



Impacts of reclaimed water irrigation on the accumulation of pharmaceutical and personal care products in soil and cereals

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Abstract

The use of reclaimed water in agriculture is an effective way to alleviate water scarcity. However, reclaimed water often contains pharmaceutical and personal care products (PPCPs), which might be introduced into agricultural soil and crops through reclaimed water irrigation. In this study, a field experiment was conducted to investigate the concentrations of PPCPs in the soil-cereal system after reclaimed water irrigation in 2015 and 2016 in the North China Plain. Three different water qualities were used for summer maize and winter wheat: reclaimed water, mixed water (reclaimed water and groundwater, 1:1), and groundwater. The total concentration of 11 PPCPs in winter wheat grain, summer maize grain and the topsoil (0–20 cm depth) was 1.37–7.65, 4.33–5.07, and 5.85–13.7 µg/kg, respectively. And the total bioaccumulation factors (BCFs) of the 11 PPCPs for summer maize and winter wheat were 3.40–5.23 and 0.11–0.21, respectively. There were no significant differences in the PPCP concentrations in the topsoil-cereal system or in the BCFs among the three irrigation treatments. The total hazard quotients for the 11 PPCPs were 0.019 and 0.053 for adults and toddlers, respectively, suggesting that consumption of the cereal grains in the current research (irrigation with groundwater, mixed water, or reclaimed water) presented little risk to human health.

Introduction

Wastewater recycling is an effective way to alleviate water scarcity in agricultural production (ICID 2017). Approximately 2.57×10^9 m³ of reclaimed water was used as irrigation water in agricultural production in the Yangtze River valley (Hu and Wu 2018). As the reclaimed water has been used in agricultural irrigation in some arid and semi-arid regions, there are three main sources of irrigation water in these areas, i.e., groundwater, surface water, and reclaimed water. However, some harmful substances (such as Pharmaceutical and personal care products, PPCPs) cannot be completely removed by wastewater treatment plants (WTPs) (Christou et al. 2017). Biel-Maeso et al. (2018) found that the concentrations of sulfamethoxazole (SMX) and trimethoprim (TMP) in the raw wastewater (influent of WTPs) were 55–1300 and 59–1310 ng/L, respectively; whereas the concentrations of SMX and TMP in the reclaimed water (effluent of WTPs) were 26–633 and 33–788 ng/L, respectively. The levels of TMP and SMX in treated wastewater collected from Cyprus were 22–73 and 26–55 ng/L, respectively (Christou et al. 2017). PPCPs are, therefore, introduced into environment via wastewater and reclaimed water discharge (Mordechay et al. 2021). Sun et al. (2018) found that there

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was a reduction in root activity and photosynthesis pigments when the concentration of PPCPs in nutrient solutions was 5–50 µg/L. PPCPs in human and wildlife tissues can disrupt endocrine functions (Ziylan-Yavas et al. 2022). PPCPs have now emerged as contaminants in the environment due to their harmful effects on plants and humans (Patel et al. 2019).

PPCPs have been detected in water (surface water, groundwater, and so on), soil, and plants in many areas. For example, sulfamethoxazole (SMX), trimethoprim (TMP), and ibuprofen (IP) have been detected in both surface water and groundwater in India (Sengar and Vijayanandan 2022); Hanna et al. (2018) reported that SMX has been detected in both drinking water and river water in Shandong, China, while chloramphenicol (CHL) has been found in soil, sediment, and vegetables; sulfamethazine (SMT), SMX, and TMP have been found in irrigation water, soil, and agricultural products in Israel (Mordechay et al. 2021). The main sources of PPCPs in agricultural systems (products and soil) may be wastewater/reclaimed water, livestock manure, and biosolids (Gottschall et al. 2012; Jin et al. 2020; Mordechay et al. 2021; Pan and Chu 2017a).

In recent years, many studies have investigated the fate of PPCPs in the soil–plant system and the risk to human health from contaminated irrigation water or soil (Ahmed et al. 2015; Biel-Maeso et al. 2018; Manasfi et al. 2021). Carter et al. (2014) investigated the concentration of PPCPs in crops (ryegrass and radish) and their impact on human health using pot experiments with soil spiked with PPCPs. Pan and Chu (2017a) studied the concentration of PPCPs in different crop tissues, their bioconcentration factor (BCF) and translocation factor (TF), and human exposure under wastewater irrigation (water added with PPCPs at levels of 2 and 20 µg/L). Goldstein et al. (2014) studied the uptake processes of PPCPs by different vegetables under reclaimed water irrigation and fresh water irrigation using pot experiments and found that the uptake of PPCPs by vegetables was impacted by the physicochemical properties of these compounds, the soil properties, and the physiology of the plant. Meffe et al. (2021) surveyed plant uptake of PPCPs and their effects on human health under field conditions irrigated using surface water highly contaminated with WTP effluents in Spain and found that positively ionized chemicals accumulated the most. Mordechay et al. (2021) reported the accumulation of PPCPs in soils and crops obtained from commercial fields under reclaimed water/wastewater irrigation in Israel. Pan et al. (2014) reported the accumulation of PPCPs in crops or soil with fishpond water/wastewater irrigation in some areas in the Pearl River Delta, China. Other researchers have also studied the impacts of irrigation with tertiary treated wastewater on PPCP accumulation in soil and vegetables using field experiments (Al-Farsi et al. 2018; Christou et al. 2017; Papaioannou et al. 2020).

To date, only very few studies have investigated the impact of exposure to complex, realistic reclaimed water (effluent of WTPs) on topsoil PPCP concentrations and their uptake by cereal grains using field experiments. Hence, this experiment was conducted in a winter wheat–summer maize field in the North China Plain to (1) study the PPCP concentrations in topsoil and cereal grains under a realistic reclaimed water irrigation system, and (2) determine the accumulation ability of PPCPs in cereal grains (i.e., the BCFs) and human health risks.

