Distribution and ecological risk assessment of typical antibiotics in surface waters of seven major rivers, China

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Table S1 Summary of the extraction and separation methods for antibiotics in surface waters of seven major rivers in china reported in the literature between 2010 and 2020

Pretreatment method	Extraction	Separation	Detection	Reference
Water samples (1 L) were filtered through 0.22 μm micropore membranes.	NA	BEH C18 column (50 mm × 2.1 mm, 1.7 μm) The mobile phase contained 0.01% formic acid in pure water (A) and 100% acetonitrile (B).	LOD: 0.071～0.432 ng/L; LOQ: 0.21～1.369 ng/L; Recovery: 81.12～115.69%	1
Water samples were filtered through 0.45 μm glass fiber filters and then acidified to pH 3 by adding HCl. 0.2 g/L Na$_2$EDTA was added into water samples.	Oasis HLB cartridges (500 mg, 6mL) Pretreatment: methanol (6 mL) and ultrapure water (6mL). Rinse: 6 mL ultrapure water. Elution: 6mL ultrapure water and 12 mL methanol.	BEH C18 column (100 mm × 2.1 mm, 1.7 μm)	LOQ: 0.05～5 ng/L; Recovery: 84.5～107.3%; RSD: <5%	2
Water samples were adjusted to pH 3 using H$_2$SO$_4$, added with 5% methanol.	Oasis HLB cartridges (200mg, 6 m) Pretreatment: methanol (10 mL) and Milli-Q water (10mL). Rinse: 10 mL Milli-Q water. Elution: 12 mL methanol.	BEH C18 column (100 mm × 2.1 mm, 1.7 μm) . The mobile phase contained 0.1% formic acid in Milli-Q water (A) and acetonitrile (B).	LOD: 0.02～5.03 ng/L; Recovery: 40～140%	3
Water samples were filtered through 0.7 μm glass fiber filters and acidified to pH 4 with formic acid, followed by the addition of 0.2 g/L Na$_2$EDTA.	Elution: 10 mL methanol.	HSS T3 column (100 mm × 2.1 mm, 1.8 μm) The mobile phase contained 0.1% formic acid in ultrapure water (A) and acetonitrile with 0.1% formic acid (B).	LOD: 0.01～1.07 ng/L; LOQ: 0.03～1.71 ng/; Recovery: 63.0～126.8% ; RSD: < 5.5%	4
Water samples were filtered by glass fiber filter, and then pH was adjusted to 5.	Strata-X cartridges (200 mg, 6 mL) Pretreatment: deionized water (6 mL) and methanol (6 mL). Rinse: 6 mL deionized water. Elution: 3 mL methanol.	Supelco Discovery HS C18 column (150 mm × 4.6 mm, 3 μm) The mobile phase contained 0.1% formic acid in ultrapure water (A) and acetonitrile (B)	LOQ: 0.15～9.1 ng/L; Recovery: 77～159%	5
Water samples were acidified to pH 4.0 with citrate buffer, followed by the addition of 0.2 g/L Na$_2$EDTA.	Oasis HLB cartridges (200 mg, 6 mL) Pretreatment: methanol (6mL) and ultrapure water (10mL). Elution: 10 mL methanol.	HSS T3 column (100 mm × 2.1 mm, 1.8 μm) Ultrapure water containing 0.1% formic acid (A) and acetonitrile containing 0.1% formic acid (B) was used as the mobile phase.	LOD: 0.01～1.18 ng/L; LOQ: 0.03～1.68 ng/L; Recovery: 62.1～129.4%	6

