

Pharmaceuticals and consumer products in four wastewater treatment plants in urban and suburb areas of Shanghai

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Abstract Ten pharmaceuticals and two consumer products were investigated in four wastewater treatment plants (WWTPs) in Shanghai, China. The concentrations of target compounds in the wastewater influents ranged from below the limit of quantification (LOQ) to 9340 ng/L, with the frequency of detection of 31–100 %, and the removal efficiencies were observed to be –82 to 100 % in the four WWTPs. Concentrations of most target compounds (i.e. diclofenac, caffeine, metoprolol, sulpiride) in the wastewater influents were around three to eight times higher in urban WWTPs than in suburb ones, probably due to the different population served and lifestyles. Mean concentrations of target compounds in the wastewater influent generally decreased by 5–76 % after rainfall due to the dilution of raw sewage by rainwater, which infiltrated into the sewer system. In the WWTPs located in the suburb area, the increased flow of wastewater influent led to a shortened hydraulic retention time (HRT) and decreased removal efficiencies of some

compounds. On the contrary, the influence of rainfall was not significant on the removal efficiencies of investigated compounds in urban WWTPs, probably due to the almost unchanged influent flow, good removal performance, or by-pass system employed.

Keywords Pharmaceuticals and consumer products · Wastewater treatment · Urban · Suburb · Rainfall · Overflows

Introduction

Release of pharmaceuticals and consumer products has attracted much public attention because of their intrinsic biological activity which may cause adverse effects on aquatic and terrestrial ecosystems (Nassef et al., 2010; Melvin et al., 2014) and the frequent detection in the water environment (Focazio et al., 2008; Tran et al., 2014; Zhou and Chen 2014). Wastewater treatment plants (WWTPs) were considered as an important source of pharmaceuticals entering into the water environment. Thus, in the last decades, the occurrence and removal of pharmaceuticals have been extensively studied (Castiglioni et al. 2006; Huerta-Fontela et al. 2008; Behera et al. 2011; Sui et al. 2011; Li et al. 2013; Yu et al. 2013; Tsui et al. 2014; Guerra et al. 2014). The results showed that the behaviors of the same pharmaceutical varied much in the WWTPs located in different cities or even in different regions of the same city. For instance, Xue et al. (2010) found that concentrations of metoprolol, a type of anti-hypertensive, had a wide distribution with the mean concentration of 908 ng/L in one WWTP in Beijing, while a much lower concentration (98 ng/L) was observed in a WWTP located in another region of Beijing. Chen et al. (2012) found that the occurrence of ibuprofen, cefazolin, and penicillin V in hospital effluents were different between a metropolitan area and a county. An analysis of drugs of abuse in wastewaters from a small

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community and a large urban center indicated that the size and demographics of the population served influenced the patterns of abuse of drugs (Yargeau et al. 2014). However, the studies on the comparison of occurrence and removal performance of other pharmaceuticals in urban and suburb WWTPs are still limited.

Concentrations of pharmaceuticals also fluctuated during different sampling campaigns in the same WWTP. Rainfall may be one of the causes associated with this phenomenon (Ternes 1998; Tauxe-Wuersch et al. 2005; Vieno et al. 2007; Phillips et al. 2012; Ryu et al. 2014). That was the reason why many studies emphasized that sampling campaigns were conducted avoiding rainfall (Kosma et al. 2010; Baker and Kasprzyk-Hordern 2013). Some researchers surveyed the pharmaceuticals in the WWTPs in a period of time, during which a heavy rainfall chanced to occur, and confirmed the influence of rainfall on the occurrence and removal of pharmaceuticals in the WWTPs. For instance, Vieno and co-workers carried out a 2-week sampling investigation of pharmaceuticals and personal care products (PPCPs) in Finland, and observed a dilution of concentration in the wastewater influent by rainwater (Vieno et al. 2007). In Seoul, South Korea, the occurrence and removal of 29 endocrine-disrupting chemicals (EDCs) and PPCPs in the wastewaters under both dry and wet weather conditions were investigated, and the influence of rainfall was also found to be evident (Ryu et al. 2014). Recently, combined sewer overflows (CSOs), caused by heavy rain or snowmelt, has been identified as an environmental source of hormones and micropollutants, as the mixture of raw wastewater and stormwater can overload the capacity of WWTPs, resulting in the discharge of untreated wastewater and subsequently deteriorated quality of the receiving surface water (Weyrauch et al. 2010; Phillips et al. 2012; Madoux-Humery et al. 2013). Phillips et al. (2012) found that annual loads of trace contaminants, which could be removed by >95 % in the WWTPs, were dominated by CSO. Moreover, substances with high removal efficiencies in the WWTPs can lead to concentration peaks in the river during CSO events (Weyrauch et al. 2010). Thus, the influence of rainfall on the occurrence and removal of pharmaceuticals and consumer products in the WWTPs deserves to be studied systematically. To the best of our knowledge, the occurrence and removal of pharmaceuticals after rainfall have not been compared in between urban and suburb WWTPs in one city.