Materials and methods

Experimental setup

This research was performed at Beijing Irrigation Test Center Station, China (114°20'E; 39°20'N) from October 2014 to October 2016. Over the past 30 years, the average annual temperature was 11.5 °C, and the average annual rainfall was 565 mm, and the summer rainfall account for more than 70% of the annual rainfall. The groundwater table at the experimental station was approximately 8.0 m below the ground surface.

The research was conducted with summer maize (*Zea mays* L.) and winter wheat (*Triticum aestivum* L.). The winter wheat varieties were Jimai, Shimai, Shifu, Zhongmai, and Nongda (Jimai was replaced by Lunxuan in 2016). The summer maize varieties were Tianyumi, Jiyuan, Nianyumi, Jingdan, and Xinyu. For winter wheat, urea (46.7% N) was used at the returning green and planting stage, with an application rate of 416 kg/ha both times. For summer maize, urea was used at the planting stage (application rate was 416 kg/ha) and jointing stage (application rate was 300 kg/ha), respectively. Considering the three main sources of irrigation water and some ditches in many areas have been recharged with reclaimed water, three irrigation treatments were tested for all varieties: irrigation using groundwater, referred to as G treatment; irrigation using reclaimed water, referred to as R treatment; and mixed irrigation using reclaimed water and groundwater (v/v = 1:1), referred to as M treatment (which was similar to surface water). There were 3 repetitions for each treatment, and there were 45 plots in total. The length and width of all plots were 3 m and 2 m, respectively. Figure S1 shows the schematic of the experiment. To avoid contamination caused by horizontal water flow, a polyethylene geomembrane was installed on each side of each plot down to a depth of 100 cm. The field managements were the same among the three irrigation treatments with the exception of water quality. Surface irrigation was used in this study. No other agrochemicals (such as organic manures, fungicides, herbicides, or insecticides) were applied during the experiments. Table 1 provides the field management details. The

Table 1 Water-fertilizer management and sowing and harvesting time for cereal during 2015–2016

Variety	Sowing date (M-D-Y)	Fertilization (urea, kg/ha)	Irrigation (mm)	Harvest date (M-D-Y)
Winter wheat	10-7-2014	832	435	6-15-2015
Summer maize	6-18-2015	716	410	9-25-2015
Winter wheat	10-2-2015	832	532	6-20-2016
Summer maize	6-22-2016	716	390	10-2-2016

studied PPCPs included SMX, SMT, IP, TMP, CHL, acetyl-sulfamethoxazole (AC-SMX), sulfachloropyridazine (SCP), sulfathiazole (STZ), sulfamonomethoxine (SMM), sulfamerazine (SMZ), and sulfisoxazole (SX). The physicochemical properties of the studied PPCPs are shown in Table 2.

The secondary effluent supplied by Gaobeidian WTP was used as reclaimed water in the current research. The groundwater was supplied by a local well in this station. Total concentration of 11 PPCPs in the groundwater and reclaimed water were 0.014–0.114 (average = 0.058) and 0.335–1.14 (0.602) $\mu\text{g/L}$, respectively, and the total concentrations of 11 PPCPs in groundwater were significantly lower than these in reclaimed water (Table S1). Considering the amounts of irrigation water used and the concentrations of the 11 PPCPs at all irrigation events, the volume-weighted concentrations of the 11 PPCPs in the reclaimed water for cereals in 2015, winter wheat in 2016, and summer maize in 2016 were 0.64, 0.52, and 0.65 $\mu\text{g/L}$, respectively. Tables S1 shows

the concentration of individual PPCP in the groundwater and reclaimed water, Tables S2 shows the concentrations of other substances in the groundwater and reclaimed water. The irrigation water quality in the current research met the corresponding standards for agricultural irrigation (GB5084-2005 and GB20922-2007).

Sampling and measurements

Samples collection

At each harvest of cereals, topsoil (0–20 cm) samples and grain samples were collected. When collecting grain and soil samples, five evenly spaced samples were taken along a diagonal transect in all plots, and then the five samples were thoroughly mixed to get one sample. A vacuum drying oven was used to dry the grain and soil samples, then these dried samples were ground and sieved by a 0.2-mm

Table 2 The molecular weight, CAS number, pKa, the fraction of neutral molecules (f_n), solubility in water, $\log K_{ow}$, and the pH-dependent octanol–water partition coefficient ($\log D_{ow}$) of the PPCPs

Compound	Molecular weight (g/mol)	CAS number	pKa	Solubility in water (mg/L)	$\log K_{ow}$	f_n (pH=8.2) ^a	$\log D_{ow}$ ^b
SCP sulfachloropyridazine	285	80-32-0	1.76;5.71 ^c	35 ^d	0.31 ^c	0.997	0.31
CHL chloramphenicol	323	56-75-7	5.50;9.61 ^e	2500	0.92 ^e	0.0375	– 0.51
STZ sulfathiazole	255	72-14-0	2.2;7.24 ^f	373	0.05 ^f	0.901	0.05
SMZ sulfamerazine	264	127-79-7	2.06;6.9 ^f	202	0.44 ^f	0.952	0.44
IP ibuprofen	206	15,687-27-1	4.91	41	3.97	0.0005	0.68
AC-SMX acetyl-sulfamethoxazole	295	21,312-10-7	5.6 ^g	–	–	0.997	–
SMM sulfamonomethoxine	280	1220-83-3	2.0;6.0 ^e	4030	0.7 ^e	0.994	0.70
SMT sulfamethazine	278	57-68-1	2.07;7.49 ^e	1500	0.76 ^e	0.837	0.76
SX sulfisoxazole	267	127-69-5	5	300	1.01 ^d	0.999	1.01
SMX sulfamethoxazole	253	723-46-6	1.6;5.7 ^f	610	0.89 ^f	0.997	0.890
TMP trimethoprim	290	738-70-5	7.12 ^h	400 ^f	0.91 ^h	0.923	0.91

^aTrapp (2009)^bTanoue et al. (2012)^cShelver et al. (2010)^dChen et al. (2015)^eGao et al. (2015)^fChen and Zhou (2014)^gYuan et al. (2019)^hWu et al. (2013).

sieve (stainless steel) for subsequent PPCPs detection. During sample collection and processing (such as drying, grinding, rinsing, and sieving), no plastic was used to avoid PPCPs contamination.

