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Quaternary Ammonium Compounds (QACs) in Textiles and Laundry Wastewater: Occurrence and Environmental Risk Assessment

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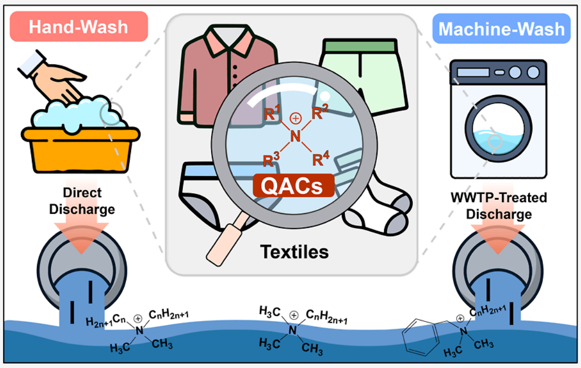
ABSTRACT: Quaternary ammonium compounds (QACs) have been used as antimicrobial additives in textiles, but information on their levels in textiles and ecological impacts from laundry wastewater is scarce. Here, 119 textile products from Chinese online vendors were analyzed for traditional and emerging QACs using target and suspect screening approaches with high-resolution mass spectrometry. The total concentrations of 18 traditional QACs ranged from 7.34 to 145,000 ng/g, and 16 emerging QACs were identified with total concentrations at 5.45 to 349,000 ng/g. Under laboratory-controlled conditions, over 76% of the selected textiles exhibited QAC migration rates exceeding 50% after washing. In simulated laundry experiments, hand-washing released a ΣQAC concentration of 42.1 ng/mL, and machine-washing released 22.4 ng/mL, with differences attributed to the water volume used by different methods. Both laundry wastewater and QACs solution at equivalent concentrations caused dose-dependent immobilization of *Daphnia magna*. Combining bioassay-based risk characterization factors with environmental dilution factors across 150 countries, 7% of regions showed high risk after WWTP-treatment, increasing to 41% for direct discharge with sewage. The high migration rates suggest that QAC-treated textiles not only fail to maintain long-term antimicrobial efficacy but also contribute to continuous low-level environmental exposures, raising concerns about their essential use in nondisposable textile materials.



KEYWORDS: *antimicrobial agents, textiles, laundry wastewater, aquatic toxicity, environmental risk*

INTRODUCTION

The textile industry consumes numerous chemicals during manufacturing and processing to achieve desired quality and performance, leading to considerable environmental burdens and potential risks to human health.[1,2] Among these chemicals, antimicrobial agents, such as silver, metal oxides, photoactive dyes, *N*-halamines, triclosan, polybiguanides, chitosan, and plant-derived bioactive agents, are widely applied to inhibit bacterial growth through their biocidal and biostatic properties.[3,4] In line with their broad applications, the global market for antimicrobial coatings was valued at $8.1 billion in 2021, with an anticipated compound annual growth rate of 13.1% from 2021 to 2028.[5] Despite their functional benefits, increasing evidence of their impacts on the environment and human health has raised concerns regarding their biocompatibility and ecological acceptability. For example, triclosan has been shown to cause severe histopathological and apoptotic damage in fish,[6] oxidative stress and cytogenotoxic effects in freshwater mussels,[7] and poses particular risks to algae in aquatic ecosystems.[8] These concerns have prompted manufacturers to actively search for alternative antimicrobial agents that can provide effective performance while minimizing environmental and health risks.[9]

Quaternary ammonium compounds (QACs), characterized by a central ammonium structure linked to alkyl and aromatic moieties, have been increasingly employed in textile antimicrobial finishes owing to their broad antimicrobial efficacy, low toxicity, and chemical stability.[3,10,11] Their amphiphilic and permanently charged structures enable integration into microbial surfaces, exerting antimicrobial effects on bacteria, fungi, and viruses by disrupting cell membranes.[12] Notably, awareness regarding antimicrobial textiles, including QAC-based finishes, has surged following the COVID-19 pandemic.[13] Despite the structural diversity of QACs, previous research has primarily focused on a few “traditional” groups, such as benzylalkyldimethylammonium compounds (BACs, C8−C18), dialkyldimethylammonium compounds (DADMACs, C8−C18), and alkyltrimethylammonium compounds (ATMACs, C8−C18), whereas many other “emerging” QACs

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documented in textile products remain insufficiently investigated. For example, cetylpyridinium chloride (CPC), a well-known antimicrobial ingredient in oral care products (e.g., mouthwash) that has also been reported in wastewater, is used in wool and cotton textiles due to its potent and durable antimicrobial efficacy.[14−16] Choline, a natural QAC, can be grafted onto cotton surfaces for antibacterial activity.[17] Modified QACs, such as polymerizable quaternary ammonium salts with acrylate groups or alkyltrialkoxysilanes, have been developed to enhance durability and antimicrobial properties in textiles.[3] However, there still remains a lack of systematic investigations on the occurrence and composition profiles of QAC analogs in textiles, including daily clothing and antimicrobial functional items.

In addition, coating techniques such as solution casting and spray coating often result in weak physical bonding between QACs and textile fibers, facilitating leaching during use and washing.[18,19] This vulnerability resembles that of other unbound textile additives (e.g., dyes, flame retardants, and plasticizers) known to migrate from fabric under similar conditions.[20] Consequently, QACs can readily release from textiles during laundering processes, entering domestic wastewater streams that may discharge directly into surface water systems without further treatment. In urban areas equipped with wastewater treatment plants (WWTPs), QACs-laden laundry wastewater is routed to these facilities, where partial removal (typically ranging from 90% to 98%) occurs through sorption to sludge and biodegradation, yet QAC residues remain in treated effluents.[21,22] Given the substantial volume of laundry wastewater generated worldwide each day, leaching of QACs from textiles has become a significant source of these compounds in surface waters, through either direct discharges or WWTP-treated effluents, thereby posing elevated ecological risks in aquatic environments.

QACs are toxic to a wide range of aquatic organisms, including fish, invertebrates, and algae.[13] Among these, *Daphnia magna* (*D. magna*) is particularly sensitive to QAC exposure, with acute EC_{50} values reported as low as 5.8 μg/L for C12−C16 BACs and 18 μg/L C10-DADMAC, respectively.[23,24] What raises particular concern is that these effect concentrations fall within the range of QAC levels detected in surface waters, such as C12−16 BACs reaching 342 μg/L in Poland and C14-BAC up to 21.6 μg/L in Korea.[25,26] Additionally, the total QAC concentration in U.S. stormwater runoff has been reported at up to 12.2 μg/L, indicating that episodic runoff inputs alone can reach levels comparable to acute effect concentrations.[27] However, most ecotoxicological studies and conventional risk assessments have focused on individual QACs, limiting the evaluation of ecological risks from complex QAC mixtures, such as those present in laundry wastewater. To address these limitations, whole-effluent toxicity (WET) approaches are required by applying bioassays on undiluted or simulated samples to directly assess aggregate mixture effects.[28] The resulting effect concentrations can be used for risk characterization ratio calculations, with dilution factors incorporated to approximate realistic environmental exposures and refine ecological risk assessments.

