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Occurrence and fate of pharmaceuticals and personal care products in Taiwan's aquatic environment

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ABSTRACT

Pharmaceuticals and personal care products (PPCPs) have raised considerable concern around the world due to their potential toxicity for ecological system and human health. PPCPs are ineffectively removed by conventional wastewater treatment processes and thus occur widely in aqueous environments. This study investigated the occurrence and removal of 28 often-used PPCPs (including selected antibiotics, estrogens, non-steroidal anti-inflammatory drugs, beta-blockers, and lipid regulators) in the primary, secondary and tertiary (ultrafiltration/reverse osmosis (UF/RO)) treatment processes of the Water Resource Recycling Center (WRRC) and surface waters in Taiwan. We have demonstrated 20 target PPCPs in WRRC influents; sulfamethoxazole (1353 ng/L), caffeine (6823 ng/L) and acetaminophen (2716 ng/L) were found at high concentrations. Secondary and chlorination processes showed inefficient removal for PPCPs (12 PPCPs had <80% removal). However, most target compounds were removed effectively (with ~90% removal) in the tertiary process (UF/RO) except for oxytetracycline and caffeine, and the overall removal efficiency by WRRC was >99%. More than 10 compounds were detected in the surveyed surface waters (from reservoirs, river waters and dams). Caffeine had the highest observed concentration (1,813 ng/L) while others were present at <260 ng/L. Sulfamethoxazole, sulfamethazine, sulfadimethoxine and caffeine were detected most frequently (>70%). Some of the PPCPs found originated from discharges from conventional wastewater treatment plants. In conclusion, RO demonstrated good overall performance and could be used to process wastewater to better ensure the health of humans and wildlife.

Keywords: Pharmaceuticals and personal care products; Reverse osmosis; Surface waters; Water reuse

1. Introduction

Pharmaceuticals and personal care products (PPCPs) are an important and emerging group of contaminants that have increasingly raised concerns due to their health effects on organisms and potential risk to ecosystems, even at low concentrations (ng/L-μg/L). In compliance with regulations set by Taiwan's Environmental Protection Administration (EPA), waste streams from wastewater treatment plants (WWTPs) are treated only to meet minimal effluent standards (BOD 30 mg/L, COD 100 mg/L, SS 30 mg/L, true color 550 and E. coli 200,000 CFU/100 mL). PPCPs are therefore not efficiently removed during treatment processes in conventional WWTPs and have been subsequently released into aqueous environments [1–3].

Many investigators have reported the presence of PPCPs at significant concentrations in aquatic environ-

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ments around the world [4–7]. In South Korea, Kim et al. [5] found antibiotics (1.7–36 ng/L), estrogens (up to 5.0 ng/L), non-steroidal anti-inflammatory drugs (NSAIDs) including acetaminophen, naproxen, ibuprofen and diclofenac (1.1–73 ng/L), and gemfibrozil (1.8–9.1 ng/L) in three rivers that received WWTP effluents. Managaki et al. [4] identified veterinary antibiotics (sulfonamides, macrolides, and trimethoprim) at concentrations of 4–448 ng/L in a Japanese river and 7–360 ng/L in Vietnamese rivers and canals.

Waste effluents from hospitals, WWTPs, and sewage treatment plants are major sources of environmental PPCP contamination. Several PPCPs have been found persisting in the effluents from conventional wastewater treatment processes [2,7–9]. Heberer [10] detected caffeine, naproxen, and ketoprofen in wastewater effluents at concentrations of 180, 80 and 230 ng/L; these were removed at more than 99%, 82% and 23% efficiencies, respectively. Brown et al. [6] found concentrations of sulfamethoxazole of 390 ng/L in WWTP influents and 310 ng/L in effluents. Effluent concentrations of macrolides have been reported to be as high as 328 ng/L and to persist through winter due to their lower biological activity and higher input [12]. Sulfonamides and macrolides were not easily adsorbed onto activated sludge for purposes of degradation [13–15]. Several studies have suggested that tertiary treatments are needed in WWTPs in order to effectively eliminate PPCPs [5,11,16–18]. Filtration processes such as reverse osmosis (RO) and nanofiltration (NF) can remove PPCPs, which tend to be neutral, with great efficiency [3].

Comprehensive studies of the occurrence and fate of PPCPs in Taiwan's aqueous environments are still very limited. The aim of this study is to investigate the occurrence and fate of 28 often-used PPCPs, including 17 antibiotics, a psychostimulant, a lipid regulator, four non-steroidal anti-inflammatory drugs (NSAIDs), four estrogens, and one beta-blocker (Table 1), in the primary, secondary and tertiary (ultrafiltration/reverse osmosis, UF/RO) treatment processes of Taiwan's Water Resource Recycling Center (WRRC) as well as to survey four surface waters in the northern, central and southern parts of Taiwan [19–26].