Reagents

The standards (> 98% purity) for all the PPCPs were obtained from Sigma-Aldrich (Shanghai, China). The isotopically labeled internal standards (SMX-d4 and IP-d3) were also obtained from Sigma-Aldrich. Each solvent used in this study was high-performance liquid chromatography grade. All solutions were used within 6 months.

Sample extraction and detection

The extraction of target PPCPs from samples was performed according to the Soxhlet extraction method. In brief, SMX-d4 and IP-d3 (100 µg/kg) were added to soil samples (10 g), then these samples were extracted by Soxhlet extraction with acetonitrile (240 mL). Anhydrous sodium sulfate was then used to dry the extracted solutions, and the supernatants were concentrated to 1.0 mL by rotary evaporators and N₂ stream blowing. A membrane filter (0.22 µm) was used to filter the extracted solution, and the final solution was stored in a refrigerator (− 20 °C) for later analysis. The extraction of PPCPs from grain samples was performed in the same way, except that 2 g of grain samples and 0.22 L of acetonitrile were used.

The PPCP concentrations in the extracted solution were analyzed using an HPLC–MS/MS system (LCMS 8040, Shimadzu, Japan). An Inert Sustain C18 column (particle size of 5 µm, 150 mm × 4.6 mm; Tokyo, Japan) was used for the separation. The injection volume for these samples was 10 µL, the flow rate was 1 mL/min, and the column temperature was 40 °C.

Quality analysis and control

Every day, each instrument was calibrated according to calibration standards, and blank samples (such as spiked, solvent blanks, and procedural) were run after every 25 grain or soil samples. No PPCPs were found in each solvent blank. The recovery of surrogates was 82–120 %, and the recovery of the studied PPCPs was 82–105 %. The limits of quantification (LOQ) for these studied PPCPs in topsoil and grain samples were 1.4–15.9 and 3.59–35.2 ng/kg, respectively. Figure S2, Tables S3 and S4 show the other parameters for the detection of all PPCPs.

Calculations and statistical analyses

The accumulation ability of PPCPs in cereal grains is often estimated according to BCFs, which are calculated using the following equation:

$$BCF = \frac{C_g}{C_s} \quad (1)$$

Where *BCF* is the bioaccumulation factors, *C_s* and *C_g* represent the concentrations of the studied PPCPs in soil and cereal grains (dry weight, µg/kg), respectively.

The hazard quotient (HQ) was used to estimate the human health risks for grain consumption, HQ was calculated by the following equation (Prosser and Sibley 2015):

$$HQ = \frac{EDI}{ADI} \quad (2)$$

$$EDI = \frac{(C_g \times I_v \times \beta_{w/d})}{W} \quad (3)$$

where EDI represents the estimated daily intake of the PPCPs, µg/(kg.day); *C_g* represents the highest accumulation of the studied PPCP in cereal grains (dry weight), µg/kg; *β_{w/d}* represents the conversion factor (wet-to-dry) for grains; *I_v* represents the total consumption of grains, g/day (wet weight); *W* represents the weight of a toddler or an adult, kg; and ADI represents the acceptable daily intake of PPCPs over a person's lifespan without causing adverse effects, µg/(kg.day). In this study, for an adult, *I_v* was 300 and 30 g/day for wheat and maize (wet weight), respectively (Li et al. 2018); for a toddler, *I_v* was 150 and 20 g/day for wheat and maize (wet weight), respectively; *β_{w/d}* was 0.88 for both wheat and maize; and *W* for a toddler and an adult were 15 kg and 70 kg, respectively. For SCP, SX, STZ, SMT, SMM, SMZ, and AC-SMX, the ADI were all 10 µg/(kg.day); while the ADI for IP, SMX, TMP, and CHL were 11.4, 5.7, 5.7, and 11 µg/(kg.day), respectively (Food and Agriculture Organization of the United Nations 2011; Prosser and Sibley 2015). Humans can be exposed to harmful compounds in several ways, such as by eating contaminated food, inhalation, ingestion, and dermal contact (Li et al. 2018). Therefore, there is no potential health risk to humans if the HQ is less than 0.1.

The statistical analysis was performed using Microsoft Excel 2010 and SPSS 20.0 (IBM Corp., New York, USA). The statistical significance (5% level) of the differences in concentrations and BCFs of the studied PPCPs among these different irrigation treatments was performed using the least significant difference test (LSD). If the studied PPCPs in samples were less than the limit of detection (LOD), a value of zero was applied to calculate the average values and sums,

and a value equal to half of the LOD was applied for the LSD test.

Results and discussion

Total accumulation of the 11 PPCPs in cereal grains and 0–20 cm soil depth

The total concentrations of the 11 PPCPs in 0–20 cm soil depth at cereal harvest are shown in Fig. 1 (the values in June 2015 are the sum of TMP, IP, and SMX). For each harvest time, the differences in the total concentrations of PPCPs in 0–20 cm soil depth among the R, G, and M treatments were not statistically significant (e.g., $P=0.337–0.47$). In addition, there were almost no significant differences in each PPCP concentration in the topsoil among the three irrigation treatments (except the concentration of SCP in June 2016). This indicates that irrigation with reclaimed water or mixed water did not affect the total accumulation of PPCPs in 0–20 cm soil depth compared to groundwater irrigation. The total accumulation of 11 PPCPs in 0–20 cm soil depth in the current research were similar to the values reported by Liu et al. (2020), who studied similar PPCPs and used similar sources of reclaimed water, and found that the total accumulation of 11 PPCPs in 0–20 cm soil treated with reclaimed water/wastewater in suburbs of Beijing (the suburb was not far from the experimental station in the current research) were 0.03–28.1 $\mu\text{g}/\text{kg}$.

