



Baseline

Occurrence and risk assessment of antibiotics in the Xi'an section of the Weihe River, northwestern China

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ABSTRACT

Fifteen antibiotics, including seven sulfonamides (SAs); three macrolides (MLs); three quinolones (QNs); one lincosamide, lincomycin (LIN); and one tetracycline (TC), were detected in the surface water of the Xi'an section of the Weihe River by using high-performance liquid chromatography-mass spectrometry (HPLC-MS/MS). The detection rates were 12.50–100%, and the detected concentrations were in the range of nd–270.60 ng/L. The average detected concentrations of the SAs, MLs, QNs, LIN and TC were 113.68, 111.79, 20.55, 23.81 and 25.66 ng/L, respectively. Among these, SAs and MLs were the dominant antibiotics detected in the Weihe River. Compared with those in other water bodies in China and abroad, the antibiotic residues in the Weihe River were at a moderate contamination level. The SAs concentration distribution followed upstream > midstream > downstream, while the MLs concentration distribution was midstream < downstream < upstream. The correlation analysis and principal component analysis (PCA) indicated that domestic sewage, livestock discharge, and aquaculture and pharmaceutical wastewater are the main sources of antibiotic residues in the Weihe River. In addition, the detected ciprofloxacin (CFX), ofloxacin (OFX) and sulfamethoxazole (SMX) pose high ecological risk in the short and long term.

Antibiotics are widely used in human therapy, livestock production and aquaculture (Kümmerer, 2009). However, a portion of the administered antibiotics can be excreted from the treated hosts in urine and faeces due to incomplete absorbed by organisms (Carvalho and Santos, 2016). Thus, antibiotics can enter the aquatic environment through subsequent discharge of effluents from wastewater treatment plants because of their incomplete removal, animal wastewater discharge, agricultural activity wastewater and surface runoff (Bouki et al., 2003; Davis et al., 2006; Deng et al., 2016). The antibiotics that enter the aquatic environment are toxic to algae and bacteria, leading to the production of resistant species and threatening watershed ecology and drinking water safety (Watkinson et al., 2009). Therefore, the pollution caused by antibiotics has become recognized as an emerging international issue (Zou et al., 2011). In China, antibiotic problems have attracted widespread attention as well.

China is the largest producer and user of antibiotics in the world (Zhu et al., 2013). It was estimated that about 2.48×10^5 tons of antibiotics were produced in 2013, $> 1.62 \times 10^5$ tons were used each year with total emission of 5.0×10^4 t into water and soil environment (Zhang et al., 2015). Furthermore, it was reported that China showed

the second highest number of antibiotics diverse types in Asia (Rico et al., 2012). Quinolones (QNs), macrolides (MLs) and sulfonamides (SAs) are the most frequently used antibiotics, contributing approximately 15, 20, and 12%, respectively (Xu et al., 2009). However, there were significant differences in the types and residue concentrations in different water systems. Some studies showed that QNs, tetracyclines (TCs) and MLs were the main antibiotic residues in the Wenyu River in Beijing, with a concentration of nd–1430.30 ng/L (Zhang et al., 2015). Erythromycin (ETM) and sulfamethoxazole (SMX) were the highest in the Shenzhen River, with the concentrations of 1142 ng/L and 907 ng/L, respectively. SMX was approximately nd–940 ng/L in Baiyangdian Lake (Li et al., 2012). The QNs concentration was found to be nd–214 ng/L in the Liaohe River (Qin et al., 2015) and nd–6800 ng/L in Bohai Bay, China (Zou et al., 2011). The norfloxacin (NFX) concentration was nd–300 ng/L in the Yellow River (Xu et al., 2009).

The Weihe River is the largest tributary of the Yellow River, with a basin area of 134,800 km², of which 49.80% is in Shaanxi Province. According to the environmental quality standard for surface water of the Peoples' Republic of China (GBCNEP, 2002), the Weihe River is severely polluted with a water quality level inferior to the V class based

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on an evaluation of chemical oxygen demand (COD), NH₃-N and other indicators. However, the existing research was mainly focused on non-point source pollution and governance, the residue of antibiotics has rarely been discussed thus far. Considering the potential adverse effects on ecosystem and human health, it is urgent to understand the occurrence, distribution, and source of antibiotic residues in the Weihe River. In addition, these results would help the local governments formulate scientific and reasonable prevention and control measures to ensure the sustainable development of the regional economy. Thus, the objectives of this paper were (1) to detect the occurrence of antibiotic pollutants in the Weihe River, (2) to determine their distribution characteristics and analyse the sources of antibiotics by principal component analysis (PCA), and (3) to assess the potential ecological risks of the detected antibiotics.

