by factors, such as temperature, storage duration and conditions, acidity, or composition of the products (Guart et al. 2011). Unfortunately, BPA can adversely interact with several hormone responsive receptors, mainly oestrogen and thyroid hormone binding, either as agonist or as antagonist, thus inducting endocrine disruption. Therefore, other bisphenol analogues, such as bisphenol S, bisphenol E, bisphenol AF, and many others, were synthesized to mitigate the endocrinedisrupting potential of BPA (Chen et al. 2016; Russo et al. 2019b). However, many BPA analogues were found to be as much or more estrogenic than BPA, as the evidence suggests that the chemical moieties responsible for the toxicological effects are those that indeed undergo the polymerisation reaction (Rosenmai et al. 2014).

For most BP analogues, the accidental release from the packaging is allowed, albeit within precisely prescribed limits. For this reason, food and beverage surveillance is crucial to assess that these limits are not exceeded. Typically, several analytical techniques such as gas and liquid chromatography with ultraviolet, fluorescence, and mass spectrometry-based detection are exploited. The present review aims at offering an overview of the analytical approaches more often applied to determine the occurrence of BPA and its analogues and their concentration levels in canned or not canned foodstuff marketed in Europe therein.

Inclusion criteria

The eligible studies investigate the occurrence of BPA starting from 2018 to February 2024 and therefore included in the current review research. Only manuscripts in English were considered. The search was performed in different database: PubMed, Sciences Direct, Web of Science, Google Scholar, and Scopus using keywords with the following terms; "bisphenol" AND ("food" OR "vegetable" OR "vegetables" OR "fruit" OR "fruits" OR "salt" OR "sugar" OR "honey"), "bisphenol" AND "seafood", "bisphenol" AND ("beverages" OR "milk" OR "wine" OR "beer" OR "alcohol" OR "soft drinks" OR "tea" OR "coffee"), "bisphenol" AND ("drinking water" OR "bottled water" OR "mineral water"), "bisphenol" AND ("meat" OR "poultry"), "bisphenol" AND ("cereals" OR "wheat" OR "pasta" OR "noodle"), "bisphenol" AND ("mixed food"), respectively. The output data were filtered by published year, document type, source type, and language as described above. Only primary, peerreviewed articles were collected, whereas reviews and conferences proceedings were excluded. The inclusion criteria for this study were: all peer-reviewed research articles investigating the occurrence of BPA into foodstuffs. In the review process, articles obtained from databases are assessed based on their titles and abstracts. All the information, such as publication year, foodstuff category and subcategory, limits of detection (LOD) and limits of quantification (LOQ), separation and detection methods, and range of concentrations retrieved, were extracted and pooled in the Tables. BPs occurrence, and key findings were extracted from the articles.

Migration of BPA and its analogues from food contact materials

From the plastics or can inner coating, BPA is released through chemical process of hydrolysis from the bonded polymer (Vom Saal and Hughes 2005) and its amount depends on temperature and/or heating duration, repeated use, brushing, type of food and packaging, manufacturing processes, irradiation, water hardness, as well as pH which could facilitate the BPA release and migration into foodstuff (Lim et al. 2009). Manufacturers are under obligation to provide information about the cleaning, sterilization methods, and product conditions on the labels of bottles or packaging. Several and various packaging materials are available in the foodstuffs industry field. These are polycarbonate (PC), polyethylene terephthalate (PET), high- and low-density polvethylene (HDPE, LDPE), metallic cans (MC), polystyrene, polypropylene (PP), and polyvinyl chloride (PVC). Several studies demonstrated the BPA releasing from these materials, even if these are labelled as BPA-free, as in the case of brushed baby bottles (Ali et al. 2018). Different packaging materials were studied for BPA migration into food by the use of simulants: amounts of BPA (PC: 1.4- 35.3 mg/kg) (Ehlert et al. 2008), (HDPE caps: $0.145 \,\mu\text{g/dm}^2$) (Guart et al. 2011), (LDPE: 0.128 µg/dm²) (Guart et al. 2011) or below the specific migration limit set for food at 0.05 mg Kg^{-1} (Krivohlavek et al. 2023) were found. Metallic cans have on their inner surface a thin layer of epoxy resins or organosols. If the polymerization is not fully completed or if there are issues during the food canning process, BPA leaches into the food. For instance, in a recent study, it has been demonstrated the occurrence of BPA concentration ranging from 8.91 to 14.01 pg/mL in canned soft drinks (Kumar et al. 2023). Hananeh et al. investigated the BPA migration from other kind of plastics, such as PP, and they found that some BPA migrated from the materials such as stainless steel, aluminum, and silicone water bottles albeit under standard limits (Astolfi et al. 2021; Hananeh et al. 2021).

Bisphenols regulations

The EU Chemicals Regulation (EC) No 1907/2006 of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) poses a responsibility on manufacturers and importers of chemicals to assess the potential risks posed to humans and the environment by all planned uses of BPA. They are required to complete and report their risk assessments in a chemical safety report, where the companies must outline the specific conditions under which BPA can be safely utilized throughout its entire life cycle. Additionally, they must implement the measures aimed at reducing the risks of contamination. As for the plastic products intended for food contact, the Regulation (EU) no. 10/2011 imposes a specific migration limit into foodstuffs of 0.05 mg kg⁻¹ for any chemical compound. BPA is a substance that may be carcinogenic, mutagenic, or toxic for reproduction (CMR), persistent, bio-accumulative and toxic (PBT), or very persistent and very bio-accumulative (vPBT). Due to its nature, notification requirements are needed under REACH, for products that contain BPA, for its potential to cause harm to the human health. On 8 July 2021, BPA has been included in the Candidate List of Substances of Very High Concern (SVHC) under REACH specifically under Category 1B, due to its toxicity as endocrine disruptor (Agencies 2023). If the concentration of BPA in EU for sale imported products exceeds 0.1% by weight, importers and manufacturers must submit notification to European Chemicals Agency (ECHA) via the "Substances of Concern" (SCIP database) under the Waste Framework Directive (WFD). The aim is to provide as much information as possible for consumers and waste operators. BPA is restricted under various regulations in the EU regarding, specifically, consumer products, FCM, and toys. Among the various European countries, some developed their own national frameworks regulating the uses and applications of BPA, such as France, that banned its usage in all food packaging including containers and utensils, as well as teethers and soother shields to be used by consumers of all ages. Indeed, the use of BPA was originally banned in the manufacture of baby bottles, import, export, and placement on the market of France under the Law 2010-729 of 30 June 2010, the limits for the global leaching into food from plastic FCM, varnishes, and coatings were set to 0.05 mg kg⁻¹. Subsequently, the Law No. 2012-1442 of 24 December 2012 expanded the restriction including all the packaging, container, or utensil intended to come into direct contact with food ((EU) 2011). Nevertheless, the import and sale of BPA-containing products for food contact purposes remain prohibited in France, and the French Constitutional Council ruled out that this ban does not apply to the marketing of such products in other countries. Since 2010, Denmark banned BPA in FCM for children under 3 years and included it in the Danish Environmental Protection Agency's List of Undesirable Substances (LOUS) (Agency 2022). In 2013, Belgium and Sweden banned the use of BPA in FCM intended for children less than 3 years old and in plastic articles like spoons and plates, as well as the Austrian law prohibited to manufacture pacifiers and teething rings with BPA (2011). In 2016, Sweden banned the use of BPA in the epoxy resins lining water pipes, but this legislative provision was withdrawn in the same year (Federal Law Gazette Part II 2011). Furthermore, in 2022, the German Federal Office for Chemicals (BfC) submitted a dossier including a proposal to ECHA to restrict the use of BPs. Successively, the dossier has been temporary withdrawn, since the German authorities advised that a significant proposal re-drafting was necessary and beneficial to achieve the intended goals. The updated Drinking Water Directive (EU) 2020/2184 requires water intended for human consumption to have a maximum of 2.5 μ g L⁻¹ of BPA and the Member States must comply with these standards by 12 January 2026. This directive also extend to the testing of polymeric materials in contact with drinking water, to ensure that they do not cause any BPA migration into water. Concerning the threshold of the tolerable daily intake (TDI) for humans, the European Food Safety Authority (EFSA) published in 2006 its first BPA-related risk assessment, setting a threshold of 50 µg kg⁻¹ body weight/day. In 2015, an updated assessment of exposure to BPA and its toxicity determined the lowering of this threshold to $4 \ \mu g \ kg^{-1}$ body weight/day (EFSA Panel on Food Contact Materials and Aids 2015). In 2023, EFSA published a scientific opinion on the de novo evaluation of public health risks related to the presence of BPA in foods, with TDI threshold lessened to 0.2 ng kg^{-1} body weight/day (Ramírez et al. 2023). As a consequence of BPA regulatory actions, industries shifted toward use of BPA alternatives or "BPA-free" products. Figure 1 represents the generic chemical structures of a bisphenol and bisphenol derivative.

