

Supporting Information

Characteristics of pharmaceutically active compounds in surface water in Beijing,

China: occurrence, spatial distribution and biennial variation from 2013 to 2017

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1. Material and methods

1.1 Target compounds

Table S1 Information of target compounds

1st Class	2nd Class	Compound	Abbreviation	CAS number	pKa*
Antibiotics	Sulfonamides	Sulfadiazine	SD	68-35-9	6.81±0.10
		Sulfathiazole	ST	72-14-0	7.24±0.10
		Sulfamerazine	SMR	127-79-7	7.35±0.10
		Sulfisoxazole	SIX	127-69-5	4.83±0.05
		Sulfisomidine	SIM	515-64-0	3.63±0.10
		Sulfamethoxypyridazine	SMP	80-35-3	7.19±0.30
		Sulfaquinoxaline	SQX	59-40-5	5.65±0.10
		Sulfamethazine	SMT	57-68-1	7.89±0.10
		Sulfadimethoxine	SDM	122-11-1	6.21±0.50
		Sulfamethoxazole	SMX	723-46-6	5.81±0.50
		Sulfamethizole	SMZ	144-82-1	5.51±0.50
		Sulfamonomethoxine	SM	1220-83-3	6.67±0.30
	Macrolides	Trimethoprim	TP	738-70-5	7.04±0.10
		Erythromycin	EM	114-07-8	13.09±0.70
		Clarithromycin	CAM	81103-11-9	13.08±0.70
		Roxithromycin	RXM	80214-83-1	13.00±0.70
	Lincoamides	Tylosin tartrate	TS	74610-55-2	-
		Clindamycin	CDM	18323-44-9	12.87±0.70
		Lincomycin	LCM	154-21-2	12.91±0.70
	Chloramphenicol	Chloramphenicol	CP	56-75-7	11.03±0.46
	Quinolones	Nalidixic acid	NA	389-08-2	3.45±0.20
Non-antibiotics	Anti-inflammatory	Diclofenac acid	DF	15307-86-5	4.18±0.10
		Indomethacin	IM	53-86-1	3.96±0.30
		Mefenamic acid	MA	61-68-7	3.73±0.36
		Ketoprofen	KP	22071-15-4	4.23±0.1
		Acetaminophen	ATP	103-90-2	9.86±0.13
	Lipid regulators	Bezafibrate	BF	41859-67-0	3.29±0.10
		Clofibric acid	CA	882-09-7	3.18±0.10
		Gemfibrozil	GF	25812-30-0	4.75±0.45
	Beta-blockers	Metoprolol	MTP	51384-51-1	13.89±0.20

	Propranolol	PHO	525-66-6	13.84±0.20
Psychiatric drugs	Carbamazepine	CBZ	298-46-4	13.94±0.20
	Sulpiride	SP	15676-16-1	9.98±0.60
Stimulant	Caffeine	CF	58-08-2	0.52±0.70
Repellent	N,N-diethyl-meta-toluamide	DEET	134-62-3	-1.37±0.70

*pKa is predicted protonation constant acquired from Scifinder database.
(<https://scifinder.cas.org/scifinder/view/scifinder/scifinderExplore.jsf>)

1.2 Beiyun River

Table S2 Information of each river basin in Beiyun River

Sub-river basin		Watershed area (km ²)	Inhabitant (10 ⁴ Capita)	Population density (Inhabitants/km ²)*
Tributaries	Sha River	1084	134.5	1241
	Lingou River	377	13.9	369
	Qing River	210	264.1	12576
	Ba River	158	142.6	9025
	Tonghui River	258	382	14806
	Liangshui River	815	319.1	3915
	Feng River	489	79	1616
Main streams	Wenyu River	407	72.3	1776
	Beiyun River	251	19.3	769

*Population density is calculated with dividing the number of inhabitant by watershed area.

1.3 Sampling sites information

Table S3 Information of sampling sites

Sampling site	Coordinate		Sampling date	Catchment
	Latitude (° N)	Longitude (° E)		
S-1	40.0773	116.1094	2017/11/30	Sha River
S-2	40.0931983	116.1743983	2017/11/30	
S-3	40.1098067	116.2038783	2017/11/30	
S-4	40.0998617	116.2066667	2017/11/30	
S-5	40.107605	116.2336267	2017/11/30	
S-6	40.1524433	116.1863883	2017/11/30	
S-7	40.1438783	116.1992883	2017/11/30	

S-8	40.1416567	116.2111017	2017/11/30	
S-9	40.143635	116.2370317	2017/11/30	
S-10	40.141075	116.2573483	2017/11/30	
LG-11	40.17194444	116.3572222	2017/11/30	
LG-12	40.1668067	116.4579267	2017/11/30	Lingou River
LG-13	40.161835	116.431465	2017/11/30	
LG-14	40.1561217	116.4312583	2017/11/30	
WY-15	40.1506617	116.4040183	2017/11/30	
WY-16	40.1313567	116.3328267	2017/11/30	
WY-38	40.093665	116.4850867	2017/12/01	
WY-39	40.0558533	116.5391867	2017/12/01	Wenyu River
WY-40	40.0308067	116.5648083	2017/12/01	
WY-41	39.9858117	116.636235	2017/12/01	
WY-42	39.9294433	116.6417867	2017/12/01	
F-17	39.72579	116.4568967	2017/11/31	
F-18	39.698945	116.51734	2017/11/31	
F-19	39.6911083	116.5542133	2017/11/31	Feng River
F-20	39.713755	116.551675	2017/11/31	
F-25	39.767085	116.7755883	2017/11/31	
F-26	39.757	116.77823	2017/11/31	
LS-21	39.7619017	116.5384517	2017/11/31	
LS-22	39.7770133	116.63001	2017/11/31	Liangshui River
LS-23	39.842645	116.699519	2017/11/31	
LS-24	39.80405834	116.7625283	2017/11/31	
BY-27	39.7636	116.8364033	2017/11/31	
BY-28	39.7926583	116.7862233	2017/11/31	Beiyun River
BY-29	39.8564217	116.753565	2017/11/31	
BY-30	39.9099	116.6750583	2017/11/31	
TH-31	39.9148717	116.656445	2017/12/01	
TH-32	39.90699	116.614265	2017/12/01	Tonghui River
TH-33	39.90511	116.50262	2017/12/01	
Q-34	40.0177067	116.3114817	2017/12/01	
Q-35	40.0287583	116.3665583	2017/12/01	Qing River
Q-36	40.0808617	116.4620817	2017/12/01	
Q-37	40.0801117	116.476565	2017/12/01	
B-43	39.9471	116.6348483	2017/12/01	
B-44	39.9701917	116.5932067	2017/12/01	Ba River
B-45	39.9683967	116.551885	2017/12/01	
B-46	39.9729217	116.5261517	2017/12/01	

