#### Supplementary Information for

### Occurrence, distribution and bioaccumulation of antibiotics in the Haihe River in China

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This file contains six tables and detailed description of sample pretreatment and instrumental methods.

Table S1 Experimental conditions of electrospray ionization tandem mass

spectrometry for the determination of 22 antibiotics and 7 surrogates

Table S2 Correlation coefficients  $(r^2)$ , linear range, method detection limits (MODs,

S/N = 3) and recovery rates of 22 antibiotics

 Table S3 Concentrations of detectable antibiotics in surface water (2010)

Table S4 Concentrations of detectable antibiotics in sediments

**Table S5** Sediment-water distribution coefficient  $(K_d)$  of antibiotics in rivers

Table S6 Concentrations of detectable antibiotics in fish samples

		Parent	Daughter	Declustering	Entrance	Collision cell	Collision	Collision cell
Groups	Analytes	ion(m/z)	ion(m/z)	Potential/V	Potential/V	ent potential/V	Energy/eV	exit potential/V
fluoroquinolones	NOR	320.1	276.3	45	8.0	11	23	10.0
			302.2 <sup>a</sup>	40	8.0	11	28	10.0
	CIP	332.1	231.1	58	4.5	14	49	9.5
			314.3 <sup>a</sup>	55	5.0	11	28	6.0
	DIF	400.0	299.1	60	4.0	13	41	12.0
			356.2 <sup>a</sup>	60	4.0	13	28	13.0
	ENR	360.0	245.2	55	5.0	12	39	9.5
			316.2 <sup>a</sup>	58	5.0	11	28	11.0
	FLE	370.0	269.2	45	4.5	20	34	10.0
			326.2 <sup>a</sup>	45	4.5	12	27	12.0
	OFL	362.2	261.2	55	5.0	24	38	10.0
			318.2 <sup>a</sup>	55	5.0	12	27	11.0
	LOM	352.0	265.2 <sup>a</sup>	53	5.0	12	34	10.0
			308.2	55	4.0	37	32	12.0
	SAR	386.0	299.2	65	4.5	13	37	6.5
			368.2 <sup>a</sup>	60	4.5	14	31	7.0
	NOR-d <sub>5</sub>	325.3	281.4	50	8.5	11	23	6.0
			307.3 <sup>a</sup>	40	7.5	11	27	6.5
	OFL-d <sub>3</sub>	365.2	261.2	55	6.0	12	39	10.0
			321.2 <sup>a</sup>	55	6.0	12	28	11.5
	SAR-d <sub>8</sub>	394.2	350.3	60	6.0	13	28	12.0
			376.2 <sup>a</sup>	59	6.0	12	33	14.0
sulfonamides	ST	256.0	108.0	43	4.5	10	36	4.5
			156.0 <sup>a</sup>	42	4.2	10	21	4.5
	SMX	254.0	156.0 <sup>a</sup>	45	4.0	9	23	5.2
			160.1	47	4.5	9	27	6.0
	SIA	268.1	108.0	46	4.5	12	37	4.5
			156.0 <sup>a</sup>	46	4.5	9	21	5.0
	SPD	250.1	108.0	38	7.0	11	36	4.5
			156.0 <sup>a</sup>	41	4.7	9	24	5.5
	SDM	311.2	108.0	55	4.5	12	41	4.5
			156.1 <sup>a</sup>	57	4.0	12	30	6.0
	SMZ	279.2	156.0	47	4.5	9	27	6.0
			186.1 <sup>a</sup>	49	4.0	10	25	6.5

## **Table S1** Experimental conditions of electrospray ionization tandem massspectrometry for the determination of 22 antibiotics and 7 surrogates

		Parent	Daughter	Declustering	Entrance	Collision cell	Collision	Collision cell
Groups	Analytes	ion	ion	Potential/	Potential/	ent potential/	Energy/	exit potential/
		(m/z)	(m/z)	V	V	V	eV	v
sulfonamides	SD	251.1	108.0	42	4.3	11	35	4.2
			156.0 <sup>a</sup>	43	3.5	9	23	5.0
	SMR	265.2	107.9 <sup>a</sup>	47	4.5	11	37	4.5
			156.0	48	5.0	10	24	5.5
	SMM	281.2	156.0 <sup>a</sup>	50	5.0	10	26	5.5
			215.1	48	5.0	9	25	7.0
	SMZ-d <sub>4</sub>	283.1	160.0 <sup>a</sup>	48	4.5	10	28	6.0
			186.0	48	4.5	10	26	6.5
	SMX-d <sub>4</sub>	257.9	112.0	44	4.5	11	36	4.8
			160.0 <sup>a</sup>	44	4.5	9	25	5.5
macrolides	SPI	843.4	174.1 <sup>a</sup>	87	10.0	30	50	8.0
			318.3	85	9.5	30	42	8.0
	JOS	828.3	174.2 <sup>a</sup>	79	10.0	30	47	7.5
			229.1	77	10.0	28	43	8.5
	TYL	916.3	174.0 <sup>a</sup>	83	10.0	33	55	8.0
			772.3	85	9.0	29	43	17.0
	ERY	734.3	158.2 <sup>a</sup>	60	4.8	27	41	6.0
			576.3	58	4.8	23	28	12.0
	ROX	837.4	158.1 <sup>a</sup>	62	7.0	30	48	7.0
			679.4	62	6.5	26	30	14.0
	SPI I-d <sub>3</sub>	846.4	174.1 <sup>a</sup>	85	10.0	30	52	8.0
			432.3	30	4.5	27	18	15.0
	ERY- <sup>13</sup> C,d <sub>4</sub>	738.0	162.1 <sup>a</sup>	55	5.0	25	45	7.0
			580.3	55	5.0	23	28	12.0

## **Table S1** Experimental conditions of electrospray ionization tandem massspectrometry for the determination of 22 antibiotics and 7 surrogates (continued)

<sup>a</sup> quantitative ion.