It can be assumed that, if a molecule has a chemical structure close to that of BPA, it may also share some of its toxicity. Understanding the potential risks of BPA substitutes is important to ensure the continued protection for human health and environment. Within the EU, three bisphenols (BPA, bisphenol B (BPB), and 2,2-bis(4'-hydroxyphenyl)-4methylpentane) (BPP) have been identified as SVHCs. BPB as endocrine disrupting chemical shows adverse effects on the male reproductive system in rodents and fishes, clear estrogenic effects in rats and fishes, and possibly anti-androgenic effects, giving rise to an equivalent level of concern to those of other substances listed in points (a) to (e) of Article 57 of the REACH Regulation (2022) (Serra et al. 2019). ECHA gathered and catalogued bisphenol derivatives obtaining a total of 148 substances including 17 "bisphenols" with the generic "bisphenol" structure, and "bisphenol derivatives" having common structural features. This list was published in December 2021 in "Assessment of regulatory needs-Group Name: Bisphenols" (2022).

According to ECHA opinion, 34 BPs of this group should be restricted under the EU's chemicals legislation REACH as these may interfere with hormonal systems, even if the number may change after the acquisition of new data. However, a group of 26 suspected BPs may still be regulated in consumer goods due to their skin sensitisers action. Germany, that has proposed to restrict BPA in products to 0.001% by weight to reduce the amount of endocrine-disrupting chemicals released into the environment, has extended this rule also to other BPs, such as Bisphenol B, S, F, and AF. The European commission set a migration limit for certain epoxy derivatives in material and articles intended to come into contact with foodstuffs and food simulants up to 9 mg kg⁻¹ and 9 mg dm⁻² respectively, for the sum of BADGE (CAS No [1675-54-3]) and its adducts with water (BADGE. H₂O [76002-91-0] and BADGE.2H₂O [5581-32-8]) and up to 1 mg kg⁻¹ and 1 mg dm⁻², respectively, for the sum of BADGE and its adducts with hydrochloric acid (HCl) (BADGE.HCl [13836-48-1], BADGE.2HCl [4809-35-2] & BADGE.H₂O.HCl [227947-06-0]) ((EU) 2005). Instead, Bisphenol F diglycidyl ether (BFDGE) CAS No [39817-09-9] has been prohibited since 2005 ((EU) 2005).

Toxicological aspects and human health effects

Exposure to BPA has gained considerable global health attention due to its potential adverse health effects on human health. Depending on exposure extent, several studies indicated a relationship between BPA exposure and negative health outcomes (Liao and Kannan 2012). Bisphenol A has been linked to various human health hazards, mostly related to reproductive, metabolic, and immune system disorders. Indeed, many studies on humans showed that the most noticeable effect is its disruption of sex hormone activity, with direct influence on the development and function of the reproductive system, both on males and females.

Additionally, BPA causes alteration in thyroid function, stimulating the pituitary gland to produce thyroid hormones. These hormones regulate many activities in the body such as glucidic/lipidic metabolism and heartbeats. Indeed, the exposure to BPA affects the cardiovascular system (Rochester 2013) as well as may contribute to the development of hormone-dependent pathologies like obesity, or type 2 diabetes (Molina-López et al. 2023). Furthermore, due to its xenoestrogen properties, it has been linked to various types of cancer, such as liver, gastrointestinal, breast, and skin tumours (Dueñas-Moreno et al. 2023). To date, a substantial amount of literature suggests that maternal BPA exposure can have a negative impact on the outcomes of offspring



Fig. 1 a Bisphenol generic structure, and b structure of bisphenol generic derivatives; "X" represents a portion of the molecule that can be variously replaced with additional bonded groups; "Rn" indicates potential additional or replacement groups

in developing systems; for instance, it increases the risk of behavioural problems, associated with hyperactivity disorder, antisocial behaviour, sleep related issues, and language development (Jensen et al. 2019; Kanlayaprasit et al. 2021). Primary route of BPA is through the diet, i.e., canned foods and water bottles, secondary routes include inhalation, and transdermal intake. After ingestion, BPA is rapidly absorbed in the bowel as monoester forms due to their rapid hydrolysis by the gut lipases or esterase. As part of the phase II metabolism, BPA is converted into namely BPA glucuronide and BPA sulphate within the gastrointestinal tract and liver of humans and eliminated through urine, faeces, and sweat (Andra et al. 2016). The bioavailability of BPA is dependent on the exposure route and, therefore, it is an important factor for assessing BPA exposure risks in humans. BPA can be measured in serum as well as urine. Urine BPA testing is an alternative to more invasive methods; however, because of the rapid metabolism and excretion of BPA (Dekant and Völkel 2008), it merely measures excreted BPA and does not take into account current in vivo exposure (Dekant and Völkel 2008). Thus, serum may provide a more accurate and realistic indicator of exposure.

Analytical method for BPs analysis

Food and beverages can contain BPA and/or its analogues in both fresh and canned commodities because of migration from the food contact material or due to the previous contamination in the food chain (Russo et al. 2018, 2019a, b). Sample preparation is particularly crucial in food analysis as it allows signal suppression of the matrix, enhanced sensitivity, and concentration of the target analytes, thereby allowing trace analysis.

Tables 1 and 2 summarise the main sample preparation approaches. Solid food is typically subjected to homogenization, while liquid samples undergo degassing, filtration, and/ or centrifugation. Samples with high protein content may require protein removal through precipitation, which can be acid, salt, or solvent mediated (Polson et al. 2003). Drinking waters are added of ascorbic acid to remove the excessive residual chlorine by products (Petraccia et al. 2006). Canned foods that contain both liquid and solid components are usually filtered and treated separately.

BPs are commonly extracted by liquid–liquid extraction (LLE) and solid-phase extraction (SPE) either in cartridge format or dispersed (dSPE or QuEChERS) (Russo et al. 2019a). These sample preparation techniques have been established since at least 4 decades and they are based on the selected partitioning of the target analytes in a water immiscible organic solvent layer (typically n-hexane, ethyl acetate, or their mixtures) in LLE and on selective absorption/partitioning on a solid sorbent in (d) SPE. It should

be noted that both LLE and (d) SPE tend to have an environmental impact particularly when the latter is operated in reversed phase. Newer, more selective and with reduced carbon footprint sample preparation approaches are Microwave-Assisted Extraction (MAE), Dispersive Liquid Micro-Liquid Extraction (DLLME), and molecularly imprinted solid-phase extraction (MISPE).