This study focused on occurrence, distribution and risk assessment of fifteen antibiotics, belonging to five classes in surface water of the Weihe River Xi'an section. Antibiotic standards used in this study were as follows: Strong anion exchange (SAX) cartridges (6 mL, 500 mg) were provided by Varian (Lake Forest, CA, USA). Oasis hydrophilic-lipophilic balance (HLB) cartridges (6 mL, 200 mg or 6 mL, and 500 mg) were supplied by Waters (Milford, MA, USA), and Agilent provided Zorbax eclipse plus C₁₈ column (100 mm × 2.1 mm, 1.8 μm). Glass fibre filters (GF/F, pore size 0.7 μm) were purchased from Whatman (Maidstone, England) and pyrolysed at 450 °C for 4 h prior to use.

High-performance liquid chromatography (HPLC)-grade methanol was purchased from Merck (Darmstadt, Germany). Formic acid was obtained from Tedia Company (Fairfield, OH, USA). Oxalic acid and ammonium acetate were supplied by Sigma-Aldrich (St Louis, MO, USA). Disodium ethylenediamine tetraacetate (Na₂EDTA), citric acid and sodium citrate were of analytical grade and were purchased from Yaohua Chemical Reagent Factory (Tianjin, China). Ultrapure water was prepared with a Milli-Q water system.

The target standards were the following: sulfapyridine (SPD), sulfadiazine (SDZ), sulfamethazine (SMZ), sulfamethoxazole (SMX), sulfamonomethoxine (SMM), sulfaquinolaxine (SQX), trimethoprim (TMP), oxytetracycline (OTC), norfloxacin (NFX), ciprofloxacin (CFX), ofloxacin (OFX), clarithromycin (CTM), and lincomycin (LIN)) were purchased from Dr. Ehrenstorfer GmbH (Germany). Antibiotic roxithromycin (RTM) and erythromycin (ETM) were obtained from Sigma-Aldrich (St. Louis, MO, USA). Internal standard meclocycline (MC) was purchased from Sigma-Aldrich (St Louis, MO, USA), while sulfamerazine (SMR) and chloramphenicol-D₅(CAP-D₅) were obtained from Dr. Ehrenstorfer GmbH (Germany). Other isotope-labelled internal standards, sulfamethoxazole-D₄ (SMX-D₄), erythromycin-¹³C-D₃ (ETM-¹³C-D₃), ciprofloxacin-D₈, trimethoprim-D₃ (TMP-D₃) and lincomycin-D₃ (LIN-D₃), were obtained from Toronto Research Chemicals (North York, ON, Canada), while sulfamethazine-¹³C₆(SMZ-¹³C₆) was purchased from Cambridge Isotope Laboratories (Andover, MA, USA). Standard products (purity > 95%) are pure solid material. Approximately 10 mg of individual standard was accurately weighed. Quinolones (QNs) were first dissolved in 0.5 mL NaOH and then diluted to 100 mL with methanol. Other agents were dissolved in 100 mL of methanol, and stock solutions were stored at -20 °C. From these stock solutions, the working solutions were prepared by gradient dilution.

The sampling was conducted in May 2016 from 8 sites on the Weihe River Xi'an section (including Lintong, Gaoling, Xi'an and Xianyang), and the detailed sampling sites are shown in Fig. 1 and Table S1. The climate in the region is characterized as temperate continental monsoon with a mean annual precipitation of 610 mm approximately, 80% of which falls between June and October (Chang et al., 2015). Therefore, during the sampling period, the Weihe River was in the dry season; there was no rain, and the average monthly flow in Xianyang and Xi'an were 49.30 and 115 m³/s, respectively.

All the water samples were collected using a water grab sampler from approximately 50 cm below the surface. The volume of the collected samples was 2.5 L. The samples were stored in brown glass

bottles, which were rinsed with the water samples three times before the final sample was collected. The water samples were kept at 4 °C in a cold storage room until further treatment and analysis.

