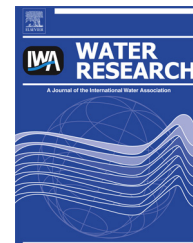




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Occurrence of organophosphate flame retardants in drinking water from China

Jun Li^a, Nanyang Yu^a, Beibei Zhang^b, Ling Jin^c, Meiyong Li^a,
Mengyang Hu^a, Xiaowei Zhang^a, Si Wei^{a,*}, Hongxia Yu^{a,*}

^a State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing University, Nanjing 210023, China

^b State Environmental Protection Key Laboratory of Monitoring and Analysis for Organic Pollutants in Surface Water, Jiangsu Provincial Environmental Monitoring Center, Nanjing 210036, China

^c National Research Centre for Environmental Toxicology (Entox), The University of Queensland, QLD 4108, Australia

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ABSTRACT

Several organophosphate flame retardants (OPFRs) have been identified as known or suspected carcinogens or neurotoxic substances. Given the potential health risks of these compounds, we conducted a comprehensive survey of nine OPFRs in drinking water in China. We found total concentrations of OPFRs in tap water ranging from 85.1 ng/L to 325 ng/L, and tris(2-butoxyethyl) phosphate (TBEP), triphenyl phosphate (TPP), and tris(2-chloroisopropyl) phosphate (TCPP) were the most common components. Similar OPFR concentrations and profiles were observed in water samples processed through six different waterworks in Nanjing, China. However, boiling affected OPFR levels in drinking water by either increasing (e.g., TBEP) or decreasing (e.g., tributyl phosphate, TBP) concentrations depending on the particular compound and the state of the indoor environment. We also found that bottled water contained many of the same major OPFR compounds with concentrations 10–25% lower than those in tap water, although TBEP contamination in bottled water remained a concern. Finally, we concluded that the risk of ingesting OPFRs through drinking water was not a major health concern for either adults or children in China. Nevertheless, drinking water ingestion represents an important exposure pathway for OPFRs.

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1. Introduction

Organophosphate flame retardants (OPFRs) are widely used in a variety of commercial products, including plastics, foams, paints, textiles, and furniture (Marklund et al., 2003). Previous experiments have demonstrated that OPFRs can have toxic effects on living organisms. For example, tris(2-chloroethyl) phosphate (TCEP) is carcinogenic (WHO, 1998) and acts as a

neurotoxin in rats (Tilson et al., 1990) and mice (Umezue et al., 1998). Tris(1,3-dichloro-2-propyl) phosphate (TDCPP) and tris(2-chloroisopropyl) phosphate (TCPP) (WHO, 1998) are also suspected carcinogens. Triphenyl phosphate (TPP) is acutely toxic to aquatic organisms (Leisewitz et al., 2000) and is a suspected neurotoxin (Andresen et al., 2004; Ni et al., 2007).

Because OPFRs are typically water-soluble and are added to products instead of being chemically bonded to them, these substances can easily diffuse into surrounding environments,

* Corresponding authors. Tel./fax: +86 25 8968 0356.

E-mail addresses: weisi@nju.edu.cn (S. Wei), yuhx@nju.edu.cn (H. Yu).

particularly in water. OPFRs have been detected in aquatic environments in many countries, including the United States (U.S.) (Benotti et al., 2009), Germany (Fries and Puttmann, 2001), Austria (Martinez-Carballo et al., 2007), Japan (Fukushima et al., 1992), Italy (Bacaloni et al., 2008), Spain (Rodil et al., 2012), and China (Wang et al., 2011). In addition, according to the published literature (Benotti et al., 2009; Stackelberg et al., 2004), drinking water treatment processes are often not effective in removing OPFRs, and these substances are commonly detected in tap water. In one study, TCEP and TDCPP were detected at maximum concentrations of 200 ng/L and 240 ng/L, respectively, in tap water tested from 19 U.S. water utilities (Benotti et al., 2009). Therefore, drinking water consumption can be considered an important pathway through which humans can be exposed to OPFRs, and the contamination of drinking water by OPFRs is of significant concern. Many OPFRs are high-production-volume chemicals with production levels at approximately 200,000 tons/year globally (EFRA, <http://www.cefic-efra.com>). In China, more than 70,000 tons of OPFRs were produced in 2007 (Wang et al., 2010), accounting for 35% of global production (EPA, 2006; Reemtsma et al., 2008). Although OPFRs have been detected in aquatic environments in China, including the Songhua River (Wang et al., 2011) and Taihu Lake (Yan et al., 2012), a broad survey of OPFRs in Chinese drinking water has not been previously conducted. In this study, we used an established ultra performance liquid chromatography-tandem mass spectrometry method to measure concentrations of nine OPFRs in drinking water in China, including bottled water and tap water from eight representative key cities. We also tested the concentrations of OPFRs in tap water before and after boiling, a common ‘treatment’ practice in Chinese households. Finally, we assessed the health risks of OPFRs ingested through drinking water.