						MODs			Recovery (%)	
Groups	Analytes	Surrogates	r <sup>2</sup>	Linearity range	Water (ng/L)	Sediment (µg/kg, dw)	Fish (µg/kg, dw)	Water	Sediment	Fish
fluoroquinolones	NOR	NOR-d <sub>5</sub>	0.9995	0.1-500	0.25	0.5	0.1	87.5	87.6	90.8
	CIP	NOR-d <sub>5</sub>	0.9969	0.05-500	0.1	0.2	0.1	63.2	64.8	78.2
	DIF	OFL-d <sub>3</sub>	0.9939	0.05-500	0.1	0.2	0.1	59.2	55.5	77.9
	ENR	OFL-d <sub>3</sub>	0.9975	0.05-500	0.1	0.2	0.2	70.7	84.4	89.1
	FLE	OFL-d <sub>3</sub>	0.9980	0.1-500	0.15	0.3	0.2	61.0	108.4	96.2
	OFL	OFL-d <sub>3</sub>	0.999	0.1-500	0.2	0.4	0.1	61.7	102.2	90.7
	LOM	OFL-d <sub>3</sub>	0.9947	0.05-500	0.1	0.2	0.3	65.4	71.7	79.5
	SAR	SAR-d <sub>8</sub>	0.9992	0.05-500	0.1	0.2	0.2	72.4	113.0	91.8
sulfonamides	ST	SMX-d <sub>4</sub>	0.9958	0.01-500	0.015	0.03	0.05	84.2	174.0	100.5
	SMX	SMX-d <sub>4</sub>	0.9993	0.1-500	0.15	0.3	0.05	117.0	92.2	96.8
	SIA	SMX-d <sub>4</sub>	0.9969	0.05-500	0.05	0.1	0.06	88.3	85.3	120.1
	SPD	SDMD-d <sub>4</sub>	0.9985	0.05-500	0.045	0.09	0.06	95.3	159.8	99.3
	SDM	SDMD-d <sub>4</sub>	0.9992	0.01-500	0.01	0.02	0.06	121.0	59.6	108.7
	SMZ	SDMD-d <sub>4</sub>	0.9984	0.01-500	0.02	0.04	0.06	121.0	106.1	104.9
	SD	SDMD-d <sub>4</sub>	0.9969	0.1-500	0.15	0.3	0.2	121.0	206.6	92.8
	SMR	SDMD-d <sub>4</sub>	0.9993	0.05-500	0.05	0.1	0.1	93.8	147.2	103.6
	SMM	SDMD-d <sub>4</sub>	0.9957	0.05-500	0.1	0.2	0.1	124.0	81.8	101.5
macrolides	SPI	SPI I-d <sub>3</sub>	0.9972	0.1-500	0.1	0.2	1.0	76.0	91.9	106.3
	JOS	SPI I-d <sub>3</sub>	0.9927	0.05-500	0.05	0.1	0.9	126.0	47.1	85.5
	TYL	SPI I-d <sub>3</sub>	0.9924	0.05-500	0.05	0.1	1.0	131.0	39.0	85.5
	ROX	SPI I-d <sub>3</sub>	0.9996	0.05-500	0.1	0.2	0.3	119.0	36.2	71.8
	ERY	ERY- <sup>13</sup> C, $d_3$	0.9810	0.1-500	0.15	0.3	0.1	130.0	89.7	94.8

# **Table S2** Correlation coefficients ( $r^2$ ), linear range, method detection limits (LODs, S/N = 3) and recovery rates of 22 antibiotics