Table 1
LC-ESI-tandem MS conditions by MRM in positive and negative mode

Ionization mode	ESI+	ESI–
Dwell time, ms	10	50
Ion spray voltage (IS), kV	5.5	–4.5
Curtain gas (CUR), L/h	10	10
Gas1 (GS1), L/h	60	50
Gas2 (GS2), L/h	50	60
Temperature (TEM), °C	500	500
Interface heater (ihe)	ON	ON
Collisionally activated dissociation (CAD)	5	5

2. Methods

2.1. Site description and wastewater treatment process

Fig, 1a is a map of the sampling locations in Taiwan, including the WRRC and four surface waters A–D (Location A is in Longtan Township, Taoyuan County, Taiwan; Location B is in Shihgang Township, Taichung County, Taiwan; Location C is in Jiji Township, Nantou County, Taiwan; and Location D is in Dashu Township, Kaohsiung County, Taiwan.) Samples were taken on different dates (Location A: 6/4, 7/23, and 8/20; Location B: 7/22, 8/20, and 8/27; Location C: 6/4; and Location D: 5/30, 7/22, and 8/21; all dates are from 2009.) All analyses were done in triplicate, and averaged values are reported in Fig. 2. Fig. 1b shows the wastewater treatment processes in WRRC, the location of sampling sites, and the sampling date. The WRRC mainly includes primary (primary clarifier (PC)), secondary (aeration tank (AT) + secondary clarifier (SC)) and chlorination processes, and tertiary (UF/RO) treatment processes. Water samples were collected from the influent (S1), the effluent of the secondary and chlorination processes (S2), and the effluent of the UF/RO process (S3) according to the hydraulic retention time (HRT) preferred by the WRRC. These surveyed surface waters in northern, central and southern parts of Taiwan constitute the sources for drinking-water treatment processing plants and are distributed to large civilian populations.

2.2. Sample collection and preparation

Grab samples (1 L) were collected from each sampling site in amber-glass bottles (rinsed with methanol and deionized water) and stored in ice-packed coolers. Water samples were all collected between May and August, 2009, and analysis was completed within two weeks of sample collection. Eight mL of 0.125 M EDTA-2Na were added to amber-glass bottles before collecting samples meant for analysis of tetracycline antibiotics. Tetracyclines (tetracycline, oxytetracycline, and chlortetracycline) can be sorbed by residual metals in the sample matrix, causing irreversible binding and lower recovery. Therefore, EDTA-2Na was employed to chelate metals sufficiently and prevent interference with extraction of the tetracycline antibiotics [19]. All samples were vacuum-filtered through 0.45-μm and 0.22-μm cellulose acetate membrane filters and adjusted to pH 4 with sulfuric acid (2 N). Samples were then stored at 4°C until analysis. Oasis HLB (500 mg, 6 mL, Waters, Milford, MA, USA) and Oasis MCX (150 mg, 6 mL, Waters, Milford, MA, USA) cartridges were employed for solid phase extraction (SPE). All target compounds were extracted using SPE (HLB cartridges for all antibiotics and NSAIDs, as well as

