Environmental and human health risk assessment of antibiotic residues in drinking water sources: case study of a fast-developing megacity in southern China

Qiao Zhang, Sheng Yang, Bangmi Xie, Jian Zhang, Chen Deng and Rong Hu

ABSTRACT

The occurrence, distribution, ecological and health risk assessment of five antibiotics (CFM, ERY, LCM, SMD and SMX) in 32 drinking water reservoirs of a megacity (Shenzhen) in Guangdong–Hong Kong–Macao Greater Bay Area (GBA), were investigated. Among the above antibiotics, CFM was not detected, ERY, SMD, LCM and SMX were detected in 65.6%, 46.9%, 43.7% and 40.6% of samples, with concentrations of 1.594–7.605 ng·L^{-1}, 0.335–6.695 ng·L^{-1}, 0.288–7.878 ng·L^{-1} and 0.770–5.355 ng·L^{-1}, respectively. The maximum concentration of ERY was detected in GT Reservoir, and those of the other 3 antibiotics were detected in SZ Reservoir. The ecological risk quotient (*RQ*) values for SMX in SZ, XL, YT, EJ, SY, TG, XK, GK, MK and GT reservoirs ranged from 0.1 to 1.0, indicating a median risk to aquatic organisms, the others posed insignificant risk or low risk. Considering the joint effects of detected antibiotics, the highest overall *RQ* value was 0.38, obtained in SZ Reservoir, in other words, it was exposed to greater risk. Based on health risk assessment, the health *RQ* values for the detected antibiotics were significantly lower than 0.01, indicating insignificant risk to human health. These findings could provide a scientific basis for the government to ensure the drinking water safety of a megacity in GBA.

Key words | antibiotics, distribution characteristics, drinking water source, ecological risk assessment, health risk assessment, risk quotients

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INTRODUCTION

The extensive use of antibiotics has caused growing international concern over their fate after their consumption and excretion in feces, and has been recognized as one of the main environmental issues in the 21st century (Zou *et al.* 2011; Yuan *et al.* 2013). Generally, antibiotics from domestic sources and manufacturers are transported to wastewater treatment plants (WWTPs), but are poorly eliminated due to their maintenance of bacterial resistance. The residues are then discharged into the ambient environment representing a point source in effluents (Xu *et al.* 2014; Li *et al.* 2019). This new contaminant can be introduced into different environmental matrices due to their variable physicochemical properties, and the types and concentrations in the environment vary among areas (Kolpin *et al.* 2002; Zou *et al.* 2011).

China is the biggest manufacturer and consumer of antibiotics, with an annual production of about two hundred and ten thousand tons, of which 85% is utilized in veterinary medicine in livestock and poultry farming. This could result in more serious antibiotic pollution in the aquatic environment than in other countries (Zou *et al.* 2011; Guo *et al.* 2016; Feng *et al.* 2019). The potential contamination of drinking water supplies due to the increased presence of antibiotics is also of concern, ascribed to the propensity of antibiotics to facilitate the establishment and

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amplification of pathogenic reservoirs that threaten public health (Rysz & Alvarez 2004; Pruden *et al.* 2006).

Shenzhen, located in the southeast of China, is an area which has seen very rapid economic development and urbanization over recent decades, and, where reservoir water is the main source of drinking, agricultural and industrial water. The five largest reservoirs (TG, SY, XL, YT and SZ reservoirs) account for over 80% of the water column of surface water (Wang *et al.* 2004). Most reservoirs accumulate the runoff from rainwater, such as TG and SY Reservoirs, some reservoirs collect water from rivers with long distance drainage, for instance, the main water supply of SZ Reservoir is from the Dong-jiang River. With local industrial developments and population increases, the reservoir ecosystems and water quality have been affected by pollutants. It has been reported that several antibiotic residues (metronidazole, cefalexin, lincomycin, sulfadimidine, sulfamethoxazole, erythromycin-H_2O, roxithromycin and cefuroxime) were detected in SY and XL Reservoirs with concentrations from 1.1 to 130 ng·L^{-1} (Zhu *et al.* 2013, 2014).

