water

Supplementary material

Contents

Text Sections

Figures

Tables

Section S1 SPE process and UPLC-MS/MS analysis of target PPCPs.

Samples were collected using a pre-cleaned 250 mL amber glass bottles, filtered through glass microfiber filters (0.7 μm, Whatman UK), adjusted to pH 3.0 with 40% H_2SO_4 (v/v), added with 1 mL 25g/L ascorbic acid, 0.1 g Na_2EDTA, spiked with internal standards (100 ng OLF-D_3, 100 ng CAF-$^{13}C_3$, 100 ng SMN-$^{13}C_6$). HLB cartridge (500 mg/6 mL CNW) were preconditioned by 5 mL MeOH, 5 mL 0.5 mol/L HCl, 5 mL Milli-Q water. Then the sample was extracted at a flow rate of approximately 3 mL/min. Afterwards the cartridge was rinsed by 5 mL 5% (v/v) methanol aqueous, 5 mL Milli-Q water, dried under vacuum for 10 min, eluted by 10 mL MeOH. The eluate was collected with a 10 mL glass tube and dried under a gentle stream of N_2 in a 40 °C water bath, then reconstructed to 1 mL with 40% methanol aqueous. The resulting extract was filtered through 0.2 μm PES filters for UPLC-MS/MS analysis.

The chromatographic separation of target PPCPs was performed on an Waters I-class ultra-performance liquid chromatography (UPLC) equipped with an Waters BEH-C18 column (100 mm × 2.1 mm, 1.7 μm). The column was maintained at 30 °C and the injection volume was 10 μL. Ultrapure water containing 0.1% formic acid (v/v) (A) and MeOH containing 0.1% formic acid (v/v) (B) were used as the mobile phases at a total flow rate of 0.25 mL min^{-1}. The elution program was set as 10 % mobile phase A and 90 % mobile phase B in 8 minutes. A Sciex QTrap mass spectrometer (MS), equipped with an electrospray ionization (ESI) source and operated in the positive ion mode, was

employed to analyze the target PPCPs. The MS system was operated under the following conditions: Ionspray Voltage (IS) : 5500; Ion Source Gas1 (GS1) : 50; Collision Gas (CAD) : Medium; Ion Source Gas2 (GS2) : 50; Curtain Gas (CUR) : 20; Temperature (TEM) : 500. For each compound, the fragmentor voltage and product ions are listed in Table S2.

Section S2 DNA extraction and PCR amplification.

E.Z.N.A.® Soil DNA kit (Omega Bio-Tek, Inc., Norcross, GA, U.S.) were used to extract DNA of the samples and the quality of DNA was assessed using a 1.0 % (w/v) agarose gel electrophoresis. To amplify the 16S rRNA from the samples the following primers were used:338F (5'-ACTCCTACGGGAGGCAGCA-3') and 806R (5'-GGACTACHVGGGTWTCTAAT-3').

Each PCR reaction was conducted in a 20 μL reaction volume containing 4 μL of 5 × FastPfu Buffer, 2 μL of 2.5 mM dNTPs, 0.8 μL of each primer (5 μM), 10 ng of template DNA and 0.4 μL of FastPfu Polymerase (TransGen AP221-02, Beijing, China). Reactions were carried out on a ABI GeneAmp® System 9700 (Perkin-Elmer Applied Biosystems, Foster City, CA, U.S.) under the following thermocycling steps: 95 °C for 2 min, followed by 25 cycles at 95°C for 30 s, 55°C for 30 s and 72 °C for 30 s, and a final extension at 72°C for 5 min and 10°C until halted by user. PCR products of three replicates were combined and purified using AxyPrep DNA gel extraction kit and Tris_HCl for each sample. Amplicons from the two samples were mixed at the equal concentrations.