Table S4 Meteorological information at the time of sampling

Date	Temperature	Weather
2017/11/30, Wednesday	-6 ~ 1 °C	Cloudy
2017/11/31, Thursday	-5 ~ 4 °C	Sunny
2107/12/01, Friday	-5 ~ 5°C	Sunny

1.4 Analysis method

The analysis method was based on our previous study (Ma et al., 2017) with minor modifications. All the pretreated samples were filtered through 0.22 µm PTFE filters (Whatman, Puradisc, 13mm) before analysis. Agilent Eclipse XDB-C18 3.5 µm, 2.1×150 mm column was used to separate different PhACs and three analytical modes (POS, NEG and IT) were applied to identify the target compounds (Zhang et al., 2018). The injection volume was 10 µL and other information of each mode was showed in Table S4.

Table S5 Column temperature and mobile phase of each mode

Mode	POS ^a	NEG ^b	IT ^c
Acquisition parameters	Electronic Spray Ion (ESI) mode, positive ionization ; multiple reaction monitoring (MRM)	ESI, negative ionization , MRM	ESI, positive ionization , MRM
Column temperature	30 °C		40 °C
Mobile phase	A= 0.01% Formic acid	A= 10 mM Ammonium acetate	A= 0.01% Formic acid
	B= 100% Methanol	B= 100% Methanol	B= 100% Acetonitrile

^a POS: positive analytical mode

^b NEG: negative analytical mode

^c IT: another positive analytical mode with different mobile phase

1.5 Quality assurance and quality control (QA & QC)

Table S6 Recovery rate and instrument detection limits (IDL), instrument quantification

limits (IQL), method detection limits (MDL) and method quantification limits (MQL)

Mode	Compounds	Quantitative		IDL (µg/L)	IQL (µg/L)	MDL (ng/L)	MQL (ng/L)
		isotopically labelled internal standard	Average recovery rate (%)*				
POS	CBZ	¹³ C-Phenacetine	106	0.05	0.18	0.05	0.18
	CF		92	0.18	0.60	0.18	0.60
	DEET		104	0.08	0.28	0.08	0.28
	MTP		112	0.22	0.72	0.22	0.72
	NA		118	1.42	4.72	1.42	4.72
	PHO		104	0.08	0.26	0.08	0.26
	SP		81	0.08	0.26	0.08	0.26
	TP		100	0.52	1.72	0.52	1.72
NEG	BF	D ₅ -Chloramphenicol	79	0.19	0.65	0.19	0.65
	CA		78	0.10	0.32	0.10	0.32
	CP		80	0.17	0.57	0.17	0.57
	DF		81	0.52	1.72	0.52	1.72
	GF	D ₆ -Gemfibrozil	86	0.08	0.26	0.08	0.26
	IM		102	0.33	1.11	0.33	1.11
	KP	D ₅ -Chloramphenicol	77	0.35	1.16	0.35	1.16
	MA	D ₆ -Gemfibrozil	83	0.31	1.03	0.31	1.03
IT	ATP	¹³ C ₂ -Erythromycin	86	0.46	1.52	0.46	1.52
	CAM		101	0.35	1.16	0.35	1.16
	CDM		77	0.08	0.27	0.08	0.27
	EM		103	0.65	2.17	0.65	2.17
	LCM		85	0.08	0.27	0.08	0.27
	RXM		82	1.00	3.33	1.00	3.33
	SD		89	0.26	0.88	0.26	0.88
	SDM		76	0.09	0.30	0.09	0.30
	SIM	¹³ C ₆ -Sulfmethazine	78	0.05	0.18	0.05	0.18
	SIX		79	0.30	1.00	0.30	1.00
	SM		79	0.18	0.60	0.18	0.60
	SMP		94	0.14	0.46	0.14	0.46
	SMR		96	0.17	0.57	0.17	0.57
	SMT		83	0.15	0.50	0.15	0.50
	SMX		79	0.29	0.98	0.29	0.98
	SMZ		80	0.42	1.40	0.42	1.40
	SQX		88	0.15	0.49	0.15	0.49
	ST		86	0.69	2.31	0.69	2.31

*Recovery rate formula:

$$\text{Recovery rate (\%)} = \frac{C(\text{spiked}) - C(\text{non-spiked})}{\text{The known spiked concentration (100 } \mu\text{g/L)}} \times 100\%$$

C (spiked): Detected concentrations of the PhAC in spiked sample (μg/L)

C (non-spiked): Detected concentrations of the PhAC in non-spiked sample (μg/L)

All concentrations in the formula specify the concentration after volume.