Water sample was filtered through 0.45 μm fiber filters and acidified to pH 3 with H_3PO_4, followed by the addition of 100 g/L Na_2EDTA.	Oasis HLB cartridges Pretreatment: methanol (6 mL) and ultrapure water (3 mL) and sodium dihydrogen phosphate solution (6 mL). Rinse: 6 mL ultrapure. Elution: 3 mL methanol and 6 mL methanol with 2% ammonia	Poroshell 120 EC-C18 (2.1 mm × 75 mm, 2.7 μm) The mobile phase contained acetonitrile (A) and 0.2% formic acid in ultrapure water (B).	LOD: 0.1～1.6 ng/L; Recovery: 74.3～95.8%; RSD: 4.7～9.5%	7
Water sample (1L) was filtered through 0.45 μm fiber filters, and then 5 mL of 100g/L Na_2EDTA was added and the pH was adjusted to 4 with 50% H_3PO_4.	Pretreatment: methanol (6 mL) and ultrapure water (3 mL) and sodium dihydrogen phosphate solution (6 mL). Rinse: 6 mL ultrapure water. Elution: 6 mL methanol with 2% ammonia.	ZORBAX Eclipse Plus C18 (150 mm × 2. 1 mm，3.5 μm) The mobile phase contained 0.2% formic acid in ultrapure water (A) and acetonitrile (B).	LOD: 0.03～15 ng/L; Recovery: 41.3～112. 6%; RSD: 0.2～12. 7%	8
Water samples were filtered through 0.22 μm fiber filters and then acidified to pH 3 by adding HCl, followed by the addition of 0.5g Na_2EDTA.	Poly-Sery HLB cartridges Pretreatment: methanol (10 mL) and ultrapure water (10 mL). Rinse: 5 mL methanol with 5% ammonia and 10 mL ultrapure water. Elution: 10 mL methanol.	C18 (2.1 mm× 50 mm, 1.8 μm) The mobile phase contained 0.2% formic acid (A) and acetonitrile (B).	Recovery: 69～125%	9
Water sample (1L) was filtered through 0.45 μm glass fiber filters and acidified to pH 3.	Oasis HLB cartridge Rinse: ultrapure water. Elution: methanol.	NA	LOQ: 1～20 ng/L; Recovery: 61～86%	10
Water samples (1L) were filtered through 0.45 μm glass fiber filters and acidified to pH 3 by adding H_2SO_4, followed by the addition of 10 mL of 5% Na_2EDTA.	HLB cartridges Pretreatment: methanol (6 mL) and ultrapure water (6 mL).	C18 (4.6 mm × 150 mm, 3.5 μm) The mobile phase contained 0.1% formic acid in ultrapure water (A) and acetonitrile with 0.1% formic acid (B).	LOQ: 0.1～1.4 ng/L; Recovery: 73～92%; RSD: <20%	11
Water samples were filtered through 0.7 μm fiber filters and then 0.5g Na_2EDTA was added. Samples were acidified to pH 3 by adding 4mol/L HCl.	Oasis HLB (200 mg，6mL) Pretreatment: methanol (10 mL) and ultrapure water (10 mL). Rinse: 10 mL ultrapure. Elution: 10 mL methanol.	Agilent ZORBAX Eclipse Plus C18 (100 mm × 2.1 mm, 1.8 μm) The mobile phase contained 0.2% formic acid and 2 mmol/L ammonium acetate (A) and acetonitrile (B).	Recovery: 31.4～158.8%; RSD: <20%	12