In the future, more highly quality water will be needed in Shenzhen, therefore we need to have a clear picture of antibiotics in drinking water sources, and their potential ecological and health risks in a city with such a large population, and pay more attention to the huge annual consumption of pharmaceuticals. Yet the occurrence of antibiotics in drinking water sources in Shenzhen at the regional scale are not fully understood, and limited data exist on the related environmental and health risks they may pose. The objectives of this paper were to investigate the occurrence of five selected antibiotics (lincomycin (LCM); cefuroxime (CFM); erythromycin (ERY); sulfamethoxazole (SMX); sulfadimidine (SMD)) in 32 drinking water reservoirs across Shenzhen using liquid chromatography-electrospray ionization-tandem mass spectrometer (LC-MS/MS) coupled with solid-phase extraction (SPE) for quantification, and further assess the potential risks to the ecosystem and human health through the calculation of risk quotients (*RQ*).

MATERIALS AND METHODS

Chemicals and reagents

Standards of LCM, CFM, ERY, SMX and SMD were purchased from ANPEL Laboratory Technologies (Shanghai, China) Inc., and their structures and chemical properties are shown in Figure 1. Unless otherwise indicated, chemicals

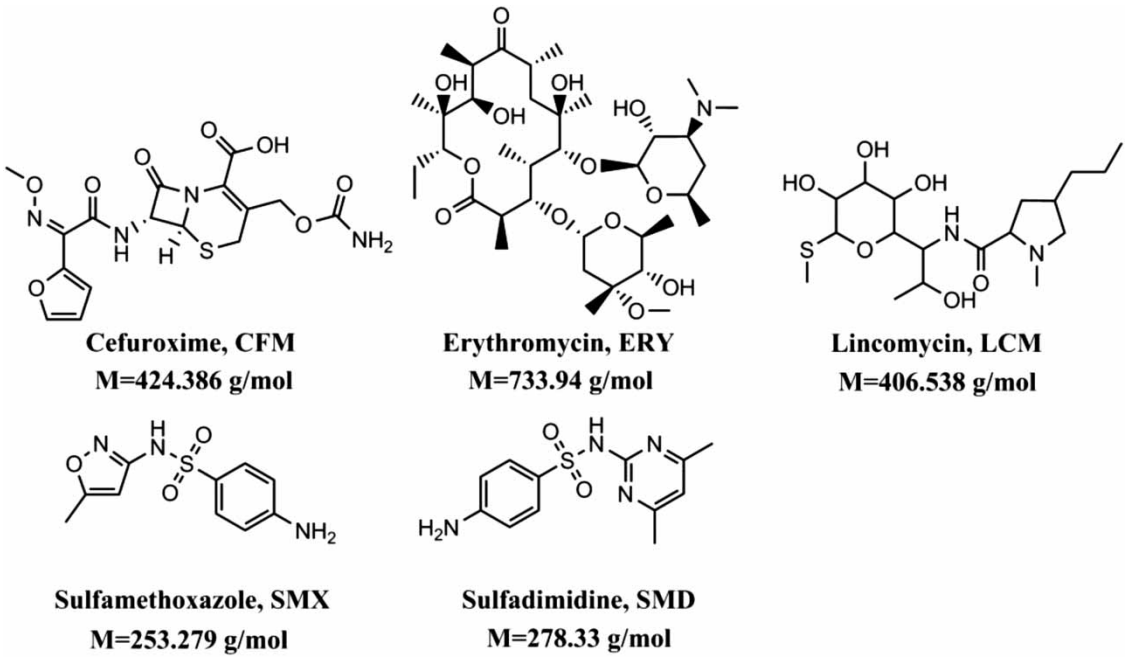

Figure 1 | Chemical structures of the antibiotics investigated.

used in the analysis were purer than the analytical grade. All reagents of high-performance liquid chromatography (HPLC) grade used for sample processing and analysis (methanol, toluene and dichloromethane) were obtained from Merck Chemicals (Shanghai, China) Co., Ltd. De-ionized water was prepared with a Milli-Q water purification system (Direct 8, Millipore, USA).

The cartridges used for SPE were Oasis HLB cartridges (N-vinylpyrrolidone-m-divinylbenzene copolymer, 6 cc/500 mg) obtained from Agilent (California, USA). Glass microfiber filters (GF/F, pore size 0.7 μm) were obtained from Whatman (Maidstone, UK).