Due to the development of analytical columns of smaller i.d. (particularly for LC), liquid-liquid microextraction (LLME), DLLME, vortex-assisted liquid-liquid microextraction (VALLME), and single-drop microextraction (SDME) have been widely adopted in food analysis providing enhanced cost-effectiveness and increased recovery rates, with the use of only minimal solvent volumes. DLLME requires two-step process: first, the analyte is extracted and dispersed, and then, the resulting mixture undergoes centrifugation. It employees a ternary blend consisting of an extraction solvent, a dispersion solvent, and the aqueous sample; the choice of the extraction solvents is pivotal for a good recovery and selectivity. The limitations of DLLME in solvent selection have been overcome by VALLME, introduced by Psillakis (2019), being the method easier in operation, more cost-effective, and the additional dispersing solvents-free. QuEChERS is another option in food sample preparation employing extraction salts, followed by the clean-up of supernatant using dSPE. On the other hand, SPE separates analytes by capturing them between a solid phase (sorbent) and a liquid phase (sample). The method requires the column conditioning, sample loading, washing, and elution, with various options related to the chosen sorbents, formats (cartridges, disks, 96-well plates, and pipette tips), and whether automated (online SPE) or conventional (off-line SPE). SPE offers the possibility to simultaneously enrich trace compounds, eliminate matrix interferences providing high pre-concentration factors. This technique facilitates efficient pre-concentration, sample clean-up, and compatibility with diverse detection techniques, making it a preferred choice in modern food analytical chemistry. Furthermore, it has undergone significant improvements, such as dispersive solid-phase extraction (d-SPE), solid-phase microextraction (SPME), magnetic solid-phase extraction (MSPE), and molecularly imprinted polymers (MIP) extraction. The latter is a particular SPE based on molecular recognition principles. MIP sorbents possess recognition sites that match the shape and physicochemical features of the target analyte, known as the template molecule. Considering both GC and LC methods to identify and quantify BPA and/or its analogues, we summarized in Tables 1 and 2 all the studies gathered in the scientific literature from 2018 to 2024.

By examining Fig. 2, it is evident that LC is the most commonly applied analytical approach accounting for more than two-thirds of the sourced articles. This is not surprising as overall LC allows analysis of compounds over a wider

| Table | 1 Liquid chromatograf | ohic methods for Bl | Ps determination | | | | | |
|-------|------------------------------------|---------------------|---------------------|--|---|--|------------|---|
| Y 20 | Matrix | PKG | Analytes | Sample amount extraction | Separation (column, elution) Detection | LOD and LOQ | % Recovery | Ref |
| 18 | Fish | NC | BPA | 0.5 g QuEChERS Kit ACN/H ₂ O 4:3 | Waters Acquity BEH C18 H ₂ O/MeOH pH 9 Gradient MS | 0.003 and 0.008 ngg ⁻¹ | 55.6-109.9 | Pico et al. (2019) |
| 18 | Mixed | C+Pp | BPA + 16 6 BDGEs | 1 g SPE (Oasis HLB) | Waters Acquity BEH C18 H ₂ O/ACN 90:10 (v/v) MS | <0.0007 and 0.0108 ng mL ⁻¹ | 90-104 | van Leeuwen et al. (2019) |
| 18 | Vegetables | NS | BPA | 0.2 g <i>d</i> -SPE (Alumina, Florisil and silica sorbents) | HALO C18 H ₂ O/ MeOH Gradient MS | 0.025 and 0.167 ng g ⁻¹ | 94-102 | Aparicio et al. (2018) |
| 18 | Milk and Dairy (Infant formula) | NS | Tetrabromobisphenol | 1 g DLLME (MeCN, MgSO4 and NaCl in acidic conditions) | Kinetex C18 MeOH/ H ₂ O 90/10 (v/v) MS | $0.04 \text{ and } 1 \text{ ng g}^{-1}$ | 88.5 | Martinez et al. (2019) |
| 18 | Beverages | NS | BPA | 0.5 L SPE (Oasis HLB) | Hypersil Gold H ₂ O/ACN Gradient MS | $0.025~{\rm and}~0.05~{\rm ng}~{\rm g}^{-1}$ | 101.8 | Huysman et al. (2019) |
| 18 | Raw and Cooked Seafood | NC | BPA | 1 g Pressurized liquid extraction with MeOH + gel per- meation chromatog- raphy | Acquity BEH C18 H ₂ O/MeOH pH 9 Gradient MS | $0.03 \text{ and } 0.20 \text{ ng g}^{-1}$ | 62.6–109.9 | Alvarez-Munoz et al. (2018), Jakimska et al. (2013) |
| 18 | Milk and Dairy, Raw Milk | NC | BPA | 2.5 mL SPE (Chromabond C18) | Synergi Fusion-RP 80 Å, ACN/H ₂ O 70:30 (v/v) FLD | 0.01 and 0.03 ng g ⁻¹ | du | Santonicola et al. (2018) |
| 18 | Vegetable | NC | BPA and BPF | 1.0 g matrix solid-phase dispersion | Ascentis Express RP- Amide H ₂ O/ACN Gradient MS | $240 \text{ and } 0.77 \text{ ng g}^{-1}$ | 38 | Margenat et al. (2018) ^b |
| 19 | Beverages | C | BPA+6 | 20 mL Strata TMX | Supelco C18 ACN/H ₂ O 55:45 FLD | 2.06–21.37 ng mL ⁻¹ | 70.2–106.7 | Russo et al. (2019b) |

| Table | 1 (continued) | | | | | | | |
|-------|----------------|-------------|---------------------------------|---|---|---|------------|---|
| Y 20 | Matrix | PKG | Analytes | Sample amount extraction | Separation (column, elution) Detection | LOD and LOQ | % Recovery | Ref |
| 19 | Beverages | U | BPA, BPB, BPF BADGE BFDGE | 5 mL BPA Affinimip® SPE | AscentisExpressRP- Amide H ₂ O/ACN Gradient FLD | 0.15/0.50 ng mL ⁻¹ | 76-103 | Cirillo et al. (2019), Gallo et al. (2017) |
| 19 | Milk and Dairy | NC | BPA | 2.5 mL SPE (Chromabond C18) | Synergi Fusion-RP 80 Å, ACN/H ₂ O 70:30 FLD | 0.005-0.016 ng g ⁻¹ | 70-100 | Santonicola et al. (2019) |
| 19 | Fish | NC | BPA | 0.5 g QuEChERS,(PSA, C18; GCB, graphitized carbon black, MgSO ₄) | Acquity BEH C18 H ₂ O/MeOH pH 9 Gradient MS | 5×10 ⁻⁴ -15 ng g ⁻¹ | 53-102 | Jakimska et al. (2013), Pico et al. (2019) |
| 19 | Beverages | U | BPA, BPB,BPF | 5.0 mL SPE (Strata® C18) | Kinctex® PFP H ₂ O/MEOH Gradient MS | 0.15 and 1.0 ng mL^{-1} | 74-98 | Gallo et al. (2019) |
| 19 | Mixed | P, G, C, Pp | BPA+12 | SPE (DSC-18) LLE (ACN/n-pentane) | Kinetex Phenyl-hexyl H ₂ O/MEOH Gradient MS | n.p. and -85 ng g^{-1} | 37-120 | Vavrouš et al. (2019) |
| 20 | Mixed | U | BPA | All cans content MAE-MISPE poly- propylene cartridges | Discovery C18 RP H ₂ O/MeOH (70/30 v/v) MS | 0.9 and 4.6 ng g ⁻¹ | 51–57 | Maragou et al. (2020) |
| 20 | Mixed | υ | BADGE+3 BFDGE+3 | 0.1 g Ultrasound-assisted solvent extraction of porous membrane | Kinetex® XB-C8 column Ammonium formate/ MeOH Gradient MS | 0.27 and 0.49 ng g ⁻¹ 0.78 and 1.5 ng g ⁻¹ | 78.3–112.6 | Szczepańska et al. (2020) |
| 20 | Fish | NP | BPA | 2.00±0.01 g AFFINIMIP® SPE | Kinctex® PFP H ₂ O/MeOH Gradient MS | 0.15 and 0.5 ng g^{-1} | 101.1 | Di Marco Pisciottano et al. (2020) |
| 20 | Vegetables | U | BPA | SPME | Luna® C18 H ₂ O/ACN Gradient FLD+MS | 5 and 10 ng g^{-1} | 72-90 | Vilarinho et al. (2020) |