Thus, the major objective of the present study was to figure out how the location of WWTPs and demographics of the population served influenced the occurrence and removal performances of pharmaceuticals and consumer products in urban and suburb WWTPs in Shanghai. A secondary objective was to gain an insight to the influence of rainfall on the occurrence and removal of target compounds in these WWTPs.

Materials and methods

Chemicals and analysis

Twelve high-purity (96–99.9 %) standards (Table S1), including chloramphenicol, trimethoprim, diclofenac, indomethacin, mefenamic acid, bezafibrate, gemfibrozil, metoprolol, carbamazepine, caffeine, *N,N*-diethyl-meta-toluamide (DEET), and sulphiride, were purchased from Sigma-Aldrich (USA) and five related isotopically labeled compounds used as internal standards, atrazine-⁵D, DEET-⁷D, chloramphenicol-⁵D, phenacetin-¹³C, and gemfibrozil-⁶D, were purchased from Dr. Ehrenstorfen (Augsburg, Germany).

A detailed description of the analytical method, including the extraction of target compounds using solid-phase extraction (SPE) and the analysis by high-performance liquid chromatography with tandem mass spectrometry (HPLC-MS/MS), has been reported previously (Wang et al. 2014) and briefly described in supporting information.

Sample collection

Four WWTPs (A, B, C, and D) serving varied communities in Shanghai were chosen: WWTP A and B are situated in the suburb area, while WWTP C and D are in the urban area. One of the obvious differences between the urban and suburb area is the population density. According to the Shanghai Bureau of Statistics (2012), the population density in the urban districts of Shanghai ranged from 18,016 to 36,269 population/km², while in the suburb area, the number was only 1295–6700 population/km². Besides, in the suburb area, the ratio of wastewater treated to wastewater released is lower than that in the urban area of Shanghai. The main characteristics of each WWTP are summarized in Table 1. The WWTPs, mainly receiving domestic wastewater, employ similar treatment processes: primary (mechanical) treatment, secondary biological treatment, and disinfection. Schematic diagrams of treatment processes and sampling points in the four WWTPs are shown in Fig. 1.

Four sampling campaigns were conducted during July 2012 to June 2013. The first two sampling campaigns were carried out just before and after a heavy rain event in July 2012, to investigate the effect of rainfall on the occurrence and removal of pharmaceuticals and consumer products in the WWTPs. Grab sampling strategy was employed in the first three sampling campaigns due to the lack of auto-samplers. In the last sampling campaign, both grab and 24-h composite samples were collected and analyzed. Grab samples were collected in duplicate (500 mL for influents and 1000 mL for effluents) in prewashed amber glass bottles, kept in a cooler, and transported to the laboratory. Twenty-four-hour composite samples (the same volume as grab samples) were collected every 4 h manually by time-proportional mode. The

Table 1 Operational parameters of the wastewater treatment plants investigated

WWTPs	A	B	C	D
Population equivalent	95,000	180,000	250,000	230,000
Treatment process	MSBR	A ² /O	A ² /O	A/O
Average flow (m ³ /day)	16,000	32,000	50,000	44,000
Average flow after rainfall (m ³ /day)	23,870	42,260	54,250	43,450
SRT (days)	15	13–15	20	14–16
Designed HRT (h)	11	13	13.3	13
Location	Suburb	Suburb	Urban	Urban

SRT sludge retention time, HRT hydraulic retention time, MSBR modified sequencing batch reactor, A²/O anaerobic/anoxic/oxic process, A/O anoxic/oxic process

results showed that the relative difference of the concentrations of target compounds in most grab and 24-h composite samples did not exceed 30 % (Fig. S1). Therefore, the grab sampling strategy was still deemed acceptable in this case, probably due to the low inter-day variability of investigated compounds in the surveyed WWTPs.