The total accumulations of the 11 PPCPs in wheat/maize grains during 2015 and 2016 are shown in Fig. 2. It shows that the total accumulations of the 11 PPCPs in summer maize and winter wheat were 4.33–49.2 and 1.37–6.77 $\mu\text{g}/\text{kg}$, respectively. The values in the current research are comparable to the values in Liu's study, in which the accumulations of 11 PPCPs in agricultural products treated with reclaimed water/wastewater in Beijing ranged from <0.01 to 28.0 $\mu\text{g}/\text{kg}$ (Liu et al. 2020). For each variety of cereal in this study, no statistically significant differences were obtained in the concentrations of 11 PPCPs in grains among the R, G, and M treatments ($P=0.299–0.933$). This indicates that irrigation with mixed water or reclaimed water did not significantly impact the accumulations of 11 PPCPs in cereal grains compared to groundwater irrigation.

Accumulation of individual PPCP in cereal grains and topsoil

The concentration of individual PPCP in 0–20 cm soil depth and cereal grains for each variety and each treatment was shown in Tables S5–S7. It shows that there were almost no significant differences in the individual PPCP concentrations in topsoil and cereal grains among the three irrigation treatments with four exceptions (SCP for topsoil in 2016, SMX for Shifu (winter wheat in 2015) and Jingdan (summer maize in 2015), and AC-SMX for Nianyumi). Figure 3 shows the concentrations of each studied PPCP in 0–20 cm soil depth and cereal grains for all treatments. For these soil samples, the concentrations

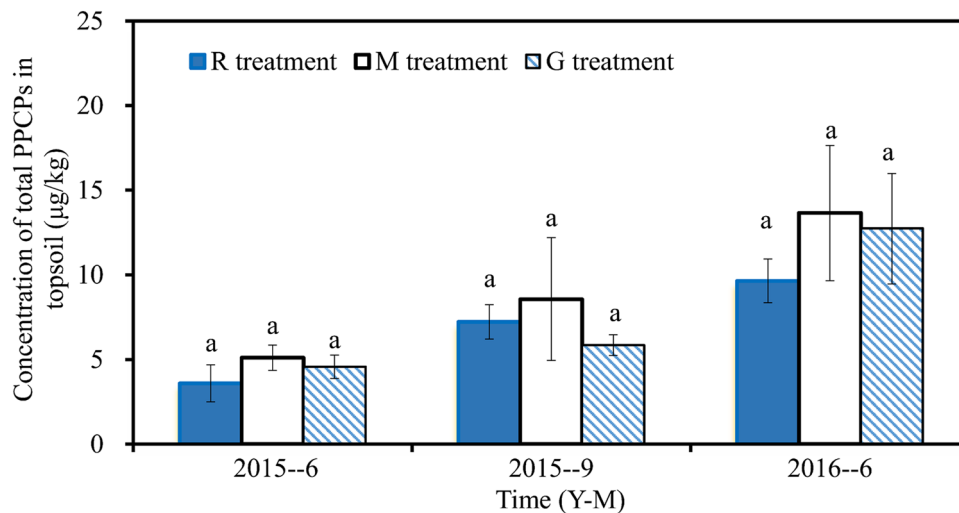
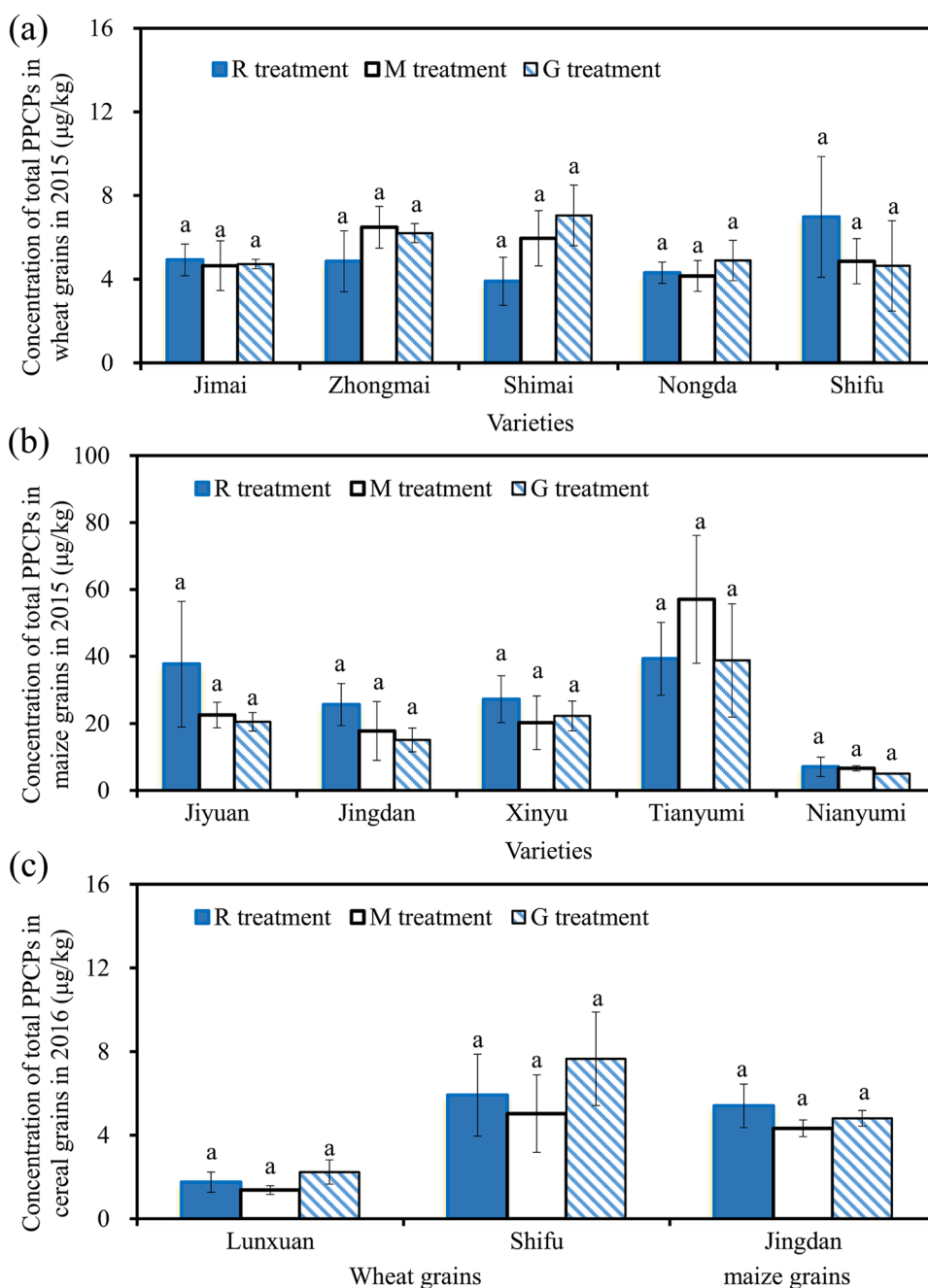


