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Persistence, temporal and spatial profiles of ultraviolet absorbents and phenolic personal care products in riverine and estuarine sediment of the Pearl River catchment, China

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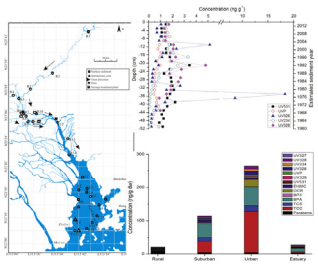
HIGHLIGHTS

- The PCPs were widely present in the sediment.
- UV stabilizers and TCC were fairly persistent in sediment.
- PCPs' temporal trend mirrored industrial developments and waste treatment.
- No significant in-situ TCC dechlorination in the sediment.

GRAPHICAL ABSTRACT



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ABSTRACT

A variety of personal care products have been classified as emerging contaminants (ECs). Occurrence, fate, spatial and vertical profiles of 13 ultraviolet absorbents, triclocarban (TCC) and its dechlorinated products, triclosan (TCS), 2-phenylphenol and parabens were investigated in riverine and estuarine sediment of the Pearl River catchment, China. Bisphenol A (BPA), a widely applied plasticizer, was also investigated. The ECs were widely present in the bed sediment. TCC was the most abundant with a maximum concentration of 332 ng g^{-1} dry weight. The other prominent ECs included BPA, TCS, octocrylene, and benzotriazole UV stabilizers UV326 and UV328. Treated wastewater effluent was the major source of the ECs in the riverine sediment. TCC, BPA, TCS, methyparaben, UV531, UV326, and UV328 were also detected throughout the estuarine sediment cores, indicating their persistence in the sediment. Temporal trends of the ECs in the sediment cores reflected a combined effect of industrial development, population growth, human life quality improvement, and waste treatment capacity in the Pearl River Delta over the last decades. TCC dechlorination products were frequently detected in the bed sediment with higher levels near treated effluent outlets but only occasionally observed in the sediment cores, suggesting insignificant in-situ TCC dechlorination in the sediment.

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Table 1
The investigated emerging contaminants.

Class	Compound/abbreviation	CAS No	$LogK_{ow}$
preservative	methylparaben/MP	99–76-3	1.96
preservative	ethylparaben/EP	120–47-8	2.47
preservative	propylparaben/PP	94–13-3	3.04
preservative	butylparaben/BP	94–26-8	3.57
disinfectant	2-phenylphenol/PHP	90–43-7	3.09
disinfectant	triclosan/TCS	3380–34-5	4.76
disinfectant	triclocarban/TCC	101–20-2	4.9
plasticizer	bisphenol A/BPA	80–05-7	3.32
metabolite	nonchlorinated carbanilide/NCC	102–07-8	–[a]
metabolite	dichlorocarbanilide/DCC	1219–99-4	–[a]
UV filter	2-hydroxy- 4-methoxybenzophenone/BP3	131–57-7	3.52
UV filter	3-(4-methylbenzylidene)camphor/4MBC	36861–47-9	5.47
UV filter	octocrylene/OCR	6197–30-4	6.88
UV filter	octyldimethylparaaminobenzoate/ODPABA	21245–02-3	5.77
UV filter	avobenzone/AVO	70356–09-1	2.41
UV filter	2-ethylhexyl 4-methoxycinnamate/EHMC	5466–77-3	5.80
UV stabilizer/plasticizer	UV531	1843–05-6	6.42
UV stabilizer	UVP	2440–22-4	3.00
UV stabilizer	UV329	3147–75-9	6.21
UV stabilizer	UV326	3896–11-5	5.52
UV stabilizer	UV234	70321–86-7	7.67
UV stabilizer	UV328	25973–55-1	7.22
UV stabilizer	UV327	3864–99-1	6.75

[a] no report.