Sampling sites and sample collection

Thirty-two sampling points covering all the drinking water conservation areas in Shenzhen were established to investigate the occurrence of the selected antibiotics (Figure 2). At each sampling site, three subsamples were collected from the top 0.5 to 1.0 m of the surface water and then were mixed together to obtain the composite samples. Each sample was placed into a 5 L clean brown glass bottle pre-rinsed with reservoir water three times, and immediately chilled in an icebox, transported to the laboratory and stored in the dark at 4 °C until filtration and extraction.

Sample preparation and analysis

A water sample of 1,500 mL was filtered through glass microfiber filters, and 500 mL of the filtrate was transferred to a 500 mL-polypropylene bottle. The pH value was adjusted to 3.2 using oxalic acid (5 mol·L^{-1}). Na_2EDTA (0.05 g), sodium azide (0.1 g) and 30 ng of surrogate standards (LCM-d3 and CFM-d3) as the internal standards were added to the solution.

The method for water sample extraction used in this study were according to the method described in previous studies (Song *et al.* 2015). Briefly, all the water samples were extracted with the SPE method (AutoTrace280, Thermo, USA) by using Waters Oasis HLB cartridges loaded at a flow rate of 10 mL/min, which were

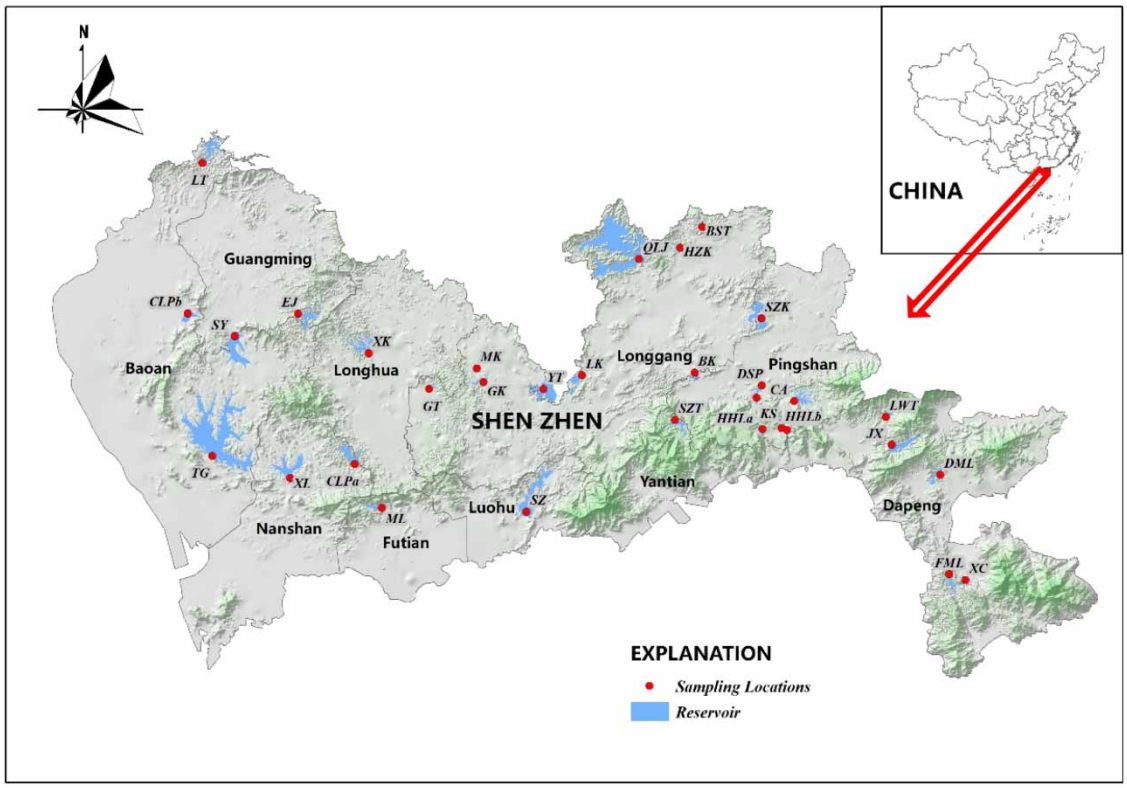

Figure 2 | Map of the sampling sites in Shenzhen city, Guangdong province.

preconditioned sequentially with 4 mL methanol, and 4 mL water. Then the cartridges were eluted with 4 mL methanol, and the extracts were concentrated and dried under a gentle nitrogen stream using a termovap sample concentrator (N-EVAP, Organomation, USA), the extracts were redissolved in 0.5 mL methanol and transferred into auto sampling vials. The extracted antibiotics were analyzed using the LC-MS/MS system (Agilent 1260–6460, USA) with optimized conditions described in a previous study (Li *et al.* 2009).

