



Do high levels of PPCPs in landfill leachates influence the water environment in the vicinity of landfills? A case study of the largest landfill in China



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ABSTRACT

Landfill leachates are identified as a significant source of pharmaceutical and personal care products (PPCPs), which might pose a threat to groundwater and surface water nearby the landfill. However, knowledge of PPCP contamination in the surrounding water environment of landfills is very limited. Here we investigated eighteen PPCPs in water environment near the largest landfill in China, focusing on their occurrences and spatial distribution, as well as the environmental risks. The results showed the concentration of target PPCPs was below the limit of quantification (< LOQ) to 53.6 ng/L in the adjacent groundwater samples, decreasing with the distance of sampling sites from landfill. The composition pattern of PPCPs in the groundwater was similar to that in raw landfill leachate. These observations indicated a considerable impact of raw landfill leachates on PPCP contamination in the nearby groundwater. In surface water samples, the occurrence of PPCPs was not consistent with that in raw or treated landfill leachates, but similar to that in the same watershed far away from landfill. Spatially, no obvious difference in the PPCPs concentrations was observed, indicating negligible contribution of landfill on PPCPs in surrounding surface water. The findings allowed the first insights into the impacts of well-constructed and managed landfills on PPCP contamination in the surrounding water environment.

1. Introduction

Besides flushed into the sewer system, unwanted or expired pharmaceutical and personal care products (PPCPs) were usually discarded into municipal solid wastes (MSWs), leading to a high concentration of PPCPs residues in the MSWs. For instance, approximately 8.1 mg/kg of pharmaceutical compounds occurred in the typical MSWs of Florida, USA (Musson and Townsend, 2009). In China, the average concentration of pharmaceuticals (oxytetracycline, tetracycline, and sulfamethoxazole) in the collected MSWs were 100.9, 63.8, and 47.9 µg/kg, respectively (Song et al., 2016). Till now, landfill has been the commonly used disposal method in many countries (Eggen et al., 2010, Masoner et al., 2014, Renou et al., 2008). As PPCPs in the MSWs may enter the leachate percolating through the landfill (Musson and Townsend, 2009), high concentration levels of PPCPs have been found

in landfill leachates globally (Clarke et al., 2015, Eggen et al., 2010, Peng et al., 2014, Sui et al., 2017).

In modern designed landfills, leachate is collected and treated on-site by physical/chemical treatment, biological treatment and combined (Gao et al., 2015). However, as the design of landfill leachate treatment mainly focused on conventional water qualities such as chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammonium, etc. (Kjeldsen et al., 2002), treated leachate still exhibited high concentration levels of PPCPs. For instance, the average removal efficiency of PPCPs in landfill leachates was limited (< 15%) for diclofenac (DF), gemfibrozil (GF) and carbamazepine (CBZ) during various treatment processes in Taiwan (Lu et al., 2016). Although some treatment processes, such as membrane bioreactor (MBR), demonstrated better performance for PPCPs removal (Sui et al., 2017, Yi et al., 2017, Zhao et al., 2018), their concentrations in the treated leachates

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are still high due to the extremely high concentrations in the raw leachates.

Thus, even after treatment, PPCPs released to the surrounding water environment cannot be ignored; besides, in some landfills, raw leachate might be directly discharged into the adjoining water environment unintentionally. Recently, high concentrations of PPCPs in groundwater, soil and surface water near the old and historic landfills have been observed, indicating that landfill leachates might have considerable impacts on the neighboring environment (Barnes et al., 2008, Buszka et al., 2009, Erickson et al., 2014, Lu et al., 2016). For instance, the average concentration of DF achieved up to 2.06 µg/L in the groundwater downstream of an MSW landfill in north-eastern Poland (Kapelewska et al., 2018). While in some other cases, the nearby groundwater was barely impacted by the landfill leachates. Peng et al. (2014) demonstrated that PPCPs detected in groundwaters near landfills in Guangdong, China had no significant difference compared to groundwater free from the impact of landfilling.

Therefore, it is difficult to draw from previous studies firm conclusions about whether or not the landfill leachates were the main source of PPCPs in the surrounding water environment. Further studies on the impact of landfills with different sizes, types, and locations on PPCP contamination in the adjacent environment are needed.