| Table | 1 (continued) | | | | | | | |
|-------|-------------------------------|----------------|---------------|--|---|--|--|---|
| Y 20 | Matrix | PKG | Analytes | Sample amount extraction | Separation (column, elution) Detection | LOD and LOQ | % Recovery | Ref |
| 21 | Mixed food and bever- ages | U | BPA+19 | Whole content of canned food/1500 uL SUPRAS hexanol/THF/water hexanol/ THF | ACE 3 C18-PFP H ₂ O/MeOH Gradient MS | n.p and 0.06– 0.81 ng g ⁻¹ n.p. and 0.019– 0.19 ng g ⁻¹ | 73-114 | Caballero-Casero and Rubio (2021) |
| 21 | Mixed and infant food | G,C,Pp,P | BPA+7 | 5.0±0.05 g for milks, fruit mixes and vegetable mixes 2.0±0.02 g of infant cereals Liquid extraction (MeOH or MeOH/ acetic acid) -SPE online | C18 column H ₂ O/ACN Gradient MS | $0.2-2$ and $1-10 \text{ ng g}^{-1}$ | du | Sirot et al. (2021) |
| 21 | Mixed | P, C, Pp | BPA+6 | 2 g Liquid extraction (ACN/H ₂ O) | Acquity UPLC® BEH C18 column H ₂ O/MeOH Gradient MS | 0.1–1 ng/g and 0.4–4.0 ng g ⁻¹ | 91–105 | Gálvez-Ontiveros et al. (2021) |
| 21 | Milk | C+P+Pp | BPA | 2 g Matrix solid-phase dispersion (MSPD) Florisil® | Acquity UPLC® BEH Phenyl column H ₂ O/ACN Gradient MS | 0.00097 and 0.0032 ng g ⁻¹ | du | Herrero et al. (2021) |
| 21 | Beverages | U | BPA+12 | 5 g Heptane solution and ACN | Phenomenex® Phenosphere ODS columns H ₂ O/ACN:MeOH Gradient FLD | 5 and 12.5 ng g^{-1} | 75-102 | Lestido-Cardama et al. (2021) |
| 21 | Milk | TP (Pure-Pak®) | BPA and BPF | 1 mL Chromabond C18 SPE | Synergy Fusion-RP column ACN/H ₂ O (70:30, v/v) FLD | 0.03 and 0.1 ng mL ⁻¹ | 78.4-107.2 (BPA) 97.60-107.16 (BPF) | Mercogliano et al. (2021), Santonicola et al. (2021a) |
| 21 | Seafood | ЧN | BPA, BPF, BPS | 0.1 g Matrix solid-phase dispersion-Florisil® | Symta ACE 5 C18- PFP column H ₂ O/ACN Gradient DAD | 0.07–0.29 ng g ⁻¹ 0.25–1.12 ng g ⁻¹ | 78.5–87.0 | Cañadas et al. (2021) |

| Y 20MatrixFKGAnalysesSample anothSepandion (column, LOD and LOQ% RecoveryRef2BecrengesCBPA, BPFBAHHOMGHOMGHOMGHOMGHOMG21BiotCBADGEHNDHLMEHOMGHOMGHOMGHOMG22BiotCBADGEHNGNSNorbelHOMGHOMG23BiotCBADGEHNGNSNorbelHOMGHOMG24BADGEHNGNSNorbelNorbelNorbelNorbelHOMG25BADGEHNGNSNorbelNorbelNorbelNorbelNorbel26BADGEHNGNSNorbelNorbelNorbelNorbelNorbel27BHDGEBADGEHNorbelNorbelNorbelNorbelNorbelNorbel28Dary and insketNorbelNorbelNorbelNorbelNorbelNorbel29BevergesNorbelNorbelNorbelNorbelNorbelNorbel20Dary and insketNorbelNorbelNorbelNorbelNorbelNorbel21Buy and insketNorbelNorbelNorbelNorbelNorbelNorbelNorbel22BevergesNorbelNorbelNorbelNorbelNorbelNorbelNorbelNorbel22BevergesNorbelNorbelNorbelNorbelNorbelNorbelNorbelNorbel< | Table | e 1 (continued) | | | | | | | |
|---|-------|-------------------|-------------|---|--|--|--|------------|--|
| | Y 20 |) Matrix | PKG | Analytes | Sample amount extraction | Separation (column, elution) Detection | LOD and LOQ | % Recovery | Ref |
| $ \begin{array}{c c c c c c c c c c c c c c c c c c c $ | 22 | Beverages | υ | BPA, BPF,BPAF | 10 mL VA- DLLME MeOH/, octanoic acid 1:1 pH 6.00 | BEH C 18 H ₂ O/MeOH Gradient MS | 0,045–9.45 ng mL ⁻¹ | 70–120 | Baute-Pérez et al. (2022) |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | 22 | Fish | υ | BADGE, BADGE-H ₂ O BADGE-HC, BADGE-2H2O BADGE-HCI BADGE-H ₂ O-HCI BFDGE | 10 g QuEChERS The dispersive kit used for disper- sive solid-phase extraction (dSPE) contained 150 mg magnesium sulfate, 25 mg primary-secondary amine (PSA), and 25 mg C18 | Poroshell 120 SB-C18 H ₂ O /MeOH Gradient MS | $0.2 \text{ and } 5.2 \text{ ng g}^{-1}$ | 85-105 | Lapviboonsuk and Lee- pipatpiboon (2014), Toptancı et al. (2022) |
| $ \begin{array}{c ccccccccccccccccccccccccccccccccccc$ | 22 | Dairy and mixed | NC | BPA+8 | 5 g/3 g ACN:QuEChERS (MgSO4, NaCl,NaOAc) | Agilent Zorbax SB-C18 H ₂ O/ACN Gradient MS | 0.30 and 1.5 ng g^{-1} | du | Liotta et al. (2022), Xiong et al. (2018) |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | 22 | Beverages | U | BPA,BPB,BPS | 2 mL Strata X-PRO | Luna Polar H ₂ O/MeOH Gradient MS | 0.26–0.78 ng mL ⁻¹ | 78–105 | Schiano et al. (2022) |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | 22 | Beverages Milk | NC | BPA+7 BADGE BFDGE | 1 mL LLE 5 mL AFFINIMIP | Kinetex Phenyl-Hexyl H ₂ O/MeOH Gradient MS | 0.003–0.01 ng mL ⁻¹ 0.15–5 ng mL ⁻¹ | 64.2–106.3 | Di Marco Pisciottano et al. (2022a) |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | 22 | Milk | NC+Pp+P | BPAF, BPB, BPF, BPAF, BPB, BPE | 1 mL QuEChERS (ace- tic buffer/ACN/ NaCIMgSO4) | Hypersil Gold C18 ammonium acetate/ ACN Gradient MS | 0.19 ng mL ⁻¹ and n.p | > 80 | Frankowski et al. (2022) |
| | 52 | Beverages Milk | NC + TP + P | BPA+19 | 1 mL LLE 5 mL AFFINIMIP | polar-embeddedC18 H ₂ O/ACN Gradient FLD MS | 0.2–3 ng mL ⁻¹ W 0.003–1 ng mL ⁻¹ M 0.03–5 ng mL ⁻¹ –1 ng mL ⁻¹ | du | Di Marco Pisciottano et al. (2022b) |

