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Frequency of use controls chemical leaching from drinking-water containers subject to disinfection

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ABSTRACT

Microbial-, and chemical-based burden of disease associated with lack of access to safe water continues to primarily impact developing countries. Cost-effective health risk-mitigating measures, such as of solar disinfection applied to microbial-contaminated water stored in plastic bottles have been increasingly tested in developing countries adversely impacted by epidemic water-borne diseases. Public health concerns associated with chemical leaching from water packaging materials led us to investigate the magnitude and variability of antimony (Sb) and bromine (Br) leaching from reused plastic containers (polyethylene terephthalate, PET; and polycarbonate, PC) subject to UV and/or temperature-driven disinfection. The overall objective of this study was to determine the main and interactive effects of temperature, UV exposure duration, and frequency of bottle reuse on the extent of leaching of Sb and Br from plastic bottles into water. Regardless of UV exposure duration, frequency of reuse (up to 27 times) was the major factor that linearly increased Sb leaching from PET bottles at all temperatures tested (13–47 °C). Leached Sb concentrations (~360 ng L^{-1}) from the highly reused (27 times) PET bottles (minimal Sb leaching from PC bottles, <15 ng L^{-1}) did not pose a serious risk to human health according to current daily Sb acceptable intake estimates. Leached Br concentrations from both PET and PC containers (up to ~15 μg L^{-1}) did not pose a consumer health risk either, however, no acceptable daily dose estimates exist for oral ingestion of organo-brominated, or other plasticizers/additives compounds if they were to be found in bottled water at much lower concentrations. Additional research on potential leaching of organic chemicals from water packaging materials is deemed necessary under relevant environmental conditions.

1. Introduction

Bottled water market has shown a 25% global increase in average consumption per capita between 2004 and 2009 (Rodwan, 2010). Increases in global population and urbanization along with climate change effects on water supply and availability have been charged with fluctuating consumer preference toward bottled water in both developed and developing countries. For example, Mexico, a developing country leads the list with the highest consumption of bottled water per capita, worldwide (Rodwan, 2010). Lack of access to safe water, sanitation and hygiene primarily impacts

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developing countries *via* water-borne bacterial and other emerging infectious diseases. Abundant, although often carrying a heavy microbial load, drinking-water in developing countries is a primary cause of elevated prevalence's of gastrointestinal illnesses (Ezzati et al., 2002). Solar disinfection of water packaged in plastic bottles (SODIS) is an increasingly adopted intervention measure aimed at reducing the public health risk from oral ingestion of contaminated water (Sommer et al., 1997; du Preez et al., 2010; Graf et al., 2010). The SODIS method is widely used around the world, enjoying use in 33 developing countries (Meierhofer and Landolt, 2009). In brief, SODIS-treated water is produced by exposure of drinking-water containers to solar UV radiation that inactivates microorganisms (SANDEC, 2002). While the primary working principle of SODIS is the germicidal effect of solar UV-A radiation, the associated elevated temperatures can also induce pasteurization effects. The efficacy of the SODIS process, therefore, is driven by both solar-, and temperature-based effects.

