



# Occurrence and risk assessment of organophosphate esters in drinking water from Eastern China



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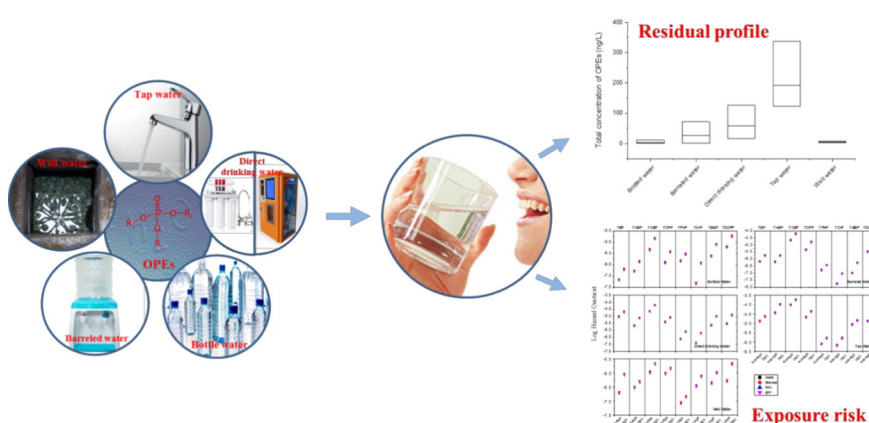
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## HIGHLIGHTS

- Residues of OPEs in drinking water were investigated in Eastern China.
- The concentrations of OPEs in tap water were higher than other drinking waters.
- Uptake of OPEs via drinking water was similar to airborne exposure for adults.
- Non-cancer and carcinogenic risks of OPEs for people via drinking water were low.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Organophosphate esters (OPEs) are ubiquitous in the environment and may pose potential health risks to humans. Drinking water is suspected as one possible exposure pathway of OPEs to humans. In this study, we investigated the residues of 9 OPEs in five types of drinking water in Eastern China. The median concentrations of  $\Sigma_9$ OPEs were determined to be 3.99, 4.50, 27.6, 59.2 and 192 ng/L in the bottled, well, barreled, direct drinking and tap waters, respectively. Triethyl phosphate (TEP) was the most abundant OPE in the tap water and filtered drinking water with median concentrations of 50.2 and 30.2 ng/L, respectively. The mixture of tri(chloropropyl) phosphate (TCPP) and tri(chloroisopropyl) phosphate (TCIPP), named here as TCPP, dominated in the barreled and well water with median concentrations of 8.04 and 2.49 ng/L, respectively. The calculated average daily doses of OPEs ranged from 0.14 to 7.07 ng/kg bw/day for people consuming the five different types of drinking water. Among the drinking water, the tap water exhibited the highest exposure doses of OPEs. The calculated non-cancer hazard quotients ( $10^{-4}$ – $10^{-7}$ ) from OPEs were much lower than the theoretical threshold of risk. The carcinogenic risks posed by TCEP were very

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low ( $<10^{-7}$ ) for all types of drinking water. The results revealed that there was currently low risk to human health from exposure to OPEs through drinking water in Eastern China.

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## 1. Introduction

Organophosphate esters (OPEs), also known as organophosphate flame retardants and plasticizers, are increasingly used since the restriction and phase-out of brominated flame retardants (BFRs), such as polybrominated diphenyl ethers (PBDEs) (USEPA, 2005). The market demand for OPEs in Europe was 93,000 tons in 2006, accounting for 20% of the total annual flame retardants (FR) consumption (van der Veen and de Boer, 2012). In China, the OPE production was more than 70,000 tons in 2007 and was estimated to increase 15% annually (Ou, 2011).

However, a growing number of toxicological studies have revealed that several OPEs may cause adverse effects in organisms. Chlorinated alkyl phosphates, such as tri(2-chloroethyl) phosphate (TCEP) and tri(1,3-dichloroisopropyl) phosphate (TDCIPP), were observed to cause tumor growth in different organs in rodents after long-term exposure and thus are suspect carcinogens (WHO, 1998). TCEP has been classified as a “potential human carcinogen” (carcinogen category 3) by the EU in 2008 (European Union, 2008). TDCIPP was added to California’s Proposition 65 List of Potential Carcinogens in 2011 (Stapleton et al., 2012). Moreover, TDCIPP was also identified to have neurotoxic properties in PC12 cells (Dishaw et al., 2011), and caused reproductive and developmental toxicity in zebrafish (Fu et al., 2013; Liu et al., 2013). In addition, several studies have suggested that OPEs may be associated with certain effects on human health, such as mucosal irritation and hormone level alteration (Kanazawa et al., 2010; Meeker and Stapleton, 2009). Even so, the current information is still limited regarding the toxicological effects of OPEs. To better understand their adverse effects, the U.S. Environmental Protection Agency (USEPA) is currently conducting full risk assessments on some OPEs (USEPA, 2013a).