In order to address the limited understanding of QAC analogs in textiles, their release during laundering, and the associated ecological risks, the present study aims to (1) identify and (semi)quantify both conventional (C8−18 BACs, DADMACs, and ATMACs) and emerging QACs in textiles with diverse functions by a combined target and suspect screening approach using high-resolution mass spectrometry; (2) evaluate the readily releasable fraction of QACs from textiles in early wash through one single cycle laundry simulated experiments and assess the toxic effects of leached QAC mixtures using whole-effluent toxicity assays with *D. magna*; (3) characterize the ecological risks of QAC-containing laundry wastewater using a risk characterization ratio framework that integrates bioassay results with modeled environmental dilution scenarios of WWTP-treated discharges or direct discharges. Collectively, this study advances the understanding of QAC occurrence and release from textiles, and offers a more realistic assessment of their ecological risks in aquatic environments, thereby providing new insights for sustainable antimicrobial textile production and wastewater management.

■ MATERIALS AND METHODS

Chemicals and Reagents

The detailed information on analytical standards is listed in Table S1. All solvents applied in the present study were HPLC grade or higher.

Sample Collection

A total of 119 textile samples, including face masks (*n* = 20), socks (*n* = 20), towels (*n* = 20), sportswear (*n* = 10), underwear (*n* = 20), clothing (*n* = 20), quilt (*n* = 4) and medical textiles (*n* = 5) were purchased from online stores in April 2023. The selected textile products were manufactured from 2019 to 2023 and are made of organic cotton, polyester, spandex, polypropylene, polyamide, or polyethylene. The 119 textile samples were categorized by clothing material into natural fiber (*n* = 35) and synthetic fiber (including chemical fiber and chemical fiber blended, *n* = 84), or antibacterial textile (*n* = 69) and nonantibacterial textile (*n* = 50) based on their labeling information. The details of the textiles are shown in Table S2 in the Supporting Information, including fabric composition, clothing type, antibacterial properties, and year of production. All samples were transported to the laboratory and sealed in polypropylene (PP) bags for further chemical analysis.

Sample Analysis

The sample pretreatment method was adapted from previous studies with minor modifications.[29,30] A square piece of cloth (2 × 2 cm) was randomly cut out of each textile sample and then cut into 2 × 2 mm pieces to improve extraction efficiency. These pieces were weighed into a 15 mL polypropylene tube and spiked with 50 ng of surrogate standards (d_7-C12-BAC and d_9-C10-ATMAC). The samples were extracted with 4 mL of methanol containing 1% formic acid, then sonicated for 30 min at room temperature. The extraction procedure was repeated 2 times. Subsequently, the supernatants were combined, concentrated to 500 μL under a gentle stream of nitrogen, filtered through a 0.22 μm nylon syringe filter, and spiked with 50 ng of internal standards (d_7-C14-BAC) before instrumental analysis.

500 mL of wastewater from machine- and hand-washing were mixed with 20 ng of surrogate standards (d_7-C12-BAC and d_9-C10-ATMAC) and loaded on Oasis WCX cartridges (3 cc, 60 mg, 30 μm), which were preconditioned with 3 mL of methanol and 3 mL of deionized water. The cartridges were washed with 3 mL of a 5% ammonia solution in water and 3 mL of a methanol−water mixture (1:9, v/v) to remove impurities. The cartridges were allowed to dry completely under a vacuum and then eluted with 6 mL of 2% formic acid in methanol. The extracts were concentrated to dryness under a stream of nitrogen, reconstituted to a final volume of 1 mL with methanol, filtered through a 0.22 μm nylon syringe filter, and spiked with internal standards (d_7-C14-BAC) before instrumental analysis.

The samples were analyzed using an Agilent 1290 Infinity ultrahigh-performance liquid chromatograph system coupled to an Agilent 6546 quadrupole time-of-flight mass spectrometer (UPLC-QTOF/MS, Santa Clara, CA). Instrumental parameters and the target and suspect screening workflow followed the established method from our previous study.[31] The confidence level of the identified emerging QACs was adapted from Schymanski et al.[32]

Simulated Laundering Experiments under Laboratory Conditions

This laundering experiment included three categories of textiles with high demands for antimicrobial functionalities (i.e., socks, towels, and underwear), each including labeled antibacterial and nonantibacterial properties. For each textile category, three samples exhibiting the highest total QAC concentrations ($\sum$QAC) were selected for analysis. Fabric pieces, approximately 4 × 4 cm, were cut from these selected samples and placed into separate beakers containing 400 mL of preheated Milli-Q water at 20 °C. These beakers were then shaken at 120 rpm in an incubator shaker for 1 h. After laundering, the fabric pieces were gently squeezed to remove excess water and air-dried at room temperature under aluminum foil within a fume hood. The concentrations of QACs in the laundered fabric samples were subsequently measured using the methods previously described.[31]

The migration rate of a chemical after the laundering experiment was calculated using eq 1

$$\text{migration rate (\%)} = \frac{(C_{\text{before}} - C_{\text{after}}) \times 100\%}{C_{\text{before}}} \quad (1)$$

where C_{before} is the $\sum$QAC concentration of a textile sample before laundering (ng/g), C_{after} is the $\sum$QAC concentration of a textile sample after laundering (ng/g).

Simulated Laundering Experiments under Real-Life Scenarios

To determine the concentration of QACs released into wash water under conditions representative of actual use, two experiments were performed: machine-washing and hand-washing, which reflect the predominant mechanical actions during typical household laundering. Specifically, four categories of textiles, including socks, towels, sportswear, and clothing, were selected to represent typical daily wash scenarios, in which they are commonly mixed and washed together. The samples with the highest QAC concentrations were selected within each textile category. Subsequently, to enable a comparative analysis of machine-washing and hand-washing processes, each selected sample was evenly divided into two portions by scissors, weighed, and then subjected to either machine-washing or hand-washing experiments, respectively. The machine-washing process was conducted using a 3.5 kg capacity washing machine (XQB35−688G, Changhong, Sichuan, China), set to a single wash mode according to the manufacturer's instructions. Prior to the experiment, 50 L of QAC-free dechlorinated tap water was collected in a food-grade plastic bucket (100 L, with a lid, prerinsed with DI water) and left to dechlorinate for over 2 days. This dechlorinated water was then used for both the washing experiment and the acute exposure assay with *D. magna*. In the machine-washing process, the four clothing items were soaked together in tap water without detergent for 5 min, then washed in single wash mode with 12 L of tap water, followed by two rinses with 4.5 L of tap water. A total of 21 L of wastewater was collected for further chemical analysis.[33]

The hand-washing process was simulated in the laboratory without detergent as well according to a protocol established previously.[33] First, four clothing items were soaked for 5 min in a 15 L stainless steel basin containing 6 L of dechlorinated tap water. Next, the garments were scrubbed 90 times on a wooden washboard to ensure thorough cleaning. Following this, the textiles were wrung by hand to remove excess water and then rinsed twice with 3 L of dechlorinated tap water each time.[34] A total of 12 L of wastewater was subsequently combined for further analysis.