Consumer concerns about drinking water quality often stem from episodic events of chemical leaching from water-contact materials (bottled water) (Sax, 2010), but they are often considered unfounded (IBWA, 2011). Examples of chemical leaching from bottled water are: antimony (Shotyk et al., 2006; Shotyk and Krachler, 2007; Westerhoff et al., 2008; Krachler and Shotyk, 2009; Keresztes et al., 2009; Andra et al., 2012), bisphenol A (Casajuana and Lacorte, 2003; Cao and Corriveau, 2008; Le et al., 2008), phthalates (Biscardi et al., 2003; Casajuana and Lacorte, 2003; Criado et al., 2005; Bosnir et al., 2007; Schmid et al., 2008; Leivadara et al., 2008), adipates (Schmid et al., 2008), and 4-nonylphenol (Amiridou and Voutsa, 2011). Little, if any, attention has been paid upon possible contaminant migration during solar disinfection of plastic bottles containing microbially-contaminated water. While the solar disinfection method mentions "aging reduces UV transmittance and hence use fresh bottles" (SANDEC, 2002), it is unlikely that individuals in the developing countries follow this recommendation, because it is not economically viable or convenient to purchase new bottle after each use, leading to frequent reuse of bottles. Studies conducted using PET containers under recommended SODIS conditions claim no potential human threat, despite release of DEHA, DEHP (Schmid et al., 2008) and aliphatic aldehydes (Wegelin et al., 2001). However, though statistically non-significant, slightly elevated levels of DEHA and DEHP in water were observed from reused (unknown frequency of reuse) SODIS containers compared to the fresh ones (Schmid et al., 2008). The effect of the frequency of bottle reuse on the magnitude of chemical leaching as well as on the disinfection effectiveness is currently unknown. However, aged and reused plastic bottle containers (19 L) by bottled water distributing companies may be subject to relatively harsh cleaning procedures, i.e., reuse of water containers for as much as 50 times after bottle washing with sodium hydroxide and hydrogen peroxide at 75 °C (personal communication, Xristodoulou Bottled Water Company, Limassol, Cyprus). In India, these bottles were used at least for 100 cycles with mild soap and sodium hypochlorite wash between cycles, followed by drying with hot air (personal communication, Ram Das, Sri Krishna Bottling Company, Delhi, India). Similarly, up to 80 times of consecutive reuse of PC bottled water containers after sun exposure for 1 h, autoclaving and chemical disinfection has been demonstrated in Bangladesh (personal communication, Shwapon Biswas, ICDDRB, Dhaka, Bangladesh).

Several studies evaluated antimony leaching behavior from water-contact materials as a function of either temperature, or storage time, or container material, or UV exposure (Table SI-1 with references). However, none of these studies took bottle reuse into consideration, except for Schmid et al. (2008) who compared leaching of DEHA and DEHP between fresh and reused (unknown frequency) containers exposed to solar water disinfection (SODIS) procedures. We decided to focus on antimony (Sb), so far the only inorganic contaminant leaching from plastics for which induction of neoplasias, or endocrine disrupting effects have been reported (Choe et al., 2003; WHO, 2003), and bromine (Br) representing brominated compounds being part of water and food contact materials that has received little attention. Polybrominated diphenyl ethers (PBDE), in particular decabromodiphenyl oxide (BDE-209), are used as flame retardants in the preparation of both PET and PC plastics (Albemarle, 2011), while tetrabromobisphenol A (TBBPA) is used in epoxy and PC resins for the same purposes (Talsness et al., 2009). The presence of PBDEs (BDE-153, 183, 196, 197, 203, 206, 207, and 209 congener) in plastics, such as high density polyethylene (HDPE), polystyrene (PS), polypropylene (PS) has been reported by Mingwu et al. (2010), and tetrabromodiphenyl ether in PET by CDS Analytical, Inc. (2010). Other brominated compounds used as plastics additives in the making of PET and PC materials are pentabromobenzyl acrylate, pentabromotoluene, ethylene bis-tetrabromo phthalimide, pentabromobenzyl acrylate in PET, and ethylene bis-tetrabromo phthalimide, 1, 2-bis(2, 4, 6-tribromophenoxy)ethane in PC material (Covaci et al., 2011).

Using a central composite experimental design (CCD) (Mason et al., 1989), this study concomitantly investigated the effects of frequency of bottle reuse, temperature, and UV exposure on the extent of leaching of Sb and Br from UV-disinfected (SODIS) plastic containers (PET and PC). The generated knowledge of this study may be extremely useful to developing countries widely using solar disinfection as a means of improving access to safe water, sanitation and hygiene. The specific aims of this study were to (i) determine the main and interactive effects of temperature, UV exposure, and frequency of bottle reuse on soluble Sb and Br levels in bottled water, and (ii) estimate human daily Sb and Br intakes associated with leaching from water packaging materials under different usage conditions (SODIS), including a worst-case scenario.

2. Materials and methods

2.1. *Solar disinfection simulated experimental setup*

Recommendations for solar disinfection of microbial-contaminated water include filling colorless and transparent 2 L plastic bottles made of PET, not exceeding 10 cm water depth when laid horizontally for solar UV exposure either 1 h at >50 °C, or 6 h if up to 50% cloudy day, or for 2 consecutive days if >50% clouds (SANDEC, 2002). Following SODIS