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Occurrence, migration, and risk assessment of PPCPs in water bodies and sediments of river-type drinking water sources in eastern China

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ABSTRACT

Pharmaceuticals and personal care products (PPCPs) are receiving attention as emerging pollutants due to their extensive applications and persistent emissions. The Qiantang River Basin, a representative region in eastern China that relies on surface water for drinking purposes, experiences the movement and accumulation of PPCPs in its water and sediment, which can directly affect the safety of drinking water in the basin. This study focuses on the Qiantang River Basin's surface water, sediment, and drinking water to determine the occurrence and potential risks of 31 PPCPs. It aims to address whether PPCPs in the environment could migrate and accumulate, thereby affecting human health. The findings indicated that PPCPs are ubiquitous in various environmental media, with surface and pore water showing distinct spatial distribution characteristics, specifically, concentrations escalated with urban scale expansion, indicating that domestic sewage discharge is the primary source of PPCP input. Bisphenol A (BPA), ketoprofen (KTP), and diethyltoluamide (DEET) were the primary contaminants. The movement of PPCPs within the surface water-sediment-pore water system was affected by various circumstances. Substances like Sulfamethoxazole (SMX) (RQ>10^3) and KTP (RQ=22.3) present in surface water and sediment pose significant ecological concerns, and KTP and atrazine (ATZ) (0.6<RQ<2.1) also pose a high risk in drinking water. This study performed an extensive analysis of the distribution and risks associated with typical PPCPs in the Qiantang River Basin, offering a scientific foundation and theoretical support for research on the environmental behavior of PPCPs in this area and the development of targeted pollution control strategies.

1. Introduction

Pharmaceuticals and Personal Care Products (PPCPs) are emerging organic pollutants that predominantly encompass human and veterinary antibiotics, hormones, anticonvulsants, lipid-lowering medications, and sunscreens (Liu JL et al., 2013; Zhang ZH et al., 2023). In contrast to traditional organic pollutants, PPCPs have relatively short half-lives and are readily degradable (Lin K et al., 2023). However, due to their widespread use globally and the presence of their metabolites and transformation products, PPCPs exhibit a phenomenon known as "pseudo-persistence" in the environment (Hernando MD et al., 2006).

Surface water serves as a primary receptor for the environmental release of PPCPs. PPCPs that enter aquatic systems are conveyed by water flow to drinking water sources and may persist in drinking water even after treatment, subsequently entering the human body through direct ingestion, dermal contact, and the consumption of fish and other aquatic organisms (Subramanian A et al., 2023); the remainder associated with suspended particles in water bodies and accumulates in sediments. These PPCPs will be resuspended and reintroduced into the water upon disturbance of sediments. The environmental hazards of PPCPs to surface and groundwater mostly rely on their migratory interactions within the water-sediment system (Tian YJ et al., 2023).

In China, 144 types of PPCPs have been identified in the water bodies and sediments (Liu N et al., 2015). PPCPs at

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trace concentrations (ng/L–μg/L) may drive antibiotic resistance gene proliferation through chronic exposure across water, sediment, and soil matrices. Furthermore, certain PPCPs are known for their bioaccumulation properties and have been extensively detected in aquatic organisms (Tanoue R et al., 2020; Nozaki K et al., 2023; Xu XM et al., 2023) as well as in crops (Wu XQ et al., 2012, 2013; Dodgen LK et al., 2013). This poses a potential risk to both ecological systems and human health. Recent studies suggest that while the likelihood of acute toxicity from PPCPs is relatively low, the risk of chronic toxicity is a substantial concern. Extended exposure to low concentrations of PPCPs can lead to the expression of antibiotic-resistance genes and potentially produce endocrine disruptors (Guo ZN et al., 2022; Liu JL, 2013), which may adversely affect reproduction and development.

The Qiantang River Basin is a quintessential surface water drinking water source area, including the Xin'anjiang Reservoir and river water sources along the mainstream. This basin supplies drinking water to Hangzhou and parts of Shanghai while receiving industrial, agricultural, and domestic wastewater from the surrounding urban areas. Consequently, the issue of water contamination is intricately linked to human activities and lifestyles. This study seeks to systematically analyze the migration patterns of PPCPs at the water-sediment-pore water-drinking water interface in the Qiantang River basin and to identify the main contaminated areas and their sources to elucidate the effects of PPCP pollution on human life.

2. Materials and methods

2.1. Reagents and chemicals

Budgetary constraints necessitated a focus on thirty-one PPCPs, which were analyzed, comprising 11 antibiotics, 17 non-antibiotic pharmaceuticals, and three personal care products (PCPs) (Table S1). Reference standards (purity >94.0%) were obtained from Dr. Ehrenstorfer and AccuStandard, with isotope-labelled internal standards (SMX-D_4, CIP-^{13}C, IBU-^{13}C, ERY-^{13}C-D_3) sourced from Wako, Japan. Stock solutions (100 mg/L) were prepared in methanol and stored at −18°C in the dark. HPLC-grade solvents (Merck, Germany), formic acid, ammonium acetate, Na_2-EDTA (Fluka, USA), and Milli-Q water comprised the analytical reagents.

2.2. Sample collection and pretreatment

Sample collection in this study was chosen to occur during the summer months to obtain maximum pollutant input when surface runoff is greatest. This sampling approach was implemented along the Qiantang River system, extending from Qiandao Lake to the Haining estuary, as illustrated in Fig. 1. To address anthropogenic PPCP inputs, strategic sampling sites were selected near major urban centers (Chun'an, Jiande, Tonglu, Fuyang, Hangzhou, Haining) and key discharge zones, including wastewater treatment plants (WWTPs), industrial complexes, and livestock facilities. Surface and sediment samples were collected from riverine and lacustrine drinking water sources supplying municipal-

