



Phthalates in Beverages – A Review

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Abstract

Phthalates as esters of phthalic acid and aliphatic alcohol are environmental contaminants which are commonly observed due to their widespread use as plasticizers. Their chemical and physical properties allow them to migrate into the environment and also into foods and beverages easily. The objective of this paper is a summarized overview of phthalate properties, their impact on human health, their content in beverages, especially in beer, including the ways of contamination, and last but not least also legislative regulations.

Keywords: Phthalates, Dibutyl phthalate, Bis(2-ethylhexyl) phthalate, Beverages, Beer

1 Introduction

Phthalates are a group of diesters of ortho-phthalic acid (dialkyl or alkyl aryl esters of 1,2-benzenedicarboxylic acid) as shown in [Figure 1](#).

These synthetic compounds, since their first use in the 1920s, have become the most commonly used plasticizers to improve the softness, flexibility, and extensibility of a variety of plastic products ([Holadová et al., 2007](#)). As phthalate-based plasticizers are not firmly bonded through covalent bonding in material, they are slowly released into the surrounding environment by volatilisation, leaching or migration ([Wormuth et al., 2006](#)). These compounds, have nowadays become ubiquitous pollutants in the environment ([Wittassek et al., 2011](#)).

Phthalates are used in many industries and are produced in extremely large quantities worldwide ([Wang et al., 2008](#)). Generally, phthalates can be added to numerous products of general use such as toys, personal care and household products, car cosmetics, various solvents, adhesives, glues, pesticides, food packaging, medical devices, electronics, tubing, and building materials ([Schettler 2006](#); [Heudorf et al., 2007](#)). Phthalates with low molecular weight, such as diethyl phthalate and dibutyl phthalate, are widely used as solvents to keep colour and scent in various consumable and personal care products, while phthalates with high molecular weight, such as

bis(1-ethylhexyl) phthalate, are primarily used to soften PVC products ([Cao, 2010](#)).

Foods can be contaminated either primarily during primary production as a result of contaminated water, soil and air, or secondarily during subsequent processing and all kinds of handling ([Corea-Tellez et al., 2008](#)). The materials with which food comes into contact form the main source of food contamination ([Wagner et al., 2009](#)). Food, drinking water, beverages and raw materials can be contaminated by means of plastic equipment used during production or by migration from the packaging material and from the printing ink ([Corea-Tellez et al., 2008](#); [Fierens et al., 2012](#)).

In case of beer and wine, phthalates contamination may occur from plastic gaskets, lids and stoppers ([Sendón et al., 2012](#)), as well as tetra packs, cans ([Fasano et al., 2012](#)) and bottles, depending on how the product is stored and sold. During brewing/fermentation, maturation and bottling of beer and wine, the liquids are in contact with tubes and joints made from plastic. During their production, they are also stored in large containers for a certain period that may last up to several weeks or months in case of wine, where maturation may also be done in steel or coated concrete containers. These can be additional sources of contamination. ([Carnol et al., 2017](#)).

Due to the potential hazard to human health and the environment, several international health and environment organisations have classified phthalates as priority pollutants.

The EU issued a list of chemicals with proven or potential endocrine system disrupting activity, such as dibutyl phthalate, benzylbutyl phthalate, and bis(2-ethylhexyl) phthalate (DEPH) (Regulation (EC) No. 1907/2006 of the European Parliament and of the Council).

2 Attributes of phthalates

Although phthalates are commonly used worldwide, they are prone to bio, photo and anaerobic degradation. This means that these pollutants do not survive long in the environment (Rudel et al., 2009; US EPA, 2012). Esters of phthalic acid with short alkyl groups, for example methyl and butyl groups, are water-soluble. Phthalates with long alkyl or aromatic parts in side chains are less water-soluble. Due to the long chain lipophilicity, the highest levels of phthalates are found in fatty foods, such as fish, meat, dairy products, and vegetable oils (Jurica et al., 2013).