				Table	<b>55</b> CU	neemiai				Jones III	Surrace		$(\Pi g/L)$	2010)					
Sites	NOR	CIP	DIF	ENR	FLE	OFL	LOM	SAR	ST	SMX	SPD	SMZ	SD	SMR	SMM	SPI	TYL	ROX	ERY
Haihe	River, m	ainstrea	m																
S5	82.5	8.7	nd <sup>a</sup>	nd	5.7	95.2	nd	nd	nd	137.5	5.4	21.8	31.9	nd	7.3	nd	nd	24.3	9.2
<b>S</b> 6	99.4	9.6	nd	nd	1.3	53.7	0.9	nd	nd	137.0	4.4	30.7	38.8	nd	7.9	nd	nd	15.4	10.3
<b>S</b> 7	128.9	13.4	nd	1.0	7.3	37.0	nd	nd	nd	137.0	5.5	44.0	52.0	nd	8.9	nd	nd	15.0	10.2
<b>S</b> 8	nd	19.7	nd	1.0	nd	22.9	nd	11.9	nd	135.5	3.2	33.6	41.3	nd	8.7	nd	nd	4.7	8.9
S9	35.3	1.1	nd	0.7	1.6	14.9	nd	10.8	nd	136.5	nd	49.6	48.1	nd	14.2	nd	nd	3.8	3.4
S10	58.5	19.2	0.9	nd	nd	9.1	nd	7.5	nd	201.0	nd	53.5	43.2	nd	19.4	nd	nd	5.5	3.1
S11	55.4	14.7	3.4	nd	nd	30.9	nd	nd	nd	54.5	nd	10.0	18.7	nd	15.2	nd	nd	10.0	3.5
S12	117.7	15.3	nd	0.7	3.1	49.4	nd	9.8	nd	63.0	1.3	6.6	17.2	nd	13.7	nd	nd	14.9	5.6
S13	11.6	nd	nd	nd	0.7	14.3	nd	nd	nd	22.1	nd	nd	3.1	nd	2.3	1.0	nd	1.5	4.1
Mean	65.5	11.3	0.5	0.4	2.2	36.4	0.1	4.4	nd	113.8	2.2	27.7	32.7	nd	10.8	0.1	nd	10.5	6.5
Med <sup>b</sup>	58.5	13.4	nd	nd	1.3	30.9	nd	nd	nd	136.5	1.3	30.7	38.8	nd	8.9	nd	nd	10.0	5.6
Freq <sup>c</sup>	8/9	8/9	2/9	4/9	6/9	9/9	1/9	4/9	0/9	9/9	5/9	8/9	9/9	0/9	9/9	1/9	0/9	9/9	9/9
Haihe	River, tri	ibutaries	5																
<b>S</b> 1	53.5	7.2	nd <sup>a</sup>	nd	9.8	11.5	nd	10.4	nd	18.6	nd	3.0	5.9	nd	nd	nd	nd	1.9	2.0
S2	94.0	35.4	nd	nd	1.4	14.1	nd	20.2	nd	93.5	3.0	8.7	52.0	nd	6.8	nd	nd	4.4	3.4
S3	28.7	16.0	nd	nd	nd	8.2	nd	nd	nd	32.1	nd	nd	2.5	nd	nd	nd	nd	2.4	1.9
S4	nd	24.1	nd	5.1	5.8	37.0	nd	142.0	nd	211.0	5.4	6.2	71.5	nd	nd	1.2	nd	20.2	10.1
S14	91.3	59.0	nd	nd	2.5	72.3	nd	41.1	nd	102.0	14.0	60.5	41.9	nd	10.0	nd	nd	13.2	8.0
S15	104.9	39.5	nd	2.2	3.6	112.2	nd	22.3	nd	74.5	4.7	49.4	39.7	nd	16.1	nd	nd	128.0	27.6
Mean	62.1	30.2	nd	1.2	3.8	42.5	nd	39.3	nd	88.6	4.5	21.3	35.6	nd	5.5	0.2	nd	28.3	8.8
Med	72.4	29.7	nd	nd	3.0	25.5	nd	21.2	nd	84.0	3.9	7.4	40.8	nd	3.4	nd	nd	8.8	5.7
Freq	5/6	6/6	0/6	2/6	5/6	6/6	0/6	5/6	0/6	6/6	4/6	5/6	6/6	0/6	3/6	1/6	0/6	6/6	6/6
Dagu I	Drainage	River																	
S16	301.0	22.4	nd	nd	nd	529.9	3.9	nd	1.9	660.0	135.5	13.9	82.5	nd	3.2	1.7	nd	333.5	52.0

**Table S3** Concentrations of detectable antibiotics in surface water (ng/L) (2010)

Sites	NOR	CIP	DIF	ENR	FLE	OFL	LOM	SAR	ST	SMX	SPD	SMZ	SD	SMR	SMM	SPI	TYL	ROX	ERY
S17	345.5	66.0	nd	nd	5.2	386.4	5.8	nd	nd	570.0	56.0	12.9	62.5	nd	3.4	nd	nd	965.0	54.0
S18	275.2	69.3	nd	1.2	17.6	216.9	nd	9.3	3.4	550.0	68.0	10.7	72.5	nd	5.8	nd	nd	595.0	77.5
S19	82.9	27.4	nd	nd	7.1	51.2	nd	nd	3.3	346.0	14.4	14.7	44.6	nd	4.6	nd	nd	181.5	32.2
S20	99.6	19.7	nd	nd	nd	22.8	nd	nd	nd	266.5	13.8	37.4	165.5	nd	8.6	nd	nd	494.0	32.0
S21	243.1	37.1	10.1	nd	nd	27.8	nd	68.6	nd	222.0	11.1	34.6	84.5	nd	11.1	nd	nd	253.5	16.2
Mean	224.5	40.3	1.7	0.2	5.0	205.8	1.6	13.0	1.4	435.8	49.8	20.7	85.4	nd	6.1	0.3	nd	470.4	44.0
Med	259.1	32.2	nd	nd	2.6	134.1	nd	nd	1.0	448.0	35.2	14.3	77.5	nd	5.2	nd	nd	413.8	42.1
Freq	6/6	6/6	1/6	1/6	3/6	6/6	2/6	2/6	3/6	6/6	6/6	6/6	6/6	0/6	6/6	1/6	0/6	6/6	6/6
Chenta	aizi Draiı	nage Riv	ver																
S22	463.9	15.6	4.9	3.5	32.8	292.7	39.4	nd	nd	216.0	102.5	5.2	25.8	nd	3.3	nd	75.0	940.0	83.5
S23	650.0	18.6	nd	nd	4.8	301.4	3.2	nd	nd	413.0	37.7	6.0	53.0	2.1	4.3	nd	2.8	1265.0	67.0
S24	405.4	18.2	nd	2.8	nd	332.7	nd	19.1	nd	520.0	29.2	10.8	34.9	3.5	4.3	nd	nd	1095.0	94.8
S25	329.8	32.5	nd	11.2	12.9	176.8	nd	nd	11.2	575.0	26.2	114.5	33.6	nd	4.6	nd	nd	885.0	64.0
Mean	462.3	21.2	1.2	4.4	12.6	275.9	10.7	4.8	2.8	431.0	48.9	34.1	36.8	1.4	4.1	nd	19.4	1046.3	77.3
Med	434.6	18.4	nd	3.1	8.8	297.0	1.6	nd	nd	466.5	33.5	8.4	34.2	1.1	4.3	nd	1.4	1017.5	75.3
Freq	4/4	4/4	1/4	3/4	3/4	4/4	2/4	1/4	1/4	4/4	4/4	4/4	4/4	2/4	4/4	0/4	2/4	4/4	4/4
Duliuji	ian River	•																	
S26	255.7	11.6	nd	2.0	1.2	89.4	nd	nd	nd	361.0	16.7	111.0	32.9	nd	8.6	nd	nd	473.0	41.5
S27	nd	51.0	nd	6.2	nd	49.2	nd	nd	nd	76.0	nd	10.5	20.2	0.8	12.6	nd	nd	65.5	20.5
Fish po	onds																		
F2	32.1	nd	nd	nd	nd	48.4	nd	nd	nd	24.9	nd	nd	3.3	nd	nd	nd	nd	2.5	2.0
F3	236.8	28.8	10.3	nd	nd	34.9	0.9	nd	nd	39.4	nd	6.8	10.8	4.4	nd	nd	nd	2.8	2.5