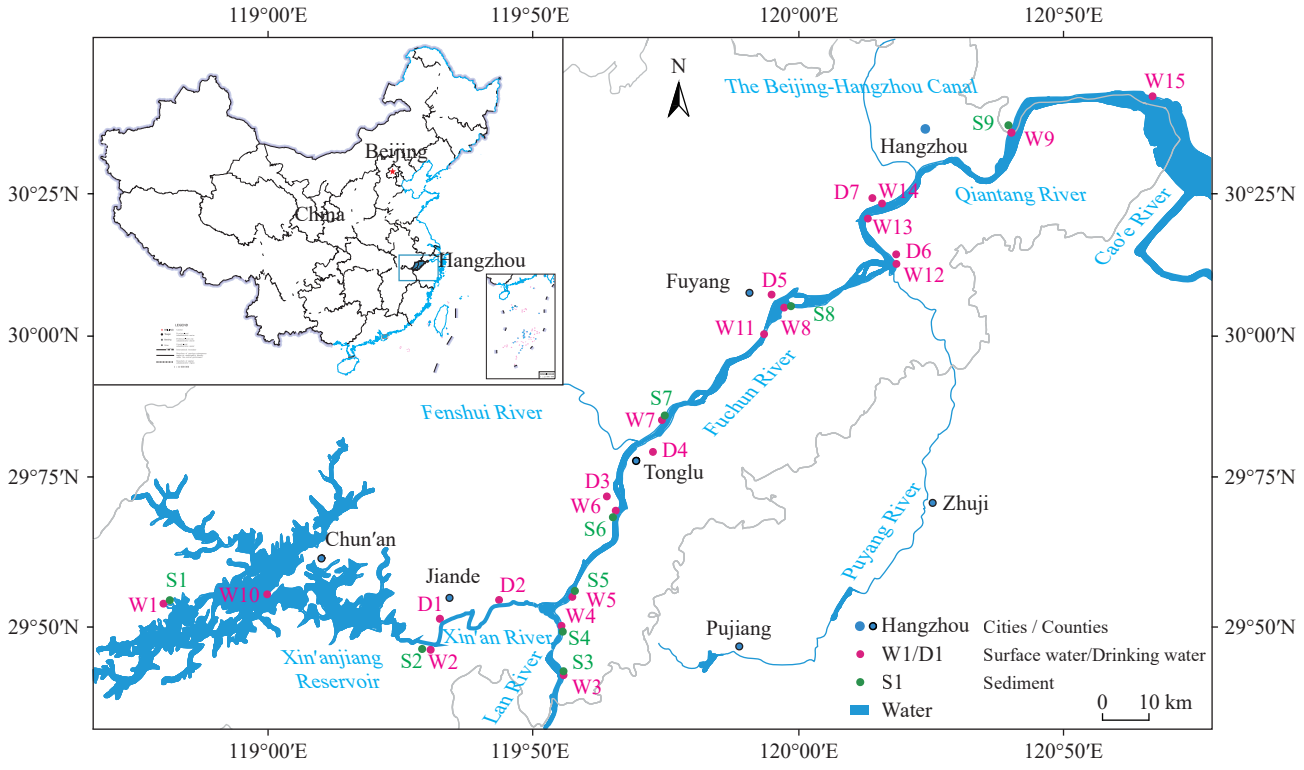

Fig. 1. Sampling sites in the Qiantang River Basin.

level populations.

Surface water was collected using stainless steel buckets pre-rinsed with sampled water and then transferred to pre-cleaned polypropylene bottles (250 mL, 500 mL, or 1 L). Drinking water samples were directly collected in polypropylene bottles. Sediment samples were retrieved with a dredge bucket, surface debris removed, and stored in sealed polyethylene bags. All samples were immediately wrapped in aluminium foil, refrigerated to inhibit microbial activity, and transported to the laboratory for analysis. Samples underwent SPE pretreatment. Surface water was filtered through filters to remove suspended solids and plankton. Pore water was obtained by high-speed centrifugation of sediment samples. The Oasis HLB cartridges (500 mg, 6 mL, Waters) were sequentially activated with 5 mL methanol containing 0.1% ammonia, 5 mL methanol, and 5 mL Milli-Q water. Following activation, a polypropylene reservoir was attached to the column's upper port to regulate sample flow at 5–10 mL/min. Post-sample loading, the column underwent centrifugation (4000×g for 15 min), followed by sequential rinsing with 10 mL and 5 mL methanol. The target analytes were eluted with 15 mL of methanol, dried under a gentle nitrogen stream, vortex-mixed, and transferred (200 μL aliquots) to injection vials for analysis.

Freeze-dried sediments were homogenized and transferred to 50 mL centrifuge tubes. Each sample underwent three sequential 3 mL aliquots of methanol and acetonitrile, with 30 s vortex-mixing followed by 20 min sonication. After centrifugation (4000×g, 10 min), supernatants were pooled in new tubes. The extract was diluted to 250 mL with Milli-Q water and then processed identically to water samples via solid-phase extraction, elution, and nitrogen evaporation.

2.3. Instrumental analysis

PPCPs were analyzed using an Agilent 1200 series HPLC coupled to an Applied Biosystems API 4000 triple quadrupole mass spectrometer with electrospray ionization (ESI). Both positive (ESI+) and negative (ESI−) ionization modes were employed to accommodate the diverse physicochemical properties of the target compounds. Separation utilized an XBridge C18 column (4.6×150 mm, 3.5 μm, Waters) with a guard column, maintained at 40°C.

The ESI+ mobile phase comprised (A) 2 mM ammonium acetate/0.1% formic acid in water and (B) methanol: Acetonitrile (1∶1) at 400 μL/min. A 10 μL injection volume was subjected to the following gradient: Initial 90% A held for 0.5 min, ramped to 95% B over 13 min, maintained for 7 min, then re-equilibrated to initial conditions for 7 min. For ESI− analysis, the mobile phase consisted of (A) 2 mM ammonium acetate in water and (B) a 1∶1 methanol-acetonitrile mixture, delivered at 400 μL/min with a 10 μL injection volume and column temperature maintained at 40°C. The gradient program was initiated at 5% B for 1 min, increased linearly to 95% B over 12 min, held for 8 min, and re-equilibrated to initial conditions for 7 min.

Mass spectrometry parameters were optimized separately for ESI+ and ESI− modes: In ESI+, nebulizing gas (GS1) pressure was 241 kPa, auxiliary gas (GS2) 103 kPa, spray voltage 5.0 kV, and ion source temperature 550°C; in ESI−, GS1 pressure was 276 kPa, GS2 138 kPa, spray voltage −4.5 kV, and ion source temperature 450°C. Both modes utilized 34.5 kPa collision gas, 69 kPa curtain gas, and multiple reaction monitoring, with compound-specific transitions detailed in Table S1.

2.4. Quality assurance and control

Quality assurance protocols included processing procedural blanks and blank spikes with every eighth sample to monitor background interference, with spike solutions prepared by adding 50×10^{-9}–100×10^{-9} target analytes to 250 mL Milli-Q water. Method validation utilized nine-point calibration curves (1−500 μg/L) incorporating isotope-labeled internal standards for recovery assessment, while quantification relied on external standardization. Accuracy was verified through spike-recovery tests in both ambient and laboratory water matrices. Method detection limits were established as the lowest concentrations producing signal-to-noise ratios ⩾3 with chromatographic peaks distinguishable from blanks, with complete sensitivity data provided in Table S1.

2.5. Risk assessment methods

Ecological and human health risks posed by PPCPs in the Qiantang River Basin were assessed using risk quotients (RQs). For ecological evaluation, measured environmental concentrations were compared to predicted no-effect concentrations (PNEC) derived from chronic toxicity data of aquatic organisms.

$$RQ = MEC/PNEC_w \quad (1)$$

$$PNEC_w = EC_{50}/AF or PNEC_w = NOEC/AF \quad (2)$$

where the MEC values were mean concentrations of PPCPs, PNEC values for surface water ($PNEC_w$) were derived by dividing acute (EC_{50}) or chronic (NOEC) toxicity data obtained from USEPA ecotoxicological databases by assessment factors (AF) of 1000 or 100, respectively.