Toxicity Assays Using *D. magna*

D. magna (obtained from the State Key Laboratory of the Chinese Research Academy of Environmental Sciences) were cultured in 2 L glass vessels containing 1.5 L of reconstituted water under controlled laboratory conditions (temperature of 22 ± 1 °C, humidity of 25%, 16 h/8 h light/dark cycle, pH of 7.5 ± 0.2, dissolved oxygen concentration of 8.2 ± 0.3 mg/L, conductivity of 560 μS/cm). The daphnids were fed daily with a suspension of *Chlorella vulgaris* algae (obtained from the Institute of Hydrobiology, Chinese Academy of Sciences). The acute testing procedure was conducted in accordance with the OECD guideline *D. magna* Acute Immobilization Test.[35] To perform toxicity evaluation by whole-effluent toxicity (WET) approaches, two exposure media were prepared: hand-washing laundry wastewater collected from simulated washing experiments, and a standard mixture solution containing equivalent total QAC concentration (prepared in QAC-free, dechlorinated tap water by dissolving the QAC congeners detected in the simulated laundry wastewater that each contributed >1% to total QACs at their respective measured concentrations). A preliminary range-finding test revealed that undiluted laundry wastewater could cause acute immobilization and mortality in all *D. magna* individuals. Therefore, exposure gradients in the definitive test were established by diluting the laundry wastewater with reconstituted water. This approach enabled the derivation of EC_{50} values through dilution percentages, providing a practical method to evaluate the toxicity of complex mixtures without the need to identify individual constituents.

In the finalized experimental setup, *D. magna* neonates (<24 h old) were exposed to various dilutions of both test solutions, with three replicates per concentration. Each replicate consisted of 8 neonates placed in a 50 mL glass beaker containing the test solution (providing approximately 6.25 mL per organism), resulting in a total of 24 daphnids per concentration level. The control groups were maintained in reconstituted water without added QACs, and clean washing water collected from the same washing procedure without textiles added, while no immobilization effects were observed in the controls. The exposure was conducted in an environmental chamber under controlled conditions (temperature of 22 ± 1 °C, humidity of 25%, 16 h/8 h light/dark cycle, pH of 7.5 ± 0.2, dissolved oxygen concentration of 8.2 ± 0.3 mg/L). Test solutions were monitored daily for pH and dissolved oxygen to ensure stable water quality. No food was provided during the acute toxicity test. Immobilization was recorded at 48 h, with individuals considered immobilized if unable to swim within 15 s after gentle agitation of the test vessel.

Ecological Risk Assessment of QACs in Laundry Wastewater

Predicted environmental concentrations (PEC) were calculated based on measured QAC concentrations in laundry wastewater and their subsequent dilution in sewage and receiving water bodies. In addition, two discharge scenarios were considered:

(i) WWTP-treated discharge, in which QAC removal efficiencies during wastewater treatment were applied;

$$\text{PEC} = C_{\text{lw}} \times f \times (1 - \text{R}) \times \frac{1}{\text{DF}} \quad (2)$$

(ii) Direct discharge, in which laundry wastewater-mixed sewage was assumed to enter the environmental receiving waters without prior treatment.

$$\text{PEC} = C_{\text{lw}} \times f \times \frac{1}{\text{DF}} \quad (3)$$

where C_{lw} (ng/mL) represents the concentrations of QAC mixtures in the wastewater from laundry activities determined in the current study, f (10%) is the proportion of laundry wastewater in the domestic sewage influent,[36] R (90%) stands for the QACs removal rate by WWTP,[37] DF is the dilution factor of WWTP-treated effluent in receiving surface waters, which was derived for 150 countries by integrating population data, per capita water use, and modeled river runoff to estimate the ratio of river flow to domestic wastewater discharge.[38]

To further clarify the ecological risks of QACs from wastewater effluents, risk characterization ratios (RCRs), often used to assess the risk of pollutants in the environment, were calculated by PEC and predicted no-effect concentrations (PNEC) according to the following equations

$$\mathrm{PNEC} = \frac{\mathrm{EC}_{50}}{\mathrm{AF}} \tag{4}$$

$$\mathrm{RCR} = \frac{\mathrm{PEC}}{\mathrm{PNEC}} \tag{5}$$

The PNEC values were derived from the EC_{50} of 84% C_{lw} based on the results of the *D. magna* toxicity test. The value of 1000 was applied to the assessment factor (AF) to account for interspecies variation and extrapolation from acute to chronic effects, following the guidelines set by the European Chemical Agency for deriving PNECs from acute toxicity data.[39] The risk levels based on RCR are defined as follows: an RCR of ≥1.0 indicates high risk, an RCR between 0.1 and 1.0 indicates moderate risk, and an RCR of <0.1 indicates low risk.

Quality Control and Quality Assurance (QA/QC)

For each trial of the machine and hand-washing procedures, the apparatus, including the washing machine, stainless-steel basin, and wooden washboard, was precleaned using the same washing protocol to remove any potential QAC residues and prevent cross-contamination. For the chemical analysis, one procedural blank containing precleaned cotton was included in each batch of 15 samples. Procedure blanks ($n = 6$) and matrix spike recovery samples ($n = 6$, spiked with 50 ng target analytes) were analyzed across each pretreatment process of both the textile analysis and laundry simulation analysis. All reported concentrations were subtracted from the average procedural blank concentrations, but were not corrected by surrogate recovery. The method detection limit (MDL) was defined as three times the standard deviation of the target analyte detected in procedural blanks. If compounds were not observed in the procedural blanks, MDL was set as 3 times the signal-to-noise (S/N) based on the lowest calibration point. For samples with detected concentrations above the MDL, the reported concentrations were corrected by subtracting the corresponding procedural blank concentration. The details on matrix spike recoveries, surrogate recoveries, and procedural blanks were presented in Tables S3 and S4.

Data Analysis

Descriptive statistics were computed using Microsoft Excel. Plots were generated using Origin, Minitab, R Studio, and Adobe Illustrator. The Kruskal−Wallis test was performed by SPSS to evaluate differences in QAC concentrations among textile categories and materials after data non-normal distributions were examined by Shapiro−Wilk test, while the Mann−Whitney test was applied to compare concentrations between textiles with or without antimicrobial functions among categories of underwear, towels, socks, sportswear, and clothing (remaining categories excluded due to the limited number of samples).

RESULTS AND DISCUSSION

QAC Concentrations in Textiles

Overall, 18 traditional QACs (C8−18 BACs, C8−18 DADMACs, and C8−18 ATMACs) and 16 emerging QACs were found in the collected textiles (Table 1 and Figure S1). Among the emerging QACs, 11 were confirmed with commercially available standards (i.e., confidence level S1), and 5 matched reference MS/MS fragment ions (i.e., confidence level S2). The MS^2 spectra of representative emerging QACs, including benzethonium (BEC), C8:10-DADMAC, methyltrioctylammonium (MTOAC), and C6-DADMAC, are shown in Figure S2.