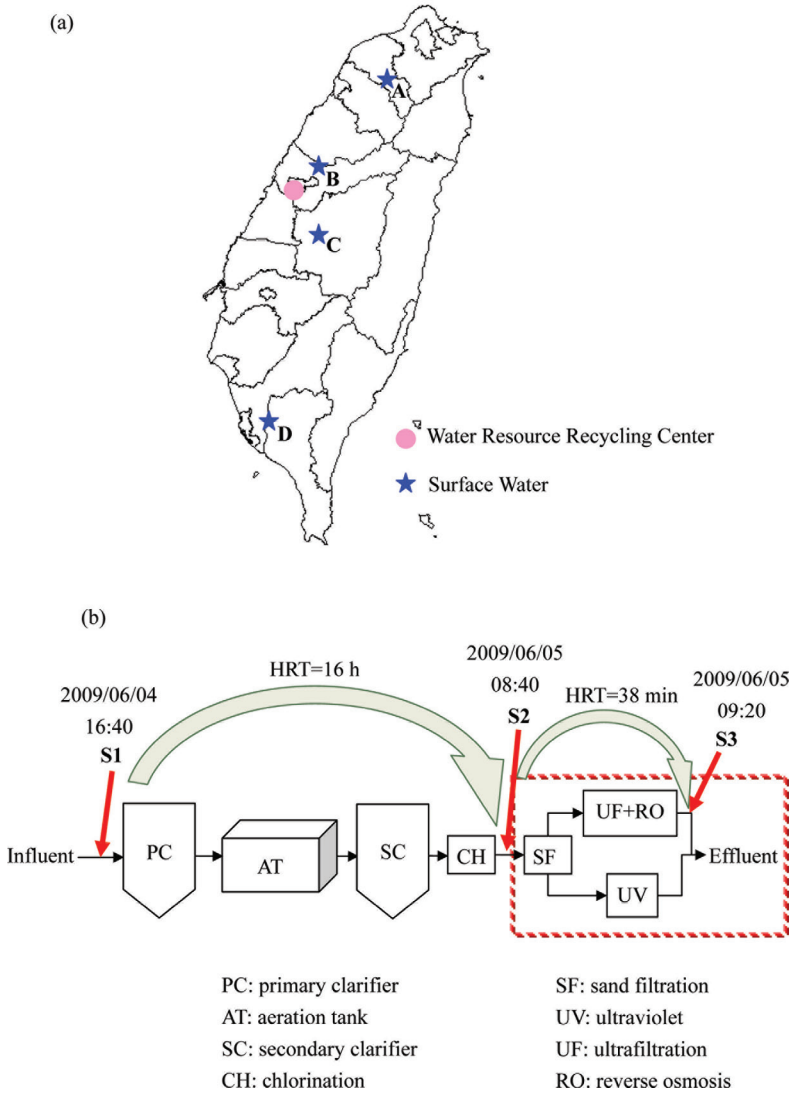

Fig. 1. (a) Sampling locations; and (b) Wastewater treatment processes in the WRRC, the location of sampling sites, and date of sampling.

the psychostimulant and lipid regulator; MCX cartridges were used for the estrogens and beta-blocker). SPE cartridges were preconditioned with 6 mL each of methanol and DI water. Four-hundred and 120 mL aliquots of the water samples for the HLB and MCX cartridges respectively were spiked with $^{13}C_6$-sulfamethazine (Cambridge Isotope Laboratories) and D5-chloramphenicol (Fluka) as surrogates and loaded into the cartridges. After sample passage, cartridges were rinsed with 6 mL DI water to remove excess EDTA–2Na. The analytes were eluted with 4 mL of methanol and 4 mL of methanol–diethyl-ether (50:50, v/v). The eluates were collected, evaporated to dryness with a flow of nitrogen gas, reconstituted to 0.4 mL with 25% aqueous methanol, and finally filtered through a 0.45-μm PVDF membrane filter before liquid chromatography/tandem mass spectrometry (HPLC–MS/MS) analysis. All analyses were performed in triplicate.

2.3. LC-ESI-MS/MS analysis

Chromatographic separation was performed using an Agilent 1200 HPLC (Agilent, Palo Alto, CA, USA) equipped with a ZORBAX Eclipse XDB-C18 column (150 × 4.6 mm, 5 μm). Mobile phase A contained 0.1% formic acid (v/v) in water. Mobile phase B contained 0.1% formic acid (v/v) in methanol. The gradient started with 0% of mobile phase B for 0.5 min, increased to 40% from 0.5–3 min, to 70% from 3.0–7.5 min, to 95% from 7.5–9.0 min, remained at 95% until 11 min, decreased to 0% from 11–12 min, and finished at 0%. All target com-