There are two types of phthalate contamination: contamination with phthalate residues, which are undesired in products (for example phthalates in beverages and food), and contamination with phthalates intentionally added to products (for example phthalates in soft PVC products).

3 Human health risk

Ingestion, inhalation, dermal and intravenous exposure present the main route of human exposure (Heudorf et al., 2007). The main human exposition is through food products contaminated by DEPH from plastic container or wrappers. Exposure from drinking water and ambient air makes a minor contribution to the total daily intake (Karačonji et al., 2017)

Human exposure to phthalates is associated with endocrine disruption, delays in fertility, impairment in foetal development, increased risk of allergies, asthma, and cancer. The current tolerable daily intake values for DEPH, dibutyl phthalate and benzylbutyl phthalate established by the EU Scientific Committee for Toxicity, Ecotoxicity and the Environment (CSTEE) are based on studies of reproductive toxicity, since reproductive effects seem to have greater relevance for humans than the proliferation of peroxisome in rat liver causing cancer (CSTEE, 1998; EFSA, 2005a; EFSA, 2005b; EFSA, 2005c). One should be aware that phthalate levels determined in general population are far below the threshold levels at which the

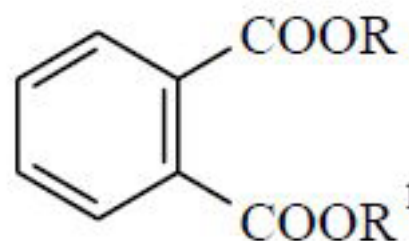


Figure 1 General structure of phthalates, R and R^1 is the same or different alkyl or aryl groups. For example - dibutyl phthalate $R = R^1 = (CH_2)_3CH_3$, for bis(2-ethylhexyl) phthalate $R = R^1 = CH_2CH(CH_2CH_3)(CH_2)_3CH_3$

first effects were observed in animals (Duty et al., 2003; Koch et al., 2004). However, due to the shared sources and routes of human exposure to different chemical classes (e.g. polychlorinated biphenyls, polybrominated diphenyl ethers, currently used pesticides – organophosphates and pyrethroids, bisphenol A, and phthalates), it is challenging to identify which chemical or metabolite is responsible for certain health effect (Robinson and Miller, 2015; Karačonji et al., 2017).

Phthalates with a long alkyl side-chain in the *ortho* position have a potential for reproductive and developmental toxic effects in humans. These include DEPH > dibutyl phthalate > benzylbutyl phthalate (in the order of their toxic potential), as well as diisononyl phthalate, di-*n*-hexyl phthalate, and diisobutyl phthalate (Fabjan et al., 2006; Lyche 2011; Karačonji et al., 2017).

Phthalates also prove a respiratory toxicity. Exposure to phthalates during childhood is often associated with impaired lung function, risk of bronchial obstruction, asthma and allergies in children, and asthma in adults (Hoppin and Ulmer, 2004; Jaakkola and Knight, 2008). The lungs of newborn rats exposed to DEPH exhibited histological changes similar to those observed in the lungs of children with bronchopulmonary dysplasia, a chronic lung disease typical for preterm infants (Magliozzi et al., 2003). Wheezing, rhinitis, and asthma in children could be associated with higher concentrations of benzylbutyl phthalate and DEPH in house dust (Bornehag et al., 2004; Kolarik et al., 2008; Bornehag and Nanberg, 2010; Karačonji et al., 2017).

Other studies (Hauser et al., 2007; Hauser, 2008) reported that DEPH and diethyl phthalate levels correlated with sperm DNA damage in exposed humans. However, as there were no available data for DEPH carcinogenicity in humans, but sufficient evidence of DEPH carcinogenicity in experimental animals, it has been classified as possibly carcinogenic to humans (Group 2B) (IARC, 2013; Karačonji et al., 2017).

4 Phthalate content in bottled water and beverages

Plastic bottles should be made as strong and rigid as possible, so plasticizers such as phthalates are not used in their production.