In this study, we investigated a wide range of pharmaceuticals in surface water and groundwater adjacent to the largest MSW landfill in China, which received a total of 8.8 million ton's MSW, accounting for greater than 50% of the MSWs in Shanghai (Sui et al. 2017). The investigated landfill has advanced leakage prevention systems, such as sufficient liners, leachate-collection system, and on-site treatment, representing a typical modern landfill in China. Spatial distribution, seasonal variations and potential risks of PPCPs in the groundwater and surface water were discussed. This work would provide the first evidence for the impact of well-constructed and managed landfills on the PPCP contamination in the nearby aquatic environment.

2. Material and methods

2.1. Chemicals

Chemical standards including carbamazepine (CBZ), gemfibrozil (GF), diclofenac (DF), bezafibrate (BF), sulphiride (SP), trimethoprim (TP), and nine sulfonamides (sulfadiazine (SD), sulfamerazine (SMR), sulfisoxazole (SIX), sulfisomidine (SIM), sulfamethoxyypyridazine (SMP), sulfamethazine (SMT), sulfadimethoxine (SDM), sulfamonomethoxine (SM), sulfamethoxazole (SMX)) were purchased from Sigma-Aldrich (Germany). Caffeine (CF) and metoprolol (MTP) were purchased from Dr. Ehrenstorfer (Germany) and chloramphenicol (CP) was from TCI (China). Six isotopically-labeled compounds, including phenacetin-¹³C and GF-⁶D from CIL (USA), atrazine-⁵D and mecoprop-¹³D from CDN (Canada), CP-⁵D from Witega (Germany) and SMT-¹³C from Sigma-Aldrich (Germany) were used as internal standards (IS).

HPLC-grade methanol and acetonitrile were purchased from J&K (USA). Ultra-pure water was generated by water purification system Classic DI from ELGA (UK). Stock solutions of individual compounds were prepared in methanol. Working solutions with different concentrations were prepared by mixing and diluting the stock solutions. All the solutions were stored at 4 °C in the dark.

2.2. Study area and sampling

The investigated landfill, located at the entrance where the Yangtze River joins the East China Sea, is the largest MSW landfill in China. It features a warm and humid subtropical climate with an annual precipitation of 1056 mm and an average temperature of 15.5 °C. The landfill, equipped with advanced leakage prevention systems and the leachate-collection system, generates 2700 m³ leachates per day. The

leachates first undergo an external MBR treatment process and then are discharged to a nearby municipal wastewater treatment plants for further treatment.

Landfill leachates, groundwater and surface water samples in the vicinity of landfills were collected in three sampling campaigns in January, May and October. Raw landfill leachates were obtained from the equalization tank prior to the treatment process. Treated landfill leachates were sampled in the final effluent. Groundwater samples were collected in ten wells from four villages located in the west of the landfill with different distances from the landfill (2.6–4.0 km) given that there was no groundwater well in the east of the landfill. The four villages have similar population density and residential size. The wells in the villages, providing waters for washing and drinking for the inhabitants, have the depths of 5–10 m. Since the depths of confined groundwater level in this area varies seasonally, ranging from 3 to 11 m underground, the collected groundwater samples were from shallow confined aquifer. The flow direction of groundwater in the investigated region is uncertain (Tian 2005, Zhao et al. 2017) due to the seawater intrusion. Sampling of surface water in the vicinity of landfills was conducted in Suitang River, which is 2 km away from the landfill. Three sampling sites, near landfill (N), 2.1 km upstream (U) of sampling site N, and 3.0 km downstream (D) of sampling site N, were selected. In addition, to evaluate the influence of landfill leachates on the surface water, we simultaneously collected surface water samples far away from the landfill in the same watershed, facilitating the comparison between surface water samples in different locations.

Sampling sites of groundwater and surface water are shown in Fig. 1. The distances of villages from the landfill were 2.6 (A), 2.7 (B), 3.5 (C) and 4.0 (D) km, respectively. All the samples were stored in prewashed amber glass bottles, kept in a cooler, transported to the laboratory, and stored at 4 °C before analysis. The sample holding time was less than 24 h before pretreatment.