Water samples (1L) were filtered through 0.7 μm fiber filters and then acidified to pH 3 by adding H_2SO_4, followed by the addition of 5% methanol.	HLB (1000 mg) Elution: 10 mL methanol.	Agilent ZORBAX Eclipse Plus C18（100 mm × 2. 1 mm，1. 8 μm) The mobile phase contained 0.1% formic acid and 5 mmol/L ammonium acetate (A) and acetonitrile (B).	LOD: 0.4～10 ng/L; Recovery: 61.4～131.5% RSD: <20%	13
The pH values of the water samples were adjusted to 3 by adding H_2SO_4 and then filtered through 0.7 μm fiber filters, followed by the addition of 1 g Na_2EDTA.	Oasis HLB (500 mg, 6 mL) Pretreatment: methanol (10 mL) and ultra-pure water (10 mL). Rinse: 10 mL Milli-Q water. Elution: 12 mL methanol.	XDB-C18 column (2.1 mm × 50 mm, 1.8 μm) The mobile phase contained acetonitrile (A) and 5mM oxalic acid (B).	LOQ: 0.077～5.3 ng/L; Recovery: 66～158%; RSD: 1～10%	14
Water samples were filtered through 0.7 μm fiber filters by the addition of 10 mL of 5% Na_2EDTA. The pH value was adjusted to 3 using formic acid.	HLB (500 mg, 6 mL) Pretreatment: methanol (6 mL) and Milli-Q water (6 mL). Rinse: 10 mL Milli-Q water. Elution: 6 mL methanol.	Agilent ZORBAX C18 (2.1 mm × 150 mm, 5 μm) The mobile phase contained acetonitrile (A) and 0.2% formic acid (B).	LOQ: 0.22～9.96ng/L; Recovery: 55.3～108%; RSD: <14%	15
Water samples were filtered through 0.7 μm glass fiber filters and then acidified pH to 3 by adding H_2SO_4, followed by the addition of 0.5 g Na_2EDTA.	HLB (500 mg, 6 mL) Pretreatment: methanol (10 mL) Milli-Q water (10 mL). Rinse: Milli-Q water (10 mL). Elution: 10 mL methanol.	ZORBAX Eclipse Plus C18 (2.1 mm × 50 mm, 1.8 μm) and ZORBAX SB C18 (2.1 mm × 100 mm, 1.8 μm) The mobile phase contained water (A) and acetonitrile (B).	LOD: 0.004～0.6 ng/L; Recovery: 70～120%	16
NA	HLB (200mg, 6mL) Pretreatment: methanol (5 mL) and ultrapure water (2 mL). Rinse: 12 mL ultrapure water. Elution: 6 mL methanol.	XTerra MS C18 (2.1 mm × 100 mm, 3.5 mm)	MDL: 0.01～0.25 ng /L; Recovery: 57.4～138%	17
Water samples were filtered through 0.45 μm fiber filters, and adjusted to pH 5 with citrate buffer. 0.2 g Na_2EDTA was added into samples.	Oasis HLB (500 mg, 6mL) Pretreatment: methanol (5 mL) and pure water (5 mL). Rinse: 5 mL deionized sterile water. Elution: 6 mL methanol.	Intersil ODS-3 (2.1 mm × 250 mm, 5 μm) The mobile phase contained acetonitrile (A) and purified water with 0.3% formic acid (B).	Recovery: 58～91%; RSD: 0.5～2.5%	18
NA	NA	NA	LOD: 0.01～8 ng/L; LOQ: 0.1～20 ng/L; Recovery: 53.79～138%;	19