At least one analyte was found in each textile sample. The total QAC concentrations (sum of $\Sigma_{\mathrm{traditional}}$ QAC and $\Sigma_{\mathrm{emerging}}$ QAC) ranged from 27.3−351,000 ng/g at a median concentration of 1050 ng/g. ATMACs were the most abundant traditional QAC congeners in these samples, with a median concentration of 55.9 ng/g, followed by BACs (38.6 ng/g) and DADMACs (5.98 ng/g). The dominance of ATMACs in textiles can be attributed to their widespread use as fabric softeners and antistatic agents in textile finishing processes.[40,41] Specifically, C12-BAC and C16-ATMAC exhibited the highest median concentrations, at 17.6 and 14.8 ng/g, with detection frequencies of 67% and 98%, respectively. C12-BAC is used to impart antimicrobial properties to textiles at the fiber stage or during the finishing process.[42] In addition, combining C12-BAC with other antimicrobial agents, such as silver nanoparticles or other QACs, can enhance its effectiveness against SARS-CoV-2.[43] C16-ATMAC, also known as cetyltrimethylammonium bromide (CTAB), is frequently applied to polyester, polypropylene, and viscose nonwoven fabrics for its activity against bacteria such as *Staphylococcus aureus*, *Escherichia coli*, and *Pseudomonas aeruginosa*, especially when combined with fluorochemical water repellents.[44,45] In contrast, DADMACs were less prominent, with only C8-DADMAC and C10-DADMAC being detected in more than half of the samples. Their concentrations (medians 1.27 and 0.819 ng/g, respectively) were generally lower than those of BACs and ATMACs, reflecting their more limited role in textile applications.

Emerging QACs were also frequently detected in these textile samples. Among them, diallyldimethylammonium (DDA) was found in 60% of the textiles with a median concentration of 12.2 ng/g. Although its median level was not as high as some other emerging QACs, DDA exhibited extremely high concentrations in a subset of samples: 30 out of 119 textiles contained concentrations higher than 1000 ng/g, of which 20 exceeded 10,000 ng/g, with the maximum reaching 349,000 ng/g. This skewed distribution implies DDA as a compound of particular concern, likely reflecting its targeted application in specific fabrics, consistent with previous reports of its incorporation into cotton and silk fabrics to enhance reactive dye adsorption and impart antibacterial properties.[46,47] Apart from DDA, several emerging DADMACs (C2:4-DADMAC, C4:6-DADMAC, C6-DADMAC, and C6:8-DADMAC) were also frequently detected (detection frequencies: 93−100%), with median concentrations of 43.4, 0.761, 6.33, and 0.463 ng/g, respectively. The co-occurrence of these mixed short-chain DADMACs and traditional long-chain DADMACs (C8−C18) is not fully understood. Our previous study suggested that mixed DADMACs may inadvertently manifest as impurities alongside their paired DADMAC counterparts during the subsequent stages of catalytic hydrogenation and quaternization synthesis processes.[31] In addition, benzyltriethylammonium (BTEAC), a BAC analogue, was detected in 74% of the samples with a median concentration of 21.4 ng/g. It is reported that BTEAC is often combined with other finishing agents to create protective barriers on fabrics, providing resistance to microbial growth through repeated washing.[48−50] By contrast, 4 ATMAC analogues, including MTOAC, acetylcholine (AChCl), tetrapentylammonium (Pen_4NCl) and tributylmethylammonium (TMABC), were detected in less than half of the samples. The concentrations of emerging QACs were nearly twice as high as those of traditional QACs, with medians of 346 ng/g compared to 176 ng/g, underscoring their widespread occurrence in textiles.

The increasing demand for antimicrobial, odor-resistant, and antistatic functions in textiles due to growing awareness of hygiene standards may have driven the use of more efficient and durable emerging QACs in the textile industry.[33] Previous studies found that some novel QACs may offer broader-

Table 1. Detection Frequencies (DF, %) and Median Concentrations of QACs in Various Types of Textiles (ng/g)