2.3. Pretreatment and analysis

Target PPCPs were extracted using solid-phase extraction (SPE) and analyzed by high-performance liquid chromatography with tandem mass spectrometry (HPLC-MS/MS), which was reported by Sui et al. (2017). Briefly, after samples were filtered through prebaked (400 °C, > 4h) glass fiber filters (Whatman, UK), an aliquot of the filtrate (100 mL for landfill leachate, 400 mL for surface water, and 1000 mL for groundwater) was spiked with 100 µL IS solution (1000 µg/L for sulfonamides and 400 µg/L for other PPCPs) and adjusted to pH 4 (for sulfonamides) or 7 (for other PPCPs) with 1 M sulfuric acid. Then, the samples were loaded on SPE cartridges (HLB, 6 mL, 500 mg, Waters, USA), and 6 mL acetonitrile (for sulfonamides) or methanol (for other PPCPs) was used to elute the cartridges at a rate of 1–2 mL/min. The extracts were concentrated to approximately 1 mL by a gentle flow of high purity nitrogen and filtered through a 0.2 µm filter (Millex-FG, Millipore, USA) followed by LC-MS/MS analysis. The instrumental analytical method was described in Tables S1 and S2.

2.4. Quality assurance and quality control (QA/QC)

Spiked and un-spiked samples (n ≥ 3) were pretreated and analyzed identical to real samples to evaluate the recoveries of the analytical method. Relative recoveries of most PPCPs in the landfill leachate, surface water and groundwater were satisfactory (60–130%). The limits of quantification (LOQ) based on instrumental quantification limits and recoveries, were 0.42–1.4 µg/L for landfill leachates, 10–150 ng/L for surface water and 3–61 ng/L for groundwater. Field, procedural and instrumental blanks were set to monitor potential contamination during the sampling, pretreatment and instrumental analysis. None of the target analytes was quantifiable in the blanks. The relative standard deviation of duplicate environmental samples was within 20%. Detailed information about QA/QC was summarized in Table S3.

