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Occurrence, migration and health risks of fluorescent whitening agents and phthalates in bottled water

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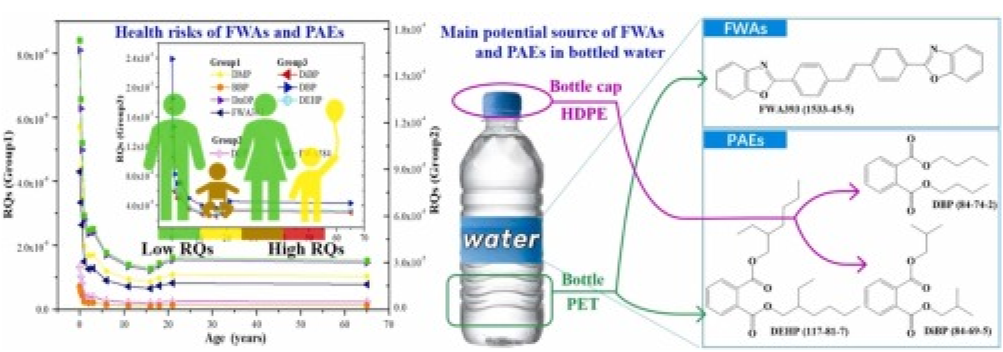
Highlights

- Fluorescent whitening agents (FWAs) were ubiquitous but at trace level (< 10 ng L^{-1}).
- Phthalates' concentrations (up to 1.2 μg L^{-1}) were much higher than those of FWAs.
- Plastic bottles and bottle caps were the main source of these plastic additives.
- Early life stages exhibited 3–5 times higher risk quotient values than adulthood.

Abstract

The occurrence and health risks of fluorescent whitening agents (FWAs) in bottled water were reported for the first time. FWA184 and FWA393 were the most frequently detected FWAs, with mean concentrations of 3.99–17.00 ng L^{-1}. Phthalates (PAEs) such as dibutyl phthalate (DBP), di-iso-butyl phthalate (DiBP), and diethylhexyl phthalate (DEHP) were prevalent in bottled water, with mean levels of 40.89–716.66 ng L^{-1}, and their concentrations in bottled water were much higher than those of FWAs. FWAs and PAEs in bottles and caps were extracted using organic solvent, and the correlation analysis showed that FWA393 and DEHP most likely originated from bottles, while bottle caps were the main sources of DBP and DiBP. The calculated risk quotients (RQs) of target substances and all age groups were considerably lower than the threshold of 0.1, indicating that consuming bottled water containing these plastic additives was unlikely to pose health risks for people of all ages. However, RQ values for underage people were several times higher than those for adults and hence cannot be neglected; therefore, special attention should be paid to understand the potential risks posed by the exposure to these plastic additives during early life stages, especially the infant stage.

Graphical Abstract



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Introduction

It is widely acknowledged by the public that the quality, taste, safety, and portability of bottled water are considerably better than those of tap water. Therefore, the global production and consumption of bottled water increasing every year [1], [2], [3], [4], [5], [6]. According to International Bottled Water Association statistics, the annual per capita consumption of bottled water has rapidly grown, increasing from 117.0 L in 2012 to 159.4 L in 2017. The top 10 countries with the highest bottled water consumption consumed 303 million tons of bottled water in 2017, accounting for 80.4% of the global bottled water consumption [7]. China has the highest bottled water consumption in the world, and 96.4 million tons of bottled water were consumed in China in 2017, accounting for 25.6% of the global bottled water consumption [7]. Bottled water has been an important part of the Chinese diet.

Plastics are widely used to make packaging containers of bottled water because they are cheap, durable, and recyclable [1]. Because of the growing popularity of bottled water, the occurrence and health hazards of chemical additives and microplastics in bottled water have caused concerns [8], [9], [10], [11], [12], [13]. However, chemical additives are indispensable in plastic production, for example, plasticizers are used to increase the strength, toughness, and plasticity of plastics [14], [15], [16], [17]. Among the many plasticizers commonly used during plastic production, phthalates (PAEs) are the most common and have the highest proportion [18], [19], [20]. More than 7.5 million tons of PAEs were globally consumed in 2017, accounting for ~65% of the global plasticizer consumption in the same year [21]. PAEs are combined with plastic polymers via simple physical mixing rather than strong chemical bonding [22]. Therefore, PAEs can easily migrate or discharge into environmental mediums during the manufacturing, storage, use, and disposal of plastic products [23], [24], [25], [26]. To date, PAEs have been detected in various environmental mediums, such as atmospheric particulate matters, garbage leachates, soil, surface water, and drinking water [27], [28], [29], [30], [31]. The concentration of PAEs in bottled water is reported to have reached as high as several hundred micrograms per liter [23]. PAEs are endocrine-disrupting chemicals and have estrogenic effects and certain biological toxicity [32]. This class of substances can be transferred to babies through diets, skin contact, and respiration, and can interfere with their endocrine system and affect development and growth, metabolism, and nervous and immune systems [33], [34]. Therefore, PAEs should be given sufficient attention even though they have low concentrations in bottled water and environmental mediums. Several PAEs have been listed as “priority pollutants” by China in 2023 [35].