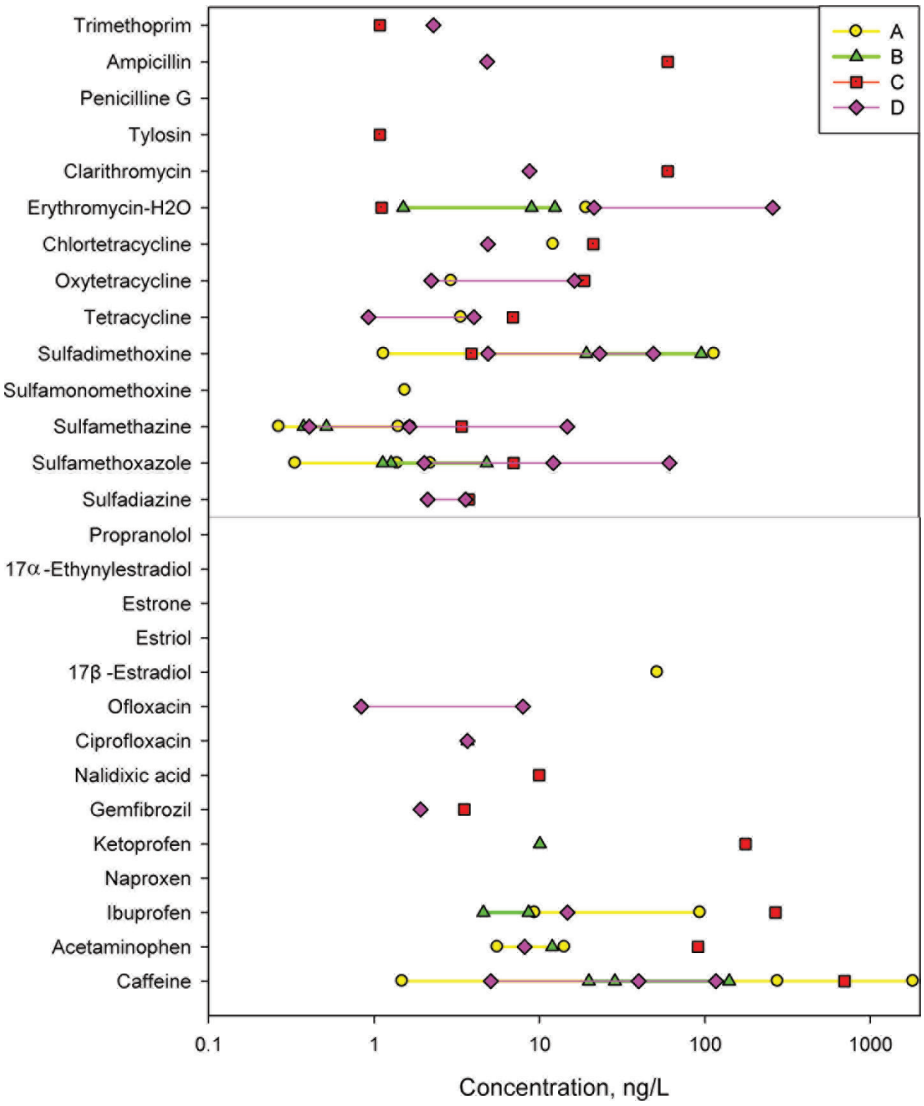

Fig. 2. Occurrence of 28 target compounds in surface waters.

pounds were eluted out of the column within 15 min. The sample injection volume was 50 μL. The autosampler was operated at room temperature. Mass spectrometric measurements were carried out on a Sciex API 4000 (Applied Biosystems, Foster City, CA, USA) equipped with an electrospray ionization (ESI) interface in negative mode for 4 compounds (gemfibrozil and all NSAIDs except for acetaminophen) and positive mode for all others. Table 1 shows the LC-ESI-tandem MS conditions by MRM in positive and negative ion mode. Data acquisition was performed in multiple reaction monitoring (MRM) mode with a dwell time of 10 ms for positive mode and 50 ms for negative mode and unit mass resolution on both mass analyzers. Two MRM pairs were used to identify the target compounds (except for ibuprofen), and pair 2 was used for quantification (Table 2).

3. Results and discussion

3.1. Quantification and method validation

Table 2 shows 28 target compounds, their MRM pairs, recoveries in DI and real water, linearity, and method detection limits (MDLs). Quantification of PPCPs was performed by means of HPLC–MS/MS with MRM, using the two highest-intensity typical precursor-ion/product-ion transition pairs. Recovery experiments were performed with an aliquot of 50 ng/L target analytes spiked into both DI and real water to determine recoveries of the spiked target compounds by comparing recovery before and after SPE extraction. The recoveries of target compounds were in the acceptable ranges for DI water (62.9–133.7%) and real water (61.0–117.3%). Recoveries for both DI and real waters were similar, indicating an insignificant ma-

Table 2
The 28 target compounds, their MRM pairs, recoveries in DI and real water, linearities and method detection limits (MDL)