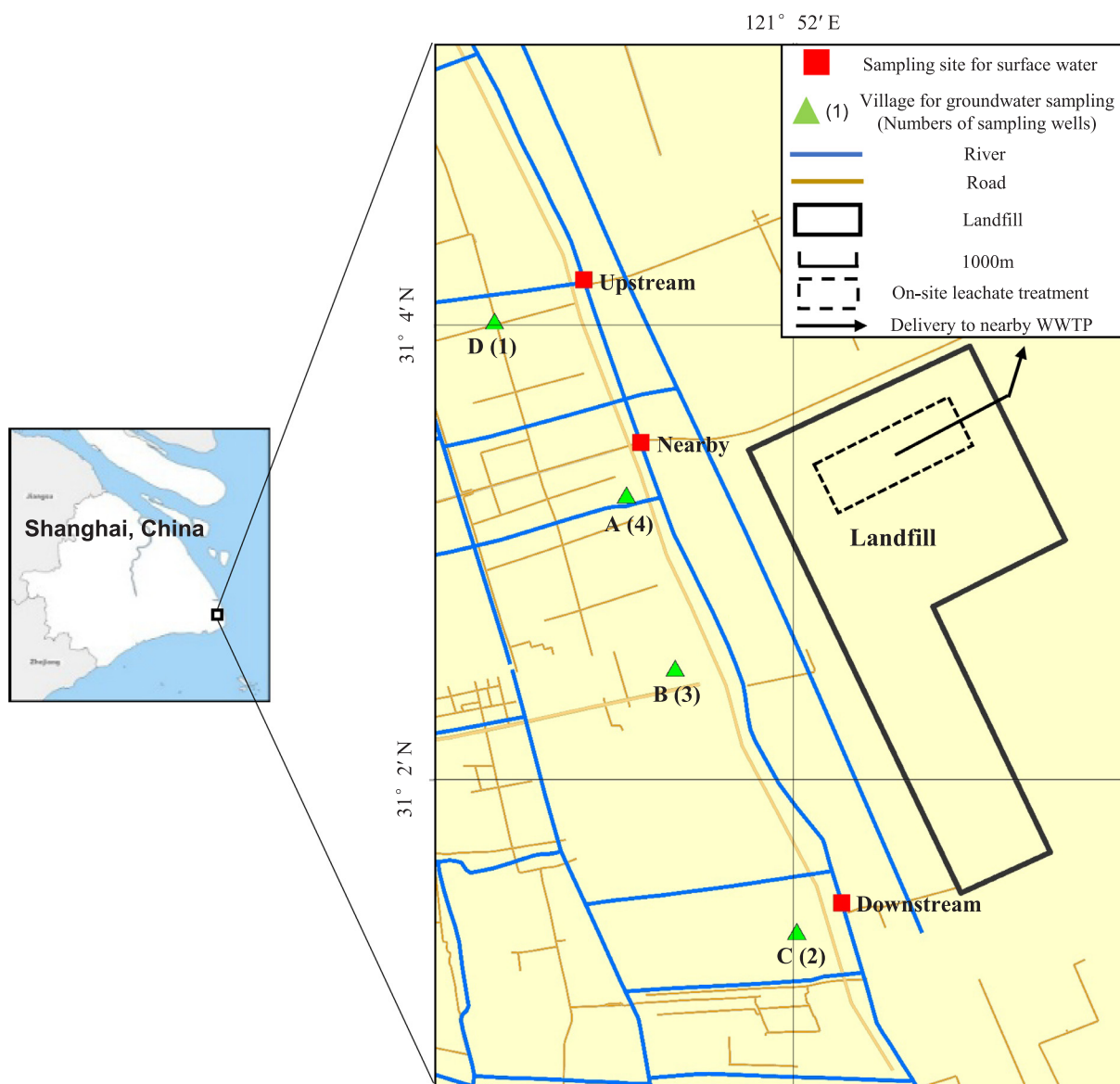


Fig. 1. Sketch map of the municipal landfills and sampling sites for groundwater and surface water.

2.5. Statistical analysis

Data processing was performed by Origin 9.5 (OriginLab, Northampton, MA, USA) and SPSS 20 (SPSS, Chicago, IL, USA). The level of significance was set at $\alpha = 0.05$. For target PPCPs, once the detected concentrations were below LOQ, half of the LOQs were used in the statistical analysis.

3. Results and discussion

3.1. Groundwater

Concentrations and detection frequencies of target compounds in groundwater samples adjacent to the landfill were presented in Fig. 2. In 70% of the groundwater samples, target compounds were detected at different concentration levels. Relatively high detection frequencies were observed for DF (70%), followed by CF (61%) and SIM (43%), while five compounds, namely CP, SD, SMR, SIX and SDM were not detected in any groundwater samples. The most abundant compound detected was CF ($< \text{LOQ}$ -53.6 ng/L). SM and SMP followed, of which the maximum concentrations were 28.2 and 26.8 ng/L, respectively.

Raw landfill leachate had extremely high concentration level of PPCPs, ranging from 0.39 to 349 $\mu\text{g/L}$, and slightly lower concentration was detected in treated landfill leachates ($< \text{LOQ}$ to 10.6 $\mu\text{g/L}$) (Sui et al., 2017). Comparing the concentrations and detection frequencies of target PPCPs in raw leachates, treated leachates, surface water and groundwater samples (Fig. 3), we could find that PPCP residues in the groundwater were associated with the investigated landfill. The composition pattern of PPCPs in groundwater was quite similar to that in raw leachate. CF and DF were the most abundant contaminants, while BF exhibited the lowest concentration level in both raw leachates and groundwater samples. CF and DF also showed higher detection frequency while CP, SMR, SIX and SDM could not be detected in neither raw leachate nor groundwater. Pearson correlation analysis further demonstrated the significant correlation between the concentrations and detection frequencies of PPCPs in groundwater and raw leachate sample ($p < 0.05$). On the contrary, the composition pattern of treated leachate and surface water, as shown in Fig. 3, deviated to some extent from the groundwater samples. Therefore, raw landfill leachates were proposed as one of the important sources of PPCPs in the investigated groundwater.