2. Materials and methods

2.1. Chemicals

Standard for TDCPP was purchased from Tokyo Chemical Industry, and standards for the other eight OPFRs (tris(2,3-dibromopropyl) phosphate (TDBPP), tris(2-ethylhexyl)

phosphate (TEHP), tributyl phosphate (TBP), tris(2-butoxyethyl) phosphate (TBEP), 2-ethylhexyl diphenyl phosphate (EHDPP), TPP, TCPP, and TCEP) were purchased from Sigma–Aldrich. Deuterated tributyl phosphate (d₂₇-TBP, 98–99%) obtained from Cambridge Isotope Laboratories was used as a surrogate standard. Methanol (HPLC grade) and Acetonitrile (ACN, HPLC grade) were purchased from Tedia Company (Fairfield, OH, USA). Formic acid (HPLC grade) was purchased from ROE scientific INC. (Newark, DE, USA). Ultra-pure water was produced by a Milli-Q Advantage A10 system (Millipore, Bedford, MA, USA).

2.2. Sample collection

Thirty-nine tap water samples were collected from eight cities of China in 2012 (Fig. 1a). These eight cities were selected as representative of the broad array of cities in China, including inland/coastal and developed/less developed cities. We sampled water from three sites near the center of each city (Table S1, Supporting Information) with the exception of Nanjing, where we collected eighteen tap water samples from local households (Fig. 1b). Samples collected from Nanjing originated from six regions corresponding to six waterworks (Table S2, Supporting Information), representing drinking water for more than 6 million people in Nanjing. We collected 1 L of water for each sample, and all samples were stored at 4°C until pretreatment within 24 h. We also purchased eight popular brands of bottled water from local supermarkets, and they were all packaged in 550–600 mL polyethylene terephthalate (PET) bottles. Two replicates were analyzed for each brand.

2.3. Sample pretreatment

We spiked 1 L samples (or 550–600 mL samples for bottled water) with 5 µL of the surrogate standard solution d₂₇-TBP (10 mg/L in methanol) and performed solid phase extraction (SPE) using ENVI-18 (Supelco, 1 g, 6 mL). Cartridges were activated and conditioned by elution with 10 mL of acetonitrile and deionized water sequentially. After the passage of the water samples under a vacuum at a flow rate of about 5 mL/min, the cartridges were dried and eluted. Analytes were then eluted twice with 6 mL ACN. The eluents were concentrated by rotatory evaporimeter, reduced to dryness under a gentle

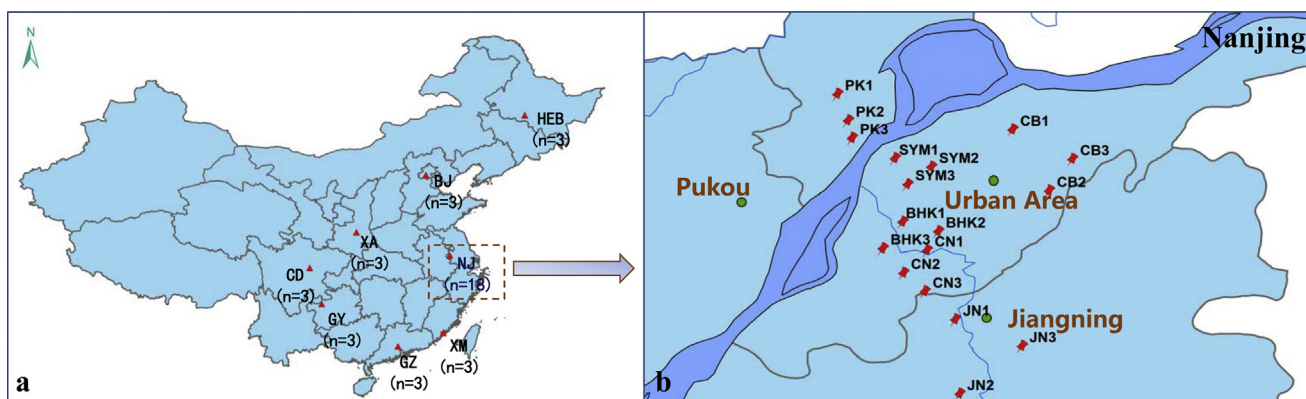


Fig. 1 – Sampling locations of tap water in China.

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