Risk evaluation

An effort was made to assess the ecological and health risks of the antibiotics measured.

(1) Ecological Risk Assessment (ERA)

For an ecological risk assessment of the detected antibiotics, the risk quotient (RQ_E) method was adopted, which was calculated as the following formula: RQ_E = MEC/PNEC, where MEC is the measured environmental concentration, and PNEC is the predictive non-effect concentration in water. No observed effect concentration (NOEC) is normally the first choice to calculate PNEC by dividing by a factor of 100, but if there is a lack of NOEC values, EC_{50} or LC_{50} values from standard ecotoxicological tests can be used (PNEC = L(E)C_{50}/AF), eventually after correction by an appropriate standard assessment factor of 1,000 to overcome the uncertainty from the extrapolation from single species toxicity to ecosystem toxicity (Park & Choi 2008). The PNECs of the selected antibiotics (CFM, ERY, LCM, SMX, and SMD) used in this study collected from the literature were 76,000, 2,000, 70, 26.8 and 1,740 ng·L^{-1}, respectively (Agerstrand & Rudén 2010; Ginebreda *et al.* 2010; Białk-Bielińska *et al.* 2011). Commonly, the RQ_E values were classified into four risk levels: ≤0.01, 0.01–0.1, 0.1–1 and ≥1, indicating insignificant risk, low risk, median risk, and high risk, respectively (Huang *et al.* 2019).

Even if the levels of single antibiotics in the aquatic environment are lower than the safety criteria, the ecological risk of antibiotics concurrently exposed as a mixture may threaten the aquatic ecosystem (Cleuvers 2004). While previous efforts focused on the environmental risk of single antibiotics, recent attention has shifted to evaluating the overall risk for the antibiotic mixtures. In this study, the RQ_E of each antibiotic was aggregated by simple addition to assess the overall ecological risk (Quinn *et al.* 2008).

(2) Health Risk Assessment (HRA)

The evaluation of the potential risk of antibiotics in source water to humans is presented by calculating the RQ for human health based on the following formula (Salgot *et al.* 2006): $RQ_H = EC \times IngR_{DW} \times k_T \times EF \times ED/(ADI \times BW \times AT)$, where RQ_H is quotient risk for human health, EC is the concentration of antibiotics in source water (μg/L), k_T is the proportion of antibiotics left after the drinking water treatment process, $IngR_{DW}$ is the child or adult drinking water ingestion rate (L/person-day), EF is the exposure frequency (days/year), ED is the exposure duration (years), ADI is acceptable daily intakes (μg/kg-day), AT is the average time (days), BW is the child or adult body weight (kg/person).

The ADI values of the five antibiotics were obtained from the literature, as summarized in Table 1 (Zhu *et al.* 2013; Tang *et al.* 2018). The other human exposure parameters used in the above equations were recommended by the US Environmental Protection Agency (US EPA) guidance to derive the Ambient Water Quality Criteria (AWQC) (Zhu *et al.* 2014), as presented in Table 2. The

Table 1 | ADI of five antibiotics (μg/kg-d)

Compounds	Acronym	Adult	Child
Cefuroxime	CFM	8.00	29.0
Erythromycin	ERY	40.0	90.0
Lincomycin	LCM	25.0	56.8
Sulfamethoxazole	SMX	130	295
Sulfadimidine	SMD	50.0	114

Table 2 | Parameters relating to adult and child receptors recommended by US EPA

		Receptor	
Parameter (Acronym)	Units	Adult	Child
EF	days/year	350	350
ED	years	30	6
AT	days	10,950	2,190
BW	kg	70	14
$IngR_{DW}$	L/person-day	2	1
k_T	–	1	1

RQ_H values were classified into four risk levels: ≤0.01, 0.01–0.1, 0.1–1 and ≥ 1, indicating insignificant risk, low risk, median risk, and high risk, respectively (Feng *et al.* 2019).