Sediment $PNEC_s$ were estimated through equilibrium partitioning using $PNEC_w$ values adjusted by sediment-water partition coefficients (K_d), calculated from organic carbon content (f_{oc} = 0.03 g/g) and octanol-water distribution coefficients as referenced in established methodologies.

$$PNECs = K_d \times PNEC_w \quad (3)$$

$$K_d = K_{oc} \times f_{oc} \quad (4)$$

$$\lg K_{oc} = 0.623 \lg K_{ow} + 0.873 \quad (5)$$

The health RQ was calculated by dividing the maximum concentration of each PPCP by the drinking water equivalent level (DWEL). Where ADI stands for acceptable daily intake (μg/kg·day), BW stands for the 50th percentile of body weight, and HQ is the hazard quotient (HQ), which is assumed to be 1; DWI is the intake of tap water (liters/day), AB is the gastrointestinal absorption rate, which is assumed to be 1; and FOE is the frequency of exposure (350 days /365 days=0.96).

$$RQ = C_s/DWEL \quad (6)$$

$$DWEL = (ADI \times BW \times HQ)/(DWI \times AB \times FOE) \quad (7)$$

The assessed ecological and human health risks were categorized into four categories: RQ < 0.01 (no risk), 0.01 < RQ < 0.1 (low risk), 0.1 < RQ < 1 (medium risk), and RQ > 1 (high risk).

3. Results and discussion

3.1. Occurrence and distribution characteristics of PPCPs in water and sediments

3.1.1. Surface water

A total of 18 PPCPs were identified in 15 surface water samples collected from the river basin. The detected concentrations were as follows: Metoprolol (MPL) ranged from 0.12 μg/L to 3.46 μg/L, atrazine (ATZ) from 2.13 μg/L to 23.29 μg/L, diethyltoluamide (DEET) from 2.46 μg/L to 54.89 μg/L, ketoprofen (KTP) from not detected (ND) to 1381.62 μg/L, sulfamethoxazole (SMX) from ND to 27.94 μg/L, bisphenol A (BPA) to 77.76 μg/L, and thiamphenicol (TAP) from ND to 10.26 μg/L. Notably, these compounds exhibited detection rates greater than 50%.

Non-steroidal anti-inflammatory drugs (NSAIDs) were the primary substances detected in surface water in relatively high concentrations. KTP constituted the majority of the total PPCPs, with a mean concentration of 363.36 μg/L, significantly surpassing levels reported in water sources in Pakistan (147 ng/L) (Khan HK et al., 2022), surface water in South Africa (390–437 ng/L) (Agunbiade FO et al., 2016), and surface water in the seven principal river basins in China (1.52–24.15 ng/L) (Xu MJ et al., 2019). KTP is widely employed as an antipyretic and analgesic because of its negligible side effects, low toxicity, and potent therapeutic efficacy (Mendes FS et al., 2023). A peak concentration of KTP (1381.62 μg/L) was observed downstream of the Haining Dingqiao WWTP effluent, strongly suggesting inadequate pharmaceutical removal during treatment (Jiang XS et al., 2019).

MPL was detected in all surface water samples. This compound is widely prescribed as a β-receptor antagonist in developed countries (Wilde ML et al., 2014), and its prevalence in aquatic ecosystems is typically elevated (Hawash HB et al., 2023). The concentration of iopromide (IPM), as an X-ray contrast agent, is frequently associated with the scale and density of urban hospitals (Heberer T, 2002). This study found IPM exclusively at sample locations adjacent to the metropolitan region of Hangzhou (21.62 μg/L), surpassing levels recorded in Japan (ND–137 ng/L) (Azuma T et al., 2019) and Serbia (75.2 ng/L) (Petrović M et al., 2014). The detection rates of carbamazepine (CBZ), atorvastatin (ATO), and benzafibrate (BZF) were all below 10%. The concentration of antibiotics (ND–27.94 μg/L) in this study varied along the route and correlated with the population of towns and cities, particularly sulfonamide antibiotics (SAs). Prior research has similarly identified SAs at a significant prevalence (Bu QW et al., 2013). As a human and animal-shared antibiotic, SAs are widely used in livestock and aquaculture. Due to their high solubility and chemical stability in water (Tang XY et al., 2021), they have strong anti-degradation ability and can be transported over long distances in water bodies, often dominating aquatic environments (Shi JY et al., 2022). The primary hormone was 17-α-ethinylestradiol (EE2), a derivative of the natural estradiol, extensively utilized in human medicine, livestock, and aquaculture (Aris AZ et al., 2014). Because conventional sewage treatment methods are ineffective in removing EE2 (Ren HY et al., 2006), significant concentrations were observed at sampling locations downstream of the Hangzhou Qige WWTP.

The widespread use of PCPs has made them prevalent in surface water, with concentrations varying from 18.66 μg/L to 144.24 μg/L. These levels increase with urban size and population density along the river basin, indicating a direct link between residual PCPs in water and the discharge of domestic sewage. DEET is resistant to natural degradation or transformation processes (Sun J et al., 2015) and is frequently identified. ATZ, a widely utilized herbicide, typically infiltrates aquatic systems via diffuse agricultural runoff with routine application (De CV et al., 2023). In this study, the concentration of ATZ in surface water ranged from 2.13μg/L to 23.29 μg/L, significantly higher than the levels observed in surface water in Saudi Arabia (2.2 ng/L) (Ali AM et al., 2017) and Brazil (5.2–516 ng/L) (Acayaba RD et al., 2021). Furthermore, its concentration variation exhibited a weak correlation with the size of cities along the river basin and was minimally influenced by domestic sewage. BPA is primarily utilized to manufacture polycarbonate, epoxy resin, and other polymeric materials (Shen J et al., 2020), with its concentration typically elevated in industrial regions (Yao WR et al., 2024).