Another type of plastic additive commonly added to plastics is fluorescent whitening agents (FWAs), also known as optical brighteners (OBs), but these materials have long been neglected by researchers. FWAs are widely used in many industrial production processes, such as the production of plastics, detergents, paper, textiles, and coatings [36], [37]. They are used to improve the whiteness, brightness, and brilliance of materials [36], [38]. FWAs can be classified into two broad categories, that is, ionic and nonionic compounds. Ionic FWAs are mainly used to whiten synthetic detergents, cotton fibers, and cotton fabrics, and FWA351 and FWA71 are the typical ionic FWAs used. Nonionic FWAs, such as FWA184 and

FWA393, are mainly used in plastic products and polyester fibers. FWAs pose a potential hazard to human and animal health, and exposure to high concentrations of FWAs can irritate the respiratory mucous membrane and skin and even cause irritant dermatitis [39], [40]. FWAs can easily combine with certain proteins in wounds and cause a certain degree of damage to blood and immune systems [41]. Additionally, FWAs with styrene or aromatic amine structures have a potential risk of cancer. Therefore, FWAs are attracting increasing attention from researchers in recent years. Zhou, et al. [42] studied four FWAs in 10 plastic children's toys and detected FWA367 and FWA184 in high frequencies (60% for both) and concentrations (the maximum concentration detected = 108.1 and 403.3 $mg\,kg^{-1}$, respectively) in plastic children's toys. Wu, et al. [43] measured seven FWAs in 40 commercial flours and reported that FWA184 was present in three samples in concentrations of 31–60 $\mu g\,kg^{-1}$. Furthermore, FWAs have been detected in cosmetics, facial masks, food contact paper products, plastic food contact materials, and medicinal composite films [44]. However, so far, there have been no reports on the occurrence and health hazards of FWAs in bottled water.

As a type of most widely used plastic additive, FWAs pose potential risks to human health. However, existing researches have not covered the pollution characteristics and behaviors of FWAs in aquatic environment, especially there are no studies focused on the occurrence and risk of FWAs in drinking water. In order to reveal the occurrence and health risks of FWAs in drinking water, we conducted the bottled water survey of FWAs and PAEs. The aims of this study were to (1) determine the concentrations and profiles of eleven selected FWAs (commonly used in plastic products) and 16 PAEs in bottled water by using solid-phase extraction coupled to liquid/gas chromatography–tandem mass spectrometry, (2) understand the migration characteristics of FWAs and PAEs released from plastic bottles into water under specific storage conditions, and (3) estimate the potential health risks of these FWAs and PAEs. The findings of this study will improve the understanding of FWAs in bottled water and will promote further researches on their toxicity as well as release and migration mechanisms in the near future. To the best of our knowledge, this is the first study on the occurrence and potential human health risks of FWAs in bottled water.

Section snippets

Chemicals and reagents

In total, 11 FWA standards, 16 PAE standards, chromatographically pure methanol, acetonitrile, hexane, chloroform, and mobile-phase additives were purchased from ANPEL

Laboratory Technologies (Shanghai) Inc. (Shanghai, China). Deionized water used in experiments was prepared using the Milli-Q IQ7000 ultrapure system from Millipore. 21 brands of bottled water were purchased from supermarkets and department stores in Shijiazhuang, Hebei, China in May 2022. The bottle volume varied from 330 mL to…

Occurrence of FWAs in bottled water

Different concentrations of FWAs were detected in bottled water samples (Fig. 1a). Among all detected FWAs, FWA184, FWA393, and OB-2 were the top three FWAs detected with high total concentrations in 21 samples, and their total concentrations were 254.99, 79.79, and 60.37 ng L^{-1}, respectively. The concentration of FWA184 in a single sample was as high as 143.81 ng L^{-1}. The total concentrations of remaining FWAs were well below or slightly higher than 1.0 ng L^{-1}. Among 21 bottled water samples,…

Conclusions

Plastic additives are indispensable in plastic production, including plastic water bottles. However, FWAs, another type of commonly used plastic additives have long been neglected by most researchers, therefore, this study reports, for the first time, the occurrence and potential human health risks of FWAs in bottled water. The commonly used FWAs and PAEs in plastic products were detected in high levels or frequencies. PAEs' concentrations in bottled water were much higher than those of FWAs.…

Environmental implication

As a class of important plastic additives, fluorescent whitening agents (FWAs) and phthalates (PAEs) are indispensable in plastics like plastic water bottles. FWAs exhibit high persistence, bioaccumulation, and toxicity, while PAEs are endocrine-disrupting chemicals and have estrogenic effects and certain biological toxicity. These substances could easily migrate into bottled water from plastic container, and then threaten human health through drinking bottled water.

FWAs have long been…

CRediT authorship contribution statement

Bo Yao: Writing – review & editing, Investigation, Funding acquisition, Formal analysis, Data curation. **Shuwen Yan:** Visualization, Methodology, Formal analysis. **Yonghui Han:**

Investigation, Conceptualization. **Jiansheng Cui:** Validation, Supervision, Project administration. **Ruiyao Guo:** Methodology, Investigation, Formal analysis. **Xiaoge Liang:** Methodology, Formal analysis. **Mengfei Su:** Methodology, Formal analysis.…

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

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