2. Results and discussions

2.1 Detection frequencies and concentrations

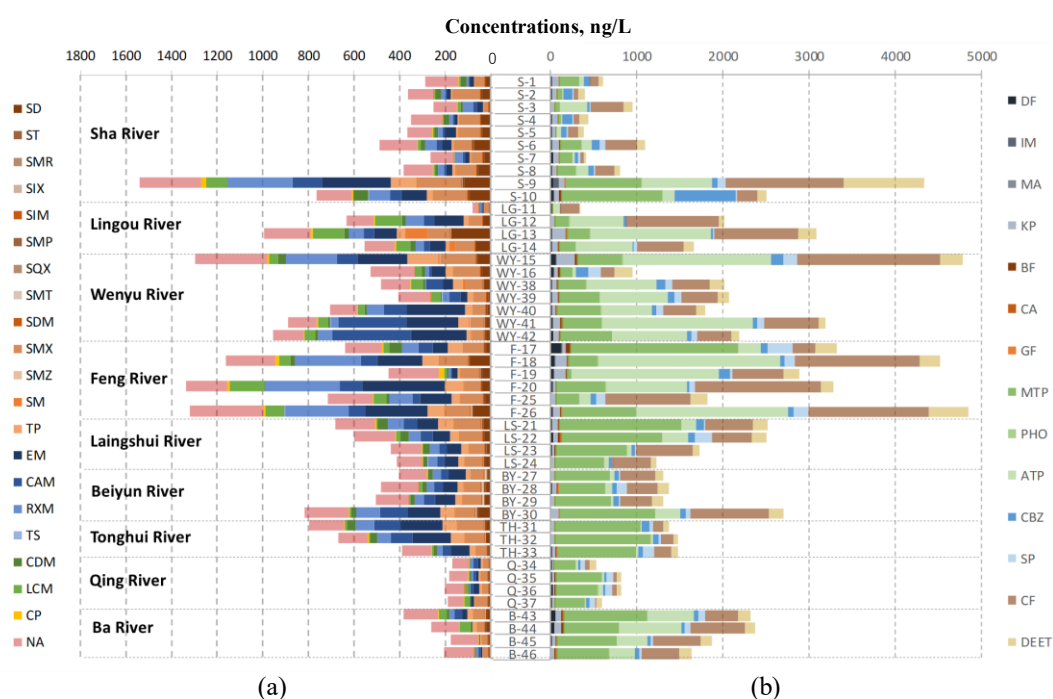


Fig. S1 Concentrations of (a) antibiotics and (b) non-antibiotics in each sampling site

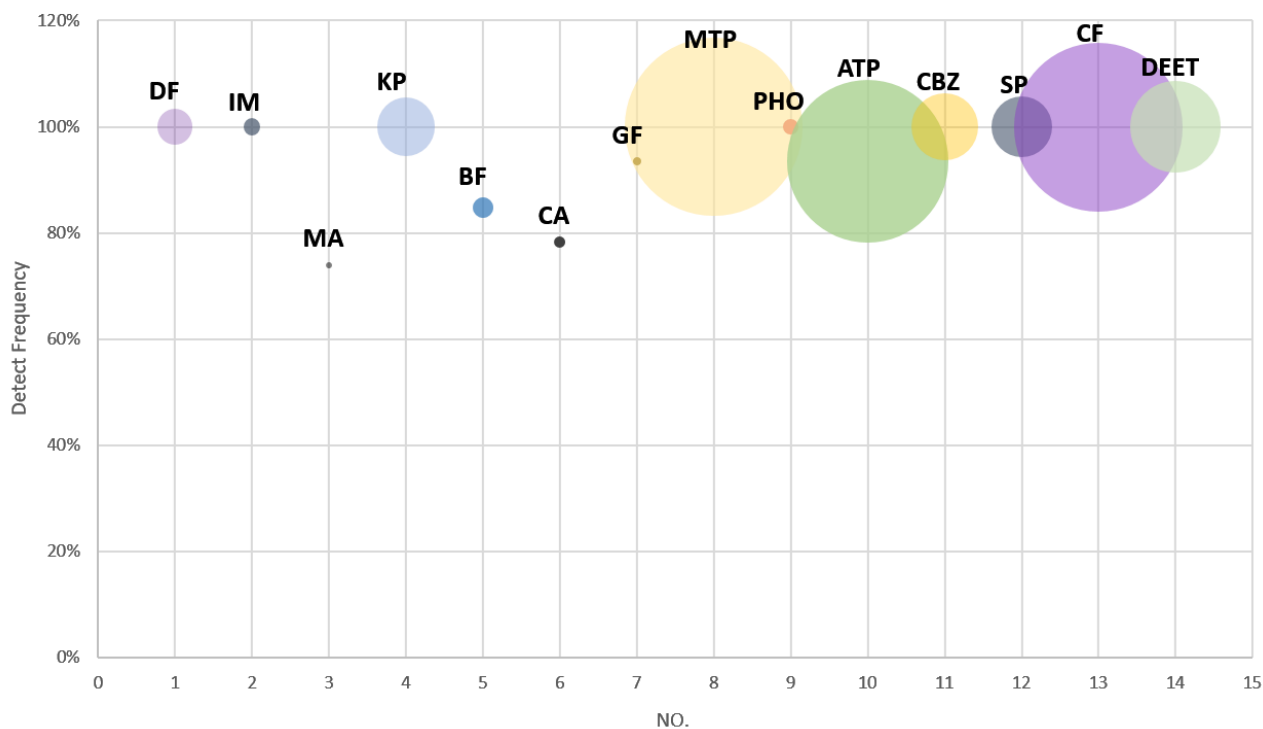


Fig. S2 Average concentrations and detection frequencies of non-antibiotics. The height of bubble is detection frequency and the size of bubble is average concentration.