Quality assurance and quality control

Strict quality assurance and quality control (QA/QC) was implemented to ensure accurate quantification of the target compounds. All the samples were analyzed in duplicate, and the relative difference of replicated samples was less than 20 %. In each sampling campaign, two laboratory blanks and one field blank were extracted and analyzed identically to wastewater samples. All the blanks were found below the

limit of quantification (LOQ), and only concentrations detected three times above the LOQ were reported (Sui et al. 2011). For the statistical analysis, the concentrations below the LOQ were replaced by zero. The LOQ, recoveries of pharmaceuticals, and consumer products were compiled in Table S1.

Result and discussion

Overall occurrence and removal

Concentrations and frequencies of detection of investigated pharmaceuticals and consumer products in the influents of four WWTPs during the four sampling campaigns are summarized in Fig. 2a. Five out of 12 target compounds, namely

Fig. 1 Schematic diagram of the treatment processes in the four WWTPs and sampling site location (●). MSBR modified sequencing batch reactor; A²/O anaerobic/anoxic/oxic process; A/O anoxic/oxic process

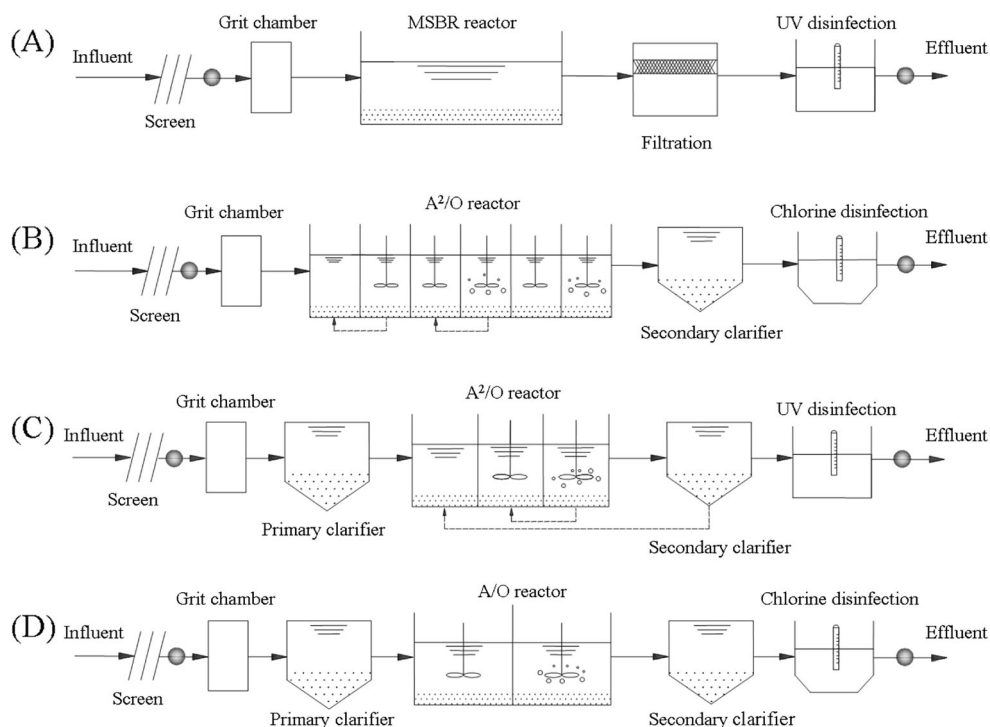
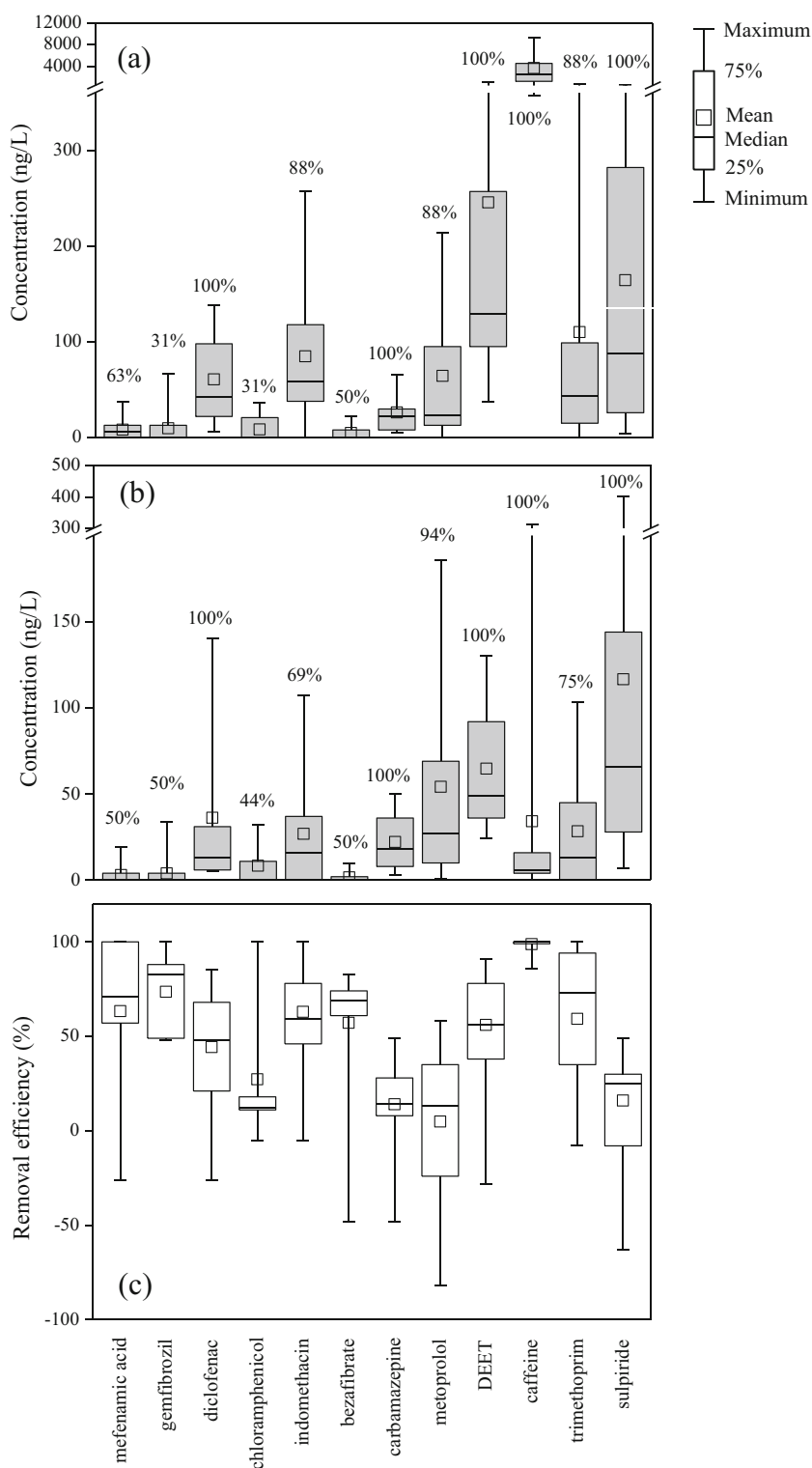