<sup>a</sup>: not detected; <sup>b</sup>: median; <sup>c</sup>: frequency.

					Table S	54 Concei	ntrations (	of detect	able an	tibiotics	s in seai	ments (	ug/kg, d	W)					
Sites	NOR	CIP	DIF	ENR	FLE	OFL	LOM	SAR	ST	SIA	SPD	SDM	SMZ	SD	SMM	SPI	JOS	ROX	ERY
Haihe R	iver, mains	tream, 20	10																
S6	125.5	24.7	nd <sup>a</sup>	69.7	nd	33.4	3.8	nd	nd	nd	2.0	nd	3.1	nd	nd	nd	nd	16.8	nd
<b>S</b> 8	103.3	11.0	nd	2.7	nd	64.6	2.7	nd	nd	nd	0.6	nd	5.0	nd	nd	nd	nd	40.2	nd
<b>S</b> 9	32.3	18.1	6.3	15.5	7.1	26.4	4.0	9.5	nd	nd	nd	nd	1.2	nd	nd	7.0	0.9	8.2	4.4
S10	141.0	88.3	9.7	23.8	24.2	71.3	16.2	35.9	nd	nd	0.6	nd	5.8	nd	0.9	2.4	nd	5.9	nd
S11	56.6	6.5	nd	nd	nd	47.9	nd	nd	nd	nd	nd	nd	1.5	nd	nd	nd	nd	0.7	nd
S12	3.9	nd	2.3	1.0	nd	nd	nd	nd	nd	nd	nd	nd	3.6	nd	nd	nd	nd	1.1	5.8
S13	7.7	13.8	16.9	26.1	5.7	13.8	7.2	16.3	0.2	nd	nd	nd	0.5	nd	nd	4.3	nd	nd	nd
Mean	67.2	23.2	5.0	19.8	5.3	36.8	4.8	8.8	nd	nd	0.5	nd	2.9	nd	0.1	2.0	0.1	10.4	1.5
Med <sup>b</sup>	56.6	13.8	2.3	15.5	nd	33.4	3.8	nd	nd	nd	nd	nd	3.1	nd	nd	nd	nd	5.9	nd
Freq <sup>c</sup>	7/7	6/7	4/7	6/7	3/7	6/7	5/7	3/7	1/7	0/7	3/7	0/7	7/7	0/7	1/7	3/7	1/7	6/7	2/7
Haihe R	iver, tribut	aries, 201	0																
<b>S</b> 1	118.5	29.4	nd <sup>a</sup>	5.3	5.1	27.3	3.2	nd	nd	nd	5.8	nd	4.2	nd	nd	1.6	nd	2.6	nd
S2	1135.5	104.6	nd	13.2	6.6	181.5	15.9	nd	nd	nd	1.7	nd	4.9	1.6	1.1	nd	nd	31.1	nd
S3	19591.0	1930.0	nd	19.0	160.0	5807.5	1340.0	nd	nd	nd	11.9	nd	0.9	1.2	nd	nd	nd	55.2	nd
S4	252.5	29.0	nd	2.1	3.3	67.0	12.4	nd	nd	nd	1.0	nd	1.6	1.1	nd	nd	nd	6.7	40.7
S14	164.0	24.3	nd	5.8	4.0	208.5	5.6	nd	nd	nd	0.7	nd	4.6	nd	nd	nd	nd	9.5	nd
S15	70.3	12.7	nd	2.4	nd	56.0	3.1	nd	nd	nd	nd	nd	0.1	nd	nd	nd	nd	154.3	14.9
Mean	3555.3	355.0	nd	8.0	29.8	1058.0	230.0	nd	nd	nd	3.5	nd	2.7	0.6	0.2	0.3	nd	43.2	9.3
Med	208.3	29.2	nd	5.6	4.5	124.2	9.0	nd	nd	nd	1.4	nd	2.9	0.5	nd	nd	nd	20.3	nd
Freq	6/6	6/6	0/6	6/6	5/6	6/6	6/6	0/6	0/6	0/6	5/6	0/6	6/6	3/6	1/6	1/6	0/6	6/6	2/6
Haihe R	iver, mains	tream, 20	08																
S5	92.2	11.7	nd	nd	nd	72.0	nd	nd	nd	nd	nd	nd	8.4	1.1	0.9	nd	nd	7.2	nd
<b>S</b> 6	128.2	25.4	nd	3.4	nd	182.1	5.3	nd	nd	nd	nd	nd	4.3	1.1	nd	nd	nd	15.8	nd
<b>S</b> 7	84.6	11.5	nd	4.3	nd	50.2	nd	nd	nd	nd	nd	nd	7.6	nd	1.3	nd	nd	4.2	nd