		total		mask (n = 20)		sock (n = 20)		towel (n = 20)		sportswear (n = 10)		underwear (n = 20)		quilt (n = 4)		medical textile (n = 5)		clothing (n = 20)	
compounds	CL	DF	median	DF	median	DF	median	DF	median	DF	median	DF	median	DF	median	DF	median	DF	median
								Traditional QACs											
C8-BAC	S1	98	4.29	100	2.61	100	2.45	95	9.37	90	4.84	100	5.23	100	4.96	100	11.2	100	3.53
C10-BAC	S1	78	1.34	100	1.94	35	<MDL	75	1.09	50	0.243	85	2.22	100	39.5	100	3.20	100	1.42
C12-BAC	S1	67	17.6	70	13.4	35	<MDL	100	109	60	53.0	65	18.2	100	538	100	22.6	55	9.87
C14-BAC	S1	79	7.17	90	7.39	65	2.51	100	46.4	100	6.56	75	6.17	75	102	40	<MDL	65	3.57
C16-BAC	S1	76	1.15	100	5.97	75	0.934	80	4.62	100	1.32	50	0.0159	100	3.01	100	2.85	50	0.0154
C18-BAC	S1	58	0.745	100	14.4	55	0.310	55	0.181	90	5.05	25	<MDL	100	2.08	80	1.09	25	<MDL
ΣBAC		**100**	**38.6**	**100**	**53.6**	**100**	**12.0**	**100**	**213**	**100**	**70.2**	**100**	**33.7**	**100**	**690**	**100**	**43.5**	**100**	**31.0**
C8-DADMAC	S1	79	1.27	100	1.02	80	1.61	90	2.44	60	1.33	90	1.99	75	14.6	60	0.625	50	0.102
C10-DADMAC	S1	87	0.819	100	6.49	80	0.341	95	1.03	100	0.681	85	0.829	100	10.0	40	<MDL	80	0.232
C12-DADMAC	S1	31	<MDL	95	2.11	10	<MDL	10	<MDL	10	<MDL	20	<MDL	100	2.49	0	<MDL	25	<MDL
C14-DADMAC	S1	3	<MDL	10	<MDL	0	<MDL	0	<MDL	0	<MDL	0	<MDL	25	<MDL	0	<MDL	0	<MDL
C16-DADMAC	S1	5	<MDL	30	<MDL	0	<MDL	0	<MDL	0	<MDL	0	<MDL	0	<MDL	0	<MDL	0	<MDL
C18-DADMAC	S1	28	<MDL	65	170	5	<MDL	75	185	20	<MDL	10	<MDL	0	<MDL	0	<MDL	0	<MDL
ΣDADMAC		**94**	**5.98**	**100**	**192**	**100**	**3.03**	**100**	**188**	**100**	**2.85**	**95**	**3.25**	**100**	**29.3**	**60**	**0.625**	**80**	**0.675**
C8-ATMAC	S1	92	3.49	100	4.57	85	4.22	90	2.03	90	4.00	95	7.66	100	3.67	100	3.68	85	2.69
C10-ATMAC	S1	98	9.64	100	9.23	100	12.6	100	10.4	100	9.05	90	10.1	100	9.09	100	20.1	100	5.01
C12-ATMAC	S1	47	<MDL	30	<MDL	20	<MDL	55	29.0	30	<MDL	70	30.5	100	160	60	14.2	55	22.6
C14-ATMAC	S1	20	<MDL	5	<MDL	30	<MDL	20	<MDL	0	<MDL	20	<MDL	75	28.9	20	<MDL	25	<MDL
C16-ATMAC	S1	98	14.8	100	11.5	90	3.64	100	23.7	100	5.57	100	23.9	100	90.0	100	15.7	100	18.5
C18-ATMAC	S1	91	4.83	95	11.1	80	1.71	90	3.82	90	4.04	90	1.82	100	69.6	100	7.28	95	4.80
ΣATMAC		**100**	**55.9**	**100**	**41.7**	**100**	**30.1**	**100**	**76.2**	**100**	**30.2**	**100**	**94.1**	**100**	**378**	**100**	**106**	**100**	**68.4**
Σtraditional QACs		**100**	**176**	**100**	**287**	**100**	**48.9**	**100**	**626**	**100**	**111**	**100**	**195**	**100**	**1560**	**100**	**166**	**100**	**100**
								Emerging QACs											
AChCl	S1	23	<MDL	5	<MDL	35	<MDL	50	13.3	10	<MDL	30	<MDL	25	<MDL	0	<MDL	5	<MDL
BEC	S1	42	<MDL	85	1.36	10	<MDL	65	5.30	10	<MDL	45	<MDL	75	5.72	40	<MDL	15	<MDL
BTEAC	S1	74	21.4	100	25.6	45	<MDL	90	70.1	100	58.7	50	4.94	100	9.30	100	52.3	60	5.21
CPC	S1	41	<MDL	90	5.95	5	<MDL	20	<MDL	50	0.538	40	<MDL	50	3.47	60	1.48	40	<MDL
C2:4-DADMAC[a]	S2	93	43.4	100	20.3	95	114	90	95.1	100	57.2	100	54.6	75	20.4	60	10.2	90	22.2
C4:6-DADMAC[a]	S2	99	0.761	100	1.91	100	0.723	100	0.613	100	0.586	95	0.341	100	0.302	100	0.534	100	0.806
C6-DADMAC[a]	S2	100	6.33	100	12.3	100	7.54	100	4.20	100	3.37	100	7.31	100	3.90	100	6.89	100	3.24
C6:8-DADMAC[a]	S2	98	0.463	100	1.01	100	0.498	95	0.212	100	0.445	100	0.613	75	0.234	100	0.990	100	0.293
C8:10-DADMAC	S1	30	<MDL	40	<MDL	10	<MDL	45	<MDL	10	<MDL	30	<MDL	75	9.19	0	<MDL	35	<MDL
C10:12-DADMAC[a]	S2	24	<MDL	70	0.0459	10	<MDL	20	<MDL	50	0.00744	15	<MDL	0	<MDL	0	<MDL	0	<MDL
DB	S1	71	4.69	70	3.38	70	9.42	65	4.65	80	11.5	60	7.32	50	1.72	100	9.16	85	4.43
DDA	S1	60	12.2	15	<MDL	85	429	50	2.41	80	13.5	85	262	50	39.3	40	<MDL	60	21.09
EDDAB	S1	28	<MDL	5	<MDL	50	0.692	45	<MDL	0	<MDL	15	<MDL	100	47.0	20	<MDL	25	<MDL
MTOAC	S1	32	<MDL	75	1.55	25	<MDL	5	<MDL	10	<MDL	30	<MDL	75	1.13	0	<MDL	35	<MDL
Pen_4NCl	S1	22	<MDL	50	0.116	5	<MDL	15	<MDL	0	<MDL	35	<MDL	0	<MDL	20	<MDL	20	<MDL
TMABC	S1	13	<MDL	40	<MDL	0	<MDL	10	<MDL	0	<MDL	10	<MDL	25	<MDL	0	<MDL	15	<MDL
Σemerging QACs		**100**	**346**	**100**	**78.0**	**100**	**1460**	**100**	**1000**	**100**	**210**	**100**	**783**	**100**	**816**	**100**	**104**	**100**	**209**

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Table 1. continued

compounds	CL	total		mask (*n* = 20)		sock (*n* = 20)		towel (*n* = 20)		sportswear (*n* = 10)		underwear (*n* = 20)		quilt (*n* = 4)		medical textile (*n* = 5)		clothing (*n* = 20)	
		DF	median	DF	median	DF	median	DF	median	DF	median	DF	median	DF	median	DF	median	DF	median
Emerging QACs																			
Σtraditional + emerging QACs			**1050**		**624**		**3000**		**2610**		**533**		**6360**		**2340**		**271**		**345**

[a] Semiquantification was performed using calibration curves of structurally similar analogues, as commercial standards were not available. MDL: method detection limit; CL: confidence level adapted from Schymanski et al.[32]

spectrum antimicrobial activity, enhanced durability, and improved fiber adherence, which could explain their higher concentrations in textiles.[51,52] In parallel, traditional QACs such as BACs, DADMACs, and ATMACs (with C8−C18 alkyl chains) may be facing stricter regulations due to potential environmental and health concerns.[13] This regulatory pressure could drive manufacturers to adopt emerging QACs, such as shorter chain analogues designed to mitigate ecological impact, reduce environmental persistence, or lower toxicity.[18]

Variations of QACs across Textile Categories

Among the 8 textile categories (Figure 1 and Table 1), the highest total QAC concentrations (ΣQAC concentrations) were found in underwear with a median value of 6360 ng/g, followed by socks (3000 ng/g), towels (2610 ng/g), and quilts (2340 ng/g). The elevated ΣQAC concentrations in these textile categories correspond to the high antimicrobial demands imposed by their respective usage environments. Underwear and socks are worn daily in direct contact with skin and sweat, which support microbial growth and odor formation.[53] Towels are frequently used with moisture retained, creating favorable conditions for bacterial and fungal growth.[54,55] Quilts, which are in prolonged contact with the human body but are usually laundered less frequently, may accumulate sweat and skin debris, further promoting microbial colonization.[56,57] These usage-related factors provide a strong rationale for the extensive use of QACs in these textile products to impart enhanced antimicrobial functionality. Additionally, this result is consistent with our previous findings, where elevated QAC concentrations in indoor dust were attributed to unventilated microenvironments enriched with textile materials (i.e., cinemas with extensive carpets, curtains, and upholstered seats), which have heightened antimicrobial requirements.[31] This suggests that the presence of QACs in indoor dust could also be a result of their release