Fig. 2 Overall concentrations of target compounds in the influents (a) and effluents (b) of the four WWTPs in Shanghai as well as corresponding removal efficiencies (c) ($n=16$). Numbers in a and b indicate the frequencies of detection



caffeine, DEET, sulpiride, carbamazepine, and diclofenac, were quantified in all the influent samples, with the concentrations ranging from 4 to 9340 ng/L. The other seven pharmaceuticals were not detected in all the samples (31–88 %) and at relatively lower concentration levels. Generally, the contamination level of pharmaceuticals and consumer

products in the wastewater influents in this study was slightly lower than those previously reported (Loraine and Pettigrove 2006; Kim et al. 2007; Schirmer and Schirmer 2008; Yang et al. 2011; Behera et al. 2011; Sui et al. 2011; Li et al. 2013; Yu et al. 2013). For instance, the average concentrations of gemfibrozil and mefenamic acid were found to be 10 and 8 ng/

L in this study, but 222 and 121 ng/L in the wastewater influents of Korea (Behera et al. 2011). Diclofenac, trimethoprim, and carbamazepine were previously reported at concentrations ranging from 130 to 770 ng/L (Yang et al. 2011), while in this study, the average concentrations were 61, 110 and 26 ng/L, respectively.

A low contamination level of investigated compounds was also observed in the wastewater effluents, as shown in Fig. 2b. Mefenamic acid, gemfibrozil, chloramphenicol, and bezafibrate were found only occasionally in the final effluent, with the frequency of detection of <50 %, and the other pharmaceuticals and consumer products were detected in the range of <LOQ–402 ng/L.

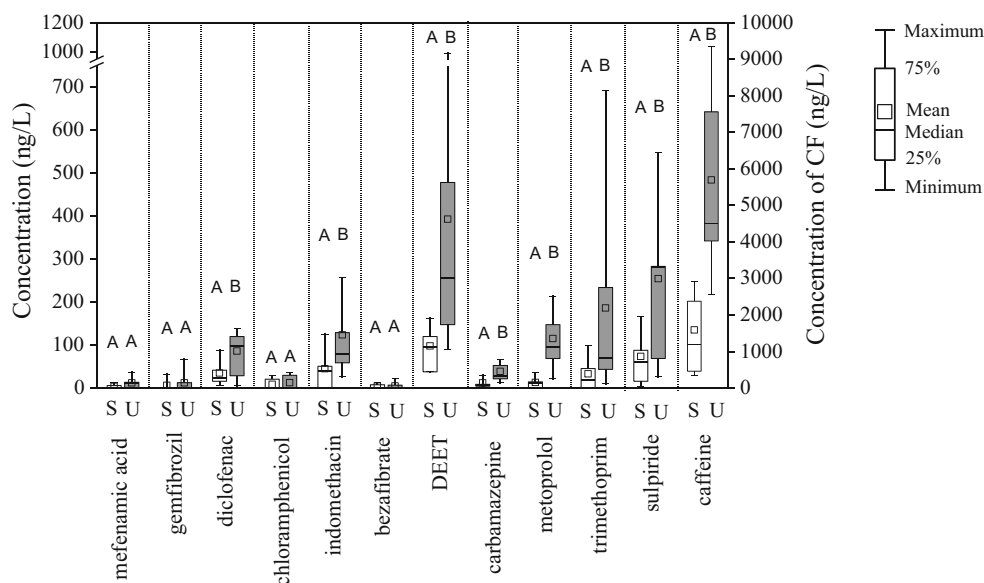
The removal efficiencies of investigated compounds were computed by comparing concentrations in the influent and effluent samples. As shown in Fig. 2c, the removal efficiencies ranged from –82 to 100 % (Fig. 2c), in good agreement with the results found elsewhere (Quintana et al. 2005; Loraine and Pettigrove 2006; Kim et al. 2007; Huerta-Fontela et al. 2008; Sui et al. 2011; Li et al. 2013; Yu et al. 2013). The apparent negative removal efficiencies were more commonly found for three investigated compounds, carbamazepine, metoprolol, and sulpiride. The phenomenon might be explained by the cleavage of conjugates (glucuronides, sulfates) of target analytes by the enzymatic processes during the secondary treatment process, producing additional amounts of target analytes in the wastewater effluent (Vieno et al., 2007; Carballa et al. 2004; Galán et al. 2012). Besides, the analytical errors caused by the grab sampling strategy and pretreatment might be another reason for the increased concentrations from influents to effluents. Among the four WWTPs, WWTP C exhibited the most efficient performance in removing pharmaceuticals and consumer

products. Removal efficiencies of five pharmaceuticals and consumer products achieved >60 % in WWTP C and mostly higher than those in the other three WWTPs (Figure S1). The longer sludge retention time (SRT) (20 days) in WWTP C might be the main cause of the better removal performance. Many researches indicated that a long SRT could promote the adaptation of different kinds of microorganisms, as well as a greater capacity for biodegradation (Kimura and Harah 2007; Suarez et al. 2010; Sui et al. 2011). A controlled study conducted by Kimura and Harah (2007) confirmed that a greater removal of diclofenac was achieved in a membrane bioreactor (MBR) operating at a longer SRT (up to 65 days) with respect to a conventional activated sludge (CAS) reactor with a SRT of 7 days due to different composition of sludge. Besides, ultraviolet (UV) light disinfection employed in WWTP C, where the UV dosage was larger (95 mW/cm²) and the total suspended solids were less (approximately 20 mg/L) than in WWTP A, would be considered as another reason for the better removal performance. Enhanced removal of some PPCPs in facilities that utilize UV light as a disinfection technique was found in our previous study (Wang et al. 2014).