Sample pretreatment	SPE	Chromatographic conditions	Method validation	Ref
Na₂EDTA was added in to samples and then filtered through 0.7 μm glass fiber.	Oasis HLB and Oasis MAX. Pretreatment: ultrapure water. Elution: methanol.	The mobile phase contained methanol (A) and methanol with 1% formic acid (B).	NA	20
Water sample was filtered through 0.45 μm fiber filters and then acidified to pH 2.5 by adding HCl, followed by the addition of 0.2 g Na₂EDTA.	Qasis HLB. Pretreatment: methanol (5 mL) and ultrapure water (5 mL). Rinse: ultrapure water (5 mL). Elution: methanol (6mL).	Eclipse Plus C18 (3.0 mm × 100 mm, 1.8 μm). The mobile phase contained 0.1% formic acid (A) and acetonitrile with 0.1% formic acid (B).	LOD: 0.4～1.9 ng/L Recovery: 64.5%～132%	21
Water samples were acidified to pH 3 with H₂SO₄, followed by the addition of 0.5% methanol and 0.5 g Na₂EDTA. Water samples were filtered through glass fiber filters finally.	HLB (500mg, 6mL). Pretreatment: methanol (20 mL) ultrapure water (6 mL). Rinse: methanol and 10 mL ultrapure water. Elution: 12 mL methanol.	RRHD Zorbax Stable Bond-C18 (2.1 mm × 100mm, 1.8 μm). The mobile phase contained 0.2% formic acid in ultrapure water (A) and methanol (B).	LOD: 0.0015～1.6 ng/L; Recovery: 51～101%	22
Water sample (1 L) was filtered through 0.45 μm fiber filters and acidified to pH 4 with diluted HCl, followed by the addition of 0.5g/L Na₂EDTA.	C18. Pretreatment: methanol (12 mL) and ultrapure water (12 mL). Rinse: ultrapure water (5 mL). Elution: methanol (6 mL).	C18 (4.6 mm × 250mm, 5 μm). The mobile phase contained 0.2% formic acid (A) and acetonitrile (B).	Recovery: 91.4～98.6%; RSD: 3.3～9.6%	23
Water sample (400 mL) was filtered through 0.45 μm fiber filters and then acidified to pH 3～4 by adding 3%～5% HCl.	Oasis HLB. Pretreatment: methanol (10 mL) and ultra-pure water (10 mL). Rinse: ultrapure water (5 mL). Elution: methanol (10mL).	BEH C18 (50 mm × 2.1 mm, 1.7 μm). The mobile phase contained methanol with 0.1% formic acid (A) and water (B).	LOD: 2.5～20.36 ng/L	24
Water sample (500mL) was filtered through 0.45 μm fiber filters and then acidified to pH 4 by adding 1mol/L HCl, followed by the addition of 0.2 g Na₂EDTA.	Oasis HLB. Pretreatment: methanol (6 mL) and ultra-pure water (6 mL). Rinse: ultrapure water (6 mL). Elution: methanol (6mL).	Dionex C18 (2.1 mm × 150 mm, 3 μm). The mobile phase contained methanol (A) and 0.1% formic acid (B).	MQL: 0.3～3.3ng/L; Recovery: 66.2～109.2%; RSD: 4.1%～15.3%	25
Water sample (1L) was filtered through two fiber filters: one at 0.8 μm and the one at 0.22 μm and then 0.5 g of Na₂EDTA was added. The pH was adjusted to 4 using 1mol/L HCl	Oasis HLB. Pretreatment: methanol (5 mL) and ultrapure water (5 mL). Rinse: 10 mL ultrapure water containing 5% methanol. Elution: 10 mL methanol containing 1% formic acid.	BEHC18 (2.1 mm × 50 mm, 1.7 um). The mobile phase contained acetonitrile (A) and 0.05% formic acid in ultrapure water (B).	LOD: 0.01～0.95 ng/L; Recovery: 72.1～104.6%; RSD<10%	26

Water samples were acidified to pH 3.	HLB (500 mg, 6mL)	RP18 (150 mm × 2.1 mm, 3.5 μm). The mobile phase contained formic acid (A) and acetonitrile (B).	LOD: 0.003～0.016 ng/L; Recovery:71～105% RSD: 2.7～12.9%	27
Water sample (400mL) was filtered through 0.45 μm fiber filters and 2mL of 100g/L Na₂EDTA was added. The pH was adjusted to 3 with H₃PO₄.	HLB (500mg, 6mL）Pretreatment: methanol (6 mL) and ultrapure water (3 mL) and Sodium dihydrogen phosphate solution (6mL). Rinse: ultrapure water (6 mL). Elution: methanol (3 mL) and methanol with 2% ammonia (3 mL).	XBridge C18(150 mm × 2.1 mm，5 μm) The mobile phase contained acetonitrile (A) and methanol with 1% formic acid (B).	LOD:0.05～0.32 ng/L Recovery: 69.5～122.6% RSD: 1.43～9.75%	28
Water samples were filtered through 0.45 μm glass fiber filters and then acidified to pH 3 by adding HCl, followed by the addition of 1.2 g Na₂EDTA.	Oasis HLB (500 mg, 6 mL) Pretreatment: methanol (5 mL) and ultrapure water (5mL). Rinse: 15 mL ultrapure water. Elution: 10 mL methanol containing 5% ammonium hydroxide.	Agilent Zorbax Eclipse XDB-C18 (4.6 mm × 150 mm, 5 μm) The mobile phase contained acetonitrile (A) and 0.1% formic acid (B).	LOD: 0.04～1.1 ng/L; LOQ: 0.1～3 ng/L; Recovery: 57.8～133.9%	
Water samples were filtered through 0.45 μm fiber filters and acidified to pH 3 with 0.1 mol/L HCl, followed by the addition of 0.2 g Na₂EDTA	Oasis HLB (200 mg, 6mL) Pretreatment: methanol (6 mL) and ultrapure water (6 mL). Rinse: 10 mL ultrapure water. Elution: 6 mL methanol.	Xbridge C18 (150 mm × 2.1 mm, 3.5 μm) The mobile phase contained acetonitrile (A) and 0.1 % formic acid (B).	LOQ: 0.1～0.5 ng/L; Recovery: 60.4～99.6 %; RSD: 0.1～21.7%	29
Water samples were filtered through 0.45 μm glass fiber filters and then 0.4 g Na₂EDTA was added into water samples. The pH value was adjusted to 3 by adding H₂SO₄ and NaOH.	HLB (500 mg, 6 mL) Pretreatment: methanol (2 mL) and 0.1% formic acid in ultrapure water (2 mL). Rinse: 2 mL ultrapure water. Elution: 10 mL ultrapure water.	Atlantis ®T3 (150 mm × 2.1 mm, 3 μm) The mobile phase contained 0.2% formic acid in ultrapure water (A) and methanol (B).	LOD: 0.18-0.52 ng/L; Recovery: 79.91～91.31%	30
NA	NA	Agilent Zorbax Eclipse XDB-C18 (4.6 mm × 150 mm, 5 μm)	Recovery:98.7～102.5%	31