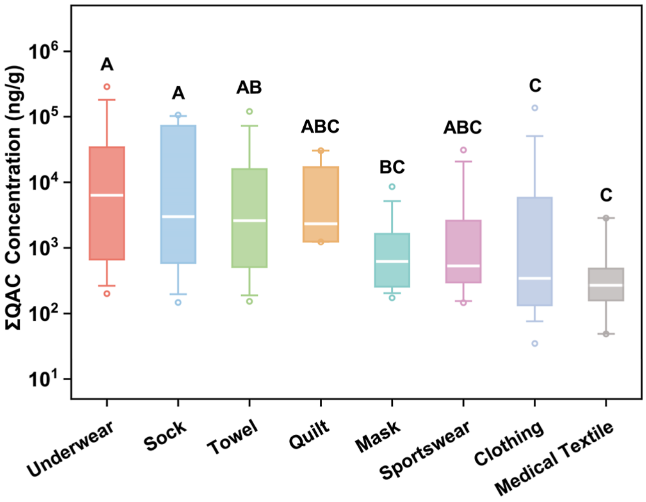

Figure 1. Concentrations of ΣQAC in different types of textiles (ng/g). Concentrations are shown as boxplots representing the 25th and 75th percentiles; white lines represent the median; the whiskers represent the 10th and 90th percentiles; and the dots represent the 5th and 95th percentiles. The letters represent the results of the Kruskal−Wallis test. Groups sharing the same letter are not significantly different, whereas groups with different letters differ significantly ($p < 0.05$); letters were assigned according to concentration levels, starting from the highest to the lowest.

from treated textile products, warranting further investigation into the potential human exposure pathways and associated health risks.

In contrast, lower QAC concentrations were observed in masks, sportswear, general clothing, and medical textiles, with median $\sum$QAC concentrations ranging from 271 to 624 ng/g. For masks, particle filtration generally takes precedence over antibacterial functionality in product design, which primarily relies on electret melt-blown polypropylene layers.[58] Surfactant-based coatings can dissipate the electret charge and thereby impair filtration efficiency, discouraging the application of high QAC loadings, although some specialized finishing techniques can enable limited incorporation of QACs.[59] Sportswear often achieves microbial growth control through moisture-management fabric constructions and functional finishing, where non-QAC biocides such as silver-based agents are commonly used as the main antimicrobial components.[60] Medical textiles, as mature healthcare protection products, generally have more established sterilization strategies and alternative non-QAC antimicrobial finishing technologies available.[61] For general clothing, antimicrobial demand is relatively low compared with textiles used in high-moisture or close-skin contact scenarios.

In light of QACs serving as active constituents in antimicrobial finishes for textiles, samples were grouped based on their labeled antibacterial and nonantibacterial functions for comparison of total QAC concentrations. Overall, the median $\sum$QAC concentration in antimicrobial textiles was 1.7 times higher than that in nonantimicrobial textiles (Figure S3A). When further stratified by textile category (underwear, towels, socks, sportswear, and clothing), a more pronounced pattern was observed (Figure S4): clothing and socks exhibited the largest median differences between antimicrobial and nonantimicrobial products at 6.0-fold ($p = 0.393$) and 5.8-fold ($p = 0.481$), respectively, followed by sportswear (2.1-fold, $p = 0.151$), towels (2.0-fold, $p = 0.218$), and underwear (1.6-fold, $p = 0.579$). While sample sizes likely contribute to the observed p-values, the overall trend still suggests that certain product types may require substantially higher QAC loadings when marketed with highlighted antibacterial functionality. In contrast, other products without antibacterial labeling may already contain background levels of QACs introduced through routine finishing processes such as fabric softening and antistatic treatment, thereby reducing the concentration difference between antibacterial and nonantibacterial items.[62]

Meanwhile, when the samples were grouped by fabric types (natural and synthetic), no significant differences in $\sum$QAC concentrations were found as well (Figure S3B), indicating that QAC use is not confined to specific fiber types but rather reflects the desired usage of textiles.

Beyond variations in $\sum$QAC concentrations among textile categories, distinct compositional patterns of QACs were also observed (Figure S5A). For traditional QACs, quilts and sportswear were dominated by BACs, particularly C12-BAC (contributing 50% and 55% of the traditional QAC fraction, respectively). Underwear, socks, and clothing contained higher contributions from ATMACs (contributing 68%, 73%, and 74% of the traditional QAC fraction, respectively), with C10-, C12-, and C16-ATMAC as major contributors. In contrast, masks and towels showed a distinct enrichment of long-chain DADMACs, especially C18-DADMAC (65% in masks and 43% in towels), which might be due to the specific product requirements for the soft texture of towels and masks, since DADMACs can serve as antistatic fabric softeners.[63] Emerging QAC profiles also varied among textile categories (Figure S5B): DDA accounted for 76−78% of emerging QACs in socks and underwear, whereas BTEAC together with short-chain DADMAC analogues (e.g., C2:4-DADMAC) dominated masks, towels, sportswear, and medical textiles (with BTEAC up to 64% in medical textiles and C2:4-DADMAC up to 49% in towels). In aggregate (Figure S5C), the contribution of emerging QACs to total QAC burdens differed substantially among textile categories, ranging from minor in quilts (12%) to predominant in socks (95%). These findings emphasize pronounced differences in the commercial QAC formulations applied across the textile market, further underscoring that the use of QACs in textile finishing is primarily driven by intended product type and functional demands.

Overall, the high concentrations and widespread occurrence of QACs in textiles strongly suggest that textile products serve as significant sources of QAC release. Except for direct contact during daily use, textiles are also subjected to repeated laundering, which can further promote the migration of QACs through mechanical actions and water immersion. Therefore, it is necessary to characterize the release behavior of QACs during laundering, which is critical for evaluating subsequent environmental risks.

Release of QACs from Textiles during Laundering

Mechanical action is a crucial driving force for the washing efficiency during textile laundry, as well as the main factor influencing the release of textile finishing chemicals.[64,65] To isolate the roles of other variables, a lab-controlled washing simulation was first performed on three skin-contact textile categories with high QAC contents to assess the effects of different factors except mechanical actions (i.e., textile types and washing temperature). After one hour of washing, extensive migration of QACs from the textile products was observed (Figure 2A), leading to a significant decrease in their residual concentrations in the fabrics. In general, over 76% of the textile samples across the three categories exhibited QAC migration rates exceeding 50%. No significant difference in QAC migration rates was observed between laundering in cool (20 °C) and warm water (50 °C), while all three textile types exhibited comparable QAC migration rates, indicating that the variations of temperature and fabric types do not accelerate the migration process. In addition, high QAC migration rates were detected in textile samples without antimicrobial labeling (65% to 91%), which is comparable to those of antimicrobial-functional textiles (37% to 78%). Thus, we hypothesize that the consistently high migration rates indicate that QACs in commercial textiles, whether used for antimicrobial purposes or other finishing applications, are commonly applied through relatively simple coating methods with limited mechanical stability, rather than through more sophisticated grafting techniques.[51,54]

To verify this release potential under conditions of real-life use, experiments of performing machine- and hand-washing, the two most representative mechanical actions in our daily laundering behavior, were conducted to analyze the concentrations of QACs released into laundry wastewater. As shown in Figure 2B, the average concentrations of $\sum$QAC released in hand-washing and machine-washing wastewater were 42.1 ng/mL and 22.4 ng/mL, respectively. This difference in measured concentrations is largely attributable to dilution due to the larger wastewater volume used by machine washing than hand