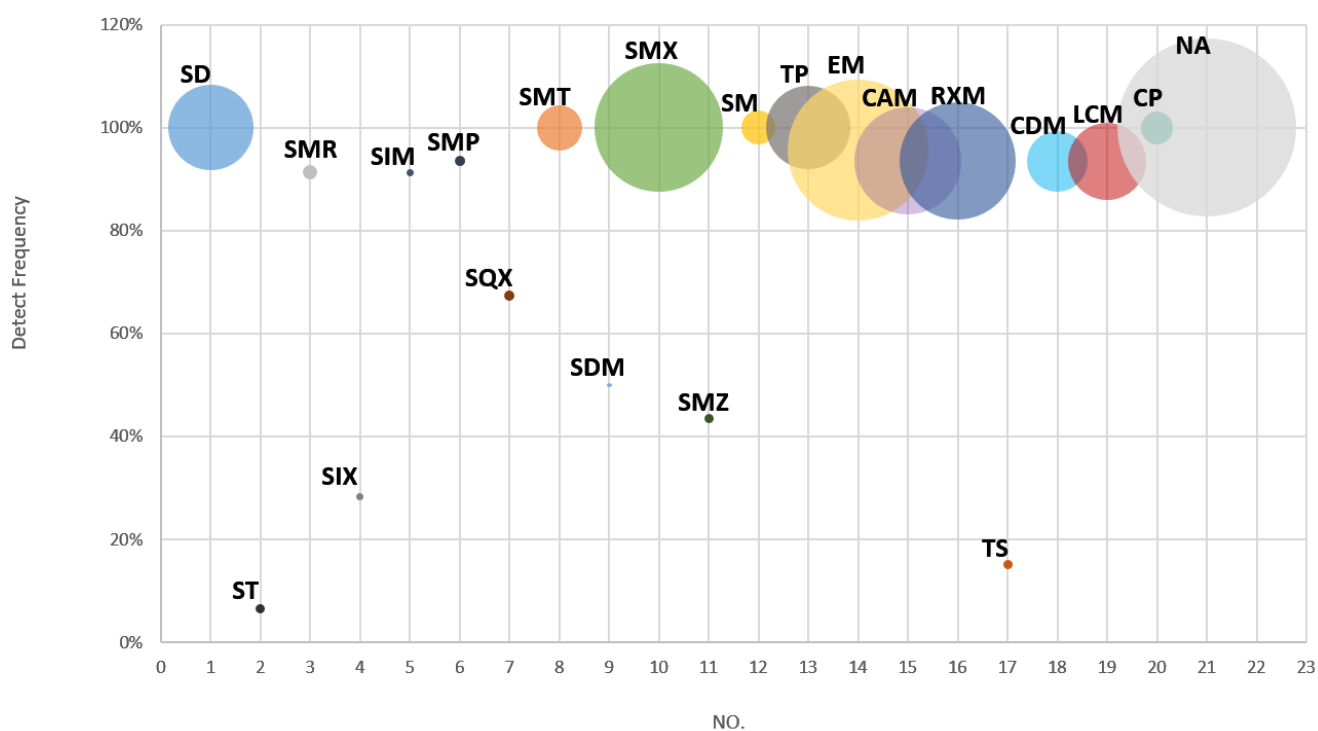


Fig. S3 Average concentrations and detection frequencies of antibiotics. The height of bubble is detection frequency and the size of bubble is average concentration.

Table S7 Concentrations of target compounds and comparison with other studies

		Concentration range (ng/L)								
		This study	Other studies in Beijing area ^a					Other studies in other area ^b		
Sampling information	Study Area	Beiyun River Basin	Beiyun River Basin	Liangshui, Yongding & Chaobai River	Beiyun River Basin	Qing & Tonghui River (Densely populated area)	Urban area river	Qinzhou Bay, South China	Xianjiang River, China	China (Review)
	Sampling Time	2017 Nov. -Dec.	2015 Jul. & Nov.	2015 Jul. & Dec.	2013 Mar., Jun. & Sep.	2012 Oct. & Dec.	2008 May. -Jul.	2017 Aug. & Dec.*	2017 Jan. & Aug.	2005-2016
Compounds	Abb.	Min-Max (Median)	Min-Max	Min-Max* (Median)	Min-Max (Median)	Mean (Median)-Max	Min-Max	Min-Max *	Min-Max **	Min-Max (Median)***
Sulfadiazine	SD	0.59-168 (23.9)	2.2-157.4	-	-	-	-	-	0.93-68	n.d.-870 (46.4)
Sulfathiazole	ST	n.d.-1.13 (0.77)	n.d.-2.1	-	-	-	-	-	-	n.d.-30.4 (0.4)
Sulfamerazine	SMR	n.d.-2.56 (0.82)	n.d.-5.3	-	-	-	-	-	-	n.d.-10.8 (n.d.)
Sulfisoxazole	SIX	0.15-1.38 (0.15)	n.d.-3.4	-	-	-	-	-	-	-
Sulfisomidine	SIM	n.d.-1.13 (0.19)	n.d.-1.3	-	-	-	-	-	-	-
Sulfamethoxypyridazine	SMP	n.d.-1.26 (0.41)	-	-	-	-	-	-	-	n.d.-630 (1.7)
Sulfaquinoxaline	SQX	n.d.-2.38 (0.26)	-	-	-	-	-	-	-	n.d.-14 (2.2)
Sulfamethazine	SMT	0.59-261 (3.28)	-	-	-	-	-	-	<MQL-60	n.d.-3900 (13)