The collected water samples were filtered through Whatman GF/F (0.7 μm) to remove suspended particles. A total of 1000 mL of surface water was extracted by solid-phase extraction (SPE) with Oasis HLB cartridges (6 mL, 500 mg). The target antibiotic compounds were analysed by HPLC-mass spectrometry (MS/MS) (Agilent Liquid Chromatography 1200 series HPLC system coupled with an Agilent 6460 triple quadrupole MS equipped with an electrospray ionization (ESI) source (Agilent, Palo Alto, CA, USA)) in the multiple-reaction monitoring (MRM) mode. The analyses were performed in the positive mode for the target compounds. The liquid chromatography (LC) parameters were as follows: an Agilent Zorbax eclipse plus C₁₈ column (100 mm × 2.1 mm, 1.8 μm) with the corresponding pre-column filter (2.1 mm, 0.2 μm); sample quantity: 5 μL; total flow: 0.3 mL/min; column temperature: 40 °C; mobile phase A: 0.2% formic acid and 2 mM ammonium acetate; mobile phase B: acetonitrile; and gradient elution programme: 0 min 10% B, 5 min 15% B, 7 min 20% B, 11 min 40% B, 15 min 60% B, 16 min 95% B, and 25 min 95% B. The MS conditions were as follows: gas temperature and gas flow: 325 °C and 6 L/min, respectively; nebulizer: 45 psi; sheath gas flow and temperature: 11 L/min and 350 °C, respectively; and capillary and nozzle voltages: 3500 V and 0 V, respectively. The optimized parameters for mass spectrometry are shown in Table S2.

Concentrations of the target compounds in the samples were performed using an internal standard method. Under the optimized conditions, the antibiotic standard solution showed a good linear relationship in the range of 5–200 μg/L. Different concentrations of the individual antibiotics were used as standards and the squared correlation coefficients for the calibration curves were not lower than 0.995 (P < 0.05). For the antibiotics that existed initially, the method quantitation limit (MQL) and method detection limit (MDL) were estimated by determining the signal-to-noise ratio (S/N) of the minimum measured concentrations and extrapolating to S/N values of 10 and 3. The MDLs were 0.19–1.09 ng/L, and the MQLs of the antibiotics from the surface water were 0.63–3.63 ng/L. The details are shown in Table S3. Taking the surface water of Xi'an section of the Weihe River as the base and adding the target mixed antibiotic standard, the samples with 10 ng/L and 100 ng/L concentrations were set up with 3 parallel samples each sample. The recoveries and standard deviations of the various antibiotics were determined according to the above optimized experimental method. The results are shown in Table S3. The recoveries of the target compounds were 87%–170%.

A correlation analysis was used to determine whether the pollution sources of different antibiotics are the same. And principal component analysis (PCA) can recombine the complex antibiotic components in surface water and sum several PCs to explain the main pollutant sources of antibiotics (Deng et al., 2017). Six SAs (SDZ, SMZ, SMX, SMM, SPD and SQX), three QNs (CFX, OFX and NFX), three MLs (CTM, RTM and ETM), LIN, OTC and TMP with high detectable rates in the water samples were analysed by PCA. The correlation analysis and PCA were completed using SPSS 20 statistical software. The correlation analysis was performed using the Pearson correlation method. The PCA of the analytical data was executed to obtain a visual representation of the main characteristics, and the principal components (PCs) were extracted for eigenvalues that were > 1. Microsoft office Excel 2010 was used to process data.

The ecological risks are generally evaluated by risk quotients (RQs). RQs of the targeting aquatic organisms were calculated according to the Eq. (1) (BielMaeso et al., 2018), and the European Commission (EC, 2003) has released technical guidance documents on risk assessments.

$$RQs = MEC/PNEC \quad (1)$$

where MEC and PNEC are the measured environmental concentrations and the predicted no-effect concentrations, respectively. The PNEC is