1. Introduction

Personal care products (PCPs), such as sunscreens and lotions, are produced to improve quality of human lives. Production and consumption of PCPs keep increasing due to growing population and people's pursuit for better quality of life. Bisphenol A (BPA) is widely applied as a stabilizing agent in plastics and as an additive in thermal paper and paper coatings. A variety of PCPs as well as BPA may find their way to the environment after consumption and have been listed as emerging contaminants [1]. Among the big family of emerging contaminants, chemicals such as BPA, preservatives (e.g. parabens), antimicrobials (e.g. triclosan and triclocarban), and various organic ultraviolet absorbents (UVAs) have caused particular concerns due to their confirmed or suspected endocrine disrupting impacts on organisms, posing threats to the health of ecological systems [2–7]. In addition, their general toxic effects on non-target organisms were also reported [4].

Many of these emerging contaminants are high-production chemicals. For example, BPA has an annual production of over 4.6 million tons globally [8,9]. Triclosan (TCS) and triclocarban (TCC) are widely used as disinfectants in such products as soaps, shampoos, body lotions, toothpastes, paints, and plastics at levels of up to 2% and 0.3% (w/w), respectively [10]. Parabens are preservatives added in cosmetics, pharmaceuticals, and foodstuffs [11,12]. UVAs are widely applied in sunscreens, cosmetics, etc., comprising up to 20% of mass weight for protecting humans from harmful UV radiation. They are also added in a variety of industrial products, such as textiles and paints, to protect the products from yellowing [13]. Significant amounts of these emerging contaminants are being discharged into the environment via treated and untreated wastewater, recreational activities, and other human activities. As a result, they have been widely detected in rivers, lakes, seas, groundwater, and organism tissues [9,14–27].

Sediment can be a significant sink for these emerging contaminants in the environment considering their moderate to high lipophilicity indicated by octanol-water partition coefficients ($\log K_{ow}$) between 1.96-7.67 (Table 1). Available research revealed the presence of TCC, TCS, BPA, parabens, and several UVAs in sediment [9,15,23,28–31]. Some of these compounds (e.g. TCC) were found to be fairly persistent [7,24,32]. Accumulation of these emerging contaminants in sediment may pose risks to wildlife, especially benthic and sediment-dwelling organisms because these chemicals can be bio-accumulative in organisms and even be biotransferable in food chains [4,17,33,34].

Pearl River Delta (PRD) is one of the most industrialized areas in China with a population of more than 50 million. The annual wastewater production has been about 9 billion m^3, of which about 7 and 2 billion m^3 are domestic wastewater and industrial wastewater, respectively, in recent years. All the industrial wastewater and about 90% of the domestic wastewater are treated before discharge. The treated and untreated wastewater is finally discharged into the Pearl River and its tributaries. Furthermore, located in the south end of the subtropical zone, the PRD has warm and humid weather with annual sunshine of 1600–2000 h [35]. Products containing disinfectants, preservatives, and UVAs are therefore expected to be consumed in large quantities. Previous research has revealed high concentrations of TCS, TCC, parabens, and BPA in wastewater and river water of the PRD [36–38]. However, only few work studied some phenolic endocrine disrupting emerging contaminants (e.g., TCC, TCS, and BPA) in sediment in the Pearl River catchment [36,38]. Furthermore, occurrence and fate of the UV absorbents in the sediment in this area have not been revealed yet.

In this context, the objectives of this study are to (1) illustrate occurrence and spatial distribution of the emerging contaminants and their dechlorination products in bed sediment of the Pearl River and Pearl River Estuary (PRE), (2) delineate sedimentary profiles of the emerging contaminants in the PRE, and (3) investigate mechanisms of accumulation of these emerging contaminants and their fate in the sediment of the Pearl River and the PRE. The 21 emerging contaminants in this work belonged to commonly used disinfectants, preservatives, ultraviolet filters, ultraviolet stabilizers, and plasticizer as detailed in Table 1. Two dechlorination products of TCC were also included to demonstrate fate of TCC in the sediment. To the best of our knowledge, the benzophenone UV stabilizer UV531 in the environment was not reported so far. This is also the first work to reveal dechlorination products of TCC in the environment of China. Fate of TCC was consequently discussed, such as when and where the dechlorination happened. Furthermore, records of the UV absorbents in sedimentary cores were delineated to reveal environmental persistence of these compounds, which has been rarely reported. This work provided insight into distribution, temporal trends, and fate of the emerging contaminants in the

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