Fig. 1 Total accumulations of the 11 PPCPs in 0–20 cm soil depth at cereal harvest during 2015–2016 (the values in June 2015 were the sum of TMP, IP, and SMX). R represents irrigation using reclaimed water, G represents irrigation using groundwater, M represents irrigation using mixed water (reclaimed water and groundwater, v/v = 1:1).

No significant differences ($P=0.337–0.47$) were found in the average concentration of the 11 PPCPs in 0–20 cm soil depth among these irrigation treatments for harvest. Error bar on the histogram indicates the standard deviation ($n=3$)

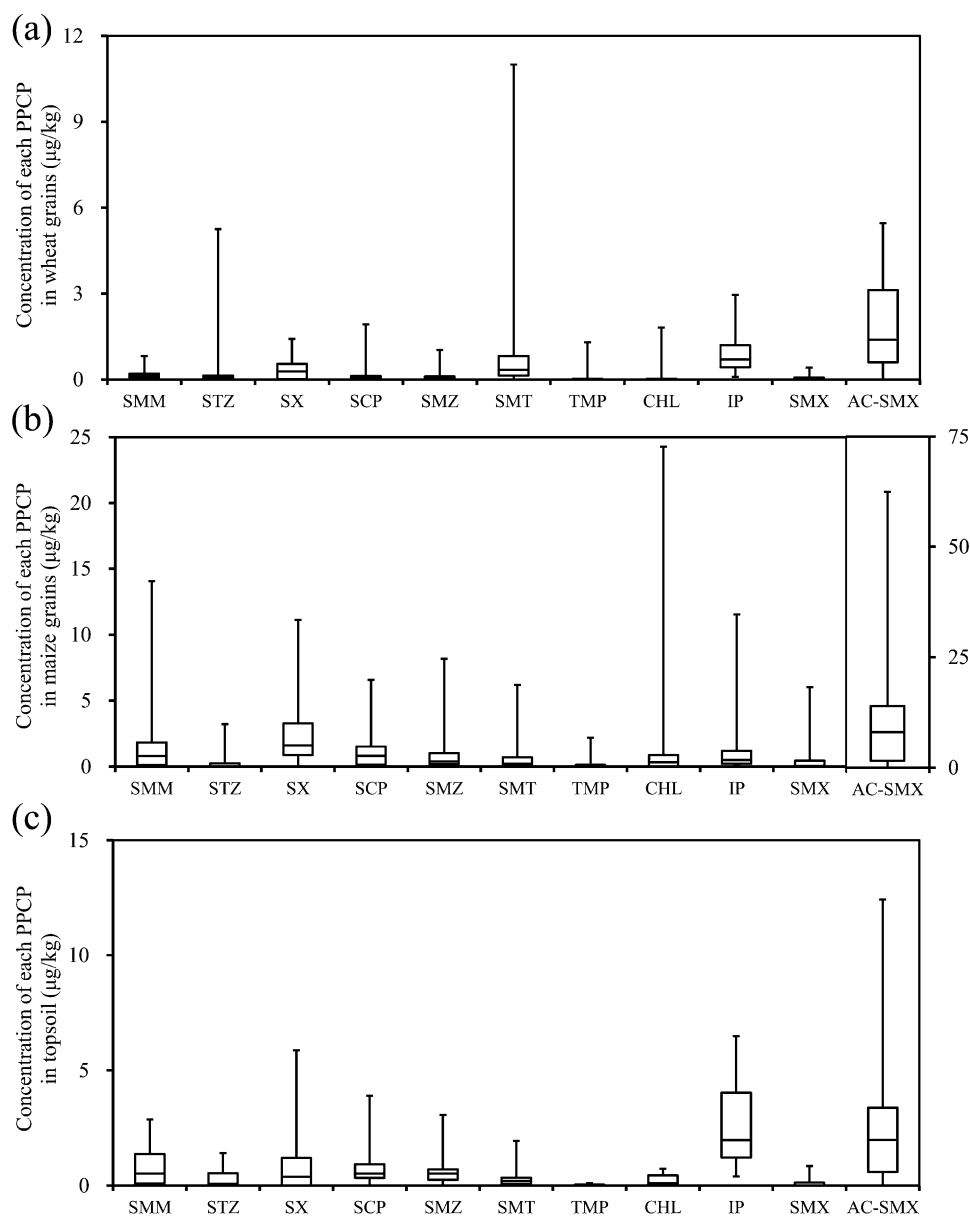
Fig. 2 Concentrations of the 11 PPCPs in cereal grains during 2015–2016. R represents irrigation using reclaimed water, G represents irrigation using groundwater, M represents irrigation using mixed water (reclaimed water and groundwater, v/v=1:1). No significant differences ($P=0.299\text{--}0.933$) were obtained in the average concentrations of the 11 PPCPs in grains among these irrigation treatments for all varieties. Error bar on the histogram indicates the standard deviation ($n=3$)



of AC-SMX, IP and SX were higher than the concentrations of the other PPCPs; the lowest concentrations were obtained for TMP, SMX, CHL, and SMT. A high input of PPCPs from irrigation water can result in high accumulation of these compounds in the soil (Biel-Maeso et al. 2018). The properties of the soil and the physicochemical properties of the PPCPs (i.e., adsorption, degradation, octanol–water partition coefficient ($\log K_{ow}$), and water solubility) might also affect the sorption of the PPCPs into the soil (Xu et al. 2021). Hu et al. (2019) reported that PPCPs with low water solubility and high $\log K_{ow}$