RESULTS AND DISCUSSION

Antibiotics distribution in reservoirs

Through investigation in this study (Figure 3), CFM was not detected in the 32 drinking water reservoirs, while the other target antibiotics (ERY, SMX, SMD and LCM) were widely detected at the ng·L^{-1} level with frequencies of 65.6%, 46.9%, 40.6% and 43.7%, respectively. It is worth noting that the four antibiotics were synchronously detected in 12 reservoirs (SZ, XL, YT, CLPa, SZK, CLPb, EJ, SY, TG, XK, GK, MK Reservoirs), accounting for about 37.5% of the samples, yet not detected in 10 reservoirs (DML, JX, DSP, KS, HHLb, QLJ, BST, HZK, BK, LK Reservoirs), accounting for 31.3%. In contrast to other sampling sites, SZ Reservoir was the most polluted by antibiotics with concentrations of 3.747 ng/L ERY, 5.355 ng/L LCM, 7.878 ng/L SMX, and 6.695 ng/L SMD, and of these the concentrations of SMX, SMD and LCM were the highest, the ERY concentration ranked second only to GT Reservoir.

It is recognized that SZ, SZK, XL, TG, SY, XK, EJ and LK Reservoirs are mainly supplied with water from rivers with long distance drainage. Apart from LK Reservoir, the ERY, LCM, SMX, and SMD were clearly detected in these reservoirs, and this is ascribed to the high concentration of antibiotics detected in the main source of water from Shawan tunnel and the Eastern channel of Xili (Table S1, Supplementary Material). A similar conclusion was confirmed in our previous investigation, the antibiotics introduced by Dongjiang River into SZ Reservoir and East water supply project into XL Reservoir accounted for above 96% in both dry and wet seasons. Additionally, antibiotics in CLPb, GK, MK and GT Reservoirs, depending on the water produced locally, were also detected.

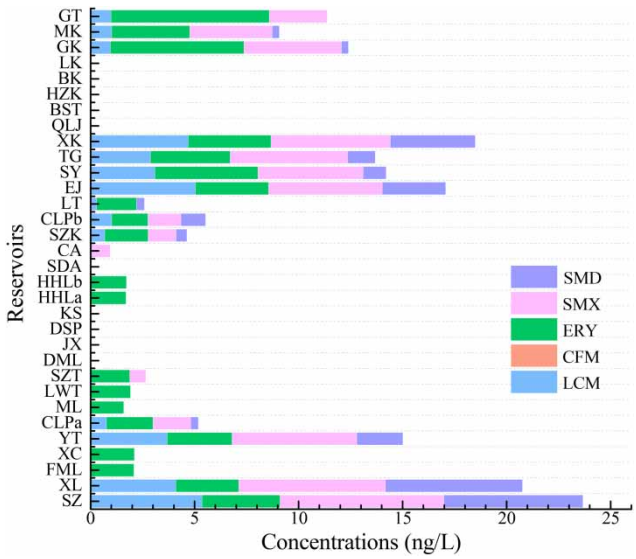

Figure 3 | Antibiotic concentrations (ng/L) in the water samples from the thirty-two reservoirs.

ERA of antibiotics in drinking water sources

It is known that antibiotic residues in the aquatic environment are of special concern principally because of their potential to accelerate widespread bacterial resistance and to have a negative influence on some key bacterial species in the ecosystem (Cleuvers 2004). In this case, previous efforts have mainly focused on this goal and much less attention has been paid to other possible effects, which has resulted in significant ecotoxicological uncertainties. Usually, the purpose of environmental risk assessment is to form the basis of decisions to decrease or otherwise manage the risks. In general, reliable knowledge about exposures and the toxicity of the target chemical is vital to establish a reasonable environmental risk assessment of the substance. In order to estimate the ecological risk posed by certain contaminants in aquatic ecosystems, the WFD (Water Framework Directive) states the convenience of assessment using taxa of three different representative trophic levels of the ecosystem, such as algae, daphnids and fish.

