Distribution of antibiotics in wastewater-irrigated soils and their accumulation in vegetable crops in the Pearl River Delta, southern China

Min PAN[†], Chris KC WONG[‡], LM CHU[*†]

[†] *School of Life Sciences, The Chinese University of Hong Kong, Shatin, NT, Hong Kong SAR, China.*

[‡]*Department of Biology, Hong Kong Baptist University, Kowloon, Hong Kong SAR, China.*

Supplementary Material:

Table S1. Properties of soils from the study sites in the Pearl River Delta, southern China.

	Depth (cm)	pH	Organic C (%)	Sand (%)	Silt (%)	Clay (%)
Huizhou	0-10	7.44±1.04	0.48	23.8±2.7	44.8±6.8	31.4±1.7
	10-20	7.66±0.82	0.51	25.6±1.5	43.4±7.2	31.0±4.1
Foshan	0-10	7.76±1.46	0.52	37.7±3.4	32.0±3.1	30.3±2.6
	10-20	7.94±0.62	0.63	39.7±3.8	29.8±2.9	30.5±2.1
Zhongshan	0-10	7.53±0.21	0.81	24.5±1.7	50.8±8.2	24.7±4.2
	10-20	7.83±0.08	0.91	27.9±2.1	49.8±5.8	22.3±1.7
Guangzhou	0-10	6.98±0.61	0.63	35.0±3.8	39.7±8.1	25.3±1.9
	10-20	7.50±0.54	0.64	34.8±6.1	40.1±6.6	25.1±3.0
Dongguan	0-10	6.80±1.19	0.44	32.7±4.6	36.1±2.7	31.2±3.2
	10-20	7.29±0.99	0.52	39.2±3.2	37.3±5.6	23.5±2.6
Shenzhen	0-10	6.83±0.01	0.5	50.4±9.5	29.9±6.9	19.7±1.0
	10-20	7.35±0.25	0.53	48.9±7.7	29.5±7.2	21.6±2.3

[*]Corresponding author phone: 852-39436378; fax: 852-26037732; email: leemanchu@cuhk.edu.hk.

Table S2. Optimised MRM parameters for the analysis of antibiotics by HPLC-MS/MS.

Compound	Abbr.[a]	R.T.[b] (min)	MRM-transition[c]	Fragmentor (V)	CE (eV)[d]	Dwell
ESI+						
Tetracycline	TC	2.123	445.2→410.3	100	25	50
			445.2→154.3	100	40	50
Norfloxacin	NOR	1.591	320.1→302.1	115	20	50
			320.1→276.1	115	12	50
Sulfamethazine	SMZ	3.163	279.1→186.1	90	12	50
			279.1→124.1	90	24	50
Erythromycin	ERY	6.741	734.5→158	135	32	50
			734.5→83.1	135	60	50
ESI-						
Chloramphenicol	CAP	1.047	323→152.2	95	13	200
			323→151.4	95	16	200

a:abbreviation; b:retention times; c: compounds for ESI+ were $[M+H]^+$ and chloramphenicol were $[M-H]^-$; d: collision energy.

Table S3. Extraction recoveries of antibiotics in different matrixes at two spiked concentrations and the method limit of detection (LOD) and the method limit of quantification (LOQ).

Matrix	Compound	Mean ±SD (%)		LOD	LOQ
		10 ng/L	100 ng/L	(ng/L)	(ng/L)
Water	TC	104±1	116±7.0	0.26	0.87
	SMZ	97.2±0.6	94.3±1.3	0.12	0.40
	NOR	84.2±1.7	70.1±6.1	0.34	1.13
	ERY	95.8±1.4	80.0±3.0	0.18	0.60
	CAP	73.6±1.6	89.3±2.3	0.17	0.57
		10 ng/g	100 ng/g	(ng/g)	(ng/g)
Soil	TC	124±1	112±9	0.33	1.10
	SMZ	100±4	103±1	0.21	0.70
	NOR	88.2±7.1	78.6±3.4	1.39	4.63
	ERY	87.8±3.4	80.9±6.7	0.32	1.07
	CAP	84.6±7.6	87.1±7.5	0.85	2.83
		10 ng/g	100 ng/g	(ng/g)	(ng/g)
Crop	TC	115±2	114±3	1.32	4.40
	SMZ	98.8±3.3	98.3±1.3	0.24	0.80
	NOR	80.8±4.1	87.2±2.7	0.35	1.17
	ERY	73.6±2.9	79.1±4.8	0.43	1.43
	CAP	79.3±4.8	89.2±5.9	0.72	2.40

Table S4. Concentrations (μg/kg dw) of antibiotics in crop tissues (mean ± SD, n= 5).