Compound	Retention time (min)	Pair 1	Pair 2	DI water			Real water			Correlation		
				Recovery ± SD (n = 3) (%)		R.S.D (%)	Recovery ± SD (n = 3) (%)		R.S.D (%)	Coefficient (r)	MDLs (ng/L)	Linear range (ng/L)
Sulfadiazine	5.62	251/92	251/156	93.1 ±	7.6	8.2	90.8 ±	4.6	5.1	0.9959	0.1	0.1~2500
Sulfamethoxazole	7.03	254/92	254/156	115.0 ±	6.2	5.4	103.0 ±	7.0	6.8	0.9965	0.1	0.1~2500
Sulfamethazine	6.40	279/156	279/186	102.2 ±	8.7	5.6	90.5 ±	13.0	14.4	0.9982	0.1	0.1~2500
Sulfamonomethoxine	7.26	281/92	281/156	111.7 ±	4.9	4.4	96.1 ±	8.1	5.3	0.9992	0.5	0.5~2500
Sulfadimethoxine	8.26	311/92	311/156	94.7 ±	12.6	13.3	87.1 ±	12.3	14.1	0.9966	0.1	0.1~2500
Tetracycline	7.75	445/154	445/410	79.3 ±	17.5	22.1	71.9 ±	3.5	4.9	0.9977	1	1.0~2500
Oxytetracycline	5.81	461/337	461/426	66.9 ±	1.8	2.6	70.9 ±	2.0	2.9	0.9958	1	1.0~2500
Chlortetracycline	6.58	479/154	479/444	62.9 ±	10.5	16.9	73.3 ±	5.3	7.2	0.9952	2.5	2.5~2500
Erythromycin-H2O	8.51	716.5/158	716.5/558	85.6 ±	5.0	5.8	96.8 ±	14.3	14.7	0.9954	0.1	0.1~2500
Clarithromycin	9.01	748.5/158	748.5/590.5	78.2 ±	1.5	1.9	97.0 ±	9.6	9.9	0.9990	0.5	0.5~2500
Tylosin	8.13	916.7/174	916.7/772.5	74.7 ±	6.5	8.6	96.0 ±	7.0	7.3	0.9986	1	1.0~2500
Penicillin G	10.5	335/160	335/176	113.7 ±	2.3	2.0	98.7 ±	6.7	6.7	0.9992	0.5	0.5~2500
Ampicillin	8.38	350/106	350/160	103.8 ±	3.9	3.8	117.3 ±	16.2	13.8	0.9982	1	1.0~2500
Nalidixic acid	9.77	233/187	233/215	97.2 ±	6.0	6.2	109.0 ±	5.3	4.9	0.9912	5	5~2500
Ciprofloxacin	5.63	332/288	332/314	105.7 ±	14.2	13.4	69.3 ±	5.7	8.2	0.9971	0.1	0.1~2500
Ofloxacin	5.46	362/261	362/318	97.8 ±	4.5	4.6	80.8 ±	7.2	8.9	0.9993	1	1~2500
Trimethoprim	5.25	291/230	291/261	77.8 ±	8.5	10.9	99.2 ±	2.4	2.5	0.9971	0.1	0.1~2500
Caffeine	6.62	195/110	195/138	92.9 ±	16.7	18.0	112.3 ±	10.6	9.4	0.9962	0.1	0.1~2500
Acetaminophen	5.79	152/93	152/110	110.8 ±	11.3	10.2	93.3 ±	2.1	2.2	0.9955	0.1	0.1~2500
Ibuprofen	11.9	205/161	—	118.3 ±	6.4	5.4	107.7 ±	12.4	11.5	0.9953	5	5~1000
Naproxen	11.2	229/170	229/185	98.6 ±	9.7	9.8	107.2 ±	11.5	10.7	0.9971	5	5~1000
Ketoprofen	11.0	253/197	253/209	89.8 ±	9.7	10.8	87.8 ±	8.7	9.9	0.9951	5	5~1000
Gemfibrozil	12.3	249/121	249/127	133.7 ±	24.6	18.4	89.2 ±	7.1	8.0	0.9924	1	1.0~500
17β-estradiol	11.3	255/133	255/159	115.0 ±	12.1	10.5	110.7 ±	10.1	9.1	0.9980	50	50~2500
Estriol	9.55	271/227	271/253	89.8 ±	14.9	16.6	87.9 ±	2.4	2.7	0.9994	100	100~2500
Estrone	11.3	271/133	271/253	106.0 ±	8.6	8.1	61.0 ±	8.9	14.6	0.9993	100	100~2500
17α-ethynylestradiol	11.2	279/133	279/159	110.0 ±	6.1	5.5	115.0 ±	1.7	1.5	0.9950	50	50~2500
Propranolol	7.15	260/116	260/183	100.6 ±	18.6	18.5	89.1 ±	4.5	5.1	0.9955	5	5~2500

trix effect on our analysis of target compounds in a real water matrix. The linearity of the calibration curve was examined by spiking analytes into real water and fitted to a linear mode, least-squares linear regression analysis (y=a+bx) in the studied concentration range. The MDLs were determined from the minimum detectable concentration of analyses in the linear range with a signal-noise ratio of at least 3 in a real water matrix. The MDLs for all PPCPs were from 0.1 ng/L to 100 ng/L, and the correlation coefficients (linearity) were all more than 0.9912, with a linear range from 0.1 ng/L to 2500 ng/L.

3.2. Occurrence and fate of PPCPs in a wastewater treatment plant

Table 3 shows the occurrence and fate of 28 target PPCPs in the WRRC. 20 target PPCPs were detected in WWRC influents; sulfamethoxazole (1353 ng/L), caffeine (6823 ng/L), and acetaminophen (2716 ng/L) were found at the highest concentrations, while tetracycline, erythromycin-H_2O, ampicillin, nalidixic acid, ofloxacin, ibuprofen, naproxen, ketoprofen, and gemfibrozil were present at concentrations of several hundred ng/L. In our previous investigations of the occurrence of 97 PPCPs in