Previous researches also showed that landfill leachate had

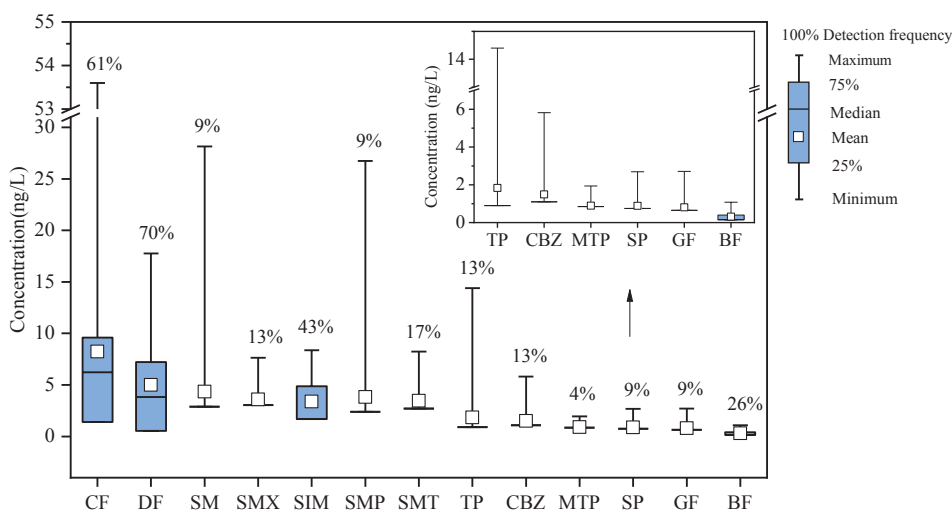


Fig. 2. Box-plots of the detected PPCPs in the adjacent groundwater of the landfill (n = 23).

considerable impacts on nearby groundwater (Kapelewska et al., 2018, Lu et al., 2016, Rodriguez-Navas et al., 2013). For example, the concentration of DF in the groundwater near a landfill reached approximately a quarter of that in the landfill leachates in Poland (Kapelewska et al., 2018). In two simple and old landfills in Taiwan, concentrations of ketoprofen, CBZ, and DF in the nearby groundwater fell in their concentration range in the landfill leachates (Lu et al., 2016). Compared to these studies, PPCP contamination level in the investigated groundwater was lower. For instance, CF was detected at a concentration of < LOQ-53.6 ng/L in the groundwater in the present study, while < LOQ-130 ng/L in groundwater downgradient from the landfill in Indiana, USA (Buszka et al., 2009). The relatively lower PPCP contamination level may be attributed to the modern design of studied landfill, having sufficient liners and leachate-collection system that reduce the possibility of PPCPs permeating into soil and groundwater effectively. On the contrary, unlined or insufficiently lined landfills were identified to be the sources of emerging contaminants to adjoining groundwater, even after the landfills were closed for many decades (Andrews et al., 2012, Barnes et al., 2004). In addition, the relatively long distance of groundwater sampling sites from landfill (2.6–4.0 km), which weakened the impact of landfill leachate on the groundwater, might be another reason for the lower concentration of PPCPs in the groundwater (Buszka et al., 2009, Kapelewska et al., 2018, Kapelewska et al., 2016, Lu et al., 2016).

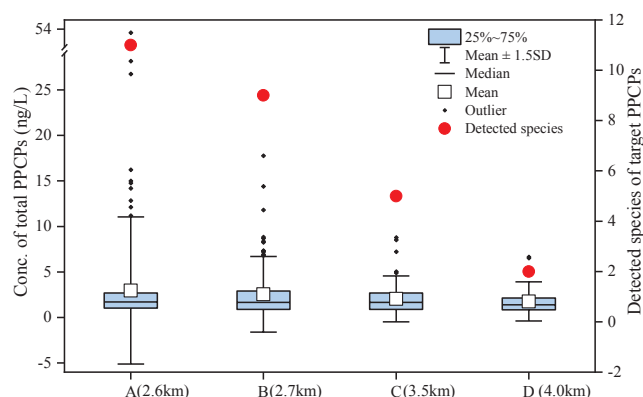


Fig. 4. Spatial distribution of sum PPCPs detected in the groundwater from different sampling sites.