Sulfadimethoxine	SDM	n.d.-0.31 (0.15)	n.d.-2.0	-	-	n.d.	-	-	-	n.d.-31.8 (n.d.)
Sulfamethoxazole	SMX	20.7-196 (77)	n.d.-276.2	-	-	39.6-78.9	-	-	<MQL- 100	n.d.-145290 (56.4)
Sulfamethizole	SMZ	n.d.-1.29 (0.6)	-	-	-	-	-	-	-	n.d.-3.47 (n.d.)
Sulfamonomethoxine	SM	0.24-97.9 (2.47)	-	-	-	-	-	-	-	-
Trimethoprim	TP	1.69-137 (27.1)	n.d.-140.6	91.7-528 (302)	n.d.-538.5 (54.2)	n.d.	<LOQ-48	-	1.35-93	n.d.-1515970 (46)
Erythromycin	EM	n.d.-364 (64)	n.d.-1320	-	-	16.9-22.3	269-1153	-	<MQL-43	n.d.-4200 (14.8)
Clarithromycin	CAM	n.d.-347 (33.2)	n.d.-96.9	-	-	28.4-59.7	-	-	0.55-100	0.02-59.7 (4.16)
Roxithromycin	RXM	n.d.-327 (37)	-	-	-	53.2-109	-	-	1.4-190	n.d.-3700 (47.5)
Tylosin tartrate	TS	n.d.-1.89 (1.15)	n.d.-4.1	-	-	-	-	-	-	n.d.-570 (n.d.)
Clindamycin	CDM	0.16-65.8 (15.6)	-	-	-	-	-	-	-	-
Lincomycin	LCM	0.13-152 (11.3)	-	-	-	50.4-180	-	-	-	n.d.-919530 (30.5)
Chloramphenicol	CP	0.3-25.9 (3.26)	1.1-22.5	100-249 (158.5)	n.d.-32.3 (8.2)	-	-	-	n.d.-6.1	n.d.-1700 (3.8)
Nalidixic acid	NA	30.9-323 (135)	n.d.-34.2	14.1-83.5 (45.7)	n.d.-116.0 (36.7)	-	-	-	-	n.d.-231 (1.3)
Diclofenac acid	DF	0.69-128 (16.7)	1.8-121.6	213-1300 (810)	4.3-150.6 (65.3)	-	55-636	n.d.-7.17	n.d.-32	-

Indomethacin	IM	0.48-68.7 (2.54)	n.d.-74.9	28.2-160 (58.6)	5.8-63.6 (32.6)	56.9-200	-	n.d.-1.04	n.d.-2.7	-
Mefenamic acid	MA	n.d.-4.15 (0.55)	2.0-7.3	1.4-31.3 (24.7)	n.d.-9.9 (4.2)	-	-	-	n.d.-3.9	-
Ketoprofen	KP	3.52-219 (51.6)	n.d.-65.0	-	n.d.-509.0 (67.6)	-	43-249	-	-	-
Acetaminophen	ATP	6.2-2110 (156)	n.d.-3577	-	-	-	-	-	-	-
Bezafibrate	BF	n.d.-43.3 (4.04)	1.4-42.9	23.4-169 (83.9)	n.d.-72.7 (22.6)	n.d.	-	-	-	-
Clofibric acid	CA	n.d.-13.2 (0.84)	2.3-11.8	34.5-187 (87.1)	-	-	33-187	n.d.-0.281	-	-
Gemfibrozil	GF	n.d.-11.1 (0.74)	n.d.-8.1	2.6-57.4 (20.1)	n.d.-63.4 (9.1)	-	-	n.d.-0.303	-	-
Metoprolol	MTP	8.15-1940 (524)	55.3-495.2	91.9-772 (452)	-	66.5-134	<LOQ-65	-	-	-
Propranolol	PHO	0.34-12.5 (3.93)	n.d.-4.5	n.d.-3.5 (2.7)	n.d.-37 (n.d.)	-	-	n.d.-0.142	-	-
Carbamazepine	CBZ	1.77-718 (58.7)	10.1-199.5	58.1-183.0 (93.1)	n.d.-189.0 (56.4)	12-21.3	64-665	n.d.-0.588	-	-
Sulpiride	SP	0.09-288 (50.6)	4.4-127.3	129-450 (355)	-	141.6-200	73-719	-	-	-
Caffeine	CF	12.5-1660 (390)	31.3-2714.1	689-4690 (3020)	33.3-9785 (169.5)	-	902-7051	0.703-26.8	-	-
N,N-diethyl-meta-toluamide	DEET	4.65-924 (107)	2.5-1356.1	19.2-50.7 (36.3)	30.3-546.5 (169.5)	2.3-5.8	712-2514	0.14-2.01	-	-

a Reference (Dai et al., 2015; Ma et al., 2017; Wang et al., 2015; Yang et al., 2017; Zhou et al., 2010)

b Reference(Cui et al., 2019; Li et al., 2018; Lin et al., 2018; Yang et al., 2019)

*According to the paper published, concentrations of December are showed.

** According to the paper published, concentrations of January are showed.

*** According to the paper published, concentrations of Haihe River are showed.

n.d. No detection in research.