Concentration comparison between urban and suburb WWTPs

Concentrations of pharmaceuticals and consumer products in the wastewater influents in all the four sampling campaigns were adopted to determine the differences between the urban and suburb WWTPs, as illustrated in Fig. 3. It is very interesting that the concentrations of most target compounds in the influents of urban WWTPs (C and D) were significantly higher than those in suburb WWTPs (A and B) according to

Fig. 3 Concentrations of pharmaceuticals and consumer products in the influents of suburb and urban WWTPs in four sampling campaigns ($n=8$). *S* indicates suburb WWTPs and *U* indicates urban WWTPs. *AB* indicates that concentrations in the two types of samples are significantly different by the nonparametric Kruskal–Wallis test at the 0.05 level, while *AA* indicates that the difference is not significant under the same condition



the nonparametric Kruskal–Wallis test at the 0.05 level. For example, metoprolol, a kind of anti-hypertensive, was measured at 69–214 ng/L in WWTP D and <LOQ–15 ng/L in WWTP A. This finding suggested that the urban area was expected to be a “hotspot” of investigated PPCPs. Similarly, Veach and Bernot (2011) quantified pharmaceutical concentrations in a suburban and agriculturally influenced stream in central Indiana, and put forward that pharmaceutical concentrations would be higher in the urban-influenced stream due to human waste contribution. Sponberg and Witter (2008) characterized the spatial distribution of selected PPCPs from three WWTPs in Northwest Ohio, USA, and also found some variability of PPCP concentrations among WWTPs in urban and suburb areas. It was suggested that the served population was a good predictor of human-use pharmaceutical release into receiving waters by comparing the pharmaceutical concentrations and loadings among treated wastewater from one rural and two urban wastewater treatment processes (MacLeod and Wong 2010). As the population density is much higher in the urban area of Shanghai (Shanghai Bureau of Statistics 2012), it might lead to higher concentrations of pharmaceuticals and consumer products in the raw wastewater received from the urban area than the suburb area. Besides, the medical security systems in urban and suburb areas of Shanghai are distinct, resulting in a higher cost of medicine for the population in the suburb area. Therefore, less consumption of pharmaceuticals in the suburb area might be predicted. Furthermore, different lifestyles and philosophies between

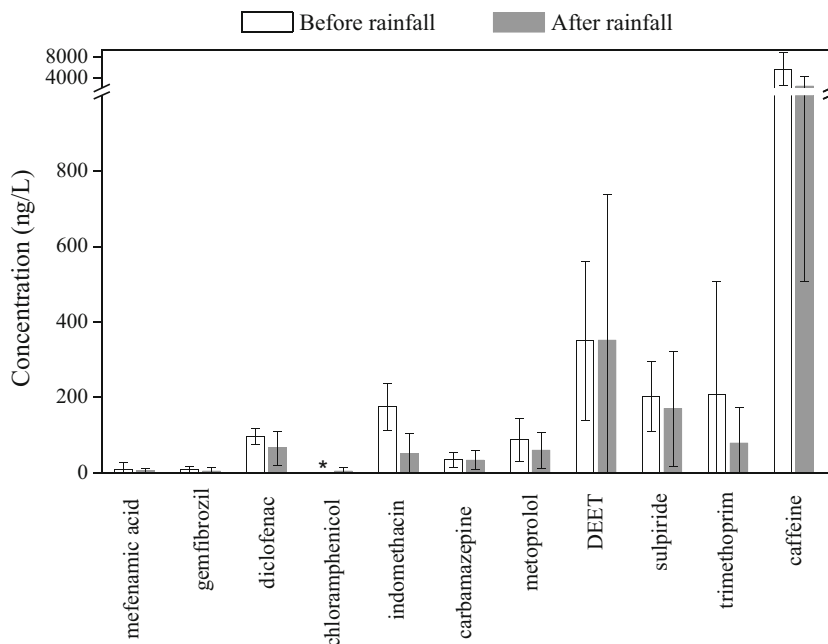
inhabitants in urban and suburb areas might also be the explanation for the different contamination levels of target compounds.