The spatial distribution of PPCPs in surface waters exhibited a pronounced downstream accumulation, with concentrations increasing from 40.62 μg/L in headwater regions to 1548.99 μg/L in urban estuaries (Fig. 2). This longitudinal gradient reflects two principal mechanisms: (1) Cumulative basin-wide nonpoint inputs and (2) concentrated urban point-source discharges. While only marginal concentration increases were observed between Jiande and Fuyang sampling stations, the Hangzhou metropolitan area demonstrated a 38-fold escalation in PPCP loading, directly

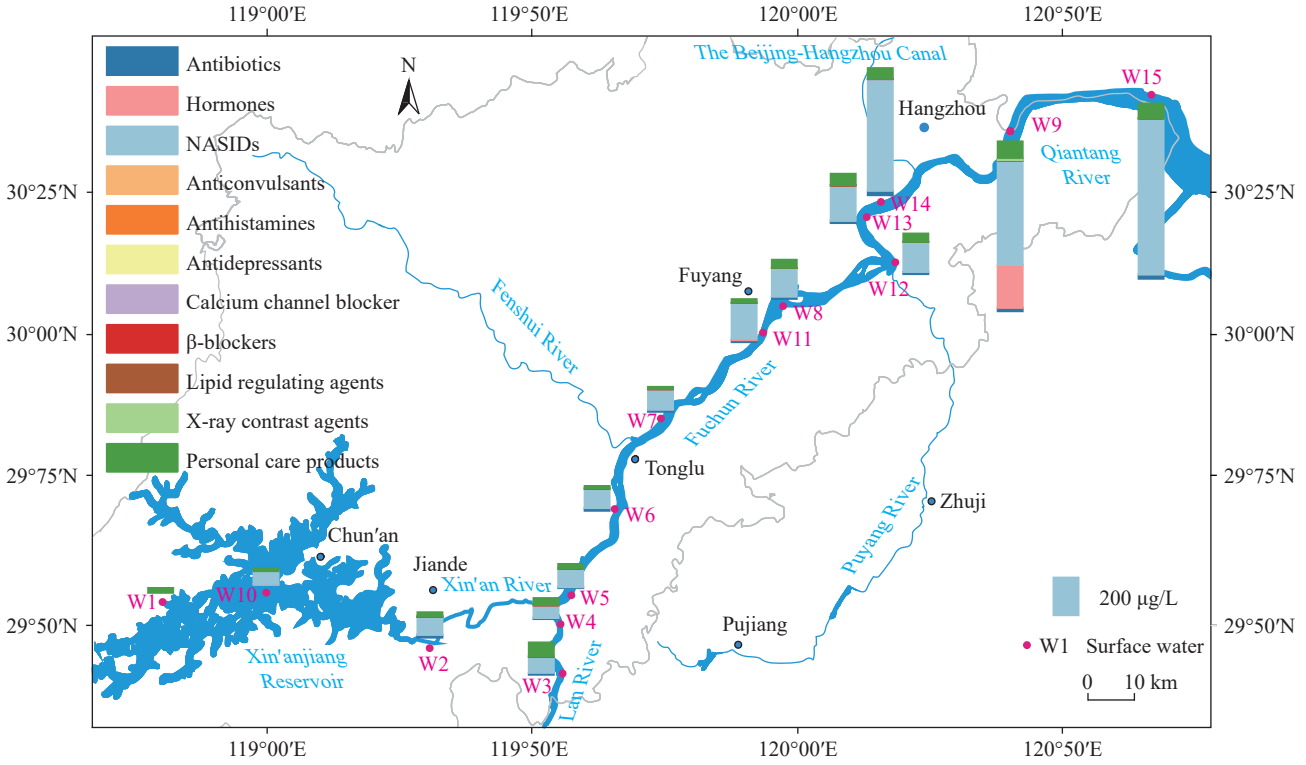

Fig. 2. Occurrence and distribution of PPCPs in the surface water of Qiantang River Basin.

correlating with population density. The downstream reaches of the Qiantang River and urban areas exhibited significantly greater PPCP diversity than upstream regions. EE2 and IPM were exclusively detected in downstream samples, providing clear chemical evidence of medical and domestic wastewater inputs.

3.1.2. Drinking water

PPCPs entering drinking water sources cannot be completely removed after drinking water treatment (Leung HW et al., 2013; Song Z et al., 2019; Meng T et al., 2021). While certain processes exhibit selective removal capabilities (Wang HZ et al., 2016), the inherent complexity of the water matrix often diminishes the overall efficiency of PPCP removal (Wu QH et al., 2012). As a result, numerous PPCPs commonly persist in tap water (Ben YJ et al., 2020; Feng L et al., 2020; He P et al., 2022). In the Hangzhou area, surface water from the Qiantang River is typically designated as the primary drinking water source. This study focused on sampling drinking water from key municipalities within the Qiantang River Basin as the main analytical targets.

Five PPCPs were detected in seven drinking water samples, with concentrations ranging from 46.14 μg/L to 161.08 μg/L. KTP, ATZ, and DEET were present in over 85% of the samples. The average concentration of KTP was 56.06 μg/L, which was higher than that in the Yangtze River Basin (29.65 ng/L) (Jiang XS et al., 2019) and Poland (28.1 ng/L) (Kot-Wasik A et al., 2016). The average concentration of ATZ was 34.37 μg/L, exceeding the limit set by China's drinking water hygiene standards threshold (2 μg/L). Traditional drinking water treatment processes usually struggle to eliminate ATZ from tap water efficiently (Wang R et al., 2023). Research has indicated that ATZ poses a significant threat to the aquatic ecosystems in the lower segments of the Yangtze River (Su C et al., 2020). Consequently, further extensive research on ATZ residues in potable water should be undertaken. DEET is resistant to natural degradation and has persistence in drinking water (Sun J et al., 2015). This investigation identified a DEET content of 14.46 μg/L, exceeding the residual levels found in drinking water in Guangzhou (844 ng/L) (Wang YQ et al., 2022) and the United States (63 ng/L) (Benotti MJ et al., 2009).

The concentration of PPCPs in treated drinking water is significantly lower than in untreated raw water, with removal efficiencies primarily influenced by the drinking water treatment process, initial concentration, and the physicochemical properties of the compounds. Substances frequently identified in raw water, such as KTP and ATZ, were typically found in treated drinking water. Certain research indicates that the treatment process influences the levels of PPCPs persisting in drinking water more than the source water quality (Liu M et al., 2019). Traditional drinking water treatment methods can efficiently eliminate SMX (Couto CF et al., 2019), with MPL removal rates reaching up to 85% (Padhye LP et al., 2014). Consequently, these two chemicals were undetected in potable water. New disinfection by-products in drinking water may emerge following the treatment of contaminated raw water (Mukhopadhyay A et al., 2022).