Table 3
Occurrence and fate of the 28 target PPCPs in the WRRC

Compounds	S1	S2		S3		
	ng/L	ng/L	RE-1	ng/L	RE-2	TRE
Sulfadiazine	15.9 ± 1.2	10.8 ± 1.0	32	ND	>99	>99
Sulfamethoxazole	1353.3 ± 57.9	328.7 ± 11.4	76	4.3 ± 2.8	99	>99
Sulfamethazine	10.4 ± 1.2	2.7 ± 0.0	74	ND	>99	>99
Sulfamonomethoxine	ND	ND	—	ND	—	—
Sulfadimethoxine	8.5 ± 3.7	2.7 ± 0.4	68	1.2 ± 0.1	57	86
Tetracycline	222.0 ± 2.4	36.1 ± 4.4	84	2.2 ± 1.3	94	99
Oxytetracycline	61.9 ± 1.0	17.2 ± 0.4	72	16.2 ± 0.5	6	74
Chlortetracycline	ND	ND	—	ND	—	—
Erythromycin-H2O	591.7 ± 47.4	85.3 ± 24.0	86	ND	>99	>99
Clarithromycin	85.4 ± 2.1	34.0 ± 2.8	60	ND	>99	>99
Tylosin	ND	ND	—	ND	—	—
Penicillin G	ND	ND	—	ND	—	—
Ampicillin	270.7 ± 22.5	153.0 ± 7.5	43	ND	>99	>99
Nalidixic acid	172.3 ± 15.8	148.0 ± 11.0	14	19.4 ± 4.8	87	89
Ciprofloxacin	21.1 ± 1.1	12.4 ± 1.9	41	ND	>99	>99
Ofloxacin	156.0 ± 7.5	24.2 ± 7.4	85	ND	>99	>99
Trimethoprim	96.1 ± 1.6	22.1 ± 0.3	77	ND	>99	>99
Caffeine	6823.3 ± 906.1	24.6 ± 6.9	>99	148.0 ± 16.0	NA	98
Acetaminophen	2716.7 ± 346.2	ND	>99	ND	-	>99
Ibuprofen	657.0 ± 110.1	115.5 ± 30.7	82	5.0 ± 0.6	96	99
Naproxen	143.0 ± 17.3	45.5 ± 9.1	68	ND	>99	>99
Ketoprofen	220.0 ± 28.3	91.9 ± 42.4	58	ND	>99	>99
Gemfibrozil	248.7 ± 63.3	ND	>99	ND	—	>99
17β-Estradiol	ND	ND	—	ND	—	—
Estriol	ND	ND	—	ND	—	—
Estrone	ND	ND	—	ND	—	—
17α-Ethynylestradiol	ND	ND	—	ND	—	—
Propranolol	18.2 ± 1.4	ND	>99	ND	—	>99
pH	7.3	6.4	◎	6.6		

Sampling date: 6/4-6/5 2009. Triplicate samples were obtained and measured.

ND: not detected; NA: not available; S1, S2 and S3: The concentration (ng/L) detected at sampling sites; RE-1 (%): removal efficiency was calculated by the concentration difference between S1 and S2; RE-2 (%): removal efficiency was calculated by the concentration difference between S2 and S3; TRE (%): total removal efficiency was calculated by the concentration difference between S1 and S3

residential, industrial, and agricultural waste streams in Taiwan (Lin et al. [20]), sulfamethoxazole, caffeine, acetaminophen and ibuprofen were the compounds detected most often, present in more than 96% of the samples and at high concentrations (μg/L levels). In addition, we previously calculated total consumptions for individual medications for the year 2004 using data from the National Health Research Institute and found that acetaminophen was the number one drug prescribed in Taiwan (approximately 5.7×10^9 doses; 500 mg, 24 mg/mL) [21].

In addition, secondary and chlorination processes did not show good removal efficiencies for PPCPs (12 PPCPs had <80% removal efficiency). Tetracycline, erythromycin-H_2O, caffeine, acetaminophen, ibuprofen, gemfibrozil, ofloxacin, and propranolol removals were greater than 82%, while sulfadiazine, nalidixic acid and ciprofloxacin removals were lower than 40%. Many studies have reported that NSAIDs, estrogens, and caffeine are easily removed by biological treatment processes (secondary process), but nevertheless some antibiotics (including the sulfonamides, macrolides, and quinolones) cannot be effectively removed by biodegradation [6, 22–25]. In S2 (the effluents via secondary and chlorination processes), the residual concentrations of sulfamethoxazole, ampicillin, nalidixic acid, and ibuprofen were higher than 100 ng/L despite the high removal efficiency of ibuprofen (>80%). Therefore, it has been suggested that residual PPCP concentrations in effluents should be monitored because even in low concentrations (erythromycin-H_2O ~40 ng/L; ampicillin ~75 ng/L), some PPCPs may still affect the natural environment [26,27].

However, most target compounds (with the exceptions of oxytetracycline and caffeine) were removed effectively (with removal efficiency around 90%) in the tertiary process (UF/RO), and the overall removal efficiency for most compounds by the WRRC was >99%. The reason for the poor removal of oxytetracycline and caffeine during the tertiary process is still unknown. Due to technical difficulties, we were unable to obtain composite samples at the wastewater treatment plant. Therefore, even though each unit was grab-sampled according to its hydraulic retention time, small errors may still have occurred. Further investigation is required to verify and explain this observation.