Table C4 Ca , <u>,</u>. a of data atable antibiotion in addimenta (us/lea dur)

Sites	NOR	CIP	DIF	ENR	FLE	OFL	LOM	SAR	ST	SIA	SPD	SDM	SMZ	SD	SMM	SPI	JOS	ROX	ERY
<b>S</b> 8	69.7	12.8	nd	2.9	4.4	29.0	nd	nd	nd	nd	nd	nd	6.9	0.7	1.8	nd	nd	2.0	nd
S10	70.7	11.8	nd	nd	5.7	41.5	3.2	nd	nd	nd	nd	nd	1.1	nd	nd	nd	nd	9.8	nd
S11	40.4	nd	nd	nd	nd	6.0	nd	nd	nd	nd	nd	nd	1.0	81.2	nd	nd	nd	nd	nd
S12	1024.8	159.0	nd	nd	nd	259.2	24.6	nd	nd	nd	nd	nd	1.8	nd	nd	nd	nd	104.6	nd
S13	129.0	27.6	nd	13.7	3.6	86.6	0.6	nd	nd	nd	nd	nd	4.4	1.4	1.6	nd	nd	2.6	nd
Mean	205.0	32.5	nd	3.0	1.7	90.8	4.2	nd	nd	nd	nd	nd	4.4	10.7	0.7	nd	nd	18.3	nd
Med	88.4	12.3	nd	1.4	nd	61.1	0.3	nd	nd	nd	nd	nd	4.4	0.9	0.4	nd	nd	5.7	nd
Freq	8/8	7/8	0/8	4/8	3/8	8/8	4/8	0/8	0/8	0/8	0/8	0/8	8/8	4/8	4/8	0/8	0/8	7/8	0/8
Haihe R	iver, tribut	aries, 200	8																
<b>S</b> 3	837.2	89.1	nd	nd	4.6	274.1	21.4	nd	nd	nd	1.2	nd	10.2	1.6	nd	nd	nd	8.8	nd
S14	80.1	5.9	nd	nd	nd	34.0	2.4	nd	nd	nd	nd	nd	0.9	nd	nd	nd	nd	0.8	nd
S15	108.8	32.1	nd	nd	nd	31.4	6.7	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	4.9	nd
Mean	342.0	42.4	nd	nd	1.5	113.2	10.2	nd	nd	nd	0.4	nd	3.7	0.5	nd	nd	nd	4.9	nd
Med	108.8	32.1	nd	nd	nd	34.0	6.7	nd	nd	nd	nd	nd	0.9	nd	nd	nd	nd	4.9	nd
Freq	3/3	3/3	0/3	0/3	1/3	3/3	3/3	0/3	0/3	0/3	1/3	0/3	2/3	1/3	0/3	0/3	0/3	3/3	0/3
Dagu Dr	ainage Riv	ver, 2010																	
S16	9331.0	688.0	nd	38.6	164.0	5427.5	637.0	nd	nd	nd	14.7	nd	nd	3.7	nd	nd	nd	30.3	56.8
S17	2872.3	250.6	nd	9.1	21.7	1248.2	114.0	nd	nd	nd	4.9	nd	nd	nd	nd	nd	nd	22.0	nd
S18	509.4	69.3	11.0	25.2	11.1	331.9	18.7	10.6	0.4	nd	4.1	nd	0.7	nd	nd	29.2	nd	24.1	4.8
S19	100.0	9.8	nd	nd	nd	90.3	2.2	nd	2.3	nd	1.2	nd	2.3	nd	nd	nd	nd	7.1	nd
S20	299.0	99.7	12.2	36.7	36.1	178.5	30.6	34.0	3.2	nd	1.1	1.0	0.7	1.1	nd	nd	nd	31.6	nd
S21	242.3	26.9	nd	nd	nd	129.2	4.5	nd	nd	0.7	0.9	nd	10.3	nd	nd	nd	nd	15.7	nd
mean	2225.7	190.7	3.9	18.3	38.8	1234.3	134.5	7.4	1.0	0.1	4.5	0.2	2.3	0.8	nd	4.9	nd	21.8	10.3
med	404.2	84.5	nd	17.1	16.4	255.2	24.7	nd	0.2	nd	2.6	nd	0.7	nd	nd	nd	nd	23.0	nd
Freq	6/6	6/6	2/6	4/6	4/6	6/6	6/6	2/6	3/6	1/6	6/6	1/6	4/6	2/6	0/6	1/6	0/6	6/6	2/6