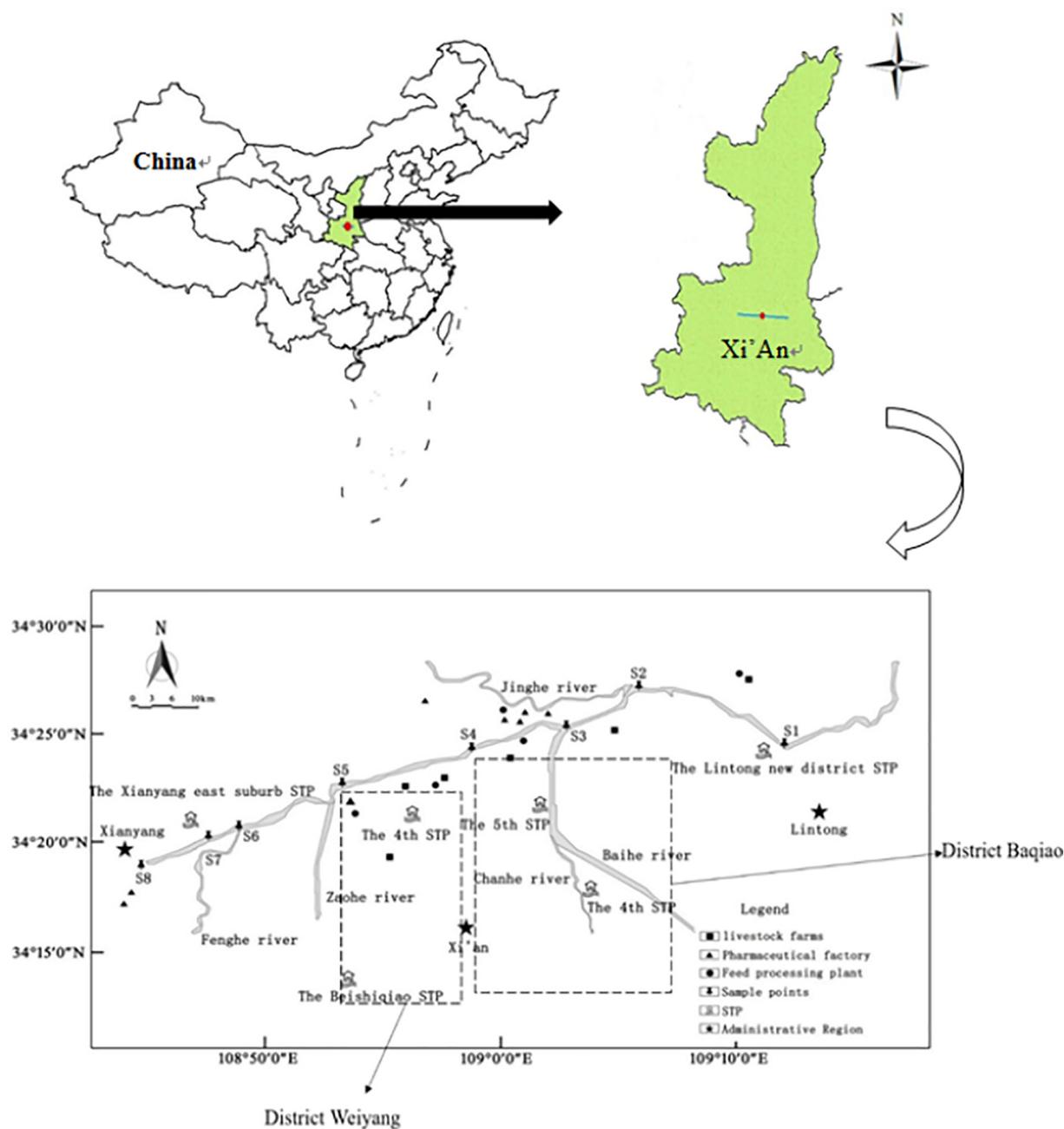


Fig. 1. Map of sampling sites in the mainstem and tributaries of the Xi'an section of the Weihe River.

obtained from EC₅₀ or LC₅₀ divided by assessment factor from acute toxicity data, or from chronic no observed effect concentration (NOEC) values divided by assessment factor. Ecological risks are determined as low risk with $RQ \leq 0.1$, moderate risk with $0.1 < RQ \leq 1$, and high risk with $RQ > 1$ (Hernando et al., 2006).

The types and residues of antibiotics in the Weihe River are summarized in Table 1 and Fig. 2. As shown in Table 1, the detection rate of 10 antibiotics was higher than 50%, and which ETM, RTM, SMX, SPD, TMP and LIN were 100%; SMM and SDZ were 87.50%; and OFX and SMZ were 75%.

As shown in Fig. 2, the residues with a 100% detection rate followed a trend of $ETM > SMX > RTM > LIN > TMP > SPD$, and the corresponding average concentrations were 83.99, 70.81, 25.52, 23.81, 12.25 and 7.19 ng/L, respectively.

In this study, six antibiotics with detection rates higher than 75% were compared with those in other river bodies in China and abroad

(Table 2). Compared with that in northern Chinese rivers (Wenyu River, Liaohe River, and the Yellow River), RTM concentration was lower, SDZ, SMX, SMZ and TMP concentrations were higher, while OFX concentration was at a moderate level in the Weihe River. Compared with concentrations in southern Chinese rivers (the Zhujiang River and the Yongjiang River), in the Weihe River, RTM, OFX and SMX were at a moderate level. By comparison with that in rivers abroad (the Seine River, the Elbe River, and the Mekong River), RTM and SMX concentrations were at a moderate level, while OFX concentration was higher in the Weihe River. OFX and TMP concentrations in Weihe River were higher than that in lakes and bays, such as Baiyangdian Lake and Qinzhou Bay.