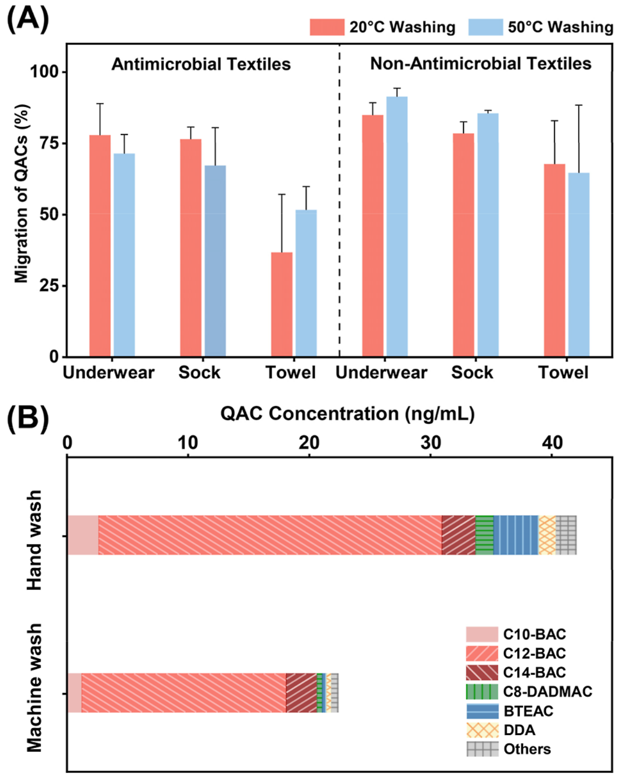

Figure 2. (A) Migration rates of QACs from three typical textile categories containing the highest QACs after lab-conditioned laundry experiments in cold water (20 °C) and warm water (50 °C). (B) Concentrations of QACs in laundry wastewater after simulation of hand-washing and machine-washing. Others include all other detected QACs with relative contribution <1%.

washing (21 L vs 12 L), whereas the total mass of QACs released was comparable between the two scenarios (i.e., 504 μg for hand washing vs 470 μg for machine washing). Similar patterns of QAC released were observed in wastewater from both washing methods, with C10-BAC, C12-BAC, C14-BAC, C8-DADMAC, BTEAC, and DDA as the dominant QAC congeners, together accounting for over 95% of the total QAC concentrations. Notably, C12-BAC was the most predominant compound, with concentrations of 28.3 ± 2.1 ng/mL in hand-washing wastewater and 16.9 ± 0.8 ng/mL in machine-washing wastewater, accounting for 67% and 75% of the total QAC concentrations, respectively. The predominance of C12-BAC likely reflects its high initial abundance in textile products and the use of simple antimicrobial application techniques (e.g., solution casting or spray-coating).[66] These results suggest that QACs in textiles are readily releasable and not strongly associated with the mechanical stress applied during laundering, which further supports the interpretation that a substantial fraction of QACs have not been chemically bonded to textile fabrics, thus facilitating their release into the aquatic environment.[67] Considering the widespread and frequent nature of domestic laundering activities, such emissions may represent a continuous source of QACs to aquatic environments. Moreover, because the QAC concentrations are influenced by the water volume used in different laundry methods, these scenario-related concentrations will be directly relevant to the subsequent toxicity and risk levels. Therefore, it is essential to evaluate the potential toxicity and ecological risks of QACs in laundry wastewater to aquatic organisms.

Toxicity of Laundry Wastewater to *D. magna* and Ecological Risk

The toxicity evaluation by WET using *D. magna* as the representative aquatic organism revealed that both the original concentration of hand-washing laundry wastewater (defined as 100% C_{lw}) and the standard solution containing equivalent QACs (defined as 100% C_{ss}) caused complete immobilization of *D. magna* (Figure 3). Notably, the standard solution exhibited a classical sigmoidal dose−response relationship, with an EC_{50} of 39.0% C_{ss}. In contrast, the dose curve of laundry wastewater showed a less inhibitory effect compared with the standard solution, with an EC_{50} of 84.3% C_{lw} (Figure 3). The reduced toxicity observed in laundry wastewater compared to the standard QAC solution might be due to the presence of other chemicals released from textiles, such as surfactants and bleaching agents, that could attenuate the toxic effects on *D. magna* through antagonistic interactions.[68,69] However, further investigation is needed to identify the specific components responsible for this phenomenon. These results confirm that QACs released during the laundry process can directly cause toxicity to aquatic organisms, highlighting the potential ecological risks associated with the discharge of laundry wastewater.

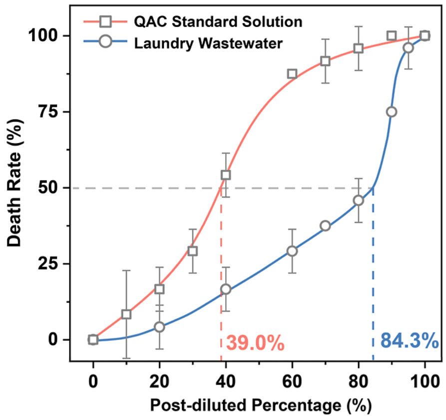

Figure 3. Concentration−response curves of the toxicity of gradient-diluted laundry wastewater (blue line) and standard solution containing equivalent QACs (red line) to *Daphnia magna* after 48 h exposure. Data indicate mean ± SD (n = 3).

Furthermore, two different scenarios were applied to evaluate the ecological risk of QACs from laundry wastewater, including WWTP-treated discharge and direct discharge. In the first scenario with treatment from WWTPs (Figure 4A and Table S5), 10 out of 150 regions exhibited high risk (RCR >1.0), with median RCR values ranging from 1.08 to 1267, primarily due to low dilution factors (medians 0.005 to 6). These regions, most located in Asia and Africa, are characterized by arid or semiarid climates with limited surface water resources, such as Saudi Arabia and Libya, which severely constrain the dilution capacity of receiving waters, thereby elevating risk levels.[70] Notably, most of these regions fall into the upper-middle income category,[71] indicating that economic development and advanced wastewater treatment infrastructures alone cannot offset the elevated environmental risks driven by unfavorable hydrological and climatic conditions. An additional 37 regions showed moderate risk ($0.1 \leq$ RCR < 1.0), with median RCR values ranging from 0.103 to 0.957. These regions were mainly distributed in