In this study, multi-antibiotics were found in drinking water sources, which probably pose a potential risk to aquatic organisms. The ecological risk of the target antibiotics was assessed on the basis of *RQ*. The ERA evaluation of antibiotics in drinking water sources was conducted, and the single risk quotient (RQ_i) combined with the overall ecological risks (RQ_m ($\sum RQ_i$)) of antibiotics are presented in Table 3. The *RQ* values of CFM, ERY and SMD in 32 reservoirs were below 0.01, as stated above, which is indicative of insignificant risk. The *RQ* value of LCM in 12 reservoirs (SZ, XL, YT, CLPa, CLPb, EJ, SY, TG, XK, GK, MK, GT)

Table 3 | The risk quotient (RQ_i) of single antibiotic and the overall risk quotient (RQ_m) of antibiotic mixtures in sampling reservoirs

Sampling location	RQ_i (MEC/PNEC ratio)					RQ_m ($\sum RQ_i$)
	CFM	ERY	LCM	SMX	SMD	
SZ	0	0.00187	**0.0765**	**0.294**	**0.00385**	**0.376**
XL	0	0.00152	0.0585	**0.263**	0.00379	**0.327**
FML	0	0.00104	0	0	0	0.00104
XC	0	0.00105	0	0	0	0.00105
YT	0	0.00156	0.0526	**0.224**	0.00127	**0.279**
CLPa	0	0.00112	0.0108	0.0678	0.000210	0.0799
ML	0	0.000797	0	0	0	0.000800
LWT	0	0.000956	0	0	0	0.000960
SZT	0	0.000939	0	0.0287	0	0.0297
DML	0	0	0	0	0	0
JX	0	0	0	0	0	0
DSP	0	0	0	0	0	0
KS	0	0	0	0	0	0
SDA	0	0.000850	0	0	0	0.000850
HHLa	0	0.000861	0	0	0	0.000860
HHLb	0	0	0	0	0	0
CA	0	0	0	0.0351	0	0.0351
SZK	0	0.00104	0.00981	0.0502	0.000301	0.0613
CLPb	0	0.000863	0.0146	0.0596	0.000678	0.0758
LT	0	0.000956	0.00411	0	0.000220	0.00528
EJ	0	0.00176	**0.0720**	**0.204**	0.00175	**0.279**
SY	0	0.00248	0.0441	**0.189**	0.000639	**0.236**
TG	0	0.00192	0.0410	**0.211**	0.000757	**0.254**
XK	0	0.00199	0.0670	**0.214**	0.00234	**0.286**
QLJ	0	0	0	0	0	0
BST	0	0	0	0	0	0
HZK	0	0	0	0	0	0
BK	0	0	0	0	0	0
LK	0	0	0	0	0	0
GK	0	**0.00321**	0.0137	**0.175**	0.000192	**0.192**
MK	0	0.00188	0.0146	**0.148**	0.000200	**0.164**
GT	0	**0.00380**	0.0141	**0.104**	0	**0.122**
Maximum	0	0.00380	0.0765	0.294	0.00385	0.376

Lightest shade: insignificant risk; medium shade: low risk; darkest shade: median risk.

ranged between 0.01 and 0.1, indicating a low ecological risk, the other reservoirs showed an insignificant risk. As for SMX, the *RQ* value ranged from 0 to 0.294, 31.3% of the sampling reservoirs (SZ, XL, YT, EJ, SY, TG, XK, GK, MK, GT) were between 0.1 and 1.0, indicating a median risk, and 15.6% of reservoirs (CLPa, SZT, CA, SZK, CLPb) with *RQ* values in the range 0.01–0.1, showed a low risk.

As there is usually more than one compound present in water, their combined effects must be considered. The RQ_m of the antibiotics involved was summarized following a

Table 4 | Health risk of antibiotic residues in thirty-two reservoirs to adults and children