Fig. 4 shows the spatial distribution of target PPCPs in terms of types and concentration levels. More types and higher concentrations of target PPCPs were detected in the groundwater collected in the villages with a shorter distance to the landfill. Specifically, out of eighteen PPCPs, eleven and nine types were detected in the groundwater in village A and B, respectively, which were 2.6–2.7 km away from the landfill. In comparison, only five and two PPCPs could be found in

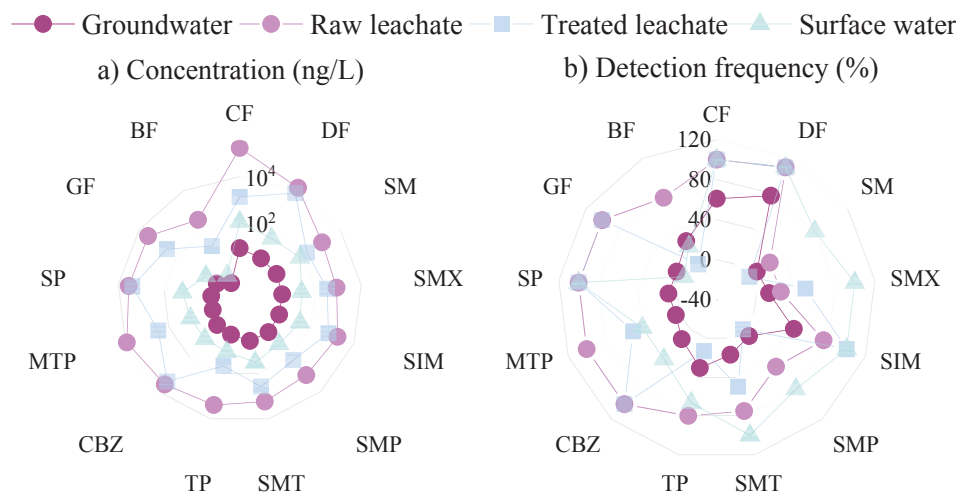


Fig. 3. Comparison of PPCPs concentrations (a) and detection of frequency (b) in raw leachates, treated leachates, surface water, and groundwater.

village C and D with the distance of 3.5–4.0 km between the sampling sites and landfill. In addition, the concentration of PPCPs were also higher in village A and B. The concentration of sum PPCPs in the groundwater in village A and B were 0.15–53.6 ng/L and 0.15–17.8 ng/L, while in village C and D, they were 0.15–8.78 ng/L and 0.15–6.66 ng/L, respectively.

Given the similar residential size and population density in the investigated villages, the variation among the four sampling sites was mainly caused by the different distances between the sampling sites and landfill. Sampling sites for groundwater closer to the landfill were also found to have higher concentrations of PPCPs (Barnes et al., 2004, Holm et al., 1995). In their studies, the distance between sampling sites for groundwater and landfill sites was less than 500 m, making the differences more prominent. Longer distance led to less remarkable differences between sampling sites. For instance, Peng et al. (2014) found that no clear correlation could be generated between PPCPs concentration and the distance of the sampling sites from the landfill in the range of 582–3178 m. The spatial distribution of PPCPs in the groundwater suggested the contribution of landfill leachates to PPCP contamination in the nearby groundwater. Even though the investigated landfill has a composite liner and leachate-collection system, there might be a risk of leakage during the long-term application, leading to the adverse impacts to the surrounding groundwater.

3.2. Surface water

In the surface water samples, all the target compounds except GF, CP, SMR, and SIX could be detected. Among the detected PPCPs, six PPCPs (CF, SP, SD, SIM, SMT, and SMX) showed the detection frequency of 100%. The mean concentrations of individual target compounds ranged from < LOQ to 112 ng/L. CF, DF, and SM exhibited higher concentrations. Their maximum concentrations reached 199–399 ng/L.

We compared the occurrence of PPCPs in the surface water collected in the vicinity of the landfill with those far away from the landfill in the same watershed (Mei et al. 2018), as showed in Fig. 5. There was no significant difference in the PPCPs concentrations between the two groups of samples ($p = 0.597$) by conducting the evaluation using the Mann-Whitney U test based on a significance level of 5%. For individual PPCPs, 11 out of 14 detected PPCPs exhibited no significant difference in their concentrations between the two types of surface water samples (Fig. 5). Thus, the potential sources of PPCPs in the surface water in the vicinity of the landfill were suggested to be similar to those far away from the landfill.