- Not available

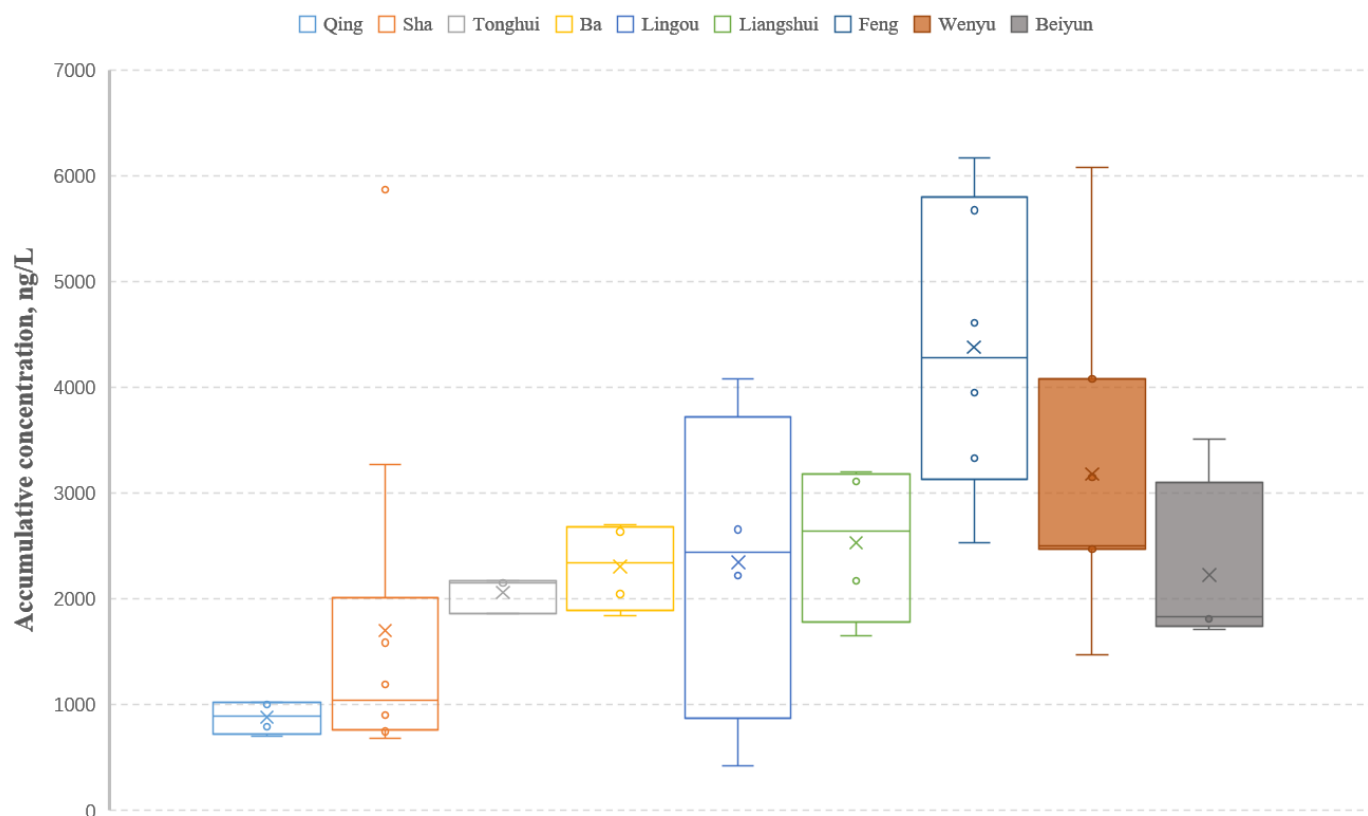


Fig. S4 Sum concentrations of main streams (Wenyu and Beiyun River) and tributaries (Qing, Sha, Tonghui, Ba, Lingou, Liangshui and Feng River)

2.2 Clustering analysis based on PhAC compositions

Heat map-hierarchical clustering **was** often used to reflect the regulation pattern of a large amount of data, for example, to observe the time variation (Zhang et al., 2018) and identify source with specific micropollutants (Carpenter and Helbling, 2018).

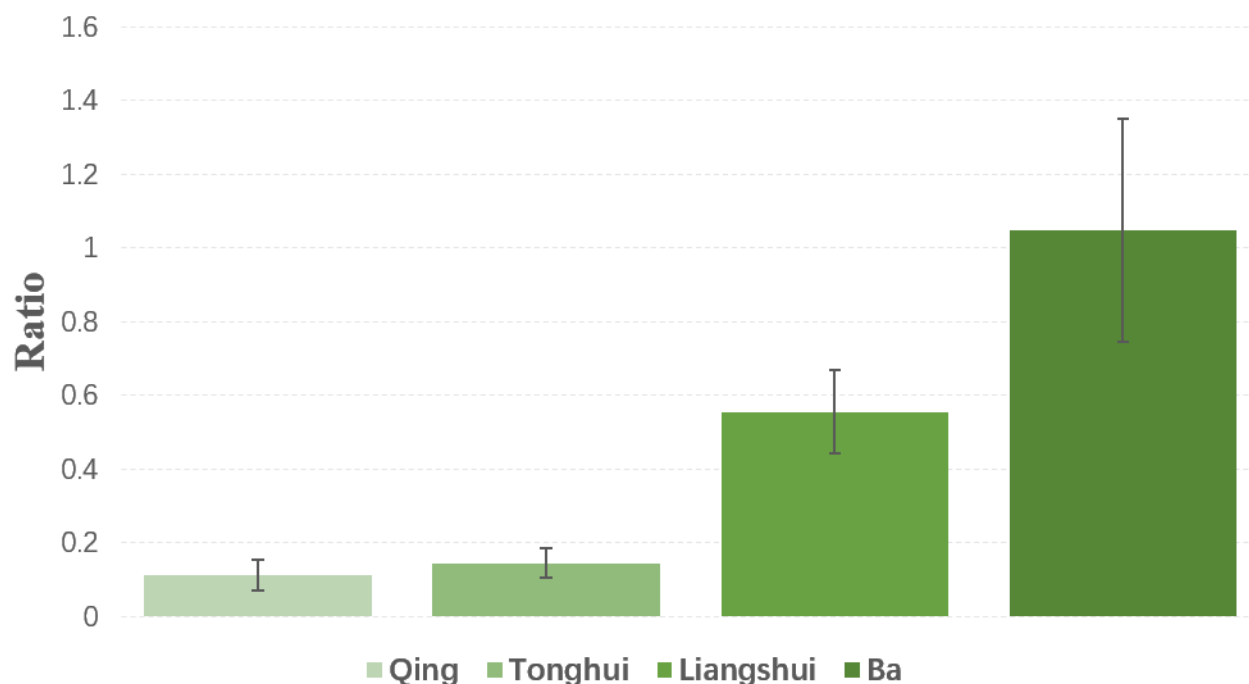


Fig. S5 Ratio of sum of concentrations of 5 easy-removal PhACs (CF, ATP, GF, CP and BF) to sum of difficult-removal groups (PHO, CBZ, TP, MTP, NA and SP) in urban rivers.