Effect of rainfall

Effect of rainfall on concentration level in the influent

Mean concentrations of target compounds generally decreased by 5–76 % after rainfall compared to those before rainfall in the influents of four WWTPs (Fig. 4). For instance, the concentration of indomethacin in the influents dropped from 71 to 257 ng/L before the rain to <LOQ–129 ng/L after the rain. For caffeine, the average concentration in the influents (2348 ng/L) after the rain was about 1/2 of that (5442 ng/L) before the rain. Although a separate sewer system covers most of the sewer service area in Shanghai, sanitary sewer overflows (SSOs) may occur because of blockages and breaks in sewer lines (Sier and Kevin 2005). The extremely heavy rain in the weather records led to an increased flow in the sewer ascribed to the rainwater flowing into the sewer system (Table 1). Accordingly, lower concentrations of pharmaceuticals and consumer products in the wastewater influents would be expected due to dilution. This is similar to the pattern of decreasing hormones and wastewater micropollutant concentrations with increasing discharge caused by the storm runoff under the CSO condition (Phillips et al. 2012). It should be mentioned that the almost unchanged wastewater flow of WWTP D after rainfall was because extra sewage reaching WWTP D was directly pumped to the receiving water and the

Fig. 4 Mean concentrations of target compounds in the wastewater influents before and after rainfall. The asterisk indicates that the concentration of the compound was <LOQ



recorded wastewater influent was lower than that actually in the sewer system.

One of the exceptions was chloramphenicol, which was only detectable at 21 ng/L in WWTP B after rainfall. As WWTP B was located in the area famous for aquaculture, and chloramphenicol was sometimes used in aquaculture, the presence of chloramphenicol in the wastewater influent after rainfall might reflect the contribution of runoff from aquaculture containing chloramphenicol to the sewage during stormflows. Elevated concentration of DEET after rainfall in WWTP D was also not expected; however, the cause was not clear.

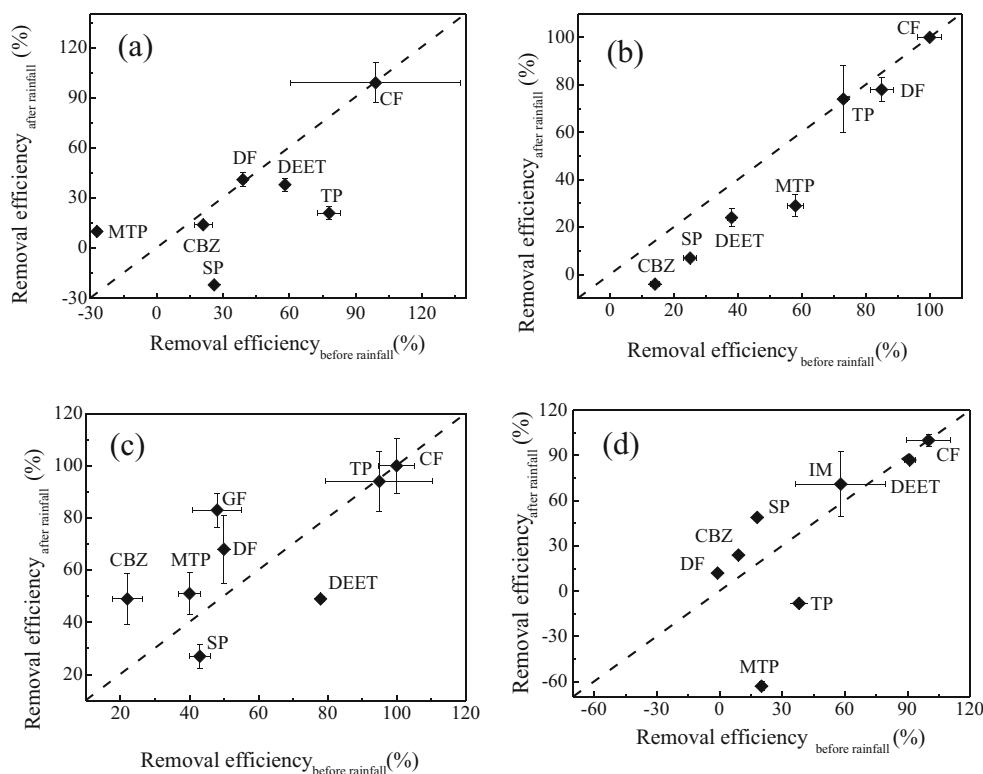
Effect of rainfall on removal efficiencies in suburb and urban WWTPs