Sampling location	RQ_H (Adults)						RQ_H (Children)					
	CFM	ERY	LCM	SMX	SMD	$\sum RQ_i$	CFM	ERY	LCM	SMX	SMD	$\sum RQ_i$
SZ		2.567×10^{-6}	5.869×10^{-6}	$\mathbf{1.660\times10^{-6}}$	$\mathbf{3.668\times10^{-6}}$	$\mathbf{1.376\times10^{-5}}$		2.852×10^{-6}	$\mathbf{6.458\times10^{-6}}$	$\mathbf{1.829\times10^{-6}}$	$\mathbf{4.022\times10^{-6}}$	$\mathbf{1.516\times10^{-5}}$
XL		2.076×10^{-6}	4.489×10^{-6}	1.484×10^{-6}	3.612×10^{-6}	1.166×10^{-5}		2.306×10^{-6}	4.940×10^{-6}	1.635×10^{-6}	3.960×10^{-6}	$\mathbf{1.284\times10^{-5}}$
FML		1.428×10^{-6}				1.428×10^{-6}		1.587×10^{-6}				1.587×10^{-6}
XC		1.438×10^{-6}				1.438×10^{-6}		1.597×10^{-6}				1.597×10^{-6}
YT		2.138×10^{-6}	4.035×10^{-6}	1.263×10^{-6}	1.213×10^{-6}	8.650×10^{-6}		2.376×10^{-6}	4.440×10^{-6}	1.392×10^{-6}	1.330×10^{-6}	9.538×10^{-6}
CLPa		1.535×10^{-6}	$\mathbf{8.291\times10^{-6}}$	3.828×10^{-7}	1.998×10^{-7}	2.947×10^{-6}		1.706×10^{-6}	9.123×10^{-7}	4.218×10^{-7}	2.191×10^{-7}	3.259×10^{-6}
ML		1.092×10^{-6}				1.092×10^{-6}		1.213×10^{-6}				1.213×10^{-6}
LWT		1.310×10^{-6}				1.310×10^{-6}		1.455×10^{-6}				1.455×10^{-6}
SZT		1.286×10^{-6}		1.622×10^{-7}		1.448×10^{-6}		1.429×10^{-6}		1.787×10^{-7}		1.607×10^{-6}
DML												
JX												
DSP												
KS												
SDA		1.16474×10^{-6}				1.165×10^{-6}		1.294×10^{-6}				1.294×10^{-6}
HHLa		1.17893×10^{-6}				1.179×10^{-6}		1.310×10^{-6}				1.310×10^{-6}
HHLb												
CA				1.985×10^{-7}		1.985×10^{-7}				2.187×10^{-7}		2.187×10^{-7}
SZK		1.424×10^{-6}	7.524×10^{-7}	2.834×10^{-7}	2.868×10^{-7}	2.747×10^{-6}		1.582×10^{-6}	8.279×10^{-7}	3.122×10^{-7}	3.145×10^{-7}	3.037×10^{-6}
CLPb		1.182×10^{-6}	1.120×10^{-6}	3.368×10^{-7}	6.462×10^{-7}	3.285×10^{-6}		1.313×10^{-6}	1.232×10^{-6}	3.711×10^{-7}	7.085×10^{-7}	3.625×10^{-6}
LT		1.310×10^{-6}	3.151×10^{-7}		2.098×10^{-7}	1.835×10^{-6}		1.456×10^{-6}	3.467×10^{-7}		2.301×10^{-7}	2.033×10^{-6}
EJ		2.412×10^{-6}	5.521×10^{-6}	1.151×10^{-6}	1.670×10^{-6}	1.075×10^{-5}		2.680×10^{-6}	6.076×10^{-6}	1.268×10^{-6}	1.831×10^{-6}	$\mathbf{1.185\times10^{-5}}$
SY		3.395×10^{-6}	3.383×10^{-6}	1.066×10^{-6}	6.090×10^{-7}	8.453×10^{-6}		3.77×10^{-6}	3.722×10^{-6}	1.174×10^{-6}	6.678×10^{-7}	9.337×10^{-6}
TG		2.633×10^{-6}	3.148×10^{-6}	1.1909×10^{-6}	7.214×10^{-7}	7.693×10^{-6}		2.925×10^{-6}	3.464×10^{-6}	1.311×10^{-6}	7.910×10^{-7}	8.492×10^{-6}
XK		2.727×10^{-6}	5.139×10^{-6}	1.2109×10^{-6}	2.234×10^{-6}	$\mathbf{1.131\times10^{-5}}$		3.030×10^{-6}	5.655×10^{-6}	1.333×10^{-6}	2.449×10^{-6}	$\mathbf{1.247\times10^{-5}}$
QLJ												
BST												
HZK												
BK												
LK												
GK		4.391×10^{-6}	1.049×10^{-6}	9.882×10^{-7}	1.833×10^{-7}	6.611×10^{-6}		4.879×10^{-6}	1.153×10^{-6}	1.089×10^{-6}	2.010×10^{-7}	7.322×10^{-6}
MK		2.569×10^{-6}	1.117×10^{-6}	8.343×10^{-7}	1.909×10^{-7}	4.711×10^{-6}		2.854×10^{-6}	1.229×10^{-6}	9.191×10^{-7}	2.094×10^{-7}	5.211×10^{-6}
GT		$\mathbf{5.209\times10^{-6}}$	1.084×10^{-6}	5.859×10^{-7}		6.879×10^{-6}		$\mathbf{5.788\times10^{-6}}$	1.192×10^{-6}	6.454×10^{-7}		7.626×10^{-6}
Maximum		$\mathbf{5.209\times10^{-6}}$	$\mathbf{8.291\times10^{-6}}$	$\mathbf{1.660\times10^{-6}}$	$\mathbf{3.668\times10^{-6}}$	$\mathbf{1.376\times10^{-5}}$		$\mathbf{5.788\times10^{-6}}$	$\mathbf{6.458\times10^{-6}}$	$\mathbf{1.829\times10^{-6}}$	$\mathbf{4.022\times10^{-6}}$	$\mathbf{1.516\times10^{-5}}$