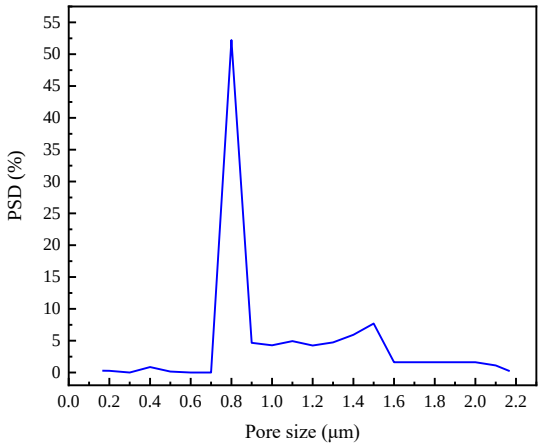


Figure S1. Average pore size of ceramic membrane.

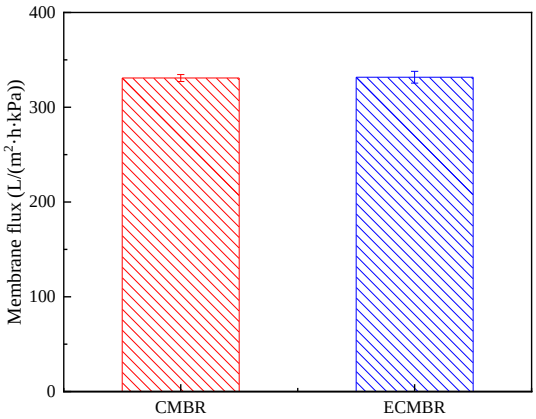


Figure S2. Average pure water permeability of the conductive membrane.

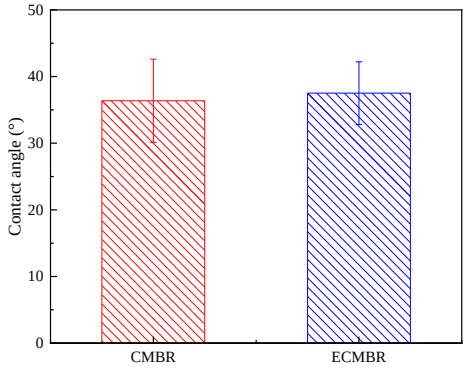


Figure S3. Water contact angle of ceramic membrane.