The consistently low spatial variability of residual PPCP concentrations in treated drinking water (Fig. 3) contrasts sharply with the pronounced urban-rural gradients observed in

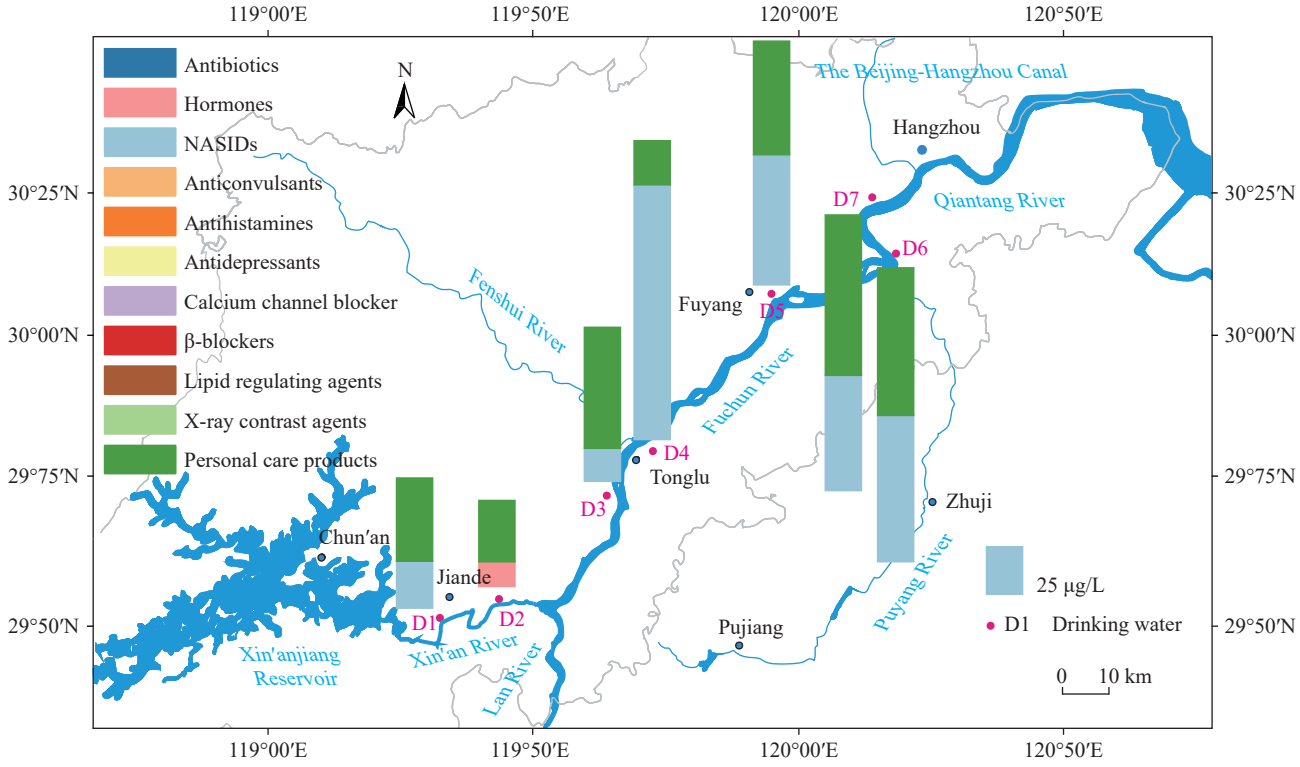

Fig. 3. Occurrence and distribution of PPCPs in the drinking water of Qiantang River Basin.

raw water. This disparity suggests that municipal drinking water treatment plants (DWTPs) achieve more uniform contaminant removal efficiencies than suburban facilities. The findings are consistent with previous reports from Shanghai's water systems (Liu M et al., 2019), confirming that advanced treatment processes in urban DWTPs effectively reduce location-dependent differences in PPCP contamination.

3.1.3. Sediments and pore water

Nine PPCPs were identified throughout the nine sediment sampling locations, with KTP, DEET, and BPA showing 100% detection frequency. Estrone (E1) and MPL were detected at >85% of sites. KTP concentrations (40.86 ng/g) exceeded those reported in the Han River (34.76 ng/g) (Gao Y et al., 2018) and Ebro River Basin (0.5−8.7 ng/g; da Silva BF et al., 2011). DEET levels (0.18−1.39 ng/g) were lower than in the Great Lakes (ND−8.2 ng/g) (Blair BD et al., 2013). SMX and clarithromycin (CLA) dominated antibiotics (ND−2.19 ng/g). PPCP concentrations in sediments showed minimal spatial variation between upstream and downstream regions (Fig. 4), indicating complex aqueous-solid phase partitioning influenced by compound properties and environmental conditions. The highest concentrations occurred at site S9 downstream of Hangzhou Qige WWTP, where wastewater effluent contributed substantial PPCPs loading despite rapid flow and minimal particle content.

Pore water represents a critical interface between surface water and sediments, potentially releasing PPCPs into surface water under environmental disturbances. Analysis of centrifuged pore water samples detected 10 PPCPs, with KTP, MPL, DEET, and BPA showing the highest detection frequencies. Concentrations ranged from 510.3 μg/L to 6068.3 μg/L, demonstrating a clear upstream-downstream increasing gradient that suggests more direct PPCPs exposure in pore water than in sediments.

3.2. Migration and enrichment of PPCPs in the surface water-sediment-pore water system

PPCPs enter sediments through adsorption and sedimentation with particulate matter, with subsequent desorption into pore water influenced by sediment organic content, pH, and redox conditions. These compounds may return to surface water via diffusion or resuspension. While PPCPs partitioning typically correlates with octanol-water coefficients (K_{ow}), this study found no significant linear relationship between sediment concentrations and logK_{ow} values for the nine detected PPCPs (Fig. 5). This indicates that K_{ow} alone cannot fully explain PPCPs' behavior in the water-sediment-pore water system.

Correlation analysis of PPCPs across surface water, pore water, and sediments (Fig. 6) revealed that only KTP exhibited strong inter-media relationships, indicating that its aqueous-phase dominance significantly influences sediment and pore water concentrations. Peak concentrations occurred downstream of a Hangzhou sewage treatment plant, reflecting direct effluent inputs. Other PPCPs showed weaker correlations, suggesting their distribution depends more on sediment properties (specifically organic content) and hydrodynamic conditions (Tian YJ et al., 2023).