As shown in Fig. 3, the antibiotics detected at sampling points 1, 3 and 4 presented more types and higher concentrations, with 12 antibiotics detected and a total concentration in the range of 2.07–276.60 ng/L. Six SAs, three MLs, one QNs, one TC and LIN were

Table 1
Antibiotics species and concentration in the surface water of the Weihe River Xi'an section.

Antibiotics	Abbreviation	Concentration range (ng/L)	Average (ng/L)	Mid (ng/L)	Detection rate (%)
Ciprofloxacin	CFX	nd~7.32	7.32	7.32	12.5
Clarithromycin	CTM	nd~10.10	9.11 ± 1.41	9.11	25
Erythromycin	ETM	23.30–276.60	83.99 ± 8.53	53.07	100
Lincomycin	LIN	3.63–125.33	23.81 ± 4.14	9.10	100
Norfloxacin	NFX	nd~39.21	18.21 ± 1.82	8.14	37.5
Ofloxacin	OFX	nd~71.99	17.08 ± 2.70	6.37	75
Oxytetracycline	OTC	nd~103.81	102.63 ± 1.68	102.63	25
Roxithromycin	RTM	1.57–59.49	25.52 ± 2.12	23.98	100
Sulfadiazine	SDZ	nd~7.95	5.41 ± 1.93	5.22	87.5
Sulfamethazine	SMZ	nd~7.32	3.87 ± 1.76	3.29	75
Sulfamethoxazole	SMX	7.6–114.46	70.81 ± 3.93	78.16	100
Sulfamonomethoxine	SMM	nd~14.74	9.70 ± 3.68	9.66	87.5
Sulfapyridine	SPD	2.07–16.77	7.19 ± 5.23	5.53	100
Sulfaquinolaxline	SQX	nd~17.91	10.36 ± 1.07	10.36	25
Trimethoprim	TMP	3.46–36.04	12.25 ± 1.05	8.24	100

Note: nd, not detected.

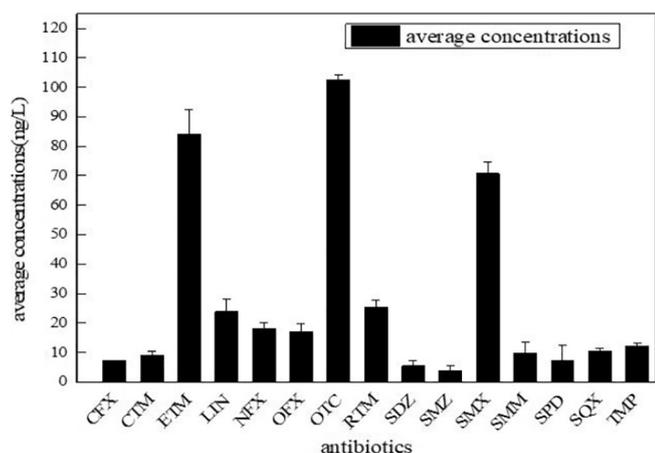


Fig. 2. Average concentrations of antibiotics.

detected at sampling point 1. Six SAs, two QNs, three MLs, and one LIN were at sampling point 3. Six SAs, three QNs, two MLs, and one LIN were at sampling point 4.

Based on the population, the distribution of factories and aquaculture, Xi'an section of the Weihe River was divided into three sections: upstream (Xianyang urban area, including sampling points 7 and 8); midstream (Xi'an area, including sampling points 3, 4, 5 and 6); and downstream (Gaoling and Lintong area, including sampling points 1 and 2). The population density increases from upstream to downstream, and the antibiotics detected showed the same pattern, with the total concentrations of 105.31 ng/L, 175.60 ng/L and 289.67 ng/L, respectively (Fig. 3).

Antibiotics types at each sampling point are shown in Figs. 4 and S1.

Table 2
Global comparison of six antibiotic concentrations (ng/L) in surface water.