(hydrophobicity) is readily absorbed by the soil. Its retransformation from the parent PPCPs (Yuan et al. 2019) and its high level in the water (García-Galán et al. 2013) can explain the high accumulations of AC-SMX in these topsoil samples. Its high level in the water, high $\log K_{ow}$ (shown in Table 2 and Table S1), and low water solubility (IP) could explain the high concentrations of IP and SX in topsoil. The adsorption coefficients (K_d) of SMX and SMT were only 1.13–2.41 and 1.37 L/kg, respectively, due to their weak interactions and sorption in soil (Hu et al. 2019; Pan and Chu 2016). Kodešová et al. (2016) reported

Fig. 3 Concentrations of each studied PPCP in 0–20 cm soil depth and cereal grains during 2015–2016. SCP, SMT, AC-SMX, SX, CHL, IP, SMZ, SMX, TMP, SMM, and STZ represent sulfachloropyridazine, sulfamethazine, acetyl-sulfamethoxazole, sulfisoxazole, chloramphenicol, ibuprofen, sulfamerazine, sulfamethoxazole, trimethoprim, sulfamonomethoxine, and sulfathiazole, respectively. The solid line indicates the median value, the box indicates the 25th and 75th quartiles, the lower whisker indicates the minimum, and the upper whisker indicates the maximum



that the half-life of SMX in soil (pH range 7.17–8.07) was only 4.66–5.83 days. Koba et al. (2017) found that TMP in nearly each type of studied soil degraded rapidly. Therefore, the low concentrations of SMX, SMT, CHL and TMP in topsoil might be explained by their low K_d (SMX and SMT), high degradation rate (SMX and TMP), and high water solubility (SMT and CHL).

The accumulations of SMX in topsoil in the current research ranged from ND to 0.846 $\mu\text{g}/\text{kg}$, which were comparable to values reported by Bigott et al. (2022), who reported that the concentrations of SMX in soil with wastewater irrigation were ND–0.31 $\mu\text{g}/\text{kg}$. The concentrations of SMX in topsoil in current study were also comparable with those (0.38–0.98 $\mu\text{g}/\text{kg}$) obtained from fields irrigated with reclaimed water in Cyprus (Christou et al. 2017). The

comparable levels of SMX in the irrigation water (22.2 and 34.4 ng/L in the current research and Cyprus, respectively) could largely explain this result. Santiago-Martín et al. (2020) found that the concentrations of IP in soil irrigated with contaminated surface water in central Spain were 0.58–12.5 $\mu\text{g}/\text{kg}$. The concentrations of IP in topsoil in this study (0.4–6.49 $\mu\text{g}/\text{kg}$) were similar to the values in central Spain, which was likely due to the comparable levels of IP in the water used in central Spain (100–250 ng/L) and in the current research (16.1–331 ng/L , Table S1). Pan et al., (2014) found the concentrations of CHL (average concentration was 8.3 $\mu\text{g}/\text{kg}$) in fields treated with domestic wastewater and fishpond water in the Pearl River Delta (PRD) were higher than the corresponding values (ranged from ND to 0.73 $\mu\text{g}/\text{kg}$) in the current research, which could be mostly

explained by the higher concentrations of CHL (ranged from 40.5 to 154 ng/L) in the irrigation water as well as the long-term wastewater irrigation (> 20 years) in the PRD.

TMP, SMX, and STZ had the lowest concentrations of the studied PPCPs in both summer maize and winter wheat in the current research. The PPCPs with the highest concentrations in grains were AC-SMX and IP in wheat and SX in maize. Studies have reported that high levels of PPCPs in soil or irrigation water could result in a high accumulation of PPCPs in crops (Ahmed et al. 2015; Pan and Chu 2017b). PPCPs can be anionic, neutral, zwitterionic, or cationic; the forms of the PPCPs are related to their pKa and the pH of the environment (Boxall et al. 2012; Malchi et al. 2014). Trapp (2004) reported that roots could easily take up neutral chemicals by diffusion, which decreases with decreasing log K_{ow} or log D_{ow} (pH-adjusted octanol–water partition coefficient). Once inside the roots, neutral chemicals can translocate from the roots to the shoots according to the water gradient (Dodgen et al. 2015). However, it is difficult for ionic chemicals to cross the permeable membranes, resulting in lower concentrations of ionic than neutral chemicals in plants (Sunyer-Caldú and Diaz-Cruz 2021). These results indicate that the high concentrations of AC-SMX and IP in cereal grains are mainly related to their high concentrations in both the irrigation water (Table S1) and 0–20 cm soil depth. Its high log D_{ow} (Table 2) and high levels in the topsoil and water may account for the relatively high concentrations of SX in the grains. The low accumulation of TMP in these grains in this research were mainly due to the very low levels of TMP in the topsoil. The low accumulations of STZ in cereal grains could be explained by its low log D_{ow} (0.05, Table 2). Bigott et al. (2022) reported that the concentration of SMX in the leaves of lettuce was less than that in the roots, since SMX could be considered almost non-ionic at the given soil pH (8.2) but anionic in the plant cytosol (pH=7.2). Therefore, it could be taken up by roots easily but might be trapped in the roots and have reduced translocation

from the roots to the leaves. Bhalsod et al. (2018) also found that SMX tends to accumulate in the roots due to its low TF (0.07). Therefore, the low concentrations of SMX in cereal grains in the current research were mostly due to the very low TF and the low accumulations in the topsoil.