| Table 1 | (continued) | | | | | |
|---------|-------------|-----|-----------------------------|-----------------------------|--|----------------------------|
| Y 20 | Matrix | PKG | Analytes | Sample amount extraction | Separation (column, elution) Detection | LOD and LOQ |
| 23 1 | Mixed | NS | BPA, BADGE+7 derivatives | 1 g SPE (ENVI- Carb®) | Raptor Biphenyl H,O/ACN | 0.2–0.6 ng g ⁻¹ |

| derivatives | SPE (ENVI- Carb®) and QuEChERS | H ₂ O/ACN Gradient MS |
|---|-----------------------------------|--|
| NS Not Specified, NP Not Packed, TP Tetrapack, C Canned, NC Not Can | anned, G Glass, P Plastic, | <i>Pp</i> Paper, <i>np</i> not present |

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85-107

Ref

Recovery

28

derivatives

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range of polarity and molecular volume, which is not the case for GC for which molecules need to feature inherent volatility or otherwise be derivatised. Due to its moderate hydrophobicity, BPA is hereby determined in reversed phase and the selectivity that is more frequently exploited is certainly C18 as such or sometimes polar embedded, typically used to enhance the retention of the more hydrophilic analyte which is relevant when BPA and its derivatives are included in multiresidue analysis. Some research groups use commercially available C18 columns using sterically hindered silanes to protect the siloxane bond, allowing to operate the chromatographic experiment at a wider pH range (typically pH 2-10). Again, this is not relevant for BPA as its phenolic OH moieties have pKa values around 10 and these are predominantly undissociated at the experimental pH. However, it could be useful for improving the peak shape and resolution of basic substances, which may be relevant when BPA is analysed along with some pharmaceuticals, for instance. A good share of phenyl stationary phases can also be observed. This is a reasoned choice as such a stationary phase depicts the interactions occurring between π -systems, that are widely represented in bisphenols, and allow exploitation of distinctive selectivities particularly when operated with alcohols (such as methanol and ethanol) as organic modifiers. With these regards, phenyl-hexyl stationary phases provide usefulness as they combine both hydrophobic and π -stacking interactions that are particularly ideal for branched BPs. Another selectivity that is now emerging for BP analysis is pentafluorophenyl, which was introduced as it affords orthogonality when compared to alkyl bonded phases. PFP features different selectivity because of the original overlapping of π - π , dipole, hydrogen bonding, and ionic interactions.

In pure principle, GC, which was conducted in 33% of the examined articles, is the technique allowing the highest resolution and efficiency. Of note, GC-MS is typically a more cost-effective alternative when compared with LC-MS of identical resolution. Moreover, matrix effects are typically minimised by the intrinsic selectivity of the analytical technique that filters out polar interferents. In terms of the selectivity that are conventionally exploited in GC, these are typically nonpolar (5%-phenyl)-methylpolysiloxane capillary column (HP-5Ms) or its equivalent (5%-diphenyl)dimethylpolysiloxane (DB-5Ms). The format, particularly film thickness and length, can be different depending on the applied temperature gradient, the matrix, and the carrier has employed. BPA can be also analysed without any derivatisation; however, this step is performed in the 59% of the scientific literature examined as it often provides sharper peaks and increased selectivity, particularly in complex mixtures. The most widely used derivatisation procedures are silvlation and acetylation. The former is performed by reacting BPA and its derivatives with bis(trimethylsilyl)

| Table | 2 Gas chromatographic | c methods for | r BPs determin | ation | | | | | |
|-------|-----------------------------------|---------------|----------------|---|----------------------------------|--|---|------------|--|
| Y 20 | Matrix | PKG | Analytes | Sample amount extraction | Separation (column) detection | Derivatisation (tech- nique) | LOD and LOQ | % Recovery | Ref |
| 18 | Ready to eat baby food | പ | BPA+6 | 2.0 g QuenChers (Disper- sive PSA solvent) | ZB-5MS capillary column MS | Yes (Silylation) | $0.1/1 \text{ ng g}^{-1} \text{ and} 0.5/4 \text{ ng g}^{-1}$ | 91–110 | Garcia-Corcoles et al. (2018) |
| 18 | Herbs and spices | NP | BPA | 0.5 g QuenChers (Disper- sive PSA solvent, MgSO4 and PSA) | Supelco SPB-5MS MS | No | 1.303 and 3.013 ng g ⁻¹ | 83-110 | Di Bella et al. (2018) |
| 18 | Vegetable | NC | BPA andBPF | 5.0 g matrix solid-phase dispersion | Sapiens X5-MS MS | Yes, (methylation of the acidic carbox- ylic groups) | dN | 38 | Margenat et al. (2018) ^b |
| 18 | Milk and Dairy, Infant formula | Z | BPA | 1 g of homogenised sample DLLME 20 μL of HCl 3 N until pH <5 fol- lowed by 2.5 mL of MeCN, 1 g of anhydrous MgSO4 and 0.25 g of NaCl, shake vigorously by hand for 5 and centrifuge the tube | MS MS | °Z | 0.02 and 0.05 ng mL ⁻¹ | 88 | Martinez et al. (2019) |
| 19 | Red wine | NC | BPA | 10 mL CH2Cl2/NaOH USVADLLME | VF-1 ms Varian MS | No | 1.05 and 5.7 ng mL ⁻¹ | 85–95 | Cinelli et al. (2019) |
| 19 | Milk | U | BPA | 2 g DLLME | DB-5MS MS | Yes, acetylation | $0.02 \text{ and} 0.05 \text{ ng mL}^{-1}$ | 88^{a} | Cunha et al. (2011), Martinez et al. (2019) |
| 20 | Spices and aromatic herbs | NC | BPA + 18 | SPE (Cao et al. 2015) | Supelco SPB-5MS MS | No | $0.005 \text{ and} 3.013 \text{ ng g}^{-1}$ | 83-110 | Lo Turco et al. (2020) |
| 20 | Honey | NS | BPA | 2.5 g UVA-DLLME | Fused silica TRB- 5MS MS | No | 11 and 22 ng g^{-1} | 71.5-100.4 | Notardonato et al. (2020) |
| 20 | Mixed | C and NC | BPA+8 | DLLME tetrachloroethylene and acetic anhydride | DB-5MS column MS | No | du | du | González et al. (2020) |
| 50 | Meat | U | BPA+8 | 5.0 g QuEChERS-DLLME tetrachloroethylene and acetic anhydride | DB-5MS column MS | Yes, acetylation | 0.15 and 0.5 ng g ⁻¹ | 67–101 | Cunha et al. (2020) |
| | | | | | | | | | |

