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Concentrations of several phthalates contaminants in Egyptian bottled water: Effects of storage conditions and estimate of human exposure

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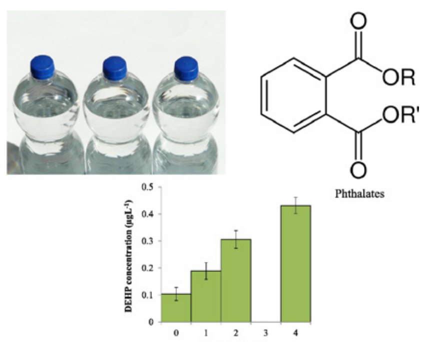
HIGHLIGHTS

- First time measurements of phthalates in Egyptian bottled water
- DEHP and DBP detected in 50 and 58% of samples respectively 2 weeks after production
- Positive correlation obtained between storage time and phthalate concentrations.
- Estimated intake of DEHP and DBP from bottled water were far below their TDI values.

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence and concentrations of six common phthalates were investigated for the first time in bottled water locally produced in the Egyptian market. The compounds investigated were dimethyl phthalate (DMP), diethyl phthalate (DEP), dibutyl phthalate (DBP), n-butyl benzyl phthalate (BBP), diethyl hexyl phthalate (DEHP), and Di-n-octyl phthalate (D-n-OP). A set of 108 bottled water samples from six different commercial brands of water bottled in transparent polyethylene terephthalate (PET) plastic bottles with high density polyethylene (HDPE) plastic caps were investigated. Water samples were analyzed immediately after purchasing (~2 weeks after production), after being stored at room temperature (25 ± 5 °C), in a refrigerator (4 ± 1 °C) and outdoor under sun exposure (daylight temperature of 40 ± 5 °C). Samples were stored up to six months depending on the tested condition. Among the target compounds, only DEHP and DBP were detected in the samples analyzed immediately after purchasing with a detection frequency of 50 and 58% and mean concentrations of 0.104 and 0.082 $\mu g\, l^{-1}$ respectively. Significant positive correlation was obtained between the storage time, temperature and the concentration of phthalate compounds detected in the bottled water, indicating possible migration from the PET plastic material as the source. The estimated contribution of bottled water consumption to the tolerable daily intake (TDI) levels of the two most abundant phthalates observed here for adults and toddlers did not exceed 0.16 and 0.72% for DBP while these values were 0.04 and 0.16% for DEHP respectively. These estimated daily intake values from PET bottled water consumption were far below their respective TDI values and therefore should constitute no adverse health effects.

1. Introduction

Packaging materials protect foods and beverages from chemical and environmental contaminations (Bhunia et al., 2013; De Fátima Pocas

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and Hogg, 2007). However, the packaging material itself may also present a potential source of chemical contamination to the many packaged products through the possible migration of some of their chemical constituents into the packaged products. These migrating constituents may include substances intentionally or non-intentionally added to the packaging materials (De Fátima Pocas and Hogg, 2007; Grob et al., 2006). Monitoring exposure of chemicals from packaging materials into foods has thus become a fundamental part of ensuring food safety and protecting human health (De Fátima Pocas and Hogg, 2007). Di-esters of ortho-phthalic acid are among such chemicals which require firm monitoring (Bhunia et al., 2013; Cao, 2010; De Fátima Pocas and Hogg, 2007).

Di-esters of ortho-phthalic acids, generally recognized as phthalates, are a class of synthetic organic chemical compounds that are extensively used in the manufacturing of polymers and many commonly used commercial products (Guo and Kannan, 2012; Swan, 2008; Latini, 2005). Phthalates are used primarily as plasticizers, which are added to the polymeric materials, especially polyvinyl chloride (PVC), to enhance their softness, durability and ease of processing (Meeker et al., 2009; Schettler, 2006). To date, phthalates are the most frequently used plasticizers accounting for over 80% of the plasticizers used globally (Guo and Kannan, 2012).

Phthalates are not chemically bound to the polymer matrices to which they are added. Accordingly, they can easily leach out from the products, during their use or after their disposal, migrating into air, dust, soil, water and food (Cao, 2010; Wormuth et al., 2006). Exposure to phthalates can cause serious adverse health effects on the kidney, liver, respiratory and endocrine systems (Meeker et al., 2007; Bornehag et al., 2004; Hoppin et al., 2004; ATSDR, 2002). Phthalates exhibit serious hepatotoxic effects and the International Agency for Research on Cancer classified DEHP as a class 2B probable carcinogen (Erkekoglu et al., 2014; ATSDR, 2002). The main health concerns are related to the endocrine disrupting potency of phthalates and their metabolites. Animal and in-vitro studies demonstrated that phthalates have anti-androgenic and weak estrogenic activities that consequently resulted in hazardous reproductive and developmental effects (Hauser et al., 2007; Lee and Koo, 2007; Meeker et al., 2007). Human studies are limited however, epidemiological studies seem to show correlations between phthalates exposure and adverse reproductive outcomes, including sexual hormones disorder, infertility, premature breast development and preterm labor (Swan, 2008; Hauser et al., 2007; Zhang et al., 2006; Latini et al., 2003). Studies have also shown that exposure to phthalates is associated with altering thyroid hormones levels, increased abdominal obesity, insulin resistance, asthma and respiratory allergic symptoms (Meeker et al., 2007; Stahlhut et al., 2007; Hoppin et al., 2004).