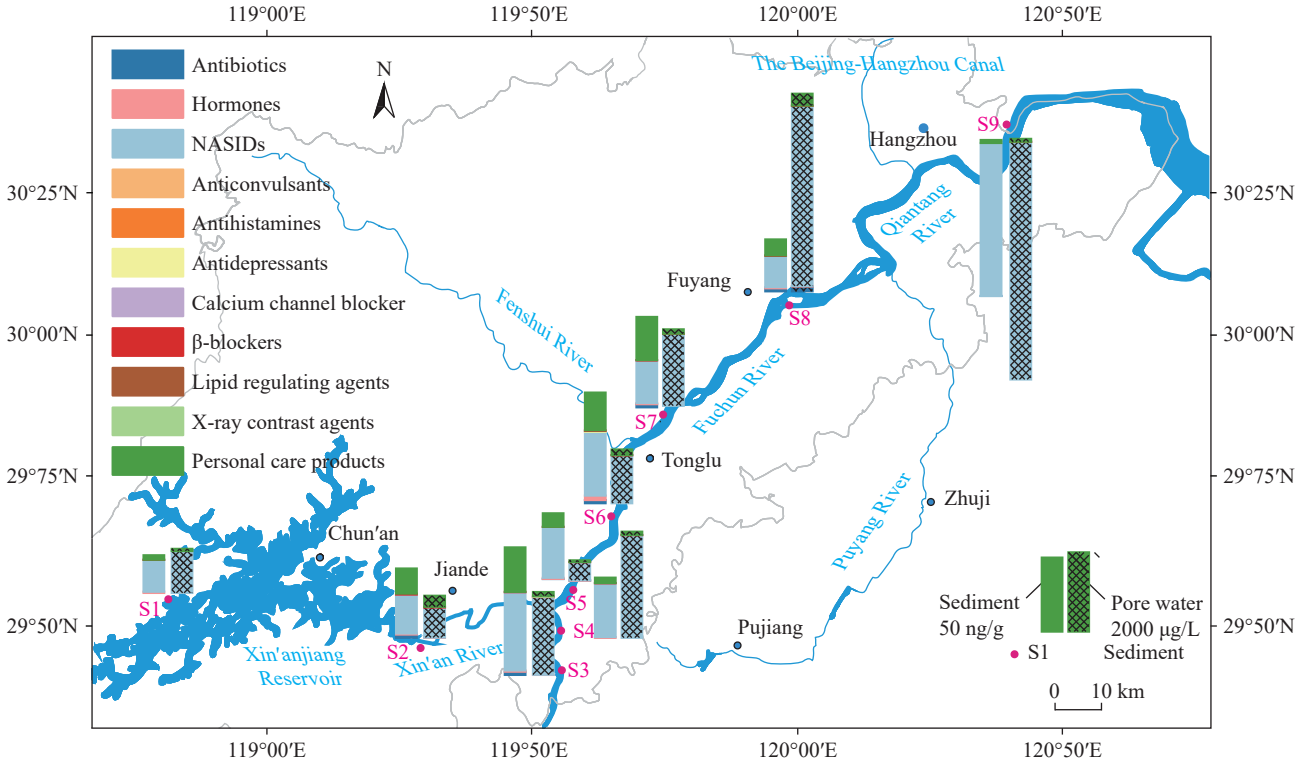

Fig. 4. Occurrence and distribution of PPCPs in sediment and pore water of Qiantang River Basin.

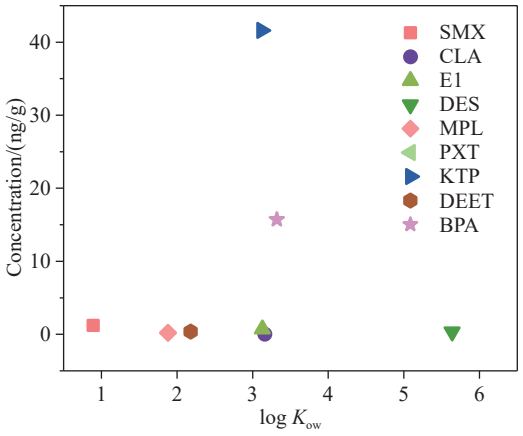

Fig. 5. Correlation between the concentration of PPCPs detected in sediments and the corresponding *K*ow was analyzed.

The pseudo-partition coefficient (P-PC) was developed to characterize PPCP distribution and migration in the surface water-sediment-pore water system. While k_{sw} (sediment-surface water) and k_{sp} (sediment-pore water) values are reported (Table. 1), P-PCs cannot represent true equilibrium partition coefficients due to dynamic river-sediment interactions (Kim SC et al., 2007).

Antibiotic concentrations in pore water were generally lower than in surface water, likely due to continuous inputs elevating surface levels. SAs (notably SMX) and chloramphenicol (CAP) dominated the system. SMX showed greater surface water and sediment occurrence, with $k_{sw} < k_{sp}$ indicating sustained surface water influx. Its aqueous-phase dominance aligns with lower logK_{ow} values and reduced organic affinity. In contrast, hydrophobic macrolides preferentially adsorbed to sediments ($k_{sw} > k_{sp}$), with pore water concentrations exceeding surface levels via desorption (Yan CX et al., 2013).

BPA and DEET were the dominant PCPs, exhibiting concentration gradients of pore water > surface water > sediment. Although ATZ has a logK_{ow} of 2.61 (Table. S1), it was only detected in aqueous phases (surface/pore water), indicating that sediment properties (including clay content, organic matter, and pH) critically control its environmental behavior. Therefore, the adsorption of ATZ cannot be accurately predicted based solely on P-PC or logK_{ow} values.

Flood season sampling in the lower Qiantang River revealed key hydrological conditions influencing PPCPs distribution: High water volume, fast water flow, less sediment at the bottom of the river, and higher sand content. High EE2 levels in Hangzhou Sewage Treatment Plant's downstream surface water (attributed to effluents and industrial discharges) contrasted with undetected sediment/pore-water concentrations. It is high logK_{ow} and water disturbance susceptibility. At the same time, a large amount of DEET was detected in the water phase at this position, while the concentration in the solid phase was low. The highest value of DEET in the sediment appeared at the S3 sampling point, where the water flow was gentle, indicating that the water flow did affect the environmental behavior of PPCPs.

While previous research has primarily examined antibiotic behavior in surface water-sediment-pore water systems, this study reveals distinct PPCPs distribution patterns influenced by domestic and industrial wastewater inputs. These

Table 1. P-PC value and octanol-water partition coefficient of PPCPs were detected in sediments.

PPCPs	k_{sw}, L/kg	k_{sp}, L/kg	logK_{ow}	PPCPs	k_{sw}, L/kg	k_{sp}, L/kg	logK_{ow}
SMX	0.11–0.55	–	0.89	KTP	0.08–0.47	0.01–0.07	3.12
CLA	0.11–0.12	0.05	3.16	DEET	0.01–0.07	0.01–0.05	2.18
E1	–	0.08–0.18	3.13	BPA	0.02–1.31	0.07–0.63	3.32
MPL	0.08–0.28	0.04–0.24	1.88				