In most cases, OPEs are used as additives and are not chemically bonded to materials, being thus able to diffuse easily into the environment (Marklund et al., 2003; Wensing et al., 2005). The occurrence of OPEs has been proven in various environmental matrices, including air (Saito et al., 2007; Salamova et al., 2014), dust (Van den Eede et al., 2011), water (Cristale et al., 2013a; Schaidler et al., 2014; Wang et al., 2014), sediment (Cristale et al., 2013b), and soil (Fries and Mihajlović, 2011). Furthermore, organisms may be exposed to OPEs through various pathways such as inhalation, ingestion and dermal contact (Wei et al., 2015). OPEs and their metabolites have been detected in wildlife (Greaves and Letcher, 2014) and human tissues (Sundkvist et al., 2010; Van den Eede et al., 2013).

Given the relatively high water-solubility of most OPEs, their occurrence in various water bodies, especially in drinking water, is an increasing issue of concern worldwide. The technologies currently employed in the production of drinking water are believed to have limited OPE removal capacity, since these substances have been detected in drinking water from the United States (Benotti et al., 2009), Spain (Rodil et al., 2012), South Korea (Kim et al., 2007), and China (Li et al., 2014). The consumption of drinking water may be an important pathway of OPE exposure for humans. However, the risk has not been well investigated to date.

In China, tap water is available in most urban areas. However, in some cases, people drink tap water after boiling to reduce the risk of microbiological contamination and improve the taste and smell of water. Additional devices with activated carbon filtration and/or membrane processes are often used to produce filtered drinking water from tap water in some households. In addition, commodity water (packed in bottles or barrels) is also popular, especially in the work environment. In rural areas, untreated well water (groundwater) is the main drinking

water source. A description of the various drinking waters is detailed in the Supplementary material (Appendix A).

In the study, we have collected five types of drinking water samples including tap water, filtered drinking water, bottled water, barreled water, and well water from both urban and rural areas in Eastern China. The residues and profiles of OPEs were analyzed in these water samples. A preliminary exposure risk was also estimated for the local residents by consumption of drinking water based on the measured concentrations of OPEs.

## 2. Materials and methods

### 2.1. Chemicals and reagents

Triethyl phosphate (TEP, 99.9%), tri-n-butyl phosphate (TnBP, 99.5%), tri-n-propyl phosphate (TnPP, 99.5%), TCEP (98.5%), tributoxylethyl phosphate (TBEP, 93.0%) were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Triphenyl phosphate (TPhP, 100%), TCPP (99.9%), TDCIPP (97.8%) were obtained from Accustandard (New Haven, CT, USA). Tricresyl phosphate (TCrP, 99%, mixture of isomers) was obtained from J & K Scientific (Beijing, China). Isotope-labeled standards tributyl phosphate (TBP-d27, 98–99%) and TPhP-d15 were purchased from Cambridge Isotope Laboratories (Andover, MA, USA) and Sigma-Aldrich (St. Louis, MO, USA), respectively.

Acetonitrile and methanol were LC/MS grade from J.T. Baker (Phillipsburg, NJ, USA) and Fisher Scientific (Fair Lawn, NJ, USA), respectively. Ultra-pure water (UPW, 18.2 M $\Omega$ ) was produced with a Milli-Q Gradient system (Millipore, Bedford, USA).

Individual stock solutions of analytes and isotope-labeled standards were prepared in acetonitrile and kept at 4 °C. Working standard solutions were prepared by mixing the stock solutions and diluting with methanol.

### 2.2. Sample collection

All water samples were collected in Hangzhou and Quzhou, Eastern China during April to July 2014. Tap water ( $n = 21$ ) was sampled in kitchens from 21 households in urban communities. In the same households or communities, filtered drinking water ( $n = 17$ ) which was produced from tap water by household filtering apparatuses (seven samples, H1–H7) or public purifying machines (ten samples, P1–P10) were also collected when available. Well water ( $n = 19$ ) was sampled from rural areas. Bottled water ( $n = 23$ ) and barreled water ( $n = 19$ ) was purchased from local markets. The bottled water was packaged in 550–1000 mL polyethylene terephthalate bottles. The barreled water was packaged in 18.9 L polycarbonate plastic barrels. Detailed description of the drinking waters is given in the Supplementary material (Appendix A). All samples were stored at 4 °C until analysis. The analysis was conducted within 48 h after sampling.

### 2.3. Analytical methods

One liter of water sample was spiked with 10 ng TBP-d27 as internal standard and extracted using solid phase extraction with cartridge (Oasis® HLB, 60 mg, 3 cc: Waters, USA) which was conditioned sequentially with 3 mL acetonitrile and 3 mL water prior to loading. The water sample was loaded on the cartridge at a flow rate of 4–5 mL/min. After loading, the cartridge was washed with 3 mL UPW and then dried under vacuum. Analytes were then eluted with  $2 \times 2$  mL acetonitrile at a flow rate of approximately 1 mL/min. The eluent was concentrated to near

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