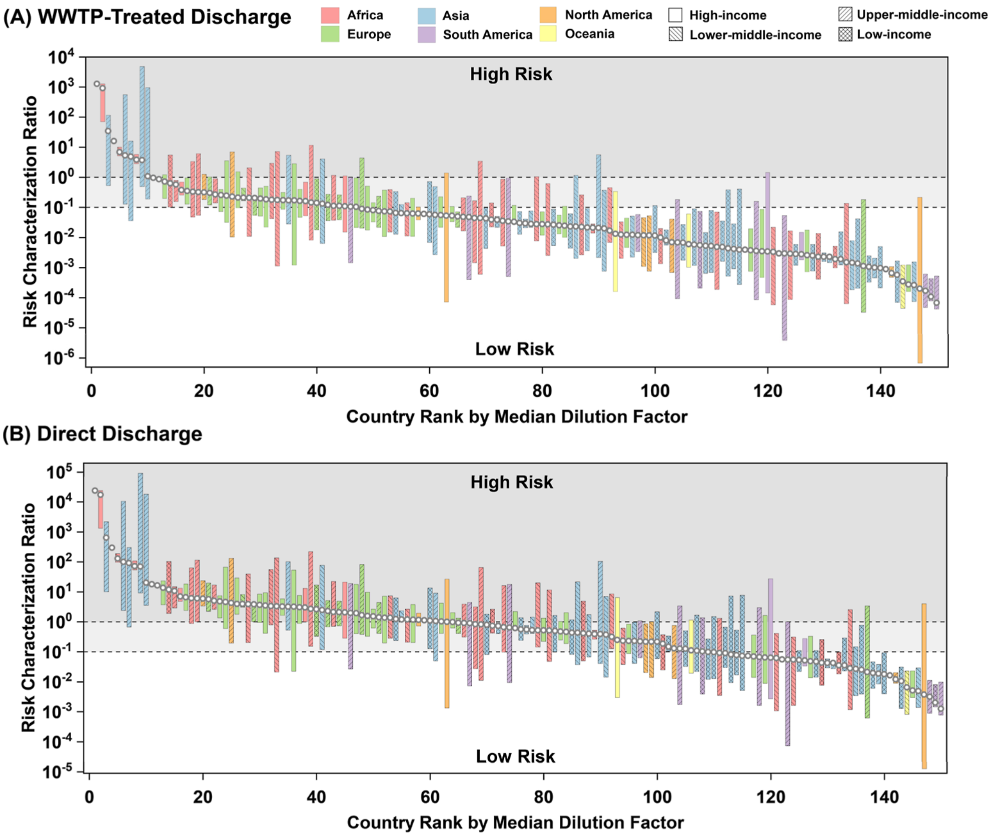

Figure 4. Risk characterization ratios (RCRs) of 150 countries for QACs released to rivers from the laundry process in the scenarios of (A) WWTP-treated discharge, and (B) direct discharge. Gray dots represent the median, and the floating columns represent the 5th and 95th percentiles.

Europe and Asia, with more than half classified as upper-middle-income or high-income economies. Although such countries are not located in arid regions and generally benefit from stronger dilution capacities and more developed wastewater treatment systems, the observed moderate risk values suggest that the high load of QACs in textiles and their release via laundry effluents remain a significant concern. In contrast, 103 regions showed low risk (RCR < 0.1), largely because abundant freshwater resources ensure exceptionally high dilution capacities that effectively attenuate QAC emissions.[38]

Alarmingly, under the scenario without WWTPs (Figure 4B and Table S6), the risk levels increased across all 150 regions due to the conservative assumption of no WWTP removal on QACs, resulting in 41% of the regions with median RCR values exceeding 1.0. In many cases, laundry effluents from hand-washing are discharged directly into receiving waters without treatment,[72] thereby amplifying the environmental release of QACs. For instance, in regions such as India and Bangladesh (median RCRs of 3.31 and 1.36, respectively), where hand-washing is a common practice,[73,74] the lack of adequate wastewater treatment facilities leads to the direct discharge of QAC-laden laundry wastewater into rivers and streams, posing significant risks to aquatic ecosystems.

■ ENVIRONMENTAL IMPLICATION

Our study has several limitations. First, the variability of QAC concentrations in laundry wastewater can be influenced by factors such as the type and amount of QAC-containing products used, as well as the specific washing conditions. Second, the laundering experiments in this study focused on a single wash cycle to evaluate the readily releasable fraction of QACs during the initial stage of laundering. Future work should incorporate sequential wash cycles to better simulate the actual lifecycle of textiles and quantify cumulative QAC emissions over repeated laundering, along with the changes of residual QAC burdens in fabrics, to assess their claimed durable functionality that justifies their essential use. Additionally, the dilution factors used in the PEC calculations are based on published literature and may not always accurately represent the actual dilution capacities of receiving waters in different regions. Another source of uncertainty lies in the use of *D. magna* as the sole test organism for evaluating the acute toxicity of QACs in wash water. While *D. magna* is a widely accepted model species for ecotoxicological studies, the results may not generalize across other sensitive aquatic species or adequately reflect chronic toxicity or ecosystem-wide effects. Incorporating additional test organisms and extending the exposure periods in future studies would provide a more comprehensive assessment of the ecological risks associated with QACs in laundry wastewater.

Nonetheless, this study provides the first comprehensive evidence that both traditional and emerging QACs are widely present in everyday textiles and can be released in substantial amounts during laundering. Additionally, QACs were detected in textiles without antibacterial labeling, suggesting that routine finishing processes (e.g., softening and antistatic treatments) may introduce background QAC levels and broaden consumer exposure beyond explicitly antimicrobial products, highlighting the widespread use of QACs in the textile market. Risk characterization ratio modeling revealed that laundry-derived QACs pose moderate to high ecological risks in many regions, especially where limited water resources or insufficient wastewater treatment reduce dilution capacity. These findings indicate that textiles represent a previously underestimated pathway for QAC release into aquatic systems, contributing to mixture effects alongside other wastewater contaminants. In addition to the contamination pathway via wastewater emissions, the findings of this study imply a potential indoor exposure route whereby elevated QAC concentrations in indoor dust may arise not only from the widespread use of QAC-containing disinfectant products, but also from QAC-treated textiles. Given that indoor dust contains a substantial fraction of textile fibers,[75,76] further studies are warranted to quantify the contribution of QAC-treated textiles to indoor dust burdens and to evaluate the associated indoor exposure.

Since there is a high potential for QAC release during actual laundering activities, their antimicrobial coatings appear short-lived and may not provide the durable protection that consumers expect. This raises questions about whether QAC-treated textiles can maintain long-term antimicrobial efficacy or effectively prevent microbial growth as intended. Meanwhile, leached QACs enter laundry wastewater, WWTPs, and ultimately the environment, leading to continuous, low-level human exposures and raising ecotoxicological and antimicrobial resistance concerns. In this sense, the QAC case may give consumers a false sense of hygiene protection. A chemical additive should be used only when it demonstrably provides a durable benefit under realistic conditions, and when that benefit outweighs potential health and environmental risks. Therefore, it is essential to critically evaluate whether such chemical additions are truly necessary or merely create a false sense of safety at the expense of long-term well-being and environmental integrity.

■ ASSOCIATED CONTENT

Data Availability Statement

A version of this paper prior to peer review is available on ChemRxiv.[77]

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c15617.

> Detailed information on chemicals and reagents, textile sample information, matrix spike recoveries of analytes, method detection limits of analytes and blank concentrations, estimated risk characterization ratios of 150 countries, chemical structures, chromatograms and MS^2 spectra of representative emerging QACs, and total QAC concentration in samples grouped by labeled antimicrobial functions/materials (PDF)

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Notes

The authors declare no competing financial interest.

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