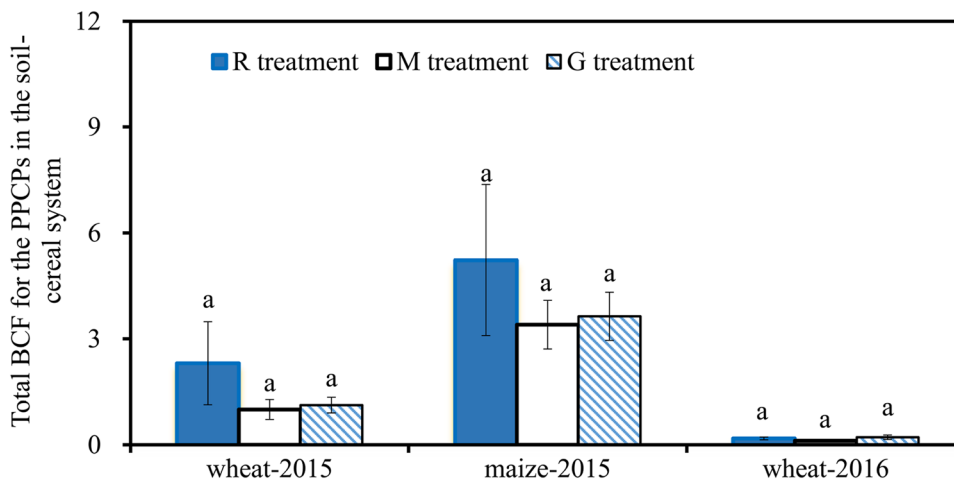
The accumulations of SMX in wheat grains in the current research were 0.05 ± 0.03 $\mu\text{g}/\text{kg}$, which are lower than those (0.64 ± 0.37 $\mu\text{g}/\text{kg}$) in wheat grains irrigated with treated wastewater in the United States (Franklin et al. 2016). This is mostly due to the levels of SMX in the irrigation water and the method of irrigation; surface irrigation was used in the current research and the SMX concentrations in the irrigation water were 0.61–32.7 ng/L (Table S1), whereas spray irrigation was used in Franklin's study and the SMX concentrations in the irrigation water were 580–22,000 ng/L. The average concentration of IP in maize grains in this study was 0.99 ± 0.84 $\mu\text{g}/\text{kg}$, which is comparable to the concentration (0.2–1.1 $\mu\text{g}/\text{kg}$) in maize grains irrigated with contaminated surface water in central Spain (Santiago-Martín et al. 2020), which is likely related to the similar levels of IP in the soil and irrigation water for the two studies (see Lines 247–251).

BCFs of PPCPs

The total BCFs of the 11 PPCPs are shown in Fig. 4. For both summer maize and winter wheat, there were no significant ($P=0.301$ – 0.666) differences in the total BCFs of the 11 PPCPs among the R, G, and M treatments. In addition, there were also no significant differences in the BCFs of individual PPCP among the R, G, and M treatments. This indicates that irrigation with reclaimed water or mixed water did not affect the BCFs of the PPCPs compared to groundwater irrigation.

The BCFs of each PPCP is shown in Fig. 5. For individual PPCP in the current research, AC-SMX and SX obtained the highest BCFs, while STZ, IP, CHL, and SMX obtained the lowest BCFs. Dudley et al. (2018) investigated

Fig. 4 Total bioaccumulation factors (BCFs) for the 11 PPCPs in the soil-cereal system during 2015–2016. R represents reclaimed water irrigation, G represents groundwater irrigation, M represents mixed irrigation (reclaimed water and groundwater, v/v = 1:1). At each harvest, no significant ($P=0.301$ – 0.666) difference was found in average BCF of the 11 PPCPs in the soil-cereal system among these different treatments. Error bar on the histogram indicates the standard deviation ($n=3$)



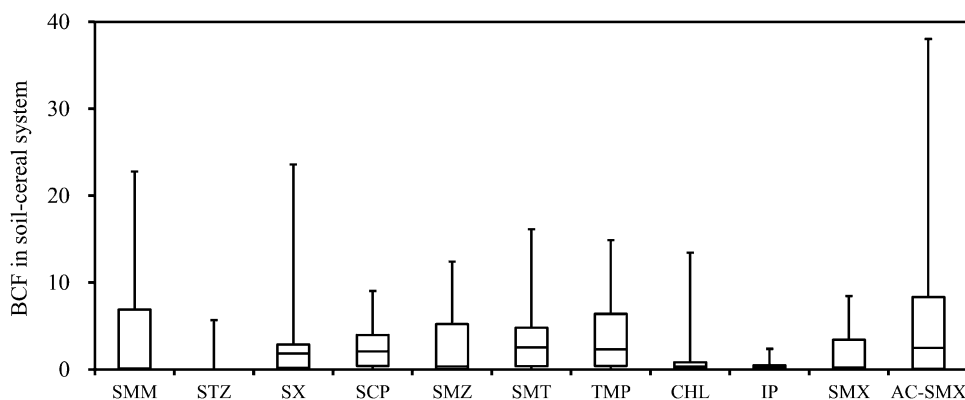


Fig. 5 The bioaccumulation factors (BCFs) of individual PPCP in the soil–cereal system during 2015–2016. SCP, SMT, SX, AC-SMX, CHL, IP, SMZ, SMX, TMP, SMM, and STZ were sulfachloropyridazine, sulfamethazine, sulfisoxazole, acetyl-sulfamethoxazole, chloramphenicol, ibuprofen, sulfamerazine, sulfamethoxazole, trimetho-

prim, sulfamonomethoxine, and sulfathiazole, respectively. The solid line indicates the median value, the box indicates the 25th and 75th quartiles, the upper whisker indicates the maximum, the lower whisker indicates the minimum