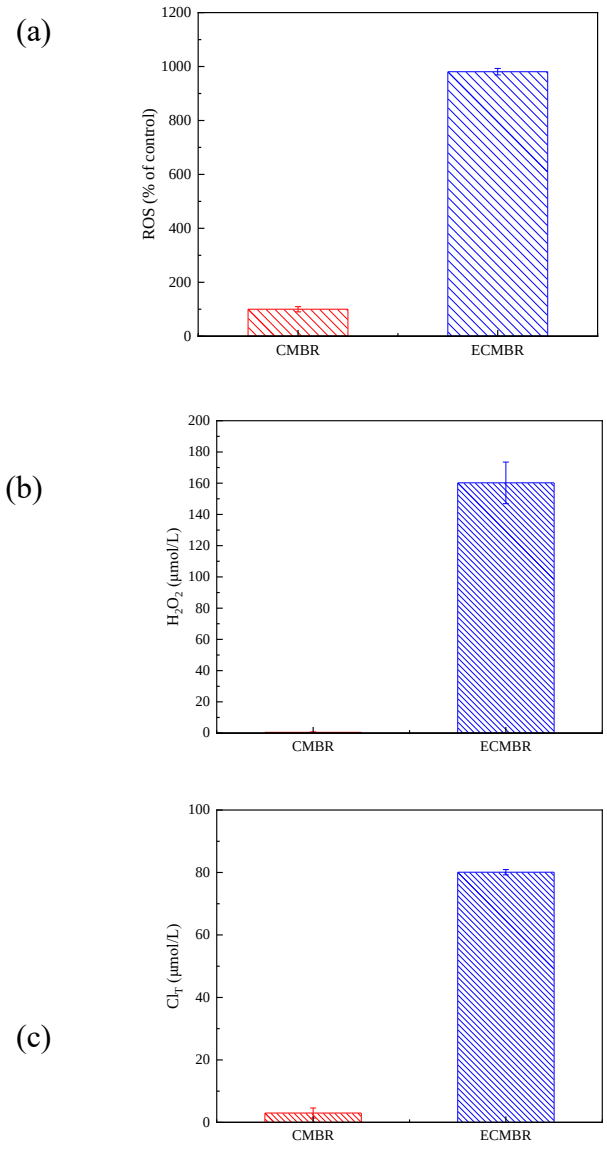
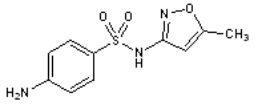
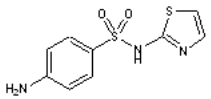
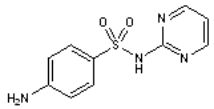
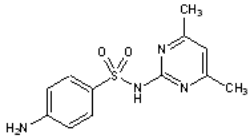
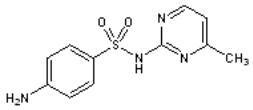
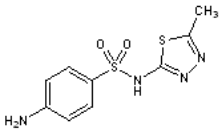
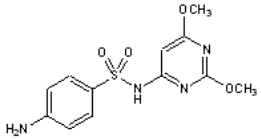
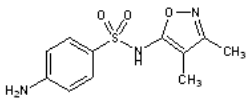
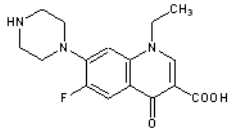
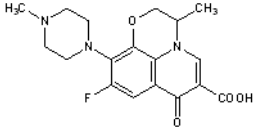
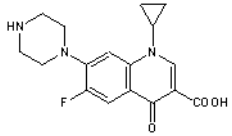
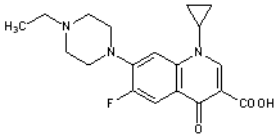
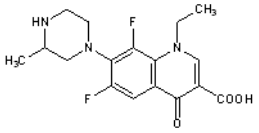
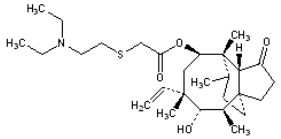
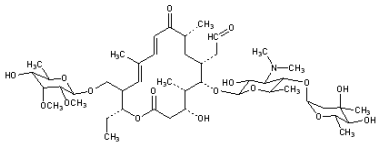
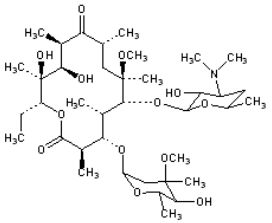
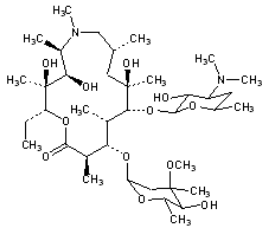
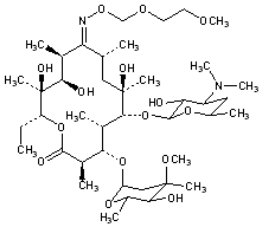
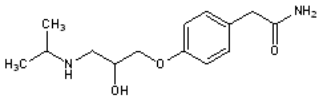
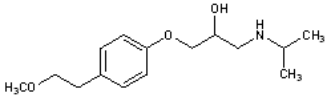


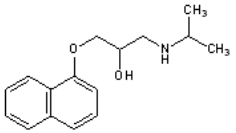
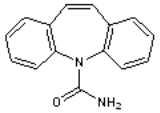
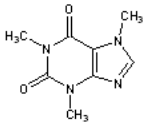
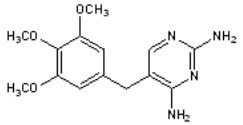
Figure S4. (a) ROS production in the batch tests (n = 3); (b) Concentrations of H_2O_2 in CMBR and ECMBR in the batch tests (n = 3); (c) Concentrations of Cl_T in CMBR and ECMBR in the batch tests (n = 3).