| Table | 2 (continued) | | | | | | | | |
|-------|-----------------------------|-------------|----------------|---|---|---|--|------------|-----------------------------------|
| Y 20 | Matrix | PKG | Analytes | Sample amount extraction | Separation (column) detection | Derivatisation (tech- nique) | LOD and LOQ | % Recovery | Ref |
| 20 | Cereal grains | NS | BPA+1 | 2.0 g UAE with AcEt:MeOH (90:10 v/v) containing 3% of NH4OH -dSPE with PSA | Agilent HP-5MS column MS | Yes, silylation | 0.05-0.4 and 0.1-1.2 ng g ⁻¹ | 30–119 | Albero et al. (2020) |
| 20 | Fish | NS | BPA+6 | 1 g muscle liver DLLME with tetra- chloroethylene and acetic anhydride | DB-5MS column MS | Yes, silylation | $0.9 \text{ and } 1.3 \text{ ng g}^{-1}$ | 63-106 | Barboza et al. (2020) |
| 20 | Cereal-based food- stuff | NS | BPA | 2.0 g Polymeric sorbent LiChrolut EN and Oasis HLB (UAE- ASPE) | DB-5MS fused silica capillary column MS | Yes, silylation | 0.006 ng g ⁻¹ and n.p | 82–105 | Azzouz et al. (2020) |
| 21 | Vegetables and fruits | NS | BPA | 2.0 g UAE-SPE (hydro- philic Millex-LG PTFE) | DB-5MS column MS | Yes, silylation | 0.006–0.025 ng g ⁻¹ | 83-110 | Hejji et al. (2021) |
| 21 | Honey | P, G | BPA | 1 g DLLME (ACN and chloroform) | Agilent HP-5MS column MS | No | 0,6 and 2.0 ng g^{-1} | 82 | Peñalver et al. (2021) |
| 22 | Fish | NC | BPA+6 | 0.5 g QuEChERS (NaCl, NaSO4,dSPE EMR- Lipid sorbet) (DLLME (ACN/AA/ CCl4 | ZB-XLB MS | 4,4-DDT-d8/tonalide- d3 | 0.5 and 100 ng g^{-1} | 70-120 | Petrarca et al. (2022a, 2022b) |
| 23 | Food and beverages | NS | BPA+15 | 10 g QuEChERS (mag- nesium sulfate) method | OPTIMA-5 MS column MS | Yes, with bis(trimethylsilyl) trifluoroacetamide (BSTFA) | 0.03–1.66 and 0.10–5.55 ng mL ⁻¹ | 25-120 | Lucarini et al. (2023) |
| NS N | ot Specified, NP Not Pac | ked, TP Tet | rapack, C Cann | ied, NC Not Canned, G G | lass, P Plastic, Pp Paper | , <i>np</i> not present | | | |

 $^{\rm a}$ Average value $^{\rm b}GC\text{-}MS/MS$ for matrix-matched calibration purposes



Fig. 2 Percentages of the separation techniques used to determine BPA and its analogue from 2018 to date. In pie to pie: LC detection techniques in LC (right) and derivatisation (Y/N) in GC (left)

trifluoroacetamide (BSTFA) often with trimethylchlorosilane to favour the formation of a single derivatives which for BPA is trimethylsilyl BPA, [M-15]+, detectable at m/z 357 in electron impact mode (EI). The latter is achieved by reaction with acetic or trifluoroacetic anhydride leading to a base peak, [M-15]+, detectable at m/z 405 in EI due to the loss of a methyl group (Peñalver et al. 2021).

Occurrence of BPs in food

The diet is the main exposure route for BPs, as these can migrate from FCM, even if food can also extract BPs during processing and transportation, especially when PVC gloves or tubing are used. Even storage containers and cookware can accidentally release BPs into food through inks, adhesives, or coatings. According to the available monitoring European research articles from 2018 until nowadays, we reported the BPs occurrence in various foodstuffs in Table 3 along with their concentration levels, even if, for some food categories such as meat and cereals, we found only few monitoring assessments and we therefore gathered them under the "mixed food" heading.

Bisphenols in beverages

Several studies investigating BPs in mineral water and beverages were retrieved. Van Leeuwen et al. (2019) found trace levels of BPs with BPA being the most frequently detected in a limited selection of canned foods and beverages, along with BPS, BADGE, and some of its derivative (BADGE·HCl, BADGE·H₂O, BADGE·2H₂O) at relatively low concentration level. Di Marco et al. analysed over the total investigated 108 samples, 62 drinking water used in the milk production chain and the results showed very low concentrations $(0.01-1.0 \text{ ng mL}^{-1})$ of 17 investigated BPs (Di Marco Pisciottano et al. 2022a), while BPs were not found in the 20 bottled water analysed by Baute-Pérez et al. (2022). Only 14 samples of the 40 canned beers analysed by Cirillo et al. collected from the Italian market (Cirillo et al. 2019) were found to be contaminated by BPA or one of its analogues (concentrations ranging from 0.5 to 2.5 ng mL^{-1}), while Russo et al. found that all of the 39 analysed canned beer but one single sample were positive for one or more BPs along with four energy drinks. BPA was found in 33% of the 46 fruit juice samples packaged in PET or Tetra PakTM (0.50–2.85 ng mL⁻¹) by Gallo et al. (2019). Detected levels of BPs in tonic water, tea, cola, and beer, along bottled water, were found to be ranging from 6.4 to 9.1 ng mL⁻¹ in these investigated beverages marketed in Spain, according to a study authored by Caballero-Casero and Rubio (Caballero-Casero and Rubio 2021). Among beverages, also Italian plant-based beverages, such as soya, coconut, almond, oats, and rice, were found contaminated by some BPs with BPA occurring in 32% of the 34 samples screened (Schiano et al. 2022). Overall, the BP occurrence was attributed to various factors, such as the use of recycled PET, cross-contamination in bottling manufacturing, and even the occurrence of BPs in the water source itself. The BP levels lower (<LOQ) than previous years or absence in such liquid matrices, as reported in the Lucarini et al. research (Lucarini et al. 2023), may indicate the effectiveness of preventive measures