NA, Not available

Table S2 Summary of the aquatic toxicity data and lowest PNEC values of target antibiotics

Compound	Taxonomic group		NOEC (mg/L)	L(E)C$_{50}$ (mg/L)	AF	Lowest PNEC (ng/L)	Reference
SMZ	Bacteria	*V. fischeri*		344.7	1000		3
	Algae	*P. subcapitata*	0.001		100	10000	3
	Invertebrate	*D. magna*		158.8	1000		3
	Fish	*O. latipes*		>100	1000		3
SDZ	Algae	*S. capricornutum*		2.19	1000		3
	Algae	*C. vulgaris.*		1.226	1000	1226	3
SMX	lgae			0.52	1000		3
	Algae	*C. vulgaris.*		1.57	1000		3
	Algae	*S. capricornutum*	0.614		100		3
	Algae	*P. subcapitata*	<0.0005		100		3
	Algae	*S. leopoliensis*		0.027	1000	27	3
	Rotifers			9.63	1000		3
	Crustaceans	*C.dubia*		0.21	1000		3
	Bacteria	*V. fischeri*		78.1	1000		3
	Invertebrate	*D. magna*		177.3	1000		3
	Fish	*O. latipes*		562.5	1000		3
	Cnidarian	*H. attenuata*	5		100		3
TMP	Algae	*S. capricornutum*		130	1000		3
	Algae	*R. salina*		16	1000	16000	3
	Algae	*S. capricornutum*	25.5		100		3
	Algae	*P. subcapitata*		40	1000		3
	Cnidarian	*H. attenuata*	>100		100		3
	Invertebrate	*D. magna*			1000		3
	Fish	*B. Rerio*			1000		3
NOR	Bacteria	*Vibrio fischeri*	0.01038		100	103.8	3
	Algal	*S. capricornutum*	4.01		100		3
	Algae	*P. subcapitata*		18	1000		3
OFX	Bacteria	*Vibrio fischeri*	0.00113		100	11.3	3
	Algae			1.44	1000		3
	Algae	*S. leopoliensis*		0.016	1000		3
	Algae	*M. aeruginosa*		0.021	1000		3
	Rotifers			0.53	1000		3

	Crustaceans			3.13	1000		3
CIP	Algae	*P. subcapitata*		6.7	1000		3
	Algae	*M. aeruginosa*		0.017	1000		3
	Algae	*M. aeruginosa*		0.005	1000	5	32
	Algae	*Pseudokirchneriella*	0.5		100		33
ENR	Bacteria	*Vibrio fischeri*	0.00288		100	28.8	3
	Algae	*M. aeruginosa*		0.049	1000		3
	Invertebrate	*D. magna*			1000		3
	Fish	*O. mykiss*			1000		3
OTC	Algae			0.17	1000	170	3
	Algae	*S. capricornutum*	0.342		100		3
	Rotifers			1.87	1000		3
	Crustaceans	*C.dubia*		0.18	1000		3
	Cnidarian	*H. attenuata*	50		100		3
TC	Algae	*M. aeruginosa*		0.09	1000		3
		M. aeruginosa		0.05	1000	50	29
	Algae	*S. capricornutum*		2.2	1000		3
	Algae	*P. subcapitata*	0.0005		100		3
CTC	Algae	*M. aeruginosa*		0.05	1000		3
	Algae	*S. capricornutum*		3.1	1000		3
	Algae	*P. subcapitata*	0.0005		100	5	3
	Invertebrate	*D. magna*		>400	1000		3
ETM-H$_2$O	Algae	*P.subcapitata*		0.020	1000	20	3
	Rotifers			0.94	1000		3
	Crustaceans	*C.dubia*		0.22	1000		3
ROX	Algae	*P. subcapitata*		0.047	1000	47	3
	Alage	*P.subcapitata*	0.01		100		35