ROX         ERY           4.0         nd           7.2         2.3           25.3         nd           13.9         nd           20.3         nd           14.1         0.5           13.9         nd           5/5         1/5           21.1         nd	JOS nd nd nd nd nd nd o/5	SPI nd nd nd nd nd nd nd o/5	SMM nd nd nd nd nd nd nd o/5	SD nd nd 2.6 4.1 2.9 1.9 2.6	SMZ 0.3 0.4 0.5 2.3 1.2 1.0 0.5	SDM nd nd nd nd nd nd nd	SPD nd nd nd nd nd nd	SIA nd nd 1.2 nd nd 0.2	ST nd nd nd nd nd	SAR nd nd nd nd	LOM nd 2.9 149.0	OFL 26.3 56.6 878.6	FLE nd 1.4 40.9	ENR nd nd	DIF nd nd	CIP er, 2008 43.6 16.1	NOR rainage Riv 99.0 158.0	Sites Dagu D S16
4.0       nd         7.2       2.3         25.3       nd         13.9       nd         20.3       nd         14.1       0.5         13.9       nd         5/5       1/5         21.1       nd	nd nd nd nd nd nd 0/5	nd nd nd nd nd nd nd 0/5	nd nd nd nd nd nd nd	nd nd 2.6 4.1 2.9 1.9 2.6	0.3 0.4 0.5 2.3 1.2 1.0 0.5	nd nd nd nd nd nd	nd nd nd nd nd	nd nd 1.2 nd nd 0.2	nd nd nd nd nd	nd nd nd nd	nd 2.9 149.0	26.3 56.6 878.6	nd 1.4 40.9	nd nd	nd nd	<b>er, 2008</b> 43.6 16.1	rainage Riv 99.0 158.0	Dagu D S16
4.0       nd         7.2       2.3         25.3       nd         13.9       nd         20.3       nd         14.1       0.5         13.9       nd         5/5       1/5         21.1       nd	nd nd nd nd nd nd 0/5	nd nd nd nd nd nd 0/5	nd nd nd nd nd nd nd	nd nd 2.6 4.1 2.9 1.9 2.6	0.3 0.4 0.5 2.3 1.2 1.0 0.5	nd nd nd nd nd nd	nd nd nd nd nd	nd nd 1.2 nd nd 0.2	nd nd nd nd	nd nd nd nd	nd 2.9 149.0	26.3 56.6 878.6	nd 1.4 40.9	nd nd	nd nd	43.6 16.1	99.0 158.0	S16
7.2       2.3         25.3       nd         13.9       nd         20.3       nd         14.1       0.5         13.9       nd         5/5       1/5         21.1       nd	nd nd nd nd nd 0/5	nd nd nd nd nd 0/5	nd nd nd nd nd 0/5	nd 2.6 4.1 2.9 1.9 2.6	0.4 0.5 2.3 1.2 1.0 0.5	nd nd nd nd nd	nd nd nd nd	nd 1.2 nd nd 0.2	nd nd nd nd	nd nd nd	2.9 149.0	56.6 878.6	1.4 40.9	nd	nd	16.1	158.0	017
25.3       nd         13.9       nd         20.3       nd         14.1       0.5         13.9       nd         5/5       1/5         21.1       nd	nd nd nd nd 0/5	nd nd nd nd 0/5	nd nd nd nd 0/5	2.6 4.1 2.9 1.9 2.6	0.5 2.3 1.2 1.0 0.5	nd nd nd nd	nd nd nd	1.2 nd nd 0.2	nd nd nd	nd nd	149.0	878.6	40.9	15				517
13.9       nd         20.3       nd         14.1       0.5         13.9       nd         5/5       1/5         21.1       nd	nd nd nd 0/5	nd nd nd 0/5	nd nd nd 0/5	4.1 2.9 1.9 2.6	2.3 1.2 1.0 0.5	nd nd nd nd	nd nd nd	nd nd 0.2	nd nd	nd	222.0			1.5	nd	320.6	3604.0	S18
20.3 nd 14.1 0.5 13.9 nd 5/5 1/5 21.1 nd	nd nd nd 0/5	nd nd nd 0/5	nd nd nd 0/5	2.9 1.9 2.6	1.2 1.0 0.5	nd nd nd	nd nd	nd 0.2	nd		255.0	4042.6	128.5	10.4	nd	950.6	11534.0	S20
14.1       0.5         13.9       nd         5/5       1/5         21.1       nd	nd nd 0/5	nd nd 0/5	nd nd 0/5	1.9 2.6	1.0 0.5	nd nd	nd	0.2		nd	141.5	2465.8	39.7	6.9	nd	622.6	6726.2	S21
13.9 nd 5/5 1/5 21.1 nd	nd 0/5	nd 0/5	nd 0/5	2.6	0.5	nd	1		nd	nd	105.3	1494.0	42.1	3.8	nd	390.7	4424.2	mean
5/5 1/5 21.1 nd	0/5	0/5	0/5			1164	nd	nd	nd	nd	141.5	878.6	39.7	1.5	nd	320.6	3604.0	med
21.1 nd			0/5	3/5	5/5	0/5	0/5	1/5	0/5	0/5	4/5	5/5	4/5	3/5	0/5	5/5	5/5	Freq
21.1 nd															10	River, 20	zi Drainage	Chentai
	nd	nd	nd	2.4	2.6	nd	14.4	3.6	nd	nd	230.0	2128.5	42.1	7.2	nd	503.6	5995.5	S22
51.1 nd	nd	nd	nd	nd	nd	nd	2.2	nd	nd	nd	13.2	196.5	nd	25.2	nd	64.2	587.0	S23
299.1 nd	nd	nd	1.2	4.4	2.7	nd	10.3	nd	nd	nd	349.0	3875.9	83.2	50.9	nd	1017.9	13885.4	S24
155.0 nd	nd	nd	nd	nd	11.3	nd	16.6	nd	nd	nd	98.5	1937.5	nd	80.7	nd	775.0	6291.0	S25
131.6 nd	nd	nd	0.3	1.7	4.2	nd	10.9	0.9	nd	nd	172.7	2034.6	31.3	41.0	nd	590.2	6689.7	mean
103.1 nd	nd	nd	nd	1.2	2.7	nd	12.3	nd	nd	nd	164.3	2033.0	21.1	38.1	nd	639.3	6143.3	med
4/4 0/4	0/4	0/4	1/4	2/4	3/4	0/4	4/4	1/4	0/4	0/4	4/4	4/4	2/4	4/4	0/4	4/4	4/4	Freq
															08	e River, 20	zi Drainage	Chentai
34.4 3.4	nd	nd	nd	1.4	0.9	nd	nd	1.0	nd	nd	13.5	525.8	4.7	2.1	nd	154.6	1376.2	S22
23.1 nd	nd	nd	nd	nd	3.1	nd	nd	nd	nd	nd	26.9	1365.8	nd	40.7	nd	1514.6	11366.2	S23
31.4 nd	nd	nd	nd	10.8	11.1	nd	24.9	nd	nd	nd	76.5	2075.8	nd	83.1	nd	957.6	6626.2	S24
101.0 nd	nd	nd	nd	nd	7.4	nd	nd	nd	nd	nd	212.5	2735.8	nd	156.8	nd	1454.6	11066.2	S25
47.5 0.9	nd	nd	nd	3.0	5.6	nd	6.2	0.3	nd	nd	82.3	1675.8	1.2	70.7	nd	1020.4	7608.7	mean
32.9 nd	nd	nd	nd	0.7	5.2	nd	nd	nd	nd	nd	51.7	1720.8	nd	61.9	nd	1206.1	8846.2	med
	0/4	0/4	0/4	2/4	4/4	0/4	1/4	1/4	0/4	0/4	4/4	4/4	1/4	4/4	0/4	4/4	4/4	Freq
4 d d d d d d	0/ nd nd nd nd nd nd nd nd nd nd nd	0/4 nd nd nd nd nd nd 0/4	1/4 nd nd nd nd nd nd 0/4	2/4 1.4 nd 10.8 nd 3.0 0.7 2/4	3/4 0.9 3.1 11.1 7.4 5.6 5.2 4/4	0/4 nd nd nd nd nd nd 0/4	4/4 nd 24.9 nd 6.2 nd 1/4	1/4 1.0 nd nd nd 0.3 nd 1/4	0/4 nd nd nd nd nd nd 0/4	0/4 nd nd nd nd nd nd 0/4	4/4 13.5 26.9 76.5 212.5 82.3 51.7 4/4	4/4 525.8 1365.8 2075.8 2735.8 1675.8 1720.8 4/4	2/4 4.7 nd nd nd 1.2 nd 1/4	4/4 2.1 40.7 83.1 156.8 70.7 61.9 4/4	0/4 008 nd nd nd nd nd nd nd 0/4	4/4 <b>River, 20</b> 154.6 1514.6 957.6 1454.6 1020.4 1206.1 4/4	4/4 zi Drainage 1376.2 11366.2 6626.2 11066.2 7608.7 8846.2 4/4	Freq Chentai S22 S23 S24 S25 mean med Freq