| Table 3 Foodstuffs moni | itoring studies in the 2018-2 | 2023 time window | | | | |
|---------------------------------|---|--|----------------|---|---|--------------------------------|
| Country (year) | Matrix | Analytes | No. of samples | Min concentration | Max concentration | Ref |
| Spain (2018) | Fish, River Fish | BPA | 59 | <٢٥٥ | 59.09 ng g ⁻¹ dw | Pico et al. (2019) |
| The Netherlands (2018) | canned food, beverages and drinkware | BPA + 16 6 BDGEs | 22 | <0.07 ng/mL | 68 ng g ⁻¹ | van Leeuwen et al. (2019) |
| Spain (2018) | leafy and root vegetables | BPA | 24 | <001> | 1.3 ng g ⁻¹ | Aparicio et al. (2018) |
| Spain (2018) | Milk and Dairy, Infant formula | Tetrabromobisphenol (TBBPA) and BPA | 50 | 0.77 ± 1.01 ng/mL free BPA 0.50 ± 0.02 ng/mL TBBPA | 0.88 ± 1.01 ng mL ⁻¹ free BPA 0.58 ± 0.34 ng mL ⁻¹ TBBPA | Martinez et al. (2019) |
| Belgium (2018) | Freshwater | BPA | 8 | <001> | n.r | Huysman et al. (2019) |
| 11 European Countries (2018) | Fish, Raw and Cooked Seafood | BPA | 65 | 8.26 ng/g | 69.1 ng g ⁻¹ | Alvarez-Munoz et al. (2018) |
| Italy (2018) | Milk and Dairy, Raw Milk | BPA | 22 | <001> | 2340 ng mL^{-1} | Santonicola et al. (2018) |
| Italy (2018) | Milk and Dairy, Cow Milk | BPA | 72 | 0.035 ng/mL | 2.776 ng mL ⁻¹ | Santonicola et al. (2019) |
| Spain (2018) | Vegetables, Lattuce | BPA and BPF | 25 | (0.59–2.32) 2.16 ng/g fw | 199 ng g ⁻¹ | Margenat et al. (2018) |
| Spain (2018) | Ready to eat baby food | BPA+6 | 15 | 1.0 ng /g | 49.2 ng g ⁻¹ | Garcia-Corcoles et al. (2018) |
| Spain (2019) | Milk | BPA | 50 | £ | £ 0.88 ng mL ⁻¹ | Martinez et al. (2019) |
| Italy (2019) | Beverages | BPA+6 | 52 | >LOQ | 1.358 ng mL ⁻¹ | Russo et al. (2019b) |
| Italy (2019) | Beverages | BPA, BPB,BPF+BADGE+BFDGE | 40 | >LOQ | 2.5 ng mL ⁻¹ | Cirillo et al. (2019) |
| Spain (2019) | Fish | BPA | 59 | 59.9 ng g ⁻¹ | 223.91 ng g ⁻¹ | Pico et al. (2019) |
| Italy (2019) | Fruit Juices | BPA, BPB, BPF | 46 | 0.50 ng mL^{-1} | 2.85 ng mL ⁻¹ | Gallo et al. (2019) |
| Italy (2019) | Milk | BPA | 72 | 0.035 ng mL ⁻¹ | 2.776 ng mL ⁻¹ | Santonicola et al. (2019) |
| Czech Republic (2019) | Various (edible oils, but- ter, and chocolate) | BPA+13 | 60 | 50 ng g^{-1} | 85 ng g ⁻¹ | Vavrouš et al. (2019) |
| Greece (2020) | Canned food | BPA | 6 | 7.3 ng g^{-1} | 42.3 ng g ⁻¹ | Maragou et al. (2020) |
| Poland (2020) | Canned food | BADGE+4; BFDGE+2 | .07 | 1.0 ng g ' | 093.0 ng g | Szczepanska et al. (2020) |

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| Table 3 (continued) | | | | | | |
|-------------------------------------|--|------------------------------------|----------------------|--|---|--|
| Country (year) | Matrix | Analytes | No. of samples | Min concentration | Max concentration | Ref |
| Italy (2020) | Fish | BPA | 44 | <lod< td=""><td>7.1 ng g⁻¹</td><td>Di Marco Pisciottano et al. (2020)</td></lod<> | 7.1 ng g ⁻¹ | Di Marco Pisciottano et al. (2020) |
| Spain-Portugal (2020) | Canned vegetables | BPA | 19 | nd | nd | Vilarinho et al. (2020) |
| Italy, Algeria, Tunisia (2020) | Spices and aromatic herbs | BPA | 53 26 65 | <l0q< td=""><td><pre>>COD</pre></td><td>Lo Turco et al. (2020)</td></l0q<> | <pre>>COD</pre> | Lo Turco et al. (2020) |
| Italy (2020) | Honey | BPA | 9 | <lod< td=""><td>5.05 ng g⁻¹</td><td>Notardonato et al. (2020)</td></lod<> | 5.05 ng g ⁻¹ | Notardonato et al. (2020) |
| Spain (2020) | Mixed | BPA+8 | 40 | 0.17 ng g^{-1} | 33.19 ng g ⁻¹ | González et al. (2020) |
| Portugal (2020) | Meat | BPA+8 | 30 | 4.3 ng g^{-1} | 202.3 ng g^{-1} | Cunha et al. (2020) |
| Spain (2020) | Cereal grain | BPA, BPF | 16 | 1.6 ng g^{-1} | 1740 ng g^{-1} | Albero et al. (2020) |
| North East Atlantic Ocean (2020) | Fish | BPA+6 | 150 | 1.0 ng g ⁻¹ | 25.3 ng g ⁻¹ | Barboza et al. (2020) |
| Spain (2020) | Cereal-based foodstuff | BPA | 12 | 0.022 ng g^{-1} | 0.620 ng g^{-1} | Azzouz et al. (2020) |
| Spain (2021) | Food and beverages | BPA+18 | 10 | 0.071 ng g^{-1} 0.06 ng mL^{-1} | 84 ng g ⁻¹ 9.1 ng mL ⁻¹ | Caballero-Casero and Rubio (2021) |
| Italy (2021) | Milk | BPF | 84 | <l0q< td=""><td>2.686 ng mL⁻¹</td><td>Santonicola et al. (2021a)</td></l0q<> | 2.686 ng mL ⁻¹ | Santonicola et al. (2021a) |
| Italy (2021) | Milk | BPF | 20 | <pre>>COO</pre> | 2.956 ng mL ⁻¹ | Santonicola et al. (2021b) |
| Turkey (2022) | Fish | BADGE + 5 deriva- tives + BFDGE | 33 | 60 ng g^{-1} | 220 ng g ⁻¹ | Toptancı et al. (2022) |
| Italy (2022) | Plant based beverages | BPA, BPB, BPS | 34 | <lod< td=""><td>18.17 ng mL⁻¹</td><td>Schiano et al. (2022)</td></lod<> | 18.17 ng mL ⁻¹ | Schiano et al. (2022) |
| Italy (2022) | Drinking water, milk | | 108 | 0.01 ng mL ⁻¹ (water), 0.1 ng ml ⁻¹ (milk) | 1.0 ng mL ⁻¹ in water,2.0 ng mL ⁻¹ milk | Di Marco Pisciottano et al. (2022a) |
| Spain (2022) | Water and beverages | BPA, BPF, BPAF | 20 | nd | nd | Baute-Pérez et al. (2022) |
| Italy (2022) | Cheese Provola and olive cake samples | BPA+8 | 20 | 1.69(provola) 3.27(olive cake) ng g ⁻¹ | 2.84/20.99 ng g ⁻¹ | Liotta et al. (2022) |
| Italy (2022) | Milk | BPA+19 | 46+15 with packaging | 0.5 ng mL^{-1} | 5.6 ng mL^{-1} | Di Marco Pisciottano et al. (2022b) |
| Portugal (2022) | Fish | BPA + 6 | 238 | 0.1 ng g^{-1} | 52.4 ng g ⁻¹ | Petrarca et al. (2022b) |
| Poland (2022) | Milk | BPA+6 | 19 | 0.12 ng mL ⁻¹ | 1.71 ng mL ⁻¹ | Frankowski et al. (2022) |
| Portugal (2022) | seafood | BPA+6 | 20 | 4.8 ng g ⁻¹ | 12.3 ng g ⁻¹ | Petrarca et al. (2022a) |
| Turkey (2022) | Milk | BPA | = | 2.1 ng mL ⁻¹ | 11.8 ng mL ⁻¹ | Kartal Temel and Gürkan (2022) |

| Table 3 (continued) | | | | | | |
|------------------------|---|----------------------------------|----------------|---|---------------------------|--|
| Country (year) | Matrix | Analytes | No. of samples | Min concentration | Max concentration | Ref |
| France (2022) | MixedVegetables | BPA BADGE + 2 derivatives | 12 | 43 ng g ⁻¹ | 1600 ng g ⁻¹ | Cariou et al. (2022) |
| Turkey (2022) | Convenience foods (54 samples), canned veg- etable oils (4 samples), olives (4 samples), and soft drinks (17 samples) samples) | BPA BADGE + derivatives | 79 | р | 1056 ng g ⁻¹ | Toptancı (2023) |
| Poland (2022) | Milk | BPA, BPS, BPF, BPAF, BPB, BPE | 14 | 0.35 ng mL^{-1} | 0.87 ng mL^{-1} | Frankowski et al. (2022) |
| Italy (2022) | Milk | BPA + 19 | 46 | 0.1 ng g ⁻¹ | 8.7 ng g ⁻¹ | Di Marco Pisciottano et al. (2022b) |
| Switzerland (2023) | Canned food and bever- ages | BPA+15 | 22 | <lod< td=""><td>40,650 ng g⁻¹</td><td>Lucarini et al. (2023)</td></lod<> | 40,650 ng g ⁻¹ | Lucarini et al. (2023) |
| <i>nd</i> not detected | | | | | | |

implemented by the various governments, differently from the past. Since BPs are lipophilic compounds, they are often found in fatty and or oily foods, while trace levels are typically occurring in beverages. Various researchers looked into the presence of BPA in beverages and, generally, compared to other food categories, these were evaluated to contain relatively low concentrations of this chemical and/or of its analogues, but, on average, higher concentration levels were detected in tap waters as compared to bottled plastic water (Russo et al. 2022, 2019b).