simple additive model. Ten reservoirs were exposed to median risk in the following order: GT < MK < GK < SY < TG < EJ = YT < XK < XL < SZ, with RQ_m values ranging from 0.122 to 0.376. It can be assumed that SZ Reservoir was exposed to greater ecological risk by antibiotics.

HRA of antibiotics in drinking water sources

The HRA evaluation of antibiotic residues in thirty-two reservoirs to adults and children is presented in Table 4. The health risk quotients of the target antibiotics, except CFM, in thirty-two sampling sites (including adults and children) were in the 10^{-7}–10^{-6} order of magnitude, much less than 0.01, indicating an insignificant risk to human health in these reservoirs. It may be worthwhile to mention that the maximum RQ_H values of ERY, LCM, SMX, and SMD for adults were obtained in GT, CLPa, SZ and SZ Reservoirs, respectively, which was slightly different to the results for children with a maximum of LCM in SZ Reservoir.

The overall risk of antibiotics in several reservoirs reached up to 10^{-5} orders of magnitude. For example, the overall risk in SZ and XK to adults was 1.376×10^{-5} and 1.131×10^{-5}, which in SZ, XL, EJ, and XK to children was 1.516×10^{-5}, 1.284×10^{-5}, 1.185×10^{-5} and 1.247×10^{-5}, respectively. Apparently, the peak value for both adults and children was attained in SZ Reservoir, in other words, SZ Reservoir presents a larger health risk compared with the other reservoirs. However the values induced by the antibiotics in the investigated drinking water sources were considerably lower than 0.01, which indicates the antibiotic residues did not pose a significant health hazard to humans, which is in accordance with the research published by Zhu *et al.* (2014).

It should be noted that the trace residual antibiotics in source water will be partially removed after the drinking water treatment process, yet they can't be removed completely (de Jesus Gaffney *et al.* 2015). The antibiotics enter the human body via drinking water, especially for school-age children, and long-term exposure to low-dose antibiotics is a risk factor for obesity (Wang *et al.* 2016). At present, a wide variety of residual antibiotics exist in source water, their potential comprehensive risks and long-term risks deserve further attention.

CONCLUSIONS

With the exception of CFM, the other four antibiotics (SMD, SMX, LCM and ERY) were detected in 46.9%, 40.6%, 43.7% and 65.6% of samples, respectively. The maximum concentration of ERY was detected in GT Reservoir, the other three were in SZ Reservoir. The CFM, ERY, LCM and SMD in reservoirs posed a low risk, yet the SMX in SZ, XL, YT, EJ, SY, TG, XK, GK, MK and GT Reservoirs posed a median ecological risk. Taking the joint effects of the antibiotics involved into consideration, the above ten reservoirs also posed a moderate risk with RQ_m 0.1– < 1.0, notably a greater ecological risk that the SZ Reservoir was exposed to. Trace levels of residual antibiotics detected in the drinking water sources did not pose a significant risk to human health.

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SUPPLEMENTARY MATERIAL

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