Sites	NOR	CIP	DIF	ENR	FLE	OFL	LOM	SAR	ST	SIA	SPD	SDM	SMZ	SD	SMM	SPI	JOS	ROX	ERY
Duliujia	n River, 20	010																	
S26	1411.0	162.0	nd	28.3	nd	802.5	31.8	nd	nd	nd	8.9	nd	3.9	5.0	2.5	nd	nd	34.8	nd
S27	45.4	5.7	nd	2.2	nd	27.5	nd	nd	nd	nd	0.7	nd	0.9	nd	nd	nd	nd	3.9	nd
Fish po	nds, 2010																		
F2	12.4	2.4	nd	9.1	nd	2.8	nd	nd	nd	nd	nd	nd	0.5	nd	nd	nd	nd	nd	nd
F3	166.0	24.3	nd	3.0	nd	10.6	nd	nd	nd	nd	nd	nd	2.8	nd	nd	nd	nd	nd	nd

<sup>a</sup>: not detected; <sup>b</sup>: median; <sup>c</sup>: frequency.

Table S5 Sediment-water distribution coefficient (K<sub>d</sub>) of antibiotics in rivers (L/kg)

Antibiotic	K <sub>d</sub>	LogK <sub>d</sub>	Antibiotic	K <sub>d</sub>	LogK <sub>d</sub>
NOR	37542	4.57	ST	409	2.61
CIP	14176	4.15	SPD	230	2.36
ENR	8128	3.91	SMZ	245	2.39
FLE	2934	3.47	SD	118	2.07
OFL	33973	4.53	SMM	195	2.29
LOM	39838	4.60	ROX	1991	3.30
SAR	2268	3.36	ERY	1338	3.13

	Fish	Sites	NOR	CIP	DIF	ENR	FLE	OFL	LOM	SAR	ST	SIA	SPD	SMZ	SMM	SPI	ERY
	crucian carp, 2008	F6	0.9	nd <sup>a</sup>	nd	nd	0.6	0.4	nd	nd	nd	nd	nd	nd	nd	278.0	nd
	common carp, 2008	F1	0.5	nd	nd	1.1	0.2	0.5	nd	nd	nd	nd	nd	nd	nd	nd	nd
		F4	nd	nd	nd	nd	nd	nd	nd	nd	0.1	0.3	nd	0.4	0.3	nd	nd
Fish ponds		F5	nd	nd	nd	nd	nd	nd	nd	nd	0.1	nd	nd	nd	nd	nd	nd
	silver carp, 2008	F1	nd	nd	nd	0.2	nd	nd	nd	1.3	nd	0.3	3.3	0.4	nd	1020.0	nd
		F4	3.7	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.1	nd	nd	nd
		F5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	1150.0	nd
	crucian carp, 2010	S5	11.6	9.3	nd	2.7	nd	nd	2.3	3.5	nd	5.2	nd	68.0	nd	nd	45.1
		S6	63.5	12.5	nd	nd	nd	nd	nd	1.8	nd	0.5	nd	15.4	nd	nd	5.0
		S9	2.5	8.1	nd	nd	nd	nd	nd	nd	nd	996.3	nd	54.2	nd	nd	27.7
	crucian carp, 2008	<b>S</b> 1	51.6	8.4	nd	50.8	nd	10.5	nd	nd	3.2	3.3	nd	0.5	nd	1.3	nd
Haihe		S3	nd	nd	0.3	nd	nd	nd	nd	nd	nd	nd	nd	0.1	nd	430.0	nd
River		S5	nd	nd	0.2	0.6	nd	nd	0.3	nd	nd	nd	nd	nd	nd	nd	nd
		<b>S</b> 7	36.9	1.7	8.2	1.4	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.6	nd
		S13	nd	nd	nd	nd	nd	nd	nd	0.7	nd	nd	nd	nd	nd	396.0	nd
	common carp, 2008	S7	1.2	nd	nd	nd	0.3	nd	nd	0.3	nd	1.0	nd	nd	nd	nd	nd
	silver carp, 2008	<b>S</b> 1	0.5	2.3	nd	nd	nd	nd	nd	nd	nd	nd	nd	1.2	nd	nd	nd