The BPs concentration values examined from 2013 to 2018 in our previous review (Russo et al. 2019a), showed BPs concentration values in water, tap or bottled as low as 0.83-1.13 pg mL⁻¹, broadly similar to those found in the scientific assessment considered in this updated piece of work (2018–2024) showing that an albeit minimal "baseline" contamination may occur due to environmental or packaging-related factors.

Occurrence of bisphenols in fishes and seafood

More than 17% of global protein human consumption (Dawson et al. 2022) derives from the intake of seafood. Indeed, many edible fish contain various BPs analogues in their muscles and other tissues. Contamination can occur not only due to migration from packaging, but also to the presence of BPs in waterbodies. In fact, BPs were also found in river/sea fishes bought in local markets (Akhbarizadeh et al. 2021). Although both freshwater and seawater fish have been studied, most research articles focused on seafood with the most of the studies performed by countries, such as Spain and Portugal, overlooking the Atlantic Ocean. In general, BPs' occurrence in this food category ranged from a minimum of 0.008 ngg⁻¹ to a maximum 223.91 ng/ ng⁻¹. An interesting monitoring assessment was performed by Alvarez-Munoz et al. (Álvarez-Muñoz et al. 2018) who analyzed 65 different seafood samples among the 12 highly consumed fish and shellfish species from 11 European countries. They aimed to cover the seafood consumption habits in western, northern, and southern Europe. According to their study, the concentration levels spanned from 8.26 to 69.1 ng g^{-1} . Based on BPA occurrence, the human exposure assessment indicated that the Spanish population has the highest BP exposure through seafood consumption. Indeed, Barboza et al. (Alvarez-Munoz et al. 2018) reported that the highest concentration of a BP was found in the liver (BPA, 302 ng g^{-1}), while in the muscle (BPE), the levels were comparatively lower (BPE, 272 ng g^{-1}). Considering the TDI of 4 μ g kg⁻¹ bw/ day for BPA assessed by the European Food Safety Authority (EFSA Panel on Food Contact Materials and Aids 2015), the estimated daily intake for BPs from fish consumption resulted higher than the recommended oral reference dose, posing a significant risk with potentially adverse effects on the human health over a lifetime. Petrarca and co-workers studied BP contamination in river fish and seafood by authoring two articles in the very same year focusing on river fishes (Petrarca et al. 2022b) and seafood (Petrarca et al. 2022a). The outcome was that BPA levels were higher in the river fishes (52.4 ng kg^{-1}) than in seafood (23.1 ng kg^{-1}). Considering our previous monitoring study on canned tuna and the published survey (2013-2017) (Russo et al. 2019a), the concentration values are, on average, rather similar or lower than in the past, with concentration values reaching values as high as 662 pg g^{-1} (Alabi et al. 2014). The global data analysis indicates that the presence of BPs in seafood can originate both from contamination occurring in the marine environment due to "seafood ready to eat" and canned seafood.

Milk and dairy products

During the 2018–2021 period, the consumption of milk products within the European Union (EU-27) fluctuated, surging to 53.4 kg per capita in 2022, resulting in the EU-27 having one of the world's highest rates of per capita milk consumption. Even if several studies were carried out on milk and dairy products, these were unfortunately conducted mostly in Italy (9 versus 14), complicating the assessment of a wider European scenario. The highest concentration value $(2.956 \text{ ng mL}^{-1})$ of BPs in milk was found by Santonicola et al. (2021b), who developed an HPLC method for determining BPF levels during milk processing. In our previous review, we found that the highest BPs concentration in dairy was 800 μ g kg⁻¹ (Ferrer et al. 2011) in canned skimmed milk from Spanish market. Considering the high consumption of milk, especially by infants, this can be an important source of exposure to BPA and its analogues. However, the observed concentrations were below either the specific migration limit (SML) of 0.05 mg kg⁻¹ into food from varnishes or coatings applied to food contact materials ((EU) 2018) or the t-TDI BPA values fixed. Nevertheless, BP traces can represent a risk to human health due to their potential synergistic effect with other EDCs.

Mixed food

The most relevant study covering meat-based food was by Cunha et al. (2020) who investigated 30 canned meat samples (sausages, pâtés, and whole meals), gathered from Portugal markets but manufactured in three different countries (Portugal, Spain, and France). They found a total concentration sum of BPA and eight of its analogues, higher than 50 ng g⁻¹, with a maximum of 236 ng g⁻¹ in half of the analysed samples. As to cereals, we could retrieve only two studies (Albero et al. 2020; Azzouz et al. 2020). The former reported BPA in all investigated samples (barley, rice, wheat, and oat) at 1.6 to 1742 ng g⁻¹ levels, and Bisphenol F at concentration levels up to 22 ng g⁻¹ (6 samples). The latter covered the cereals rice, maize, and wheat, indicated concentration levels ranging from 0.022 pg g⁻¹ to 0.620 ng g⁻¹. The paucity of meat and cereals studies along with the limited number of analysed samples do not allow to draw any significant conclusion about this food category and, consequently, their impact on human health due to their consumption. In European Union regulations, composite foods are defined as those containing both processed animal and plant products, such as ready to eat baby food, leafy and root vegetables, and spices. We also report in Table 3 the assessment of BPs content in composite foods, even if, again, no conclusions can be drawn due to the extremely varying nature of the foodstuffs.

Conclusions

The present review aimed at offering an update on the contamination levels of BPs in food and beverages in Europe from our latest review on the topic, which covered until the year 2017. Unfortunately, at this time, we could witness a scarcity of monitoring studies and a very homogeneous origin, with most country, if we except Poland, located in the southern part of Europe. For this reason, estimating a TDI would have been extremely challenging and would have offered a geographically sided view on this topic. This aspect may suggest that new plasticizers, hopefully with a reduced impact on the human health and/or mitigated endocrine-disrupting properties, are being developed and used in northern countries or that in these regions "greener" packaging such as glass and kraft cardboard are being exploited to a superior extent than in Southern Europe, reducing the amount of BP release or the likelihood of its occurrence. Another related possibility is that in the countries enforcing more stringent restrictions on the use of BPs, the contamination of foodstuff may indeed be so negligible to make such studies practically irrelevant and therefore discourage researchers to set up new monitoring work. However, considering that (1) the food and beverage market is extremely globalised, with frequent import and export of food commodities, (2) mixtures of contaminants, even at low concentration, can exert a synergistic effect on the endocrine system and (3) the levels found were not drastically lowered as compared to the past, it is still advisable to conduct monitoring studies to evaluate whether the enforcement of more stringent regulations translated into a decrease in the human exposure toBPs.

Data availability Not applicable in this specific case as this is a review article.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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