Table S3 Sampling sites of studies on target antibiotics in surface waters of seven major rivers in China

Surface water	Sampling sites	Sampling sites	Survey period	reference
Yangtze River	S1-S2	Lower Yangtze River (Jiangsu section)	11/2018	1
	S3-S10	Yangtze River estuary (Datong, Xuliujing and Wusongkou)	6	4
	S11-S17	Yangtze Estuary(Xupu, Daxingang, Yinyang, Bailonggang, Luchao, Wusongkou, and Liuhekou)	7/2011,10/2011	6
	S18-S19	Nanjing reach of the Yangtze River	8/2018	8
	S20-S21	Middle and lower reaches of the Yangtze River	7/2016	3
	S22	Nanjing reach of the Yangtze River	4/2017	7
Yellow River	S23	Yellow river mainstream and tributaries (Luo River, Yi River, Xinmang River, Qing River, Sishui River and Jindi River)	7-10/2006	10
	S24-S27	The intertidal zones of the Yellow River Delta	4/2014, 9/2014	11
Huai River	S28-S29	Shangdong reach of the Huai River	11/2016	21
	S30-S31	Huai River tributary (Weihe River)	June	20
Songhua River	S32-S33	Harbin reach of Songhua River	NA	24
	S34-S37	Middle part of the Songhua River; sample sites included 8 mainstreams and 11 tributaries	1/2016,5/2016,7/2016,10/2016	26
	S38-S49	Jilin section of Songhua River and Songhua River tributary（Huifa River, Yinma River and Yitong River）	4/2018,7/2018/2018,10/2018,4/2019,7/2019,10/2019	25
	S50	Harbin reach of Songhua River	March, June, September, December	23
Liao River	S51-S52	Liao River Upstream(Dahuofang reservoir, guangyinge reservori) and Liao River downstream (Bohai Sea, Liaodong Bay)	26/10/2011-30/10/2011	27
	S53-S54	mainstream and tributaries of Liao River(Daliao River , Hun River andTaizi River)		28
	S55-S56	Tributary, upstream, in urban sections and downstream in the Liao River Basin	5/2012	29
	S57-S60	Mainstream and estuary of Liao River	5/2015,10,2015 october	32
	S61-S62	Upper river of the Liao River in Jilin Province(Dongliao River, Tiaozi River, and Zhaosutai River)	7/2015,11/2015	30
	S63-S64	Tributary of Liao River (Shenfu section of Hun River)	2/7/2018	31

	S65-S68	Three Pearl River tributaries (Liuxi River, Zhujiang River and Shijing River)	NA	14
Pearl River	S69-S72	Guangzhou Section of Pearl River	12/2015,4/2016	12
	S73-S74	Part of Pearl River	NA	13
	S75-S78	Pearl River Estuary (Shiziyang channel)	1/2011, 8/2010	15
	S79-S80	Pearl River estuary (rivers and coastal areas of Zhuhai City)	NA	16
Hai River	S81-S82	Tributary rivers of Hai River (Daqing River, Beiyun River, Miyun Reservoir, Jiyun River, Chaobai River, Zhangwei River and Baiyangdian Lake)	8/2007-11/2015	19
	S83-S86	mainstream and tributaries of Hai River Basin	8/2009,12/2009, 8/2010	18
	S87-S88	Hai River mainstream and tributaries(Dagu Drainage River, Chentaizi Drainage River, Duliujian River)	9/2010	17

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