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Chemical compounds and toxicological assessments of drinking water stored in polyethylene terephthalate (PET) bottles: A source of controversy reviewed

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ABSTRACT

A declaration of conformity according to European regulation No. 10/2011 is required to ensure the safety of plastic materials in contact with foodstuffs. This regulation established a positive list of substances that are authorized for use in plastic materials. Some compounds are subject to restrictions and/or specifications according to their toxicological data. Despite this, the analysis of PET reveals some non-intentionally added substances (NIAS) produced by authorized initial reactants and additives.

Genotoxic and estrogenic activities in PET-bottled water have been reported. Chemical mixtures in bottled water have been suggested as the source of these toxicological effects. Furthermore, sample preparation techniques, such as solid-phase extraction (SPE), to extract estrogen-like compounds in bottled water are controversial. It has been suggested that inappropriate extraction methods and sample treatment may result in false-negative or positive responses when testing water extracts in bioassays. There is therefore a need to combine chemical analysis with bioassays to carry out hazard assessments.

Formaldehyde, acetaldehyde and antimony are clearly related to migration from PET into water. However, several studies have shown other theoretically unexpected substances in bottled water. The origin of these compounds has not been clearly established (PET container, cap-sealing resins, background contamination, water processing steps, NIAS, recycled PET, etc.).

Here, we surveyed toxicological studies on PET-bottled water and chemical compounds that may be present therein. Our literature review shows that contradictory results for PET-

List of abbreviations: AA, acetaldehyde; APEOs, polyethoxylated nonylphenols; BBP, benzylbutyl phthalate; BHET, bis(hydroxyethyl) terephthalate; BHT, butylated hydroxytoluene; BPA, bisphenol A; DBP, dibutyl phthalate; DiBP, di-iso-butyl phthalate; DEG, diethylene glycol; DEHP, di-2-(ethylhexyl) phthalate; DEHA, bis-2-ethylhexyl adipate; DEP, diethyl phthalate; DMSO, dimethyl sulfoxide; DMT, dimethylterephthalate; DOP, di-n-octyl phthalate; EEC, European economic community; EEQs, estradiol equivalents; GC–MS, gas chromatography–mass spectrometry; HDPE, high density polyethylene; HULYs, human blood lymphocytes; IPA, isophthalic acid; LDH, lactate dehydrogenase; MEG, ethylene glycol; NIAS, non-intentionally added substances; NP, 4-nonylphenol; OP, octylphenol; PA, polyamide; PC, polycarbonate; PhA, phthalic acid; PVC, polyvinylchloride; RPE, relative proliferative effects; Sb₂O₃, antimony trioxide; SEC–HPLC, size exclusion chromatography–high performance liquid chromatography; SML, specific migration limits; SPE, solid-phase extraction; SPME, solid-phase micro-extraction; SODIS, solar water disinfection; TPA, terephthalic acid; TDI, tolerable daily intake; TNPP, tris(nonylphenyl) phosphite; TOC, total organic carbon; YES, yeast estrogen screen.

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bottled water have been reported, and differences can be explained by the wide variety of analytical methods, bioassays and exposure conditions employed.

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1. Introduction

Polyethylene terephthalate (PET) is a semi-crystalline polymer belonging to the family of polyesters. It is the most favorable packaging material for drinking water. PET bottles have been marketed for the last four decades and they have gradually replaced polyvinylchloride (PVC) and glass bottles on the markets. In Europe, packaged water sales account for 44% of the market volume for non-alcoholic drinks in 2009, with an average individual consumption of 105 L per year (EFBW, 2011).

1.1. The synthesis of PET

The prepolymerization of dimethylterephthalate (DMT) or terephthalic acid (TPA) with ethylene glycol (MEG) is the first industrial step in the synthesis of PET. Both reactions generate low-weight oligomers and an intermediate compound named bis(hydroxyethyl)terephthalate (BHET). After this step, a second polycondensation is carried out with an Sb-, Ge- or Ti-based catalyst (ILSI, 2000; Fakirov, 2002).

During PET manufacturing, several degradation and decomposition reactions can occur. High temperatures and the presence of oxygen in the PET melt process can promote thermo-mechanical and thermo-oxidative reactions (Romão et al., 2009a). Also, PET hydrolysis can be induced by the

presence of water during the melt process (Zhang and Ward, 1995; Paci and La Mantia, 1998).

PET thermal degradation generates sub-products such as oligomers and diethylene glycol. Volatile organic compounds such as carbon monoxide, aldehydes (formaldehyde, acetaldehyde, benzaldehyde), C₁–C₄ aliphatic hydrocarbons, aromatic hydrocarbons (benzene, toluene, ethylbenzene and styrene), esters (vinyl benzene, methyl acetate), methanol, and acetophenone were identified in PET samples subjected to temperatures between 200 and 300 °C (Dzięcioł and Trzeszczyński, 2000). In addition, 1,3-dioxolane and 2-methyl-1,3-dioxolane as thermal degradation products in PET bottles (Franz and Welle, 2008).

PET thermal stability depends on the type of co-monomers used for its production (Holland and Hay, 2002b). Concerning bottle-grade PET, co-polymerization with diethylene glycol (DEG) and isophthalic acid (IPA) is usually carried out to minimize polymer thermal crystallization during the production of preforms and the blow-molding process. Both co-monomers reduce the size of spherulites and, as a result the final container is transparent (Holland and Hay, 2002a). Indeed, glass-like transparency is a valued commodity for drinking-water bottles.

1.2. The manufacture of PET bottles for drinking water

Injection blow-molding is the preferred process for manufacturing PET bottles. Amorphous preforms are

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