Research region	Roxithromycin	Ofloxacin	Sulfadiazine	Sulfamethoxazole	Sulfamethazine	Trimethoprim	Reference
North China	Beijing Wenyu River	7.5–141.7	25.1–1213.6				(Zhang et al., 2015)
	Liaohu River		12–24(17)	nd~21		nd~8	(Qin et al., 2015)
	Yellow River	nd~95	nd~264		nd~56		(Xu et al., 2009)
	Weihe River	1.57–59.49	nd~71.99	nd~7.95	7.6–114.46	nd~7.32	3.46–36
South China	Zhujiang River(Guangzhou)	13–169	53–108		111–193		(Xu et al., 2007)
	Yongjing River	nd~6.1	0.77–21.8		5.6–78.8		(Wu et al., 2013)
Foreign	Seine River, France	nd~350	30		40–140		(Tamtam et al., 2008)
	Elbe River, German	nd~40			30–70		(Wiegel et al., 2004)
	Mekong River, Vietnam				20–33		(Managaki et al., 2007)
Estuaries and bays	Baiyangdian Lake	nd~155	0.38–32.6	0.86–505	nd~940	nd~16.1	(Li et al., 2012)
	Qinzhou Bay			nd~3.41	nd~10.4	nd~3.39	nd~3.77

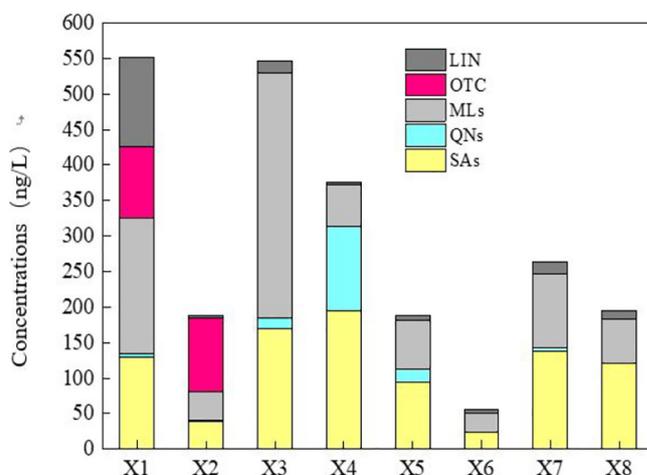


Fig. 3. Antibiotic concentration distributions in the Xi'an section of the Weihe River.

The concentrations of LIN was downstream (64.48 ng/L) > upstream (14.49 ng/L) > midstream (8.16 ng/L). The SA residue was upstream (123.92 ng/L) > midstream (115.81 ng/L) > downstream (80.27 ng/L). OTC was only detected in downstream, with an average concentration of 102.63 ng/L. The distributions of MLs and QNs followed midstream > downstream > upstream, and the residues at sampling points 3 and 4 were especially high, with the total concentrations achieving 344.20 ng/L and 118.52 ng/L, respectively. Which was attributed to the poultry and livestock breeding areas around the Baqiao District, and dairy cattle and aquaculture area near the Weiyang District. MLs and QNs were more easily detected in the aquatic

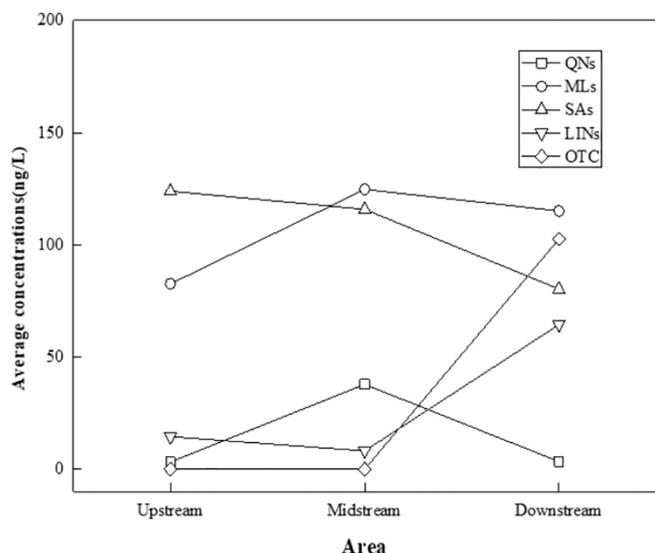


Fig. 4. Antibiotic distributions along different regions.

environment near livestock breeding and aquaculture areas (Zhang et al., 2015).

SAs antibiotics are the main clinical drugs widely used in China (Tang et al., 2015). SAs cannot be effectively removed through the current sewage treatment process (Yuan et al., 2019). The relatively high residue of SAs in upstream was attributed to the dense population in Xianyang City, where more antibiotic sewage, industrial waste and medical wastewater were discharged into the Weihe River than other locations. The average concentration of SAs at sampling point 4 in midstream achieved the highest, 194.84 ng/L. That is due to the surrounding livestock breeding area.