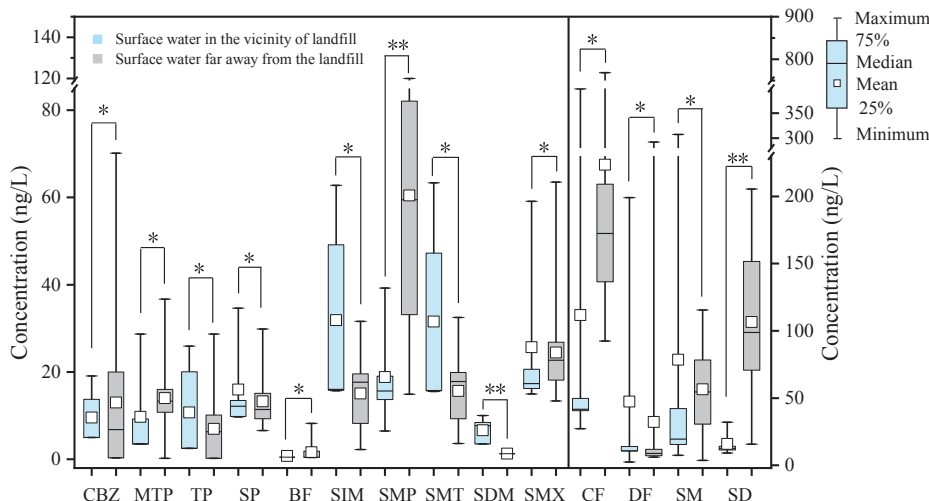


Fig. 5. Comparison of detected PPCPs in the surface water in the vicinity of the landfill and far away from the landfill. ** indicates that concentrations in the two types of samples are significantly different by U test at the 0.05 level, while * indicates that the difference is not significant under the same condition.

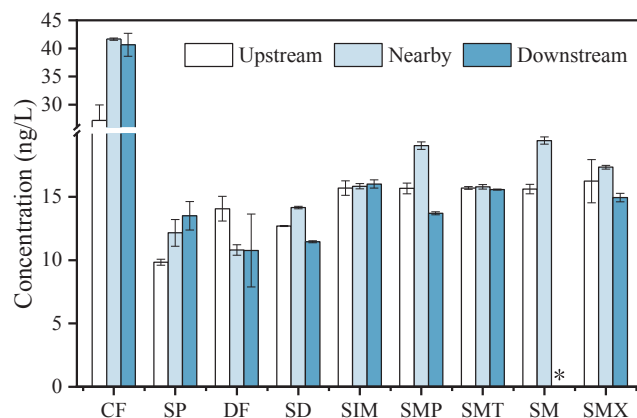


Fig. 6. Concentration of frequently detected PPCPs in the surface water upstream, nearby and downstream the investigated landfill (* indicates that the detected concentrations were below LOQ).

Besides, the composition patterns of target PPCPs in the studied surface water were inconsistent with raw or treated landfill leachates (Sui et al., 2017). For instance, the main contributors of PPCPs in the surface water were CF (26%) and SM (18%), quite different from those in the treated landfill leachates, where DF, CBZ, and SP made the largest contribution (43%, 15% and 14%, respectively) to the total PPCP contamination (Fig. S1). Therefore, it can be inferred that the contribution of landfill leachates to the PPCPs in the surrounding surface water was negligible.

This speculation could be verified by comparing the occurrence of frequently detected PPCPs in the surface water upstream, nearby and downstream the investigated landfill (Fig. 6). For most investigated PPCPs, the occurrence of the three sampling sites had no significant difference ($p = 0.277$ according to T-test, $\alpha = 0.05$).

Generally, influence of landfill leachate on the ambient surface water is mainly caused by i) direct discharge or leak of landfill leachate, whether treated or untreated, into the surrounding surface water, and ii) natural exchange process between the surface water and the hyporheic zone (Lewandowski et al. 2011). In the present landfill, the collected leachates are not discharged into ambient surface water. They undergo an effective treatment process (Sui et al. 2017), and the effluent is delivered to a nearby WWTP. Thus, the direct release of landfill leachate to the surface water was negligible. Although landfill leachate had some impacts on the surrounding groundwater, the contaminants