2.3 Urban wastewater treatment plants (WWTPs) and their receiving rivers

Table S8 Information of urban WWTPs and their receiving rivers

WWTP	Receiving River	Capacity (10 ⁴ m ³ /d)	Bio-treatment	Clarification	Disinfection
A	Tonghui	100	A/A/O	Sedimentation	Sodium hypochlorite
B	Qing	60	A/A/O	MBR	Ozone; Sodium hypochlorite
C	Liangshui	60	A/A/O	Sedimentation	Sodium hypochlorite
D	Ba	20	Oxidation ditch +A/O	Cloth-media filter	Ozone; Sodium hypochlorite

The information refers to Zhang et al. (Zhang et al., 2018).

Table S9 Concentrations of influent and effluent (Zhang et al., 2018) (ng/L)

Compounds	WWTP A		WWTP B		WWTP C		WWTP D	
	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent
DF	163.1	146.8	128.6	146.8	202.8	156.4	1027.1	115.4
IM	64.6	18.2	42.1	4.9	51.3	31.3	67.2	6.2
MA	10.4	4.5	5.8	1	4.8	3.1	56.7	12.7
KP	184	68.3	100.6	37.7	161.2	205.5	7881	1712.7

BF	191	35.2	106.6	16.1	212.5	81.5	433.7	63.7
CA	19.8	15.6	20.4	4.5	24.4	19.3	28.7	10
GF	13.7	3.8	4.7	0.3	13.7	6.9	220.3	3.6
MTP	662.4	838.9	626.1	48.6	1175	1372.8	794.4	321.2
PHO	0.97	0.97	8.7	1.9	11.8	17.2	6.6	2.4
ATP	6733.7	3.8	4496.1	3.5	6626.7	58.4	8983.9	4.9
CBZ	139.2	180.5	118.6	80.8	164.5	195.4	148.8	43.4
SP	245.7	376.7	343.9	18.6	547.2	426.4	279	71.5
CF	32638.9	93	14454.1	35.6	33039.7	1790.9	32193.8	45.2
DEET	844.5	321.2	983.1	186.5	774.9	250.2	985.1	409.7
SD	64.57	21.49	39.14	1.74	78.94	41.03	33.61	1.22
ST	3.4	0.46	5.3	0.46	3	0.46	15.9	1.4
SMR	3.4	0.79	4.7	0.8	3.6	0.79	2.7	0.79
SIX	2.9	0.76	2.3	0.76	1.8	0.76	13.4	0.8
SIM	1.7	0.66	1.6	0.66	1.3	0.66	0.7	0.66
SMP	0.69	0.69	1.2	0.69	0.69	0.69	2.1	0.69
SQX	1.7	0.87	1.1	0.87	1.2	0.87	0.9	0.87
SMT	9.7	6.3	4.3	0.16	6.8	0.16	9.7	1
SDM	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01
SMX	365	121	601	150	982	366	254	41
SMZ	3.6	1.35	2.8	1.35	3.1	1.35	3.9	1.35
SM	3.9	1.37	2	1.37	3.5	1.37	4	1.37
TP	11.2	18	192.9	4.3	423.2	427.8	252.8	10.8
EM	342.7	271.3	299.9	5.6	408.8	189	309.4	23.9
CAM	509.8	319.6	374.4	3.7	639.3	342.6	661.4	22.4
RXM	317	217.3	373.5	5	457.7	269	375.8	25.1
TS	2.04	2.04	2.04	2.04	2.04	2.04	2.04	2.04
CDM	7.8	71.2	76.7	1.5	87.7	353.6	131.3	3.3
LCM	173	197.5	264.3	1.1	814.2	507.9	332.6	3.3
CP	31.9	7.6	29.3	18.4	47.3	18.4	45.6	6
NA	143.8	199.7	135.1	4.7	212.9	131.7	166.2	6