Table S1. physical-chemical properties and molecular structures of target PPCPs.

Category	Compound (name/acronym/formula)	Chemical Structure	CAS No.	MW	pK_a	logK_{ow}
Sulfonamides (SAs)	Sulfamethoxazole (SMX) $C_{10}H_{11}N_3O_3S$		723-46-6	253.28	1.85 ± 0.30 5.60 ± 0.04	0.89
	Sulfathiazole (STZ) $C_9H_9N_3O_2S_2$		72-14-0	255.32	2.01 ± 0.30 7.11 ± 0.09	0.05
	Sulfadiazine (SDZ) $C_{10}H_{10}N_4O_2S$		68-35-9	250.30	1.57 ± 0.10 6.50 ± 0.30	–0.12
	Sulfamethazine (SMN) $C_{12}H_{14}N_4O_2S$		57-68-1	278.33	2.07 ± 0.30 7.49 ± 0.13	0.80
	Sulfamerazine (SMR) $C_{11}H_{12}N_4O_2S$		127-79-7	264.30	2.82 ± 0.31 6.84 ± 0.30	0.34
	Sulfamethizole (SML) $C_9H_{10}N_4O_2S_2$		144-82-1	270.33	1.86 ± 0.30 5.29 ± 0.04	0.51
	Sulfadimethoxine (SDM) $C_{12}H_{14}N_4O_4S$		122-11-2	310.33	2.13 ± 0.30 6.08 ± 0.09	1.48

	Sulfisoxazole (SFX) $C_{11}H_{13}N_3O_3S$		127-69-5	267.30	1.52 ± 0.10 4.83 ± 0.50	1.01
Fluoroquinolones (FQs)	Norfloxacin (NOR) $C_{16}H_{18}FN_3O_3$		70458-96-7	319.33	3.59 ± 0.70 8.38 ± 0.25	1.48
	Ofloxacin (OFL) $C_{18}H_{20}FN_3O_4$		82419-36-1	362.15	2.27 ± 0.40 6.41 ± 0.30	1.41
	Ciprofloxacin (CIP) $C_{17}H_{18}FN_3O_3$		85721-33-1	331.34	2.68 ± 0.20 8.38 ± 0.25	1.31
	Enrofloxacin (ENR) $C_{19}H_{22}FN_3O_3$		93106-60-6	359.4	3.85 ± 0.30 6.19 ± 0.18	N/A [a]
	Lomefloxacin (LOM) $C_{17}H_{19}F_2N_3O_3$		98079-51-7	351.35	5.64 8.47	–0.39
Macrolides (MLs)	Tiamulin (TIA) $C_{28}H_{47}NO_4S$		55297-95-5	493.82	7.60	6.43

	Tylosin (TYL) $C_{46}H_{77}NO_{17}$		1401-69-0	916.1	3.31 ± 0.30 7.50 ± 0.13	N/A
	Clarithromycin (CLA) $C_{38}H_{69}NO_{13}$		81103-11-9	747.95	7.25	3.16
	Azithromycin (AZN) $C_{38}H_{72}N_2O_{12}$		83905-01-5	748.98	7.34	4.02
	Roxithromycin (ROX) $C_{41}H_{76}N_2O_{15}$		80214-83-1	837.53	9.17 ± 0.30	3.73
β-blockers	Atenolol (ATE) $C_{14}H_{22}N_2O_3$		29122-68-7	266.34	9.17 ± 0.38	0.10
	Metoprolol (MET) $C_{15}H_{25}NO_3$		37350-58-6	267.36	9.18 ± 0.38	1.79

	Propranolol (PROP) $C_{16}H_{21}NO_2$		525-66-6	259.35	9.45 ± 0.03	3.10
Antiepileptic	Carbamazepine (CBZ) $C_{15}H_{12}N_2O$		298-46-4	236.27	13.94 ± 0.20	2.67
Stimulant	Caffeine (CAF) $C_8H_{10}N_4O_2$		58-08-2	194.19	10.40	–0.13
Dihydrofolate reductase inhibitor	Trimethoprim (TMP) $C_{14}H_{18}N_4O_3$		738-70-5	290.32	7.20 ± 0.17	0.79

a: not available

Table S2. Operational parameters of tandem MS and LOQs of target PPCPs in wastewater.