4.1 Water

Phthalates migrate to tap and bottled water (Peñalver et al., 2000; Bošnjir et al., 2007; Keresztek et al., 2013; Khedr 2013). The dominant source of phthalates in bottled water is from environmental contamination, and the levels of phthalates in bottled water are low in general. In the 3 samples of bottled water analysed by Yano et al. (2002) levels of dibutyl phthalate and DEPH were below 0.02 mg/kg. In another study (Cao, 2008), phthalates were analysed in samples of bottled water products in various container types (glass, PET, polycarbonate). Dibutyl phthalate was detected at levels ranging from 0.075 to 1.717 µg/l and DEPH in levels from 0.075 to 1.717 µg/l. Considerable differences in phthalate levels were not observed among the bottled water in different container types.

On the other hand, some studies show that migration from PET bottles could be the source of phthalates in bottled water contained in PET bottles. For example, the levels of phthalates in bottled water in glass containers were below or close to the detection limits, while slightly higher levels of phthalates were observed for bottled water in PET containers: dibutyl phthalate at 0.52 µg/l, and DEPH at 0.02 µg/l. (Montuori et al., 2008; Cao, 2010).

The report of the analysis of 30 samples of bottled mineral water in the Czech Republic showed no positive results (SZPI, 2004).

4.2 Soft drinks

Phthalates migrate also to soft and alcoholic drinks (Leibowitz et al., 1995; Bošnjir et al., 2007; Cao, 2008; Carrillo et al., 2008; Del Carlo et al., 2008; Russo et al., 2012; Cinelli et al., 2013; Chatonn et al., 2014; March and Cerdà, 2015; Wang et al., 2015; Jurica et al., 2016). Migration from plastic packaging to soft drinks is 5–40 times higher than migration to mineral water (Bošnjir et al., 2007). One of the reasons may be the difference in pH: lower than 3 in soft drinks and higher than 5 in all mineral waters (Karačonji et al., 2017).

4.3 Beer

Phthalates in beer can mainly come from raw materials, but can also be released from plastic materials, plastic coated equipment that is in a direct contact with beer or intermediates. Furthermore, they can be transferred to beer from

containers intended for transport and storage of beer. Metal crowns of glass bottles include a plastic seal made from polyvinyl chloride. In aluminium cans and bottles phthalates may originate from the polymer coating covering the aluminium inside the bottle or can. The alcohol content in beer may aid in releasing the phthalate into beer (Ye et al., 2009; Carnol et al., 2017; Olšovská et al., 2019).

Analyses of fifteen different beer samples stored in glass or aluminium bottles and cans from seven different Luxembourgish breweries showed that phthalates were detected in all samples with concentrations as high as 61.56 µg/l for total phthalates. The concentration range was from 0.05 µg/l for DEPH to 37.14 µg/l for dibutyl phthalate. DEPH presents the most commonly found phthalate in beer. The conclusion from this study indicates that there are no significant signs of phthalates content in beers stored in glass or aluminium bottles compared to cans (Carnol et al., 2017).

4.4 Wine and more alcoholic beverages

Phthalate content in some more alcoholic beverages is shown in Table 1. The most frequent phthalates detected in alcoholic beverages are dibutyl phthalate and DEPH (Karačonji et al., 2017). Phthalates can contaminate fruits through the deposition of dust and rain. They can also pollute fruit as early as being picked and stored into plastic bags for transport to production manufacturers. Another source of contamination can be during production by plastic and rubber components of pumps and other equipment, especially the ones in contact with the distillate. Dibutyl phthalate and diisobutyl phthalate are major phthalate contaminants from epoxy coatings for storage and fermentation tanks. Finally, alcoholic beverages are often contaminated with phthalates from packaging. In some synthetic corks used for wine and spirit bottling there was found diisobutyl phthalate. Concentrations about 200 µg/l of dibutyl phthalate was determined in vodka samples in plastic packaging (Leibowitz et al., 1995). Other studies suggested that phthalate contamination is not caused by plastic cork or packaging, but by the technological process of production itself, as there was no significant difference in phthalate content regardless the plug or packaging, specifically, between wines plugged with a plastic or normal cork, and those stored in plastic or glass (Carrillo et al., 2008). Likewise, no significant differences between plum spirit stored in plastic or glass bottles were found, in fact, phthalate concentrations were slightly higher in the spirit samples stored in glass (Jurica et al., 2016; Karačonji et al., 2017).