Removal efficiencies of investigated compounds in four WWTPs before and after the rain are exhibited in Fig. 5. In WWTP A and B, removal of most pharmaceuticals and consumer products deteriorated dramatically during the rainfall day. For example, the removal efficiencies of trimethoprim in WWTP A were 78 % before rainfall and 21 % after rainfall. In WWTP B, the removal of sulphiride and metoprolol decreased from 25 and 58 % to 7 and 29 % after rainfall, respectively. This phenomenon was in accordance with the findings of Tauxe-Wuersch et al. (2005). During the long-term monitoring of PPCPs in the WWTPs, they found that removal of

ibuprofen and ketoprofen significantly decreased after a long raining period, and deduced that the rainfall event may be presumably responsible for the decreased elimination rates. Similarly, Ternes (1998) reported that in the WWTPs of Berlin, elimination rates of some pharmaceuticals, such as bezafibrate, diclofenac, naproxen, and clofibric acid, were significantly reduced on the days of heavy rainfall and did not recover until a week later. Recently, a research conducted by Phillips et al. (2012) has indicated decreased treatment efficiency that occurred for many hormones and wastewater micropollutants with increasing flows probably resulting from the lesser hydraulic retention times (HRT) during higher flows. As shown in Table 1, the heavy rain led to an elevated flow rate of about 30–50 % in WWTP A and B, which may be ascribed to the worse sewer system and more severe SSO in the suburb area. Accordingly, the shortened HRT was expected in WWTP A and B during the rainfall days. The decreased contact time of compounds and the microorganisms during the rain event might be the main cause of the deteriorated removal in WWTP A and B.

Unlike WWTP A and B, the influence of rainfall on the removal of pharmaceuticals and consumer products was not evident for WWTP C and D. The flow rate of wastewater influent in WWTP C increased by <10 %, meaning the HRT during the rainfall in WWTP C was not significantly reduced. Besides, as mentioned above, WWTP C exhibited the best performance in removing pharmaceuticals and consumer

Fig. 5 Comparison of removal efficiencies of pharmaceuticals and consumer products before and after rainfall in suburb WWTPs: **a** WWTP A, **b** WWTP B and urban WWTPs, **c** WWTP C, **d** WWTP D. *CF* caffeine, *CBZ* carbamazepine, *MTP* metoprolol, *SP* sulphiride, *GF* genfibrozil, *DF* diclofenac, *IM* indomethacin, *BF* bezafibrate, *TP* trimethoprim, *DEET* N,N-diethyl-metoluamide



products among the four WWTPs. It could be inferred that the well-operated treatment process in WWTP C was still capable of efficiently removing most investigated pharmaceuticals and consumer products even under slightly reduced HRT. This speculation could be indirectly proven by the unchanged removal performance of caffeine in all the four WWTPs after the rainfall. Caffeine could be easily eliminated from all the four WWTPs (~100 %), and the excellent removal performance was not sensitive to the external perturbations. In WWTP D, as the incoming extra sewage was pumped to the receiving water from bypass, no significant increase of the wastewater influent was observed (Table 1) after rainfall, and subsequently, the HRT during the rainfall remained unchanged. Thus, it could be understood that the removal of pharmaceuticals and consumer products after rainfall was not obviously influenced in WWTP D.

Conclusion

Ten pharmaceuticals and two consumer products were analyzed in the WWTPs located in urban and suburb areas of Shanghai. The overall concentrations of target compounds ranged from <LOQ to 9340 ng/L in the wastewater influents and <LOQ to 402 ng/L in the wastewater effluents, both of which were slightly lower than those previously reported. The urban area was found to be a hotspot of investigated PPCPs, as significantly higher concentrations of most investigated compounds were observed in the influents of urban WWTPs than suburb WWTPs. Even in the cities covered by the separate sewer system, rainfall may influence the occurrence and removal performance of pharmaceuticals and consumer products. The decreased concentrations of most target pharmaceuticals and consumer products were observed in the wastewater influents by 5–76 % due to the dilution of rainwater. The removal efficiencies were severely affected by the rainfall, decreasing by 8–73 % in either suburb WWTP, while in the urban WWTPs, the removal performances remained stable after rainfall due to the barely unchanged influent flow, good removal performance, or the employed bypass system. This study indicated that a higher risk might be posed by the investigated trace contaminants in WWTPs located in the suburb area during rainfall events. Further investigation using improved sampling strategies for pharmaceuticals and consumer products in both aqueous phase and sludge phase is required to better understand the evolution of target compounds during the wastewater treatment under different conditions.

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