	Vegetables	TC	SMZ	NOR	ERY	CAP
HZ	Chinese white cabbage (leaf)	5.5±0.6	<LOQ	9.6±1.2	<LOQ	2.6±0.7
	Chinese white cabbage (root)	<LOQ	0.8±1.3	5.9±1.2	<LOQ	<LOQ
	Water spinach (leaf)	<LOQ	<LOQ	8.6±0.6	<LOQ	<LOQ
	Water spinach (root)	<LOQ	0.7±0.8	4.6±0.7	<LOQ	<LOQ
	Rice (fruit)	5.6±2.5	<LOQ	10.5±0.8	<LOQ	6.4±1.1
	Rice (stem)	<LOQ	<LOQ	7.7±0.8	<LOQ	5.2±1.0
	Rice (root)	<LOQ	<LOQ	6.6±1.1	<LOQ	2.7±0.7
	Chinese radish (leaf)	<LOQ	<LOQ	5.7±0.7	<LOQ	18.9±2.7
	Chinese radish (root)	<LOQ	2.0±1.3	4.9±0.6	1.5±0.9	<LOQ
FS	Chinese white cabbage (leaf)	6.3±2.0	<LOQ	12.8±0.4	<LOQ	2.7±0.5
	Chinese white cabbage (root)	4.2±1.6	0.8±1.0	7.9±0.8	<LOQ	<LOQ
	Corn (fruit)	6.6±1.5	<LOQ	17.5±1.1	<LOQ	16.2±3.6
	Corn (stem)	4.4±0.9	<LOQ	14.0±0.3	<LOQ	13.6±2.3
	Corn (root)	<LOQ	<LOQ	12.6±1.6	<LOQ	13.1±3.6
	Rice (fruit)	8.0±1.5	<LOQ	10.4±1.4	<LOQ	17.2±5.0
	Rice (stem)	<LOQ	<LOQ	9.3±1.3	<LOQ	14.5±0.7
	Rice (root)	<LOQ	<LOQ	7.7±0.6	<LOQ	13.4±1.1
ZS	Chinese white cabbage (leaf)	4.9±1.6	<LOQ	16.5±0.6	<LOQ	2.7±0.5
	Chinese white cabbage (root)	<LOQ	0.8±0.9	10.6±1.6	<LOQ	5.8±1.7
	Rice (fruit)	8.5±1.1	<LOQ	13.8±1.0	<LOQ	20.4±4.8
	Rice (stem)	4.8±1.3	<LOQ	10.5±0.8	<LOQ	15.6±0.7
	Rice (root)	<LOQ	<LOQ	9.2±1.5	<LOQ	13.8±1.7
GZ	Chinese white cabbage (leaf)	4.0±1.4	<LOQ	13.8±0.6	<LOQ	4.1±2.8
	Chinese white cabbage (root)	<LOQ	0.5±1.3	9.7±1.1	<LOQ	<LOQ
	Corn (fruit)	<LOQ	<LOQ	10.2±0.6	<LOQ	15.1±1.5
	Corn (stem)	<LOQ	<LOQ	8.2±0.8	<LOQ	12.9±0.2
	Corn (root)	<LOQ	<LOQ	6.4±1.4	<LOQ	9.9±0.9
DG	Chinese white cabbage (leaf)	10.1±0.8	<LOQ	23.6±0.7	<LOQ	12.7±0.7
	Chinese white cabbage (root)	5.9±0.6	2.3±1.2	20.4±1.3	1.7±0.4	5.5±0.5
	Water spinach (leaf)	6.3±1.1	<LOQ	21.8±1.1	<LOQ	10.1±0.9

	Water spinach (root)	4.8±1.3	1.7±1.4	16.9±2.5	1.3±0.1	3.0±1.9
	Chinese radish (leaf)	9.2±1.0	1.9±1.0	21.3±2.1	<LOQ	8.0±1.0
	Chinese radish (root)	6.5±0.6	4.2±2.0	17.5±0.7	2.2±0.2	5.2±3.5
SZ	Chinese white cabbage (leaf)	5.3±1.0	<LOQ	15.7±0.6	<LOQ	7.1±1.4
	Chinese white cabbage (root)	<LOQ	1.2±0.8	10.7±1.9	<LOQ	3.4±1.0
	Corn (fruit)	<LOQ	<LOQ	16.3±0.9	<LOQ	22.4±2.4
	Corn (stem)	<LOQ	<LOQ	14.9±0.1	<LOQ	14.6±1.3
	Corn (root)	<LOQ	<LOQ	13.4±0.8	<LOQ	8.4±2.4
	Water spinach (leaf)	<LOQ	<LOQ	11.7±1.3	<LOQ	5.5±0.9
	Water spinach (root)	<LOQ	<LOQ	10.3±0.5	<LOQ	<LOQ