Table S6 Concentrations of detectable antibiotics in fish samples (µg/kg, dw)

<sup>a</sup>: not detected.

#### Sample pretreatment for surface water, sediment and fish muscle

Water: Water samples were filtered through nylon film (0.45  $\mu$ m) to remove particles. Before extraction with Oasis HLB cartridge (6 cc, 200 mg; Waters Corp. Milford, USA), 0.2 g of chelating reagent, Na<sub>2</sub>EDTA, and 20 ng of surrogate standards (Norfloxacin-d<sub>5</sub>, Ofloxacin-d<sub>3</sub>, Sarafloxacin-d<sub>8</sub>, Sulfamethoxazole-d<sub>4</sub>, Sulfamethazine-d<sub>4</sub>, Spiramycin I-d<sub>3</sub> and Erythromycin-<sup>13</sup>C<sub>3</sub>) were added to 200 mL water sample. The HLB cartridges were conditioned with 5 ml methanol and 5 ml ultra-pure water. After loading of samples, cartridges were washed with 15 mL of ultra-pure water and vacuum-dried for 20 min. Finally, the cartridges were eluted with 6 mL of methanol containing 5% ammonium hydroxide. All the eluates were concentrated to 1 mL with a stream of nitrogen at 35 °C, and 15  $\mu$ L of this solution was injected into the high-performance liquid chromatography-electrospray ionization tandem mass spectrometry (HPLC-ESI MS/MS) system for analysis.

Sediment: Approximately 0.1 g of the dry sludge was extracted with 15 mL of the mixture of acetonitrile and EDTA-McIlvaine buffer (pH $\approx$ 4.0) (2:1) in 50 mL polypropylene centrifuge tube (20 ng of surrogate was added). After vortex mixing for 30 s, the sludge sample was sonicated for 4 min, and then centrifuged for 10 min at 10000 rpm, and the supernatant was collected. The procedure was repeated three times. The supernatant was combined, concentrated to about 15 mL by rotatory vaporization, diluted to approximately 100 mL with ultra-pure water, and then extract by SPE with a HLB cartridge. The SPE procedure was the same as the wastewater sample. The eluate was concentrated to 1 mL with a stream of nitrogen at 35 °C, and 15 µL of this solution was injected into the high-performance liquid chromatography-electrospray ionization tandem mass spectrometry (HPLC-ESI MS/MS) system for analysis. **Fish:** 0.1 g of pretreated lyophilized fish muscle was spiked with 20 ng of surrogate before being thoroughly mixed with 7 g of diatomite from Varian (Walnut Creek, KS, USA). The mixture was put into a 34 mL stainless steel extraction cell with 100% methanol as the extracting solvent.

The ASE 350 extraction conditions were: extraction temperature, 70 °C; extraction pressure, 1500 psi; preheating period, 5min; static extraction, 10 min; final extraction volume, 60 mL; flush volume 60% of the cell volume; nitrogen purge, 120 s; and number of extraction cycles, 2. Each PLE extract was concentrated by RE-2000 rotary evaporator (Yarong, Shanghai, China) to a final volume of about 1 mL at 37 °C and 0.08 MPa in 100 mL round-bottom flasks. Immediately after concentration, the extract was transferred to a 100 mL conical tube and the round-bottom flasks were rinsed twice with 0.5 ml of methanol. After that, 100 ml of regent water was added to this conical tube, and then the solution was extracted by SPE with a HLB cartridge. The SPE procedure was the same as the wastewater sample. The eluate was concentrated to 1 mL with a stream of nitrogen at 35 °C, and 15  $\mu$ L of this solution was injected into the high-performance liquid chromatography-electrospray ionization tandem mass spectrometry (HPLC-ESI MS/MS) system for analysis.

#### Analysis methods of HPLC-MS/MS

Separation of analytes was carried out on a XTerra MS  $C_{18}$  column (2.1 mm× 100 mm i.d., 3.5 µm) with mobile phase of A, methanol: acetonitrile (1:1, V/V), and B, 0.3% formic acid/water (containing 0.1% ammonium formate, V/V, pH = 2.9). The gradient elution was performed as follows: A 10% in the first 2 min, 10-70% in 10 min, then increased to 100 % in 4min and maintained for 3 min, after that, the mobile phase was returned to the initial conditions in 0.1 min and maintained for 13.9 min. The flow rate of the mobile phase was 0.2 mL/min, and the column temperature was 25 °C. The injection volume was 15 µL. The MS/MS parameters for the instrument were optimized. Curtain gas pressure: 0.14 MPa, Collision gas pressure: 0.02 MPa, Ion spray voltage: 5000 V, Tempreture: 600 , Gas1: 0.38 MPa, Gas2: 0.45 MPa. The detailed parameters of MS/MS and ion pair of the analytes are shown in **Table S1**.