the metabolism of SMX in cucumber seedlings and found that AC-SMX was the major metabolite, which could explain the high BCF of AC-SMX. The extent of ionization (f_n) and lipophilicity ($\log K_{ow}$ or $\log D_{ow}$) of PPCPs have a great impact on their accumulation, migration, and translocation in soil–plant systems (Pan and Chu 2017b). Generally, neutral chemicals (high f_n) cross cell membranes faster than ionic chemicals (Malchi et al. 2014). Wu et al. (2013) reported that there was a positive relationship between $\log BCF_{root}$ of neutral PPCPs and $\log D_{ow}$ ($\log K_{ow}$). The TF of neutral chemicals often increased with increasing $\log K_{ow}$, then decreased when the $\log K_{ow}$ was higher than 1.5–2.0 (Briggs et al. 1982); thus, hydrophilic compounds (i.e., low $\log K_{ow}$) would be taken up more slowly than hydrophobic compounds. Therefore, the high BCFs of SX are due to the high f_n (0.999) (Table 2) and high $\log D_{ow}$. The low BCFs for CHL, STZ, and IP could be explained by their low f_n (0.037 for CHL, 0.0005 for IP) and low $\log D_{ow}$ (-0.51 for CHL, 0.05 for STZ) (Table 2). The root concentration factors of SMX for lettuce were 7.8–19.2, whereas the TF was only 0.07 (Bhalsod et al. 2018), therefore, the very low TF and metabolism of SMX could mostly explain its low BCFs.

The BCFs of IP in the current research were 0.09–0.47, which agrees with the results reported by Santiago-Martín (Santiago-Martín et al. 2020), who found that the BCFs (soil to grain) of IP were less than 1 (mean values of 0.05) in central Spain. The average BCF of each PPCP in wheat grains in the current research was 0.08–2.0, which agrees with those in Liu's study (Liu et al. 2020), in which the BCFs of PPCPs were 0.01–5.0 in the soil-wheat system under wastewater/reclaimed water irrigation in Beijing.

Public health risk assessment

Table 3 shows the adult/toddler EDI and HQs of each studied PPCP in summer maize and winter wheat. The results show that the HQ of individual PPCP for adults and toddlers ranged from 0.0001 to 0.0041 and 0.0004 to 0.0097, respectively, and all the values were lower than 0.1, suggesting that the consumption of cereal grains has little potential risk to human health. For both summer maize and winter wheat, the highest HQs were found for AC-SMX, mostly due to its highest concentrations in cereal grains (Fig. 3). Liu et al. (2020) found that AC-SMX had the highest HQs due to its highest concentrations in agricultural products. Generally, crops are more likely to take up mixtures of PPCPs rather than individual PPCP under reclaimed water irrigation, and the human diet often contains different kinds of crops, therefore, a conservative assessment of the potential risks to human health should be the total HQs of all detected PPCPs in all cereal grains (Prosser and Sibley 2015). In the current research, the total HQs of all the studied PPCPs and all the grains was 0.019 and 0.053 for adults and toddlers, respectively, suggesting little potential risk to human health from grain consumption.

Conclusions

No significant differences were found in the concentrations of PPCPs in topsoil-cereal grains or in the BCFs among these irrigation treatments. For both topsoil and cereal grains, AC-SMX, SX, and IP were presented at higher concentrations than the other studied PPCPs, and the

Table 3 HQ and EDI of each PPCPs in summer maize and winter wheat during 2015–2016

PPCP compound	Cereal	ADI ($\mu\text{g}/(\text{kg}\cdot\text{day})$)	Toddler EDI ($\text{ng}/(\text{kg}\cdot\text{day})$)	Adult EDI	Toddler hazard quotient	Adult hazard quotient
SMM	Wheat	10	7.24	3.10	0.0007	0.0003
	Maize	10	16.5	5.30	0.0016	0.0005
STZ	Wheat	10	46.2	19.8	0.0046	0.0020
	Maize	10	3.96	1.21	0.0004	0.0001
AC-SMX	Wheat	10	48.0	20.6	0.0048	0.0021
	Maize	10	80.6	23.6	0.0081	0.0024
SX	Wheat	10	12.5	5.36	0.0012	0.0005
	Maize	10	15.0	4.19	0.0015	0.0004
SCP	Wheat	10	17.0	7.27	0.0017	0.0007
	Maize	10	9.25	2.48	0.0009	0.0002
SMZ	Wheat	10	9.07	3.89	0.0009	0.0004
	Maize	10	12.0	3.08	0.0012	0.0003
SMT	Wheat	10	96.8	41.5	0.0097	0.0041
	Maize	10	9.45	2.34	0.0009	0.0002
TMP	Wheat	5.7	11.4	4.89	0.0020	0.0009
	Maize	5.7	3.48	0.828	0.0006	0.0001
CHL	Wheat	11	16.0	6.84	0.0015	0.0006
	Maize	11	39.9	9.15	0.0036	0.0008
IP	Wheat	11.4	26.0	11.1	0.0023	0.0010
	Maize	11.4	19.6	4.35	0.0017	0.0004
SMX	Wheat	5.7	3.67	1.57	0.0006	0.0003
	Maize	5.7	10.6	2.27	0.0019	0.0004
Total	–	–	–	–	0.053	0.019

ND not detected, NA not applicable

higher BCFs in this system were obtained by SX and AC-SMX. Consumption of the grains in the current research (irrigation with groundwater, mixed water or reclaimed water) has little potential risk to adults and toddlers. Considering the different PPCP concentrations in reclaimed water in different regions and the variation in the bioaccumulation ability among crop varieties and individual PPCP, further field experiments on reclaimed water irrigation are required, and more attention should be paid to the PPCPs, which could easily transfer in soil-crop system.

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Data availability The datasets generated during and/or analysed during the current study are available upon request from the corresponding author.

Declarations

Conflict of interest There was no conflict of interest among the authors.

Ethical approval Not applicable.

Consent to publish and participate All the authors listed have approved the manuscript that is enclosed. The manuscript is approved by all authors for publication.

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