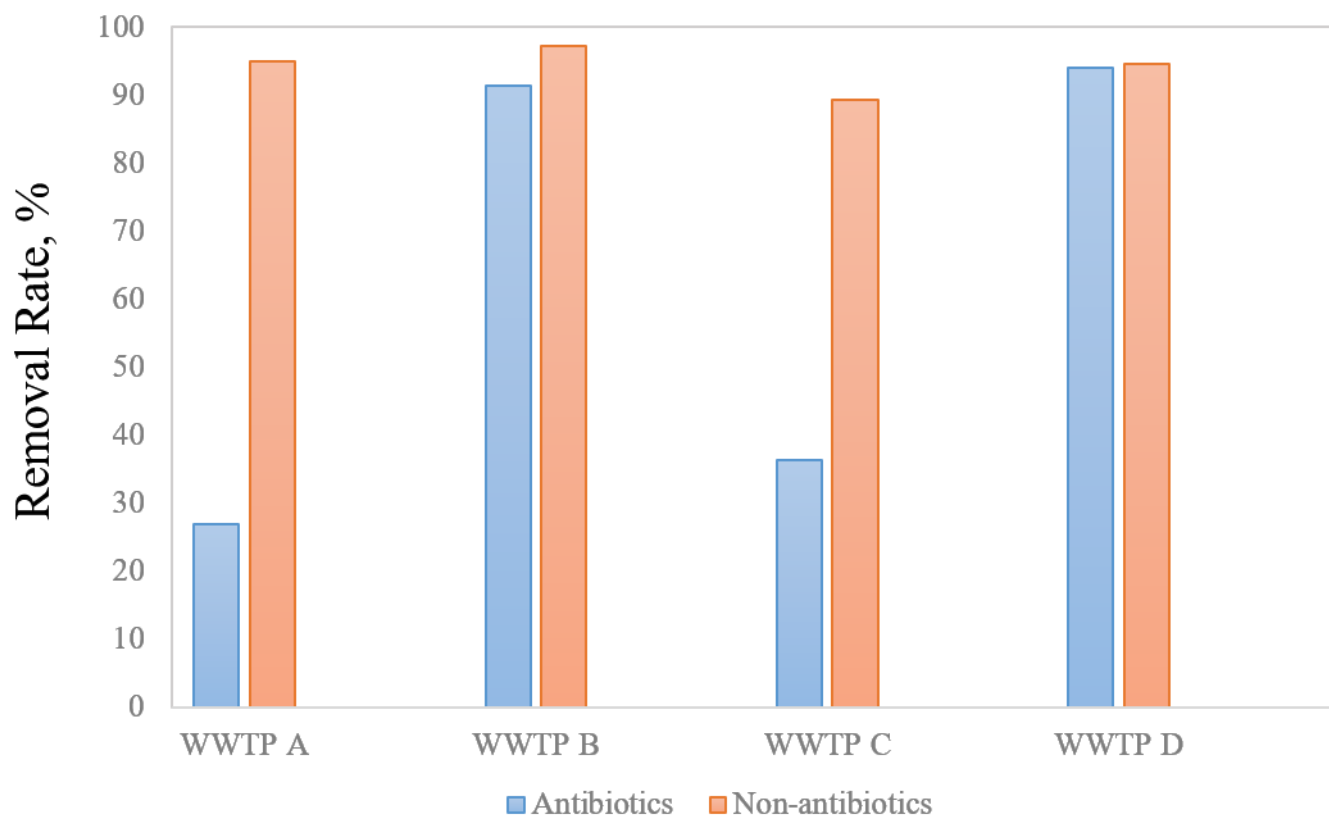


Fig. S6 The removal rates of antibiotics and non-antibiotics in the four urban WWTPs.

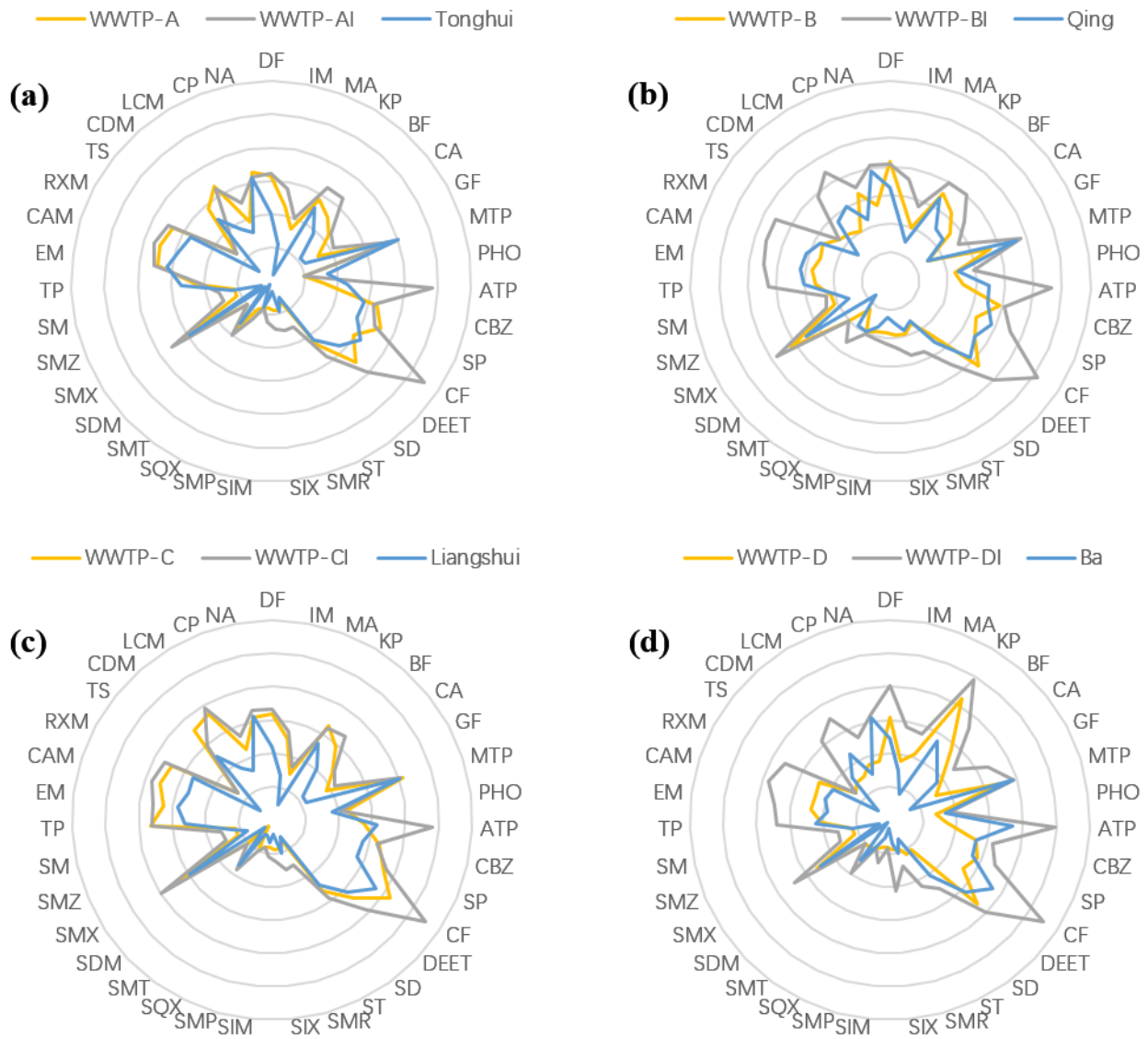


Fig. S7 The distribution profile in influents, effluents of four urban WWTPs and their receiving rivers. The yellow line means the effluent from WWTP; the gray line means the influent and the blue line means the river.

2.4 Principal component analysis (PCA) on source apportionment

Principal component analysis is useful for source apportionment. Dai et al. identified three sources of PhACs in Beiyun River: freshly discharging untreated domestic sewage, bioconversion source from wastewater treatment processes or unknown source and treated sewage or naturally attenuated untreated sewage (Dai et al., 2015). Lin et al. also did source analysis with PCA and identified four sources of PhACs in Xiangjiang River (Lin et al., 2018).

3. Reference

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