Compound	Q1 (amu)	Q3 (amu)	DP (volts)	CE (volts)	Recovery (%)	LOQ [a] (μg/L)	LOD [b] (μg/L)
SMR	264.8	155.9	50	23	121	0.18	0.05
SMX	253.9	108.0	50	31	104	0.33	0.10
SDZ	250.9	108.0	40	31	105	0.36	0.11
SMN	279.0	186.0	60	23	130	0.31	0.09
SML	270.9	156.1	20	19	123	0.50	0.15
SFX	268.0	113.1	40	20	91	0.47	0.14
SDM	310.9	156.0	20	19	83	0.34	0.10
STZ	255.8	156.0	60	20	116	0.47	0.14
LOM	352.2	265.2	117	21	127	0.35	0.11
OLF	362.1	260.9	90	38	102	0.49	0.15
ENR	360.2	316.0	30	27	72	0.30	0.09
CIP	332.1	288.1	40	23	100	0.29	0.09
NOR	320.0	233.0	30	33	101	0.47	0.14
CLA	748.5	158.0	80	34	90	0.62	0.19
ROX	837.4	679.4	20	31	116	0.66	0.20
AZN	749.3	158.0	40	46	134	0.26	0.08
TYL	916.6	772.3	90	39	117	0.41	0.12
TIA	494.2	285.4	90	30	94	0.68	0.20
CBZ	237.1	194.0	60	26	126	0.25	0.07
CAF	195.0	138.0	70	28	79	0.38	0.11
PROP	260.3	116.0	40	23	85	0.40	0.12
MET	268.1	116.2	60	25	77	0.48	0.14
ATE	260.0	116.3	52	25	81	0.36	0.11
TMP	291.0	261.0	110	31	96	0.47	0.14

a: limits of quantification

b: limits of detection

Table S3. Concentrations of target PPCPs in Influent of EMBR and CMBR.

Compound	Influent (ng/L)			
	Min	Med	Max	Freq
SAs				
SMR	29.6	54.9	219.2	7
SMX	165.0	277.4	1003.2	7
SDZ	149.9	230.9	709.6	7
SMN	34.4	48.4	120.5	7
SML	59.4	162.7	428.5	7
SFX	24.6	35.8	182.9	7
SDM	26.2	48.7	124.2	7
STZ	116.4	223.1	977.9	7
FQs				
LOM	102.7	151.0	748.3	7
OLF	2920.5	3540.2	4294.8	7
ENR	74.8	97.7	218.6	7
CIP	340.4	400.2	811.6	7
NOR	436.6	590.2	817.7	7
MLs				
CLA	73.8	668.9	2874.2	7
ROX	25.1	200.6	887.3	7
AZN	12.2	209.8	490.5	7
TYL	ND	49.4	99.8	6
TIA	224.4	262.2	472.7	7
β-blockers				
PROP	ND	74.6	86.2	5
MET	2199.7	3609.7	4065.6	7
ATE	ND	57.4	71.8	5
Antiepileptic				
CBZ	119.7	145.8	473.3	7
Stimulant				
CAF	12127.6	14633.2	19273.7	7
dihydrofolate reductase inhibitor				
TMP	194.9	253.9	552.8	7

*ND: Not detected.

Table S4. Richness and diversity estimators of the bacteria phylotypes in the two MBR systems.

Sample	OTU	Chao1	Shannon	Coverage
Inoculum	913	1058	5.15	0.993829
EMBR	964	1155	5.08	0.993136
CMBR	853	1083	4.59	0.992674