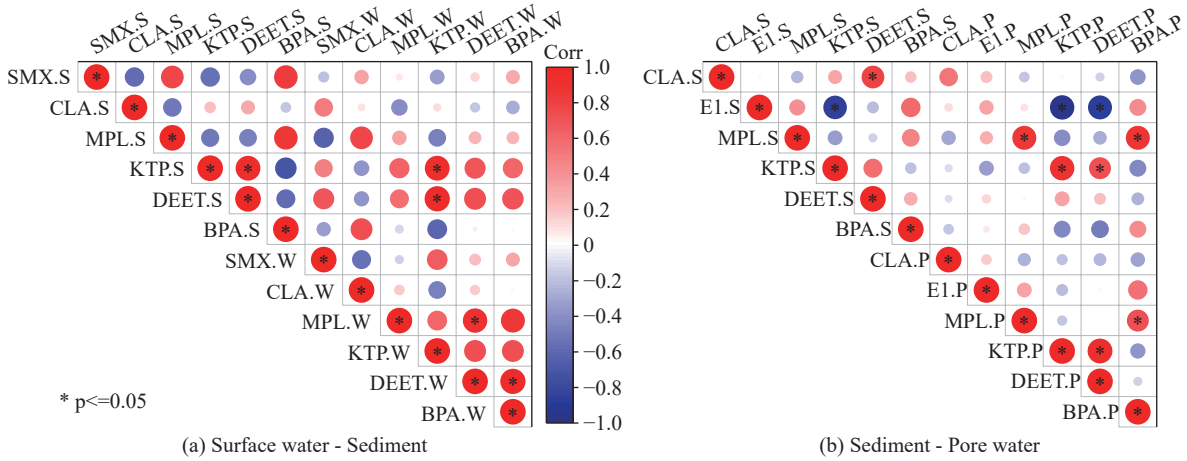

Fig. 6. Correlation of PPCPs concentration detected in surface water, sediment, and pore water.

anthropogenic sources led to elevated PPCPs concentrations and reduced P-PC values compared to prior studies. Sediment characteristics varied significantly along the Qiantang River, where marine sediment influences increased silt content in the lower reaches. The increased silt content affects PPCPs' adsorption in sediments (Zhang M et al., 2021).

E1, diethylstilbestrol (DES), and paroxetine (PXT) were exclusively detected in sediments due to their hydrophobic properties and high K_{ow} values, demonstrating strong sediment affinity (Lei BL et al., 2008). These compounds were absent in drinking water, confirming their environmental persistence in sediments. In contrast, ATZ showed aqueous-phase dominance, detected only in surface water but not in sediments, indicating high mobility and resistance to conventional drinking water treatment. These findings highlight the need for improved treatment strategies targeting water-soluble contaminants like ATZ while recognizing the sediment as a major sink for hydrophobic PPCPs.

The absence of mechanistic modeling (such as QWASI for multimedia partitioning) limits the quantitative interpretation of PPCPs' phase transfer. Future work should integrate process-based modeling with field measurements to resolve the coupled hydro-bio-geochemical controls on contaminant fate.

3.3. Risk assessment of PPCPs in the Qiantang River Basin

3.3.1. Ecological risk assessment

Considering only a single species in risk assessment may ignore the impacts of pollutants on other trophic levels and lead to inaccurate assessment, therefore, in this study, the authors selected three typical trophic level organisms in the water body, namely algae, invertebrates and fish (Li L et al., 2021), to comprehensively and accurately assess the ecological risk caused by PPCPs. Table. S2 presents the $PNEC_w$ values (μg/L) of target PPCPs.

Fig. 7a presents the RQ values for PPCPs in surface water. While IPM and E1 showed no significant risk to aquatic organisms, other detected compounds demonstrated varying risk levels. KTP and ibuprofen (IBU) exhibited medium-to-high risk across all tested organisms, and PCPs also displayed potential ecological hazards. Even in relatively low concentrations, antibiotics pose substantial ecological risks to algae.

SMX and KTP emerged as the highest-risk PPCPs in surface water. Spatial analysis revealed three distinct risk zones (Figs. 7b–d): (1) The Xin'anjiang and Lanjiang sections showed minimal ecological risks, with KTP posing low risk to algae/invertebrates and SMX displaying significant algal toxicity. (2) The Fuchunjiang section exhibited moderate risks, particularly KTP's algal toxicity from industrial/domestic wastewater inputs. (3) The Qiantangjiang section demonstrated the most severe risks, including high-risk antibiotics and hormones. This gradient strongly correlates with anthropogenic pressures, such as population density and urban development, intensifying downstream.

Table S3 presents the PNEC values for the PPCPs that exhibit higher detection rates in the sediments. While PPCP concentrations in the sediments examined in this study were comparatively low, potential risks were still present (Fig. 8). The ecological risk assessment revealed that SMX and CLA exhibit significant toxicity to algal communities in localized areas while demonstrating minimal to negligible effects on invertebrate and fish populations. KTP and BPA are

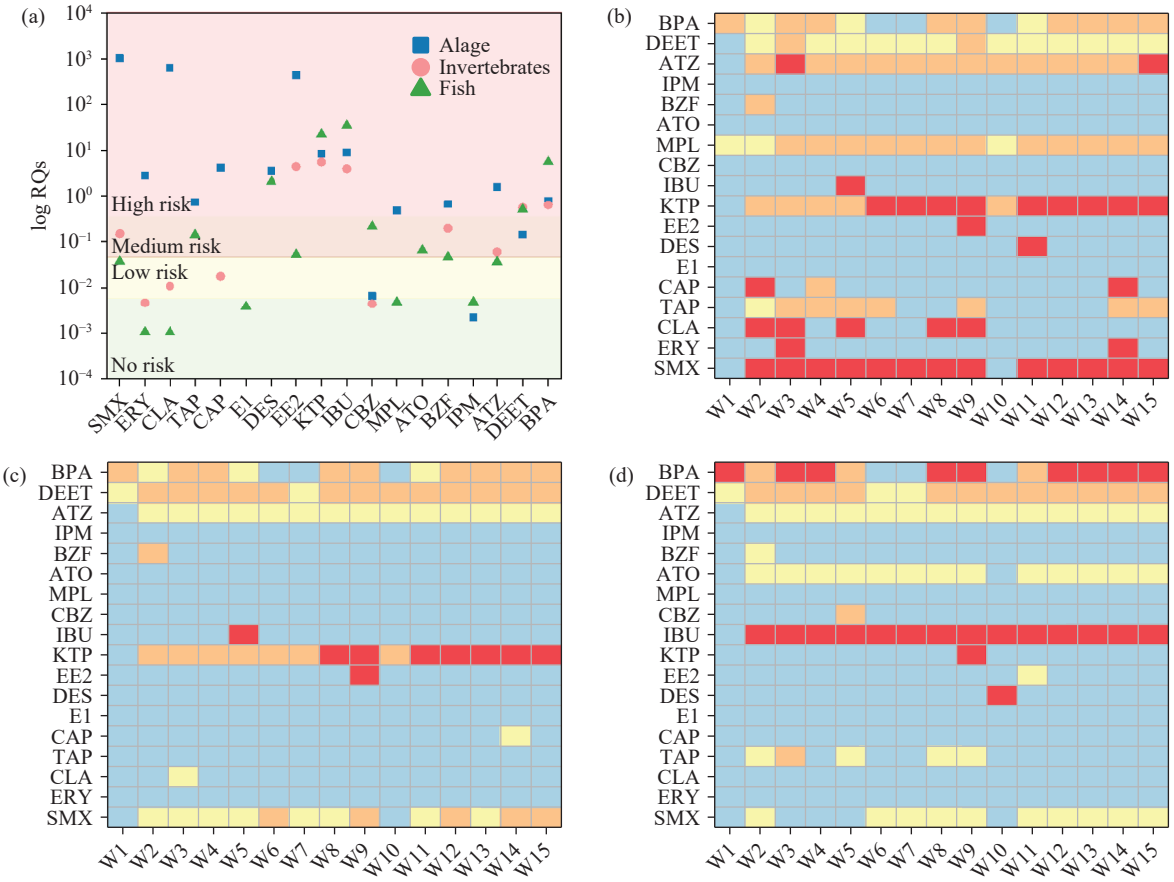

Fig. 7. Ecological risk of detecting PPCPs in surface water. The figure includes (a) the risk level of the main detected substances to the three trophic levels and the risk level of the detected substances to algae (b), invertebrates (c), and fish (d) in each sampling site of surface water.