Czech monitoring study of three PET bottled wines proved no positive phthalate results. In the same study 20 samples of fruit distillates reported 10% positive results.

Table 1 Phthalate concentration in some alcoholic beverages, NA – not analysed (Leibowitz et al., 1995; Chatonnet et al., 2014; March and Cerdà, 2015; Wang et al., 2015; Jurica et al., 2016; Karačonji et al., 2017)

Matrix	Number of samples	Dibutyl phthalate (µg/L)	DEPH (µg/L)
Vodka	50	< LOQ=204	62–492
Plum spirit	20	25.2–822	16–1638
Chinese spirit	6	5–1946	156–1955
Brandy	1	65	NA
Wine (French)	100	< 4–2212	< 4–132
Sangria	1	30	NA

DEPH maximum concentration was 1.21 mg/l, dibutyl phthalate reached maximum at level 0.07 mg/l (SZPI, 2020).

5 Phthalate regulations

The Czech Head Public Health Officer set strict limits for phthalates expressed as a sum of dibutyl phthalate and DEPH in 1994. For non-alcoholic and also alcoholic beverages including beer and wine this limit was determined as 1.0 mg/kg (Gajdůšková et al., 1996). Later, in 1997 the Regulation No. 298/1997 of the Czech Ministry of Health defined a mild maximum limit for the sum of dibutyl phthalate and DEPH as 1 mg/kg for spirits, 2.0 mg/kg for beer and wine, and 4.0 mg/kg for dessert wine. As the European Union does not specify any limits for phthalates in food, the Czech Republic also has not been placing any restrictions on phthalates in food since the adoption of the European Union legislation in 2004.

However, the Czech Regulation No. 409/2005 about hygienic requirements for products in a direct contact with water and with water treatment defines a restriction for phthalates migration to water. This maximum migration concentration limit in water is 0.008 mg/l for DEPH (Jarošová, 2000). Hence, it reflects limits for the most common phthalate DEPH in drinking water set by the World Health Organisation (WHO) and US EPA that are 8 µg/l and 6 µg/l, respectively (WHO, 2005; US EPA, 2012).

Moreover, in January 2011 the European Union adopted a regulation that limits the migration (called specific migration limits) of some phthalates from a food contact material to food to quite low amounts, with maximum residue limits of 1.5 mg/kg for DEPH, 0.3 mg/kg for dibutyl phthalate, 30 mg/kg for butylbenzyl phthalate and 18 mg/kg for di(ethylhexyl) adipate (EC, 2011).

The tolerable daily intake set by the European Food Safety Authority (EFSA) is 50 µg/kg of body weight for DEPH (EFSA, 2005d), 10 µg/kg of body weight for dibutyl phthalate (EFSA, 2005b), 150 µg/kg of body weight for diisononyl phthalate and diisodecyl phthalate (EFSA, 2005a;

EFSA, 2005c), and 500 µg/kg of body weight for butylbenzyl phthalate (EFSA, 2005e; Karačonji et al., 2017).

6 Conclusion

Phthalates have become ubiquitous environmental contaminants and so their contamination is a widespread problem. Since foods, including beverages, present the main source of human exposure to phthalates, it is essential to monitor concentration levels of various food and beverages to protect human health. Attention must be focused not only on the technology process but also on the possible migration from beverage-packing materials plasticized with phthalates.

7 Acknowledgments

This study was supported by the Ministry of Agriculture of the Czech Republic within the institutional support MZE-RO1918.

The authors also thank to close colleagues for their help and unique creative atmosphere in the laboratory.

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