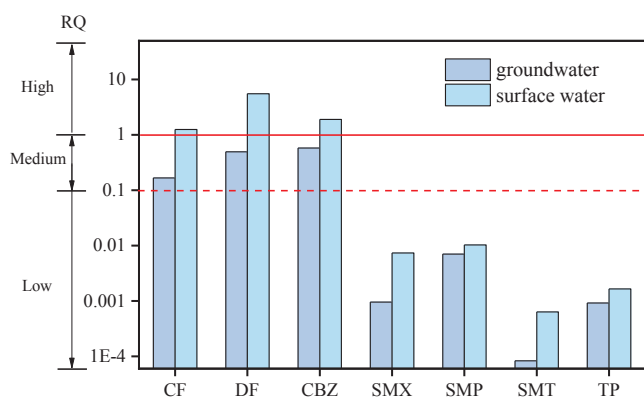


Fig. 7. Preliminary ecological risk assessment of PPCPs detected in the surface water and groundwater in the vicinity of investigated landfill.

exchange in the water cycle between surface water and groundwater seems to be limited.

3.3. Environmental risk assessment

Environmental risk assessment, based on risk quotients (RQ) method, is adopted to evaluate the potential risks of PPCPs to aquatic organisms in the groundwater and surface water proximal to the investigated landfill. The RQ value was defined as the ratio between the measured environment concentration (MEC) and predicted no-effect concentration (PNEC) of pollutants, and the risk levels are defined according to the value of RQ: $0.01 < RQ < 0.1$ indicating low risk, $0.1 < RQ < 1.0$ indicating medium risk and $RQ > 1.0$ indicating high risk (Hernando et al. 2006). To consider the worst case, the maximum concentration detected was calculated as MEC. Detailed environmental risk assessment method was described in supporting information and Table S4.

Among the target PPCPs, CF, DF, and CBZ posed a high environmental risk to sensitive aquatic organisms in the surface water (Fig. 7). While in the groundwater, RQ of CF, DF, and CBZ, nevertheless, posed medium risks ($RQ > 0.1$). Other PPCP showed RQ values of lower than 0.1, demonstrating limited adverse ecological consequences in the surrounding water environment nearby the investigated landfill.

Very few studies focused on the environmental risk assessment of PPCPs in the water environment adjacent to landfill previously (Zhao et al. 2016). Some studies demonstrated that SMX and DF posed environmental risks to the surrounding groundwater near the landfills (Kapelewska et al. 2018, Peng et al. 2014). For instance, in Poland, the RQ of DF exceeded 10 for all the trophic levels considered in groundwater downgradient of a landfill (Kapelewska et al. 2018). While other studies indicated no significant toxicity in the groundwater downstream of the landfills, i.e. in Taiwan, China (Lu et al. 2016). Our investigation provided useful information on the environmental risks caused by PPCPs in the water environment near modern landfills and is helpful to clarify the inconsistencies of previous studies.

4. Conclusion

In the present study, we investigated the influence of landfill leachate generated in the largest landfill in China on the PPCP contamination in the surrounding water environment. Thirteen out of eighteen target PPCPs belonging to different therapeutic classes were detected in the adjacent groundwater samples, ranging from $< LOQ$ to 53.6 ng/L. Concentrations of PPCPs in the groundwater decreased with the distance from the landfill, and the composition pattern was similar to that in the raw landfill leachate. These observations indicated raw leachates had a considerable impact on PPCP contamination in the nearby groundwater. In the surrounding surface water samples, the

occurrence of PPCPs was inconsistent with that in the raw or treated landfill leachates, but similar to the occurrence in the same watershed far away from landfill. Spatially, no obvious difference in the PPCPs concentrations was observed. Therefore, it could be inferred that the impacts of the landfill on the PPCPs in the surrounding surface water were negligible. This study provided the first case for the influence of a well-constructed and managed landfill on the PPCP contamination in the surrounding water environment, which is helpful to comprehensively understand the impact of landfills with diverse construction and management levels on the emerging contaminants in the surrounding environment.

CRediT authorship contribution statement

Xia Yu: Methodology, Writing - original draft. **Qian Sui:** Conceptualization, Writing - review & editing, Resources, Supervision. **Shuguang Lyu:** Resources, Supervision. **Wentao Zhao:** Conceptualization, Writing - review & editing. **Xuqi Cao:** Investigation, Methodology. **Jiusi Wang:** Investigation. **Gang Yu:** Project administration.

Declaration of Competing Interest

The authors declared that there is no conflict of interest.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2019.105404>.

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