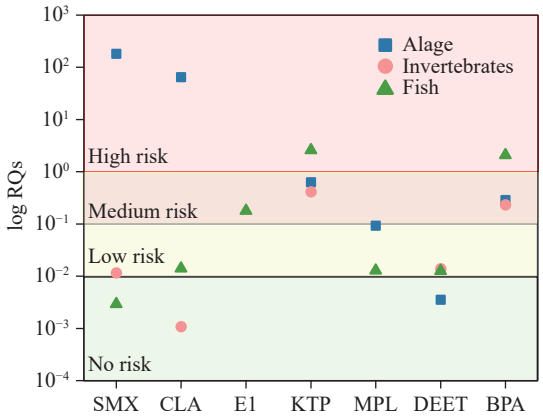

Fig. 8. Ecological risk of PPCPs detected in sediments.

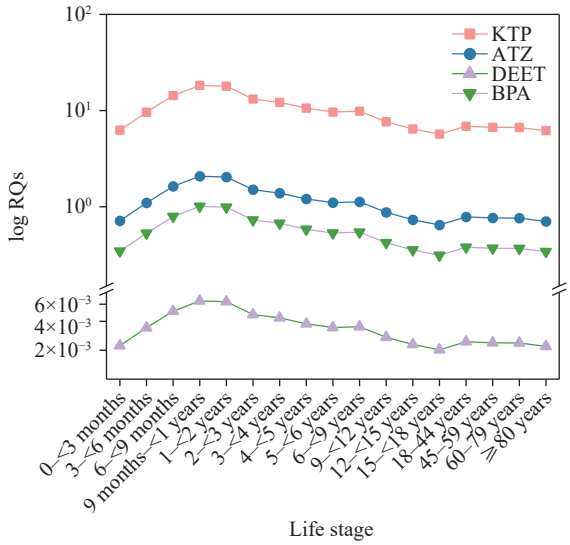

Fig. 9. Health risks of residual PPCPs in drinking water at different ages.

commonly detected in sediments and pose medium to high risks to all selected organisms. However, various pollutants and their metabolites frequently coexist in aquatic settings and can have synergistic or antagonistic impacts. Consequently, the RQ value of an individual pollutant presents a somewhat limited perspective.

3.3.2. Health risk assessment

Human health risks were assessed using RQ values across different age groups (Fig. 9). Table S4 presents each demographic's body weight and drinking water intake levels. The evaluation focused on five specific compounds detected in drinking water, as other PPCPs were undetected or lacked established ADI values (Table S5). Children aged 9 months to 2 years showed the highest RQ values, attributable to their lower body weight and higher water intake (Hebig KH et al., 2017). KTP demonstrated elevated RQs across all age groups, while ATZ and BPA exhibited increased risks, primarily in

young children.

Persistent and mobile chemicals (PMT/vPvM) threaten drinking water supplies significantly. While REACH's 2017 PMT list includes some industrial PPCPs (Arp HP et al., 2019), many remain unassessed. The Guangzhou Institute of Geochemistry evaluated 169 PPCPs detected in Chinese waters against REACH criteria (Huang C et al., 2021). This study screened five drinking water PPCPs using REACH and the Institute's PMT lists (Table 2). The assessment identified ATZ as a confirmed PMT/vPvM substance affecting drinking water quality, while DEET and KTP showed potential PMT characteristics with lower risks. DES and BPA did not meet PMT criteria. Notably, 60% of detected drinking water PPCPs qualified as PMT/vPvM substances - a higher proportion than reported in previous REACH and Guangzhou Institute of Geochemistry evaluations. This discrepancy likely reflects the inherent pseudo-persistence and mobility of PPCPs and the limited compound diversity in this study. Combined risk quotient and PMT assessments highlight KTP, ATZ, and DEET as priority contaminants requiring enhanced regulatory attention. However, the methodology for assessing the RQ value for health risk in this study only considered the exposure route of drinking water intake and ignored the components of dermal contact and aquatic organisms' transmission through the food chain (Zhu DN et al., 2022), and thus may have underestimated the risk level of PPCPs.

Table 2 PMT evaluation of main PPCPs in drinking water.

PPCPs	PMT types	PPCPs	PMT types
ATZ	vPvM/PMT	DES	Not PMT/vPvM
DEET	Potential PMT/vPvM++	BPA	Not PMT
KTP	Potential PMT/vPvM		

4. Conclusions

(i) PPCPs are prevalent in the Qiantang River Basin's surface water, sediment, and pore water. KTP is the most significant contributor among all environmental media, with personal care compounds like DEET and ATZ also frequently detected. The concentration of PPCPs in surface water and pore water exhibits a notable increasing trend from upstream to downstream, whereas in sediment and drinking water, it remains relatively constant geographically.

(ii) The migration of PPCPs in surface water, sediment, and pore water in this study is affected by many circumstances and has a weak association with K_{ow}. Their physicochemical characteristics influence the distribution of PPCPs. The concentration of antibiotics in pore water is often lower than in surface water, whereas personal care items in pore water are elevated. The widespread distribution of KTP in the aqueous phase suggests a future need to incorporate the pharmaceutical into watershed pollutant inventory management.

(iii) The ecological risk assessment findings indicate that PPCPs in surface water and sediment reveal that antibiotics present a significant risk to algae, while KTP poses a considerable risk to organisms across all trophic levels. The health risk evaluation of drinking water reveals that the possible dangers of KTP and ATZ in the Qiantang River region warrant careful consideration.

CRediT authorship contribution statement

Ge Feng and Nan Gai conceived of the presented idea. Ge Feng and Guo-hui Lu carried out the experiment. All authors discussed the results and contributed to the final manuscript.

Declaration of competing interest

The authors declare no conflicts of interest.

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Supplementary dataset

Supplementary dataset (Tables. S1–S5) to this article can be found online at doi: 10.31035/cg2025025.

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