3.3. Occurrence of PPCPs in surface waters

Fig. 2 shows the occurrence of 28 target compounds in four surface waters. 12, 10, 16, and 17 target compounds were detected in surface waters A, B, C, and D, respectively. Caffeine had the highest observed concentration (1,813 ng/L), while others were found at levels <260 ng/L. Sulfamethoxazole, sulfamethazine, sulfadimethoxine and caffeine were detected most frequently (frequency of detection >70%). Sulfamonomethoxine, tylosin, penicillin G, 17 β-estradiol, estriol, 17 α-ethynylestradiol and estrone were not detected in surface water samples. Some of these PPCPs originated from discharges of conventional wastewater treatment plants. As shown in Fig. 2, concentrations of erythromycin-H_2O and ampicillin in surface waters C and D were higher than known impact levels (erythromycin-H_2O ~40 ng/L; ampicillin ~75 ng/L). Thus our results showed good overall performance of RO, which may be used to process wastewaters to better ensure the health of humans and wildlife.

Although most PPCPs were detected at low concentrations in the effluents of conventional wastewater treatment plants, no PPCP-targeted regulations are yet in place. Because of their potential risks to humans and wildlife, it is important to make sure that reclaimed sewage waters — i.e., effluents — are free of these pharmaceuticals. In the present state, we suggest that reclaimed water should be reserved for non-potable uses, i.e., agricultural or landscape irrigation or industrial use.

4. Conclusion

This study investigated the occurrence and fate of 28 often-used PPCPs during the primary, secondary and tertiary (UF/RO) treatment processes of the WRRC and also surveyed four surface waters in northern, central and southern Taiwan. Sulfamethoxazole (1353 ng/L), caffeine (6823 ng/L) and acetaminophen (2716 ng/L) were found at higher concentrations in WRRC influents. Secondary and chlorination processes did not show good removal efficiencies for PPCPs (12 PPCPs were removed with <80% efficiency). However, most target compounds were removed effectively (with removal efficiency ~90%) in the tertiary process (UF/RO) except for oxytetracycline and caffeine, and the overall removal efficiency by the WRRC was >99%. In addition, our results indicate that many PPCPs were frequently detected at significant levels (ng/L-μg/L) in Taiwan's surface waters, while 12, 10, 16, and 17 target compounds were detected in surveyed surface waters A, B, C, and D, respectively. Caffeine had the highest observed concentration (1,813 ng/L), while others were found at levels <260 ng/L. Sulfamethoxazole, sulfamethazine, sulfadimethoxine, and caffeine were detected most frequently. Some of these PPCPs originated from discharges of conventional wastewater treatment plants. In conclusion, RO demonstrated good overall performance and may be used to process wastewater to better ensure the health of humans and wildlife.

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References

[1] P.E. Stackelberg, E.T. Furlong, M.T. Meyer, S.D. Zaugg, A.K. Henderson and D.B. Reissman, Persistence of pharmaceutical compounds and other organic wastewater contaminants in a conventional drinking-watertreatment plant, Sci. Total Environ., 329 (2004) 99–113.

[2] M. Farre, M. Petrovic and D. Barcelo, Recently developed GC/MS and LC/MS methods for determining NSAIDs in water samples, Anal. Bioanal. Chem., 387 (2007) 1203–1214.

[3] C. Zwiener, Occurrence and analysis of pharmaceuticals and their transformation products in drinking water treatment, Anal. Bioanal. Chem., 387 (2007) 1159–1162.

[4] S. Managaki, A. Murata, H. Takada, B.C. Tuyen and N.H. Chiem, Distribution of macrolides, sulfonamides, and trimethoprim in tropical waters: Ubiquitous occurrence of veterinary antibiotics in the Mekong Delta, Environ. Sci. Technol., 41 (2007) 8004–8010.

[5] S.D. Kim, J. Cho, I.S. Kim, B.J. Vanderford and S.A. Snyder, Occurrence and removal of pharmaceuticals and endocrine disruptors in South Korean surface, drinking, and waste waters, Water Res., 41 (2007) 1013–1021.

[6] K.D. Brown, J. Kulis, B. Thomson, T.H. Chapman and D.B. Mawhinney, Occurrence of antibiotics in hospital, residential, and dairy, effluent, municipal wastewater, and the Rio Grande in New Mexico, Sci. Total Environ., 366 (2006) 772–783.

[7] R. Hirsch, T. Ternes, K. Haberer and K.L. Kratz, Occurrence of antibiotics in the aquatic environment, Sci. Total Environ., 225 (1999) 109–118.