Recent reports suggest food and beverage to be the predominant source of human exposure to phthalates (Heinemeyer et al., 2013; Fierens et al., 2012; Fasano et al., 2012; Rudel et al., 2011; Cao, 2010). Bottled water as a potential source of human exposure to phthalates has drawn considerable attention lately due to its high and regular consumption despite its higher price relative to tap water, in part due to the perception of higher quality, purity and safety (Marcussen et al., 2013; Diduch et al., 2013; Bach et al., 2012). In fact the Codex Alimentarius general standard for bottled/packaged drinking waters requires compliance with the World Health Organization guideline for DEHP in drinking water not to exceed 8 $\mu g\ l^{-1}$ (WHO, 2011; Codex Alimentarius, 2001) while both the US FDA and US EPA have regulated the maximum permissible level of DEHP to be 6 $\mu g\ l^{-1}$ in bottled water and drinking water respectively (FDA, 2011; EPA, 2009).

The most common polymer used for the packaging of bottled water is polyethylene terephthalate (PET). Although PET is reported to be free from plasticizers or phthalates, several studies have shown the presence of phthalates in bottled water packed in PET containers (Jeddi et al., 2015; Guart et al., 2014; Keresztes et al., 2013; Al-Saleh et al., 2011; Amiridou and Voutsa, 2011; Montuori et al., 2008; Cao, 2008; Leivadara et al., 2008; Casajuana and Lacorte, 2003). Phthalates occurrence in PET bottled water is a controversial issue as PET polymers differ chemically from phthalates. Phthalates are esters of ortho-phthalic acids, while PET is polyester of para-phthalic acid. Thus, it is not chemically possible for PET to yield the phthalates in question through degradation or any other chemical pathway during the manufacturing or storage (Sax, 2010; Enneking, 2006). Furthermore, Phthalates are not typically used in PET manufacturing as PET bottles are required to be rigid, in order to attain good mechanical and gas barrier properties, while their elasticity can be tailored during their production through extrusion and molding (Bhunia et al., 2013).

In Egypt, the occurrence of phthalates in PET bottled water was not previously investigated. The aim of this study was to examine the occurrence and quantify the levels of phthalates in bottled water locally produced in the Egyptian market and to investigate the effects of various storage conditions on the levels of these contaminants. As it is not unusual for vendors and distributors in Egypt to transport and store bottled waters outdoors under direct sun exposure for extended periods, the levels of phthalate compounds were analyzed directly after purchasing (~2 weeks after production), the migration of phthalate compounds during storage under common conditions in the Egyptian market was investigated, comparisons of phthalate levels in the locally produced PET plastic bottled water with the global levels for PET bottled water are presented and finally an estimate of the exposure of adults and toddlers in Cairo to phthalate compounds through bottled water consumption and possible associated health effects are explored. To our knowledge, there are no previous published studies that have examined the phthalate contaminants in PET bottled water produced in Egypt.

2. Materials and methods

2.1. Samples collection and storage conditions

Bottled water samples ($n = 108$) were collected across six different brands (equivalent to 2 bottles from each brand per condition analyzed) that are locally produced and frequently consumed in the Egyptian market. The bottled water samples were geographically randomly collected in July 2013 from various local markets in Cairo, Egypt. All sampled bottled waters were of the same size (1.5 L) and those from a given brand were from the same batch. All the samples were packed in PET plastic bottles with HDPE plastic caps with no cap liners. The only brand of carbonated bottled water locally produced in Egypt was also examined to assess the effect of the presence of carbon dioxide gas on the phthalates levels, while the remaining five were non carbonated bottled drinking water.

Bottled water samples were analyzed a) directly after purchasing (~2 weeks after production) ($n = 12$); b) analyzed after 1, 2, and 4 months of storage ($n = 36$) after being stored outdoors (exposed directly to sunlight during the period of July to November 2013 where the daylight temperature was recorded to be 40 ± 5 °C); c) after 1, 2, and 4 months of storage at 4 °C ($n = 36$); and d) after 2 and 6 months of storage ($n = 24$) at room temperature (25 ± 4 °C). Each sample was analyzed in duplicates and the values for the duplicate analysis were averaged for data summaries and statistical analyses.

2.2. Chemicals and reagents

A standard mixture (Z-014A) containing standard solutions of dimethyl phthalate (DMP), diethyl phthalate (DEP), dibutyl phthalate (DBP), n-butyl benzyl phthalate (BBP), diethyl hexyl phthalate (DEHP), and Di-n-octyl phthalate (D-n-OP) dissolved in dichloromethane (each at a concentration of 2000 $\mu g\ ml^{-1}$) was obtained from AccuStandard (New Haven, USA). HPLC grade dichloromethane was purchased from Fisher Scientific (Loughborough, UK). Anhydrous sodium sulfate of analytical reagent grade was purchased from MP