The concentrations of LIN and OTC in downstream were the highest, because the district is a dominant fishery breeding area, and there are many livestock farms as well. Studies have shown that OTC remains in fish pond sediments and water (Liu et al., 2008), and LIN is also frequently associated with the treatment of respiratory diseases such as avian pneumonia and diarrhoea (Kim et al., 2013).

As shown in Table 3, CFX, NFX, OFX, SMZ and TMP were extremely significantly correlated ($P < 0.01$), while CTM, ETM, LIN and RTM were significantly correlated ($P < 0.05$). SDZ, SMM and SPD were extremely significantly correlated ($P < 0.01$); SMZ, SPD, and TMP were also extremely significantly correlated ($P < 0.01$); SMX and SPD were significantly correlated ($P < 0.05$). The above analysis indicated

Table 3
Correlation analysis of surface water antibiotics in Xi'an section of the Weihe River.

	CFX	CTM	ETM	LIN	NFX	OFX	OTC	RTM	SDZ	SMZ	SMX	SMM	SPD	SQX	TMP
CFX	1	-0.216	-0.280	-0.187	0.862**	0.977**	-0.218	0.145	0.490	0.913**	0.360	0.452	0.740*	-0.163	0.919**
CTM		1	0.796*	0.811*	-0.355	-0.264	0.409	0.779*	0.437	0.065	0.394	0.493	0.251	-0.245	0.026
ETM			1	0.328	-0.329	-0.286	0.008	0.812*	0.539	-0.040	0.546	0.525	0.408	-0.163	-0.175
LIN				1	-0.354	-0.227	0.595	0.500	0.128	0.097	0.209	0.220	-0.047	-0.121	0.100
NFX					1	0.895**	-0.090	-0.106	0.198	0.731*	0.210	0.124	0.552	0.147	0.794*
OFX						1	-0.305	0.121	0.473	0.917**	0.431	0.412	0.735*	0.016	0.869**
OTC							1	-0.040	-0.477	-0.192	-0.329	-0.400	-0.370	-0.248	0.144
RTM								1	0.767*	0.450	0.831*	0.764*	0.699	-0.277	0.262
SDZ									1	0.652	0.710*	0.988**	0.867**	-0.145	0.377
SMZ										1	0.654	0.617	0.836**	-0.049	0.887**
SMX											1	0.638	0.788*	0.070	0.372
SMM												1	0.807*	-0.198	0.359
SPD													1	-0.210	0.681
SQX														1	-0.307
TMP															1

* Significant correlation was found at the 0.05 level (bilateral).

** Significant correlation was found at the 0.01 level (bilateral).

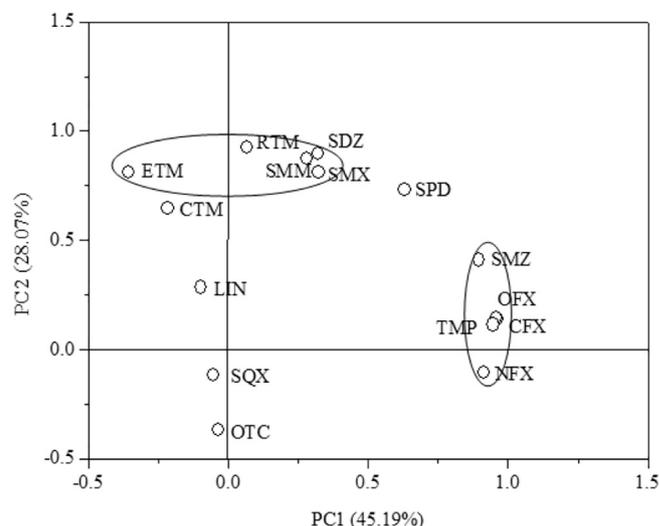


Fig. 5. Principal components analysis for the Weihe River Xi'an section.

that the pollution sources of CFX, NFX, OFX, SMZ, TMP, and SPD were the same; the pollution sources of SPD, SDZ, SMM, and SMX were the same; and that of CTM, ETM, LIN, and RTM were the same.

As shown in Table S4, the varimax rotation reduced all the variables to four different PCs, which represented 93.52% of the total variance. Among these, PC1 accounted for 45.19%, PC2 for 28.07%, PC3 for 13.29%, and PC4 for 6.97%.