[8] G.A. Loraine and M.E. Pettigrove, Seasonal variations in concentrations of pharmaceuticals and personal care products in drinking water and reclaimed wastewater in Southern California, Environ. Sci. Technol., 40 (2006) 687–695.

[9] Y.J. Zhang, S.U. Geissen and C. Gal, Carbamazepine and diclofenac: Removal in wastewater treatment plants and occurrence in water bodies, Chemosphere, 73 (2008) 1151–1161.

[10] T. Heberer, Tracking persistent pharmaceutical residues from municipal sewage to drinking water., J. Hydrol., 266 (2002) 175–189.

[11] C.H. Huang, J.E. Renew and L. Kristen, Assessment of potential antibiotic contaminants in water and preliminary occurence analysis, Water Res., 120 (2001) 30–40.

[12] C.S. Mcardell, E. Molnar, M.J.F. Suter and W. Giger, Occurrence and fate of macrolide antibiotics in wastewater treatment plants and in the Glatt Valley Watershed, Switzerland, Environ. Sci. Technol., 37 (2003) 5479–5486.

[13] A. Gobel, A. Thomsen, C.S. McArdell, A. Joss and W. Giger, Occurrence and sorption behavior of sulfonamides, macrolides, and trimethoprim in activated sludge treatment, Environ. Sci. Technol., 39 (2005) 3981–3989.

[14] S. Kim, P. Eichhorn, J.N. Jensen, A.S. Weber and D.S. Aga, Removal of antibiotics in wastewater: Effect of hydraulic and solid retention times on the fate of tetracycline in the activated sludge process, Environ. Sci. Technol., 39 (2005) 5816–5823.

[15] T.A. Ternes, N. Herrmann, M. Bonerz, T. Knacker, H. Siegrist and A. Joss, A rapid method to measure the solid-water distribution coefficient (K-d) for pharmaceuticals and musk fragrances in sewage sludge, Water Res., 38 (2004) 4075–4084.

[16] F.J. Beltran, A. Aguinaco, J.F. Garcia-Araya and A. Oropesa, Ozone and photocatalytic processes to remove the antibiotic sulfamethoxazole from water, Water Res., 42 (2008) 3799–3808.

[17] V. Matamoros, C. Arias, H. Brix and J.M. Bayona, Removal of pharmaceuticals and personal care products (PPCPs) from urban wastewater in a pilot vertical flow constructed wetland and a sand filter, Environ. Sci. Technol., 41 (2007) 8171–8177.

[18] P.H. Roberts and K.V. Thomas, The occurrence of selected pharmaceuticals in wastewater effluent and surface waters of the lower Tyne catchment, Sci. Total Environ., 356 (2006) 143–153.

[19] S.W. Yang, J.M. Cha and K. Carlson, Simultaneous extraction and analysis of 11 tetracycline and sulfonamide antibiotics in influent and effluent domestic wastewater by solid-phase extraction and liquid chromatography-electro spray ionization tandem mass spectrometry, J. Chromatogr. A, 1097 (2005) 40–53.

[20] A.Y.C. Lin, T.H. Yu and C.F. Lin, Pharmaceutical contamination in residential, industrial, and agricultural waste streams: Risk to aqueous environments in Taiwan, Chemosphere, 74 (2008) 131–141.

[21] A.Y.C. Lin and Y.T. Tsai, Occurrence of pharmaceuticals in Taiwan's surface waters: Impact of waste streams from hospitals and pharmaceutical production facilities, Sci. Total Environ., 407 (2009) 3793–3802.

[22] K.G. Karthikeyan and M.T. Meyer, Occurrence of antibiotics in wastewater treatment facilities in Wisconsin, USA, Sci. Total Environ., 361 (2006) 196–207.

[23] A.Y.C. Lin, T.H. Yu and S.K. Lateef, Removal of pharmaceuticals in secondary wastewater treatment processes in Taiwan, J. Hazard. Mater., 167 (2009) 1163–1169.

[24] C.D. Metcalfe, B.G. Koenig, D.T. Bennie, M. Servos, T.A. Ternes and R. Hirsch, Occurrence of neutral and acidic drugs in the effluents of Canadian sewage treatment plants, Environ. Toxicol. Chem., 22 (2003) 2872–2880.

[25] M. Pedrouzo, S. Reverte, F. Borrull, E. Pocurull and R.M. Marce, Pharmaceutical determination in surface and wastewaters using high-performance liquid chromatography-(electrospray)-mass spectrometry, J. Sep. Sci., 30 (2007) 297–303.

[26] O.A.H. Jones, N. Voulvoulis and J.N. Lester, Aquatic environmental assessment of the top 25 English prescription pharmaceuticals, Water Res., 36 (2002) 5013–5022.

[27] K. Kummerer and A. Henninger, Promoting resistance by the emission of antibiotics from hospitals and households into effluent, Clinical Microbiology and Infection, 9 (2003) 1203–1214.