Fig. 5 and Table S5 show that PC1 had strong positive loadings on CFX, NFX, OFX, TMP and SMZ. As mentioned above, these antibiotics were highly detected at sampling point 4. Wastewater treatment plants (WWTPs) are regarded as one of the most important sources of antibiotics in the environment. TMP, OFX, and NFX were found to be the predominant antibiotics in the WWTP effluents (Zhou et al., 2013). The Xi'an Fourth sewage treatment plant is located near point 4. Therefore, PC1 represented the source of domestic sewage pollution. PC2 had strong positive loadings on ETM, RTM, SDZ, SMX, and SMM, and the highest concentrations of these antibiotics were detected at sampling point 3, where many livestock breeding areas and pharmaceutical factories are located. It was estimated that approximately 40% of the antibiotics were used in livestock (Xu et al., 2009). SAs have been detected in livestock WWTPs (Kim et al., 2013). Therefore, PC2 represented the source of livestock wastewater pollution and pharmaceutical wastewater pollution. PC3 had strong positive loadings on LIN and OTC and represented the source of aquaculture wastewater

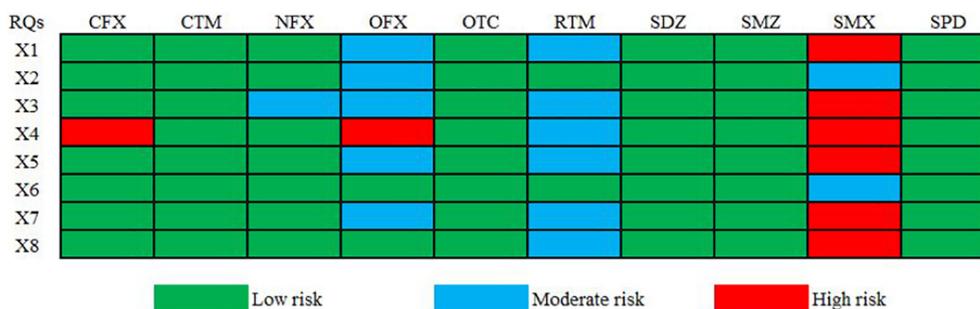


Fig. 6. Ecological risks of antibiotics in the Xi'an section of the Weihe River.

pollution. LIN and OTC were detected in high amounts at sampling points 1 and 2, where many aquaculture areas were located. In addition, > 47.4 million tons of farmed aquatic products have been produced in mainland China (Liu et al., 2017). OTC is also a typical antibiotic in the marine aquaculture farms surrounding Hailing Island, South China (Chen et al., 2015). SQX had strong positive loadings in PC4, and its highest concentration was detected at sampling point 8, where Xianyang City is located. Therefore, PC4 represented the other pollution. In summary, the main sources of antibiotics in the Xi'an section of the Weihe River are domestic sewage, livestock wastewater, pharmaceutical wastewater, and aquaculture wastewater.

Recently, with the widespread detection of antibiotics in the natural ecosystem across the world, the potential influence of antibiotics on organisms in water and sediment has become an important research issue. In this study, the calculated RQs of the antibiotics for algae organisms and the species tested were *Microcystis aeruginosa*, *Rhodomonas salina*, *Vibrio fischeri*, *Pseudokirchneriella subcapitata*, *Selenastrum capricornutum*, *Scenedesmus vacuolatus*, *Synechococcus leopoliensis* and *Lemna minor*. The specific evaluation results are shown in Table S6 and Fig. 6.

As shown in Fig. 6, CFX and OFX posed a short-term high ecological risk to the corresponding aquatic organisms at sampling point 4. NFX and RTM posed moderate ecological risk at different sampling points. In addition to sampling points 2 and 6, SMX posed moderate ecological risk to corresponding aquatic organisms, and SMX posed high ecological risks at other sampling points. SMX was the important contributor to ecological risk due to their low PNEC values. In previous reports, both short-term and long-term ecological risks posed by detected SMX were at high levels (Chen et al., 2017). It is reported that SMX was the most toxic antibiotics in Chaohu, east China (Tang et al., 2015). SMX showed relatively high RQ values for the selected organisms in the Yangtze Estuary (Yan et al., 2013), and in Korea (Lee et al., 2008), where SMX was shown to pose relatively high ecological risks. This was due to the fact that SMX inhibit phagocytic activity despite at considerably low concentrations (Park and Choi, 2008). Although the antibiotics residue in the environmental samples were quite low, its strong ability to absorb on sediment can lead to the contamination of surface water, be toxic to aquatic organisms (Boxall et al., 2003; Robinson et al., 2005). Nevertheless, it should be noted that the single-compound exposure scenarios are unrealistic. In the real environment, the mixture effect of multiple contaminants is ubiquitous, which may cause considerable ecological concerns (Leung et al., 2012).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2019.07.016>.

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